

ALL-OPTICAL QUANTUM INFORMATION PROCESSING
IN THE ULTRAFAST REGIME

by

KATE FENWICK

A thesis

submitted to the University of Ottawa
in partial fulfillment of the requirements
for the degree of
Doctor of Philosophy in Physics

Department of Physics

Faculty of Science

University of Ottawa

© Kate Fenwick, Ottawa, Canada, 2025

Thesis Examiners

Supervisor	Benjamin Sussman Adjunct Professor Department of Physics, University of Ottawa Group Leader, Ultrafast Quantum Photonics group National Research Council (NRC) of Canada
External examiner	Donna Strickland Professor Department of Physics and Astronomy, University of Waterloo
Internal examiners	Stefanie Czischek Assistant Professor Department of Physics, University of Ottawa Jeff Lundeen Associate Professor and Tier II Canada Research Chair in Quantum Photonics Department of Physics, University of Ottawa Connor Kupchak Assistant Professor Department of Electronics, Carleton University
Defense chairperson	Javier Giorgi Professor Department of Chemistry and Biomolecular Sciences, University of Ottawa

Abstract

Quantum computing offers a promising pathway to solving problems that are intractable for classical computers. Despite recent progress, the development of a *scalable* quantum information processor remains an open challenge. Several hardware platforms remain contenders; however, photonic systems have emerged as a strong candidate due to their ability to operate at room temperature and exceptional resilience to decoherence. State-of-the-art photonic quantum processors use path encoding, which often requires an exponentially increasing number of optical components, making these schemes unavoidably large or lossy in some cases. Time-bin encoding has recently proven to be a promising method to reduce the number of optical components in a photonic quantum processor by enabling operation along a single optical path. Yet, a major barrier to the widespread adoption of time-bin encoding is the challenge of maintaining phase stability across the entire device for extended periods of time. We overcome this obstacle here, by introducing a promising new contender in the photonic quantum computing landscape: *ultrafast time-bin encoding*.

The architecture proposed in this thesis encodes quantum information in the arrival time of photons on ultrafast timescales, processed using optically induced nonlinearities via Kerr gating and birefringent media. A major part of this thesis is the development of the optical Kerr gating technique, which is explored in three initial studies. We first introduce a characterization method, which also happens to enable the carving of ultrashort pulses from a continuous wave laser. With this technique, we demonstrate configurable all-optical Kerr gating down to a temporal resolution of 305 fs. We next investigate translating this high temporal resolution to the frequency domain, using our Kerr gate to measure time-stretched spectra. Finally, we employ

two Kerr gates for measurement of the temporal profiles of entangled photons, subsequently allowing for verification of entanglement. Each of these studies provides valuable insight into the optimal operation of the Kerr gate for quantum information processing.

The Kerr gate is then deployed for the demonstration of our ultrafast time-bin encoding scheme. Operating first in the single photon regime, we implement both a specific quantum algorithm and a more generalized, programmable circuit. Within the large multimode interferometers presented in these two implementations, we see remarkable phase stability over extended periods of time, high fidelity operation, and low loss—all key ingredients for scalability in quantum information processing. Finally, these capabilities are extended to include multiphoton operation, which is a crucial requirement for any photonic quantum information processing system. Taken together, these results demonstrate the potential of ultrafast time-bin encoding for the development of scalable photonic quantum information processing.

Acknowledgments

First and foremost, I am sincerely grateful to my supervisor, Benjamin Sussman, for supporting my growth as a scientist—by encouraging me to learn as much as possible, providing me the opportunity to visit other labs and attend many conferences and meetings across the world, and sharing his wealth of knowledge (often in the form of one of his many textbooks, a few of which I still need to return). The *Ultrafast Quantum Photonics (UQP)* group has grown significantly in recent years, with the high caliber of each team member a testament to his excellence as both a scientist and a person. It has been such a privilege to pursue my PhD in the *UQP* group under his supervision.

Many others in the *UQP* group have played a significant role in making my experience here so positive. I am deeply grateful to Frédéric Bouchard for his continuous support, encouragement, and belief in my abilities over these past years. His mentorship is immensely appreciated, and has been paramount to both this work and my own academic development. I have learned so much from him, and would not be where I am today without all of his guidance and advice. His passion for science is truly contagious, and I look forward to working with him in the upcoming years.

I also wish to extend my utmost thanks to Duncan England and Philip Bustard, for all of their support, guidance, and mentorship—from my time as a co-op student at the *NRC* through to the end of my PhD studies. I have learned a great deal from both of them, including how to approach various problems, and recognizing the importance of balancing the methodology of “just giving it a go in the lab to see if it works” with “thinking it through first, before jumping in”. They have each set such an excellent example of what it means to be a good scientist, heavily influencing my own growth over the years. This thesis would not have been possible without them.

Guillaume Thekkadath also played a pivotal role in my PhD journey. I have really appreciated his support, and his willingness to discuss science—whether in the office or lab, at the climbing gym, or on the ski trails. His passion for science is inspiring, and I have learned a great deal from him. Many of the projects in this thesis would not have been possible without his contributions. I am grateful for all the knowledge he has shared, his impressive coding skills (imperative to our quantum walk simulations), and his patience with me while learning to use *Strawberry Fields*.

The work presented in this thesis would not have been possible without Rune Lausten, who fixed my laser on numerous occasions. His willingness to share his insights, advice, and wealth of experiences is truly appreciated. I have greatly enjoyed all of our scientific discussions, and have learned a lot from him. I also wish to thank Denis Guay and Doug Moffatt for their invaluable technical support and assistance, for lighting up the lab with their positivity, and for teaching me some useful pool techniques.

There are many other individuals within the *UQP* group who have made my PhD experience such a pleasant one. In particular, I would like to thank our excellent summer student, Jonathan Baker—for his enthusiasm and assistance with data processing and simulation, as well as in the lab; Khabat Heshami—for his helpful theory insights; Aaron Goldberg—for his eloquent answers to my many questions and his useful theory insights; Kent Bonsma-Fisher—for his positivity and insightful suggestions during the early years of my PhD; as well as Alicia Sit, Milica Banic, Nicolas Couture, Ramy Tannous, Andrew Proppe, Yingwen Zhang, Brayden Freitas, Noah Lupu-Gladstein, Nicolas Dalbec-Constant, Nathan Roberts, and Colin Veevers—for contributing to the excellent dynamics within the *UQP* group, for their support and scientific insights, and for all the lively lunch conversations.

Beyond the *UQP* group, I wish to thank James Fraser at *Queen's University*—for many things, but ultimately for inspiring me to pursue physics. My journey would have taken a very different path if it weren't for him. I am so incredibly grateful for his support and encouragement throughout my undergraduate and Master's studies (and beyond), and for introducing me to the wonderful field of ultrafast optics. His dedication to the success and professional development of each of his students is extraordinary, and has impacted me profoundly.

I have had the good fortune of collaborating with some phenomenal people during my PhD studies. From the *Institute for Quantum Computing (IQC)* at the *University of Waterloo*, I wish to thank Andrew Cameron, Sandra Cheng, Sacha Schwarz, Benjamin MacLellan, and Kevin Resch—for their hard work and dedication to making our collaboration a fruitful one. I enjoyed working with Andrew so much that we have since collaborated again. At *Fermilab*, I wish to thank Cristián Peña, Si Xie, and Andrew Cameron—for the opportunity to visit during the final months of my PhD; as well as Raju Valivarthi, Thomas Sievert, and Lautaro Narváez at *Caltech*—for their help from afar. I have learned a lot from them in a short period of time. I also thank Andrew and Medea for making me feel so at home during my stay.

Equally important to the many colleagues I have had the pleasure of working with, are the friends and family who have supported me over the years. I am so grateful for my friendship with Thomas, Troy, James, and Tristan, and for their willingness to host me during many a visit to Kingston. I have also been surrounded by some truly wonderful friends in Ottawa, who played an instrumental role in my well-being throughout the course of my PhD; I thank Anja, Cassandra, Sebastian, Helen, Kyle, Andrea, Guillaume, Chris, Lindsay, RJ, Simon, Cat, Thomas, and Troy for their encouragement and for all the fun adventures we have shared. Additionally, I want to thank Maurine and Liam for recently introducing me to the wonderful world of trail running (and for always bringing extra snacks). I am also very grateful to Erin, Hayden, and Nicholas, as well as Tova and Erika—for their support from afar. Also, a big thanks to Michael Schuurman—for all the excellent reading recommendations; and to Albert Stolow and Karina Kraenzle—for their encouragement, and for hosting many an enjoyable gathering.

Finally, to my parents, Kelly Munro and David Fenwick, for their unconditional love and encouragement—not only during my PhD, but throughout my entire life. To my sisters, Julia Fenwick, Elizabeth Fenwick, and Jennifer Fenwick—for being my best friends, for putting up with me, and for always finding a way to make me laugh. To my grandparents, Carol and David Munro—for providing a home-away-from-home (with superb meals and the best company) to visit throughout my studies; and Susan and Marsden Fenwick—for being my biggest cheerleaders from afar. And to Lara Barnett and Nancy Vamvakas for their support and encouragement over the years.

Last, but certainly not least, I am so incredibly grateful to Savva and Andrey Boguslavskiy. Savva, it has been a privilege getting to know you. Thank you for introducing me to the world of anime (and to JoJo, in particular), for sharing your many excellent culinary creations, and for patiently teaching me how to play Halo, Noita, and many other interesting games. You are such a well-rounded, thoughtful, and genuine person, and I am so excited to see where life takes you. Andrey, you have been such a wonderful partner through this journey. Thank you for getting us lost just the right amount in Gatineau park (whether on skis, foot, or bike), for your willingness to listen to me explain my experiments during many a whiteboard session at home, for your unbridled curiosity which has often led me to solutions in the lab, for being audience to countless practice presentations, for celebrating all the little victories with me, for all the encouraging words when I needed them, and for introducing some much-needed spontaneity to my life. I could not have done this without you.

Funding

I have been incredibly lucky to have received financial support for my PhD studies from the *Vanier Canada Graduate (Vanier CGS) Scholarships* program, the *Queen Elizabeth II Graduate Scholarship in Science and Technology (QEII-GSST)*, the *National Research Council (NRC) of Canada*, the *Natural Sciences and Engineering Research Council (NSERC) of Canada*, and the *Boris P. Stoicheff Memorial Graduate Scholarship* through the *Canadian Association of Physicists (CAP)*.

I also gratefully acknowledge the financial support for travel provided by *Optica*, the *Quantum Engineering Centre for Doctoral Training (CDT)* at the *University of Bristol*, the *Okinawa Institute of Science and Technology (OIST)*, the *Mid-infrared Quantum Technologies for Sensing (MIRAQLS)* consortium, and the *University of Warsaw*. Finally, I thank the *Doctoral Mobility Program* (funded by the *Office of the Vice-President, Research and Innovation (OVPRI)*, and managed by *International Research and Experiential Learning (IREX)* at the *University of Ottawa*) and *Fermilab* for supporting my three-month research visit with the *Fermilab Quantum Network (FQNET)* group.

List of Publications and Presentations

Articles published in peer-reviewed journals

- [1] **K. L. Fenwick**, G. S. Thekkadath, P. J. Bustard, D. England, F. Bouchard, and B. Sussman, “Time-of-flight spectroscopy with ultrafast all-optical gating”, *Optics Express* **33**(3): 5914–5922 (2025).
- [2] **K. L. Fenwick**, F. Bouchard, G. S. Thekkadath, D. England, P. J. Bustard, K. Heshami, and B. Sussman. “Photonic quantum walk with ultrafast time-bin encoding”, *Optica* **11**(7): 1017–1023 (2024).
- [3] F. Bouchard, **K. Fenwick**, K. Bonsma-Fisher, D. England, P. J. Bustard, K. Heshami, and B. Sussman. “Programmable Photonic Quantum Circuits with Ultrafast Time-Bin Encoding”, *Physical Review Letters* **133**(9): 090601 (2024).
- [4] A. R. Cameron, **K. L. Fenwick**, S. W. L. Cheng, S. Schwarz, B. MacLellan, P. J. Bustard, D. England, B. Sussman, and K. J. Resch. “Ultrafast measurement of energy-time entanglement with an optical Kerr shutter”, *Physical Review A* **108**(4): L041503 (2023).
- [5] D. England, F. Bouchard, **K. Fenwick**, K. B.-Fisher, Y. Zhang, P. J. Bustard, and B. J. Sussman. “Perspectives on all-optical Kerr switching for quantum optical applications”, *Applied Physics Letters* **119**(16): 160501 (2021).
[Selected for Journal Cover].
- [6] F. Bouchard, D. England, P. J. Bustard, **K. L. Fenwick**, E. Karimi, K. Heshami, and B. Sussman. “Achieving ultimate noise tolerance in quantum communication”, *Physical Review Applied* **15**(2): 024027 (2021).
[Selected as Editor’s Suggestion].
- [7] **K. L. Fenwick**, D. G. England, P. J. Bustard, J. M. Fraser, & B. J. Sussman, “Carving out configurable ultrafast pulses from a continuous wave source via the optical Kerr effect”, *Optics Express* **28**(17): 24845–24853 (2020).

Preprints

- [1] **K. L. Fenwick**, J. Baker, G. S. Thekkadath, A. Z. Goldberg, K. Heshami, P. J. Bustard, D. England, F. Bouchard, and B. Sussman. “Multiphoton interference in a single-spatial-mode quantum walk”, *arXiv preprint arXiv:2409.11483* (2024).

Presentations

- [1] **K. Fenwick**. “All-optical quantum information processing in the ultrafast regime”, *11th International Conference on Quantum Simulation and Quantum Walks* (2025).
Oral presentation; invited.
- [2] **K. Fenwick**, F. Bouchard, D. England, P. Bustard, K. Heshami, and B. Sussman. “Photonic quantum walk with ultrafast time-bin encoding”, *Summer School on Mid-Infrared Quantum Sensing and Technologies (MIRAQLS)* (2024).
Poster presentation.
- [3] **K. Fenwick**, F. Bouchard, G. Thekkadath, J. Baker, A. Goldberg, P. Bustard, D. England, K. Heshami, and B. Sussman. “A heralded single photon and an attenuated coherent state take a quantum walk...”, *Photonics North* (2024).
Oral presentation.
- [4] **K. Fenwick**. “All-optical switching: from classical to quantum regimes”, *National Research Council (NRC) of Canada Seminar Series* (2024).
Oral presentation; invited.
- [5] **K. Fenwick**. “All-optical switching: from classical to quantum regimes”, *Department of Physics, University of Ottawa, Canada Excellence Research Chairs (CERC) Seminar Series* (2024).
Oral presentation; invited.
- [6] **K. L. Fenwick**, F. Bouchard, D. G. England, P. J. Bustard, K. Heshami, and B. J. Sussman. “Photonic quantum walk with ultrafast time-bin encoding”, *OIST School in Physics: Coherent Quantum Dynamics* (2023).
Poster presentation; received travel scholarship to attend.
- [7] **K. L. Fenwick**, F. Bouchard, D. G. England, P. J. Bustard, K. Heshami, and B. J. Sussman. “Photonic quantum walk with ultrafast time-bin encoding”, *International Summer School on Integrated Quantum Photonics* (2023).
Poster presentation; received full scholarship to attend.

- [8] **K. L. Fenwick**, F. Bouchard, D. G. England, P. J. Bustard, K. Heshami, and B. J. Sussman. “Photonic quantum walk with ultrafast time-bin encoding”, *Photonics North* (2023).
Oral presentation; won 1st Place in the COPL and STARaCom Student Best Paper Award Competition.
- [9] **K. L. Fenwick**, F. Bouchard, D. G. England, P. J. Bustard, K. Heshami, and B. J. Sussman. “Photonic quantum walk with ultrafast time-bin encoding”, *Quantum Days* (2023).
Poster presentation; won Best Overall Poster Award.
- [10] **K. L. Fenwick**, P. J. Bustard, D. G. England, and B. J. Sussman. “Mapping frequency to time: A Kerr-shutter spectrometer with ~ 50 GHz resolution”, *Siegman International School on Lasers* (2022).
Poster presentation; received Zeugel Family Scholarship and Optica Travel Scholarship to attend.
- [11] **K. L. Fenwick**, P. J. Bustard, D. G. England, and B. J. Sussman. “Mapping frequency to time: A Kerr-shutter spectrometer with ~ 50 GHz resolution”, *Photonics North* (2022).
Oral presentation.
- [12] **K. L. Fenwick**, D. G. England, P. J. Bustard, J. M. Fraser, and B. J. Sussman. “Carving-out light: Configurable ultrafast pulse generation in single-mode fibre via optical Kerr switching”, *Photonics North* (2020).
Oral presentation; 2nd place in the IEEE Ottawa Photonics Chapter Best Student Paper Award.

Statement of Originality and Collaborative Contributions

To the best of her knowledge, the author states that the work described in this thesis constitutes original research. Author contributions to each of the publications in this thesis are listed below.

Chapter 2 contains the paper entitled “Carving out configurable ultrafast pulses from a continuous wave source via the optical Kerr effect”, published in *Optics Express* in 2020. D. England, P. Bustard, and B. Sussman conceived the idea for this work. K. Fenwick built the experiment, collected and analyzed the data for the paper, and performed the modeling. K. Fenwick drafted the original manuscript and created the figures for the paper. D. England, P. Bustard, J. Fraser, and B. Sussman supervised the project. All authors provided key insights to help move the project forward. D. England handled the correspondence with the editor. All authors reviewed and contributed to the published manuscript.

Chapter 2 also contains the paper entitled “Time-of-flight spectroscopy with ultrafast all-optical gating”, published in *Optics Express* in 2025. P. Bustard, D. England, and B. Sussman conceived the idea for this work. P. Bustard and K. Fenwick built the initial version of the experiment. K. Fenwick built the final version of the experiment, collected and analyzed the data for the paper, and performed the modeling. K. Fenwick drafted the original manuscript and created the figures for the paper. P. Bustard, D. England, and B. Sussman supervised the project. All authors provided valuable insights to help move the project forward. K. Fenwick handled the paper submission process and correspondence with the editor. All authors reviewed and contributed to the published manuscript.

Chapter 3 contains the paper entitled “Ultrafast measurement of energy-time entanglement with an optical Kerr shutter”, published in *Physical Review A* in 2023. K. Resch, B. Sussman, D. England, and P. Bustard conceived the idea for this work. S. Schwarz built the preliminary experimental setup, with assistance from K. Fenwick during a week-long visit. A. Cameron and S. Cheng built the final version of the experiment, with assistance from K. Fenwick during another week-long visit. A. Cameron performed the experiment and collected the data, with remote assistance from K. Fenwick, D. England, and P. Bustard. S. Schwarz, B. MacLellan, and A. Cameron developed the theory and model, with assistance from P. Bustard, D. England, and K. Fenwick. A. Cameron and K. Fenwick drafted the original manuscript. A. Cameron created the figures for the manuscript. K. Resch, B. Sussman, P. Bustard, and D. England supervised the project. A. Cameron handled the correspondence with the editor. All authors reviewed and contributed to the published manuscript.

Chapter 4 contains the paper entitled “Photonic quantum walk with ultrafast time-bin encoding”, published in *Optica* in 2024. K. Fenwick, F. Bouchard, D. England, P. Bustard, K. Heshami, and B. Sussman conceived the idea for this work. K. Fenwick built the experiment, and collected and analyzed the data for the paper. K. Fenwick, F. Bouchard, and G. Thekkadath developed the model. K. Fenwick drafted the original manuscript and created the figures for the paper. F. Bouchard, D. England, P. Bustard, K. Heshami, and B. Sussman supervised the project. F. Bouchard handled the correspondence with the editor. All authors reviewed and contributed to the published manuscript.

Chapter 4 also contains the paper entitled “Programmable Photonic Quantum Circuits with Ultrafast Time-Bin Encoding”, published in *Physical Review Letters* in 2024. F. Bouchard, K. Fenwick, K. Bonsma-Fisher, D. England, P. Bustard, K. Heshami, and B. Sussman conceived the idea for this work. F. Bouchard built the experiment, collected and analyzed the data for the paper, and performed the modeling for the demonstration of programmability. K. Fenwick built the experiment, collected and analyzed the data for the paper, and performed the modeling for the demonstration of scalability. F. Bouchard drafted the original manuscript and created the figures for the paper. F. Bouchard, D. England, P. Bustard, K. Heshami, and B. Sussman supervised the project. F. Bouchard handled the correspondence with the

editor. All authors reviewed and contributed to the published manuscript.

Chapter 5 contains the preprint entitled “Multiphoton interference in a single-spatial-mode quantum walk”, posted to *arXiv* in 2024. K. Fenwick, F. Bouchard, G. Thekkadath, D. England, P. Bustard, K. Heshami, and B. Sussman conceived the idea for this work. K. Fenwick built the experiment and collected the data for the paper. J. Baker processed the data for the paper. K. Fenwick and J. Baker analyzed the data for the paper. J. Baker developed the preliminary modeling. G. Thekkadath developed the *Strawberry Fields* simulation, which J. Baker then expanded. K. Fenwick drafted the original manuscript and created the figures for the paper. F. Bouchard, D. England, P. Bustard, K. Heshami, and B. Sussman supervised the project. A. Goldberg provided key insight into the theory presented in the paper. F. Bouchard handled the paper submission process and correspondence with the editor. All authors reviewed and contributed to the submitted manuscript.

Table of Contents

Thesis Examiners	ii
Abstract	iii
Acknowledgments	v
List of Publications and Presentations	ix
Statement of Originality and Collaborative Contributions	xii
Table of Contents	xv
List of Figures	xviii
List of Tables	xxi
List of Abbreviations	xxii
Chapter 1: Introduction	1
1.1 The problem of solving problems	1
1.2 A brief history of quantum computing	2
1.3 Quantum computing with photons	14
1.4 Our all-optical approach	15
1.5 Organization of Thesis	16
Chapter 2: Some background on ultrafast and nonlinear optics	17
2.1 Ultrafast optics	18
2.1.1 Generating ultrashort pulses	19
2.1.2 Propagation of ultrashort pulses in media	24
2.1.3 Manipulating ultrashort pulses	30
2.2 Nonlinear optics	31
2.2.1 Nonlinear propagation of electric fields in media	33
2.2.2 Second-order nonlinear optical processes	36

2.2.3	Third-order nonlinear optical processes	50
2.2.4	Optical Kerr effect	54
2.2.5	All-optical Kerr gating	58
2.3	Configurable all-optical Kerr gating	63
2.3.1	Publication: Carving out configurable ultrafast pulses from a continuous wave source via the optical Kerr effect	65
2.4	Ultrafast spectro-temporal modulation	75
2.4.1	Submitted manuscript: Time-of-flight spectroscopy with ultrafast all-optical gating	78
Chapter 3: Some background on quantum optics		91
3.1	Quantum optics	92
3.1.1	Quantization of the electromagnetic field	93
3.1.2	Quantization of the electromagnetic field in unbounded free space*	100
3.1.3	Quadrature operators of the electric field	103
3.1.4	Different states of light	104
3.2	Generating single photons	129
3.2.1	Attenuated coherent states of light	130
3.2.2	Deterministic single-photon sources	131
3.2.3	Probabilistic single-photon sources	132
3.3	Detecting single photons*	135
3.3.1	Avalanche photodiodes*	137
3.3.2	Superconducting nanowire single-photon detectors*	138
3.3.3	Transition-edge sensors*	140
3.3.4	Time-to-digital conversion*	143
3.4	Characterizing single-photon sources	143
3.4.1	Mean photon number	144
3.4.2	Heralding efficiency	145
3.4.3	Quantum coherence and quantum correlation functions*	145
3.4.4	Second-order autocorrelation	156
3.4.5	Second-order cross-correlation	158
3.4.6	Coincidence-to-accidental ratio	160
3.4.7	Spectral and temporal correlations	160
3.5	Characterization of ultrashort photon pairs	164
3.5.1	Publication: Ultrafast measurement of energy-time entanglement with an optical Kerr shutter	166
Chapter 4: Quantum information processing with single photons		172
4.1	Linear optical quantum computing	173

4.1.1	Linear optical elements	176
4.1.2	Circuit-based approach	178
4.1.3	Boson sampling protocol	181
4.1.4	Cluster-state-based photonic quantum computing	183
4.2	Photonic quantum algorithms	184
4.2.1	Random walks: the inspiration for quantum walks	184
4.2.2	The quantum walk (QW)	186
4.3	Encoding methods in photonic quantum information processing	187
4.3.1	Ultrafast time-bin encoding (UTBE)	191
4.4	Implementing a quantum algorithm	196
4.4.1	Publication: Photonic quantum walk with ultrafast time-bin encoding	199
4.5	Implementing programmable circuits	216
4.5.1	Publication: Programmable photonic quantum circuits with ultrafast time-bin encoding	217
Chapter 5:	Multiphoton quantum information processing	236
5.1	Requirements for scalable quantum information processing	237
5.2	Multiphoton quantum interference	238
5.2.1	Two-photon Hong-Ou-Mandel (HOM) interference	239
5.2.2	Multiphoton interference	242
5.3	Implementing multiphoton capabilities	243
5.3.1	Publication: Multiphoton interference in a single-spatial-mode quantum walk	244
Chapter 6:	Concluding remarks	260
6.1	Summary and conclusions	260
6.2	Outlook	262
Bibliography		265

List of Figures

2.1	Multimode laser operation.	21
2.2	Mode-locked laser output with constant mode phase.	23
2.3	Depiction of second-harmonic generation (SHG).	37
2.4	Depiction of sum-frequency generation (SFG).	40
2.5	Wavevector mismatch in SFG.	44
2.6	Birefringence in an optical crystal.	45
2.7	Depiction of difference-frequency generation (DFG).	48
2.8	An optical parametric oscillator (OPO).	50
2.9	Depiction of degenerate four-wave mixing (FWM).	53
2.10	Effects due to the intensity-dependent refractive index.	54
2.11	Schematic of an optical Kerr shutter (OKS).	61
2.12	Pulse carving with an optical Kerr gate.	64
2.13	Spectral sampling with an optical Kerr gate.	76
2.14	Temperature-stable box housing the 1 km PMF.	77
3.1	Photon energy level spectrum.	100
3.2	Quadrature representation of the vacuum and number states.	109
3.3	Quadrature representation of the coherent state.	113
3.4	Probability of detecting n photons in a coherent state.	114

3.5	Quadrature representation of a squeezed vacuum state. . . .	117
3.6	Probability of detecting n photons in a squeezed vacuum state.	120
3.7	Probability of detecting n_1 and n_2 photons in modes a and b of a two-mode squeezed vacuum (TMSV) state.	123
3.8	Model of a black body.	124
3.9	Probability of detecting n photons in a thermal state.	128
3.10	Idealistic single-photon emitter.	131
3.11	Probabilistic photon pair source.	133
3.12	Schematic of a photodiode.	137
3.13	Schematic of a superconducting nanowire single-photon de- tector (SNSPD).	139
3.14	Schematic of a transition-edge sensor (TES).	141
3.15	Young's double-slit experiment.	146
3.16	First-order correlation, $g^{(1)}(\tau)$	151
3.17	Hanbury Brown-Twiss interferometer.	152
3.18	Second-order correlation, $g^{(2)}(\tau)$	155
3.19	Detection of photons as a function of time.	156
3.20	Second-order autocorrelation for a photon pair source.	157
3.21	Second-order cross-correlation for a photon pair source. . . .	159
3.22	Joint spectral intensity (JSI) illustration.	162
3.23	Joint spectral and temporal correlations.	163
3.24	Measurement of the temporal wave packet of a photon with an optical Kerr gate.	165
4.1	A beam splitter (BS).	176

4.2	The Bloch (or Poincaré) sphere.	177
4.3	Universal N -port interferometer.	179
4.4	Depiction of a discrete one-dimensional random walk.	185
4.5	Depiction of a discrete one-dimensional quantum walk (QW).	186
4.6	Encoding methods in photonic quantum information processing.	188
4.7	Photonic quantum circuits with ultrafast time-bin encoding (UTBE).	192
4.8	Photonic quantum walk with ultrafast time-bin encoding (UTBE).	195
4.9	Passive interferometers used in ultrafast time-bin encoding (UTBE).	197
5.1	Two-photon interference at a beam splitter.	240
5.2	A typical HOM experiment.	241

List of Tables

1.1	Summary of quantum computing platforms.	13
2.1	Mode locking methods.	25
2.2	Phase matching in uniaxial crystals.	46
2.3	Comparison of optical modulation methods.	62
3.1	Criteria of the ideal single-photon detector.	136
4.1	Approaches to linear optical quantum computing (LOQC). . .	175
4.2	Photonic quantum information encoding methods.	189
5.1	DiVincenzo's criteria for quantum computing.	237

List of Abbreviations

<i>1QBit</i>	<i>1QB Information Technologies, Inc.</i>
2D	two-dimensional
α -BBO	alpha-barium borate
APD	avalanche photodiode
AR	anti-reflection
<i>CAP</i>	<i>Canadian Association of Physicists</i>
CAR	coincidence-to-accidental ratio
c.c.	complex conjugate
<i>CDT</i>	<i>Quantum Engineering Centre for Doctoral Training</i>
CNOT	controlled-NOT
CW	continuous-wave
DFG	difference-frequency generation
DM	dichroic mirror
<i>D-Wave</i>	<i>D-Wave Quantum Systems Inc.</i>
FFT	fast Fourier transform
<i>FQNET</i>	<i>Fermilab Quantum Network group</i>
FWHM	full width at half maximum

FWM	four-wave mixing
GVD	group-velocity dispersion
HeNe	helium-neon
HOM	Hong, Ou, and Mandel
HWP	half-wave plate
IFFT	inverse fast Fourier transform
<i>IQC</i>	<i>Institute for Quantum Computing</i>
<i>IREX</i>	<i>International Research and Experiential Learning</i>
JSA	joint spectral amplitude
JSI	joint spectral intensity
JTA	joint temporal amplitude
JTI	joint temporal intensity
KLM protocol	Knill, Laflamme, and Milburn protocol
LPR	linear phase retarder
MEMS	micro-electromechanical systems
<i>MIRAQLS</i>	<i>Mid-infrared Quantum Technologies for Sensing</i>
<i>MIT</i>	<i>Massachusetts Institute of Technology</i>
ND	neutral density
NISQ	noisy intermediate-scale quantum
NMR	nuclear magnetic resonance
NOLM	nonlinear optical loop mirror
<i>NRC</i>	<i>National Research Council of Canada</i>
<i>NSERC</i>	<i>Natural Sciences and Engineering Research Council of Canada</i>

OAM	orbital angular momentum
<i>OIST</i>	<i>Okinawa Institute of Science and Technology</i>
OKS	optical Kerr shutter
OPA	optical parametric amplifier
OPO	optical parametric oscillator
<i>OVPRI</i>	<i>Office of the Vice-President, Research and Innovation</i>
PBS	polarizing beam splitter
PCF	photonic crystal fiber
PMF	polarization-maintaining fiber
PNRD	photon-number-resolving detector
POUTINE	programmable optical ultrafast temporal interferometric network element
<i>QEII-GSST</i>	<i>Queen Elizabeth II Graduate Scholarship in Science and Technology</i>
QW	quantum walk
QWP	quarter-wave plate
RSA	Rivest-Shamir-Adleman
SCG	supercontinuum generation
SFWM	spontaneous four-wave mixing
SFG	sum-frequency generation
SHG	second-harmonic generation
SMF	single-mode fiber
SNR	signal-to-noise ratio

SNSPD	superconducting nanowire single-photon detector
SPM	self-phase modulation
$SU(2)$	Special Unitary Group of degree 2
TES	transition-edge sensor
TBP	time-bandwidth product
Ti:Sapph	titanium-doped sapphire
<i>UQP</i>	<i>Ultrafast Quantum Photonics group</i>
<i>USTC</i>	<i>University of Science and Technology of China</i>
UTBE	ultrafast time-bin encoding
<i>Vanier CGS</i>	<i>Vanier Canada Graduate Scholarships</i>
XPM	cross-phase modulation

Chapter 1

Introduction

1.1 The problem of solving problems

Science and engineering revolve around problem-solving. When scientists and engineers encounter problems that are too difficult to solve by hand, they turn to computers. If these problems cannot be solved by regular computers, they turn to supercomputers. But what about problems that are intractable for even the most powerful supercomputer? For example, as Richard Feynman proposed in his seminal lecture in 1982, the task of simulating quantum systems: “Nature isn’t classical, dammit, and if you want to make a simulation of nature, you’d better make it quantum mechanical, and by golly it’s a wonderful problem, because it doesn’t look so easy” [1]. Another example is factoring large numbers—the backbone of modern-day security [2]. Such tasks require a quantum computer. From identifying subtle fraudulent patterns in financial transactions [3] to explaining new supercollider physics [4], a large-scale, fully programmable quantum computer will indeed be disruptive.

Recent years have seen extraordinary advancements in isolating [5, 6, 7], manipulating [8], and detecting [9, 10, 11] quantum systems—making physical implementations of quantum processors a reality. Although there has been a great deal of progress, platforms to date still suffer from drawbacks which limit their promise for scalability. The development of a *scalable* quantum information processor remains an open challenge, although there is still no easy answer for the question of what form the

hardware will take. In the global effort to develop quantum computers, several platforms remain contenders, including trapped atoms [12] or ions [13, 14], nuclear magnetic resonance [15], superconducting qubits [16], and photonic platforms [17, 18, 19]. The historical development of these platforms, as well as their benefits and limitations, are discussed in the following section¹.

1.2 A brief history of quantum computing

1960s–1970s: Major developments in information theory lead to the first quantum information theory proposals.

Quantum computing is rooted in the developments of classical computing and information theory. Based on the work of Szilard [20] and von Neumann [21], who connected the concept of thermodynamic entropy [22] with information theory [23, 24], in the 1960s Landauer determined a fundamental lower bound on any irreversible operation on a bit [25]. Naturally, physicists sought to determine how such a limit could be beat. In 1973, Bennett proposed the concept of reversible computation, without the erasing of information, to overcome the Landauer limit [26]. Bennett discussed his findings with Feynman, asking how they may be affected by quantum mechanics [27]. In the same year, Holevo established an important upper bound on the accessible information of a quantum state [28], leading to the 1975 demonstration from Poplavskiy of the computational infeasibility of simulating quantum systems on classical computers [29]. Ingarden then presented one of the first attempts at creating a quantum information theory in 1976 [30], which is a generalization of Shannon theory [31]. And in 1980, Manin motivated the idea of a quantum computer, describing logical operation on qubits [32]; this work did not become well-known until the 1990s, after which it significantly influenced developments in the field. A frequently cited work from Benioff in 1980 proposed a quantum mechanical model of the Turing machine [33], but it should be noted that this proposal does not exploit entanglement.

¹Brace yourself—the next section is rather long; but, it was educational for me to review these developments, and I believe there is value in having this history summarized in one place.

1980s: The “birth” of quantum computing, the first quantum algorithms, and the first proposals of qubit operations.

Although many bodies of research have contributed to the early development of quantum computing, Feynman’s 1982 lecture [1] is broadly considered the origin of the new field of quantum computing. Quantum computers are not strictly “better” than classical computers; indeed, there are many problems for which classical computers may be more suitable. It has also not been explicitly proven that quantum computers are more powerful than classical computers—this is still merely a conjecture (albeit, a strong one). Perhaps the main attribute of quantum computers, which provides them an advantage, is that they can perform quantum algorithms which are significantly faster than the best known classical algorithms. In 1985, Deutsch formalized the concept of a quantum Turing machine (universal quantum computer) [34], a device on which any quantum algorithm can be expressed. This gave way to new quantum algorithms, such as the Deutsch-Jozsa algorithm—one of the first results showing that quantum computers can solve certain problems exponentially faster than classical computers [35]. Around this time, proposals for the implementation of quantum computational gates began to emerge, based on prior work from Fredkin and Toffoli [36, 37]. Yamamoto and Igeta proposed, in 1988, quantum computing and networking protocols in which atoms perform two-qubit operations and photons transmit the qubits [38]. Then in 1989, Milburn proposed a quantum-optical method for realizing the Fredkin gate [39]. Still, at this time, the idea of quantum computation was entirely theoretical and interest was predominantly limited to the academic community.

1990s: Major breakthroughs in quantum algorithms, followed by the first experimental demonstrations.

It was not until 1994, when Shor developed an algorithm for efficiently factoring large integers, that interest in quantum computing grew beyond the academic community. Known today as “Shor’s algorithm”, it presents strong evidence of super-polynomial speedup compared to known classical algorithms [40] and could theoretically break many cryptosystems widely in use today, including the Rivest-Shamir-Adleman (RSA) cryptosystem [41]. Only one year later, in 1995, Shor proposed the

first schemes for quantum error correction [42], the need for which was first proposed by Peres [43] in 1985. Error correction is now viewed as essential to achieving the high complexity and large circuit depth graphs required for useful quantum computing, by protecting quantum information from errors arising due to decoherence and noise. Improving upon Shor’s quantum error correction, two independent proposals—one from Aharonov and Ben-Or [44] and one from Knill, Laflamme, and Zurek [45]—presented fault-tolerant quantum computation, allowing for arbitrarily long quantum computation below a certain physical error threshold rate. Around the same time Kitaev proposed an algorithm for quantum phase estimation [46] and Coppersmith proposed the quantum Fourier transform [47] (though this was only published in 2002); both are important subroutines of many quantum algorithms. Grover introduced his quantum database search algorithm in 1996, now referred to as the “Grover search algorithm”, providing a quadratic speedup which can be applied to a wide range of problems [48]. Other important quantum algorithms were developed in 1997, by Simon [49] and by Bernstein and Vazirani [50]. The first experimental demonstrations of quantum logic gates also occurred around this time. In 1995, Monroe and Wineland used trapped ions to experimentally realize the first quantum logic gate, a controlled-NOT (CNOT) gate [51]. This followed the first experimental demonstrations of laser-cooled ions in 1992 [52, 53, 54], and the 1995 proposal of Cirac and Zoller to use cold trapped ions to perform a CNOT gate [55].

More experimental implementations emerged in the late 1990s. Quantum computational gates based on nuclear magnetic resonance (NMR)—first proposed by Lloyd [56] and DiVincenzo [57, 58]—were demonstrated independently in 1997 by Cory, Fahmy and Havel [59] and Gershenfeld and Chuang [60]. This led to the work of Jones and Mosca in 1998, which implemented a two-qubit NMR quantum computer in order to solve Deutsch’s problem [61]. Shortly thereafter, Chuang and Kubinec implemented a Grover search algorithm using NMR [62]. Around the same time, Loss and DiVincenzo proposed the Loss-DiVincenzo quantum computer, which is based on controlling the spin of charge carriers in quantum dot semiconductors [63]. In the same vein, Kane proposed theoretically the first silicon-based nuclear spin quantum computer, which would use nuclear spins of individual phosphorus atoms in silicon as the qubits and donor electrons to mediate the coupling between qubits [64]. Also

in 1998, Gottesman and Knill proved that some quantum computations could be perfectly simulated in polynomial time (*i.e.*, on a classical computer) [65]. The excitement surrounding NMR qubits was dampened a bit in 1999, when it was shown that none of the quantum computational experiments of NMR qubits at the time could contain any entanglement due to poor scaling of the signal-to-noise ratio; the quantum states were too strongly mixed [66]. Yet, it still remains an open question as to whether entanglement is necessary for quantum computational speedup. Other potential quantum computational platforms began emerging in 1999, including an experimental demonstration of quantum annealing in a condensed matter system by Aeppli and Rosenbaum [67], results for which the newly-founded Canadian quantum computing company, *D-Wave Quantum Systems Inc. (D-Wave)*, based their approach on. Yet another platform emerged in this year when Nakamura, Pashkin, and Tsai showed that a superconducting circuit could be used as a qubit [68].

2000s: The development of early quantum computers and proof-of-concept demonstrations, and the announcement of the first industrial systems.

With these early successes, the interest in quantum computing started to grow more rapidly. Given the growing efforts towards full-scale quantum computing, DiVincenzo published a list of requirements for the implementation of quantum computation in 2000 [69]. Throughout the 2000s, many more proof-of-concept demonstrations emerged. Though limited in capability and scale, these early demonstrations still showed the basic principles of quantum computing. In 2000, a 5-qubit NMR computer was demonstrated at the *Technical University of Munich in Germany* [70], and a 7-qubit NMR computer was developed at *Los Alamos National Laboratory* [71]. In the same year, the concept of adiabatic quantum computing was proposed at the *Massachusetts Institute of Technology (MIT)*, further diversifying the approaches to quantum computation [72]; while the “universal” approach to quantum computing takes a gate-based model, adiabatic quantum computing prepares a quantum system in some initial state and allows it to evolve to the desired final ground state. Also in 2000, researchers at *IBM, Stanford*, and the *University of Calgary* reported the demonstration of a quantum order finding algorithm (with the same structure as Shor’s algorithm) using a 5-qubit NMR quantum computer [73].

Only a year later, in 2001, Shor’s factoring algorithm was realized experimentally, using seven NMR qubits to factor the number 15 [74]. Further doubt about the future of NMR computing were raised around this time, when Linden and Popescu proved that entanglement is necessary for a large class of quantum computational protocols [75]. On the other hand, the viability of optical quantum computing was established in a breakthrough proposal by Knill, Laflamme, and Milburn in 2001, using only single-photon sources, linear optical elements, and single-photon detectors [76]. In the same year, Raussendorf and Briegel proposed measurement-based quantum computation [77], using measurements to imprint a specific quantum logic circuit onto a particular class of entangled states, called cluster states. In 2002, the first two-dimensional (2D) array of neutral atoms, trapped using optical tweezers, was demonstrated at the *University of Hannover* [78]. In the following year, researchers led by Blatt at the *University of Innsbruck* demonstrated the Deutsch-Jozsa algorithm [79] and a CNOT quantum gate [80] on a trapped-ion quantum computer. Demonstrations of quantum CNOT gates using only linear optical elements were also demonstrated around this time, independently by Pittman *et. al.* at *Johns Hopkins University* [81] and by O’Brien *et. al.* at the *University of Queensland* [82].

In 2004, the first instance of five-photon entanglement—the minimum number of entangled qubits for universal quantum error correction—was demonstrated by Jian-Wei Pan’s team at the *University of Science and Technology of China (USTC)* [83]. Also demonstrated in this year was the first working pure state NMR quantum computer [84]. Another large step was made in 2005, this time in the direction of superconducting qubits, where the capacitance of a Josephson junction [85] was measured for the first time independently by two teams, one at *Chalmers University of Technology* in Sweden [86] and another between the *Helsinki University of Technology* and the *Landau Institute of Theoretical Physics* in Moscow [87]. Also in 2005, quantum-entangled W-states of up to 8 qubits were demonstrated using trapped ions [88]. Further developmental leaps were made in 2006 towards trapped atom and ion quantum computers, with demonstrations of seven atoms in a stable line at the *University of Bonn* [89] and the first 2D ion trap at the *National Institute of Standards and Technology (NIST)* [90]. Also in this year, a group at the *University of Illinois at*

Urbana-Champaign demonstrated a photonic implementation of counterfactual quantum computation [91], where results are inferred without actually running a quantum computation. This year also saw developments of other potential solid-state quantum computer technology, including quantum dots [92] and a silicon-phosphorus Kane quantum computer [93]. Yet another large milestone in 2006 was the demonstration of a 12-qubit NMR quantum computer by researchers at the *Institute for Quantum Computing (IQC)* at the *University of Waterloo*, the *Perimeter Institute for Theoretical Physics*, and *MIT* [94].

Developments continued through the late 2000s, in photonic platforms [8, 95, 96, 97, 98, 99], trapped atoms [100, 101] and trapped ions [102, 103, 104], as well as in superconducting [105, 106, 107, 108, 109] and other solid-state quantum computing systems [110, 111, 112, 113, 114, 115]. Notable steps in photonic quantum computing during these years include demonstrations of the first one-way (measurement-based) quantum computer [116], a compiled version of Shor’s algorithm with quantum entanglement [117, 118], and waveguide quantum circuits [119], culminating in demonstrations of Shor’s factoring algorithm on a photonic chip at the *University of Bristol* [120], and of a general technique for quantum computational gates using higher-dimensional encoding schemes [121]. Superconducting quantum information platforms also saw major steps reported, for example the synthesis of arbitrary states [122] and the demonstration of two-qubit algorithms [123]. Several demonstrations of the coupling of different types of qubits were reported [124, 125, 126, 127, 128], holding importance in the context of quantum memory [129] or even potentially linking two types of quantum computers together. In the same years, the possibility of quantum computing without entanglement was described [130], and steps towards topological quantum computers were also emerging [131, 132]. On the industrial side, *D-Wave* claimed to have developed a 128-qubit computer chip in 2008, though there was (and still remains) skepticism about whether these systems are truly quantum computers. *Google* then announced its collaboration with *D-Wave* in 2009 to develop image search technology using quantum computing [133]. This year also marked a significant step in trapped-ion quantum computing, where researchers at *NIST* published a complete method for trapped-ion quantum information processing, on a chip [134]—this was viewed, by some, as “the first quantum computer”, but it ought

to be noted that there exist several claims of this kind. Nevertheless, this was still a major step in the field and solidified the view at the time that the most promising approach to quantum computing was with trapped ions. Also in 2009, Harrow, Hassidim, and Lloyd published their quantum algorithm for solving linear systems of equations, showing exponential speedup in comparison to any classical algorithm for the same problem [135].

2010s: The race to demonstrate “quantum advantage”, both in academia and industry.

The 2010s consisted of key advancements across all potential quantum computing platforms, punctuated by announcements in the rush to get commercial quantum processors to market. In 2010, researchers at *NIST*, led by Wineland, realized a programmable two-qubit quantum processor with trapped ion technology [136]. In the same year, Lanyon *et. al.* used a linear optic quantum computing architecture to calculate the energy spectrum of the hydrogen molecule to 20 bits of precision [137], and Peruzzo *et. al.* demonstrated a particular quantum algorithm, the quantum walk, of two photons in an array of 21 waveguides [138]. Yet another work which would have a large impact in photonic quantum computing was the 2011 boson sampling proposal of Aaronson and Arkhipov [139], demonstrating a problem which cannot be efficiently simulated by classical computers. In 2011, *D-Wave* introduced their product, “D-Wave One”, operating on a superconducting 128-qubit processor [140]. The same year saw 14-qubit entanglement [141] and advancements in error correction [142, 143] in trapped-ion systems. At the same time, superconducting qubits were used to implement the first quantum von Neumann architecture [144], and demonstrations of quantum memory [145] and macroscopic entanglement [146] were reported in diamond. In 2012, new proposals emerged in surface code quantum computing for error correction [147], the world’s first dedicated quantum computing software company, *1QB Information Technologies, Inc. (1QBit)*, was founded, and diamond-based quantum memory was shown to exceed one second [148]. This was followed by a 2013 demonstration of room temperature storage in silicon defects of over 39 minutes [149]. Also in 2013, Childs, Gosset, and Webb showed that a multiparticle quantum walk algorithm is capable of universal quantum computation [150]. Note

that many other advancements were made in the early 2010s, including in trapped ions [151, 152, 153, 154, 155], solid-state systems [156, 157, 158, 159, 160, 161, 162, 163] and superconducting technology [164], photonics [165, 166, 167, 168, 169, 170], and topological implementations [171].

In 2015, *D-Wave* announced the availability of the “D-Wave 2X” system, claimed to be a 1000-qubit+ quantum processor. Skepticism remained as to whether this system could truly be called a quantum computer. This year also saw demonstrations of nuclear spin qubits with six-hour coherence times [172], quantum error detection code in superconducting qubit lattices [173], and 2-qubit logic gates in silicon defects [174]. By 2016, other players in the quantum computing market emerged. *IBM* released what they called “the Quantum Experience”, an online interface to their superconducting quantum information processing systems. Their system was immediately put to use to demonstrate new protocols in quantum information processing [175, 176]. *Google* also announced a simulation of the hydrogen molecule, performed with an array of 9 superconducting qubits developed by the Martinis group at *UC Santa Barbara* [177]. Another big collaboration, between Blatt (*University of Innsbruck*) and Chuang (*MIT*), developed an efficient protocol for implementing Shor’s algorithm in a trapped-ion quantum computer [178]. At the same time, quantum logic gates with extremely high fidelities were demonstrated in trapped-ion qubits [179]. *D-Wave* pushed a 2000-qubit quantum annealer, the “D-Wave 2000Q”, to market in 2017, while *IBM* unveiled a quantum computer with 17 superconducting qubits, later demonstrating 50 qubits where the quantum state could be preserved for 90 seconds. In the same year, *Microsoft* revealed its quantum programming language, “Q#”, which is integrated with its Visual Studio development environment; programs could be executed locally on a 32-qubit simulator, or on a 40-qubit simulator through Azure. Also in 2017, high-dimensional entangled photon states were generated on-chip [180].

By 2018, quantum processors with up to (and over) 1,000 qubits had been demonstrated, but they were not yet large enough nor advanced enough to demonstrate fault tolerance and quantum advantage. As such, Preskill coined a new term to describe the current state of the field: the “Noisy Intermediate-Scale Quantum” (NISQ) era [181]. Announcements from other quantum computing startups emerged at this

time. *QuTech* presented their silicon quantum dot qubit platform [182], *Google* announced their 72-qubit chip, “Bristlecone” [183], *Intel* confirmed the development of a 49-qubit superconducting test chip, “Tangle Lake” [184], and *IonQ* introduced the first commercial trapped-ion quantum computer, which was later benchmarked in 2019 [185]. In 2019, *IBM* unveiled the “IBM Q System One”, the first circuit-based commercial quantum computer. *Google* also revealed its “Sycamore” processor, consisting of 53 qubits, after which it published a paper claiming that it had reached “quantum supremacy” [17]. Note that nowadays, the term “quantum supremacy” has been replaced by “quantum advantage”. Many other developments in the late 2010s were published, for example in trapped ions [186, 187], superconducting qubits [16], and in photonics [188, 189]. This decade ended with an impressive demonstration of boson sampling with 14 detected photons from Jian-Wei Pan’s team at the *USTC* [190].

2020s: Interest in quantum computing is at an all-time high, with commercial systems evolving rapidly.

Jian-Wei Pan’s team followed this up in 2020 with a demonstration of quantum computational advantage using their photonic quantum computer, “Jiuzhang”, capable of performing calculations at 100 trillion times the speed of classical supercomputers [18]. In the same year, it was found that ionizing radiation from cosmic rays and radioactive materials in the environment may pose limitations to the coherence times of superconducting qubits if they are not shielded adequately [191]. *Honeywell* announced their trapped-ion quantum computer with a quantum volume of 64 and also introduced a subscription service to access it. Meanwhile, the Canadian photonic quantum computing startup, *Xanadu*, offered their own cloud service. Also in 2020, *Google* reported the largest quantum chemistry simulation performed on a quantum computer to-date, using a Hartree-Fock approximation on their “Sycamore” processor, paired with a classical computer [192]. In 2021, *Honeywell* announced an increase in their quantum volume to 512, and *IBM* published a quantum advantage paper [193], later announcing its 127-qubit processor, “IBM Eagle”. Microsoft began to offer “Azure Quantum”, their cloud quantum computing service, and *Rigetti* developed a method for networking their superconducting quantum computing chips [194].

In a similar vein, researchers at the *Max-Planck-Institute for Quantum Optics* in Germany demonstrated a prototype for distributed quantum computing [195]. Further commercially minded demonstrations included the first compact trapped-ion quantum computer, fitting into two 19-inch racks [196]. Programmable Rydberg atom arrays, a subset of neutral atom quantum computing, also saw large developments in 2021 [197, 198]. In the same year, Jian-Wei Pan’s team added yet another impressive publication to the literature, reporting a 66-qubit programmable superconducting quantum processor, “Zuchongzhi”, claimed to be one million times more complex and 10 million times faster than Google’s “Sycamore” [199].

Quantinuum (previously *Honeywell*) announced its new “H1-2” system in 2022, which they claimed to have a quantum volume of 4096. *IBM* presented its 433-qubit “Osprey” quantum processor in this year, as well. Turning its attention back to NMR-based quantum computing, Japan released the first portable 3-qubit device for educational purposes. Another exciting development in 2022, *Xanadu*, demonstrated quantum computational advantage using their programmable photonic processor [19]. In 2023, *QuiX Quantum* demonstrated an impressive 20-mode photonic quantum processor [200], *Alpine Quantum Technologies* demonstrated a quantum volume of 128 on its 19-inch rack system, “PINE”, the startup *Atom Computing* exceeded 1,000 qubits, fusion-based quantum computation was proposed [201], *Microsoft* declared its work on topological quantum computing with Majorana fermions, and *IBM* reported evidence for the utility of quantum computing in a pre-fault-tolerant era [202] and presented its 1121-qubit processor “Condor”, which is the culmination of its multi-year “Roadmap to Quantum Advantage”. This year, the difficulty of simulating Gaussian boson sampling was put to the test, with a new classical simulation algorithm [203]; however, the boson sampling community need not worry yet, as there is still evidence that noiseless Gaussian boson sampling remains classically intractable. Simmons, at *Photonic Inc.*, published results demonstrating the promise of defect centers in silicon [204], bringing this technology to join other front-runners in the field. *Quantinuum* also published its new “H2-1” quantum processor, now with 56 qubits [205], and the *Psi-Quantum* team published results from their photonic quantum information processor, boasting remarkably high interference visibility and state fidelity [206]. Most recently,

Google published a breakthrough error correction scheme [207], using its new “Willow” processor, a major step towards fault-tolerant quantum computing. There are many other notable works through the 2020s to present, including research in various solid-state quantum computing platforms [208, 209, 210, 211, 212, 213, 214, 215], trapped-ion [216, 217, 218] and trapped-atom [12, 219, 220] quantum computers, as well as in photonic quantum computing platforms [221, 222, 223].

As of today, we are still in the NISQ era of quantum computing, where noise and decoherence remain a challenge. Though a robust ecosystem is emerging in which current quantum processors have been shown to perform certain tasks which are difficult for classical computers, these devices are not yet capable of full-scale, fault-tolerant quantum computing. Nevertheless, recent breakthroughs in neutral atom, trapped ion, superconducting, and photonic quantum computing provide hope and excitement that we may see such a device in the near future. Table 1.1 summarizes and compares these quantum computing platforms.

Though efforts were made to ensure a comprehensive (yet somewhat brief) history of quantum computing, the work referenced in this section does not constitute an exhaustive list of all publications in the field; indeed, that would have been difficult to do! It is also worth noting that, by the time this thesis is published, there will likely be even more relevant work made available.

Solid-state quantum computers

Superconducting circuits [17, 199, 202, 224]

- *Pros*: Leading the race, commercially; high-precision control of qubits; fast gate operation; broad availability of control equipment
- *Cons*: Sensitive to noise, quick decoherence (sensitive to decoherence bursts); cryogenic operation; intricate wiring, which increases with number of qubits; low connectivity; anticipated need for optical interconnects; expensive

Semiconductor quantum dots (in silicon [225, 226, 227, 228] and germanium [229]) and color centers (*e.g.*, in silicon) [204]

- *Pros*: Compatibility with existing technology; potential for miniaturization; fast gate operation; can be used to design topologically protected qubits
 - *Cons*: Difficulty of controlling and reading out individual qubits; short coherence times; sensitive to environmental noise; prone to defects in fabrication (or difficult to engineer defects precisely); cryogenic temperatures (in most cases)
-
-

Atoms, ions, and molecules for quantum computing

Trapped ions [196, 230, 231]

- *Pros*: Fully connected network, long interaction ranges; one of the first technologies to demonstrate 2-qubit gates and quantum algorithms
- *Cons*: Cryogenic temperatures required; difficult to trap and control a large number of ions; sensitive to noise; bulky system requirements (in some cases)

Trapped atoms [6, 197, 198, 232, 233, 234]

- *Pros*: With optical tweezers, can place atoms as desired; possible to move atoms around for longer interaction distances; less sensitive to noise
 - *Cons*: Cryogenic temperatures required; short interaction range; bulky system requirements (in some cases)
-
-

Optical quantum computers [18, 19, 200, 206]

- *Pros*: Room-temperature operation; easy to integrate with communications infrastructure; naturally long coherence time
 - *Cons*: Generating single photons is a challenge; implementing logic gates can be complex and resource-intensive; can suffer from loss and noise; cryogenic temperatures required for most detectors
-
-

Table 1.1: **Summary of quantum computing platforms.** The current front-runners in quantum computing technology are summarized and compared.

1.3 Quantum computing with photons

Recent breakthroughs [17, 18, 19] have confirmed photonic systems as highly promising candidates for quantum information processing. Photonic platforms offer advantages such as the ability to operate at room temperature and exceptional resilience to decoherence, making them ideal for large-scale quantum computation. Photonic quantum processors would also be the most readily integrable with quantum communication platforms [235], a key requirement in the development of a quantum internet [236]. Photonic quantum information processing requires the ability to encode information in one (or several) of the many properties of photons. Information encoded in the polarization [237], frequency [238], spatiotemporal distribution [239], orbital angular momentum (OAM) [240, 241], path [18], and time of arrival [242] of photons has been demonstrated for a variety of quantum applications.

State-of-the-art photonic quantum processors are based on platforms which encode information in the path degree of freedom, using either free-space optics [18, 243] or integrated photonics [244, 245, 246, 247, 248]. Most free-space designs offer low loss but are unavoidably large, limiting their stability, scalability, and programmability. Integrated silicon-based platforms have seen universal programmability in up to an impressive 20 modes [200]; however, these systems often suffer from coupling and propagation losses, depending on the material used, potentially limiting their scalability. To demonstrate a clear quantum advantage, quantum information processing platforms must be scalable to a significantly larger number of modes (*i.e.*, system complexity) than has currently been demonstrated. An alternative platform based on time-bin encoding is emerging, with high scalability potential.

As its name suggests, time-bin encoding is a technique which encodes information in the time of arrival of a photon. Originally devised for quantum communication [249, 250, 251] due to its robustness against decoherence, there has since been a growing interest in this encoding method [252], in particular for quantum information processing [242, 253, 254, 255, 256]. The Canadian company, *Xanadu*, has recently demonstrated the potential for time-bin encoding in scaling quantum processors to achieve a quantum advantage [19]. To scale quantum processors, a critical requirement will be maintaining interferometric phase stability across the entire device for extended periods of time. This challenging requirement can be satisfied by a promising

new contender in the quantum computing landscape, which we have been developing in the *Ultrafast Quantum Photonics (UQP)* group, and has been the primary focus of my PhD research: ultrafast time-bin encoding.

1.4 Our all-optical approach

In ultrafast time-bin encoding (UTBE), information is encoded in the arrival time of a photon on ultrafast timescales. This scheme uses ultrashort pulses to prepare single photons with hundreds-of-femtoseconds (10^{-13} s) duration, encoded in time bins separated by several picoseconds (10^{-12} s). Far from the timescales of mechanical fluctuations, this operation regime offers improvements in system stability. Furthermore, time-bin states are prepared and manipulated in a common beam path, in the same spatial mode, a useful feature which supports scalability. Programmability is made possible by a series of all-optical phase shifters and mode couplers which can arbitrarily manipulate the quantum information to be processed (*i.e.*, the sequence of time bins). The building blocks that allow for this are passive birefringent crystals (α -barium borate) and active all-optical modulation (via the optical Kerr effect) in single-mode fiber. Birefringent crystals are relatively inexpensive and compact optical elements that, with the proper coating, provide minimal loss ($< 1\%$). Optical Kerr gating can modulate quantum optical signals with high speed and bandwidth, and $> 99\%$ efficiency [257, 258, 259]; it is also crucial for measuring the output of a quantum processor based on UTBE.

The development of ultrafast optical Kerr gating and UTBE has thus been the primary focus of the work presented in this thesis. Prior work in the *UQP* group at the *University of Ottawa* and *National Research Council (NRC) of Canada* laid the foundations for this work [257, 258]. Beyond their promise for quantum information processing, the optical Kerr gate and UTBE platform have also been demonstrated for other applications [259], including quantum communication [260, 261, 262].

1.5 Organization of Thesis

I hope that Chapter 1 has provided you with an enjoyable overview of quantum information processing, and has given you a feel for the current state of the field. I also hope that this has excited you to continue reading!

The remainder of this thesis will take you on a journey through the development of UTBE for quantum information processing. Chapter 2 presents a background on ultrafast and nonlinear optics, with a focus on the development and characterization of optical Kerr gating. This chapter also demonstrates implementations of the optical Kerr gate for ultrashort pulse generation [263] and frequency-selective gating [264].

Chapter 3 provides a background on quantum optics, focusing on the generation, detection, and characterization of single photon sources—all crucial capabilities for photonic quantum information processing. This chapter ends with a demonstration of the optical Kerr gate for characterization of single photons [265].

Key concepts of photonic quantum information processing are presented in Chapter 4, along with proof-of-concept demonstrations of photonic processors, based on UTBE, acting on a single photon. These demonstrations include a specific quantum computational algorithm, the quantum walk (QW) [266], as well as programmable photonic quantum circuits [267].

These proof-of-concept demonstrations are extended to the multiphoton regime in Chapter 5. The ability to operate in the multiphoton regime is a crucial capability for scalable photonic quantum information processing. This chapter presents background on multiphoton interference, which is then demonstrated for up to three photons [268].

This thesis ends with an outlook on UTBE for photonic quantum information processing in Chapter 6.

Note that some sections in this thesis are denoted with an asterisk (*). These sections offer additional background information that I consider useful; however, they can be skipped on a first reading without disrupting the overall flow of this thesis. Also note that supplemental documents are provided for some publications. These are included for completeness, but are not required reading.

Chapter 2

Some background on ultrafast and nonlinear optics

All-optical quantum information processing in the ultrafast regime requires the ability to generate, manipulate, and measure photons in ultrafast time bins. In order to achieve these capabilities, we can make use of ultrashort pulses of light and nonlinear all-optical gating. This chapter presents several important concepts in ultrafast and nonlinear optics which form the fundamental basis for the development of our all-optical approach to quantum information processing. Section [2.1](#) presents key concepts from ultrafast optics, while Section [2.2](#) presents key concepts from nonlinear optics. Sections [2.3](#) and [2.4](#) conclude this chapter with early demonstrations of the development and characterization of our ability to generate, manipulate, and measure light on ultrafast timescales.

2.1 Ultrafast optics

As its name suggests, the field of ultrafast optics deals with *ultrafast* processes and phenomena. At this point, you may be wondering what timescales the term ultrafast refers to. Generally, the term ultrafast is used to describe processes which occur on the picosecond timescale and below [269]:

$$\begin{aligned} 1 \text{ picosecond (ps)} &= 10^{-12} \text{ s} \\ 1 \text{ femtosecond (fs)} &= 10^{-15} \text{ s} \\ 1 \text{ attosecond (as)} &= 10^{-18} \text{ s} . \end{aligned}$$

To build an appreciation for just how fast these timescales are, it is useful to consider their spatial equivalent. Here, I will paraphrase a nice description from Andrew Weiner’s book, *Ultrafast optics* [269]: If it were possible to take a snapshot of a 1 s pulse of light, we would see this pulse stretched over a distance of 300,000 km (the distance light travels in one second); this distance is a little more than 3/4 of the distance between the Earth and moon (384,400 km). Skipping to the timescales relevant to most high-speed electronics, the spatial extent of a nanosecond pulse is 30 cm. Moving to *ultrafast* timescales, a picosecond has a spatial extent of 0.3 mm, about the thickness of a business card. When considering that most lasers have a beam diameter of a few millimeters, it may be better to visualize ultrashort¹ pulses as pancakes of light rather than pencils of light.

Ultrashort pulses of light have many advantages, including high timing resolution, high spatial resolution, high bandwidth, and the potential for high intensity operation. They have been applied in a wide range of applications, including ultrafast time-resolved spectroscopy [270, 271], high-precision metrology [272], machining and processing of materials [273], nonlinear microscopy [274], and optical communications [275]. This thesis extends the application space of ultrashort pulses to include quantum information processing in the ultrafast regime. The following subsections describe the generation, propagation, and manipulation of ultrashort pulses.

¹Note here the necessary distinction between *ultrafast* and *ultrashort*. While *ultrafast* is used to describe events occurring on picosecond timescales or shorter, *ultrashort* describes the temporal width of a pulse wave packet. Ultrashort pulses can be considered ultrafast events.

2.1.1 Generating ultrashort pulses

Ultrashort pulses intrinsically require a broad optical bandwidth. This statement, in itself, arises from decades of deep insight into Fourier analysis and related uncertainty principles. Fourier analysis dictates that any nonzero function and its Fourier transform cannot be simultaneously sharply localized [276]. Likewise, the variance of a function and of its Fourier transform cannot be simultaneously small, as described by Heisenberg's uncertainty principle [277] and the Gabor limit in signal processing [278]. This imposes a limit on the time-bandwidth product (TBP),

$$\Delta t \Delta f \geq \text{TBP}, \quad (2.1)$$

where Δt and Δf are the time and frequency full width at half maximum (FWHM) power (*i.e.*, squared) representations, respectively. For Gaussian pulses $\text{TBP} \approx 0.44$ and for sech^2 -pulses $\text{TBP} \approx 0.315$. Returning to the task at hand of generating ultrashort pulses: to generate a signal which is highly localized in the time domain, Δt , we need a broad optical bandwidth, Δf , which satisfies Eq. 2.1.

Ultrashort pulse generation thus requires a laser² which is capable of supporting multiple longitudinal modes. The bandwidth of operation for a given laser is determined primarily by its gain medium. A typical helium-neon (HeNe) laser has a gain bandwidth of around 1.5 GHz (a 0.002 nm range at a central wavelength of 633 nm), whereas a titanium-doped sapphire (Ti:Sapph) solid-state laser has a gain bandwidth of about 128 THz (a 300 nm range centered at 800 nm). Another important factor which determines the emission bandwidth³ of a laser is its optical cavity. Only standing modes are allowed to oscillate within the cavity, with frequencies

$$\nu_q = \frac{mc}{2L}, \quad (2.2)$$

where c is the speed of light, L is the length of the cavity, and m is an integer number; any other frequencies will be suppressed by destructive interference. In practice, the

²I will not go into details of basic laser operation in this thesis; for a nice resource on this topic, I recommend *Laser Physics* by Milonni and Eberly [279].

³Not to be confused with gain bandwidth. The emission bandwidth is the bandwidth of light generated (which is influenced by the gain bandwidth).

cavity length is usually much larger than the operating wavelength, $\lambda = c/\nu$, of a laser. This means that the relevant values of m are generally large (~ 100). It is perhaps more practical to consider the frequency separation between any two adjacent modes, m and $m + 1$, within the cavity,

$$\Delta\nu = \frac{c}{2L}. \quad (2.3)$$

The frequency separation can be compared to the gain bandwidth to determine the number of supported modes for a given laser, see Fig. 2.1. To put this into perspective, we can compare the number of allowed modes in a HeNe laser and Ti:Sapph laser. The mode frequency separation for a HeNe laser with $L \approx 30$ cm is about 0.5 GHz, so its 1.5 GHz gain bandwidth can support up to 3 longitudinal modes. On the other hand, the 128 THz gain bandwidth of a Ti:Sapph laser (assuming $L \approx 55$ cm) can support over 450,000 longitudinal modes!

We clearly see that not all lasers will be capable of ultrashort pulse generation; yet, even a laser that can support many longitudinal modes, such as Ti:Sapph, will not generate ultrashort pulses if each of these modes oscillates independently. If instead each mode has a fixed phase relation with the other modes, the modes can periodically all constructively interfere to produce an intense burst of light. This can be shown as follows; the electric field of a multimode laser, assuming operation in a single transverse mode, is

$$E(z, t) = \text{Re} \left\{ \sum_m E_m e^{i(k_m z - \omega_m t + \phi_m)} \right\}, \quad (2.4)$$

where E_m is the complex phase amplitude, ω_m is the angular frequency, k_m is the wave vector, and ϕ_m is the phase of mode m . We have that

$$\omega_m = \omega_0 + m\Delta\omega = \omega_0 + \frac{2m\pi c}{2L} \quad (2.5)$$

$$\text{and } k_m = \frac{\omega_m}{c}, \quad (2.6)$$

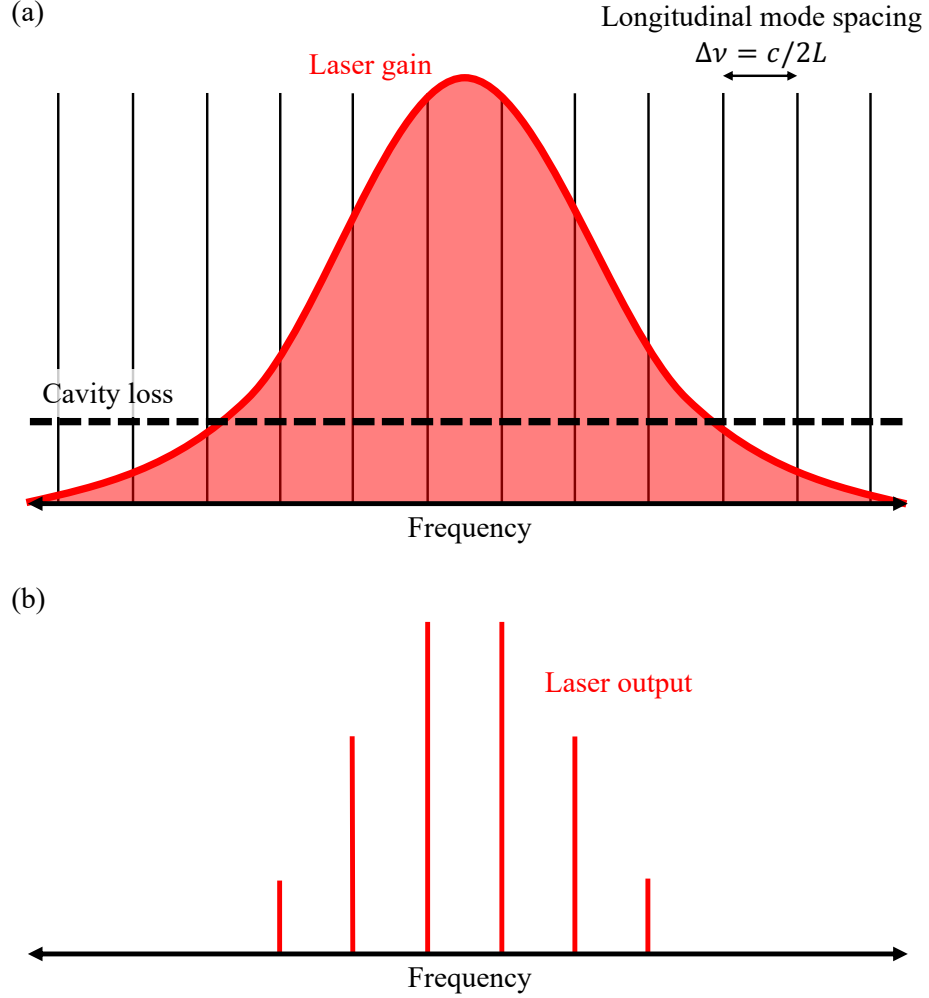


Figure 2.1: **Multimode laser operation.** The laser gain (shaded red curve), cavity loss (dashed black line), and longitudinal modes (black lines) are shown in (a); these quantities dictate the multimode laser output, shown in (b).

where ω_0 is the carrier frequency⁴, which gives

$$E(z, t) = \text{Re} \left\{ e^{i\omega_0(z/c-t)} \sum_m E_m e^{i[m\Delta\omega(z/c-t) + \phi_m]} \right\}. \quad (2.7)$$

Assuming that there are N oscillating modes with equal amplitude, E_0 , and a fixed

⁴It is worth noting that the carrier frequency is left arbitrary here. In many mode-locked lasers, the carrier frequency is allowed to drift; however, in some instances it must be stabilized.

phase of zero, we can then write

$$E(z, t) = \text{Re} \left\{ E_0 e^{i\omega_0(z/c-t)} \sum_{m=-(N-1)/2}^{(N-1)/2} e^{i[m\Delta\omega(z/c-t)]} \right\}. \quad (2.8)$$

If we substitute $m \rightarrow m + (N-1)/2$, we can then make use of the identity

$$\sum_{m=0}^{q-1} a^m = \frac{1-a^q}{1-a}, \quad (2.9)$$

and arrive at the result of

$$E(z, t) = \text{Re} \left\{ E_0 e^{i\omega_0(z/c-t)} \frac{\sin[(N\Delta\omega/2)(z/c-t)]}{\sin[(\Delta\omega/2)(z/c-t)]} \right\}, \quad (2.10)$$

which, for a given location in the cavity, say $z = 0$, the time-averaged intensity is

$$I(t) \sim |E_0|^2 \frac{\sin^2(N\Delta\omega t/2)}{\sin^2(\Delta\omega t/2)}. \quad (2.11)$$

We see that, if we can somehow manage to lock together the phases, ϕ_m , of N longitudinal modes in a multimode laser cavity, we will generate a train of pulses, as shown in Fig. 2.2. The output pulses are separated in time with a period equal to the time it takes light to make a round trip of the cavity, $T_{\text{rt}} = 2L/c$, corresponding to a repetition rate, $\Delta\nu = 1/T_{\text{rt}}$, equal to the mode spacing of the laser. The pulse duration is approximately $\tau = 2\pi/N\Delta\omega = 1/N\Delta\nu$, which is about equal to the inverse of the total laser bandwidth, and agrees with the TBP in Eq. 2.1. The peak intensity is proportional to $N^2|E_0|^2$, while the average intensity is proportional to $N|E_0|^2$. The process which generates such a pulse train is known as *mode locking*.

In practice, initiating and maintaining mode-locked laser operation is achieved through modulation, which can be active or passive. Active mode locking uses a modulator that is driven externally, while in passive mode locking, the pulse of light modulates itself through nonlinear processes. In both cases, the modulation will generate a periodic loss which corresponds to the cavity round-trip time. Mode locking can be conceptualized as a “survival of the fittest” phenomenon; laser oscillation will

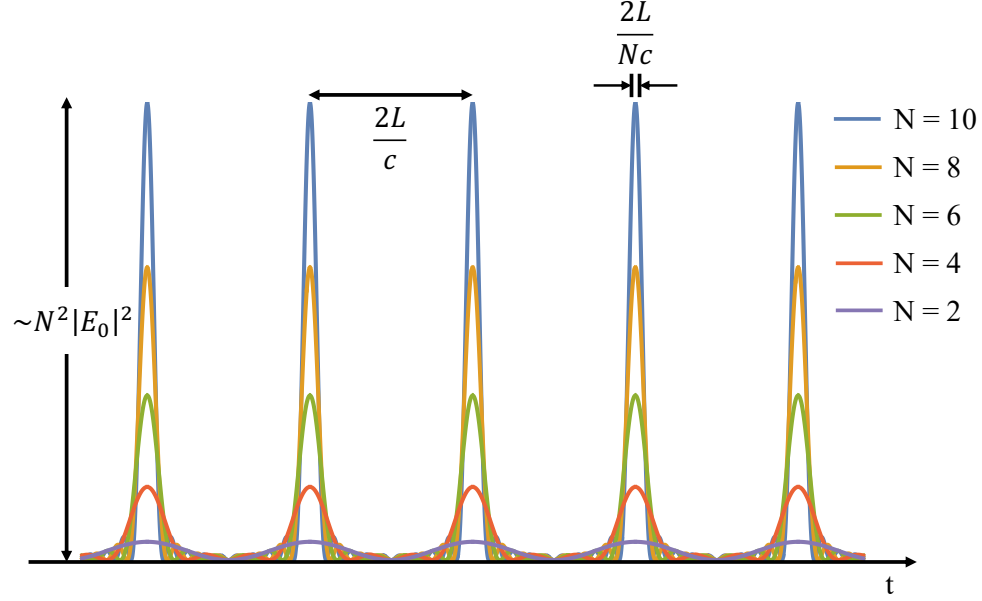


Figure 2.2: **Mode-locked laser output with constant mode phase.** Eq. 2.11 is plotted for various values of N . We see that, for increasing N , the pulse width decreases and the peak intensity increases.

only occur in the brief intervals of time where the periodically modulated loss is at a minimum. Between these time intervals, the loss will be too large for laser oscillation. Laser oscillation will thus only occur if it is possible to generate a train of short pulses separated by T_{rt} , which can only occur when that train of pulses is mode-locked.

In order to arrive at a stable mode-locked solution, we require that a pulse reproduces itself after one round trip in the laser cavity. We consider a single pulse (*i.e.*, not a pulse train), defined in time by the complex amplitude, $A(t)$, at a certain position within the cavity (*e.g.*, just before the output coupler). Note that the amplitude should be normalized such that taking the squared modulus is either the optical intensity or optical power in the gain medium. We then consider all possible changes to the pulse, $\Delta A(t)$, which can occur within a single round trip, including changes caused by laser gain, optical losses, chromatic dispersion, and optical nonlinearities. The consideration of these effects will depend on the mode locking method.

After n round trips through the cavity, the pulse amplitude can be written as $A(T, t)$, where $T = nT_{rt}$. Taking T as a continuous variable, our requirement for

stable, mode-locked operation is

$$T_{\text{rt}} \frac{\partial}{\partial T} A(T, t) = \Delta A(T, t). \quad (2.12)$$

In the case of an actively mode-locked laser with intra-cavity dispersion and a Kerr nonlinearity, this equation becomes

$$T_{\text{rt}} \frac{\partial}{\partial T} A(T, t) = \left[g + (D_g + iD_2) \frac{\partial^2}{\partial t^2} - l - \frac{M}{2} \omega_m^2 t^2 - i\delta |A|^2 \right] A(T, t), \quad (2.13)$$

where the terms on the right-hand side describe (from left to right) the saturated gain, the net gain filtering (*i.e.*, the frequency dependence of the gain) and second order dispersion, the cavity losses, the time-dependent active modulator losses (in the parabolic approximation for small time arguments), and the Kerr nonlinearity. Eq. 2.13 is referred to as the Haus master equation [280], and can be seen as a generalization of the nonlinear Schrödinger equation. Some of these terms may be neglected for other mode locking scenarios.

We see that care must be taken to ensure that pulse broadening effects are balanced by pulse compression effects. It is also important to recognize that the changes to the pulse profile per round trip should be small; this condition is satisfied in many mode-locked lasers, but is often problematic when considering few-cycle pulses. In the case of few-cycle pulse generation, changes to the pulse parameters during a single round trip are usually large enough that the ordering of optical components within the laser cavity is an important consideration. There are many different ways in which to achieve mode locking; the most common methods are outlined in Table 2.1.

2.1.2 Propagation of ultrashort pulses in media

At an early point in our physics education, we are taught about the concept of light “slowing down” when it travels, for example, in water compared to in air. This effect is described by the refractive index, n , of an optical medium, which is often defined as the ratio between the speed of light in vacuum, c , and the phase velocity

Active mode locking

Electro-optic modulator

- When driven with an electrical signal, this produces a sinusoidal amplitude modulation of light in the cavity. This modulator acts as a weak “shutter”, synchronized to the cavity round-trip time. The strength of the modulation need not be large, since the same part of the light is repeatedly attenuated as it traverses the cavity.

Acousto-optic modulator

- When driven with an electrical signal, this induces a small, sinusoidally varying frequency shift in the light passing through it. If the frequency of modulation matches the cavity round-trip time, some light will experience repeated frequency upshifts and some light will experience repeated frequency downshifts. After many round trips, the upshifted and downshifted light is swept out of the gain bandwidth of the laser, and only the unaffected light (with zero frequency shift) remains.
-
-

Passive mode locking

Saturable absorber

- This device exhibits an intensity-dependent transmission; it will selectively absorb low-intensity light, but transmit high-intensity light. This process repeats as the light traverses the cavity many times, leading to selective amplification of the high-intensity light, and the formation of a pulse train.

Kerr-lens

- This makes use of a nonlinear optical process (the optical Kerr effect) which results in high-intensity light being focused more tightly than low-intensity light. Careful placement of an aperture in the laser cavity can then be used to select only the high-intensity, more focused light, leading to an ultrashort pulse train.
-
-

Table 2.1: **Mode locking methods.** The most common mode locking methods are presented and briefly explained.

of light within the medium, $n = c/v_p$ ⁵. In most cases, the index of refraction, and thus also the phase velocity of light, will depend on its frequency; this phenomenon is referred to as dispersion.

⁵The physics which describes this is nicely detailed in Volume I, Chapter 31 of the *Feynman Lectures on Physics* [281]. This reading material was recommended to me, in my first year of PhD studies, by Ben; I enjoyed it, and so I also pass along the recommendation here.

The phase velocity,

$$v_p = \frac{c}{n}, \quad (2.14)$$

describes only one frequency component, often referred to as the carrier frequency of some signal. Yet, in many cases—particularly in communications—it is not the carrier frequency which transmits information, but rather the modulation of the carrier frequency (*e.g.*, the envelope of an ultrashort optical pulse⁶). The velocity at which a pulse propagates in an optical medium is described by its group velocity,

$$v_g = \frac{c}{n + \omega \frac{dn}{d\omega}} = \frac{c}{n - \lambda \frac{dn}{d\lambda}}. \quad (2.15)$$

We see that, when an optical signal consisting of more than one wavelength travels through a dispersive medium for which the index of refraction is frequency-dependent (a fairly standard case), the group velocity will be different than the phase velocity. Furthermore, we see that the group velocity itself will vary with frequency; this phenomenon is known as group-velocity dispersion (GVD) and can cause an ultrashort pulse to be temporally broadened.

GVD is an important consideration for the work presented in this thesis, which exploits effects that occur when ultrashort pulses are transmitted through optical fibers. As such, we take the time here to more formally describe GVD. For an input pulse, $E_{\text{in}}(t)$, with spectrum, $\tilde{E}_{\text{in}}(\omega)$, passing through a dispersive medium which imposes a spectral phase, $\phi(\omega)$, the output pulse can be written as^{7,8,9}

$$E_{\text{out}}(t) = \text{Re} \left\{ \frac{1}{\sqrt{2\pi}} \int d\omega \tilde{A}_{\text{in}}(\omega - \omega_0) e^{-i\omega t} e^{i\phi(\omega - \omega_0)} \right\}. \quad (2.16)$$

⁶In some cases, the group velocity of a pulse can be superluminal (larger than the speed of light in vacuum, c) [282], and the group velocity cannot be interpreted as a velocity of information transfer. The information velocity will actually always be equal to c —even if the refractive index and group index are larger than one [283].

⁷The properties of a linear time invariant system allow an output signal in time to be described as the weighted integral over the input signal (*i.e.*, a convolution between the input signal and the transfer function of the system). This can be simplified by operating in Fourier space, where a convolution in the time domain is equivalent to a multiplication in the frequency domain.

⁸The output pulse, $E_{\text{out}}(t)$ can be obtained directly by either taking the inverse Fourier transform of the double-sided spectrum, $\tilde{E}_{\text{in}}(\omega)$, or from the real part of the single-sided spectrum, as we use here. See Section 1.5 of *Ultrafast Optics* for useful analytical aspects of Fourier transforms [269].

⁹Unfortunately, there are many conventions used for taking the forward and backward Fourier transforms, which can get confusing. Here we use the convention common in modern physics [284].

For propagation through a dispersive medium of length, L , we can write

$$\phi(\omega) = \beta(\omega)L, \quad (2.17)$$

where

$$\beta(\omega) = \frac{\omega n(\omega)}{c}, \quad (2.18)$$

is the propagation constant of the medium. In general, $n(\omega)$, and thus also $\beta(\omega)$, are arbitrary functions of ω . This can make it difficult to find an analytical solution for $E_{\text{out}}(t)$; however, if the pulse bandwidth is narrow relative to the curvature of $n(\omega)$, then a good approximation can be made with a Taylor series expansion of $\beta(\omega)$, centered about the carrier frequency, ω_0 ,

$$\begin{aligned} \beta(\omega) &\approx \beta(\omega_0) + \beta'(\omega_0)(\omega - \omega_0) + \frac{1}{2}\beta''(\omega_0)(\omega - \omega_0)^2 + \frac{1}{6}\beta'''(\omega_0)(\omega - \omega_0)^3 + \dots \\ &\approx \beta_0 + \beta_1(\omega - \omega_0) + \frac{\beta_2}{2}(\omega - \omega_0)^2 + \frac{\beta_3}{6}(\omega - \omega_0)^3 + \dots \end{aligned} \quad (2.19)$$

where we denote $\beta_n = \frac{\partial^n \beta}{\partial \omega^n} \big|_{\omega_0}$. Substituting Eq. 2.19 into Eq. 2.16, and redefining $(\omega - \omega_0) \rightarrow \omega$, we can write the output pulse as

$$E_{\text{out}}(t) = \text{Re} \left\{ \frac{e^{i(\beta_0 L - \omega_0 t)}}{\sqrt{2\pi}} \int d\omega \tilde{A}_{\text{in}}(\omega) e^{i([\beta_1 \omega + \frac{\beta_2}{2}\omega^2 + \frac{\beta_3}{6}\omega^3 + \dots]L - \omega t)} \right\}. \quad (2.20)$$

We see that $E_{\text{out}}(t)$ is the product of a carrier term, $e^{i(\beta_0 L - \omega_0 t)}$, (which propagates at the phase velocity, $v_p = \omega_0/\beta_0$, and is not affected by the variation of β with ω) and an envelope function,

$$A_{\text{out}}(t) = \frac{1}{\sqrt{2\pi}} \int d\omega \tilde{A}_{\text{in}}(\omega) e^{i([\beta_1 \omega + \frac{\beta_2}{2}\omega^2 + \frac{\beta_3}{6}\omega^3 + \dots]L - \omega t)}, \quad (2.21)$$

which is evidently affected by the form of $\beta(\omega)$. To build up an intuition of how each of the terms in $\beta(\omega)$ will influence the propagating pulse, we first consider the simple

case where $\beta(\omega)$ is a linear function of ω ,

$$\beta(\omega) = \beta_0 + \beta_1\omega. \quad (2.22)$$

Taking the Fourier transform, we arrive at

$$A_{\text{out}}(t) = A_{\text{in}}(t + \beta_1 L), \quad (2.23)$$

where we see that the output pulse is an undistorted replica of the input pulse, delayed by $\tau = \beta_1 L$. This is consistent with the Fourier transform identity stating that the Fourier transform of a delayed pulse, $A(t + \tau)$, is given by $\tilde{A}(\omega)e^{i\omega\tau}$. The delay imparted by β_1 is related to the group velocity,

$$v_g = \frac{1}{\beta_1} = \left(\frac{\partial \beta}{\partial \omega} \bigg|_{\omega_0} \right)^{-1}. \quad (2.24)$$

where Eq. 2.24 is consistent with our previous definition of v_g , Eq. 2.15.

When $\beta(\omega)$ is not a linear function of ω , the higher-order terms alter $A_{\text{out}}(t)$ relative to $A_{\text{in}}(t)$. The β_2 term contributes a quadratic spectral phase variation, which imparts a chirp to the output pulse (*i.e.*, a linear variation in delay with frequency). This term leads to GVD, and it is often acceptable to truncate the expansion of $\beta(\omega)$ here; however, when dealing with very broad optical spectra (*e.g.*, in sub-10 fs laser design and chirped pulse amplification), higher-order terms (β_3 and beyond) must be retained. These higher-order terms contribute additional distortions to the overall pulse shape; for example, the β_3 term contributes a cubic spectral phase variation, resulting in an asymmetric distortion with an oscillatory structure. To understand the work presented in this thesis, we only require knowledge of up to β_2 , so the rest of this section will focus on the effects of quadratic dispersion.

For a given frequency component, ω , the time it will take to propagate through a length, L , of a dispersive medium is $\tau(\omega) = L/v_g(\omega)$. It is useful to think of this

propagation time relative to that of frequency ω_0 ,

$$\begin{aligned}\tau(\omega) - \tau(\omega_0) &= \Delta\tau(\omega) = \frac{\partial\tau}{\partial\omega}(\omega - \omega_0) \\ &= \frac{\partial(v_g^{-1})}{\partial\omega}(\omega - \omega_0)L = \beta_2(\omega - \omega_0)L.\end{aligned}\quad (2.25)$$

We see that we can relate this propagation time difference to the quadratic phase term in our expansion of $\beta(\omega)$. It is often more convenient to work with wavelength, λ , as opposed to angular frequency, ω , which are related by

$$\lambda = \frac{2\pi c}{\omega}, \quad (2.26)$$

allowing us to write $\Delta\tau$ in terms of the wavelength offset, $\Delta\lambda$,

$$\Delta\tau(\lambda) = \frac{\partial(v_g^{-1})}{\partial\lambda}(\lambda - \lambda_0)L = \frac{-2\pi c\beta_2\Delta\lambda L}{\lambda^2}. \quad (2.27)$$

In fiber optics, GVD is commonly quantified by the dispersion parameter,

$$D = \frac{\partial(v_g^{-1})}{\partial\lambda} = \frac{-2\pi c\beta_2}{\lambda^2}, \quad (2.28)$$

which is customarily given in units of $[\text{ps}][\text{nm}]^{-1}[\text{km}]^{-1}$.

For a Gaussian pulse, it is straightforward to express the broadening due solely to second-order spectral phase. We consider an unchirped input pulse with a pulse-width parameter of t_p ,

$$A_{\text{in}}(t) = A_0 \exp(-t^2/t_p^2), \quad (2.29)$$

which, in the frequency domain is given by

$$\tilde{A}_{\text{in}}(\omega) = A_0 \sqrt{t_p^2/2} \exp(-t_p^2 \omega^2/4). \quad (2.30)$$

After propagation through a length L of dispersive medium with dispersion β_2 ¹⁰, this

¹⁰Here, β_2 is assumed to take on a positive value, and so the remainder of this description falls within the normal dispersion regime.

pulse has an envelope of

$$A_{\text{out}}(t) = \frac{A_0}{\sqrt{1 + 2iL\beta_2/t_p^2}} \exp\left(-\frac{t^2/t_p}{1 + 2iL\beta_2/t_p^2}\right). \quad (2.31)$$

The intensity profiles of the input and output pulse envelope are, respectively,

$$|A_{\text{in}}(t)|^2 = |A_0|^2 \exp(-2t^2/t_p^2) \quad (2.32)$$

and

$$|A_{\text{out}}(t)|^2 = \frac{|A_0|^2}{\sqrt{1 + (2L\beta_2/t_p^2)^2}} \exp\left(-\frac{2t^2/t_p^2}{1 + (2L\beta_2/t_p^2)^2}\right), \quad (2.33)$$

and we see that the pulse is lengthened to

$$t_{p,\text{out}} = \sqrt{t_{p,\text{in}}^2 + (2L\beta_2/t_{p,\text{in}})^2}. \quad (2.34)$$

Until this point, we have intentionally left vague the definition of the pulse-width parameter, t_p ; however, here we note that we can relate t_p to two common definitions of pulse width, either the intensity standard deviation, $\sigma = \sqrt{2}t_p$, or the intensity FWHM, $t_{\text{FWHM}} = 2\sqrt{\ln 2}t_p$ ¹¹. It is worth noting that material dispersion can be either normal or anomalous, leading to pulse lengthening or compression, respectively.

2.1.3 Manipulating ultrashort pulses

In many situations it is desirable to have control over the properties of an ultrashort pulse (or sequences of ultrashort pulses). As discussed in Section 2.1.2, dispersive media can be used to stretch or compress ultrashort pulses which is useful in mode-locked pulse generation, chirped pulse amplification¹², and ultrashort pulse characterization [286]. Pulse shaping can also be used to carefully control chemical

¹¹I reiterate here that σ and t_{FWHM} are two ways of defining the **intensity** envelope of the pulse, and should be adjusted accordingly when working with the field envelope.

¹²The 2018 Nobel Prize in Physics was awarded to Donna Strickland and Gérard Mourou, for the practical implementation of chirped pulse amplification [285].

reactions [287], or to produce a train of pulses for high-speed optical communications [288].

Pulse shapers are effectively modulators, defined by some modulating function in the time (direct pulse shaping) or frequency (Fourier transform pulse shaping) domain. The speed of direct pulse shaping is limited to the speed of its driving electronics, which can be problematic for ultrashort pulse shaping. Current approaches circumvent this issue by using dispersive pulse shaping, for example with chirped mirrors, fiber Bragg gratings, liquid crystal modulators, or acousto-optic modulators. Another option is to exploit the timescales of ultrashort pulse sources themselves, which this thesis presents. For this, nonlinear optical interactions are required, as discussed in more detail in Section 2.2.

2.2 Nonlinear optics

When working in an experimental optics lab, it is of practical interest to understand the physics describing light in matter. To simplify things, we will confine our discussion here to light-matter interactions in which the magnetic field strength is much weaker than the electric field strength, and consider only electric fields in matter¹³. Of course, matter comes in many varieties, but we will restrict our discussion to the most relevant here: dielectric material. Dielectrics are electrical insulators. When placed in an external electric field, the electrons within a dielectric are not free to flow through the material; rather, their average position within the atom shifts, very slightly but enough to induce a dipole moment between the electron and atomic nucleus¹⁴. Although these shifts are small for a single atom, the cumulative effects within the dielectric as a whole cause it to become polarized, with a polarization

¹³This is generally a good assumption—the force exerted by the electric field on a charged particle is $e\mathbf{E}$, while the force exerted by the magnetic field is $e\mathbf{v} \times \mathbf{B}$. The magnetic force will only be comparable to the electric force when the velocity of the particle becomes relativistic. Furthermore, most materials used in optics experiments are not magnetic, and so they have a magnetic susceptibility of zero.

¹⁴If the field is **very** large, then it can pull the electron away from the atomic nucleus completely, “ionizing” it. We are not working in this regime here.

density (dipole moment per unit volume) of

$$\mathbf{P}(t) = \epsilon_0 \chi^{(1)} \mathbf{E}(t), \quad (2.35)$$

where ϵ_0 is the electric permittivity of free space and $\chi^{(1)}$ is the linear electric susceptibility of the dielectric, which measures how easily it can be polarized. We see that an oscillating electric field, $\mathbf{E}(t)$, leads to an oscillating material polarization, $\mathbf{P}(t)$.

The induced polarization is approximately proportional to the incident electric field, provided the electric field is not too strong. This is indeed the case for conventional (linear) optics; however, with access to lasers (and in particular, ultrafast lasers which can have very high peak field intensities) the incident electric field can modify the optical properties of the material system through which it travels, and the polarization will respond nonlinearly to the incident electric field. The field of nonlinear optics studies phenomena that arise from this effect¹⁵. Such phenomena are crucial to the work in this thesis—from measuring ultrafast time-bin signals to providing sources of photons.

In nonlinear optics, the optical response of a material can be described as a generalization of Eq. 2.35, where the polarization is expressed as a power-series expansion of the electric field strength,

$$\mathbf{P}(t) = \epsilon_0 [\chi^{(1)} \mathbf{E}(t) + \chi^{(2)} \mathbf{E}^2(t) + \chi^{(3)} \mathbf{E}^3(t) + \dots]. \quad (2.36)$$

For very small $\mathbf{E}(t)$, we see that Eq. 2.36 is well-approximated by Eq. 2.35. For large enough $\mathbf{E}(t)$, we must consider the higher-order terms, where $\chi^{(2)}$, and $\chi^{(3)}$ refer to the second- and third-order nonlinear optical susceptibilities, respectively. In general, $\mathbf{E}(t)$ and $\mathbf{P}(t)$ are vector quantities¹⁶, and the susceptibilities, $\chi^{(n)}$, are tensors of rank $n + 1$. Note that tensor indices are suppressed throughout this thesis for simplicity.

¹⁵I refer you to Robert Boyd’s book *Nonlinear Optics* [289], which is a fantastic resource for learning about this field.

¹⁶Vector quantities are denoted here by bold font, where the functional dependence on $\mathbf{r}$ is suppressed for simplicity.

We simplify our discussion here by considering only scalar fields, such that that

$$\begin{aligned} P(t) &= \epsilon_0 [\chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + \dots] \\ &= P^{(1)}(t) + P^{(2)}(t) + P^{(3)}(t) + \dots, \end{aligned} \quad (2.37)$$

where we have assumed that the instantaneous polarization at time t depends only on the instantaneous field strength (that is, we assume an instantaneous material response time). The nonlinear susceptibilities generally depend on the frequencies of the applied fields, but we can take them as constants under our current assumptions.

To simplify our discussion, we will refer to $P^{(1)}(t) = \epsilon_0 \chi^{(1)} E(t)$ as the linear polarization, $P^{(2)}(t) = \epsilon_0 \chi^{(2)} E^2(t)$ as the second-order nonlinear polarization, $P^{(3)}(t) = \epsilon_0 \chi^{(3)} E^3(t)$ as the third-order nonlinear polarization, and so on. In some cases, it is convenient to group the nonlinear polarization terms into one and write

$$P(t) = P^{(1)}(t) + P^{\text{NL}}(t), \quad (2.38)$$

where $P^{\text{NL}}(t) = P^{(2)}(t) + P^{(3)}(t) + \dots$. We will see in the coming sections how the nonlinear polarization components can generate new fields with different frequency components. It should be noted that the power-series expansion of the electric field in Eqs. 2.35 through 2.38 need not necessarily converge—this is indeed the case if the applied laser field strength becomes comparable to the atomic field strength (*i.e.*, for laser intensities around 10^{16} W/cm^2), where strong photoionization is possible. The work presented in this thesis does not enter this regime (referred to as “intense-field nonlinear optics”); as such, it is not discussed further.

2.2.1 Nonlinear propagation of electric fields in media

We will show in this section how nonlinearities induced in the response of a material to intense laser fields can lead to the generation of new frequencies of light. We

start with Maxwell's equations,

$$\nabla \cdot \mathbf{D}(t) = \rho(t) \quad (2.39)$$

$$\nabla \cdot \mathbf{B}(t) = 0 \quad (2.40)$$

$$\nabla \times \mathbf{E}(t) = -\frac{\partial \mathbf{B}(t)}{\partial t} \quad (2.41)$$

$$\nabla \times \mathbf{H}(t) = \frac{\partial \mathbf{D}(t)}{\partial t} + \mathbf{J}(t), \quad (2.42)$$

where $\mathbf{D}(t) = \epsilon_0 \mathbf{E}(t) + \mathbf{P}(t)$ is the electric displacement field, $\mathbf{H}(t) = \frac{1}{\mu_0} \mathbf{B}(t) - \mathbf{M}(t)$ is the magnetic auxiliary field (where $\mathbf{B}(t)$ is the magnetic field, μ_0 is the permeability of free space, and $\mathbf{M}(t)$ is the magnetization), $\rho(t)$ is the free charge density, and $\mathbf{J}(t)$ is the free current. All nonlinear processes we consider in this thesis occur in materials that do not have free charges, $\rho(t) = 0$, are not surrounded by wires that apply free currents, $\mathbf{J}(t) = 0$, and are non-magnetic, $\mathbf{M}(t) = 0$. We can take the curl of Eq. 2.41, and make use of 2.42, to show that

$$\begin{aligned} \nabla \times \nabla \times \mathbf{E}(t) &= \nabla \times -\frac{\partial \mathbf{B}(t)}{\partial t} \\ &= -\frac{\partial}{\partial t} (\nabla \times \mathbf{B}(t)) \\ &= -\frac{\partial}{\partial t} \left(\mu_0 \frac{\partial \mathbf{D}(t)}{\partial t} \right) \\ &= -\mu_0 \frac{\partial^2}{\partial t^2} (\epsilon_0 \mathbf{E}(t) + \mathbf{P}(t)). \end{aligned} \quad (2.43)$$

After some rearrangement, we arrive at the wave equation,

$$\nabla \times \nabla \times \mathbf{E}(t) + \frac{1}{c^2} \frac{\partial^2 \mathbf{E}(t)}{\partial t^2} = -\frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{P}(t)}{\partial t^2}, \quad (2.44)$$

where we have made use of the fact that $c^2 = \frac{1}{\mu_0 \epsilon_0}$. Using the vector calculus identity,

$$\nabla \times \nabla \times \mathbf{A} = \nabla (\nabla \cdot \mathbf{A}) - \nabla^2 \mathbf{A}, \quad (2.45)$$

we can re-write Eq. 2.44 as

$$\nabla (\nabla \cdot \mathbf{E}(t)) - \nabla^2 \mathbf{E}(t) + \frac{1}{c^2} \frac{\partial^2 \mathbf{E}(t)}{\partial t^2} = -\frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{P}(t)}{\partial t^2}. \quad (2.46)$$

The term $\nabla (\nabla \cdot \mathbf{E}(t))$ vanishes in the linear regime because $\nabla \cdot \mathbf{D}(t) = 0$ implies $\nabla \cdot \mathbf{E}(t) = 0$, but it need not necessarily vanish in the nonlinear regime, even for isotropic materials; however, we can still usually drop this term in nonlinear optics. For example, if $\mathbf{E}(t)$ is a transverse, infinite plane wave this term vanishes. Alternatively, in the slowly varying amplitude approximation,

$$\left| \frac{d^2 \mathbf{E}(t)}{dt^2} \right| \ll \left| \frac{d\mathbf{E}(t)}{dt} \right|, \quad (2.47)$$

this term will be very small and can be dropped. For the work presented in this thesis, we can assume that the contribution from $\nabla (\nabla \cdot \mathbf{E}(t))$ is negligible, and the wave equation is

$$\nabla^2 \mathbf{E}(t) - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}(t)}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{P}(t)}{\partial t^2}. \quad (2.48)$$

We can split the polarization field into its linear and nonlinear components, $\mathbf{P}(t) = \mathbf{P}^{(1)}(t) + \mathbf{P}^{\text{NL}}(t)$, and refer to the linear component of the electric displacement field as $\mathbf{D}^{(1)}(t) = \epsilon_0 \mathbf{E}(t) + \mathbf{P}^{(1)}(t)$ in order to write the wave equation as

$$\nabla^2 \mathbf{E}(t) - \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{D}^{(1)}(t)}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{P}^{\text{NL}}(t)}{\partial t^2}, \quad (2.49)$$

which is often a more convenient form to work with. For a lossless, dispersionless material, $\mathbf{D}^{(1)}(t) = \epsilon_0 \epsilon^{(1)} \mathbf{E}(t)$, where $\epsilon^{(1)}$ is a real, frequency-independent dielectric tensor. If the material is isotropic, $\epsilon^{(1)}$ is a scalar quantity and can be written as $\epsilon^{(1)}$. The wave equation is then

$$\nabla^2 \mathbf{E}(t) - \frac{\epsilon^{(1)}}{c^2} \frac{\partial^2 \mathbf{E}(t)}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{P}^{\text{NL}}(t)}{\partial t^2}, \quad (2.50)$$

which takes the form of a driven wave equation, where the nonlinear polarization acts as a source term capable of generating new fields with different frequencies.

In the absence of this source term, these equations describe free waves propagating with velocity c/n (where n is the linear index of refraction). Here, we have assumed that the electric field, $\mathbf{E}(t)$, is monochromatic; however, for the case of a dispersive medium, we must consider separately each frequency component of $\mathbf{E}(t)$. In this case, the electric, linear displacement, and polarization fields can be written as sums of their various frequency components,

$$\mathbf{E}(\mathbf{r}, t) = \sum_n \mathbf{E}_n(\mathbf{r}, t) \quad (2.51)$$

$$\mathbf{D}^{(1)}(\mathbf{r}, t) = \sum_n \mathbf{D}_n^{(1)}(\mathbf{r}, t) \quad (2.52)$$

$$\mathbf{P}^{\text{NL}}(\mathbf{r}, t) = \sum_n \mathbf{P}_n^{\text{NL}}(\mathbf{r}, t), \quad (2.53)$$

where the summations should be performed over only positive field frequencies and

$$\mathbf{E}_n(\mathbf{r}, t) = \mathbf{E}_n(\mathbf{r})e^{-i\omega_n t} + \text{c.c.} \quad (2.54)$$

$$\mathbf{D}_n^{(1)}(\mathbf{r}, t) = \mathbf{D}_n^{(1)}(\mathbf{r})e^{-i\omega_n t} + \text{c.c.} \quad (2.55)$$

$$\mathbf{P}_n^{\text{NL}}(\mathbf{r}, t) = \mathbf{P}_n^{\text{NL}}(\mathbf{r})e^{-i\omega_n t} + \text{c.c.}, \quad (2.56)$$

where c.c. stands for complex conjugate. Neglecting dissipation, a driven wave equation can be written for each frequency component,

$$\nabla^2 \mathbf{E}_n(t) - \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{D}_n^{(1)}(t)}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{P}_n^{\text{NL}}(t)}{\partial t^2}. \quad (2.57)$$

In the following sections, we consider nonlinear optical processes that arise from the source term, $\mathbf{P}^{\text{NL}}(t)$.

2.2.2 Second-order nonlinear optical processes

We first consider the $\chi^{(2)}$ term in our power-series expansion of the polarization in Eqs. 2.36 to 2.38, which is responsible for second-order nonlinear optical processes. Second-order optical nonlinearity is the lowest order of nonlinear processes, and leads to nonlinear optical effects including second-harmonic generation (SHG),

sum-frequency generation (SFG), and difference-frequency generation (DFG), the last of which is crucial in the functionality of optical parametric oscillators (OPOs) and amplifiers (OPAs). It should be noted that second-order nonlinear processes can only occur in non-centrosymmetric media (*i.e.*, crystals lacking a center of inversion). This will be demonstrated in the context of SHG.

Second-harmonic generation (SHG)

The field of nonlinear optics is generally thought to have begun with the first demonstration of SHG [290], one year after the first working demonstration of the laser by Maiman [291]. SHG is therefore a nice first example for us to consider, and is depicted schematically in Fig. 2.3(a).

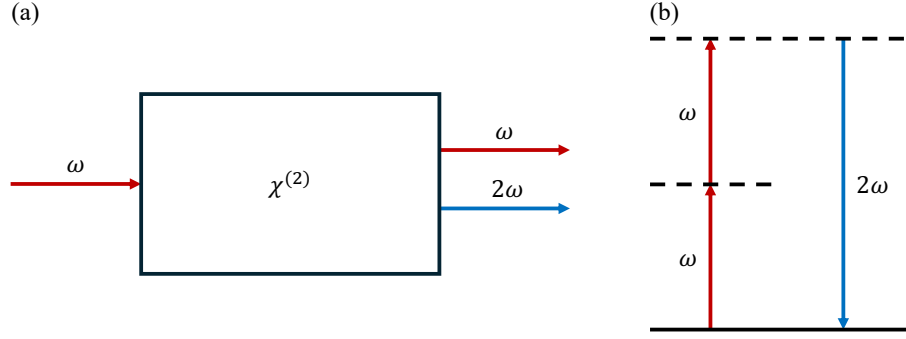


Figure 2.3: **Depiction of second-harmonic generation (SHG).** A schematic diagram of the fields involved in SHG in a $\chi^{(2)}$ crystal is shown in (a), while the corresponding energy-level diagram describing SHG is shown in (b).

A laser beam, with frequency ω , is incident upon a nonlinear optical crystal with a nonzero second-order susceptibility, $\chi^{(2)}$. The electric field of the laser is given by

$$E(t) = E_0 e^{-i\omega t} + \text{c.c.}, \quad (2.58)$$

such that the second-order nonlinear polarization will then be expressed as

$$\begin{aligned} P^{(2)}(t) &= \epsilon_0 \chi^{(2)} E^2(t) \\ &= 2\epsilon_0 \chi^{(2)} E_0 E_0^* + (\epsilon_0 \chi^{(2)} E_0^2 e^{-i2\omega t} + \text{c.c.}), \end{aligned} \quad (2.59)$$

with contributions from a zero frequency term (often referred to as the DC term) and a frequency-doubled (2ω) term. In accordance with the driven wave equation in Eq. 2.50, the frequency-doubled contribution can lead to the generation of new radiation at a frequency of 2ω (the second-harmonic). As shown in Fig. 2.3(b), SHG can be considered in terms of the exchange of photons between the frequency modes involved, where two photons at frequency ω are destroyed while one photon at frequency 2ω is created simultaneously.

Under the right conditions, SHG can be so efficient that almost all of the power in the input beam, at frequency ω , is converted to radiation at frequency 2ω . In the lab, we use SHG to convert 1064nm to 532nm in order to pump our mode-locked Ti:Sapph lasers! The DC term, on the other hand, does not lead to the generation of new electromagnetic radiation; rather, it leads to a static electric field across the nonlinear crystal, a process which is called optical rectification (OR).

Earlier we said that we would demonstrate how $\chi^{(2)}$ effects can only occur in non-centrosymmetric media using SHG as an example. We do so here by considering the second-order nonlinear polarization,

$$P^{(2)}(t) = \epsilon_0 \chi^{(2)} E^2(t), \quad (2.60)$$

where the input electric field is given by

$$E(t) = E_0 \cos(\omega t). \quad (2.61)$$

For a medium which possesses inversion symmetry, changing the sign of the electric field must also cause the sign of the induced polarization to change. This implies that

$$-P^{(2)}(t) = \epsilon_0 \chi^{(2)} [-E(t)]^2, \quad (2.62)$$

which simplifies to

$$-P^{(2)}(t) = \epsilon_0 \chi^{(2)} E^2(t). \quad (2.63)$$

This result implies that $-P^{(2)}(t)$ must be equivalent to $P^{(2)}(t)$, which can only be the case if $P^{(2)}(t) = 0$ and implies that $\chi^{(2)} = 0$ for centrosymmetric media.

A more general treatment of the second-order nonlinear polarization

A broader description of second-order nonlinear processes takes an input optical field consisting of two distinct frequency components,

$$E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + \text{c.c.}, \quad (2.64)$$

which leads to a second-order nonlinear polarization of

$$\begin{aligned} P^{(2)}(t) = \epsilon_0 \chi^{(2)} \big[& E_1^2 e^{-i2\omega_1 t} + E_2^2 e^{-i2\omega_2 t} + 2E_1 E_2 e^{-i(\omega_1 + \omega_2)t} \\ & + 2E_1 E_2^* e^{-i(\omega_1 - \omega_2)t} + \text{c.c.} \big] + 2\epsilon_0 \chi^{(2)} \big[E_1 E_1^* + E_2 E_2^* \big]. \end{aligned} \quad (2.65)$$

It is convenient to write this as a sum of polarization components over all frequencies involved (extending over both positive and negative frequencies),

$$P^{(2)}(t) = \sum_n P(\omega_n) e^{-i\omega_n t}, \quad (2.66)$$

so that the complex amplitudes of the nonlinear polarization can be written to more clearly indicate which second-order nonlinear process they lead to,

$$\begin{aligned} P(2\omega_1) &= \epsilon_0 \chi^{(2)} E_1^2 && \text{SHG from } \omega_1, \\ P(2\omega_2) &= \epsilon_0 \chi^{(2)} E_2^2 && \text{SHG from } \omega_2, \\ P(\omega_1 + \omega_2) &= 2\epsilon_0 \chi^{(2)} E_1 E_2 && \text{SFG}, \\ P(\omega_1 - \omega_2) &= 2\epsilon_0 \chi^{(2)} E_1 E_2^* && \text{DFG}, \\ P(0) &= 2\epsilon_0 \chi^{(2)} (E_1 E_1^* + E_2 E_2^*) && \text{OR.} \end{aligned} \quad (2.67)$$

We label each expression above by the physical process it describes: second-harmonic generation (SHG), sum-frequency generation (SFG), difference-frequency generation (DFG), and optical rectification (OR)¹⁷. We note, for completeness, that the complex conjugate will lead to contributions to the polarization responses at the negatives of each of the frequencies above. It becomes clear, from this general treatment of $P^{(2)}(t)$,

¹⁷We will not discuss OR effects in this thesis, but it should be noted that OR is also a useful process. For example, it can be used to generate terahertz radiation [292].

that there are four different nonzero frequency components present in the second-order nonlinear polarization; however, usually only one of these components will have appreciable intensity in the generated radiation. This occurs because the nonlinear polarization can only efficiently produce an output signal if a specific phase-matching condition is satisfied. Usually, it is not possible to simultaneously satisfy the phase-matching condition for multiple frequency components of the nonlinear polarization; in the lab, we optimize the orientation of a second-order nonlinear crystal in order to properly radiate the desired frequency component. This phase matching criterion will be discussed next, in the context of SFG.

Sum-frequency generation (SFG)

To build up our understanding of how these nonlinear processes evolve in time, we will consider the nonlinear wave equation in the context of SFG. We restrict ourselves here to a lossless nonlinear medium, to which collimated, monochromatic, continuous wave (CW) beams are input at normal incidence, as shown in Fig 2.4(a). The energy-level diagram outlining the exchange of photons between the frequency modes involved is provided in Fig 2.4(b). We will make the assumption that the two input beams are undepleted by the nonlinear interaction (*i.e.*, the nonlinear interaction is small); this can be a good approximation in the context of the probabilistic single-photon sources discussed in Section 3.2.3.

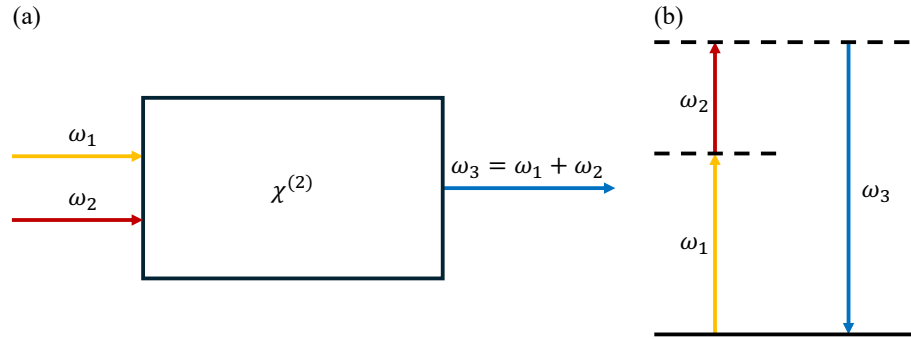


Figure 2.4: **Depiction of sum-frequency generation (SFG).** A schematic diagram of the fields involved in SFG in a $\chi^{(2)}$ crystal is shown in (a), while the corresponding energy-level diagram describing SFG is shown in (b).

As dictated by Eq. 2.57, the driven wave equation must hold for each frequency component of the field present in the nonlinear medium. In the case of SFG, the electric fields involved in the process are written as

$$E_i(z, t) = A_i e^{i(k_i z - \omega_i t)} + \text{c.c.}, \quad i = \{1, 2, 3\}, \quad (2.68)$$

where

$$k_i = \frac{n_i \omega_i}{c}, \quad \text{and} \quad n_i^2 = \epsilon^{(1)}(\omega_i). \quad (2.69)$$

Here, we take the fields to depend only on the longitudinal coordinate, z , for simplicity. We direct our focus to the sum-frequency component, at frequency ω_3 . In the absence of a nonlinear source term, A_3 is constant and the solution to the driven wave equation takes a plane wave form. If the nonlinear source term is present, but small, then A_3 will be a slowly varying function of propagation distance in the nonlinear medium, z . As such, we take $E_3(z, t)$, with A_3 as a function of z , as a trial solution to the driven wave equation with a nonlinear source term,

$$P_3(z, t) = 2\epsilon_0 \chi^{(2)} E_1(z, t) E_2(z, t) e^{-i\omega_3 t} + \text{c.c.} \quad (2.70)$$

Eqs. 2.69 and 2.70 can be substituted into the wave equation, Eq. 2.57. Since we consider only the longitudinal coordinate of the fields, we can replace ∇^2 with d^2/dz^2 and obtain

$$\begin{aligned} & \left[\frac{d^2 A_3}{dz^2} + 2ik_3 \frac{dA_3}{dz} - k_3^2 A_3 + \frac{\epsilon^{(1)}(\omega_3) \omega_3^2 A_3}{c^2} \right] e^{i(k_3 z - \omega_3 t)} + \text{c.c.} \\ & = -\frac{2\chi^{(2)} \omega_3^2}{c^2} A_1 A_2 e^{i[(k_1 + k_2)z - \omega_3 t]} + \text{c.c.} \end{aligned} \quad (2.71)$$

The equality still holds if we drop the complex conjugate (c.c.) terms from both sides of this equation, so we proceed under this simplification. We can further simplify the above by noting that $k_3^2 = \epsilon^{(1)}(\omega_3) \omega_3^2 / c^2$, and so we can cancel the third and forth terms on the left-hand side of the expression. Finally, the exponential factor can be

cancelled on both sides of the equation, and we have

$$\frac{d^2 A_3}{dz^2} + 2ik_3 \frac{dA_3}{dz} = -\frac{2\chi^{(2)}\omega_3^2}{c^2} A_1 A_2 e^{i(k_1+k_2-k_3)z}. \quad (2.72)$$

It is often permissible to apply the slowly varying amplitude approximation, *i.e.*,

$$\left| \frac{d^2 A_3}{dz^2} \right| \ll \left| k_3 \frac{dA_3}{dz} \right|, \quad (2.73)$$

since the fractional change in A_3 is generally much smaller than unity over distances on the order of an optical wavelength. Under this approximation, we arrive at

$$\frac{dA_3}{dz} = i \frac{\chi^{(2)}\omega_3}{n_3 c} A_1 A_2 e^{i\Delta k z}, \quad (2.74)$$

which is a coupled-amplitude equation, since A_3 depends on its coupling to A_1 and A_2 . Here, we have also introduced a new quantity, the wavevector mismatch,

$$\Delta k = k_1 + k_2 - k_3. \quad (2.75)$$

In general, the variation of A_1 and A_2 should also be taken into consideration. The above derivation can be repeated for these fields to find two more coupled-amplitude equations,

$$\frac{dA_1}{dz} = i \frac{\chi^{(2)}\omega_1}{n_1 c} A_3 A_2^* e^{i\Delta k z}, \quad (2.76)$$

$$\frac{dA_2}{dz} = i \frac{\chi^{(2)}\omega_2}{n_2 c} A_3 A_1^* e^{i\Delta k z}. \quad (2.77)$$

We remind ourselves that these equations assume a lossless medium, where no explicit loss terms must be included and we can make use of full permutation symmetry to take $\chi^{(2)}$ to be a scalar quantity with the same value in each equation.

In order to understand how these coupled-amplitude equations describe the evolution of SHG, we turn our attention to the wavevector mismatch introduced in Eq. 2.75. In the undepleted pump approximation, we can take A_1 and A_2 as constant values in Eq. 2.74, and solve for $A_3(z)$. Under perfect phase-matching conditions, $\Delta k = 0$,

we see that $A_3(z)$ increases linearly with z , and so the intensity of this field increases quadratically with propagation distance. Perfect phase matching implies that the generated field with frequency ω_3 has a fixed phase relative to the nonlinear polarization, and can thus efficiently extract energy from the incident fields. This can be pictured microscopically, where the individual atomic dipoles in the nonlinear medium are all in phase, and so their emitted fields each add coherently in the forward direction.

When $\Delta k \neq 0$ (*i.e.*, under imperfect phase matching), the emitted radiation at frequency ω_3 will have a smaller intensity. Under this condition, we can determine A_3 by integrating Eq. 2.74 over the length of the nonlinear medium,

$$\begin{aligned} A_3(z) &= i \frac{\chi^{(2)} \omega_3}{n_3 c} A_1 A_2 \int_0^L e^{i \Delta k z} dz \\ &= i \frac{\chi^{(2)} \omega_3}{n_3 c} A_1 A_2 \frac{e^{i \Delta k L} - 1}{i \Delta k}. \end{aligned} \quad (2.78)$$

The intensity of a field at frequency ω_i field is given by the time-averaged Poynting vector,

$$I_i = 2n_i \epsilon_0 c |A_i|^2, \text{ where } i = \{1, 2, 3\}, \quad (2.79)$$

and so the intensity of the generated field at frequency ω_3 is given by

$$I_3 = \frac{(\chi^{(2)})^2 \omega_3^2 I_1 I_2}{2n_1 n_2 n_3 \epsilon_0 c^3} L^2 \text{sinc}^2 \left(\frac{\Delta k L}{2} \right), \quad (2.80)$$

where we have used the fact that

$$\left| \frac{e^{i \Delta k L} - 1}{i \Delta k} \right|^2 = L^2 \text{sinc}^2 \left(\frac{\Delta k L}{2} \right). \quad (2.81)$$

Wavevector mismatch is only present in the $\text{sinc}^2(\Delta k/L)$ factor, which is plotted in Fig. 2.5. We see that the efficiency of this three-wave mixing process decreases with $|\Delta k|L$, with some oscillations. Intuitively, if $L > 1/|\Delta k|$, the generated wave at ω_3 can become out of phase with its driving polarization, causing power to flow from A_3 back into A_1 and A_2 . It is therefore important to consider phase-matching conditions when performing nonlinear optics in the lab.

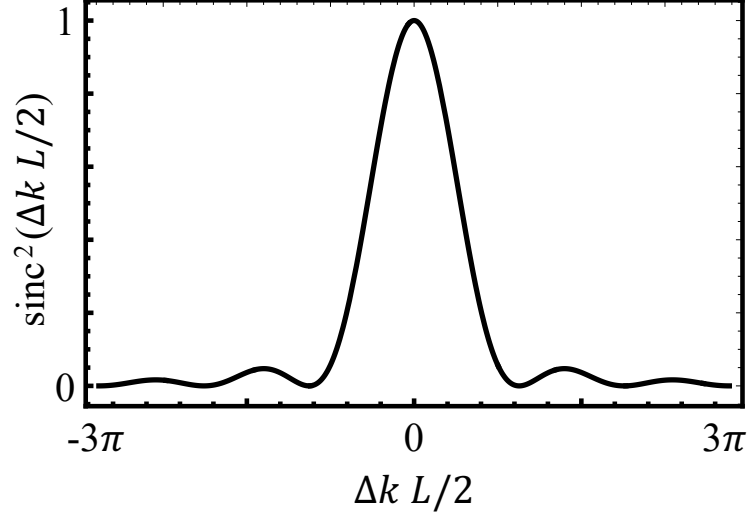


Figure 2.5: **Wavevector mismatch in SFG.** The dependence of SFG intensity, $I \propto \text{sinc}^2(\Delta k L/2)$, on wavevector mismatch, Δk .

Phase matching

When considering the time evolution of SFG in the previous section, we saw that the intensity of the generated field at ω_3 decreases, as in Fig. 2.5, when the phase-matching condition, $\Delta k = k_1 + k_2 - k_3 = 0$, is not met. Even for nonlinear processes which are efficient enough to cause depletion of the input fields, where the functional dependence of the field intensity on propagation length is no longer given by Eq. 2.80, the condition $\Delta k = 0$ must be maintained for high nonlinear conversion efficiency.

It is often difficult to meet the phase-matching condition $\Delta k = 0$. Over the range of frequencies involved in SFG, a lossless material exhibits normal dispersion; the refractive index increases with frequency. Perfect phase matching would dictate that

$$\frac{n_1 \omega_1}{c} + \frac{n_2 \omega_2}{c} = \frac{n_3 \omega_3}{c}, \quad (2.82)$$

which, for a monotonically increasing index of refraction over frequencies ω_1 to ω_3 , cannot be satisfied simultaneously with the energy conservation requirement of the SFG process, $\omega_1 + \omega_2 = \omega_3$. This can be shown explicitly by rearranging Eq. 2.82,

$$n_3 - n_2 = (n_1 - n_2) \frac{\omega_1}{\omega_3}, \quad (2.83)$$

where, for a monotonically increasing $n(\omega)$, the left-hand side should be positive, while the right-hand side should be negative, and thus cannot hold.

From this analysis, we see that it is necessary to choose a material system in which the refractive index allows us to satisfy Eq. 2.82. Although it is possible to make use of anomalous dispersion, where the refractive index decreases with frequency, this requires operating near an absorption feature. Instead, the most common method to achieve phase matching makes use of birefringence¹⁸, where the refractive index depends on the direction of an optical field's polarization. In order for a crystal to display birefringence, it cannot be optically isotropic; rather, it must exhibit different refractive indices along two different axes, as shown in Fig. 2.6. Achieving phase matching through birefringence requires the highest-frequency field, $\omega_3 = \omega_1 + \omega_2$, to be polarized along the crystal axis with the lower of the two refractive indices. Both uniaxial (trigonal, tetragonal, or hexagonal) and biaxial (triclinic, monoclinic, or orthorhombic) crystals exhibit birefringence; here, we will consider the simpler case of uniaxial crystals, to demonstrate how phase-matching requirements can be met.

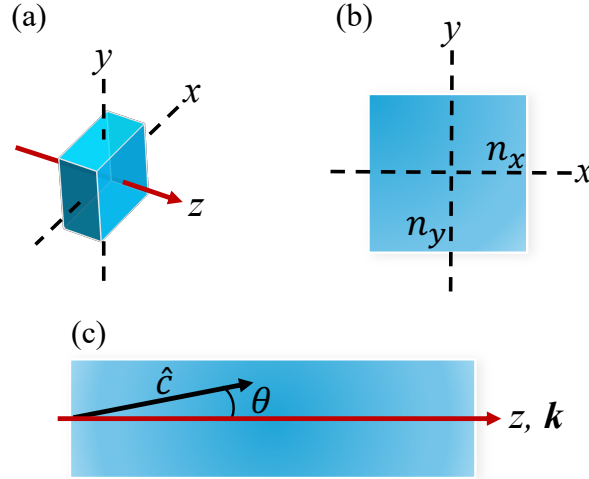


Figure 2.6: **Birefringence in an optical crystal.** A birefringent crystal, with axes defined as in (a), has different refractive indices, n_x and n_y , along the two directions, $\hat{x}$ and $\hat{y}$, which are perpendicular to the optical, z , axis (the axis along which the input beam propagates, $\mathbf{k}$). The optic axis of the crystal, $\hat{c}$, is depicted relative to $\mathbf{k}$ in (c).

¹⁸We will exploit birefringent crystals for our ultrafast time-bin encoding (UTBE) scheme for quantum computing in later sections!

Uniaxial crystals have a single crystal direction, $\hat{c}$ (the optic axis or crystal axis), which governs the optical anisotropy, whereas all other perpendicular directions are optically equivalent. In other words, as light propagates through the crystal in the direction of $\hat{c}$, the crystal behavior does not change. Light that is polarized perpendicular to the plane containing the propagation vector, $\mathbf{k}$, and the optic axis is governed by the *ordinary* refractive index, n_o , while light polarized in the plane containing $\mathbf{k}$, and the optic axis is governed by the *extraordinary* refractive index, n_e . Uniaxial crystals can be positive ($n_e > n_o$) or negative ($n_o > n_e$). For both positive and negative uniaxial crystals, there are two choices for the polarization of the lower frequency components, ω_1 and ω_2 . Type I phase matching refers to the case where the fields at ω_1 and ω_2 have the same polarization, while Type II phase matching refers to the case where the fields at ω_1 and ω_2 have orthogonal polarization [293]. Table 2.2 summarizes these details.

	Positive uniaxial ($n_e > n_o$)	Negative uniaxial ($n_o > n_e$)
Type I	$n_3^o \omega_3 = n_1^e \omega_1 + n_2^e \omega_2$	$n_3^e \omega_3 = n_1^o \omega_1 + n_2^o \omega_2$
Type II	$n_3^o \omega_3 = n_1^o \omega_1 + n_2^e \omega_2$	$n_3^e \omega_3 = n_1^e \omega_1 + n_2^o \omega_2$

Table 2.2: **Phase matching in uniaxial crystals.** The phase-matching condition for positive and negative uniaxial crystals is provided, for both Type I and Type II phase matching.

Typically, phase matching in birefringent crystals is accomplished through either angle tuning or temperature tuning. In angle tuning, the crystal is rotated to adjust the angle of incidence of the collinear input beams relative to the optic axis so as to maximize the nonlinear effect. Angle tuning has one major drawback: when the angle between the propagation direction and optic axis is not 0° or 90° , the Poynting vector is no longer parallel to the propagation vector for extraordinary polarization. This results in the fields with extraordinary polarization quickly diverging from the fields with ordinary polarization, ultimately limiting the spatial overlap of the two

waves and thus also the nonlinear mixing process. Some crystals have a temperature-dependent birefringence, which can be used to achieve phase matching while the angle is kept at either 0° or 90° [294].

There are some situations in which birefringence is not suitable to achieve phase matching. For example, it may be desirable to take advantage of the large nonlinearity in lithium niobate, which can only be accessed if all interacting fields have the same polarization. Birefringence also tends to be insufficient for high-frequency nonlinearities. In these cases, other methods must be employed to achieve phase matching; for example, quasi-phase-matching in a material which has been engineered such that the orientation of one of the crystal axes is periodically inverted as a function of the propagation distance in the crystal. A periodic inversion of the $\hat{c}$ axis changes the sign of the nonlinear susceptibility, $\chi^{(2)}$, periodically. This in turn allows for compensation of a nonzero wavevector mismatch, and the field amplitude of generated field is allowed to grow when it would have otherwise begun to decrease.

Difference-frequency generation (DFG) and optical parametric amplification

The last second-order nonlinear process we consider is DFG, which is described by the nonlinear polarization term,

$$P(\omega_1 - \omega_2) = 2\epsilon_0\chi^{(2)}E_1E_2^*. \quad (2.84)$$

and depicted in Fig. 2.7. In DFG, the frequency of the generated field is the difference between the frequencies of the applied fields, $\omega_3 = \omega_1 - \omega_2$. Although DFG and SFG may appear to be very similar processes, a key difference between the two is seen when comparing their energy-level diagrams. In SFG, energy conservation requires two input photons, at frequencies ω_1 and ω_2 be destroyed in order to generate a photon at frequency ω_3 . On the other hand, for energy conservation to hold in DFG, a photon at the higher input frequency, ω_1 , must be destroyed while a photon at the lower input frequency, ω_2 , must be created in order to generate a photon at frequency ω_3 . For this reason, DFG is often referred to as optical parametric amplification. It should be noted that the DFG process can either be stimulated by a field at

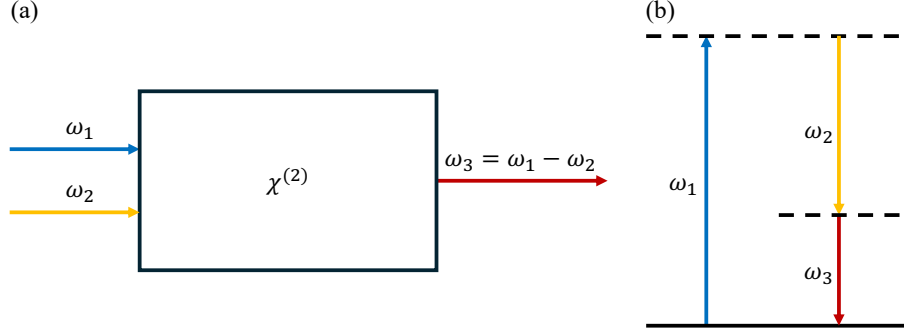


Figure 2.7: **Depiction of difference-frequency generation (DFG).** A schematic diagram of the fields involved in DFG in a $\chi^{(2)}$ crystal is shown in (a), while the corresponding energy-level diagram describing DFG is shown in (b).

frequency ω_2 , or in the absence of an applied field at this frequency. In the latter case, the process is referred to as spontaneous parametric down-conversion (SPDC) or parametric fluorescence. SPDC is often used to generate entangled photon pairs for quantum optics experiments [265, 295, 296].

The coupled amplitude equations for DFG can be obtained following a similar procedure to the one we used in describing SFG, such that

$$\frac{dA_2}{dz} = i \frac{\chi^{(2)} \omega_2}{n_2 c} A_1 A_3^* e^{i\Delta k z}, \quad (2.85)$$

$$\frac{dA_3}{dz} = i \frac{\chi^{(2)} \omega_3}{n_3 c} A_1 A_2^* e^{i\Delta k z}, \quad (2.86)$$

where $\Delta k = k_1 - k_2 - k_3$. In the undepleted pump approximation, we treat A_1 as constant. Under the initial conditions that $A_3(z = 0) = 0$ and $A_2(z = 0)$ is arbitrary, the solutions to these coupled differential equations are

$$A_2(z) = A_2(0) \cosh(\kappa z), \quad (2.87)$$

$$A_3(z) = i \sqrt{\frac{n_2 \omega_3}{n_3 \omega_2}} \frac{A_1}{|A_1|} A_2^*(0) \sinh(\kappa z), \quad (2.88)$$

where

$$\kappa = \sqrt{\frac{(\chi^{(2)})^2 \omega_2 \omega_3}{n_2 n_3 c^2}} |A_1|^2. \quad (2.89)$$

We see that the fields at both ω_2 and ω_3 experience monotonic growth, which is qualitatively very different from our results for SFG. Intuitively, from the energy-level picture, the presence of the field at ω_2 stimulates the downward transition which generates the field at ω_3 , and vice versa. In other words, the generation of one of these two fields reinforces the generation of the other field, which leads to exponential growth of both fields. For the input conditions considered here, we can say that the field at ω_2 (signal) is amplified by the nonlinear process, and the field at ω_3 (idler) is generated. Placing the nonlinear medium inside an optical resonator cavity allows oscillation to occur at frequencies ω_2 and ω_3 due to the gain of the parametric amplification process. This device is known as an optical parametric oscillator (OPO) and is briefly discussed in the following section.

Earlier we noted that even in the absence of fields at both ω_1 and ω_2 , this process can still occur (*i.e.*, in SPDC); however, we see that the coupled-amplitude equations would all go to zero under such initial conditions. We see that under this classical treatment, the DFG process cannot occur with only field at ω_3 present; a quantum mechanical description is required to allow for spontaneous generation of signal and idler photon pairs, which will be expanded on in Chapter 3.

Optical parametric oscillation

In the previous section, we explored how DFG, in the presence of radiation at frequencies ω_2 or ω_3 , can stimulate the emission of additional photons at these frequencies. We said that placing the nonlinear medium within an optical resonator, as shown in Fig. 2.8, would allow these fields to build up to much larger values, through the process of optical parametric oscillation. This process is a useful source of tunable radiation in the lab, as it can have broad tuning ranges. The output frequency of an OPO can be controlled by tuning the phase-matching condition of the nonlinear crystal within the resonator. It may be useful to know that the highest frequency field involved in the DFG process within an OPO is generally called the pump, while

the two lower-frequency fields are called the signal and idler. Generally, the desired output field is referred to as the signal.

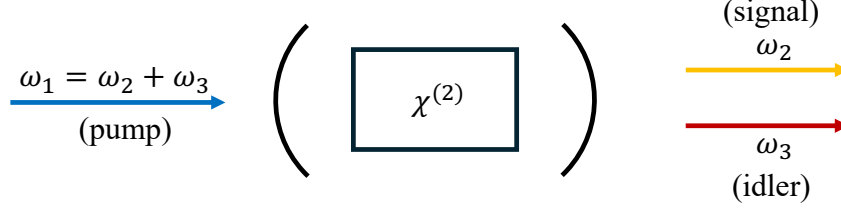


Figure 2.8: **An optical parametric oscillator (OPO).** The resonator mirrors have high reflectivities at frequencies ω_2 and/or ω_3 , depending on the desired output frequency. Tuning the crystal orientation allows for tuning of the output frequencies.

Parametric vs. nonparametric processes

In describing OPO and OPA, we have introduced the concept of a parametric process. This refers to a process where the initial and final quantum-mechanical states of the nonlinear medium are identical. In other words, an electron can only be removed from its ground state for a brief period of time, during which it resides in a virtual level. This period of time is limited, by the Heisenberg uncertainty principle, to the order of $\hbar/\Delta E$, where ΔE is the energy difference between the virtual level and nearest real level. In contrast, *nonparametric* processes involve the transition of electrons between real energy levels.

Most nonlinear processes used in this thesis are parametric, where photon energy is conserved. In nonparametric processes, such as saturable absorption (often used to achieve mode-locked lasing), two-photon absorption, and stimulated Raman scattering, energy can be transferred between the incident fields and the medium.

2.2.3 Third-order nonlinear optical processes

We next consider the third-order $\chi^{(3)}$ term in our power series expansion of the polarization in Eqs. 2.36 to 2.38. Third-order optical nonlinearities are *crucial* to the work presented throughout this thesis. In contrast to the second-order effects

discussed in previous sections, third-order effects are not constrained by material symmetry (or lack thereof); third-order effects can occur in any medium, although their strength will still be determined by the material properties. Third-order nonlinear optical interactions arise from the nonlinear polarization term,

$$P^{(3)}(t) = \epsilon_0 \chi^{(3)} E^3(t), \quad (2.90)$$

where the applied electric field can consist of up to three frequency components,

$$E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + E_3 e^{-i\omega_3 t} + \text{c.c.} \quad (2.91)$$

Expanding the expression for $P^{(3)}(t)$ leads to an expression which contains 44 different frequency components (taking the positive and negative frequencies to be distinct). If we represent the nonlinear polarization as

$$P^{(3)}(t) = \sum_n P(\omega_n) e^{-i\omega_n t}, \quad (2.92)$$

we can write the 22 possible complex amplitudes of the nonlinear polarization as, $P(\omega_n)$, for positive ω_n as

$$\begin{aligned} P(\omega_1) &= \epsilon_0 \chi^{(3)} (3E_1 E_1^* + 6E_2 E_2^* + 6E_3 E_3^*) E_1, \\ P(\omega_2) &= \epsilon_0 \chi^{(3)} (6E_1 E_1^* + 3E_2 E_2^* + 6E_3 E_3^*) E_2, \\ P(\omega_3) &= \epsilon_0 \chi^{(3)} (6E_1 E_1^* + 6E_2 E_2^* + 3E_3 E_3^*) E_3, \\ P(3\omega_1) &= \epsilon_0 \chi^{(3)} E_1^3, \quad P(3\omega_2) = \epsilon_0 \chi^{(3)} E_2^3, \quad P(3\omega_3) = \epsilon_0 \chi^{(3)} E_3^3 \\ P(\omega_1 + \omega_2 + \omega_3) &= 6\epsilon_0 \chi^{(3)} E_1 E_2 E_3, \\ P(\omega_1 + \omega_2 - \omega_3) &= 6\epsilon_0 \chi^{(3)} E_1 E_2 E_3^*, \\ P(\omega_1 + \omega_3 - \omega_2) &= 6\epsilon_0 \chi^{(3)} E_1 E_3 E_2^*, \\ P(\omega_2 + \omega_3 - \omega_1) &= 6\epsilon_0 \chi^{(3)} E_2 E_3 E_1^*, \\ P(2\omega_1 + \omega_2) &= 3\epsilon_0 \chi^{(3)} E_1^2 E_2, \quad P(2\omega_1 + \omega_3) = 3\epsilon_0 \chi^{(3)} E_1^2 E_3, \\ P(2\omega_2 + \omega_1) &= 3\epsilon_0 \chi^{(3)} E_2^2 E_1, \quad P(2\omega_2 + \omega_3) = 3\epsilon_0 \chi^{(3)} E_2^2 E_3, \\ P(2\omega_3 + \omega_1) &= 3\epsilon_0 \chi^{(3)} E_3^2 E_1, \quad P(2\omega_3 + \omega_2) = 3\epsilon_0 \chi^{(3)} E_3^2 E_2, \\ P(2\omega_1 - \omega_2) &= 3\epsilon_0 \chi^{(3)} E_1^2 E_2^*, \quad P(2\omega_1 - \omega_3) = 3\epsilon_0 \chi^{(3)} E_1^2 E_3^*, \\ P(2\omega_2 - \omega_1) &= 3\epsilon_0 \chi^{(3)} E_2^2 E_1^*, \quad P(2\omega_2 - \omega_3) = 3\epsilon_0 \chi^{(3)} E_2^2 E_3^*, \\ P(2\omega_3 - \omega_1) &= 3\epsilon_0 \chi^{(3)} E_3^2 E_1^*, \quad P(2\omega_3 - \omega_2) = 3\epsilon_0 \chi^{(3)} E_3^2 E_2^*, \end{aligned} \quad (2.93)$$

where the argument of each $P(\omega_n)$ term is the sum of frequencies associated with the field amplitudes that appear on the right-hand side of each expression. The above convention associates complex field amplitudes with negative frequency components, and the numerical factors of 1, 3, or 6 indicate the number of possible distinct permutations of the field frequencies contributing to the polarization term. The form used in Eq. 2.94 is convenient to work with, as it provides us immediate information about the nonlinear process described. Some of the third-order nonlinear processes which the terms above describe include third-harmonic generation, four-wave mixing (FWM), and the intensity-dependent nonlinear refractive index.

We will not describe every single third-order nonlinear process in this section; rather, we will focus on those which are key to the work in this thesis: degenerate FWM and the intensity-dependent refractive index.

Degenerate four-wave mixing (FWM)

Each of the terms given in Eq. 2.94 describes a specific FWM process. The degenerate FWM process of interest in this thesis, depicted in Fig. 2.9, is governed by the polarization term,

$$P(\omega_i = 2\omega_p - \omega_s) = 6\epsilon_0\chi^{(3)}E_p^2E_s^*. \quad (2.94)$$

We see that the polarization of the ω_i field depends quadratically on the amplitude of the pump field, ω_p (equivalently, the idler beam intensity increases quadratically with the pump beam intensity). It should be noted that here, we consider a FWM process which is stimulated by the presence of an input signal field at frequency ω_s , as shown in Fig. 2.9(a); however, the signal, s , and idler, i , indices can be swapped in the case of stimulation by the idler.

As we saw in our discussion of DFG, this classical description of FWM predicts that in the absence of stimulation at the signal frequency (*i.e.*, no field at ω_s), the polarization amplitude will be zero and the nonlinear process would not occur; yet, it is in fact possible to observe *spontaneous* four-wave mixing (SFWM), depicted in Fig. 2.9(c), in which only a pump field, ω_p , is applied to the medium. This process requires a quantum-mechanical description, which will be expanded on in Chapter 3.

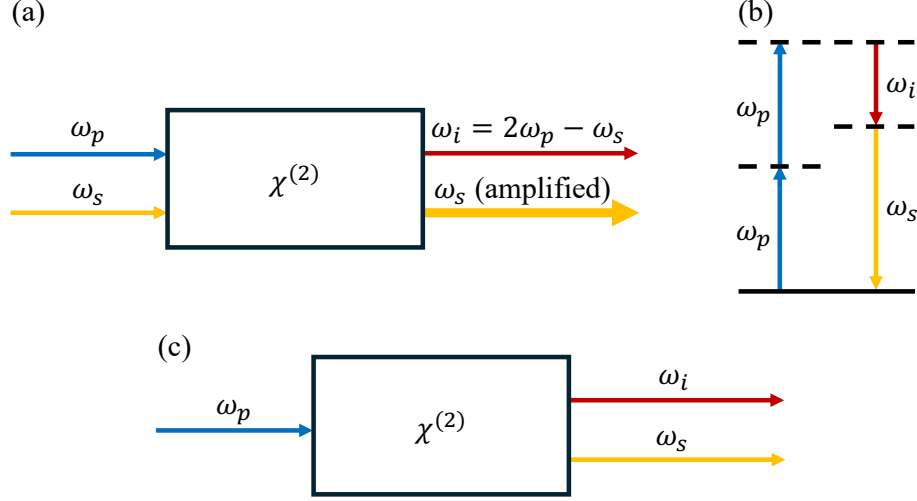


Figure 2.9: **Depiction of degenerate four-wave mixing (FWM).** A schematic diagram of the fields involved in degenerate FWM when the process is stimulated by the signal field, ω_s is shown in (a). The corresponding energy-level diagram is shown in (b). Spontaneous FWM (SFWM), in which only the pump field is incident on the crystal, is depicted in (c).

Nonlinear refractive index

The nonlinear optical effect which lies at the heart of this thesis is the intensity-dependent nonlinear refractive index arising from the $\chi^{(3)}$ nonlinearity. This is often referred to as the optical Kerr effect, and describes the process through which an applied field generates a third-order nonlinear polarization, that changes the refractive index of the medium. This change in refractive index is proportional to the intensity of the applied field, leading to a new total refractive index of

$$n = n_0 + n_2 I, \quad (2.95)$$

where n_0 is the linear refractive index, n_2 is the intensity-dependent nonlinear refractive index, and I is the intensity of the applied field. In this thesis, we exploit the Kerr effect in single-mode fiber (SMF) made up of fused silica. For fused silica, n_2 is on the order of $3 \times 10^{-16} \text{ cm}^2/\text{W}$, so ultrafast laser pulses are typically required to reach intensities that can generate a noticeable change in refractive index.

This change in refractive index can be used to induce self-action effects on the

input field (the pump) or to induce cross-action effects on a secondary field (the probe). Section 2.2.4 elaborates on how these self- and cross-action effects occur.

2.2.4 Optical Kerr effect

As shown in Fig. 2.10, there are two ways in which the optical Kerr effect can be exploited, both of which rely on the intensity-dependent refractive index arising from the $\chi^{(3)}$ nonlinear response described in the previous section. When the strong input field modulates itself, as seen in Fig. 2.10(a), this is referred to as self-phase modulation (SPM). This strong (pump) field can also be used to modulate a weaker (probe) field, as seen in Fig. 2.10(b), a process which is referred to as cross-phase modulation (XPM). The following sections describe SPM and XPM in more detail.

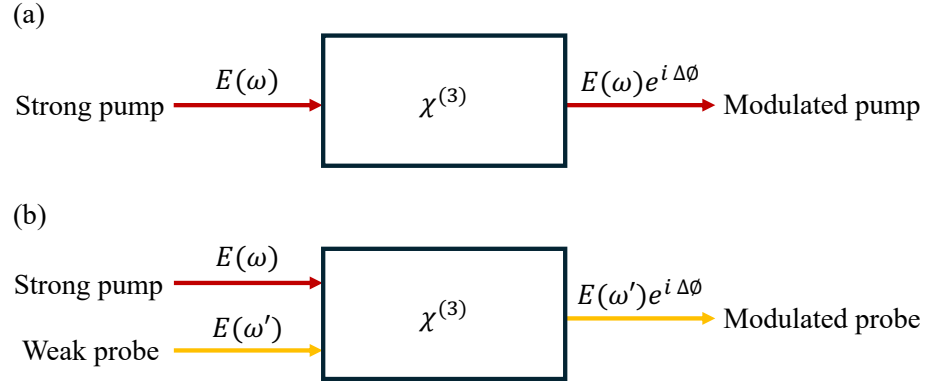


Figure 2.10: **Effects due to the intensity-dependent refractive index.** A strong pump field modifies its own propagation through self-phase modulation (SPM) in (a) while a strong pump field is used to modify the propagation of a weaker probe field through cross-phase modulation (XPM) in (b).

Self-phase modulation (SPM)

If only a strong pump beam is present in the $\chi^{(3)}$ medium, SPM will occur. The nonlinear polarization which describes SPM is given by

$$P^{(3)}(\omega = \omega + \omega - \omega) = 3\epsilon_0\chi^{(3)} |E(\omega)|^2 E(\omega), \quad (2.96)$$

where we assume linearly polarized fields for simplicity. The total polarization of this frequency-degenerate process is

$$P^{\text{TOT}}(\omega) = \epsilon_0 \chi^{(1)} E(\omega) + 3\epsilon_0 \chi^{(3)} |E(\omega)|^2 E(\omega), \quad (2.97)$$

which we can simplify by introducing the effective susceptibility,

$$\chi_{\text{eff}} = \chi^{(1)} + 3\chi^{(3)} |E(\omega)|^2. \quad (2.98)$$

In general, the refractive index of a medium is related to its susceptibility as

$$n^2 = 1 + \chi_{\text{eff}}, \quad (2.99)$$

and by introducing Eq. 2.95 on the right-hand side, we find

$$\begin{aligned} (n_0 + n_2 I)^2 &= 1 + \chi_{\text{eff}} \\ (n_0 + 2n_2 n'_0 \epsilon_0 c |E(\omega)|^2)^2 &= 1 + \chi^{(1)} + 3\chi^{(3)} |E(\omega)|^2, \end{aligned} \quad (2.100)$$

where we have made use of the relation

$$I = 2n'_0 \epsilon_0 c |E(\omega)|^2. \quad (2.101)$$

Note that here, we are expressing the refractive index in terms of its real and imaginary parts as $n = n' + in''$ (which also holds for n_0 and n_2). When expanded, Eq. 2.100 becomes (correct to order $|E(\omega)|^2$),

$$n_0^2 + 4n_0 n'_0 n_2 \epsilon_0 c |E(\omega)|^2 = (1 + \chi^{(1)}) + 3\chi^{(3)} |E(\omega)|^2. \quad (2.102)$$

As expected, the linear part of the refractive index is given by

$$n_0^2 = 1 + \chi^{(1)}, \quad (2.103)$$

while the nonlinear part of the refractive index is given by

$$n_2 = \frac{3}{4n_0 n'_0 \epsilon_0 c} \chi^{(3)}. \quad (2.104)$$

Provided the frequency of the pump is far from an absorption peak of the medium, this can be approximated as

$$n_2 = \frac{3}{4n_0^2 \epsilon_0 c} \chi^{(3)}. \quad (2.105)$$

The effects of SPM lead to a change in phase of the input optical field. If that input field is an ultrashort pulse, SPM will cause spectral broadening. In some cases this spectral broadening is exploited, *e.g.*, in supercontinuum generation (SCG)—used in this thesis. Yet, SPM is often a parasitic effect which must be minimized, for example in the optical Kerr gating technique used in all work presented in this thesis. Pulse broadening due to SPM is outlined later in this section, when discussing SCG.

Cross-phase modulation (XPM)

If both the strong pump beam and a weaker probe beam are present in the $\chi^{(3)}$ medium, both SPM and XPM will occur. The nonlinear polarization which describes XPM is given by

$$P^{(3)}(\omega' = \omega' + \omega - \omega) = 6\epsilon_0 \chi^{(3)} |E(\omega)|^2 E(\omega'), \quad (2.106)$$

where the frequency of the probe field is given by ω' . Following an analogous procedure for finding the nonlinear refractive index of the SPM process, the nonlinear refractive index experienced by the probe beam is

$$n_2^{(\text{XPM})} = \frac{3}{2n_0^2 \epsilon_0 c} \chi^{(3)}, \quad (2.107)$$

under the assumption that neither the pump nor probe frequency are near an absorption peak of the medium. The results presented here assumed co-polarized pump and probe fields; if the pump and probe fields are cross-polarized, the tensor nature of $\chi^{(3)}$ is three times less than for co-polarized fields [269].

Through XPM, a strong pump beam can be used to modulate the phase of a weaker probe beam. In the case where the pump and probe fields are cross-polarized, the probe field polarization will be rotated. It is this effect which we make use of throughout this thesis to achieve optical Kerr gating of ultrashort signals. Ultrafast optical Kerr gating will be described in Section 2.2.5.

Supercontinuum generation (SCG)

The spectral broadening induced by SPM can be used to generate a very broad optical spectrum, a process referred to as SCG. When an ultrashort pulse is applied to a $\chi^{(3)}$ nonlinear medium, it experiences a nonlinear phase shift, ϕ_{NL} , arising due to the intensity-dependent nonlinear refractive index, n_2 . The origin of this phenomenon can be understood by considering an applied optical pulse (at central frequency ω_0), with intensity

$$I(t) = I_0 \exp\left(-\frac{t^2}{t_p^2}\right), \quad (2.108)$$

propagating through a nonlinear medium of length L , with a refractive index given by¹⁹

$$n(t) = n_0 + n_2 I(t), \quad (2.109)$$

where $I(t) = 2n_0 \epsilon_0 c |A(z, t)|^2$. Here we assume an instantaneous nonlinear response. We see that the index of refraction is now a time-varying quantity, $\frac{dn(t)}{dt} = n_2 \frac{dI(t)}{dt}$, as is the instantaneous phase of the pulse,

$$\phi(t) = \omega_0 t - kz = \omega_0 t - \frac{2\pi}{\lambda_0} n(t) L. \quad (2.110)$$

The instantaneous frequency of the pulse can be written in terms of the phase as

$$\begin{aligned} \omega(t) &= \frac{d\phi(t)}{dt} \\ &= \omega_0 - \frac{2\pi L}{\lambda_0} \frac{dn(t)}{dt}, \end{aligned} \quad (2.111)$$

¹⁹Here, we assume a mechanism driven by the electronic polarization, which has a response time (10^{-15} s) much faster than the pulse (10^{-13} s) driving the process.

which, for the intensity given in Eq. 2.109, evaluates to

$$\omega(t) = \omega_0 + \frac{4\pi L n_2 I_0}{\lambda_0 t_p^2} t \exp\left(-\frac{t^2}{t_p^2}\right). \quad (2.112)$$

It is evident that, for positive n_2 , the leading edge of the pulse is shifted to lower (more red) frequencies, while the trailing edge of the pulse is shifted to higher (more blue) frequencies. We also see that spectral broadening due to SPM grows with the strength of the nonlinearity ($n_2 I_0$), and will become noticeable when the maximum frequency shift exceeds the spectral bandwidth of the input pulse.

2.2.5 All-optical Kerr gating

In order to perform all-optical quantum information processing in the ultrafast regime, we require a way to manipulate and detect quantum information on ultrafast timescales that is scalable. One highly promising means of achieving this capability is through all-optical Kerr gating, a technique which is central to this thesis.

All-optical Kerr gating relies on XPM, described in Section 2.2.4, whereby a strong pump pulse generates a localized intensity-dependent refractive index modulation in the medium through which it propagates. This change in refractive index can be used to induce a nonlinear phase shift in a secondary optical signal. If the pump is polarized 45° with respect to the signal²⁰, this nonlinear phase shift induces a birefringence in the medium which can be used to rotate the signal polarization by some amount. When the pump is an ultrashort pulse, this induced birefringence will only occur over very short timescales²¹. It is generally assumed that the probe beam is too weak to induce a significant change in the refractive index on its own.

The induced phase shift can be written as

$$\Delta\phi = \frac{2\pi}{\lambda_s} (n_x - n_y) L_{\text{eff}}, \quad (2.113)$$

where λ_s is the signal wavelength, L_{eff} is the effective length of the Kerr medium, and

²⁰Here, we assume both the pump and signal are linearly polarized at the Kerr medium input.

²¹This effect can be thought of as a highly localized wave plate, only where the pump is located within the medium.

n_x and n_y are the refractive indices of the Kerr medium, along the directions parallel and perpendicular to the pump polarization, respectively. These refractive indices are written as²²

$$n_x(t) = n_{x,0} + 2n_2I(t), \quad (2.114)$$

$$n_y(t) = n_{y,0} + 2n_2bI(t), \quad (2.115)$$

where $I(t)$ is the time-dependent intensity profile of the pump, n_2 is the nonlinear refractive index, and b is a constant which depends on the nonlinear susceptibility of the Kerr medium. If the medium is isotropic, then $n_{x,0} = n_{y,0} = n_0$ ²³, and if the susceptibility is purely electronic in nature, then $b = \frac{1}{3}$. These values are suitable in the context of this thesis, where we employ short pieces of SMF as the medium for optical Kerr gating.

We can now write the induced phase shift, along the directions parallel and perpendicular to the pump polarization, as

$$\Delta\phi_x(t) = \frac{2\pi L_{\text{eff}}}{\lambda_s} (n_0 + 2n_2I(t)), \quad (2.116)$$

$$\Delta\phi_y(t) = \frac{2\pi L_{\text{eff}}}{\lambda_s} \left(n_0 + \frac{2}{3}n_2I(t) \right), \quad (2.117)$$

and the total phase shift is

$$\begin{aligned} \Delta\phi(t) &= \Delta\phi_x(t) - \Delta\phi_y(t) \\ &= \frac{8\pi n_2 L_{\text{eff}}}{3\lambda_s} I(t). \end{aligned} \quad (2.118)$$

When the Kerr medium is sufficiently long (*e.g.*, the SMF used in this thesis) and the pump and signal are ultrashort pulses, the effective length of the nonlinear interaction must be considered in more detail. Since the pump and signal will necessarily be at different wavelengths (for detection purposes), they will have different group

²²Note that here we include a factor of 2 in the nonlinear refractive index terms because we are considering an XPM process. Usually, a medium's n_2 will be quoted as per the expression in Eq. 2.104, which considers the SPM process.

²³Technically, SMF does have a very small intrinsic birefringence; however, this is compensated for in the lab with quarter- and half-wave plates (QWPs and HWPs), so $n_{x,0} = n_{y,0} = n_0$ holds.

velocities within the fiber. This is incorporated into a more completed expression for the induced phase shift,

$$\Delta\phi(T) = \frac{8\pi n_2}{3\lambda_0} \int_0^L I_p(T - d_w z) dz, \quad (2.119)$$

where z is the propagation distance and I_p is the pump intensity profile in time, expressed in the reference frame moving with the signal, $T = t - z/v_{gs}$, where t is the laboratory frame. The temporal walk-off between the pump and signal within the medium is given by $d_w = v_{gp}^{-1} - v_{gs}^{-1}$, where v_{gp} and v_{gs} are the pump and signal group velocities, respectively.

The Kerr medium is placed between two crossed polarizers, a configuration referred to as an optical Kerr shutter (OKS)²⁴ and depicted in Fig. 2.11. Here, the transmission of the signal through the second polarizer depends on the amount of polarization rotation it experiences due to the phase shift induced by the pump. The efficiency of the OKS is thus given by [259]

$$\eta(T) = \sin^2(2\theta) \sin^2\left(\frac{\Delta\phi(T)}{2}\right), \quad (2.120)$$

where θ is the angle between the pump and signal polarization. When $\eta = 0$, no polarization rotation in the signal has occurred, and when $\eta = 1$, the signal polarization has been rotated to an orthogonal state. We also see that the OKS efficiency can only be maximized if the signal polarization is 45° with respect to the pump, a condition we meet in all experiments presented in this thesis.

In this thesis, we employ optical Kerr gating for both manipulation and detection of the ultrafast time bins in our photonic quantum computing platform. With hundreds-of-femtoseconds duration pump pulses and short pieces of SMF (length ≤ 10 cm), we are able to achieve Kerr gate resolution of ~ 1 ps or less. This is orders of magnitude faster than the fastest Pockels cells available, which have < 250 ps rise times²⁵, as indicated in Table 2.3.

²⁴An OKS is analogous to a Pockels cell, where a changing electrical field is applied to stimulate birefringence in a material, essentially turning it into a wave plate.

²⁵Here, I quote values from the EM500 series of KD*P longitudinal Pockels cells available from *Electro Optical Components Inc.* for Q-switching applications.

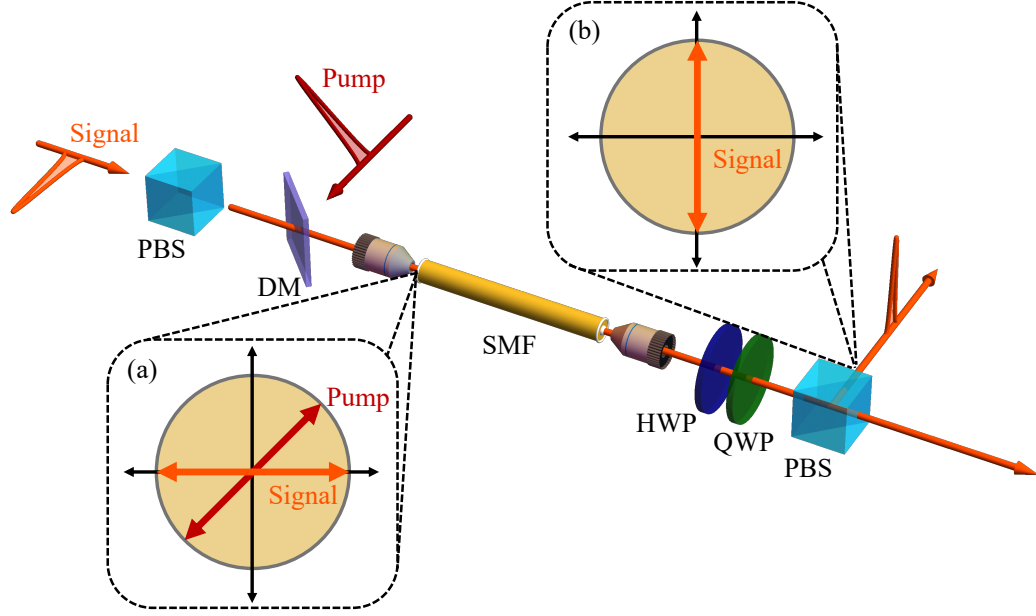


Figure 2.11: **Schematic of an optical Kerr shutter (OKS).** The Kerr medium is a short piece of single-mode fiber (SMF), placed between two crossed polarizers. A strong pump pulse is combined with a weak signal pulse on a dichroic mirror (DM) before both beams are launched into the SMF. The pump pulse induces a birefringence in the SMF which rotates the polarization of the signal. The rotated signal is redirected by a polarizing beam splitter (PBS). The pump and signal polarizations prior to rotation are shown in (a), while the signal polarization after rotation is shown in (b). HWP: half-wave plate; QWP: quarter-wave plate.

It should be noted that other methods of all-optical gating exist, included in Table 2.3, with resolutions of 450 ps seen in silicon-based ring resonators [297], and 10 ps seen in nonlinear optical loop mirrors (NOLM) [298]. The NOLM approach also uses the optical Kerr effect; however, it does not rotate the polarization of the signal, instead it induces a phase shift which re-routes signal photons to a different fiber beam splitter output port. Another approach uses plasma mirrors to achieve sub-picosecond all-optical gating [299]; however, this technique is limited to the gating of high-power lasers. In the context of modulating quantum signals, the all-optical Kerr gate can modulate a signal without depending on its intensity, is capable of high-efficiency operation, and is flexible in its configuration. For these reasons, we use optical Kerr gating in our ultrafast approach to photonic quantum computation.

The capabilities of the optical Kerr gate are first characterized in the classical regime in Sections 2.3 and 2.4, then in the quantum regime in Section 3.5. Finally, the optical Kerr gate is employed for quantum information processing with one photon in Sections 4.4 and 4.5, and with up to three photons in Section 5.3.

	Resolution	Bandwidth	Insertion loss
Electro-optical approaches			
<i>Thorlabs</i> MEMS switch	0.5 ms	—	< 0.7 dB
<i>Thorlabs</i> high-speed optical shutter	1 ns	—	—
<i>BATi</i> ultrafast switch	< 60 ns	1 MHz	< 1 dB
<i>Agiltron</i> ultrafast switch	9 ns	2 MHz	< 1.5 dB
<i>EOSpace</i> intensity modulators	—	> 20 GHz	< 3.5 dB
<i>EOSpace</i> phase modulators	—	> 30 GHz	< 4 dB
<i>Electro Optical Components Inc.</i> KD*P Pockels Cells	< 250 ps	—	—
All-optical approaches			
Silicon ring resonators [297]	500 ps	—	3.5–10 dB
NOLMs [298]	180 ps	—	< 2.1 dB
Plasma mirrors [299]	< 1 ps	—	—
SFG-based gating [300]	< 1 ps	—	17 dB
Cascaded SHG and OPA [301]	~ 46 fs	< 9.6 THz	26.6 dB
Optical Kerr gating	< 1 ps	> 1 THz	< 0.8 dB

Table 2.3: **Comparison of optical modulation methods.** Selected state-of-the-art electro-optical and all-optical methods for modulating optical signals are compared on the basis of speed, bandwidth, and efficiency. This table does not constitute an exhaustive list. Only available specs are presented.

2.3 Configurable all-optical Kerr gating

This section is based on the following publication:

K. L. Fenwick, D. G. England, P. J. Bustard, J. M. Fraser, & B. J. Sussman, “Carving out configurable ultrafast pulses from a continuous wave source via the optical Kerr effect”, *Optics Express* **28**(17): 24845–24853 (2020).

DOI: <https://doi.org/10.1364/OE.399878>

© 2020 Optical Society of America

Previous demonstrations of optical Kerr gating²⁶ of single photons with efficiencies of 97% [258, 257] outline the promise of this technique for the manipulation and detection of quantum information encoded in a photon’s arrival time, on ultrafast timescales. Before we can employ optical Kerr gating for the manipulation and detection of ultrafast time bins, it must first be well-characterized. This is best done in the classical regime.

A key characterization metric is the intrinsic Kerr gate resolution, for which we demonstrate a simple, yet effective, measurement technique²⁷. Here, we refer to the intrinsic resolution as the Kerr gate window due only to the pump pulse as it propagates through the SMF with walk-off from the signal wavelength. When being used to gate signal pulses, the total Kerr gate resolution will then be a convolution of the intrinsic Kerr gate resolution and the temporal profile of the signal. If the signal is a CW beam of light at the wavelength of interest, then a measurement of the rotated signal light provides a measure of the intrinsic Kerr gate resolution.

We measure the spectral profile of the Kerr-gated light from the CW beam to infer the Kerr gate resolution. As the pump pulse propagates through the SMF, it “carves out” ultrashort pulses of polarization-rotated light from the CW beam, as seen in Fig. 2.12. As such, we can expect to observe spectra with expanded bandwidth

²⁶Note that throughout this thesis, I refer to this technique as optical Kerr *gating*; however, some publications (including that at the end of this section) refer to the technique as optical Kerr *switching*. The reader can take these terms to be interchangeable.

²⁷Early work towards this goal was performed during my MSc studies, during which I created an interactive *Mathematica* model for the Kerr gate, built the experimental setup, and collected preliminary results [302]. Final data collection and the preparation of the publication at the end of this section were performed during my PhD studies.

from the carved pulses, relative to the narrowband spectrum of the input CW beam. These bandwidth-expanded spectra are measured and compared to the fast Fourier transform (FFT) of their respective theoretical temporal intensity profiles, based on Eq. 2.120, for the relevant experimental parameters.

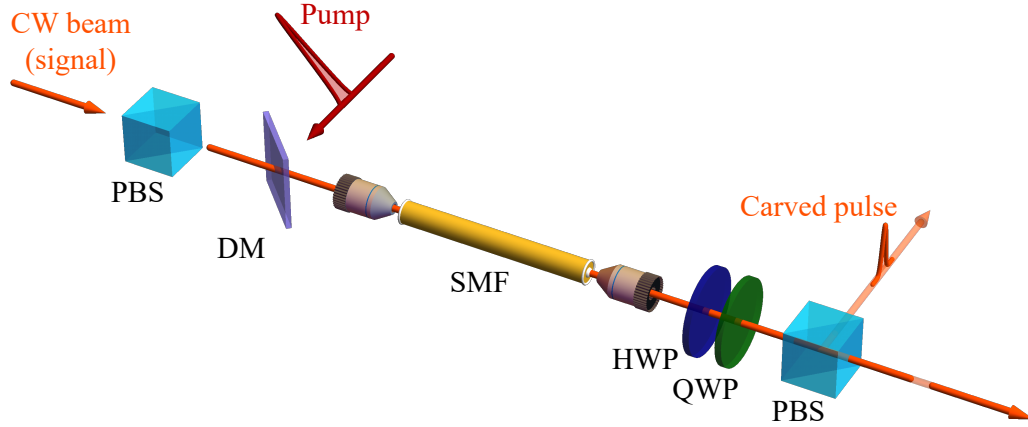


Figure 2.12: **Pulse carving with an optical Kerr gate.** The optical Kerr gate is used to carve pulses from a continuous wave (CW) input signal. The Kerr medium is a short piece of single-mode fiber (SMF), placed between two crossed polarizers. A strong pump pulse can be combined with the CW signal on a dichroic mirror (DM) before both beams are launched into the SMF. The pump pulse induces a birefringence in the SMF which then rotates the polarization of the signal. The carved pulses are redirected by a polarizing beam splitter (PBS). HWP: half-wave plate; QWP: quarter-wave plate.

It should be noted that the Kerr gate is not operated with transform-limited pump pulses. This has important consequences when defining the intrinsic Kerr gate resolution. If it were possible to generate transform-limited pulses of Kerr-gated light, we could simply take the inverse fast Fourier transform (IFFT) of the spectrum of polarization-rotated light—under the assumption of a constant phase in the signal’s electric field—to extract the Kerr gate temporal resolution; however, we cannot make this guarantee. In fact, the Kerr-gated pulses are almost certainly not perfectly transform-limited due to dispersion as they propagate in the fiber.

Despite this, we can still approximate the temporal resolution of the intrinsic Kerr gate resolution by comparing a FFT of the expected theoretical Kerr gate temporal

profile with the measured spectra of polarization-rotated signal light. It should be noted that chirp in the pump pulses does not induce chirp in the polarization-rotated signal pulses, due to the fact that the nonlinear optical effect being exploited is an intensity-dependent process. Furthermore, the effects of pump dispersion in the SMF are accounted for by taking the temporal FWHM of the pump pulses at the fiber output as the pump pulse duration parameter in the theoretical model. This is, of course, a simplification but we find it sufficient for our purposes.

We find that the measured spectra of Kerr-gated light are in good qualitative agreement with the FFT of our theoretical model. This implies that the pulses carved out from the CW beam are near transform-limited, which is pretty cool! This allows us to estimate the intrinsic Kerr gate resolution for a given set of experimental input parameters. Beyond this, we also study the dependence of Kerr gating as the pump pulse temporal profile is modulated. We demonstrate the carving out of tunable ultrashort pulses with configurable, sub-picosecond duration (305–570 fs). Furthermore, we demonstrate the carving of ultrafast time bins from the CW signal beam, by pumping the Kerr gate with various sequences of pump pulses²⁸.

Beyond characterizing the intrinsic Kerr gate resolution for a given set of experimental input parameters, we also show that the Kerr gate can be used to carve out configurable ultrashort pulse sequences. This is a promising capability in the context of optical communication and computing—in both the classical and quantum regimes. The carved pulses are time-locked to the pulsed beam pumping the process, which implies pulse-to-pulse phase stability over the coherence length of the input CW beam. The Kerr gate technique is straightforward in its implementation, and its fiber-based design makes it promising for full fiber integration (*i.e.*, with a gigahertz fiber oscillator pump). Furthermore, the exploited XPM process is intrinsically phase-matched for any wavelength, so that wavelength tuning is only restricted by waveguiding. This suggests easy adaption to various wavelengths, and scalability towards pulse generation at multiple different wavelengths, simultaneously. By exploiting XPM—often viewed as parasitic in optical telecom fiber—this work opens new application avenues, such as in time-gated microscopy and spectroscopy, or optical communications.

²⁸The birefringent α -BBO crystals used to generate these sequences are used later in our ultrafast time-bin encoding (UTBE) protocol!

Carving out configurable ultrafast pulses from a continuous wave source via the optical Kerr effect

KATE L. FENWICK,^{1,2,3} DUNCAN G. ENGLAND,^{2,*} PHILIP J. BUSTARD,² JAMES M. FRASER,³ AND BENJAMIN J. SUSSMAN^{1,2}

¹Department of Physics, University of Ottawa, Ottawa, Ontario K1N 6N5, Canada

²National Research Council of Canada, 100 Sussex Drive, Ottawa, Ontario K1A 0R6, Canada

³Department of Physics, Engineering Physics & Astronomy, Queen's University, Kingston, Ontario K7L 3N6, Canada

Abstract: Wavelength-tunable, time-locked pairs of ultrafast pulses are crucial in modern-day time-resolved measurements. We demonstrate a simple means of generating configurable optical pulse sequences: sub-picosecond pulses are carved out from a continuous wave laser via pump-induced optical Kerr switching in 10 cm of a commercial single-mode fiber. By introducing dispersion to the pump, the near transform-limited switched pulse duration is tuned between 305–570 fs. Two- and four-pulse signal trains are also generated by adding birefringent α -BBO plates in the pump beam. These results highlight an ultrafast light source with intrinsic timing stability and pulse-to-pulse phase coherence, where pulse generation could be adapted to wavelengths ranging from ultraviolet to infrared.

© 2020 Optical Society of America under the terms of the [OSA Open Access Publishing Agreement](#)

1. Introduction

Ultrafast optical technology is widespread, with a highly interdisciplinary application space including time-resolved spectroscopy [1], high-precision frequency metrology and the development of optical clocks [2], material processing and machining [3], high-speed electronic testing [4], biomedical imaging [5], and optical communications [6,7]. In time-resolved spectroscopy, *pump-probe* techniques are often used to resolve ultrafast dynamics that are too fast to be directly imaged with photodetectors. A standard pump-probe experiment utilizes one laser pulse (the *pump*) to drive the process of interest, and a second time-delayed pulse (the *probe*) to interrogate the dynamics. The pump and probe are often at different wavelengths, each depending on the system under investigation. Accordingly, generating customizable, time-locked pairs of ultrafast pulses is a crucial capability.

Often, the pump pulse is generated by a mode-locked laser and the spectrally tunable probe pulse is generated by nonlinear frequency mixing, for example in an optical parametric oscillator (OPO) or amplifier (OPA) pumped by the mode-locked laser. This is an established method for generating high-intensity probe pulses, but the bulk and expense of an OPO or OPA may be prohibitive in many cases. An alternative approach uses *all-optical switching* to carve a pulse train from a continuous-wave (CW) laser. This technique offers access to wavelengths achievable by any CW light source, including low-cost laser diodes and fiber lasers, which can be chosen appropriately for the application. Furthermore, the switching process can be driven by the pump laser to provide intrinsic timing stability. Such time-locked pulses can be generated near the pump wavelength, which would usually be challenging to achieve using an OPO or OPA. Switched pulses will necessarily be weak compared to those produced by femtosecond oscillators or nonlinear mixing, but in linear pump-probe measurements such as transient absorption or reflection, where only a differential measurement is made, high probe intensity is not required

and is often avoided [8–10]. All-optical switching of CW beams is therefore a promising option for simple, customizable ultrafast pulse generation. Picosecond-scale all-optical switching has been achieved via the optical Kerr effect [11,12], and has been demonstrated as an effective method for generating picosecond pulses in the telecommunications band around 1550 nm [13]. However, due to the length of fiber involved ($\sim 10 - 1000$ m) and the need to operate near zero dispersion, this technique is not easily implemented beyond the telecommunications band. With femtosecond lasers and short (~ 10 cm) fibers, picosecond scale all-optical switching of single photons is possible in the visible domain [14,15]. In this article, we exploit the same nonlinear process to carve light out from a CW beam, generating pulses on the hundreds-of-femtoseconds timescale, in the near-infrared.

The pulse generation technique uses a commercial mode-locked oscillator to actively induce polarization modulation in a CW laser source, by exploiting the optical Kerr effect in single-mode fiber (SMF). When the fiber is placed between two crossed-polarizers, a portion of the modulated light is transmitted, producing a sub-picosecond pulse train. This generates additional bandwidth about the narrow CW laser spectrum, which is indicative of the ultrafast switching mechanism and provides a means of characterizing the generated pulses. The method is straightforward in its implementation, where all system components are readily commercially available, and active stabilization is not necessary. Sub-nanojoule pump pulse energies are sufficient to induce the switching mechanism, falling within reach of commercial fiber oscillators, thus offering the potential for a fully integrated fiber-based optical switch. The device requires only 10 cm of conventional SMF which, combined with modest pump pulse energies, minimizes parasitic nonlinear processes such as stimulated Raman scattering, two-photon absorption, and self-phase modulation (SPM).

We achieve sequences of near transform-limited switched pulses with sub-picosecond duration, where the time-domain amplitude envelope of the pump pulse is effectively "mapped" from the carrier frequency of the pump to the signal. The switched pulse duration is tuned between 305–570 fs by introducing dispersion elements to the pump. Birefringent α -BBO plates are also added to the pump to generate trains of two and four switched pulses. Furthermore, the phase coherence of the generated pulses is expected to be time-locked for the coherence length of the CW laser. In addition to the promise of this device as an easily-implementable ultrafast light source, the demonstrations in this article underline its potential in a variety of applications including communications [16], biomedical imaging [17], microscopy [18], spectroscopy [19], and quantum optics [20].

2. Methods

Ultrafast all-optical switching relies on cross-phase modulation via the optical Kerr effect. The Kerr effect is a $\chi^{(3)}$ nonlinear process where a pump field generates an intensity-dependent refractive index modulation. The induced birefringence can be used to modulate the polarization of a signal beam. When the medium is situated between two crossed-polarizers, a portion of the modulated signal will be transmitted; this arrangement is an optical Kerr shutter [21]. For a linearly polarized pump pulse co-propagating with a CW signal beam of intensity I_0 , the switched light transmitted through the shutter will have an intensity profile [22]:

$$I(t) = I_0 \sin^2(2\theta) \sin^2\left(\frac{\Delta\phi(t)}{2}\right), \quad (1)$$

where θ is the angle between the signal and pump polarization and $\Delta\phi(t)$ is the time-dependent phase difference between the polarization parallel to the pump and orthogonal to the pump. Maximal switching can only occur when $\theta = \pi/4$, so we operate under this condition. When $\Delta\phi(t) \neq 0$, the medium acts as a birefringent waveplate and allows a portion of the signal to be

transmitted through the second polarizer. The time-dependent phase difference is given by [23]:

$$\Delta\phi(T) = \frac{8\pi n_2}{3\lambda_{\text{signal}}} \int_0^L I_{\text{pump}}(T - d_w z) dz, \quad (2)$$

where n_2 is the nonlinear refractive index of the fiber, λ_{signal} is the wavelength of the signal light, and z is the propagation distance within the fiber of length L . In a dispersive medium, the pump and signal will experience temporal walkoff, given by $d_w = v_{gp}^{-1} - v_{gs}^{-1}$, where v_{gp} and v_{gs} are the pump and signal group velocities, respectively. As such, the intensity profile of the pump, I_{pump} , is expressed in the frame moving with the signal [15]: $T = t - z/v_{gs}$, where t is time in the laboratory frame. The switched light intensity profile is thus determined by its wavelength, the pump pulse shape, and the temporal walkoff.

An experimental schematic is shown in Fig. 1. An optical Kerr shutter is employed, where a pulsed pump beam is used to carve out pulses from a CW signal beam within 10 cm of SMF. This short fiber length was selected to achieve ultrashort, near transform-limited sub-picosecond switched pulses. A longer fiber would increase pump-signal temporal walkoff and group delay dispersion in the switched pulses. On the other hand, a shorter fiber would require higher pump pulse energies.

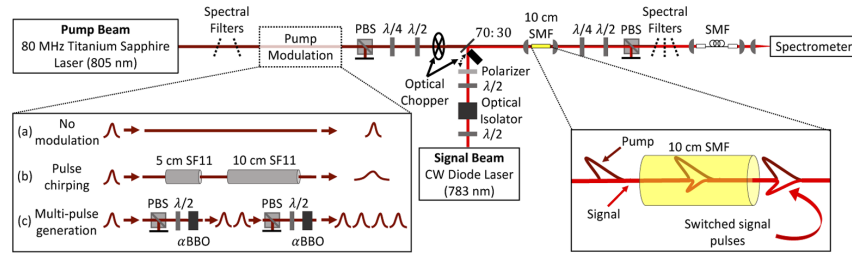


Fig. 1. Schematic diagram of the experimental setup. An 80 MHz Ti:Sapphire oscillator (pump beam) induces the optical Kerr effect in 10 cm of single-mode fiber (SMF). Shown in the bottom right inset, this mechanism carves out pulses from a continuous wave (CW) diode laser (signal beam), that are measured by a spectrometer. The pump polarization is prepared by a polarizing beamsplitter (PBS), and a quarter- ($\lambda/4$) and half-waveplate ($\lambda/2$). The signal polarization is prepared by an absorptive polarizer and $\lambda/2$ waveplate. Polarization projection of the switched light is achieved with $\lambda/4$ and $\lambda/2$ waveplates, and a PBS. Shown in the bottom left inset, pump pulses are either (a) not modulated, (b) chirped using various amounts of SF11 glass, or (c) split temporally into trains of 2 or 4 pulses using birefringent α -BBO.

The pump originates from an 80 MHz repetition rate Ti:Sapphire laser, which generates pulses of 10 nm bandwidth at a central wavelength of 805 nm. The pump is operated close to the signal spectral region to minimize temporal walkoff for sub-picosecond pulse carving. For this reason, SPM must be adequately minimized by spectral filtering of the pump prior to switching and restricting pump pulse energy. As such, the switch is operated at 30% efficiency, achieved using modest pulse energies of 0.56–1.3 nJ, depending on the pump pulse duration. After filtering, the pump pulse is measured (at the output of the SMF) to be 367 fs by frequency-resolved optical gating (FROG). As indicated by the bottom left inset in Fig. 1, the pump is prepared with either (a) no modulation, (b) temporal chirp, or (c) temporal separation into 2- and 4-pulse trains, to demonstrate the configurability of pulses generated by our switch. A polarizing beamsplitter (PBS) and a set of quarter- and half-waveplates ($\lambda/4$ and $\lambda/2$) are used to prepare the polarization state of the pump before it is combined with the signal.

The signal is generated by a 783 nm CW diode laser (Thorlabs CPS780S). An optical isolator (OI) is used to minimize any instability caused by back-reflection of the pump at the 10-cm SMF tip. In this configuration, the diode laser stability is sufficient for spectral measurement of the switched pulses on timescales of several minutes. A $\lambda/2$ waveplate and an absorptive polarizer prepare the initial polarization state of the signal: vertical with respect to the optical table (defined here as 0°). Both the pump and signal are chopped, at 4 Hz and 0.2 Hz respectively, for averaging and background subtraction purposes. The pump and signal are combined via a 70:30 beamsplitter and are then coupled into the 10-cm SMF (Thorlabs S630-HP, FC/PC connectors), with typical coupling efficiencies of 70% and 60%, respectively. Shown in the bottom right inset in Fig. 1, pump pulses induce birefringence while propagating through the 10-cm SMF, causing polarization-rotation in the CW signal where it temporally overlaps with the pump pulses. The output from this process is a train of pulses that have been rotated out from the initial CW signal. The switched pulses are polarization-filtered from the unswitched CW light using a PBS, which projects the signal onto a 90° (horizontal) polarization. A $\lambda/4$ and $\lambda/2$ waveplate combination are placed between the SMF and PBS to correct for small polarization changes due to the 10-cm SMF. With the pump beam blocked, these optics are tuned to minimize the CW light measured at the spectrometer. This ensures maximal transmission of the cross-polarized component of the switched light through the PBS. After polarization filtering, three angle-tuned spectral filters remove the remaining light from the pump, and the switched light is coupled into a 1-m SMF for spatial filtering of cladding modes which were not fully attenuated in the 10-cm SMF. The time-frequency relationship of optical pulses anticipates that the pulses carved from the CW signal will have larger spectral bandwidth than the narrowband input signal. Switched pulses are thus measured using a spectrometer.

Sufficient polarization extinction is required to overcome the short duty cycle of the switch. For perfect switching, a polarization extinction of $\sim 10 \text{ ns}/100 \text{ fs} = 10^5$ would ensure the intensities of the switched and unswitched (leakthrough) signal are on the same order of magnitude. Polarization extinction of $1.5 - 6 \times 10^4$ is achieved, causing some of the narrowband input light to leak through the second PBS. Small fluctuations in the polarization of light exiting the SMF are present due to changing air currents and vibrations, which leads to the range in measured polarization extinction values.

The integrated spectral intensity of the leakthrough is $\sim 75\times$ that of the switched signal pulse (see Fig. 2). Although this may seem insurmountable for many applications, the short duty-cycle of the switch ($\sim 300 \text{ fs} \times 80 \text{ MHz} = 2.4 \times 10^{-5}$) actually generates switched pulses with a peak intensity $\sim 3200\times$ higher than the leakthrough. Many options exist to reduce the effects of leakthrough including implementing polarizers with higher extinction ratio, increasing the repetition rate of the pump [12], employing active feedback control on the $\lambda/4$ waveplate after the SMF [24], performing nanosecond electro-optic modulation before the switch, electronic filtering using a fast detector, or even implementing a second switch. Here, background subtraction is employed to overcome the bright integrated spectral intensity of the leakthrough, where the signal and pump are periodically chopped while continuously collecting spectra. These spectra are sorted into four categories, averaged, and used to generate the switched spectrum:

$$I_{\text{switched}} = (I_1 - I_2) - (I_3 - I_4), \quad (3)$$

where I_1 , I_2 , I_3 , and I_4 correspond to spectral data respectively sorted into categories: (1) signal on, pump on; (2) signal on, pump off; (3) signal off, pump on; and (4) signal off, pump off. Despite improvements from this method, a variable leakthrough signature still obscures the $\sim 783.0 - 783.5 \text{ nm}$ region and is thus excluded from all switched spectra presented in this article.

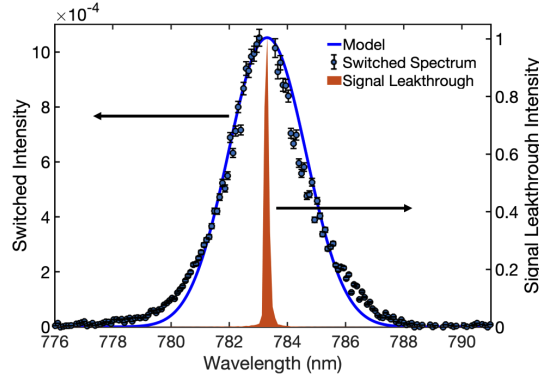


Fig. 2. Switched spectrum of generated pulses (blue data points) about the CW signal spectrum (shaded in orange) with 30% switching efficiency. Switched spectrum agrees with modelled spectrum (solid blue curve) for ~ 0.6 nJ pump pulses with 367 fs duration (measured via FROG at the 10-cm SMF output) and shows bandwidth expansion about the narrowband CW signal spectrum.

3. Results and discussion

The anticipated bandwidth expansion of the switched signal light (configuration (a) in pump modulation inset of Fig. 1) is shown in Fig. 2, where the switched spectrum is evidently broadened with respect to the signal leakage (input CW spectrum). Experimental data is compared with the fast-Fourier transform (FFT) of the temporal model (Eq. (1), assuming a Gaussian pump pulse of duration 367 fs) for verification and estimation of the generated pulse duration. The data and model are normalized relative to the signal leakage for comparison. The switched spectrum agrees with the modelled spectrum, though the model underestimates the intensity of the switched light at its tails. This is attributed to chirping of the pump pulse within the 10-cm SMF, which is not fully accounted for in the model. Given that the FFT of the model and the measured switched spectrum are in good agreement, it is inferred that the switched pulses are near transform-limited. For the experimental parameters associated with Fig. 2 ($\Delta\tau_{\text{pump}} = 367$ fs, $\lambda_{\text{pump}} = 805$ nm, $\lambda_{\text{signal}} = 783$ nm, $L_{\text{fiber}} = 10$ cm, and pump pulse energy 0.6 nJ) the model estimates a generated pulse duration of 305 fs. For 30% switching efficiency, this switching window leads to generated pulse energies of ~ 0.03 fJ.

With the demonstrated ability to carve out ultrafast pulses from a CW beam and a reliable method for estimating the generated pulse duration, pump pulse modulation is introduced to demonstrate customizability of generated pulses. First, chirp is introduced in the pump pulses to control the duration of generated pulses. Pump pulses are chirped by adding 5, 10, or 15 cm of SF11 glass to the pump beam path (see (b) in pump modulation inset in Fig. 1). The addition of SF11 glass stretches the pump pulses, measured via FROG at the 10-cm SMF output, from 415 fs (no SF11 glass) to 550 fs, 675 fs, or 735 fs (with 5, 10, or 15 cm SF11, respectively). Pump pulse energies are 0.56 nJ, 0.75 nJ, 0.94 nJ, and 1.3 nJ for 0, 5, 10, and 15 cm SF11 to maintain 30% switching efficiency. As anticipated by the time-frequency relationship, narrowing in the switched spectra is observed for increasing pump pulse duration. This is shown in Fig. 3, where spectral bandwidth tunability from 1.6–2.9 nm is demonstrated. By comparing measured switched spectra to an FFT of the corresponding temporal model, measurements of generated pulse duration are again estimated (provided in Table 1). With 0–15 cm of SF11 glass, the switch is capable of generating pulses with customizable duration from 330–570 fs, where generated

pulses are shorter than associated pump pulses due to the quadratic sinusoidal dependency of the switching profile in Eq. (1).

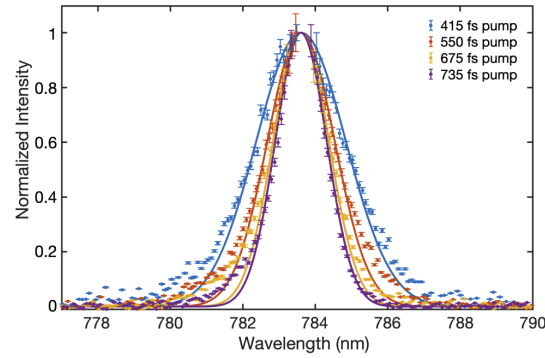


Fig. 3. Spectral bandwidth of generated pulses controlled with pump pulse duration. Switched spectra (coloured data points) agree with respective modelled spectra (matching coloured curve) for pump durations of 415 fs (blue), 550 fs (red), 675 fs (yellow), and 735 fs (purple).

Table 1. Modulation of generated pulse duration by chirping pump pulses (measured by FROG at SMF output) with SF11 glass.

SF11 Glass Added	Pump Pulse Duration (measured via FROG)	Generated Pulse Duration (estimated from model)
0 cm	415 fs	330 fs
5 cm	550 fs	435 fs
10 cm	675 fs	525 fs
15 cm	735 fs	570 fs

In addition to generating ultrafast pulses with tunable spectral bandwidth, sequences of two and four switched pulses are achieved by adding birefringent media (α -BBO crystals) to the pump beam path (see (c) in pump modulation inset in Fig. 1). Two-pulse trains are prepared using a single α -BBO crystal (with a thickness of either 5 or 10 mm), and a four-pulse train is prepared using two α -BBO crystals (of thickness 5 and 10 mm). The fast axis of the α -BBO crystals are oriented at 45° with respect to the pump polarization and components of a single pump pulse that are polarized along the fast and slow axes of a single α -BBO crystal are temporally separated into two pulses that are cross-polarized. The temporal separation is set by the thickness of the α -BBO crystal. Following each α -BBO crystal, the orthogonally polarized pulse pair is projected onto a single polarization by a PBS.

Pump pulse trains separated by some known delay, $\Delta\tau_{\text{delay}}$, carve out trains of switched pulses separated with the same delay. A train of switched pulses leads to spectral interference with fringe spacing: $\Delta\omega = 2\pi/\Delta\tau_{\text{delay}}$ [25,26]. Spectral interference in the switched spectra for three different pump pulse train sequences is measured (left column in Fig. 4) and converted to the frequency-domain using a linear interpolation, after which the IFFT can then be taken (right column in Fig. 4). The IFFT results show: (a) one peak at 2.10 ps, (b) one peak at 4.38 ps, and (c) three peaks at 2.13 ps, 4.36 ps, and 6.50 ps. These results are consistent with the generated delays predicted by the polarization-dependent group velocity of α -BBO: (a) two pulses separated by 2.15 ps, (b) two pulses separated by 4.30 ps, and (c) four pulses separated by 2.15 ps. The peak at

0 ps in each IFFT corresponds to the sum of the spectral profiles for all pulses present, and thus has a higher intensity than subsequent peaks corresponding to the interference [26]. These results demonstrate control over pulse sequence which, with tunable wavelength, duration, and temporal separation, could be useful, for example, in low-intensity Ramsey-comb spectroscopy [27,28].

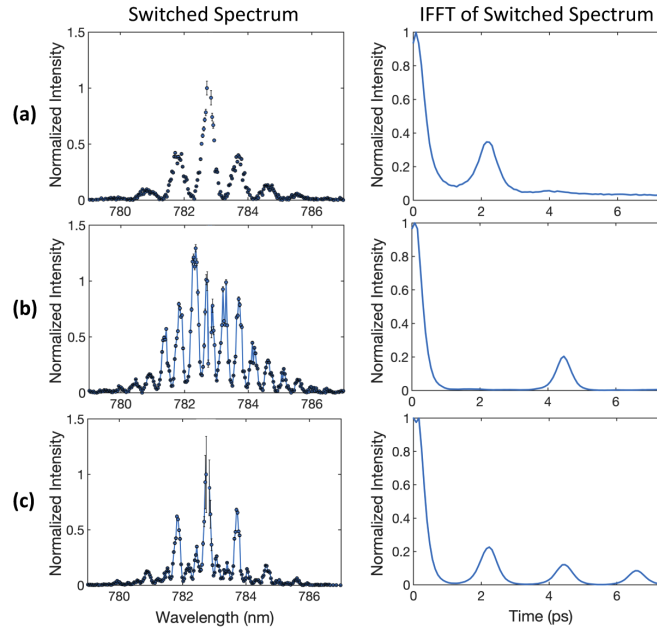


Fig. 4. Switched spectra (left) and respective IFFTs (right) for input pump trains of: (a) two pulses, separated by 2.15 ps; (b) two pulses, separated by 4.30 ps; and (c) four pulses, separated by 2.15 ps.

4. Conclusion

In summary, we demonstrated ultrafast pulse generation from a CW diode laser via the optical Kerr effect in only 10 cm of commercially available SMF. The device achieves temporally configurable pulses, and the technique provides a reliable means of switch speed characterization which can be extended to any all-optical switch [29–32]. Sub-nanojoule pump energies are sufficient and no active stabilization is required, making the implementation simple. Though switched pulse energies are low (sub-femtojoule), they are sufficient for transient pump-probe spectroscopy, and pulse characterization in the time-domain could be achieved using *temporal analysis, by dispersing a pair of light electric fields* (TADPOLE) [33]. It is expected that a range of CW sources could be implemented to achieve pulse generation over a wide range of wavelengths (i.e., 0.375–1.5 μm), noting that the signal, pump, and Kerr medium should be chosen appropriately for desired wavelength and duration of the generated pulses. The generated pulse duration is limited by the pump pulse duration and pump-signal walkoff in the Kerr medium; therefore, employing shorter pump pulses or exploiting dispersion engineering in photonic crystal fiber (PCF) could be used to generate shorter pulses. Another prospect for this device includes full fiber integration with, for example, a CW fiber laser input signal and a gigahertz fiber oscillator pump, which provides another means of reducing signal leakthrough. With some

modifications, this switching technique is anticipated to be beneficial in ultrafast time-gating applications including, for example, improving timing selectivity in both microscopy [34] and spectroscopy [35]. Beyond the classical realm, this is expected to be a useful tool in quantum optics, such as in the conversion of photonic qubits [14] and quantum metrology [36,37].

Funding

Natural Sciences and Engineering Research Council of Canada; University of Ottawa-NRC Joint Centre for Extreme Photonics.

Acknowledgments

The authors wish to thank Frédéric Bouchard, Khabat Heshami, Yingwen Zhang, Kent Bonsma-Fisher, Rune Lausten, Denis Guay, and Doug Moffatt for useful discussion and technical assistance.

Disclosures

The authors declare no conflicts of interest.

References

1. V. Blanchet, M. Z. Zgierski, T. Seideman, and A. Stolow, "Discerning vibronic molecular dynamics using time-resolved photoelectron spectroscopy," *Nature* **401**(6748), 52–54 (1999).
2. M. Takamoto, F.-L. Hong, R. Higashi, and H. Katori, "An optical lattice clock," *Nature* **435**(7040), 321–324 (2005).
3. K. M. Davis, K. Miura, N. Sugimoto, and K. Hirao, "Writing waveguides in glass with a femtosecond laser," *Opt. Lett.* **21**(21), 1729–1731 (1996).
4. A. Y. Elezzabi and M. R. Freeman, "Ultrafast magneto-optic sampling of picosecond current pulses," *Appl. Phys. Lett.* **68**(25), 3546–3548 (1996).
5. A. Adhikari, J. K. Eliason, J. Sun, R. Bose, D. J. Flannigan, and O. F. Mohammed, "Four-dimensional ultrafast electron microscopy: Insights into an emerging technique," *ACS Appl. Mater. Interfaces* **9**(1), 3–16 (2017).
6. H. C. H. Mulvad, M. Galili, L. K. Oxenløwe, H. Hu, A. T. Clausen, J. B. Jensen, C. Peucheret, and P. Jeppesen, "Demonstration of 5.1 Tbit/s data capacity on a single-wavelength channel," *Opt. Express* **18**(2), 1438–1443 (2010).
7. T. Morioka, K. Uchiyama, S. Kawanishi, S. Suzuki, and M. Saruwatari, "Multiwavelength picosecond pulse source with low jitter and high optical frequency stability based on 200 nm supercontinuum filtering," *Electron. Lett.* **31**(13), 1064–1066 (1995).
8. M. Wesseli, C. Ruppert, S. Trumm, H. J. Krenner, J. J. Finley, and M. Betz, "Nonlinear optical response of a single self-assembled InGaAs quantum dot: A femtojoule pump-probe experiment," *Appl. Phys. Lett.* **88**(20), 203110 (2006).
9. M. Namboodiri, T. Khan, K. Karki, M. M. Kazemi, S. Bom, G. Flachenecker, V. Namboodiri, and A. Materny, "Nonlinear spectroscopy in the near-field: time resolved spectroscopy and subwavelength resolution non-invasive imaging," *Nanophotonics* **3**(1-2), 61–73 (2014).
10. K. Karki, M. Namboodiri, T. Zeb Khan, and A. Materny, "Pump-probe scanning near field optical microscopy: Sub-wavelength resolution chemical imaging and ultrafast local dynamics," *Appl. Phys. Lett.* **100**(15), 153103 (2012).
11. N. A. Whitaker, H. Avramopoulos, P. M. W. French, M. C. Gabriel, R. E. LaMarche, D. J. D. Giovanni, and H. M. Presby, "All-optical arbitrary demultiplexing at 2.5 Gbits/s with tolerance to timing jitter," *Opt. Lett.* **16**(23), 1838–1840 (1991).
12. E. Yamada, K. Suzuki, and M. Nakazawa, "Subpicosecond optical demultiplexing at 10 GHz with zero-dispersion, dispersion-flattened, nonlinear fibre loop mirror controlled by 500 fs gain-switched laser diode," *Electron. Lett.* **30**(23), 1966–1968 (1994).
13. L. Moller, Y. Su, X. Liu, J. Leuthold, and C. Xie, "Ultrahigh-speed optical phase correlated data signals," *IEEE Photonics Technol. Lett.* **15**(11), 1597–1599 (2003).
14. C. Kupchak, P. J. Bustard, K. Heshami, J. Erskine, M. Spanner, D. G. England, and B. J. Sussman, "Time-bin-to-polarization conversion of ultrafast photonic qubits," *Phys. Rev. A* **96**(5), 053812 (2017).
15. C. Kupchak, J. Erskine, D. England, and B. Sussman, "Terahertz-bandwidth switching of heralded single photons," *Opt. Lett.* **44**(6), 1427–1430 (2019).
16. S. Matsuo, A. Shinya, T. Kakitsuka, K. Nozaki, T. Segawa, T. Sato, Y. Kawaguchi, and M. Notomi, "High-speed ultracompact buried heterostructure photonic-crystal laser with 13 fJ of energy consumed per bit transmitted," *Nat. Photonics* **4**(9), 648–654 (2010).
17. S. Andersson-Engels, R. Berg, S. Svanberg, and O. Jarlman, "Time-resolved transillumination for medical diagnostics," *Opt. Lett.* **15**(21), 1179–1181 (1990).

18. G. Vicidomini, G. Moneron, K. Y. Han, V. Westphal, H. Ta, M. Reuss, J. Engelhardt, C. Eggeling, and S. W. Hell, "Sharper low-power STED nanoscopy by time gating," *Nat. Methods* **8**(7), 571–573 (2011).
19. P. Matousek, M. Towrie, A. Stanley, and A. W. Parker, "Efficient rejection of fluorescence from Raman spectra using picosecond Kerr gating," *Appl. Spectrosc.* **53**(12), 1485–1489 (1999).
20. R. Prevedel, P. Walther, F. Tiefenbacher, P. Böhi, R. Kaltenbaek, T. Jennewein, and A. Zeilinger, "High-speed linear optics quantum computing using active feed-forward," *Nature* **445**(7123), 65–69 (2007).
21. J. Etchepare, G. Grillon, R. Muller, and A. Orszag, "Kinetics of optical Kerr effect induced by picosecond laser pulses," *Opt. Commun.* **34**(2), 269–272 (1980).
22. H. Kanbara, H. Kobayashi, T. Kaino, T. Kurihara, N. Ooba, and K. Kubodera, "Highly efficient ultrafast optical Kerr shutters with the use of organic nonlinear materials," *J. Opt. Soc. Am. B* **11**(11), 2216–2223 (1994).
23. G. Agrawal, *Applications of nonlinear fiber optics* (Academic Press, 2001).
24. A. V. Kuhlmann, J. Houel, D. Brunner, A. Ludwig, D. Reuter, A. D. Wieck, and R. J. Warburton, "A dark-field microscope for background-free detection of resonance fluorescence from single semiconductor quantum dots operating in a set-and-forget mode," *Rev. Sci. Instrum.* **84**(7), 073905 (2013).
25. C. Iaconis and I. A. Walmsley, "Self-referencing spectral interferometry for measuring ultrashort optical pulses," *IEEE J. Quantum Electron.* **35**(4), 501–509 (1999).
26. L. Lepetit, G. Chériaux, and M. Joffe, "Linear techniques of phase measurement by femtosecond spectral interferometry for applications in spectroscopy," *J. Opt. Soc. Am. B* **12**(12), 2467–2474 (1995).
27. J. Morgenweg, I. Barmes, and K. S. E. Eikema, "Ramsey-comb spectroscopy with intense ultrashort laser pulses," *Nat. Phys.* **10**(1), 30–33 (2014).
28. L. S. Dreissen, C. Roth, E. L. Gründeman, J. J. Krauth, M. Favier, and K. S. E. Eikema, "High-precision Ramsey-comb spectroscopy based on high-harmonic generation," *Phys. Rev. Lett.* **123**(14), 143001 (2019).
29. H. Lu, X. Liu, L. Wang, Y. Gong, and D. Mao, "Ultrafast all-optical switching in nanoplasmonic waveguide with Kerr nonlinear resonator," *Opt. Express* **19**(4), 2910–2915 (2011).
30. K. Nozaki, T. Tanabe, A. Shinya, S. Matsuo, T. Sato, H. Taniyama, and M. Notomi, "Sub-femtojoule all-optical switching using a photonic-crystal nanocavity," *Nat. Photonics* **4**(7), 477–483 (2010).
31. Z.-G. Zang and W.-x. Yang, "Theoretical and experimental investigation of all-optical switching based on cascaded LPPGs separated by an erbium-doped fiber," *J. Appl. Phys.* **109**(10), 103106 (2011).
32. W. Yoshiki and T. Tanabe, "All-optical switching using Kerr effect in a silica toroid microcavity," *Opt. Express* **22**(20), 24332–24341 (2014).
33. D. N. Fittinghoff, J. L. Bowie, J. N. Sweetser, R. T. Jennings, M. A. Krumbügel, K. W. DeLong, R. Trebino, and I. A. Walmsley, "Measurement of the intensity and phase of ultraweak, ultrashort laser pulses," *Opt. Lett.* **21**(12), 884–886 (1996).
34. J. C. Blake, J. Nieto-Pescador, Z. Li, and L. Gundlach, "Ultraviolet femtosecond Kerr-gated wide-field fluorescence microscopy," *Opt. Lett.* **41**(11), 2462–2465 (2016).
35. S. Draxler and M. E. Lippitsch, "Time-resolved fluorescence spectroscopy for chemical sensors," *Appl. Opt.* **35**(21), 4117–4123 (1996).
36. J. M. Donohue, M. Agnew, J. Lavoie, and K. J. Resch, "Coherent ultrafast measurement of time-bin encoded photons," *Phys. Rev. Lett.* **111**(15), 153602 (2013).
37. J.-P. W. MacLean, J. M. Donohue, and K. J. Resch, "Direct characterization of ultrafast energy-time entangled photon pairs," *Phys. Rev. Lett.* **120**(5), 053601 (2018).

2.4 Ultrafast spectro-temporal modulation

This section is based on the following publication:

K. L. Fenwick, G. S. Thekkadath, P. J. Bustard, D. England, F. Bouchard, and B. Sussman, “Time-of-flight spectroscopy with ultrafast all-optical gating”, *Optics Express* **33**(3): 5914–5922 (2025).

DOI: <https://doi.org/10.1364/OE.549697>

© 2025 Optica Publishing Group

In the journey towards using ultrafast time-bin encoding (UTBE) for photonic quantum computing, we came across a fun (and informative) “side quest”, which provided us with a deeper understanding of the optical Kerr gate’s capabilities. Given the high temporal resolution of the Kerr gate, we decided to explore the possibility of translating this high resolution to the conjugate (frequency) domain. Again, we work in the classical domain for simplicity, but note that the results presented here can be extended to the quantum domain.

In order to achieve this, we require a means of “mapping” the spectral content of the signal we wish to Kerr gate to the time domain for measurement or manipulation. The photonic time stretch (also called the time-of-flight spectrometer) is a well-known technique which does just this [303]. This technique makes use of the dispersive Fourier transform, in which a highly dispersive medium²⁹ is used to map the spectrum of an ultrashort input signal to the temporal envelope of its output [304, 305, 306]. The time-stretched pulse is then measured by a high-speed photodetector, where the time-dependent intensity profile informs the spectrum.

Although recent advancements in single-photon detection have seen increasingly precise timing resolution, with cutting-edge superconducting nanowire single-photon detectors (SNSPDs) now capable of sub-10 ps resolution [11, 307], there is always room for improvement. Optical Kerr gating provides a potential pathway for improved timing resolution of the photonic time-stretch measurement, which we explore here. As shown in Fig. 2.13, an ultrashort signal is chirped by ~ 1 km of polarization-maintaining fiber (PMF), to generate a frequency-to-time mapped (chirped) signal. A specific frequency band of the chirped signal can then be re-routed (*e.g.*, to a detector for measurement) by the Kerr gate.

²⁹The dispersion of ultrashort signals is explained in Section 2.1.2.

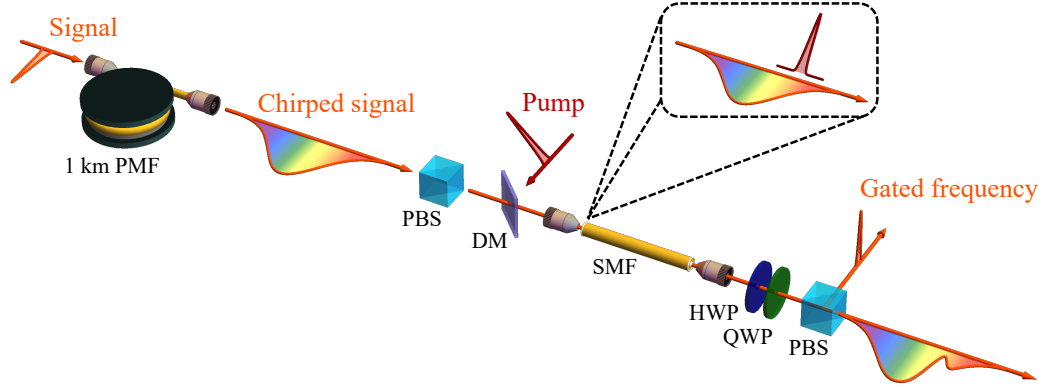


Figure 2.13: **Spectral sampling with an optical Kerr gate.** The optical Kerr gate is used to measure the spectrum of an ultrashort input signal which has been chirped by 1 km of polarization-maintaining fiber (PMF). The Kerr medium is a short piece of single-mode fiber (SMF), placed between two crossed polarizers. A strong pump pulse can be combined with the chirped signal on a dichroic mirror (DM) before both beams are launched into the SMF. The pump pulse induces a birefringence in the SMF which then rotates the polarization of the particular frequency band of the signal. This frequency band is redirected by a polarizing beam splitter (PBS). HWP: half-wave plate; QWP: quarter-wave plate.

It should be noted that the relative arrival time of the chirped signal and pump pulse at the Kerr gate SMF is important in this application. As such, fluctuations in the 1 km PMF length must be mitigated on measurement timescales. We initially observed that signal pulse arrival time drifted between 13 to 53 ps/K in the laboratory environment. This can be attributed to thermal expansion of the optical glass in the PMF, and is consistent with the range of values for the thermal expansion coefficient of optical glass provided in [308]. To slow this drift in signal arrival time, the 1 km PMF is housed in a matryoshka-style temperature-stable box with aluminum heat sinks³⁰, shown in Fig. 2.14. With this addition to the experimental setup, effects due to temperature drift are slower than data collection timescales.

Although the Kerr gate defines an absolute minimum frequency resolution, it is important to realize that the resolution of the frequency-to-time mapping technique

³⁰Note that this could be improved upon by adding material with higher heat capacity. Aluminum has a heat capacity of 887 J/kg/K, while water has a heat capacity of 4179.6 J/kg/K. Note that adding beer was also discussed, which has a heat capacity of ~ 3600 J/kg/K, depending on the mash.

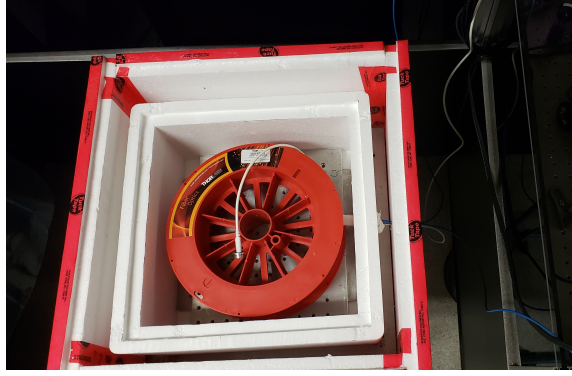


Figure 2.14: **Temperature-stable box housing the 1 km PMF.** The spool of PMF is placed within two styrofoam boxes. Two thick plates of aluminum are placed within the innermost box, and one thick aluminum plate is placed within the outermost box—these act as heat sinks. With this design, thermal expansion effects due to temperature drifts in the lab become slower than the timescale of measurement.

is not only limited by the detector resolution, but also by the TBP of the input signal. Described in detail in the manuscript and supplemental material provided at the end of this section, the input signal duration also influences the achievable resolution. We demonstrate a situation in which the technique is limited by the frequency-to-time mapping requirement, as opposed to the Kerr gate resolution. It should be noted that despite being outside the “direct” frequency-to-time mapping regime, we are still able to measure partially frequency-to-time mapped signals with the Kerr gate; this can be thought of as a fractional Fourier transform.

With input signals that do not limit the frequency-to-time mapping resolution (*e.g.*, a single photon generated by SPDC or SFWM), we can anticipate that frequency modulation or measurement will be limited by the Kerr gate resolution. For the 1 km PMF used in this thesis, and a ~ 350 fs Kerr gate resolution as seen in Section 2.3, the anticipated resolution in the spectral domain is a few picometers. This technique is demonstrably useful for characterizing optical signals with high resolution. Furthermore, the capabilities we demonstrate here are promising in the context of frequency demultiplexing in both classical and quantum optical communication, as well as in frequency bin manipulation and measurement in quantum information processing.

Time-of-flight spectroscopy with ultrafast all-optical gating

KATE L. FENWICK,^{1,2,*}  GUILLAUME S. THEKKADATH,²  PHILIP J. BUSTARD,²  DUNCAN ENGLAND,²  FRÉDÉRIC BOUCHARD,² AND BENJAMIN SUSSMAN^{1,2}

¹Department of Physics, University of Ottawa, Advanced Research Complex, 25 Templeton Street, Ottawa ON Canada, K1N 6N5, Canada

²National Research Council of Canada, 100 Sussex Drive, Ottawa, Ontario K1A 0R6, Canada

Abstract: Spectral measurement is crucial in applications ranging from the investigation of matter and its electronic properties to the wavelength-multiplexed routing of optical signals. We propose and demonstrate a spectral measurement technique based on an all-optical approach. The signal spectrum is mapped to the time domain by dispersion in 1 km of fiber and is then optically gated by an intense ultrafast pulse in 10 cm of single-mode fiber. A portion of the signal spectrum can be recovered by sweeping the gate pulse through the stretched signal. Spectral fringes are measured down to the minimum bound for frequency-to-time mapping of 51.5 GHz. Our measurement technique expands the toolbox of ultrafast measurements from the time domain to the frequency domain.

© 2025 Optica Publishing Group under the terms of the [Optica Open Access Publishing Agreement](#)

1. Introduction

Spectral measurement of light is one of the most fundamental exploratory tools in astronomy [1], chemistry [2], and physics [3]. Measuring the spectrum resulting from light-matter interactions provides information on the composition and structure of matter, from atomic [4] to astronomical [5] scales. Not only is spectral measurement crucial for understanding material properties, it is also important for characterizing optical signals, such as ultrafast pulses [6,7] or single photons [8,9]. In many cases, high spectral resolution is required, for example when characterizing optical frequency combs [10] or spectral fringes [11,12], or in optical communication where different wavelengths of light can be used to send multiple signals at once over a fiber optic communication channel (*i.e.*, wavelength-division multiplexing) [13]. Frequency-to-space mapping in a grating spectrometer is perhaps the most common method used for spectral measurement; however, numerous techniques exist. Some techniques do not even require direct measurement in the frequency-domain, allowing for high-resolution capabilities in other domains (*e.g.*, time) to be exploited, as we demonstrate here.

Fourier transform spectroscopy, for example, measures interference in the time-domain to build optical spectrograms [14]. Another time-domain implementation, which has attracted attention in the last decade, is time-of-flight spectroscopy [15,16]. This technique uses time-domain “diffraction”, where a highly dispersive medium maps the spectrum of an input ultrafast pulse to the temporal envelope of its output [17–19]. The stretched pulse is then measured by a high-speed photodetector, where the time-dependent intensity informs the spectrum [20–22]. At the single-photon level, frequency-to-time mapping has been implemented with dispersion compensating fiber [9] and chirped fiber Bragg gratings [8] to measure the spectra of single photons. Time-stretched single-photon spectra are monitored by fast single-photon counting modules, which demonstrates the potential for real-time measurement of multiple bits of information per single photon. Recent advancements in single-photon detection have led to increasingly precise timing

resolution and fewer dark counts. Superconducting nanowire single-photon detectors (SNSPDs) routinely achieve timing resolution around 100 ps, with cutting-edge systems reaching down to the sub-10 ps timescale [23,24]; but there is always room for improvement.

In particular, when detection bandwidth is limiting the resolution of a time-of-flight spectrometer, one might consider using nonlinear optical gating [7] to reach detection bandwidths beyond what can be achieved electronically [25]. Importantly, the gating should not introduce a prohibitive amount of noise in the signal, especially when this is a single photon [26]. Low-noise gating based on linear interferometric techniques can be used to overcome detection bandwidth limitations [27–31]. Nonlinear techniques can operate without the need for an interferometer, and have been demonstrated using upconversion as the gating mechanism [32], but this method inherently requires a frequency conversion and is limited in terms of efficiency. Another promising all-optical approach—which has already demonstrated broad applicability in ultrafast pulse shaping [33], characterization of energy-time entangled photons [34], quantum communications [35], quantum simulations [36], and quantum information processing [37]—uses optical Kerr gating. By extending this optical Kerr gating technique to the spectral domain via frequency-to-time mapping, we anticipate a scheme that could pave the way for new advancements in spectral characterization and manipulation.

Here, we propose and demonstrate a spectral measurement technique that uses ultrafast optical Kerr gating of a frequency-to-time mapped signal. Outlined in Fig. 1, group velocity dispersion (GVD) in 1 km of fiber is used to stretch an input signal pulse, effectively mapping its spectrum to the time-domain. This time-stretched signal is then gated by an ultrafast pump pulse through the optical Kerr effect in 10 cm of single-mode fiber (SMF), depicted in the inset of Fig. 1. The pump pulse overlaps with a narrow window of the stretched signal pulse in time, and thus also a

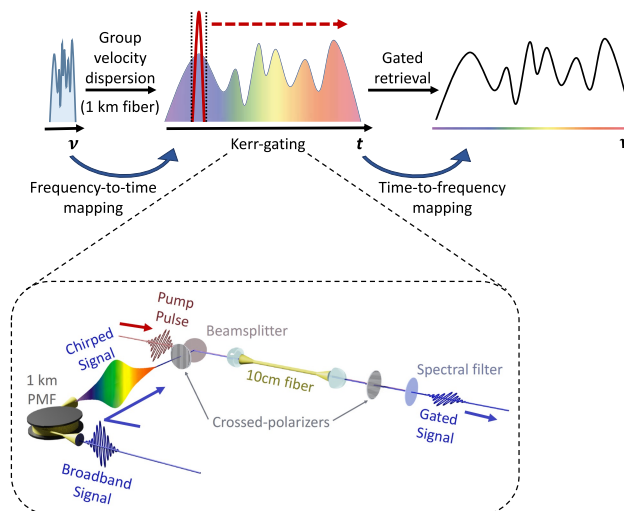


Fig. 1. Operating principle for Kerr-gated spectral measurement. A dispersive medium (such as a long optical fiber) is used to stretch an optical signal of interest and thus map its spectrum to the time-domain. Kerr-gating, shown in the inset, uses an intense ultrafast pump pulse to rotate the polarization of a narrow temporal window in the dispersed signal pulse. The gated signal is measured for various delay between signal and pump, and can be used to retrieve the optical spectrum.



narrow frequency band. This induces polarization rotation in the signal within the window of overlap. When the SMF is placed between two crossed-polarizers, only the polarization-rotated signal is transmitted, thus sampling a narrow frequency band for detection. This technique offers tunability in its operating band and the potential for full fiber-integration.

2. Theoretical requirements

We first consider the requirements of implementing frequency-to-time mapping. The output field, $E_{\text{out}}(t)$, for a pulse propagating through group delay dispersion, $\beta_2 z$ (where z is the length of the dispersive medium with quadratic dispersion β_2), is given by the convolution of the input field, $E_{\text{in}}(t)$, with a quadratic phase function: $E_{\text{out}} \sim E_{\text{in}} * e^{it^2/2\beta_2 z}$ (see [Supplement 1](#) for more details); this result is isomorphic with that for Fresnel diffraction in one dimension. In analogy to the spatial domain, the longest temporal feature duration of the input pulse, τ_0 , determines the dispersion required to be in the “far field”, where each spectral component is mapped to a different time in the output pulse. This condition is met when $|\beta_2 z| \gg \tau_0^2/8$, in which case the time-stretched output pulse is a scaled version of its input spectrum:

$$E_{\text{out}}(t) = \sqrt{\frac{-i}{2\pi\beta_2 z}} e^{it^2/2\beta_2 z} e^{i\omega_0 t} \tilde{E}_{\text{in}}(\omega = \omega_0 + \frac{t}{\beta_2 z}), \quad (1)$$

where angular frequency is given by $\omega = 2\pi f$ and $\omega_0 = 2\pi f_0$ is the carrier frequency of the pulse. Frequency, f , is thus mapped on to time with a mapping factor of [38],

$$2\pi \frac{\partial f}{\partial t} = \frac{1}{\beta_2 z}. \quad (2)$$

Given that the finest spectral feature, δf , and its duration are related by the time-bandwidth product, $\tau_0 \delta f \sim 1$, in order to obtain an accurate spectrum, the finest spectral feature, δf , must satisfy [38]

$$\delta f \gg \frac{1}{\sqrt{8|\beta_2 z|}}. \quad (3)$$

We can use this to estimate a minimum bound, δf_{min} , for frequency-to-time mapping with a given group delay dispersion,

$$\delta f_{\text{min}} = \frac{1}{\sqrt{8|\beta_2 z|}}, \quad (4)$$

with the understanding that spectral distortions will increase as δf_{min} is approached. In many cases, the spectral measurement resolution will be limited by the detector timing resolution. For a detector with timing resolution, $\Delta\tau_D$, the corresponding frequency bandwidth is given by

$$\Delta f_D = \frac{\Delta\tau_D}{2\pi|\beta_2 z|}. \quad (5)$$

It is commonly appreciated that the resolution of the frequency-to-time mapping technique is determined by the ratio of the detector (or gate) speed to the dispersion; however, care must be taken to ensure that Eq. (3) is satisfied, or the measured trace will include distortions. In order to accurately reconstruct the spectrum for a given dispersion, $\Delta f_D \lesssim \delta f_{\text{min}}$ should be met, placing a limit on the necessary detector resolution. For more details on the theory of frequency-to-time mapping, see [Supplement 1](#).

3. Experimental implementation

The experimental setup for the Kerr-gate frequency sampler is shown in Fig. 2. Both the signal and pump originate from an 80 MHz titanium sapphire (Ti:Sapph) laser, centered at 816 nm with a spectral bandwidth of 12 nm. The output pulses are split into two paths by a half-wave plate ($\lambda/2$) and polarizing beam splitter (PBS), which are used to control the pulse energy in the signal (reflected) path. The pulse energy in the pump (transmitted) path is controlled by a second $\lambda/2$ wave plate and PBS combination. Supercontinuum generation (SCG) in a 12 cm photonic crystal fiber (PCF) provides a signal pulse in a different spectral region than the pump, which is a requirement for operating the Kerr gate. The broadband light generated in the PCF is shown in Fig. 2(a). Pump pulses with energy ~ 0.055 nJ are used to generate the spectrum shown in blue, which is then spectrally filtered using a 5 nm bandpass filter (shown in red).

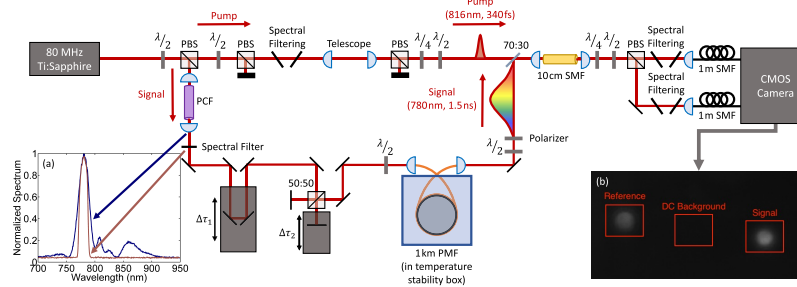


Fig. 2. Experimental setup. An 80 MHz Ti:Sapphire oscillator is split along two paths by a half-wave plate ($\lambda/2$) and polarizing beam splitter (PBS). The signal provides the spectrum to be gated, and the pump does the gating. Supercontinuum generation in a photonic crystal fiber (PCF) provides broadband signal light, depicted in (a), which is delayed with respect to the pump, $\Delta\tau_1$. A Michelson interferometer is used to prepare two signal pulses delayed by $\Delta\tau_2$, providing spectral fringes for measurement. 1 km of polarization maintaining fiber (PMF), housed in a temperature-stability box, provides the frequency-to-time mapping of the signal. The $\lambda/2$ wave plate prior to the 1 km PMF rotates the signal polarization such that it is aligned to the slow axis of the PMF. The time-stretched signal polarization is prepared by a $\lambda/2$ wave plate and absorptive polarizer. The pump polarization is prepared by a polarizing beam splitter (PBS) and a quarter- ($\lambda/4$) and half- ($\lambda/2$) wave plate. The pump and signal are recombined by a 70:30 (70% reflection, 30% transmission) beam splitter, and are then coupled into the 10 cm single mode fiber (SMF) for Kerr-gating. Only polarization-rotated light is allowed through the $\lambda/2$, $\lambda/4$, PBS combination after the SMF, to be measured by a CMOS camera. Shown in (b), the rejected light is also monitored on the CMOS camera, and allows for common-mode noise division where applicable.

The signal light is then sent through a variable delay line, $\Delta\tau_1$, to allow the control of the relative delay between the signal pulse and pump pulse. We use a Michelson interferometer with tunable delay $\Delta\tau_2$ between the two arms to impart a controllable spectral signature onto the signal. Pulses exiting the interferometer exhibit spectral fringes with frequency $f = 1/\Delta\tau_2$ [6,39]. After this, 1 km of polarization-maintaining fiber (PMF) disperses the signal pulses, with dispersion parameter $D = -146.1$ ps/nm/km (corresponding to quadratic dispersion $\beta_2 = 47.2$ ps²/km). The signal pulse energy prior to the 1 km PMF is 690 fJ. Signal pulse propagation time in the 1 km fiber can drift due to thermal expansion of the glass. Based on the range of values for the thermal expansion coefficient of optical glass [40], the signal pulse arrival time drift is expected to be between $\approx 13 - 53$ ps/K. To slow this drift, the 1 km PMF is housed in a temperature-stable box. Note that the $\lambda/2$ wave plate prior to the PMF is used to rotate the signal polarization such that it

couples to the slow axis of the PMF. After the PMF, the polarization of the time-stretched signal is then prepared by a $\lambda/2$ wave plate and nanoparticle polarizer, and recombined with the pump on a 70:30 beam splitter (70% of the signal is reflected, 30% of the pump is transmitted).

The stretched signal is then gated by the pump pulse within the 10 cm SMF (Thorlabs S630-HP). As outlined in [Supplement 1](#), it is important that the pump polarization is at 45° relative to the signal polarization for Kerr gating. The pump polarization is set by a PBS, quarter- ($\lambda/4$) wave plate, and $\lambda/2$ wave plate. The portion of the signal that is overlapped temporally with the pump will experience polarization rotation due to the Kerr effect. A PBS at the output of the 10 cm SMF allows only polarization-rotated light through to the detector. It is preceded by $\lambda/4$ and $\lambda/2$ wave plates, which compensate for any intrinsic birefringence within the 10 cm SMF. After the PBS, spectral filters are used to attenuate the pump, and a 1 m SMF is used to spatially filter any light allowed to propagate in the cladding modes of the short 10 cm SMF. The polarization rotated light (signal) is measured using a CMOS camera (Thorlabs, DCC1645C). The light reflected at the PBS can be measured simultaneously on the CMOS camera (reference), allowing for common-mode noise division, if desired. Note that, for the data presented here, we perform a background subtraction rather than make use of the common-mode noise division option; however, we include the option of measuring the reference signal in [Fig. 2](#) for completeness. For our background subtraction, we take measurements of the Kerr gated signal (both pump and signal on) and subtract the background noise due to the pump (pump on, signal blocked). The integrated DC background should be subtracted from both the integrated signal and reference spots on the camera in all cases.

The Kerr gate pump pulse can introduce noise photons in the signal spectral band due to competing parasitic nonlinear processes such as fluorescence, Raman scattering, self-phase modulation (SPM), and two-photon fluorescence [41], especially when the pump and signal are in similar spectral regions. We mitigate these noise sources by spectrally filtering the pump prior to Kerr gating and also reducing its power, the latter of which limits our gating efficiency to $\sim 30\%$. However, previous works have shown that the Kerr gate can reach near-unit efficiencies with low noise, when the signal and pump center wavelengths are well-separated [35,42].

4. Results and discussion

The setup was first operated without the Michelson interferometer to measure the dispersion parameter in the 1 km PMF. Instead of the CMOS camera, an Ocean Optics USB2000+ spectrometer was used to measure the spectrum of the gated signal. When the stretched signal is delayed relative to the gate (pump) pulse, the gate overlaps with different signal frequencies. As expected, this causes the central wavelength of the gated signal to shift with gate pulse delay. This is shown in [Fig. 3](#), where the slope of a linear fit allows for the experimental measurement of the dispersion parameter in the 1 km PMF. The experimental dispersion parameter was measured to be -146.1 ± 0.4 ps/nm (for ~ 1 km of fiber), which is close to the quoted dispersion parameter from Thorlabs of -150 ps/nm/km at 780 nm. With a measurement of dispersion, time (Kerr-gate delay $\Delta\tau_1$) can be mapped to frequency. For this amount of dispersion, the minimum bound for frequency-to-time mapping is $\delta f_{\min} = 51.5$ GHz, which corresponds to a required detector speed of 15.2 ps. Interestingly, since we operate with a sub-picosecond Kerr gate resolution, we are limited here by the amount of dispersion rather than the speed of our detection mechanism.

With the Michelson interferometer in the experimental setup, spectral fringes can be measured with the Kerr gate. By increasing the signal pulse separation, we can generate increasingly narrow spectral features down to the minimum bound for frequency-to-time mapping; for $\delta f_{\min} = 51.5$ GHz, this is given by $\Delta\tau_2 = 19.5$ ps. [Figures 4\(a\)–\(f\)](#) compare measured fringes with a model for pulse separation within the frequency-to-time mapping regime, while [Fig. 4\(g\)](#) presents the same comparison just beyond the limit of [Eq. \(4\)](#). The model follows the theory of frequency-to-time mapping presented in [Supplement 1](#), where the input signal is taken to be two

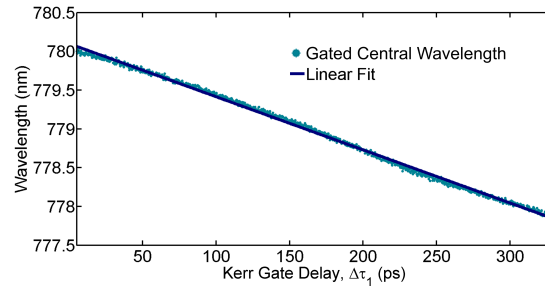


Fig. 3. Measuring the dispersion parameter. Central wavelength of polarization-rotated light is plotted against Kerr-gate delay (light blue). Over the small range of wavelengths measured, a linear fit (dark blue) can be used to extract the dispersion parameter of the 1 km fiber: -146.1 ± 0.4 ps/nm.

100 fs Gaussian pulses separated by a time delay of $\Delta\tau_2$. Note that $\Delta\tau_2$ is estimated based on the measured fringe spacing to account for any stage misalignment relative to the optical axis. The modeled temporal profile of the input signal is then converted to the frequency domain by a fast Fourier transform (FFT), after which a quadratic phase operator, associated with dispersion in the 1 km PMF, is multiplied. The result is then converted back to the time domain by inverse fast Fourier transform (IFFT), and a cross-correlation with the Kerr gate intensity profile (for $\lambda_{\text{signal}} = 780$ nm, $\lambda_{\text{pump}} = 816$ nm, and a pump pulse with 340 fs duration and energy of 0.69 nJ) is performed. This end result can then be compared with experimental measurements, as presented in Fig. 4. We observe good qualitative agreement between our experimental data and model.

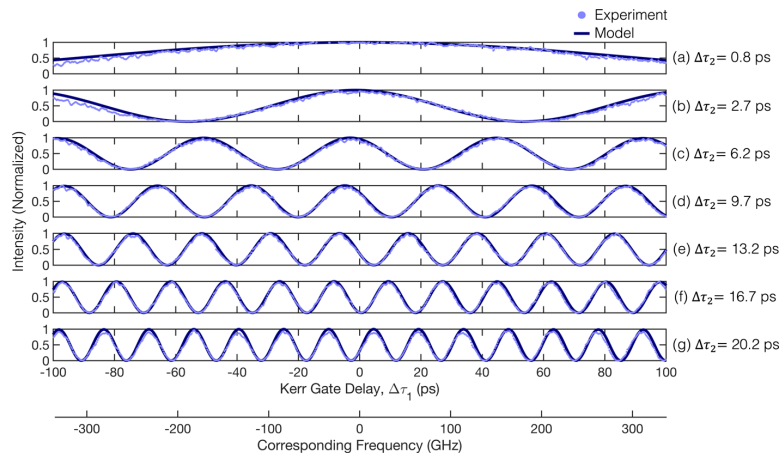


Fig. 4. Spectral fringes within and near the frequency-to-time mapping limit. Measured fringes (light purple) are compared with modeled fringes (dark blue) for various signal pulse delay, $\Delta\tau_2$. The respective frequency axis, calculated using Eq. (2), is displayed at the bottom for reference. Note that the data presented in (g) has spectral features smaller than the limit in Eq. (4), and is thus outside the frequency-to-time mapping regime.



The full measurement window is limited by the scanning range of the motorized delay line, $\Delta\tau_1$. Here, we are limited to a measurement spanning 200 ps, and thus only measure a portion of the signal spectrum; however, this window can be increased with the use of larger motorized linear delay stages. The acquisition time of a single data point (*i.e.*, the intensity of a Kerr-gated spectral component of the signal) is approximately 360 ms. Since we perform a measurement of both the gated signal (pump and signal on) and background noise due to the pump (pump on, signal off), the total acquisition time required for each spectrum presented in Fig. 4 takes about 9 minutes (2×4.5 minutes, to acquire both a signal and background). This total scan duration can be optimized to suit experimental needs, by adjusting the detector integration time and scanning resolution.

5. Conclusions and outlook

In summary, we have demonstrated a technique for time-of-flight spectroscopy using ultrafast optical Kerr gating. The spectrum of interest is first mapped to the time-domain by applying dispersion, after which an optically gated measurement is performed. Spectral fringes are measured down to the minimum bound for frequency-to-time mapping, with good agreement observed between experiment and model. Although time-of-flight spectroscopy is often limited by detector timing resolution, we stress the importance of recognizing that this is not always the case. Here, the detection resolution surpasses that of the minimum bound set by dispersion.

The high speed of ultrafast optical Kerr gating in fiber, combined with its potential for high-efficiency and low-noise operation [35,43], make this an attractive technique for operating on broadband single-photon-level signals. Indeed, optical Kerr gating has already seen application in fields such as time-resolved fluorescence emission spectroscopy [44–48], where spectral measurement of fluorescence can be performed without frequency-to-time mapping. Here, we explored the possibility of Kerr-gated measurement in the spectral domain, down to the estimated minimum resolution bound for frequency-to-time mapping, $\delta f_{\min} = 51.5$ GHz. For comparison, state-of-the-art time-of-flight spectrometers have seen approximately 213 GHz resolution in the 4.4–4.9 μm range [22], 90 GHz resolution at 1550 nm [9] 24 GHz resolution at 830 nm [8], and an impressive 1.9 GHz resolution around 1550 nm [21]. We anticipate that, with increased dispersion, the optical Kerr gate will be capable of higher-resolution measurement, competitive with state-of-the-art demonstrations. Obtaining sufficient dispersion to complement the high speed of the Kerr gate is a challenge; the temporal duration corresponding to the minimum frequency bound will also increase with dispersion. Furthermore, propagation in long fiber requires temperature stabilization. Chirped fiber Bragg gratings could provide one avenue for achieving higher group delay dispersion without the need for temperature stabilization; however, in this case, one must consider the trade-off between increased dispersion and loss. It is also important to weigh the time cost of performing a scanning measurement against the potential resolution benefits of the high-speed Kerr gate.

Nevertheless, intriguing possibilities remain: this technique could enable selective modulation or re-routing of optical signals based on frequency, for example, in frequency demultiplexing or for the manipulation of frequency-bins in quantum information processing schemes. With the implementation of multiple Kerr gates, frequency demultiplexing could be performed with minimal scanning. One may also envision combining time-bin and frequency-bin encoding in quantum information processing through this technique. Our approach could also be generalized to encompass spectro-temporal measurements; by adjusting the dispersion applied, one could apply selective phase and amplitude modulations to spectro-temporal modes. One could also explore the use of tomographic techniques [49] to better estimate the signal spectrum when the measurement resolution is limited by dispersion.

Funding. Natural Sciences and Engineering Research Council of Canada; Vanier Canada Graduate Scholarships (Vanier CGS) program.



Acknowledgments. We thank Aaron Goldberg, Alicia Sit, Andrew Proppe, Brayden Freitas, Denis Guay, Doug Moffatt, Kent Bonsma-Fisher, Khabat Heshami, Milica Banic, Nicholas Couture, Noah Lupu-Gladstein, Ramy Tannous, Rune Lausten, and Yingwen Zhang for their support and insightful discussions.

Disclosures. The authors declare no conflicts of interest.

Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

Supplemental document. See [Supplement 1](#) for supporting content.

References

1. J. B. Hearnshaw, *The analysis of starlight: one hundred and fifty years of astronomical spectroscopy* (CUP Archive, 1990).
2. J. Ingle and S. Crouch, *Prentice-Hall/Englewood Cliffs* (CUP Archive, 1988).
3. S. Dattagupta, *Relaxation phenomena in condensed matter physics* (Elsevier, 2012).
4. J. Stewart, J. Gaebler, and D. Jin, "Using photoemission spectroscopy to probe a strongly interacting Fermi gas," *Nature* **454**(7205), 744–747 (2008).
5. J. Angel, A. Cheng, and N. Woolf, "A space telescope for infrared spectroscopy of Earth-like planets," *Nature* **322**(6077), 341–343 (1986).
6. C. Iaconis and I. A. Walmsley, "Self-referencing spectral interferometry for measuring ultrashort optical pulses," *IEEE J. Quantum Electron.* **35**(4), 501–509 (1999).
7. D. J. Kane and R. Trebino, "Characterization of arbitrary femtosecond pulses using frequency-resolved optical gating," *IEEE J. Quantum Electron.* **29**(2), 571–579 (1993).
8. A. O. Davis, P. M. Saulnier, M. Karpiński, *et al.*, "Pulsed single-photon spectrometer by frequency-to-time mapping using chirped fiber bragg gratings," *Opt. Express* **25**(11), 12804–12811 (2017).
9. M. Avenhaus, A. Eckstein, P. J. Mosley, *et al.*, "Fiber-assisted single-photon spectrograph," *Opt. Lett.* **34**(18), 2873–2875 (2009).
10. A. Schliesser, N. Picqué, and T. W. Hänsch, "Mid-infrared frequency combs," *Nat. Photonics* **6**(7), 440–449 (2012).
11. J. Morgenweg, I. Barmes, and K. S. Eikema, "Ramsey-comb spectroscopy with intense ultrashort laser pulses," *Nat. Phys.* **10**(1), 30–33 (2014).
12. L. Dreissen, C. Roth, E. Gründeman, *et al.*, "High-precision Ramsey-comb spectroscopy based on high-harmonic generation," *Phys. Rev. Lett.* **123**(14), 143001 (2019).
13. A. Sharkawy, S. Shi, and D. W. Prather, "Multichannel wavelength division multiplexing with photonic crystals," *Appl. Opt.* **40**(14), 2247–2252 (2001).
14. F. Adler, P. Masłowski, A. Foltynowicz, *et al.*, "Mid-infrared Fourier transform spectroscopy with a broadband frequency comb," *Opt. Express* **18**(21), 21861–21872 (2010).
15. J. Chou, Y. Han, and B. Jalali, "Time-wavelength spectroscopy for chemical sensing," *IEEE Photonics Technol. Lett.* **16**(4), 1140–1142 (2004).
16. D. Solli, J. Chou, and B. Jalali, "Amplified wavelength-time transformation for real-time spectroscopy," *Nat. Photonics* **2**(1), 48–51 (2008).
17. Y. Tong, L. Chan, and H. Tsang, "Fibre dispersion or pulse spectrum measurement using a sampling oscilloscope," *Electron. Lett.* **33**(11), 983–985 (1997).
18. V. Torres-Company, J. Lancis, and P. Andres, "Space-time analogies in optics," *Prog. Opt.* **56**, 1–80 (2011).
19. K. Goda and B. Jalali, "Dispersive Fourier transformation for fast continuous single-shot measurements," *Nat. Photonics* **7**(2), 102–112 (2013).
20. A. Mahjoubfar, D. V. Churkin, S. Barland, *et al.*, "Time stretch and its applications," *Nat. Photonics* **11**(6), 341–351 (2017).
21. J. Mance, B. La Lone, J. Madajian, *et al.*, "Time-stretch spectroscopy for fast infrared absorption spectra of acetylene and hydroxyl radicals during combustion," *Opt. Express* **28**(20), 29004–29015 (2020).
22. A. Kawai, K. Hashimoto, T. Dougakiuchi, *et al.*, "Time-stretch infrared spectroscopy," *Commun. Phys.* **3**(1), 152 (2020).
23. I. E. Zadeh, J. W. Los, R. B. Gourgues, *et al.*, "Efficient single-photon detection with 7.7 ps time resolution for photon-correlation measurements," *ACS Photonics* **7**(7), 1780–1787 (2020).
24. B. Korzh, Q.-Y. Zhao, J. P. Allmaras, *et al.*, "Demonstration of sub-3 ps temporal resolution with a superconducting nanowire single-photon detector," *Nat. Photonics* **14**(4), 250–255 (2020).
25. B. Crockett, C. M. Rowe, and J. Azaña, "Ultrafast spectrogram with sub-THz bandwidth," in *2022 IEEE Photonics Conference (IPC)*, (IEEE, 2022), pp. 1–2.
26. O. Kuzucu, F. N. Wong, S. Kurimura, *et al.*, "Time-resolved single-photon detection by femtosecond upconversion," *Opt. Lett.* **33**(19), 2257–2259 (2008).
27. S. J. Soundararajan, L. Yang, S. Zhang, *et al.*, "Time-wavelength optical sampling spectroscopy based on dynamic laser cavity tuning," *J. Opt. Soc. Am. B* **35**(5), 1186–1191 (2018).
28. S. J. Soundararajan and L. Duan, "Time-stretch spectroscopy based on laser cavity tuning with a dual-function delay line," *IEEE Photonics Technol. Lett.* **31**(16), 1385–1388 (2019).



29. S. J. Soundararajan and L. Duan, "Real-time TWOS spectroscopy with sub-GHz resolution and high sensitivity," *Opt. Express* **29**(25), 41512–41521 (2021).
30. T. Hochrein, R. Wilk, M. Mei, *et al.*, "Optical sampling by laser cavity tuning," *Opt. Express* **18**(2), 1613–1617 (2010).
31. L. Yang, J. Nie, and L. Duan, "Dynamic optical sampling by cavity tuning and its application in lidar," *Opt. Express* **21**(3), 3850–3860 (2013).
32. Y. Cai, Y. Chen, K. Dorfman, *et al.*, "Mid-infrared single-photon upconversion spectroscopy enabled by nonlocal wavelength-to-time mapping," *Sci. Adv.* **10**(16), ead13503 (2024).
33. K. L. Fenwick, D. G. England, P. J. Bustard, *et al.*, "Carving out configurable ultrafast pulses from a continuous wave source via the optical Kerr effect," *Opt. Express* **28**(17), 24845–24853 (2020).
34. A. R. Cameron, K. L. Fenwick, S. W. Cheng, *et al.*, "Ultrafast measurement of energy-time entanglement with an optical Kerr shutter," *Phys. Rev. A* **108**(4), L041503 (2023).
35. F. Bouchard, D. England, P. J. Bustard, *et al.*, "Achieving ultimate noise tolerance in quantum communication," *Phys. Rev. Appl.* **15**(2), 024027 (2021).
36. K. L. Fenwick, F. Bouchard, G. S. Thekkadath, *et al.*, "Photonic quantum walk with ultrafast time-bin encoding," *Optica* **11**(7), 1017–1023 (2024).
37. F. Bouchard, K. Fenwick, K. Bonsma-Fisher, *et al.*, "Programmable photonic quantum circuits with ultrafast time-bin encoding," *Phys. Rev. Lett.* **133**(9), 090601 (2024).
38. A. M. Weiner, *Ultrafast optics* (John Wiley & Sons, 2011).
39. L. Lepetit, G. Chériaux, and M. Joffre, "Linear techniques of phase measurement by femtosecond spectral interferometry for applications in spectroscopy," *J. Opt. Soc. Am. B* **12**(12), 2467–2474 (1995).
40. Schott, "TIE-31: Mechanical and thermal properties of optical glass," (2010).
41. D. England, F. Bouchard, K. Fenwick, *et al.*, "Perspectives on all-optical Kerr switching for quantum optical applications," *Appl. Phys. Lett.* **119**(16), 160501 (2021).
42. C. Kupchak, J. Erskine, D. England, *et al.*, "Terahertz-bandwidth switching of heralded single photons," *Opt. Lett.* **44**(6), 1427–1430 (2019).
43. C. Kupchak, P. J. Bustard, K. Heshami, *et al.*, "Time-bin-to-polarization conversion of ultrafast photonic qubits," *Phys. Rev. A* **96**(5), 053812 (2017).
44. S. Arzhantsev and M. Maroncelli, "Design and characterization of a femtosecond fluorescence spectrometer based on optical Kerr gating," *Appl. Spectrosc.* **59**(2), 206–220 (2005).
45. S. Arzhantsev, H. Jin, G. A. Baker, *et al.*, "Measurements of the complete solvation response in ionic liquids," *J. Phys. Chem. B* **111**(18), 4978–4989 (2007).
46. S. Kinoshita, H. Ozawa, Y. Kanematsu, *et al.*, "Efficient optical Kerr shutter for femtosecond time-resolved luminescence spectroscopy," *Rev. Sci. Instrum.* **71**(9), 3317–3322 (2000).
47. J. Takeda, K. Nakajima, S. Kurita, *et al.*, "Time-resolved luminescence spectroscopy by the optical Kerr-gate method applicable to ultrafast relaxation processes," *Phys. Rev. B* **62**(15), 10083–10087 (2000).
48. B. Schmidt, S. Laimgruber, W. Zinth, *et al.*, "A broadband Kerr shutter for femtosecond fluorescence spectroscopy," *Appl. Phys. B* **76**(8), 809–814 (2003).
49. I. A. Walmsley and C. Dorrer, "Characterization of ultrashort electromagnetic pulses," *Adv. Opt. Photonics* **1**(2), 308–437 (2009).

Supplemental Document

Optics EXPRESS

Time-of-flight spectroscopy with ultrafast all-optical gating: supplement

KATE L. FENWICK,^{1,2,*}  GUILLAUME S. THEKKADATH,²  PHILIP J. BUSTARD,²  DUNCAN ENGLAND,²  FRÉDÉRIC BOUCHARD,² AND BENJAMIN SUSSMAN^{1,2}

¹Department of Physics, University of Ottawa, Advanced Research Complex, 25 Templeton Street, Ottawa ON Canada, K1N 6N5, Canada

²National Research Council of Canada, 100 Sussex Drive, Ottawa, Ontario K1A 0R6, Canada

This supplement published with Optica Publishing Group on 5 February 2025 by The Authors under the terms of the [Creative Commons Attribution 4.0 License](https://creativecommons.org/licenses/by/4.0/) in the format provided by the authors and unedited. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

Supplement DOI: <https://doi.org/10.6084/m9.figshare.28278809>

Parent Article DOI: <https://doi.org/10.1364/OE.549697>

Time-of-flight spectroscopy with ultrafast all-optical gating: supplemental document

1. THEORY OF FREQUENCY-TO-TIME MAPPING

When a short pulse propagates through a transparent medium, its group velocity will be frequency dependent. This phenomenon, called group velocity dispersion (GVD), allows for frequency-to-time mapping. The transfer function of a dispersive medium, of length z and propagation constant β_2 , can be given by [1]

$$\tilde{H}(\omega) = e^{-i\beta_2 z (\omega - \omega_0)^2 / 2}, \quad (\text{S1})$$

where $\tilde{H}(\omega)$ is a function of frequency, ω , and ω_0 is the carrier frequency of the pulse. The dispersion parameter of the medium, D , is related to the second-order propagation constant through: $D = -2\pi c \beta_2 / \lambda^2$, where c is the speed of light and λ denotes wavelength.

Following closely the mathematical description of frequency-to-time mapping presented in Refs [1, 2], the dispersed pulse can be written as the convolution between the input pulse, $E_{\text{in}}(t)$, and the transfer function of the dispersive medium, $H(t)$, in time:

$$\begin{aligned} E_{\text{out}}(t) &= E_{\text{in}}(t) * H(t) \\ &= \int dt' E_{\text{in}}(t') H(t - t'), \end{aligned} \quad (\text{S2})$$

where t' and t describe the temporal domain of the input and output pulses, respectively.

By taking the Fourier transform of $\tilde{H}(\omega)$ given in Equation S1, $H(t)$ can be found,

$$\begin{aligned} H(t) &= \frac{1}{2\pi} \int d\omega e^{-i\frac{1}{2}\beta_2 z (\omega - \omega_0)^2} e^{i\omega t} \\ &= \sqrt{\frac{-i}{2\pi\beta_2 z}} e^{it^2/2\beta_2 z} e^{i\omega_0 t}, \end{aligned} \quad (\text{S3})$$

where $e^{i\omega_0 t}$ is the carrier term. The output pulse is therefore

$$\begin{aligned} E_{\text{out}}(t) &= \sqrt{\frac{-i}{2\pi\beta_2 z}} \int dt' E_{\text{in}}(t') e^{i(t-t')^2/2\beta_2 z} e^{i\omega_0(t-t')} \\ &= A \int dt' E_{\text{in}}(t') e^{-itt'/\beta_2 z} e^{it'^2/\beta_2 z} e^{-i\omega_0 t'}, \end{aligned} \quad (\text{S4})$$

with

$$A = \sqrt{\frac{-i}{2\pi\beta_2 z}} e^{it^2/2\beta_2 z} e^{i\omega_0 t}. \quad (\text{S5})$$

Here, we assume a near-transform limited input pulse with duration τ_0 , where $E_{\text{in}}(t') \rightarrow 0$ sufficiently quickly for $|t'| > \tau_0/2$. In this case, and for large enough dispersion,

$$|\beta_2 z| \gg \frac{\tau_0^2}{8}, \quad (\text{S6})$$

$t'^2/\beta_2 z \approx 0$ for all values of t' over which the input pulse is non-zero. This is the direct frequency-to-time mapping regime. The output pulse is then given by

$$E_{\text{out}}(t) = A \int dt' E_{\text{in}}(t') e^{-itt'/\beta_2 z} e^{-i\omega_0 t'}. \quad (\text{S7})$$

The integral in Equation S7 is just the Fourier transform of the input pulse, where $\omega = \omega_0 + t/\beta_2 z$. The output pulse can therefore be written as

$$E_{\text{out}}(t) = \sqrt{\frac{-i}{2\pi\beta_2 z}} e^{it^2/2\beta_2 z} e^{i\omega_0 t} \tilde{E}_{\text{in}}(\omega = \omega_0 + \frac{t}{\beta_2 z}). \quad (\text{S8})$$

It is seen that in the large dispersion limit, the stretched output pulse is a scaled version of the input spectrum multiplied by a quadratic temporal phase. This shows the important result that the dispersive element maps frequency onto time, with a mapping factor of

$$\frac{\partial\omega}{\partial t} = \frac{1}{\beta_2 z}. \quad (\text{S9})$$

It is important to remember that Eq. S6 sets the required dispersion to reach the “far field” for frequency-to-time mapping with minimal distortions and an accurate spectrum representation. The finest spectral feature, δf , of the input waveform and its duration τ_0 are related by the time-bandwidth product $\tau_0 \delta f \sim 1$. This sets a limit on the spectral resolution in the large dispersion limit,

$$|\beta_2 z|(\delta f)^2 \gg \frac{1}{8}. \quad (\text{S10})$$

This spectral feature, δf , is stretched to a duration $\delta t = 2\pi(\delta f)|\beta_2 z|$, which must satisfy

$$\begin{aligned} \delta t &\gg \frac{\pi}{\sqrt{2}} \sqrt{|\beta_2 z|} \\ \delta t &\gg \frac{\pi}{4(\delta f)}, \end{aligned} \quad (\text{S11})$$

and we see that the smallest temporal feature, δt , which can be generated in frequency-to-time mapping must exceed the bandwidth-limited pulse duration of the corresponding spectral feature.

2. THEORY OF OPTICAL KERR-GATING

In this work, the time-stretched intensity envelope is sampled optically, through a process known as cross-phase modulation. Cross-phase modulation occurs due to a $\chi^{(3)}$ nonlinear optical process known as the optical Kerr effect. In this process, a strong ultrafast optical pulse generates an intensity-dependent refractive index along its polarization axis in the medium through which it travels. This induces a localized birefringence, moving with the pulse as it propagates through the medium, which can then be used to rotate the polarization of another optical signal in a very short temporal window (< 1 ps). When the medium is placed between two crossed polarizers, only the rotated signal light will be transmitted.

For a linearly polarized ultrafast pump pulse co-propagating with a time-stretched signal beam of intensity I_0 , the gated light will have an intensity profile of [3]

$$I(t) = I_0 \sin^2(2\theta) \sin^2\left(\frac{\Delta\phi(t)}{2}\right), \quad (\text{S12})$$

where θ is the angle between the signal and pump polarizations and $\Delta\phi(t)$ is the time-dependent phase difference between the parallel and orthogonal components of signal polarization (relative to the pump). Polarization rotation is maximal for $\theta = 45^\circ$, so the pump is set 45° relative to the signal in this work.

The time-dependent phase difference is given by [4]:

$$\Delta\phi(T) = \frac{8\pi n_2}{3\lambda_{\text{signal}}} \int_0^L I_{\text{pump}}(T - d_w z) dz, \quad (\text{S13})$$

where n_2 is the nonlinear refractive index of the fiber, λ_{signal} is the center wavelength of the stretched signal, and z is the propagation distance within the fiber of length L . In a dispersive medium, the pump and signal will experience temporal walkoff, $d_w = v_{\text{sp}}^{-1} - v_{\text{gs}}^{-1}$, where v_{sp} and v_{gs} are the pump and signal group velocities, respectively. For this reason, the temporal intensity of the pump, I_{pump} , is expressed in the frame moving with the signal [5] $T = t - z/v_{\text{gs}}$, where t is time in the laboratory frame.

3. MEASURING SPECTRAL FRINGES

Figure S1 compares what will be observed when two pulses, separated by delay $\Delta\tau_2$, are measured with a spectrometer and with the time-of-flight spectrometer. If these two pulses were to be measured with a grating spectrometer with adequate resolution, spectral fringes with spacing $\Delta f = 1/\Delta\tau_2$ would be observed [1, 6]. This is due to the geometry of the spectrometer. Specific frequencies of pulses that are localized in time will traverse different path lengths before arriving at the same spot on the detector. If the spectrometer geometry allows, two pulses separated in time can be spread such that they interfere at the detector, leading to the observation of fringes. Similarly, this can be seen if the two pulses are stretched in a dispersive medium and sampled in the time domain.

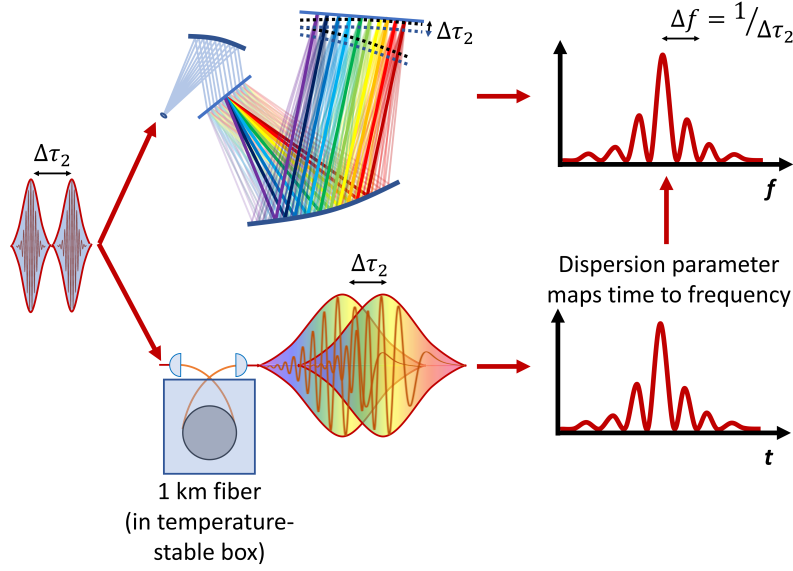


Fig. S1. Spectral fringe measurement in the spectral domain. Comparing spectral fringes measured with a grating spectrometer (top) and the time-of-flight spectrometer (bottom). Spatial dispersion from the grating in the spectrometer leads to interference in the spectral domain. Group velocity dispersion from the 1 km fiber in the time-of-flight spectrometer leads to interference in the temporal domain, which can be mapped to the frequency-domain with the fiber dispersion parameter.

REFERENCES

1. C. Iaconis and I. A. Walmsley, "Self-referencing spectral interferometry for measuring ultra-short optical pulses," *IEEE J. quantum electronics* **35**, 501–509 (1999).
2. A. M. Weiner, *Ultrafast optics* (John Wiley & Sons, 2011).
3. H. Kanbara, H. Kobayashi, T. Kaino, *et al.*, "Highly efficient ultrafast optical Kerr shutters with the use of organic nonlinear materials," *JOSA B* **11**, 2216–2223 (1994).
4. G. Agrawal, *Applications of nonlinear fiber optics* (Elsevier, 2001).
5. C. Kupchak, J. Erskine, D. England, and B. Sussman, "Terahertz-bandwidth switching of heralded single photons," *Opt. letters* **44**, 1427–1430 (2019).
6. L. Lepetit, G. Chériaux, and M. Joffe, "Linear techniques of phase measurement by femtosecond spectral interferometry for applications in spectroscopy," *JOSA B* **12**, 2467–2474 (1995).

Chapter 3

Some background on quantum optics

Photonic quantum information processing requires photons. Our all-optical approach to quantum information processing in the ultrafast regime is no different. We must be capable of generating manipulating, and detecting single photons. This chapter presents several important concepts in quantum optics, which make up the foundation of our ability to do quantum information processing with single photons. Section 3.1 introduces the field of quantum optics, starting with the quantization of the electromagnetic field and following with a discussion on different states of light. After this, Section 3.2 explains how single photons can be generated in the lab, while Section 3.3 outlines how these photons are detected. Section 3.4 then discusses how photon sources can be characterized. This chapter concludes with a demonstration of characterizing an entangled photon pair source¹ in Section 3.5.

¹This demonstration makes use of the optical Kerr gate introduced in Section 2.2.5!

3.1 Quantum optics

The field of quantum optics studies the nature and effects of light as quantized photons. Historically, the first major development which led to this new way of understanding light was Max Planck’s derivation of the correct form for the distribution function of energy emitted by an ideal black body². Planck’s remarkably accurate radiation law, published in 1901 [309], regarded the energy spectrum as consisting of “energy elements” that are directly proportional to their frequency through Planck’s constant of nature, $h = 6.62607015 \times 10^{-34}$ J·s. But, it was Einstein’s 1905 paper, which outlined his theory of light quanta (photons) in order to explain the photoelectric effect [310], which recognized the essence of quantum theory. In 1913, another major advancement was made when Bohr provided further proof of the quantization of optical radiation, by theorizing that the emission lines of hydrogen corresponded to quantized energy levels within atoms [311]. Among many others, these developments were crucial for the understanding of interactions between light and matter, and for quantum mechanics as a whole. Many technologies today rely on these scientific developments, including the transistor and the laser—both of which are hard to imagine a world without!

This quantized picture of light raises an interesting question: what exactly is a photon? The answer to this will likely be different, depending on who you ask. Some might say that a photon is an elementary particle in the standard model, while others may tell you that a photon is the smallest possible packet of electromagnetic energy. While these definitions are correct, there is yet another definition which is often used within the formalism of quantum optics: a photon is a single quantum excitation of a normal mode of the electromagnetic field. We will see that this definition becomes more obvious in the next section, where we follow Dirac [312] and combine the wave- and particle-like pictures of light³.

²Before the photon picture, classical physics predicted that an ideal black body at thermal equilibrium would emit an unbounded quantity of energy as wavelength decreased into the ultraviolet range. Yet, experimentally, this trend was not observed. This discrepancy is referred to as the *ultraviolet catastrophe*.

³There are many excellent quantum optics textbooks which are recommended for further reading. To name a few: Loudon’s *The quantum theory of light* [313], Gerry and Knight’s *Introductory quantum optics* [314], or Scully and Zubairy’s *Quantum optics* [315].

3.1.1 Quantization of the electromagnetic field

The essence of Dirac's quantum theory of radiation [312] associates each normal mode of the electromagnetic field with a quantized simple harmonic oscillator. We will demonstrate how this is done, and then outline how quantum states of light can be used to describe how many excitations (photons) are present in each of these normal modes. We begin with the simple, yet informative case, of a radiation field confined to a one-dimensional cavity of length, L , along the z axis. Provided the walls of the cavity are perfectly conducting, the electric field must vanish on the boundaries of the cavity (*i.e.*, at $z = 0$ and $z = L$). Assuming that there are no other sources of radiation (*i.e.*, no currents, charges, nor dielectric materials within the cavity), the electric field will take the form

$$\mathbf{E}(\mathbf{r}, t) = \hat{\mathbf{x}}E_x(z, t), \quad (3.1)$$

where the field is taken to be polarized along the x -direction, as indicated by the unit polarization vector, $\hat{\mathbf{x}}$. The functional form of $E_x(z, t)$ must be a solution to Maxwell's equations, as defined in Eqs. 2.39–2.42 in Chapter 2, but written again here for convenience,

$$\nabla \cdot \mathbf{E} = 0 \quad (3.2)$$

$$\nabla \cdot \mathbf{B} = 0 \quad (3.3)$$

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \quad (3.4)$$

$$\nabla \times \mathbf{B}(t) = \mu_0 \epsilon_0 \frac{\partial \mathbf{E}(t)}{\partial t}. \quad (3.5)$$

A similar protocol to the one presented in Section 2.2.1 can be followed to arrive at the wave equation, this time in the absence of a nonlinear source term,

$$\nabla^2 \mathbf{E} - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = 0. \quad (3.6)$$

Solutions which satisfy Maxwell's equations are referred to as *modes*. Under the boundary conditions we have imposed, a single-mode field solution to Maxwell's equations takes the form,

$$E_x(z, t) = \left(\frac{2\omega^2}{V\epsilon_0} \right)^{1/2} q(t) \sin(kz), \quad (3.7)$$

where ω is the frequency of the mode, and $k = \omega/c$ is the wave number. The cavity length defines the allowed frequencies of oscillation, $\omega_m = c(m\pi/L)$, where $m = 1, 2, 3, \dots$, and the effective volume of the cavity is denoted as $V = L A$, where A is the cross sectional area of the cavity. We also see a time-dependent factor, $q(t)$, in our solution, which should have dimensions of length. This factor is a time-dependent field amplitude, which here acts as the canonical position.

The magnetic field in the cavity can be found by inserting our solution for $\mathbf{E}(\mathbf{r}, t)$ into Eq. 3.5,

$$\mathbf{B}(\mathbf{r}, t) = \hat{\mathbf{y}} E_y(z, t), \quad (3.8)$$

where $\hat{\mathbf{y}}$ is the unit polarization vector in the y-direction, and

$$B_y(z, t) = \left(\frac{\mu_0 \epsilon_0}{k} \right) \left(\frac{2\omega^2}{V\epsilon_0} \right)^{1/2} \dot{q}(t) \cos(kz). \quad (3.9)$$

Here, $\dot{q}(t)$ is another time-dependent field amplitude, which acts as the canonical momentum for a “particle” of unit mass, so that $p(t) = \dot{q}(t)$.

We can calculate the classical field energy (the Hamiltonian, which we denote as $\mathcal{H}$) in terms of the canonical position and momentum,

$$\begin{aligned} \mathcal{H} &= \frac{1}{2} \int dV \left[\epsilon_0 \mathbf{E}^2(\mathbf{r}, t) + \frac{1}{\mu_0} \mathbf{B}^2(\mathbf{r}, t) \right] \\ &= \frac{1}{2} \int_0^L dz \int dA \left[\epsilon_0 E^2(z, t) + \frac{1}{\mu_0} B^2(z, t) \right] \\ &= \frac{1}{2} [p^2(t) + \omega^2 q^2(t)]. \end{aligned} \quad (3.10)$$

We can see that this Hamiltonian is of the same form as that of a classical one-dimensional harmonic oscillator,

$$\mathcal{H}_{\text{HO}} = \frac{p^2}{2m} + \frac{m\omega^2 q^2}{2}, \quad (3.11)$$

with unit mass, $m = 1$, where the electric and magnetic field amplitudes play the role of the canonical position, $q(t)$, and momentum, $p(t)$. It should be noted that, so far, we have considered only a single-mode solution to Maxwell's equations; however, a more general solution takes the form of a linear combination of these single-mode solutions⁴,

$$E_x(z, t) = \sum_j \left(\frac{2\omega_j^2}{V\epsilon_0} \right)^{1/2} q_j(t) \sin(k_j z) \quad (3.12)$$

$$B_y(z, t) = \sum_j \left(\frac{\mu_0\epsilon_0}{k_j} \right) \left(\frac{2\omega_j^2}{V\epsilon_0} \right)^{1/2} \dot{q}_j(t) \cos(k_j z). \quad (3.13)$$

In this case, the Hamiltonian of the electromagnetic field is written as

$$\mathcal{H} = \frac{1}{2} \sum_j (p_j^2(t) + \omega_j^2 q_j^2(t)). \quad (3.14)$$

The result that the Hamiltonian of the electromagnetic field is of the same form as the Hamiltonian of a harmonic oscillator is crucial to the quantization of the electromagnetic field, which we will now proceed to demonstrate. We can use Bohr's correspondence principle⁵ [316] and replace our classical variables for position and momentum, $q_j(t)$ and $p_j(t)$, with the associated Hermitian operators, $\hat{q}_j(t)$ and $\hat{p}_j(t)$,

⁴It should also be noted here that the electromagnetic field need not be polarized as we have defined here. More generally, we can include an additional sum over the two possible polarization components in the linear combination of modes we have written; however, we stick with linearly-polarized fields here for simplicity.

⁵In the limit of large numbers, the predictions of quantum physics become identical to the predictions of classical physics.

which satisfy the canonical commutation relations⁶,

$$[\hat{q}_j, \hat{p}_{j'}] = i\hbar\delta_{j,j'}, \quad (3.15)$$

$$[\hat{q}_j, \hat{q}_{j'}] = 0, \quad (3.16)$$

$$[\hat{p}_j, \hat{p}_{j'}] = 0. \quad (3.17)$$

We must now write the electric and magnetic fields as operators,

$$\hat{E}_x(z, t) = \sum_j \left(\frac{2\omega_j^2}{V\epsilon_0} \right)^{1/2} \hat{q}_j(t) \sin(k_j z) \quad (3.18)$$

$$\hat{B}_y(z, t) = \sum_j \left(\frac{\mu_0\epsilon_0}{k_j} \right) \left(\frac{2\omega_j^2}{V\epsilon_0} \right)^{1/2} \hat{p}_j(t) \cos(k_j z), \quad (3.19)$$

as well as the Hamiltonian,

$$\hat{\mathcal{H}} = \sum_j \hat{\mathcal{H}}_j = \sum_j \frac{1}{2} [\hat{p}_j^2(t) + \omega_j^2 \hat{q}_j^2(t)]. \quad (3.20)$$

Since the operators, $\hat{q}_j(t)$ and $\hat{p}_j(t)$ are Hermitian, they correspond to physically observable quantities; however, it is convenient (and traditional) to introduce the non-Hermitian operators,

$$\hat{a}_j(t) = (2\hbar\omega_j)^{-1/2} [\omega_j \hat{q}_j(t) + i\hat{p}_j(t)], \quad (3.21)$$

$$\hat{a}_j^\dagger(t) = (2\hbar\omega_j)^{-1/2} [\omega_j \hat{q}_j(t) - i\hat{p}_j(t)], \quad (3.22)$$

which are referred to as the annihilation and creation operators for mode j , respectively. The electric and magnetic field operators can then be written as

$$\hat{E}_x(z, t) = \sum_j \mathcal{E}_j [\hat{a}_j(t) + \hat{a}_j^\dagger(t)] \sin(k_j z), \quad (3.23)$$

$$\hat{B}_y(z, t) = \sum_j -i\mathcal{B}_j [\hat{a}_j(t) - \hat{a}_j^\dagger(t)] \cos(k_j z), \quad (3.24)$$

⁶The first commutation relation should technically be written as $[\hat{q}_j, \hat{p}_j] = i\hbar\delta_{j,j'}\hat{I}$; however, we follow custom here and drop the identity operator, $\hat{I}$.

where $\mathcal{E}_j = (\hbar\omega_j/\epsilon_0 V)^{1/2}$ and $\mathcal{B}_j = (\mu_0/k_j) (\epsilon_0\hbar\omega_j^3/V)^{1/2}$. The commutation relations for the operators, $\hat{a}_j(t)$ and $\hat{a}_j^\dagger(t)$, follow from those defined for $\hat{q}_j(t)$ and $\hat{p}_j(t)$, where

$$[\hat{a}_j, \hat{a}_{j'}^\dagger] = \delta_{j,j'}, \quad (3.25)$$

$$[\hat{a}_j, \hat{a}_{j'}] = 0, \quad (3.26)$$

$$[\hat{a}_j^\dagger, \hat{a}_{j'}^\dagger] = 0. \quad (3.27)$$

The Hamiltonian for the j^{th} mode can then be written in terms of the annihilation and creation operators as

$$\hat{\mathcal{H}}_j = \hbar\omega_j \left(\hat{a}_j^\dagger(t)\hat{a}_j(t) + \frac{1}{2} \right). \quad (3.28)$$

Up to this point, we have been working in the Heisenberg picture, where the state vectors are stationary and the operators evolve with time. We should therefore treat the time dependence of the operators, $\hat{a}_j(t)$ and $\hat{a}_j^\dagger(t)$. An arbitrary operator, $\hat{O}$, (with no explicit time dependence in the Schrödinger picture) evolves according to

$$\frac{d\hat{O}}{dt} = \frac{i}{\hbar} [\hat{\mathcal{H}}, \hat{O}], \quad (3.29)$$

which has the formal solution

$$\hat{O}(t) = e^{i\hat{\mathcal{H}}t/\hbar} \hat{O}(0) e^{-i\hat{\mathcal{H}}t/\hbar}. \quad (3.30)$$

The Baker-Campbell-Hausdorff lemma allows us to write

$$\hat{O}(t) = \hat{O}(0) + \frac{it}{\hbar} [\hat{\mathcal{H}}, \hat{O}(0)] \quad (3.31)$$

$$+ \frac{1}{2!} \left(\frac{it}{\hbar} \right)^2 [\hat{\mathcal{H}}, [\hat{\mathcal{H}}, \hat{O}(0)]] \quad (3.32)$$

$$+ \frac{1}{3!} \left(\frac{it}{\hbar} \right)^3 [\hat{\mathcal{H}}, [\hat{\mathcal{H}}, [\hat{\mathcal{H}}, \hat{O}(0)]]] + \dots \quad (3.33)$$

Applying this to our annihilation operator for mode j , we have

$$\begin{aligned}\hat{a}_j(t) &= \hat{a}_j(0) \left[1 - i\omega_j t - \frac{\omega_j^2 t^2}{2!} + i\frac{\omega_j^3 t^3}{3!} + \dots \right] \\ &= \hat{a}_j(0) e^{-i\omega_j t},\end{aligned}\tag{3.34}$$

and applying this to our creation operator for mode j , we have

$$\begin{aligned}\hat{a}_j^\dagger(t) &= \hat{a}_j^\dagger(0) \left[1 + i\omega_j t - \frac{\omega_j^2 t^2}{2!} - i\frac{\omega_j^3 t^3}{3!} + \dots \right] \\ &= \hat{a}_j^\dagger(0) e^{i\omega_j t}.\end{aligned}\tag{3.35}$$

As with many problems in quantum mechanics, we can solve for the energy eigenvalues of the Hamiltonian. We introduce the energy eigenstate associated with the j^{th} mode, $|\psi_n\rangle_j$, with energy eigenvalue, $E_n^{(j)}$, which together obey the equation

$$\hat{\mathcal{H}}_j |\psi_n\rangle_j = \hbar\omega_j \left(\hat{a}_j^\dagger \hat{a}_j + \frac{1}{2} \right) |\psi_n\rangle_j = E_n^{(j)} |\psi_n\rangle_j,\tag{3.36}$$

where we have dropped the time dependence of the operators for ease of reading, but note that it is still present. If we multiply both sides of Eq. 3.36 by $\hat{a}_j^\dagger$ and rearrange, we can generate a new eigenvalue equation,

$$\begin{aligned}\hbar\omega_j \left(\hat{a}_j^\dagger \hat{a}_j^\dagger \hat{a}_j + \frac{1}{2} \hat{a}_j^\dagger \right) |\psi_n\rangle_j &= E_n^{(j)} \hat{a}_j^\dagger |\psi_n\rangle_j \\ \hbar\omega_j \left[\left(\hat{a}_j^\dagger \hat{a}_j \hat{a}_j^\dagger - \hat{a}_j^\dagger \right) + \frac{1}{2} \hat{a}_j^\dagger \right] |\psi_n\rangle_j &= E_n^{(j)} \hat{a}_j^\dagger |\psi_n\rangle_j \\ \hbar\omega_j \left[\hat{a}_j^\dagger \hat{a}_j + \frac{1}{2} \right] \left(\hat{a}_j^\dagger |\psi_n\rangle_j \right) &= [E_n^{(j)} + \hbar\omega_j] \left(\hat{a}_j^\dagger |\psi_n\rangle_j \right),\end{aligned}\tag{3.37}$$

where we see a new eigenstate, $(\hat{a}_j^\dagger |\psi_n\rangle_j)$, emerge with energy eigenvalue, $E_n^{(j)} + \hbar\omega_j$. We now see why $\hat{a}_j^\dagger$ is called the creation operator for mode j ; it “creates” a quantum of energy, $\hbar\omega_j$, in mode j of the electromagnetic field. We often say, loosely, that the action of this operator is to create a photon of energy $\hbar\omega_j$. A similar protocol can be followed to understand the effects of the annihilation operator; we can multiply both

sides of Eq. 3.36 by $\hat{a}_j$ and rearrange to arrive at

$$\hbar\omega_j \left[\hat{a}_j^\dagger \hat{a}_j + \frac{1}{2} \right] \left(\hat{a}_j |\psi_n\rangle_j \right) = [E_n^{(j)} - \hbar\omega_j] \left(\hat{a}_j |\psi_n\rangle_j \right), \quad (3.38)$$

where we see another new eigenstate, $(\hat{a}_j |\psi_n\rangle_j)$, with eigenvalue, $E_n^{(j)} - \hbar\omega_j$. The $\hat{a}_j$ operator is called the annihilation operator because it “destroys” a quantum of energy, $\hbar\omega_j$, in mode j of the electromagnetic field. Repeated operation of the annihilation operator will continuously decrease the energy eigenstate by integer multiples of $\hbar\omega_j$; yet, the energy of the harmonic oscillator must always be positive. This implies that there is a lowest-energy eigenvalue, $E_0^{(j)} > 0$, with a corresponding eigenstate of $|0\rangle_j$ such that

$$\hat{\mathcal{H}}_j \left(\hat{a}_j |0\rangle_j \right) = \left(E_0^{(j)} - \hbar\omega_j \right) \left(\hat{a}_j |0\rangle_j \right) = 0, \quad (3.39)$$

where we require that

$$\hat{a}_j |0\rangle_j = 0. \quad (3.40)$$

The eigenvalue equation for the ground state is therefore

$$\hat{\mathcal{H}}_j |0\rangle_j = \hbar\omega_j \left(\hat{a}_j^\dagger \hat{a}_j + \frac{1}{2} \right) |0\rangle_j = \frac{1}{2} \hbar\omega_j |0\rangle_j. \quad (3.41)$$

The creation and annihilation operators thus provide us with an energy spectrum, as depicted in Fig. 3.1, for the j^{th} mode of the electromagnetic field, with integer multiples of $\hbar\omega_j$. The bottom of this energy spectrum is called the ground state, with energy $\hbar\omega_j/2$. Any further use of the annihilation operator, once the ground state is reached, will result in $\hat{a}_j |0\rangle_j = 0$. It is common to denote a photon in mode j as $\hat{a}_j^\dagger |0\rangle_j$, which is measured in the lab as a count at a single photon detector. We will use the term *photon* quite casually throughout this thesis, but it is important to remember our more formal description of a photon from this section: a photon is the canonically quantized energy of the electromagnetic field (which itself is a solution to Maxwell’s equations).

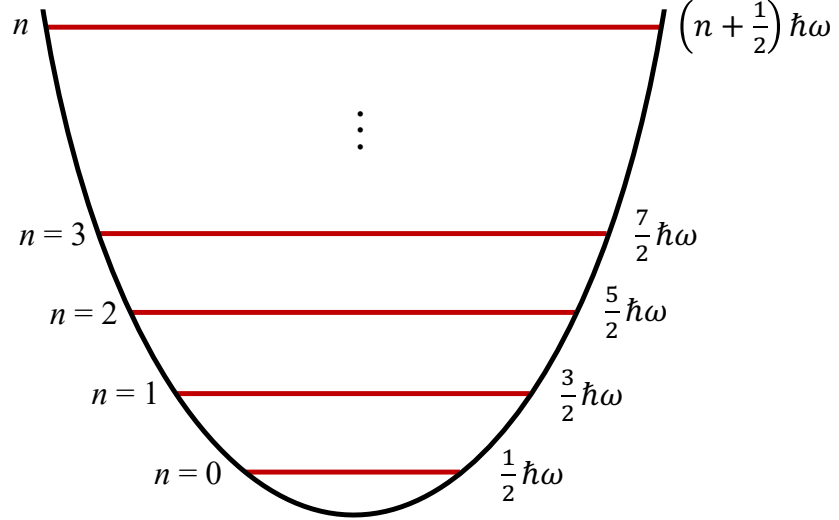


Figure 3.1: **Photon energy level spectrum.** The energy level spectrum arising from the quantization of the electromagnetic field is depicted against the harmonic oscillator potential.

3.1.2 Quantization of the electromagnetic field in unbounded free space*

It should be noted that, so far, we have considered the quantization of the electromagnetic field in a one-dimensional cavity; however, in many experiments which utilize single photons, they are not bound by a cavity. We can quantize the electromagnetic field in unbounded free space by considering a large, but finite, cubic space of side length L (where we take L to be much larger than the wavelengths of the field). Solutions to Maxwell's equations⁷ now take the form of plane waves and we impose periodic boundary conditions at the “bounds” of the L^3 cube. Here, the classical electric and magnetic field solutions can be expanded as a sum over an infinite discrete set of plane wave vectors⁸, $\mathbf{k} = (k_x, k_y, k_z)$, and a sum over two polarizations, s , such that

$$\mathbf{E}(\mathbf{r}, t) = i \sum_s \sum_{\mathbf{k}} \omega_k \hat{\epsilon}_{\mathbf{k},s} [A_{\mathbf{k},s} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega_k t)} - A_{\mathbf{k},s}^* e^{-i(\mathbf{k} \cdot \mathbf{r} - \omega_k t)}], \quad (3.42)$$

$$\mathbf{B}(\mathbf{r}, t) = \frac{i}{c} \sum_s \sum_{\mathbf{k}} \omega_k \left(\frac{\mathbf{k}}{|\mathbf{k}|} \times \hat{\epsilon}_{\mathbf{k},s} \right) [A_{\mathbf{k},s} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega_k t)} - A_{\mathbf{k},s}^* e^{-i(\mathbf{k} \cdot \mathbf{r} - \omega_k t)}], \quad (3.43)$$

⁷See Gerry and Knight's *Introductory quantum optics* [314] for a detailed derivation.

⁸Note that in free space, the sum should be replaced by a volume integral, $\sum_{\mathbf{k}} \rightarrow \frac{V}{\pi^2} \int k^2 dk$.

where $\hat{\mathbf{e}}_{\mathbf{k},s}$ is the unit polarization vector, and we note that the time-dependent field amplitude is given by $A_{\mathbf{k},s}(t) = A_{\mathbf{k},s}e^{-i\omega_k t}$. Periodic boundary conditions require that

$$\begin{aligned} k_x &= \frac{2\pi n_x}{L}, \\ k_y &= \frac{2\pi n_y}{L}, \\ k_z &= \frac{2\pi n_z}{L}, \end{aligned} \quad (3.44)$$

where n_x , n_y , and n_z are each integer numbers. A given set of (n_x, n_y, n_z) defines a mode of the electromagnetic field. Eq. 3.2 of Maxwell's equations requires that $\mathbf{k} \cdot \hat{\mathbf{e}}_{\mathbf{k}} = 0$, such that the electric and magnetic fields are transverse, and so there are two independent polarization directions of $\hat{\mathbf{e}}_{\mathbf{k},s}$ for each possible $\mathbf{k}$ (which we sum over with index s).

The energy of the field is calculated as

$$\mathcal{H} = \frac{1}{2} \int_V \left(\epsilon_0 \mathbf{E} \cdot \mathbf{E} + \frac{1}{\mu_0} \mathbf{B} \cdot \mathbf{B} \right) dV, \quad (3.45)$$

which, after some calculation, is found to be of the simple form,

$$\mathcal{H} = 2\epsilon_0 V \sum_s \sum_{\mathbf{k}} \omega_k^2 A_{\mathbf{k},s} A_{\mathbf{k},s}^*. \quad (3.46)$$

In order to quantize the fields, we must introduce the canonical position and momentum variables as in the previous section. We thus define

$$A_{\mathbf{k},s} = \frac{1}{2\omega_k (\epsilon_0 V)^{1/2}} [\omega_k q_{\mathbf{k},s} + i p_{\mathbf{k},s}], \quad (3.47)$$

$$A_{\mathbf{k},s}^* = \frac{1}{2\omega_k (\epsilon_0 V)^{1/2}} [\omega_k q_{\mathbf{k},s} - i p_{\mathbf{k},s}], \quad (3.48)$$

and upon substitution into Eq. 3.46, we arrive at

$$\mathcal{H} = \frac{1}{2} \sum_s \sum_{\mathbf{k}} (p_{\mathbf{k},s}^2 + \omega_k^2 q_{\mathbf{k},s}^2), \quad (3.49)$$

which is equivalent to the energy expression for a harmonic oscillator of unit mass,

as demonstrated in the previous section.

The quantization of the electromagnetic field requires that the canonical variables, $q_{\mathbf{k},s}$ and $p_{\mathbf{k},s}$, become operators satisfying the commutation relations

$$[\hat{q}_{\mathbf{k},s}, \hat{p}_{\mathbf{k}',s'}] = i\hbar\delta_{\mathbf{k},\mathbf{k}'}\delta_{s,s'}, \quad (3.50)$$

$$[\hat{q}_{\mathbf{k},s}, \hat{q}_{\mathbf{k}',s'}] = 0, \quad (3.51)$$

$$[\hat{p}_{\mathbf{k},s}, \hat{p}_{\mathbf{k}',s'}] = 0. \quad (3.52)$$

As in the previous section, we can write creation and annihilation operators in terms of the canonical variables,

$$\hat{a}_{\mathbf{k},s} = (2\hbar\omega_k)^{1/2} [\omega_k \hat{q}_{\mathbf{k},s} + i\hat{p}_{\mathbf{k},s}], \quad (3.53)$$

$$\hat{a}_{\mathbf{k},s}^\dagger = (2\hbar\omega_k)^{1/2} [\omega_k \hat{q}_{\mathbf{k},s} - i\hat{p}_{\mathbf{k},s}], \quad (3.54)$$

which satisfy the commutation relations

$$[\hat{a}_{\mathbf{k},s}, \hat{a}_{\mathbf{k}',s'}^\dagger] = \delta_{\mathbf{k},\mathbf{k}'}\delta_{s,s'}, \quad (3.55)$$

$$[\hat{a}_{\mathbf{k},s}, \hat{a}_{\mathbf{k}',s'}] = 0, \quad (3.56)$$

$$[\hat{a}_{\mathbf{k},s}^\dagger, \hat{a}_{\mathbf{k}',s'}^\dagger] = 0. \quad (3.57)$$

The Hamiltonian can then be written as

$$\hat{\mathcal{H}} = \sum_s \sum_{\mathbf{k}} \hbar\omega_k \left(\hat{a}_{\mathbf{k},s}^\dagger \hat{a}_{\mathbf{k},s} + \frac{1}{2} \right), \quad (3.58)$$

where it is important to note that these operators can be thought of as Heisenberg picture operators, evaluated at $t = 0$; the time dependence is included in the expressions for the quantized electric and magnetic fields,

$$\hat{\mathbf{E}}(\mathbf{r}, t) = i \sum_s \sum_{\mathbf{k}} \left(\frac{\hbar\omega_k}{2\epsilon_0 V} \right)^{1/2} \hat{\epsilon}_{\mathbf{k},s} \left[\hat{a}_{\mathbf{k},s} e^{i(\mathbf{k}\cdot\mathbf{r} - \omega_k t)} - \hat{a}_{\mathbf{k},s}^\dagger e^{-i(\mathbf{k}\cdot\mathbf{r} - \omega_k t)} \right], \quad (3.59)$$

$$\hat{\mathbf{B}}(\mathbf{r}, t) = \frac{i}{c} \sum_s \sum_{\mathbf{k}} \left(\frac{\hbar\omega_k}{2\epsilon_0 V} \right)^{1/2} \left(\frac{\mathbf{k}}{|\mathbf{k}|} \times \hat{\epsilon}_{\mathbf{k},s} \right) \left[\hat{a}_{\mathbf{k},s} e^{i(\mathbf{k}\cdot\mathbf{r} - \omega_k t)} - \hat{a}_{\mathbf{k},s}^\dagger e^{-i(\mathbf{k}\cdot\mathbf{r} - \omega_k t)} \right]. \quad (3.60)$$

These quantized fields are often written in the form

$$\hat{\mathbf{E}}(\mathbf{r}, t) = \hat{\mathbf{E}}^{(+)}(\mathbf{r}, t) + \hat{\mathbf{E}}^{(-)}(\mathbf{r}, t), \quad (3.61)$$

$$\hat{\mathbf{B}}(\mathbf{r}, t) = \hat{\mathbf{B}}^{(+)}(\mathbf{r}, t) + \hat{\mathbf{B}}^{(-)}(\mathbf{r}, t), \quad (3.62)$$

where $\hat{\mathbf{E}}^{(+)}(\mathbf{r}, t)$ and $\hat{\mathbf{B}}^{(+)}(\mathbf{r}, t)$ are referred to as the positive frequency parts of the fields, containing all the terms which oscillate as $e^{-i\omega_k t}$ for $\omega_k > 0$, while $\hat{\mathbf{E}}^{(-)}(\mathbf{r}, t)$ and $\hat{\mathbf{B}}^{(-)}(\mathbf{r}, t)$ are referred to as the negative frequency parts of the fields. The positive frequency parts of the fields are essentially a collective annihilation operator,

$$\hat{\mathbf{E}}^{(+)}(\mathbf{r}, t) = i \sum_s \sum_{\mathbf{k}} \left(\frac{\hbar \omega_k}{2\epsilon_0 V} \right)^{1/2} \hat{\epsilon}_{\mathbf{k},s} \hat{a}_{\mathbf{k},s} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega_k t)}, \quad (3.63)$$

$$\hat{\mathbf{B}}^{(+)}(\mathbf{r}, t) = \frac{i}{c} \sum_s \sum_{\mathbf{k}} \left(\frac{\hbar \omega_k}{2\epsilon_0 V} \right)^{1/2} \left(\frac{\mathbf{k}}{|\mathbf{k}|} \times \hat{\epsilon}_{\mathbf{k},s} \right) \hat{a}_{\mathbf{k},s} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega_k t)}, \quad (3.64)$$

while the negative frequency parts of the fields are essentially a collective creation operator,

$$\hat{\mathbf{E}}^{(-)}(\mathbf{r}, t) = \left[\hat{\mathbf{E}}^{(+)}(\mathbf{r}, t) \right]^\dagger, \quad (3.65)$$

$$\hat{\mathbf{B}}^{(-)}(\mathbf{r}, t) = \left[\hat{\mathbf{B}}^{(+)}(\mathbf{r}, t) \right]^\dagger. \quad (3.66)$$

3.1.3 Quadrature operators of the electric field

When a pair of operators does not commute, a corresponding uncertainty relation emerges. In general, for two Hermitian operators, $\hat{A}$ and $\hat{B}$, it can be shown through the Cauchy-Bunyakovsky-Schwarz⁹ inequality [317, 318, 319] that

$$\langle (\Delta \hat{A})^2 \rangle \langle (\Delta \hat{B})^2 \rangle \geq \frac{1}{4} \langle (\hat{C})^2 \rangle \quad (3.67)$$

where $i\hat{C} = [\hat{A}, \hat{B}]$. Operators $\hat{A}$ and $\hat{B}$ are often referred to as quadrature operators¹⁰. Our position and momentum observables, $\hat{q}$ and $\hat{p}$, are quadrature operators with an

⁹Also often called the Cauchy-Schwarz inequality.

¹⁰The word quadrature refers to the process of *squaring*. Here, the $\hat{q}$ and $\hat{p}$ operators allow us to write the Hamiltonian as a sum of squares!

uncertainty relation $\Delta\hat{q}\Delta\hat{p} \geq \frac{\hbar}{2}$. In quantum optics, it is common to instead work with the quadrature operators,

$$\hat{X}_1 = \frac{1}{2} (\hat{a} + \hat{a}^\dagger) \quad (3.68)$$

$$\hat{X}_2 = \frac{1}{2i} (\hat{a} - \hat{a}^\dagger), \quad (3.69)$$

which are essentially dimensionless position and momentum operators. The single-mode electric field operator,

$$\hat{E}_x(z, t) = \left(\frac{\hbar\omega}{\epsilon_0 V} \right)^{1/2} \sin(kz) (\hat{a}e^{-i\omega t} + \hat{a}^\dagger e^{i\omega t}), \quad (3.70)$$

can be re-written in terms of the quadrature operators as

$$\hat{E}_x(z, t) = 2 \left(\frac{\hbar\omega}{\epsilon_0 V} \right)^{1/2} \sin(kz) [\hat{X}_1 \cos(\omega t) + \hat{X}_2 \sin(\omega t)]. \quad (3.71)$$

Evidently, the quadrature operators, $\hat{X}_1$ and $\hat{X}_2$, are associated with the field amplitudes oscillating in and out of phase with each other by $\pi/2$. The quadrature operators satisfy the following commutation relation,

$$[\hat{X}_1, \hat{X}_2] = \frac{i}{2}, \quad (3.72)$$

and so it can be shown that

$$\langle (\Delta\hat{X}_1)^2 \rangle \langle (\Delta\hat{X}_2)^2 \rangle \geq \frac{1}{16}. \quad (3.73)$$

We will revisit these quadrature operators for the various states of light considered in the following section in order to better understand their properties.

3.1.4 Different states of light

In the previous section, we demonstrated how the normal modes of the electromagnetic field can be quantized, and how this description leads us to the definition

of the photon. We now turn our attention towards the distribution of electromagnetic excitations (photons) across the normal mode solutions to Maxwell's equations, otherwise referred to as *states* of light. We will see that certain states of light are classical, while others are nonclassical. The future of photonic quantum computing is expected to necessarily make use of nonclassical (quantum) states of light, and in particular, states which display entanglement between many degrees of freedom (*i.e.*, multimode quantum states)¹¹. The remainder of this section discusses several important states of light.

Number (Fock) states

We begin our exploration of quantum states with something familiar, returning our attention to the energy eigenstates of the Hamiltonian (presented in Eq. 3.36, but rewritten here for convenience),

$$\hat{\mathcal{H}} |n\rangle = \hbar\omega \left(\hat{a}^\dagger \hat{a} + \frac{1}{2} \right) |n\rangle = E_n |n\rangle. \quad (3.74)$$

Note that here we consider a single mode of the electromagnetic field for simplicity and we now write the energy eigenstate as $|n\rangle$; the reason for the latter will soon become apparent. We recall that we defined the energy eigenstate of the annihilation operator as

$$\hat{\mathcal{H}} [\hat{a} |n\rangle] = (E_n - \hbar\omega) [\hat{a} |n\rangle], \quad (3.75)$$

which implies that the state

$$C_n |n-1\rangle = \hat{a} |n\rangle, \quad (3.76)$$

where C_n is a normalization constant, is also an energy eigenstate, but one with a reduced energy eigenvalue,

$$E_{n-1} = E_n - \hbar\omega. \quad (3.77)$$

¹¹I recommend the very nice review article (originally assigned as reading material for my comprehensive exam by Jeff Lundeen), *Modes and states in quantum optics*, by Fabre and Treps [320] which outlines modes and states in quantum optics, and discusses how the two are intricately related.

This same procedure can be applied n times, decreasing the energy of the state in increments of $\hbar\omega$, until reaching

$$\hat{\mathcal{H}}[\hat{a}|0\rangle] = (E_0 - \hbar\omega)[\hat{a}|0\rangle], \quad (3.78)$$

where we have previously defined $E_0 = \frac{1}{2}\hbar\omega$ as the ground state energy and $\hat{a}|0\rangle = 0$. The state $|0\rangle$ is generally called the vacuum state. It follows from Eq. 3.77 that

$$E_n = \left(n + \frac{1}{2}\right) \hbar\omega. \quad (3.79)$$

By comparing Eq. 3.79 with Eq. 3.74, we find that

$$\hat{a}^\dagger \hat{a} |n\rangle = n |n\rangle \quad (3.80)$$

must hold; in other words, there exists another operator, $\hat{n} = \hat{a}^\dagger \hat{a}$, for which $|n\rangle$ is an eigenstate. We refer to the operator, $\hat{n}$, as the *number operator*, and the eigenstates, $|n\rangle$, as the *number (Fock) states*. These states must be normalized according to $\langle n|n\rangle = 1$. We can solve for the normalization constant, C_n , as follows

$$\begin{aligned} \left(\langle n|\hat{a}^\dagger\right)\left(\hat{a}|n\rangle\right) &= \langle n-1|C_n^* C_n |n-1\rangle = \langle n|n|n\rangle \\ |C_n|^2 \langle n-1|n-1\rangle &= n \langle n|n\rangle \\ C_n &= \sqrt{n}. \end{aligned} \quad (3.81)$$

We can follow a similar normalization procedure for the creation operator, where

$$D_n |n+1\rangle = \hat{a}^\dagger |n\rangle, \quad (3.82)$$

allows us to write

$$\begin{aligned} \left(\langle n|\hat{a}\right)\left(\hat{a}^\dagger |n\rangle\right) &= \langle n+1|D_n^* D_n |n+1\rangle = \langle n|\left(\hat{a}^\dagger \hat{a} + 1\right)|n\rangle \\ |D_n|^2 \langle n+1|n+1\rangle &= (n+1) \langle n|n\rangle \\ D_n &= \sqrt{n+1}, \end{aligned} \quad (3.83)$$

and we have the result that

$$\hat{a} |n\rangle = \sqrt{n} |n-1\rangle \quad (3.84)$$

$$\hat{a}^\dagger |n\rangle = \sqrt{n+1} |n+1\rangle. \quad (3.85)$$

We can therefore write the number states in terms of the vacuum state, by repeated use of the creation operator,

$$|n\rangle = \frac{(\hat{a}^\dagger)^n}{\sqrt{n!}} |0\rangle. \quad (3.86)$$

Since $\hat{\mathcal{H}}$ and $\hat{n}$ are Hermitian operators, different number states are orthogonal, *i.e.*, $\langle n'|n\rangle = \delta_{n,n'}$. Furthermore, the number states form a complete set,

$$\sum_{n=0}^{\infty} |n\rangle \langle n| = \mathbb{1}, \quad (3.87)$$

where $\mathbb{1}$ is the identity matrix.

The number state, $|n\rangle$, is well-defined in terms of energy; however, it is not well-defined in terms of its electric field. The expectation value of the electric field operator, as defined in Eq. 3.61, for a single-mode electric field (propagating in the z -direction, and polarized in the x -direction) is

$$\langle n | \hat{E}_x(z, t) | n \rangle = i \left(\frac{\hbar\omega}{2\epsilon_0 V} \right)^{1/2} \left[e^{i(kz-\omega t)} \langle n | \hat{a} | n \rangle - e^{-i(kz-\omega t)} \langle n | \hat{a}^\dagger | n \rangle \right] = 0, \quad (3.88)$$

implying a mean field of zero. Any fluctuations in the field about this mean field of zero will be given by its variance,

$$\langle (\Delta \hat{E}_x(z, t))^2 \rangle = \langle \hat{E}_x^2(z, t) \rangle - \langle \hat{E}_x(z, t) \rangle^2, \quad (3.89)$$

where we have already shown that $\langle \hat{E}_x(z, t) \rangle^2 = 0$. Meanwhile, the expectation value

of the square of the electric field—which contributes to the energy density—is

$$\begin{aligned}
 \langle n | \hat{E}_x^2(z, t) | n \rangle &= (i)^2 \left(\frac{\hbar\omega}{2\epsilon_0 V} \right) \langle n | \left(\hat{a}^2 e^{2i(kz-\omega t)} - \hat{a}\hat{a}^\dagger - \hat{a}^\dagger\hat{a} + \hat{a}^{\dagger 2} e^{-2i(kz-\omega t)} \right) | n \rangle \\
 &= \left(\frac{\hbar\omega}{2\epsilon_0 V} \right) \langle n | (\hat{a}\hat{a}^\dagger + \hat{a}^\dagger\hat{a}) | n \rangle = \left(\frac{\hbar\omega}{2\epsilon_0 V} \right) \langle n | (2\hat{a}^\dagger\hat{a} + 1) | n \rangle \\
 &= \left(\frac{\hbar\omega}{2\epsilon_0 V} \right) (2n + 1) \langle n | n \rangle = \left(\frac{\hbar\omega}{\epsilon_0 V} \right) \left(n + \frac{1}{2} \right), \tag{3.90}
 \end{aligned}$$

and so the standard deviation (uncertainty) in the field is equal to

$$\begin{aligned}
 \Delta \hat{E}_x &= \sqrt{\langle n | \hat{E}_x^2 | n \rangle} \\
 &= \left(\frac{\hbar\omega}{\epsilon_0 V} \right)^{1/2} \left(n + \frac{1}{2} \right)^{1/2}. \tag{3.91}
 \end{aligned}$$

Evidently, there are fluctuations in the electric field about its zero ensemble average. We note that there are also fluctuations for the vacuum state, $|0\rangle$; these *vacuum fluctuations* are responsible for many interesting quantum phenomena, including the stimulation of an atom to emit a photon spontaneously¹².

From the quadrature operators, as defined in Section 3.1.3, we see that

$$\langle (\Delta \hat{X}_1)^2 \rangle = \langle (\Delta \hat{X}_2)^2 \rangle = \frac{1}{4} (2n + 1). \tag{3.92}$$

Thus, for a number state, both quadratures have the same uncertainty. Moreover, the vacuum state minimizes the quadrature uncertainty, since

$$\langle (\Delta \hat{X}_1)^2 \rangle_{\text{vacuum}} = \langle (\Delta \hat{X}_2)^2 \rangle_{\text{vacuum}} = \frac{1}{4}. \tag{3.93}$$

The phase-space diagrams for the vacuum and photon number states can be seen in Figs. 3.2(a) and (b), respectively.

We can extend our description of the number states to the multimode picture by defining the number state across N modes as

$$|n\rangle_1 \otimes |n\rangle_2 \otimes \cdots \otimes |n\rangle_N = |n_1, n_2, \dots, n_N\rangle = |\{n_N\}\rangle. \tag{3.94}$$

¹²We think back to Chapter 2 where we discussed spontaneous parametric processes, but could not explain them with classical optics!

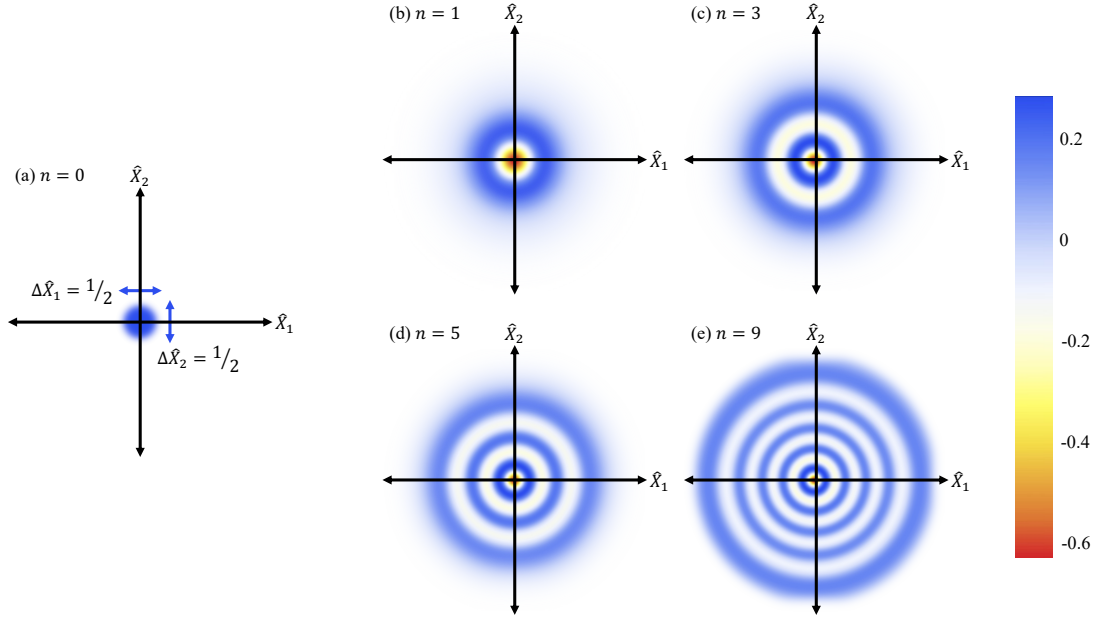


Figure 3.2: **Quadrature representation of the vacuum and number states.**

The phase-space representation for (a) the vacuum state ($n = 0$) and the number states with (b) $n = 1$, (c) $n = 3$, (d) $n = 5$, and (e) $n = 9$. Since the number states are eigenstates of the quantum harmonic oscillator, these solutions take the form of Hermite-Gaussian functions.

The Hamiltonian operates on this multimode number state as

$$\mathcal{H} |\{n_N\}\rangle = E |\{n_N\}\rangle, \quad (3.95)$$

where

$$E = \sum_{j=1}^N \hbar \omega_j \left(n_j + \frac{1}{2} \right), \quad (3.96)$$

and the action of the creation and annihilation operators are as follows

$$\hat{a}_j |n_1, n_2, \dots, n_j, \dots, n_N\rangle = \sqrt{n_j} |n_1, n_2, \dots, (n_j - 1), \dots, n_N\rangle \quad (3.97)$$

$$\hat{a}_j^\dagger |n_1, n_2, \dots, n_j, \dots, n_N\rangle = \sqrt{n_j + 1} |n_1, n_2, \dots, (n_j + 1), \dots, n_N\rangle. \quad (3.98)$$

Any number state can be generated from the multimode vacuum state, $|\{0\}\rangle = |0_1, 0_2, \dots, 0_N\rangle$, as

$$|\{n_N\}\rangle = \prod_{j=1}^N \frac{(\hat{a}_j^\dagger)^{n_j}}{\sqrt{n_j!}} |\{0\}\rangle. \quad (3.99)$$

Coherent states

Following the first laser demonstrations, there was a great deal of interest in studying the states of light which most closely describe such classical electromagnetic fields. As is often the case in quantum mechanics, we seek a state of the quantum harmonic oscillator which most closely resembles a classical harmonic oscillator. Though it might be tempting to simply take a number state in the limit of large n , we recall that $\langle \hat{E}_x(z, t) \rangle = 0$, even in this limit; we must therefore seek a different quantum state which satisfies the correspondence principle. First demonstrated by Schrödinger in 1926 [321], as a quantum state solution to the linear harmonic oscillator, the *coherent state* provides such a solution.

We first define this coherent state of the radiation field, $|\alpha\rangle$, as an eigenstate of the annihilation operator with complex eigenvalue α , thus satisfying

$$\hat{a} |\alpha\rangle = \alpha |\alpha\rangle \quad (3.100)$$

$$\langle \alpha | \hat{a}^\dagger = \alpha^* \langle \alpha |. \quad (3.101)$$

These states can be expanded in terms of the number operators¹³ (since the number operators form a complete set) as

$$|\alpha\rangle = \sum_{n=0}^{\infty} C_n |n\rangle, \quad (3.102)$$

where operation on this state with $\hat{a}$ gives

$$\hat{a} |\alpha\rangle = \sum_{n=0}^{\infty} C_n \sqrt{n} |n-1\rangle = \alpha \sum_{n=0}^{\infty} C_n |n\rangle. \quad (3.103)$$

¹³Again, we consider a single-mode field, for simplicity.

The coefficients of each term in the sums of Eq. 3.103 must be equivalent,

$$C_n \sqrt{n} = \alpha C_{n-1}, \quad (3.104)$$

and we arrive at the recurrence relation,

$$\begin{aligned} C_n &= \frac{\alpha}{\sqrt{n}} C_{n-1} = \frac{\alpha^2}{\sqrt{n(n-1)}} C_{n-2} = \dots \\ &= \frac{\alpha^n}{\sqrt{n!}} C_0. \end{aligned} \quad (3.105)$$

With this, we can write the coherent state as

$$|\alpha\rangle = C_0 \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle, \quad (3.106)$$

where the normalization requirement allows us to find C_0 ,

$$\begin{aligned} \langle \alpha | \alpha \rangle = 1 &= |C_0|^2 \sum_{n=0}^{\infty} \sum_{n'=0}^{\infty} \frac{\alpha^{*n} \alpha^{n'}}{\sqrt{n! n'!}} \langle n | n' \rangle \\ 1 &= |C_0|^2 \sum_{n=0}^{\infty} \frac{|\alpha|^{2n}}{n!} = |C_0|^2 e^{|\alpha|^2} \\ C_0 &= \exp\left(-\frac{1}{2}|\alpha|^2\right), \end{aligned} \quad (3.107)$$

and our normalized coherent states are thus written

$$|\alpha\rangle = \exp\left(-\frac{|\alpha|^2}{2}\right) \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle. \quad (3.108)$$

We can now consider the expectation value of the single-mode, linearly polarized electric field operator, as we did for the number states,

$$\begin{aligned} \langle \alpha | \hat{E}_x(z, t) | \alpha \rangle &= i \left(\frac{\hbar \omega}{2 \epsilon_0 V} \right)^{1/2} \left[e^{i(kz - \omega t)} \langle \alpha | \hat{a} | \alpha \rangle - e^{-i(kz - \omega t)} \langle \alpha | \hat{a}^\dagger | \alpha \rangle \right] \\ &= i \left(\frac{\hbar \omega}{2 \epsilon_0 V} \right)^{1/2} \left[\alpha e^{i(kz - \omega t)} - \alpha^* e^{-i(kz - \omega t)} \right]. \end{aligned} \quad (3.109)$$

Introducing the polar form, $\alpha = |\alpha|e^{i\theta}$, we can write

$$\langle \alpha | \hat{E}_x(z, t) | \alpha \rangle = -2|\alpha| \left(\frac{\hbar\omega}{2\epsilon_0 V} \right)^{1/2} \sin(kz - \omega t + \theta), \quad (3.110)$$

and we see that the mean electric field looks classical. We can also show that

$$\langle \alpha | \hat{E}_x^2(z, t) | \alpha \rangle = \left(\frac{\hbar\omega}{2\epsilon_0 V} \right) [4|\alpha|^2 \sin^2(kz - \omega t + \theta) + 1], \quad (3.111)$$

and so the uncertainty in the field is

$$\begin{aligned} \sqrt{\langle (\Delta \hat{E}_x(z, t))^2 \rangle} &= \sqrt{\langle \hat{E}_x^2(z, t) \rangle - \langle \hat{E}_x(z, t) \rangle^2} \\ &= \left(\frac{\hbar\omega}{2\epsilon_0 V} \right)^{1/2}, \end{aligned} \quad (3.112)$$

which is identical to the uncertainty of the vacuum state. The coherent state is a nearly classical-like state, as it yields the expected form of the mean field and contains only the noise of the vacuum. This result can also be seen when considering the uncertainty in the quadrature operators for the coherent state. We can show that

$$\langle (\Delta \hat{X}_1)^2 \rangle_\alpha = \langle (\Delta \hat{X}_2)^2 \rangle_\alpha = \frac{1}{4}, \quad (3.113)$$

and again arrive at the conclusion that coherent states minimize the uncertainty relation of the quadrature operators, and exhibit equal uncertainties to those of the vacuum state in each quadrature. The phase-space diagram for the coherent state is shown in Fig. 3.3(a), where we note that the uncertainty is independent of α . For this reason, the phase uncertainty, $\Delta\theta$ decreases as $|\alpha|$ increases. Fig. 3.3(b) depicts the time evolution of the $\hat{X}_1$ quadrature operator for the coherent state.

The physical importance of the parameter α can be understood by considering the expectation value of the number operator,

$$\langle \alpha | \hat{n} | \alpha \rangle = \langle \alpha | \hat{a}^\dagger \hat{a} | \alpha \rangle = |\alpha|^2, \quad (3.114)$$

where we see that $|\alpha|^2$ is just the mean photon number of the coherent state. We can

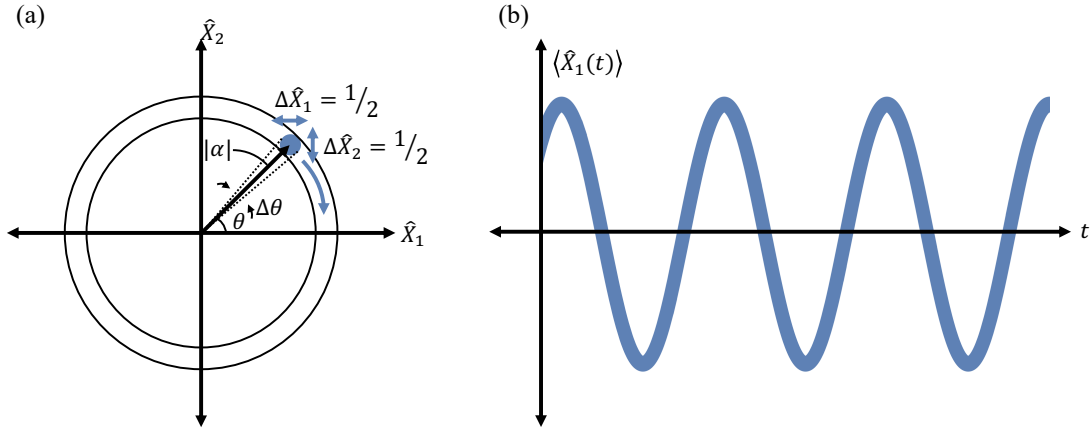


Figure 3.3: **Quadrature representation of the coherent state.** The phase-space representation for a coherent state of amplitude $|\alpha|$ and phase angle θ (with uncertainty $\Delta\theta$) is shown in (a), while the time evolution of the $\hat{X}_1$ quadrature for the coherent state is shown in (b).

calculate the fluctuations in the mean photon number, μ_α , by first determining

$$\begin{aligned}\langle \alpha | \hat{n}^2 | \alpha \rangle &= \langle \alpha | \hat{a}^\dagger \hat{a} \hat{a}^\dagger \hat{a} | \alpha \rangle = \langle \alpha | (\hat{a}^\dagger \hat{a}^\dagger \hat{a} \hat{a} + \hat{a}^\dagger \hat{a}) | \alpha \rangle \\ &= |\alpha|^4 + |\alpha|^2,\end{aligned}\tag{3.115}$$

which leads us to

$$\begin{aligned}\Delta n &= \sqrt{\langle \alpha | \hat{n}^2 | \alpha \rangle - (\langle \alpha | \hat{n} | \alpha \rangle)^2} \\ &= \sqrt{|\alpha|^4 + |\alpha|^2 - |\alpha|^4} = \sqrt{\mu_\alpha}.\end{aligned}\tag{3.116}$$

Thus, the fluctuations in the mean photon number are equivalent to the square root of the mean photon number—a characteristic of Poissonian statistics. In fact, the probability of detecting n photons in the coherent state field is

$$P_n = |\langle n | \alpha \rangle|^2 = e^{-|\alpha|^2} \frac{|\alpha|^{2n}}{n!},\tag{3.117}$$

which takes the form of a Poissonian distribution with a mean of $\mu_\alpha = |\alpha|^2$. This distribution is shown in Figs. 3.4(a), (b), and (c) for $\mu_\alpha = 0.1$, $\mu_\alpha = 1$, and $\mu_\alpha = 3$.

Coherent states, which can be used to describe laser light, are quantum states that

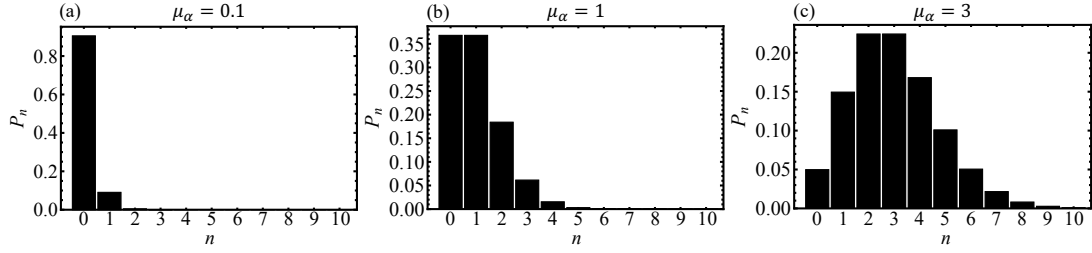


Figure 3.4: **Probability of detecting n photons in a coherent state.** Depicted for a mean photon number of (a) $\mu_\alpha = 0.1$, (b) $\mu_\alpha = 1$, and (c) $\mu_\alpha = 3$.

are very close to classical states: the expectation value of the field has the same form as the classical expression, the fluctuations in the electric field are the same as for vacuum, the fractional uncertainty decreases with increasing mean photon number, and the states become well localized in phase for a large mean photon number. Despite their nearly classical properties, these are still technically quantum states; the field clearly has the classical form of a sinusoidal wave, but with quantum fluctuations of the field superimposed. This implies some quantum features of the state. All states of light must have some quantum features, according to the quantum theory of light; however, it is not always possible to observe these quantum features—to make use of the quantum features of light, we require states of light which provide us access to these features.

Squeezed states

Previously, it was emphasized that all states of light are quantum mechanical, deriving quantum mechanical features from the discreteness of the photon; however, in practice, the nonclassical features of light are difficult to observe and exploit. One particularly important example of a nonclassical state in which the nonclassicality can be observed is the *squeezed state* of light.

We return our attention to the quadrature operators of the electric field, where a state is said to be squeezed if

$$\langle(\Delta\hat{X}_1)^2\rangle < \frac{1}{4} \quad \text{or} \quad \langle(\Delta\hat{X}_2)^2\rangle < \frac{1}{4}, \quad (3.118)$$

where we emphasize that, due to the uncertainty relation for the quadrature operators,

these two expressions cannot both simultaneously hold. States for which one of these inequalities holds will have a lower uncertainty (*i.e.*, less noise) in one quadrature than for a coherent state or a vacuum state. We can say that the fluctuations in this quadrature are *squeezed* (while the fluctuations in the other quadrature must be enhanced).

One way of generating a squeezed state is through the squeezing operator (which corresponds to the two-photon Hamiltonian of a degenerate nonlinear parametric process),

$$\hat{S} = \exp \left(\frac{1}{2} \zeta^* \hat{a}^2 - \frac{1}{2} \zeta \hat{a}^{\dagger 2} \right), \quad (3.119)$$

where $\zeta = r e^{i\theta}$ is a complex number, and r is referred to as the squeezing parameter. Using the Baker-Campbell-Hausdorff lemma, we can write

$$\hat{S}^\dagger(\zeta) \hat{a} \hat{S}(\zeta) = \hat{a} \cosh(r) - \hat{a}^\dagger e^{i\theta} \sinh(r) \quad (3.120)$$

$$\hat{S}^\dagger(\zeta) \hat{a}^\dagger \hat{S}(\zeta) = \hat{a}^\dagger \cosh(r) - \hat{a} e^{-i\theta} \sinh(r), \quad (3.121)$$

where $\hat{S}^\dagger(\zeta) = \hat{S}(-\zeta)$. For an arbitrary state, $|\psi\rangle$, we define

$$|\psi_s\rangle = \hat{S}(\zeta) |\psi\rangle, \quad (3.122)$$

where $|\psi_s\rangle$ is the state obtained after action by the squeezing operator. We can then obtain the variances of quadrature operators for this new state, by evaluating

$$\langle \psi_s | \hat{a} | \psi_s \rangle = \langle \psi | \hat{S}^\dagger(\zeta) \hat{a} \hat{S}(\zeta) | \psi \rangle \quad (3.123)$$

$$\langle \psi_s | \hat{a}^\dagger | \psi_s \rangle = \langle \psi | \hat{S}^\dagger(\zeta) \hat{a}^\dagger \hat{S}(\zeta) | \psi \rangle \quad (3.124)$$

$$\langle \psi_s | \hat{a}^2 | \psi_s \rangle = \langle \psi | \hat{S}^\dagger(\zeta) \hat{a} \hat{S}(\zeta) \hat{S}^\dagger(\zeta) \hat{a} \hat{S}(\zeta) | \psi \rangle \quad (3.125)$$

$$\langle \psi_s | \hat{a}^{\dagger 2} | \psi_s \rangle = \langle \psi | \hat{S}^\dagger(\zeta) \hat{a}^\dagger \hat{S}(\zeta) \hat{S}^\dagger(\zeta) \hat{a}^\dagger \hat{S}(\zeta) | \psi \rangle. \quad (3.126)$$

For the case where $|\psi\rangle$ is the vacuum state, $|0\rangle$, action of the squeezing operator generates a squeezed vacuum state which we write as

$$|\zeta\rangle = \hat{S}(\zeta) |0\rangle. \quad (3.127)$$

For this squeezed vacuum state, we calculate the quadrature variances as

$$\langle(\Delta\hat{X}_1)^2\rangle = \frac{1}{4} [\cosh^2(r) + \sinh^2(r) - 2\sinh(r)\cosh(r)\cos(\theta)] \quad (3.128)$$

$$\langle(\Delta\hat{X}_2)^2\rangle = \frac{1}{4} [\cosh^2(r) + \sinh^2(r) + 2\sinh(r)\cosh(r)\cos(\theta)]. \quad (3.129)$$

We note that for $\theta = 0$, we have

$$\langle(\Delta\hat{X}_1)^2\rangle = \frac{1}{4}e^{-2r} \quad (3.130)$$

$$\langle(\Delta\hat{X}_2)^2\rangle = \frac{1}{4}e^{2r}, \quad (3.131)$$

and we see squeezing in the $\hat{X}_1$ quadrature. On the other hand, for $\theta = \pi$,

$$\langle(\Delta\hat{X}_1)^2\rangle = \frac{1}{4}e^{2r} \quad (3.132)$$

$$\langle(\Delta\hat{X}_2)^2\rangle = \frac{1}{4}e^{-2r}, \quad (3.133)$$

and the squeezing now appears in the $\hat{X}_2$ quadrature. We note that, in either of these two cases, $\langle(\Delta\hat{X}_1)^2\rangle\langle(\Delta\hat{X}_2)^2\rangle = 1/16$, and the squeezed vacuum state minimizes the quadrature uncertainty relation¹⁴. Squeezing can be visualized in phase space, as in Fig. 3.5(a), where the corresponding time evolution is depicted in Fig. 3.5(b).

Squeezing need not necessarily occur along $\hat{X}_1$ or $\hat{X}_2$. We can demonstrate this by defining the rotated quadrature operators,

$$\begin{pmatrix} \hat{Y}_1 \\ \hat{Y}_2 \end{pmatrix} = \begin{pmatrix} \cos(\theta/2) & \sin(\theta/2) \\ -\sin(\theta/2) & \cos(\theta/2) \end{pmatrix} \begin{pmatrix} \hat{X}_1 \\ \hat{X}_2 \end{pmatrix}, \quad (3.134)$$

which have variances of

$$\langle(\Delta\hat{Y}_1)^2\rangle = \frac{1}{4}e^{-2r} \quad (3.135)$$

$$\langle(\Delta\hat{Y}_2)^2\rangle = \frac{1}{4}e^{2r}, \quad (3.136)$$

¹⁴We note that squeezed states need not minimize the uncertainty relation.

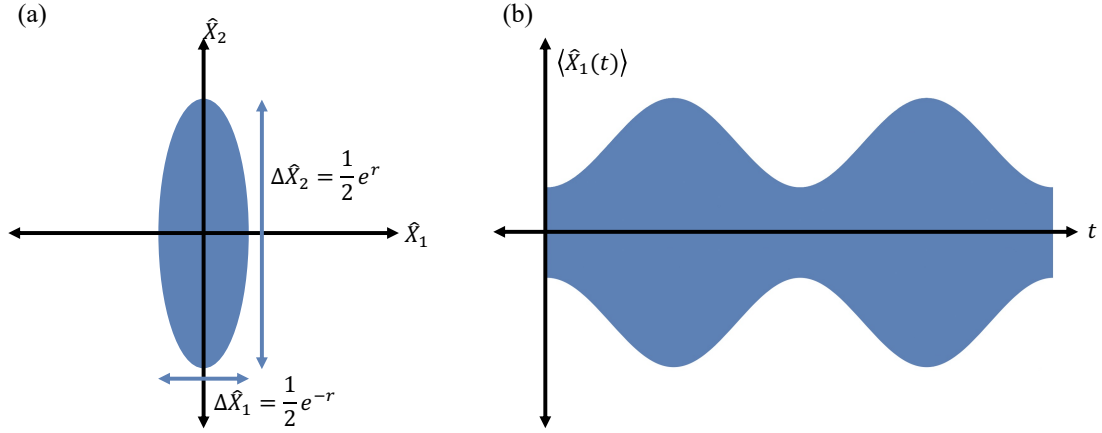


Figure 3.5: **Quadrature representation of a squeezed vacuum state.** The phase-space representation for a squeezed vacuum state is shown in (a), while the time evolution of the $\hat{X}_1$ quadrature is shown in (b).

and we see that squeezing can occur in any rotated coordinate system for the quadrature operators.

For the single-mode electric field defined in terms of the quadratures as in Eq. 3.70, we have the following commutation relation

$$\left[\hat{E}_x(z, \omega t = 0), \hat{E}_x(z, \omega t)\right] = i \left(\frac{\hbar\omega}{\epsilon_0 V}\right) \sin^2(kz) \sin(\omega t), \quad (3.137)$$

which allows us to write the uncertainty relation between the electric fields at different phases

$$\langle(\Delta\hat{E}_x(z, \omega t = 0))^2\rangle \langle(\Delta\hat{E}_x(z, \omega t))^2\rangle \geq \frac{1}{4} \left(\frac{\hbar\omega}{\epsilon_0 V}\right) \sin^2(kz) \sin(\omega t). \quad (3.138)$$

This is unlike our results for the number states and coherent states, and we will soon see it is also unlike the result for thermal states. We see that, for squeezed states of light, if the electric field uncertainty is squeezed for a phase $\omega t = 0$, then the uncertainty in the field must be enhanced for a phase of ωt . In other words, squeezed light contains phase-dependent noise, reduced below the noise of the vacuum for some phases while enhanced above the vacuum for others. This result is particularly useful in the context of measuring weak signals such as gravitational waves [322].

We next show how the squeezed vacuum state, $|\zeta\rangle$, can be decomposed in terms of the number states,

$$|\zeta\rangle = \sum_{n=0}^{\infty} C_n |n\rangle. \quad (3.139)$$

Since we already know that $\hat{a}|0\rangle = 0$, we can write

$$\begin{aligned} \hat{S}\hat{a}\hat{S}^\dagger\hat{S}|0\rangle &= 0 \\ \hat{S}\hat{a}\hat{S}^\dagger|\zeta\rangle &= 0. \end{aligned} \quad (3.140)$$

By the Baker-Campbell-Hausdorff lemma, we have that

$$\hat{S}\hat{a}\hat{S}^\dagger = \hat{a}\cosh(r) + \hat{a}^\dagger e^{i\theta}\sinh(r), \quad (3.141)$$

where we define $\mu = \cosh(r)$ and $\nu = e^{i\theta}\sinh(r)$, and can thus write

$$(\mu\hat{a} + \nu\hat{a}^\dagger)|\zeta\rangle = 0. \quad (3.142)$$

Inserting Eq. 3.142 into Eq. 3.139, we have that

$$(\mu\hat{a} + \nu\hat{a}^\dagger) \sum_{n=0}^{\infty} C_n |n\rangle = 0, \quad (3.143)$$

which gives rise to the recurrence relation,

$$C_{n+1} = -\frac{\nu}{\mu} \left(\frac{n}{n+1} \right)^{1/2} C_{n-1}, \quad (3.144)$$

which connects only every other photon number state. There are two distinct solutions to the recurrence relation in Eq. 3.144, which involve either the odd number states or the even number states. We focus our attention on the solution for the even photon number states,

$$C_{2m} = (-1)^m (e^{i\theta}\tanh(r))^m \left[\frac{(2m-1)!!}{(2m)!!} \right]^{1/2} C_0, \quad (3.145)$$

because this is the only set of solutions containing the vacuum state, which is of interest here. The normalization condition,

$$\sum_{m=0}^{\infty} |C_{2m}|^2 = 1, \quad (3.146)$$

can be used to determine C_0 . We see that we must satisfy

$$|C_0|^2 \left(1 + \sum_{m=0}^{\infty} \frac{(\tanh^{2m}(r))(2m-1)!!}{(2m)!!} \right) = 1. \quad (3.147)$$

We can then make use of the identity,

$$1 + \sum_{m=0}^{\infty} x^m \frac{(2m-1)!!}{(2m)!!} = (1-x)^{-1/2}, \quad (3.148)$$

to show that $C_0 = \sqrt{\cosh(r)}$. This result, together along with the identities,

$$(2m)!! = 2^m m! \quad (3.149)$$

$$(2m-1)!! = \frac{1}{2^m} \frac{(2m)!}{m!}, \quad (3.150)$$

allows us to write the squeezed vacuum state as

$$|\zeta\rangle = \frac{1}{\sqrt{\cosh(r)}} \sum_{m=0}^{\infty} (-1)^m \frac{\sqrt{(2m)!}}{2^m m!} e^{i\theta \tanh^m(r)} |2m\rangle. \quad (3.151)$$

From this result, we can calculate the probability of detecting $2m$ photons in the field as

$$P_{2m} = |\langle 2m | \zeta \rangle|^2 = \frac{(2m)!}{2^{2m} (m!)^2} \frac{\tanh^{2m}(r)}{\cosh(r)}, \quad (3.152)$$

while $P_{2m+1} = 0$. We see that, for a squeezed vacuum state, the photon probability distribution vanishes for all odd photon numbers. The photon number probability distributions for squeezing parameters of $r = 0.1$ and $r = 1$ are shown in Figs. 3.6(a) and (b), respectively. Aside from the oscillatory nature of the distribution, its overall

shape is similar to that of a thermal state (which we will describe later); but, it is important to remember that a squeezed vacuum state is a pure state, while a thermal state is a mixed state.

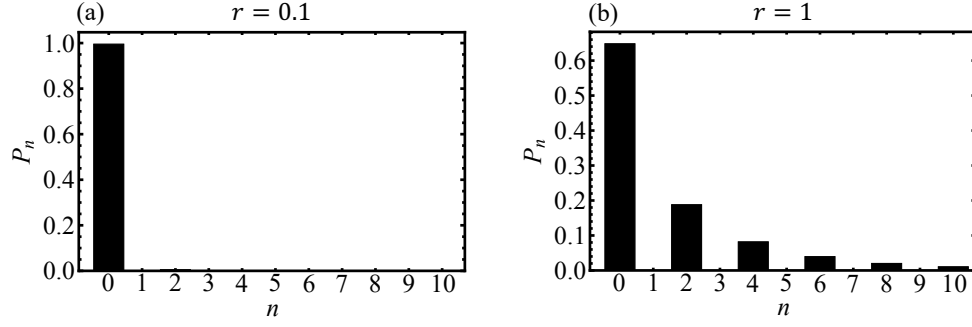


Figure 3.6: **Probability of detecting n photons in a squeezed vacuum state.** Depicted for a squeezing parameter of (a) $r = 0.1$ and (b) $r = 1$.

Two-mode squeezed vacuum (TMSV) states

We have just demonstrated the interesting nonclassical properties arising in the case of a single-mode squeezed state of light; however, the prospects for nonclassical effects become even richer when we extend our discussion to include *entangled* multimode field states. Here, we consider the simple, yet important, example of the two-mode squeezed vacuum (TMSV) state.

In analogy with our previous description of single-mode squeezing, we can introduce the two-mode squeezing operator,

$$\hat{S}_2(\zeta) = \exp \left(\zeta^* \hat{a} \hat{b} - \zeta \hat{a}^\dagger \hat{b}^\dagger \right), \quad (3.153)$$

where $\zeta = r e^{i\theta}$, as before, and where we now label our two modes with a and b (the creation and annihilation operators are denoted correspondingly). We can define the TMSV by the action of $\hat{S}_2(\zeta)$ on the two-mode vacuum state, $|0_a, 0_b\rangle$, such that

$$\begin{aligned} |\zeta\rangle_2 &= \hat{S}_2(\zeta) |0_a, 0_b\rangle \\ &= \exp \left(\zeta^* \hat{a} \hat{b} - \zeta \hat{a}^\dagger \hat{b}^\dagger \right) |0_a, 0_b\rangle. \end{aligned} \quad (3.154)$$

Importantly, $\hat{S}_2(\zeta)$ does not factor into a product of single-mode squeezing operators for each mode. As such, the TMSV state is not a product of two single-mode squeezed vacuum states, but rather an entangled state containing strong correlations between the two modes. The two-mode squeezing operator contains terms which either create or annihilate photons in a pair-wise manner. Also arising from the correlations between the two modes is the fact that the squeezing of quantum fluctuations occurs in a superposition of the two modes. This can be seen by defining the superposition quadrature operators,

$$\hat{X}_1^{(\text{TMSV})} = \frac{1}{2^{3/2}} \left(\hat{a} + \hat{a}^\dagger + \hat{b} + \hat{b}^\dagger \right) \quad (3.155)$$

$$\hat{X}_2^{(\text{TMSV})} = \frac{1}{2^{3/2}i} \left(\hat{a} - \hat{a}^\dagger + \hat{b} - \hat{b}^\dagger \right), \quad (3.156)$$

where the commutation relations as for the single-mode case remain satisfied. From the Baker-Campbell-Hausdorff lemma we obtain

$$\hat{S}_2^\dagger(\zeta) \hat{a} \hat{S}_2(\zeta) = \hat{a} \cosh(r) - e^{i\theta} \hat{b}^\dagger \sinh(r) \quad (3.157)$$

$$\hat{S}_2^\dagger(\zeta) \hat{b} \hat{S}_2(\zeta) = \hat{b} \cosh(r) - e^{i\theta} \hat{a}^\dagger \sinh(r). \quad (3.158)$$

Following much the same procedure as we did in the case of the single-mode squeezed vacuum state, the variances of the superposition quadratures, for the TMSV state with $\theta = 0$, are

$$\langle (\Delta \hat{X}_1^{(\text{TMSV})})^2 \rangle = \frac{1}{4} e^{-2r} \quad (3.159)$$

$$\langle (\Delta \hat{X}_2^{(\text{TMSV})})^2 \rangle = \frac{1}{4} e^{2r}. \quad (3.160)$$

Although these expressions are mathematically equivalent to those we derived for the single-mode squeezed vacuum state, it is important to remember that the interpretation here is in terms of the superposition of the two modes.

Following a procedure analogous to the one we used for the single-mode squeezed

state, the TMSV state can be decomposed into the two-mode number states as

$$|\zeta\rangle_2 = \frac{1}{\cosh(r)} \sum_{n=0}^{\infty} (-1)^n e^{in\theta} \tanh^n(r) |n_a, n_b\rangle. \quad (3.161)$$

Aside from the fact that this is an entangled state, another striking feature is the evident strong correlations between the two modes, a and b : only the paired states, $|n_a, n_b\rangle$, with the same number of photons in each mode are allowed to occur¹⁵. The average photon number in each mode is therefore the same, where

$$\langle \hat{n}_a \rangle = \langle \hat{n}_b \rangle = \sinh^2(r), \quad (3.162)$$

and the variances are given by

$$\langle (\Delta \hat{n}_a)^2 \rangle = \langle (\Delta \hat{n}_b)^2 \rangle = \sinh^2(r) \cosh^2(r) = \frac{1}{4} \sinh^2(2r). \quad (3.163)$$

We see that $\langle (\Delta \hat{n}_i) \rangle > \langle n_i \rangle, i \in \{a, b\}$, and so both modes exhibit super-Poissonian statistics.

The joint probability of finding n_1 photons in mode a and n_2 photons in mode b is given by

$$\begin{aligned} P_{n_1, n_2} &= |\langle n_1, n_2 | \zeta \rangle_2|^2 \\ &= \frac{\tanh^{2n}(r)}{\cosh^2(r)} \delta_{n_1, n} \delta_{n_2, n}. \end{aligned} \quad (3.164)$$

This joint probability is visualized for squeezing parameter $r = 0.1$ and $r = 1$ in Figs. 3.7(a) and (b), respectively. We see that the joint probability is a monotonically decreasing function along the diagonal, $n_1 = n_2$.

We can examine the individual properties of the two modes by introducing the density operator for the TMSV state,

$$\hat{\rho}_{ab} = |\zeta\rangle_2 {}_2\langle\zeta|, \quad (3.165)$$

¹⁵This is one reason why such states are often called *twin beams*.

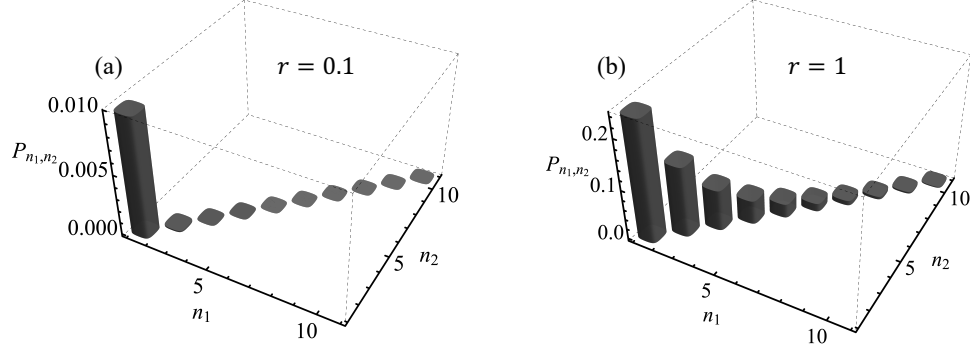


Figure 3.7: **Probability of detecting n_1 and n_2 photons in modes a and b of a two-mode squeezed vacuum (TMSV) state.** Depicted for a squeezing parameter of (a) $r = 0.1$ and (b) $r = 1$.

and the reduced density operators for the individual modes,

$$\hat{\rho}_a = \text{Tr}_b[\hat{\rho}_{ab}] = \sum_{n=0}^{\infty} \frac{\tanh^{2n}(r)}{\cosh^2(r)} |n\rangle_a \langle n| \quad (3.166)$$

$$\hat{\rho}_b = \text{Tr}_a[\hat{\rho}_{ab}] = \sum_{n=0}^{\infty} \frac{\tanh^{2n}(r)}{\cosh^2(r)} |n\rangle_b \langle n|, \quad (3.167)$$

where $\text{Tr}_i[\hat{\rho}_{ab}] = \sum_i \langle i | \hat{\rho}_{ab} | i \rangle$, $i \in \{a, b\}$. The physical interpretation behind the density operator (and its properties) will be discussed in the next subsection, where we consider thermal states. For now, we simply use it as a tool. The probability of finding n photons in a single mode, a or b , is given by

$$P_n^{(i)} = {}_i \langle n | \hat{\rho}_i | n \rangle_i = \frac{\tanh^{2n}(r)}{\cosh^2(r)}, \quad i \in \{a, b\}, \quad (3.168)$$

Taking into account Eq. 3.162, this can be written as

$$P_n^{(i)} = \frac{\langle \hat{n}_i \rangle^n}{1 + \langle \hat{n}_i \rangle^{n+1}} \quad i \in \{a, b\}, \quad (3.169)$$

which corresponds to a thermal distribution with $\langle \hat{n}_i \rangle = \sinh^2(r)$. In other words, when we measure only one of the radiation fields (*e.g.*, in mode a) and disregard the

other (*e.g.*, in mode b), then the radiation field which we measure (a) is indistinguishable from the radiation from a thermal source. We will make use of this result in Chapter 5.3.

Thermal states

We conclude our exploration of quantum states with another familiar source of light, this time turning our attention back to the radiation emitter which sparked the development of the quantum theory of radiation: the black body. A widely used black body model is a cavity with a small hole through which radiation can enter and perfectly absorbing walls, shown in Fig. 3.8. For a large enough cavity, radiation

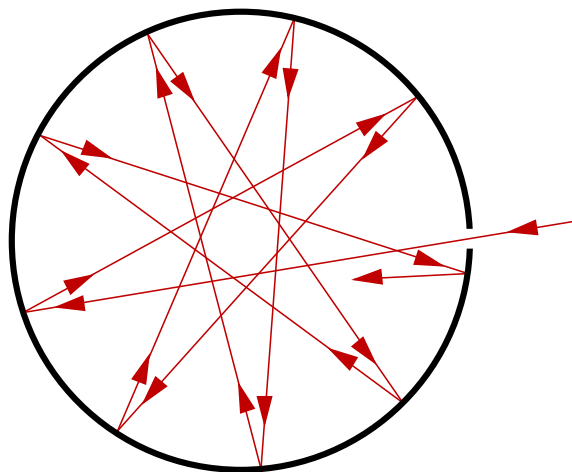


Figure 3.8: **Model of a black body.** A simple model of a black body, consisting of a cavity with a small hole through which light radiation can enter but is unlikely to exit. The cavity walls are taken to be perfect absorbers of radiation.

which enters the cavity is unlikely to exit. This lack of re-emission implies that the cavity acts as a nearly perfectly black surface. The cavity's hole will, of course, allow some radiation to escape; however, if the hole is small, the radiation which enters and exits the cavity will have a negligible effect on the system's equilibrium. If the cavity is held at a fixed temperature, T , and the radiation trapped within the cavity is at thermal equilibrium with the cavity, then the small amount of radiation which

is allowed to escape will approximate that of a black body. Such radiation will have an energy distribution that is characteristic of the temperature¹⁶.

In this black body model, the radiation is coupled to a heat bath and is thus not a truly free field; however, if we assume weak coupling, we can treat the field as an isolated system described by the microcanonical ensemble¹⁷. We first consider a single-mode field that is in equilibrium with the walls of a cavity held at temperature T . The probability of the mode being excited to the n^{th} level is given by the Boltzmann distribution [323],

$$P_n = \frac{\exp(-E_n/k_B T)}{\sum_n \exp(-E_n/k_B T)}, \quad (3.170)$$

where the energy levels are those of our photon number states, as in Eq. 3.79, and $k_B = 1.38 \times 10^{-23}$ J/K is the Boltzmann constant. In the quantum mechanical picture, this radiation is an ensemble of photons that can each be prepared in n different pure states; thus, we must now deal with the statistics of the possible state preparations. To do this, we introduce the density operator,

$$\hat{\rho} = \sum_j p_j |\psi_j\rangle \langle \psi_j|, \quad (3.171)$$

where each pure state, $|\psi_j\rangle$, is prepared with probability p_j . For a system in thermal equilibrium, if the eigenstate, $|\psi_n\rangle$, of the Hamiltonian, $\hat{\mathcal{H}}$, has energy E_n , then the relative probability for being in that state is given by $e^{-E_n/k_B T}$. The density matrix for this ensemble in thermal equilibrium can be written as

$$\hat{\rho}_{\text{Th}} = \frac{1}{Z} \sum_n e^{-E_n/k_B T} |\psi_n\rangle \langle \psi_n|, \quad (3.172)$$

where Z is the partition function, which is given by¹⁸

$$Z = \sum_n e^{-E_n/k_B T} = \text{Tr} \left[e^{-\hat{\mathcal{H}}/k_B T} \right]. \quad (3.173)$$

¹⁶Also, for wavelengths smaller than the cavity's hole, this distribution will not depend on the properties of the cavity.

¹⁷This follows from statistical mechanics.

¹⁸Here we make use of the fact that the trace of an operator, $\hat{A}$, is equivalent to the sum over its eigenvalues, λ_i , as $\text{Tr}[\hat{A}] = \sum_i \lambda_i$.

The eigenvalue expression for the Hamiltonian,

$$\hat{\mathcal{H}} |\psi_n\rangle = E_n |\psi_n\rangle \quad (3.174)$$

implies that

$$e^{-\hat{\mathcal{H}}/k_B T} |\psi_n\rangle = e^{-E_n/k_B T} |\psi_n\rangle. \quad (3.175)$$

Since the eigenvectors form a complete basis, we can write

$$|\phi\rangle = \sum_n |\psi_n\rangle \langle \psi_n | \phi \rangle, \quad (3.176)$$

and so we have that

$$e^{-\hat{\mathcal{H}}/k_B T} |\phi\rangle = \sum_n e^{-\hat{\mathcal{H}}/k_B T} |\psi_n\rangle \langle \psi_n | \phi \rangle = \sum_n e^{-E_n/k_B T} |\psi_n\rangle \langle \psi_n | \phi \rangle. \quad (3.177)$$

Thus, we can write the density matrix as

$$\hat{\rho}_{\text{Th}} = \frac{e^{-\hat{\mathcal{H}}/k_B T}}{\text{Tr} [e^{-\hat{\mathcal{H}}/k_B T}]}, \quad (3.178)$$

where $\hat{\mathcal{H}} = \hbar\omega (\hat{a}^\dagger \hat{a} + \frac{1}{2})$, and so our eigenstates, $|\psi_n\rangle$, are the number states, $|n\rangle$.

The partition function can be evaluated as

$$\begin{aligned} Z &= \text{Tr} \left[\exp \left(-\hat{\mathcal{H}}/k_B T \right) \right] = \sum_{n=0}^{\infty} \langle n | \exp \left(-\hat{\mathcal{H}}/k_B T \right) | n \rangle \\ &= \sum_{n=0}^{\infty} \exp (-E_n/k_B T) \\ &= \exp (-\hbar\omega/2k_B T) \sum_{n=0}^{\infty} \exp (-\hbar\omega n/k_B T). \end{aligned} \quad (3.179)$$

Using the geometric series identity,

$$\sum_{n=0}^{\infty} \exp(-\hbar\omega n/k_B T) = \frac{1}{1 - \exp(-\hbar\omega/k_B T)}, \quad (3.180)$$

we can write

$$Z = \frac{\exp(-\hbar\omega/2k_B T)}{1 - \exp(-\hbar\omega/k_B T)}. \quad (3.181)$$

The expectation value for an operator must now be defined as

$$\begin{aligned} \langle \hat{A} \rangle &= \sum_i p_i \langle i | \hat{A} | i \rangle = \sum_i p_i \langle i | \hat{I} \hat{A} \hat{I} | i \rangle \\ &= \sum_i p_i \langle i | \left(\sum_j |j\rangle \langle j| \right) \hat{A} \left(\sum_k |k\rangle \langle k| \right) | i \rangle \\ &= \sum_i p_i \sum_j \sum_k \langle i | j \rangle \langle j | \hat{A} | k \rangle \langle k | i \rangle = \sum_j \sum_k \langle k | \left(\sum_i p_i |i\rangle \langle i| \right) | j \rangle \langle j | \hat{A} | k \rangle \\ &= \sum_k \langle k | \hat{\rho} \left(\sum_j |j\rangle \langle j| \right) \hat{A} | k \rangle = \sum_k \langle k | \hat{\rho} \hat{A} | k \rangle = \text{Tr} [\hat{\rho} \hat{A}], \end{aligned} \quad (3.182)$$

which we make use of to calculate the average photon number of the thermal field as

$$\begin{aligned} \bar{n} = \langle \hat{n} \rangle &= \text{Tr} [\hat{\rho}_{\text{Th}} \hat{n}] = \sum_{n=0}^{\infty} \langle n | \hat{\rho}_{\text{Th}} \hat{n} | n \rangle = \sum_{n=0}^{\infty} n \langle n | \hat{\rho}_{\text{Th}} | n \rangle \\ &= \sum_{n=0}^{\infty} n \langle n | \left(\sum_{n'=0}^{\infty} p_{n'} |n'\rangle \langle n'|n\rangle \right) \\ &= \sum_{n=0}^{\infty} n p_n. \end{aligned} \quad (3.183)$$

The probability of finding n photons in the thermal field is

$$\begin{aligned} p_n &= \langle n | \hat{\rho}_{\text{Th}} | n \rangle = \langle n | \left(\frac{e^{-\hat{\mathcal{H}}/k_B T}}{Z} \right) | n \rangle \\ &= \frac{e^{-E_n/k_B T}}{Z}, \end{aligned} \quad (3.184)$$

and so the average photon number can be further evaluated as

$$\begin{aligned}\bar{n} &= \sum_{n=0}^{\infty} n p_n = \frac{1}{Z} \sum_{n=0}^{\infty} n e^{-E_n/k_B T} \\ &= \frac{e^{-\hbar\omega/2k_B T}}{Z} \sum_{n=0}^{\infty} n e^{-\hbar\omega n/k_B T}.\end{aligned}\quad (3.185)$$

We can set $x = \hbar\omega/k_B T$, and further simplify using the identity

$$\sum_{n=0}^{\infty} n e^{-nx} = \frac{e^{-x}}{(1 - e^{-x})^2}, \quad (3.186)$$

to arrive at

$$\bar{n} = \frac{1}{\exp(\hbar\omega/k_B T) - 1}. \quad (3.187)$$

With some rearrangement, it can be shown that the probability of finding n photons in the thermal field can be written in terms of the average photon number as

$$p_n = \frac{\bar{n}^n}{(1 + \bar{n})^{n+1}}. \quad (3.188)$$

Even for different values of average photon number, as in Fig. 3.9(a) and (b) for $\bar{n} = 0.1$ and $\bar{n} = 1$, respectively, we see that the most probable number of photons is zero, decreasing monotonically with n .

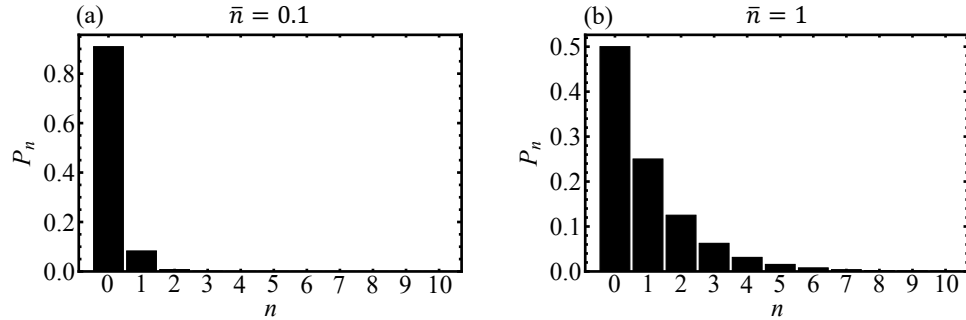


Figure 3.9: **Probability of detecting n photons in a thermal state.** Depicted for an average photon number of (a) $\bar{n} = 0.1$ and (b) $\bar{n} = 1$.

The fluctuations in average photon number can be calculated in a similar manner to the procedure we followed to determine $\bar{n}$,

$$\langle \hat{n}^2 \rangle = \text{Tr} [\hat{\rho}_{\text{Th}} \hat{n}^2] = \bar{n} + 2\bar{n}^2, \quad (3.189)$$

which then allows us to calculate the variation in photon number,

$$\langle (\Delta \hat{n})^2 \rangle = \bar{n} + \bar{n}^2. \quad (3.190)$$

We see that the fluctuations in n are larger than the average photon number, $\bar{n}$. The average photon energy within the black body cavity is $\hbar\omega\bar{n}$. Multiplying this average energy per photon by the density of modes per unit interval in ω in a unit of volume (and accounting for the two possible polarization directions), $\rho(\omega) = \omega^2/\pi^2c^3$, we arrive at an average energy density as a function of ω ,

$$\begin{aligned} \bar{U}(\omega) &= \hbar\omega\bar{n}\rho(\omega) \\ &= \frac{\hbar\omega^3}{\pi^2c^3} \frac{1}{\exp(\hbar\omega/k_B T) - 1}, \end{aligned} \quad (3.191)$$

which is equivalent to Planck's black body radiation law.

3.2 Generating single photons

Now that we have a better understanding of what a photon is and how photons may be distributed across the normal mode solutions to Maxwell's equations, we turn our attention to the practical question of how to generate photons in the lab. Ideally, single photons would be generated by an “on-demand” source which produces exactly one photon at any particular time (*i.e.*, never zero photons and never more than one photon), as desired by the user [324]. For such a source to provide a photon at any arbitrary time, it must also necessarily be capable of arbitrarily high repetition rates. In addition to these requirements, we would also like the emitted photons to be entirely indistinguishable from one another—a crucial requirement for quantum computing, among other applications.

Significant resources have been put towards developing such an ideal single photon source, and it will certainly be exciting to see how this field develops; however, single photon sources to-date do not achieve all of the qualities we have just listed. Current single photon sources can fall broadly under two categories, deterministic and probabilistic, each with their own advantages and limitations.

3.2.1 Attenuated coherent states of light

One of the first (and most straightforward) means of generating single photons was achieved by attenuating a light source—typically a laser beam. In the lab, this can be done by adding neutral density (ND) filters to the beam path, attenuating the laser intensity down to a level well below one mean photon per mode (or, one mean photon per pulse for a pulsed laser source). In this regime, most modes (or pulses) contain no photons, some contain one photon, and few contain more than one photon. We recall that the probability of detecting n photons in a coherent state is

$$P_n = e^{-|\alpha|^2} \frac{|\alpha|^{2n}}{n!}, \quad (3.192)$$

where the mean photon number per mode is defined as $\mu_\alpha = |\alpha|^2$. To get a feel for a desirable mean photon number, we consider the case where $\mu_\alpha = 0.1$, leading to $P_0 \approx 90\%$, $P_1 \approx 9\%$, and $P_2 \approx 1\%$.

This type of source is probabilistic, and so there is a trade-off between its brightness and the proportion of multiphoton events; however, with high repetition rate pulsed lasers, it is possible to produce weak coherent pulses¹⁹ at high rates, which is useful in many situations (*e.g.*, proof-of-concept experiments). It is important to remember that the distribution in Eq. 3.192 is governed by Poissonian statistics, and so attenuated coherent states cannot provide access to the sub-Poissonian number distribution that is often required in many quantum technologies, such as in practical quantum cryptography [325]. Despite this limitation, attenuated coherent states are still widely used as single-photon sources [260, 326, 327], as they are easy to implement and provide a nice mechanism to test whether an experiment can operate at the single-photon level.

¹⁹In the literature, it is common to see *attenuated coherent state* and *weak coherent pulses* used interchangeably—at least when the attenuated source is a pulsed laser.

3.2.2 Deterministic single-photon sources

Deterministic sources are so named because they most closely meet the “on-demand” requirement for an ideal single-photon source. There are many platforms which have been investigated for use as an “on-demand” source of single photons. Most of these are single-emitter quantum systems, shown idealistically in Fig. 3.10, which are selected or engineered to approximate a two-level system. Platforms in-

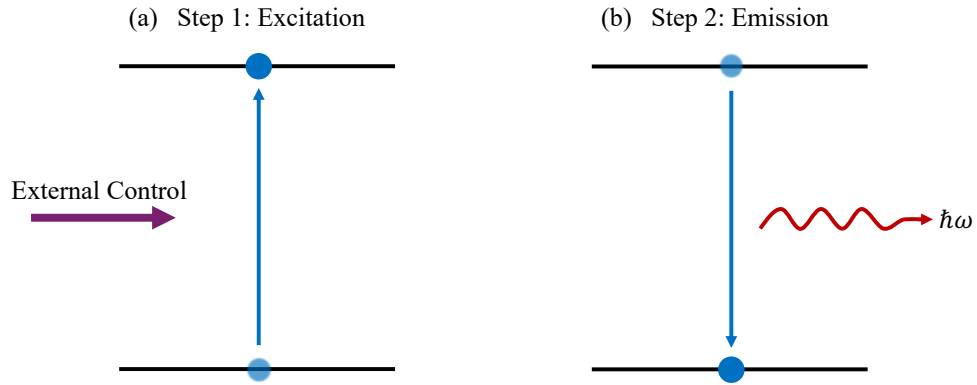


Figure 3.10: **Idealistic single-photon emitter.** A simplified energy-level diagram for a single-photon emitter system, based on a two-level system. The system is (a) excited by some means (the external control) which leads to (b) single-photon emission after some time (*i.e.*, the transition lifetime).

clude, among others, semiconductor quantum dots [328, 329, 330], mesoscopic quantum wells [331], single molecules [332, 333], single atoms [334, 335], single ions [336, 337, 338], and colour centers [339, 340]. Ensembles of atoms, which make use of more than just two energy levels, have also been used to realize deterministic sources of single photons [341, 342]. These sources use collective excitations in the ensemble as the excited state. While ensemble excitation itself is a probabilistic process, the excitation can be read out with high efficiency and so this mechanism is commonly considered a deterministic method of single-photon generation.

When a photon is desired from these deterministic systems, some external control (often a laser pulse) excites the system. Then, after some time, the system relaxes back to its ground state, emitting a photon in the process. In theory, a photon is produced every time the control excites the system and there is no possibility of emitting more than one photon per excitation; however, this is not always the case in practice.

Noise and imperfections in the system can contribute to multiphoton events, and in some cases the system may relax through a non-radiative path thus limiting the efficiency of the single-photon generation process. Furthermore, the emitted photon may not be collected in the proper mode, which reduces the overall system efficiency; however, placement of the source within a cavity [343] or waveguide [344] can significantly improve collection efficiency. Some platforms, such as quantum dots and colour centers suffer from distinguishability issues, while other approaches, such as trapped ions, have complex experimental requirements [324].

It should also be noted that single-emitter sources are necessarily defined by their excited state lifetime. Long lifetimes limit the repetition rate of the source and also define the uncertainty in the arrival time of the emitted photon. In the context of the work in this thesis, deterministic sources with lifetimes longer than several hundred femtoseconds are not viable candidates for use in our ultrafast photonic quantum computing. For a deterministic single-photon source to be compatible with our ultrafast time-bin encoding platform, its lifetime must also necessarily be ultrafast.

While some experimental challenges have yet to be overcome, deterministic sources remain an attractive option for many photonic quantum technologies. When these challenges are fully addressed, deterministic sources have the greatest potential to approach ideal single photon sources. Unlike the probabilistic sources we will discuss next, ideal deterministic sources would not have a trade-off between brightness and probability of multiphoton events. The popularity of deterministic sources will undoubtedly grow as they become increasingly compact and user-friendly, but probabilistic sources still remain (at least for now) the go-to method for generating single photons in most quantum optics experiments.

3.2.3 Probabilistic single-photon sources

As previously alluded to, probabilistic sources (also called *photon pair sources* or *heralded single-photon sources*) are the current workhorse in quantum optics experiments. These sources are based on parametric nonlinear optical processes in which pairs of photons are generated [345, 346]. As shown in Fig. 3.11, one (or more) pump beams are input to a nonlinear medium and, with some probability, a pair of correlated photons will be created while one (or two) pump photon(s) will be annihilated.

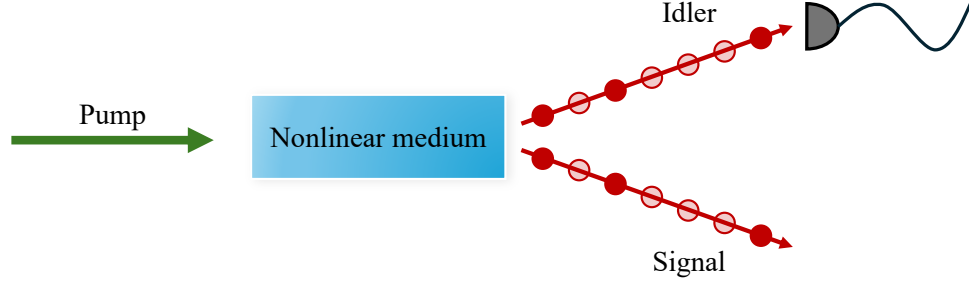


Figure 3.11: **Probabilistic photon pair source.** Pairs of correlated photons are generated in a nonlinear medium which is pumped by a high-intensity laser. The solid red circles indicate photon pair events, while the empty circles represent times at which a pair was not generated. Photon pairs are always generated simultaneously, as depicted. The idler photon is sent to a detector to herald the presence of the signal photon.

While it is not possible to predict whether a given pump photon (or pair of photons) will generate a photon pair, the nonlinear process itself will, crucially, always generate a pair of photons—never just a single photon. Since the photons are necessarily generated in pairs, detecting one of the two photons allows us to infer (or *herald*) the existence of the other photon. As discussed in Sections 2.2.2 and 2.2.3, two common nonlinear processes used for photon pair generation are spontaneous parametric down-conversion (SPDC) [347] and spontaneous four-wave mixing (SFWM) [348].

The Hamiltonian which describes the nondegenerate SPDC process is given by

$$\hat{\mathcal{H}} = \hbar\omega_s \hat{a}^\dagger \hat{a} + \hbar\omega_i \hat{b}^\dagger \hat{b} + \hbar\omega_p \hat{c}^\dagger \hat{c} + i\hbar\chi^{(2)} \left(\hat{a}\hat{b}\hat{c}^\dagger - \hat{a}^\dagger \hat{b}^\dagger \hat{c} \right), \quad (3.193)$$

where a , b , and c are used to denote the signal, idler, and pump modes, respectively. The second-order nonlinear susceptibility, which we dealt with in detail in Section 2.2.2, is given by $\chi^{(2)}$. Under the parametric approximation, where we assume that the pump is a strong classical field (coherent state) which remains undepleted, we can approximate the c -mode operators as $\hat{c} = \gamma e^{-i\omega_p t}$ and $\hat{c}^\dagger = \gamma^* e^{i\omega_p t}$, respectively. Under this approximation, the Hamiltonian becomes

$$\hat{\mathcal{H}}^{(\text{PA})} = \hbar\omega_s \hat{a}^\dagger \hat{a} + \hbar\omega_i \hat{b}^\dagger \hat{b} + i\hbar\chi^{(2)} \left(\gamma^* e^{i\omega_p t} \hat{a}\hat{b} - \gamma e^{-i\omega_p t} \hat{a}^\dagger \hat{b}^\dagger \right). \quad (3.194)$$

In the interaction picture, the time-dependent interaction Hamiltonian is written as

$$\hat{\mathcal{H}}_1(t) = i\hbar\chi^{(2)} \left(\gamma^* e^{i(\omega_p - \omega_a - \omega_b)t} \hat{a}\hat{b} - \gamma e^{-i(\omega_p - \omega_a - \omega_b)t} \hat{a}^\dagger \hat{b}^\dagger \right). \quad (3.195)$$

In SPDC, energy conservation requires $\omega_p = \omega_a + \omega_b$, so we obtain

$$\hat{\mathcal{H}} = i\hbar\chi^{(2)} \left(\gamma^* \hat{a}\hat{b} - \gamma \hat{a}^\dagger \hat{b}^\dagger \right), \quad (3.196)$$

which is time-independent. The corresponding time-evolution operator is

$$\hat{U}_1(t) = e^{-i\hat{\mathcal{H}}_1 t/\hbar} = \exp \left[\chi^{(2)} \left(\gamma^* \hat{a}\hat{b} - \gamma \hat{a}^\dagger \hat{b}^\dagger \right) t \right], \quad (3.197)$$

which takes the form of the two-mode squeezing operator, $\hat{S}_2(\zeta)$, with $\zeta = \chi^{(2)}\gamma t$. The nondegenerate SPDC process thus produces entangled TMSV states. A similar result can be shown for the nondegenerate SFWM process, defined by the Hamiltonian

$$\hat{\mathcal{H}} = \hbar\omega_a \hat{a}^\dagger \hat{a} + \hbar\omega_b \hat{b}^\dagger \hat{b} + \hbar\omega_p \hat{c}^\dagger \hat{c} + i\hbar\chi^{(3)} \left(\hat{a}\hat{b}(\hat{c}^\dagger)^2 - \hat{a}^\dagger \hat{b}^\dagger (\hat{c})^2 \right), \quad (3.198)$$

where $\chi^{(3)}$ is the third-order nonlinear susceptibility detailed in Section 2.2.3, and the squeezing parameter takes the form $\zeta = \chi^{(3)}\gamma^2 t$. Clearly, nondegenerate SPDC and SFWM generate TMSV states, thus making these processes a source for nonclassical light! When we first introduced these processes in Chapter 2, we were unable to demonstrate photon pair production using a solely classical description. We now see the importance of the quantum mechanical description of these spontaneous processes. In fact, these processes can also be thought of as the amplification of vacuum fluctuations, discussed earlier in this chapter, mediated by optical nonlinearity.

These processes are necessarily probabilistic, where not every pump photon (or pair of pump photons) will be converted into a correlated pair, and in some cases multiple pump photons (or pairs of pump photons) can generate two or more correlated photon pairs at the same time. Since most single-photon detectors are not photon-number-resolving, it is usually not possible to identify, through heralded detection, when these multiphoton pair events take place. These sources must therefore be operated with low enough pump power such that there is a high probability of generating no photon pairs, some probability of producing a pair of single photons,

and a low probability that pairs of two or more photons will be produced. Evidently, this is far from the ideal “on-demand” source, and poses limitations on the scalability of photonic quantum systems which make use of these sources. Keeping the probability of generating a photon pair low by necessity, it becomes increasingly difficult to operate quantum systems in which multiple single photons are required. Experiments must be operated with what can be prohibitively long integration times [349].

Moreover, probabilistic single-photon sources can be negatively affected by system loss and noise. If, due to system loss, a herald photon is not detected, its partner photon will not be recognized as a single-photon event (and vice versa). On the other hand, noise in the measurement (arising from, *e.g.*, detector dark counts or parasitic nonlinear processes) can lead to a heralded measurement of a noise photon. Despite these challenges, probabilistic single-photon sources are generally straightforward to implement experimentally and can be optimized to generate highly indistinguishable single photons. Furthermore, in the context of the work presented in this thesis, these sources can be pumped by ultrashort pulses of light so that the uncertainty in arrival time of generated single photons occurs on ultrafast timescales.

3.3 Detecting single photons*

Recalling that the energy of a single photon is $\hbar\omega$, which for visible red light (*e.g.*, at 633 nm) is about 3.14×10^{-19} J = 1.96 eV, an important practical question we must address is how single photons can be measured in the lab. Evidently, we require a highly sensitive detector which is capable of detecting very small optical intensities. The ideal single photon detector will meet all of the criteria outlined in Table 3.1. Of course, it is difficult to meet each of these criteria in one detector; it is oftentimes possible to forgo certain criteria in order to meet others, so the choice of detector will depend on the experimental requirements. The remainder of this section will discuss three important types of detectors in the context of quantum optics experiments.

Ideal single-photon detector criteria

High quantum efficiency (QE): QE is the percentage of photons a detector can convert into electrons, and depends largely on the spectral responsivity of the detection material. This value will ideally be as close to 100% as possible, such that each incident photon is converted into an electron.

Low noise: Noise defines how much the signal generated by a detector will fluctuate. For single-photon detection, it is crucial that this is minimized. The two main noise mechanisms in optical receivers are *shot noise* and *thermal noise*. Shot noise is due to the fact that an electric current consists of a stream of electrons generated at random times, governed by Poissonian statistics. Thermal noise (also called *Johnson noise* or *Nyquist noise*) arises from random thermal motion of electrons in a detector's load resistor, thus leading to fluctuations in current.

Low jitter time: Timing jitter refers to the timing distribution of a detector's response to single photons. In quantum optics, it is important to know, with high resolution, the arrival time of a photon at the detector; as such, this value will ideally be low.

Short dead time: Detector dead time is the minimum amount of time that must elapse after a detection event, before the detector is able to detect the next photon. For high-rate experiments, this number must be low.

High dynamic range: This refers to the range of intensities a detector can reliably detect. In some cases, it is important to have a high dynamic range.

No after pulsing: After pulsing is a phenomenon that occurs due to feedback, or from the trapping and releasing of charge carriers, within the detector. This can lead to reduction in efficiency or overestimation of count rates, and so it is important to reduce the possibility of this effect.

Long operation times: Some detection systems (especially those which require cryogenics) must be cycled, thus limiting the timescales over which they can be used for a continuous period. Some experiments require long operation times, making this an important consideration.

Photon number resolution: This refers to a detector's ability to count the number of photons that arrive in a single detection event.

Table 3.1: **Criteria of the ideal single-photon detector.** Metrics are presented and used to describe the ideal single-photon detector.

3.3.1 Avalanche photodiodes*

An avalanche photodiode (APD) is a type of photodiode with an internal gain mechanism that is achieved using a high reverse bias voltage. In general, a photodiode is a semiconductor device with a p-n junction that is placed under a reverse bias. As depicted in Fig. 3.12, a p-n junction combines p-type (positively doped, containing freely-moving holes) and n-type (negatively doped, containing freely-moving electrons) semiconducting materials. Connecting these two materials causes diffusion

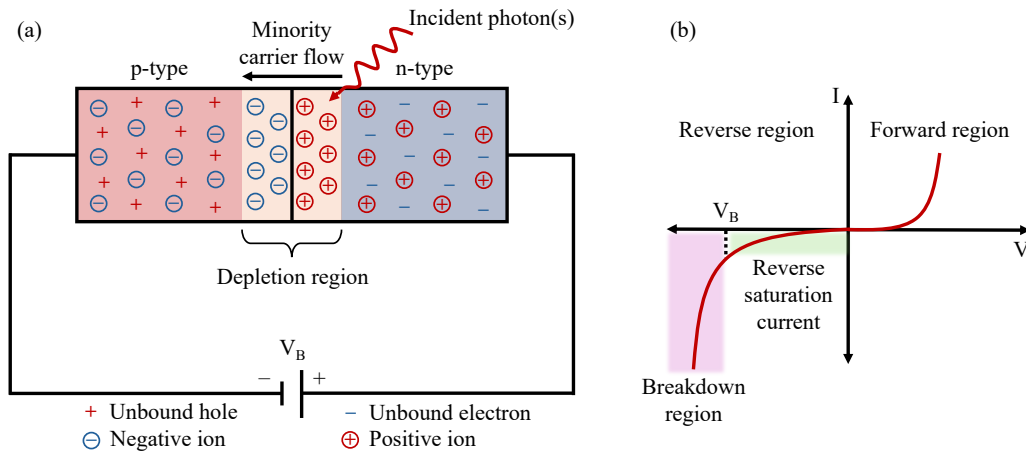


Figure 3.12: **Schematic of a photodiode.** The photodiode consists of (a) a p-n junction operated under a reverse bias voltage. Here, bound charges are enclosed in circles while free charges are not. Operation of a p-n junction under a reverse bias will (b) generate a very small reverse saturation current, shown in green. Photoexcitation of charge carriers can increase this current. In the case of APDs, the reverse bias voltage is very high, such that an incident photon can induce currents high enough that the device enters the breakdown region ($V > V_B$), shown in pink.

of free electrons from the n-type to the p-type region and diffusion of free holes from the p-type to the n-type region. This creates a *depletion region* near the boundary between the p-type and n-type regions which is free of mobile charge. Application of a reverse bias voltage (*i.e.*, the p-type and n-type regions are connected to the negative and positive terminals of a voltage supply, respectively) will increase the voltage barrier of the depletion region, thus causing a high resistance to the flow of charge

carriers, and very little current will flow—until the diode breaks down. Impact ionization, where incident photons provide the energy to separate charge carriers within the depletion region, can cause the diode the break down and generate a current.

When the electric field across the depletion zone increases beyond a critical level, the depletion zone of the p-n junction breaks down and current is allowed to flow, through either the *Zener breakdown* or the *avalanche breakdown* processes²⁰. In the avalanche breakdown effect, the electric field is strong enough such that a mobile electron or hole will be accelerated to speeds that are high enough to free other neighbouring bound electrons or holes via collision. This process will continue to occur, creating the avalanche-like effect which gives the APD its name²¹. The APDs used for single-photon detection are operated with a reverse bias voltage that is just above the nominal breakdown voltage, allowing for the very large amplification needed to create a macroscopic current from a single photon²². This gain makes the APD a very sensitive photodiode. APDs are convenient detectors, as they are low cost and can be operated at room temperature for an indefinite amount of time; however, their detection efficiency, dark count rates, and timing jitter may not always be sufficient, and they are not capable of photon-number resolution. Despite these limitations, APDs are still a common tool in quantum optics labs and have been the detection method of choice in numerous experiments due to their ease of use.

3.3.2 Superconducting nanowire single-photon detectors*

The superconducting nanowire single-photon detector (SNSPD) is a detector, capable of single-photon-level detection, based on a current-biased superconducting nanowire [350, 351]. The superconducting nanowire is typically thin (~ 5 nm) and narrow (~ 100 nm) with a total length that is typically hundreds of micrometers, patterned in a geometry as depicted in Fig. 3.13(a) to optimize detection efficiency.

The nanowire must be cooled well below the superconducting critical temperature and must be biased with a DC current that is just below its superconducting critical current. At the extremely low temperatures required for superconductivity, electrons can form *Cooper pairs*. Cooper pairing is a quantum mechanical effect in which an

²⁰Each of these breakdown processes are non-destructive and reversible, provided the current does not reach levels which can cause thermal damage to the semiconducting material. Note that the avalanche breakdown has a hysteresis effect, while the Zener breakdown does not.

²¹Functionally, APDs can be regarded as the semiconductor analog of photomultiplier tubes.

²²This regime is not optimized for generating electricity from light, but rather for detection of low-light signals.

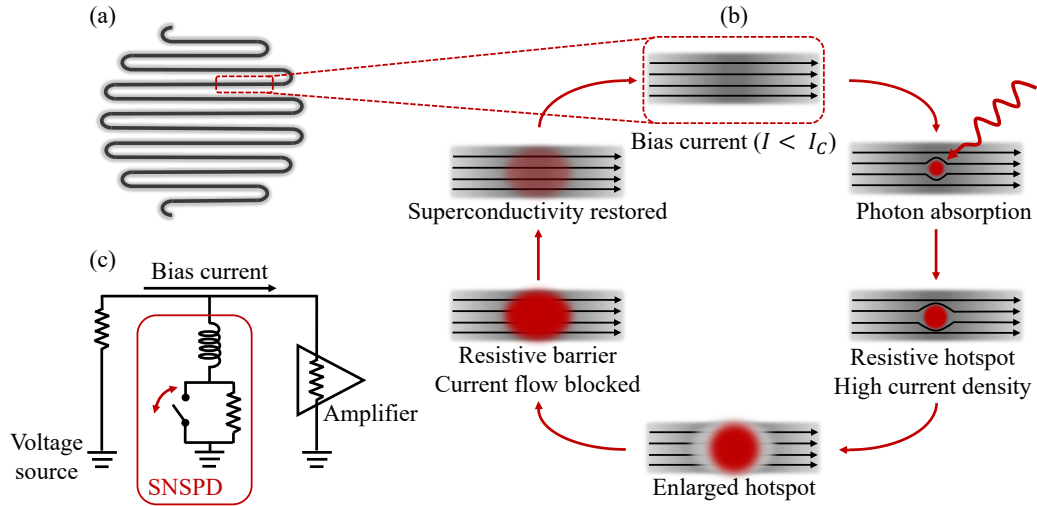


Figure 3.13: **Schematic of a superconducting nanowire single-photon detector (SNSPD).** The SNSPD nanowire is patterned in the geometry depicted in (a). When a photon is incident on the superconducting nanowire, it generates a hotspot which breaks superconductivity. This in turn leads to the cycle depicted in (b), which is used to shunt current to the amplifier as seen in (c), ultimately allowing for single-photon readout. Figure adapted from [352] and [353].

arbitrarily small attraction between electrons in a metal can cause a paired state of electrons to have a lower energy than the Fermi energy, thus implying the pair is bound. The free electrons in a metal should usually repel one another; however, they also attract the positive ions which make up the rigid lattice of the metal. This attraction causes the ion lattice to be distorted, moving slightly towards the free electrons and thus increasing the positive charge density of the lattice in the vicinity of the free electrons. This positive charge density can then attract other electrons and, at long distances²³, can overcome the repulsion between two electrons and cause them to “pair up”. A more rigorous quantum mechanical explanation²⁴ attributes Cooper pairing to electron-phonon interactions²⁵.

²³The electrons in a Cooper pair are not necessarily close together; in fact, they can be many hundreds of nanometers apart.

²⁴The 1972 Nobel Prize was awarded to John Bardeen, Leon Cooper, and John Schrieffer for the theoretical description of Cooper pair formation.

²⁵The phonon is the collective motion of the positively-charged lattice.

The energy of the Cooper pairing interaction is understandably quite weak (on the order of 10^{-3} eV), such that a small amount of thermal energy can easily break the pairs. Thus, this effect is really only significant at cryogenic temperatures (~ 2.5 K). As seen in Fig. 3.13(b), when a photon is incident on the superconducting nanowire, it will heat the device up a small amount, thus breaking Cooper pairs and reducing the local critical current below that of the bias current. This forms a localized non-superconducting region within the nanowire that has a large electrical resistance, and so most of the bias current is temporarily shunted to the readout amplifier, as seen in Fig. 3.13(c). With most of the bias current flowing through the amplifier, the non-superconducting region can cool down and return to its superconducting state. The time it takes the bias current to return to the nanowire is usually set by the inductive time constant of the nanowire [354].

Commercial SNSPDs are available with over 90% efficiency at near infrared and telecom wavelengths, maximum count rates of over 80 MHz, dark count rates of less than 1-10 counts per second, and timing jitter of less than 15 ps²⁶. In particular, SNSPDs have achieved over 98% efficiency [10] and the most precise timing resolution to-date, boasting a remarkably low jitter time of 3 ps at visible wavelengths [11]. A low timing jitter is important for quantum computing and communications applications which make use of time bins, and is thus of particular interest in the context of the work presented here. While SNSPDs do not match the intrinsic energy or photon-number resolution of the transition edge sensors discussed next, the SNSPD is significantly faster and can operate at higher temperatures than transition edge sensors. Furthermore, SNSPD arrays [9] can, to some degree, achieve photon-number resolution through time-binning [355, 356] or other advanced readout schemes [357].

3.3.3 Transition-edge sensors*

Similarly to the SNSPD, a transition-edge sensor (TES) is also a cryogenic sensor which exploits the superconducting phase transition. The TES must be operated at temperatures on the order of a milli-Kelvin (mK); although this temperature is only a few Kelvin away from the operational temperature of the SNSPD, it leads to significantly different capabilities (and is also a much more challenging cryogenic regime to

²⁶These specifications are taken from the *Single Quantum Eos* SNSPD product line.

reach). The TES is a very sensitive calorimeter made from superconducting film, as shown in Fig. 3.14(a), which is operated near its superconducting transition temperature. During the transition between superconductor and normal metallic conductor, a very small change in temperature causes a large change in resistance within the TES device, as shown in Fig. 3.14(b). The width of this transition is typically on the order of a milli-Kelvin, with a resolution that increases as temperature decreases, so it is crucial that TES devices are maintained at milli-Kelvin temperatures.

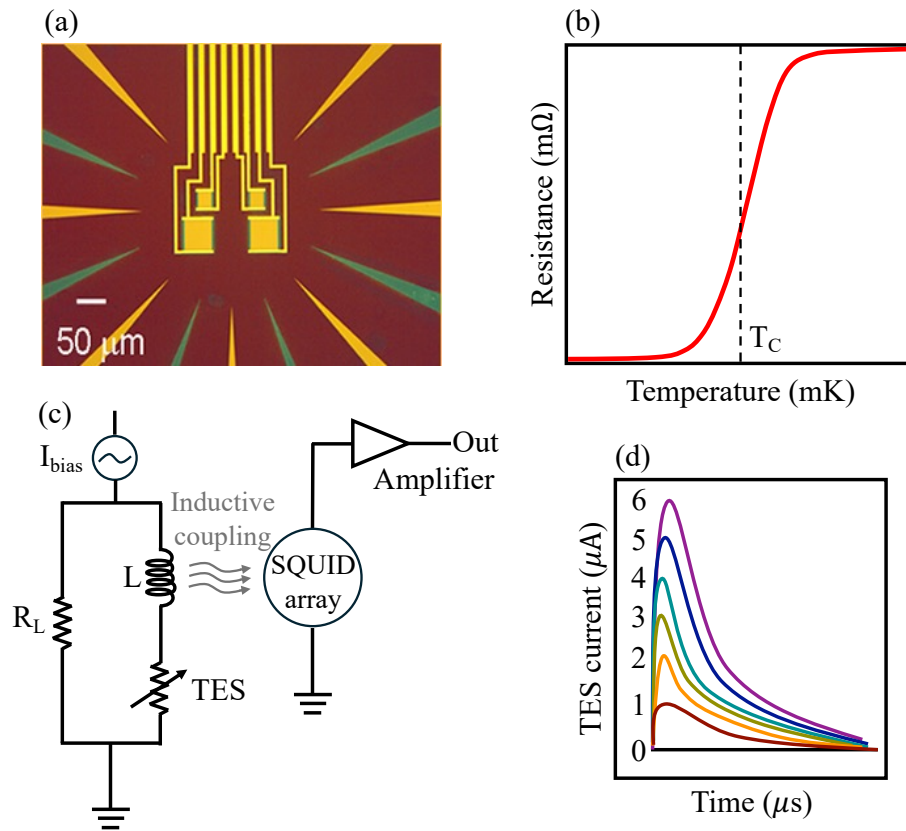


Figure 3.14: **Schematic of a transition-edge sensor (TES).** An example TES geometry [358] is shown in (a). The TES is a sensitive calorimeter, where (b) small temperature changes induce changes in resistance. These changes in resistance are measured by (c) coupling the TES circuit to a superconducting quantum interference device (SQUID) for readout. The measured current from the TES will (d) reach different, discrete, values depending on the number of incident photons.

The TES is operated in its “self-biased” region, where the power dissipated in the device remains constant by an applied bias voltage. When a photon is incident on the TES, its energy will be absorbed and increase the temperature of the device, thus increasing the resistance of the voltage-biased sensor within its transition region. This increase in resistance leads to a drop in current, which is proportional to the energy absorbed by the TES. Once this occurs, the power dissipated in the device will drop and the device can then return back to equilibrium. As seen in Fig. 3.14(c), the most common readout system uses a superconducting quantum interference device (SQUID) [359], which is a highly sensitive magnetometer based on superconducting loops with Josephson junctions. Here, the TES is operated in series with an input coil that is inductively coupled to a SQUID series-array, such that a small change in TES current can be read out as a change in the input flux to the SQUID. The SQUID output is then amplified and read out by room-temperature electronics.

In our quantized picture of radiation, finite amounts of energy will be absorbed for a finite number of absorbed photons. With high enough sensitivity, the TES will be capable of determining the total number of photons absorbed to produce a given increase in temperature (and thus also a decrease in current), depicted in Fig. 3.14(d). For maximal temperature sensitivity, the TES should have a low heat capacity and a narrow transition. The thermal conductance of the TES material is also an important property to consider. Taking all of these properties into consideration, it is clear that the choice of TES material and transition temperature are both critical to the design of the device. Of course, the transition temperature must also accommodate available cryogenic systems. To-date, thin-film tungsten has been a popular material system for TES devices operating in the near infrared and optical regimes [360, 361]. Thin-film tungsten has two superconducting phases, one with a critical temperature of ~ 15 mK and another with a critical temperature of $\sim 1 - 4$ K, which can be combined to allow for fine-tuning of the final overall critical temperature of the device [362].

TESs are attractive for a multitude of reasons, including high detection efficiency across a very broad range of wavelengths (millimeter to gamma rays) [363, 364] and theoretically negligible dark count rates²⁷ (less than one event in 1000 seconds) [365].

²⁷In practice, background photons in a quantum optics experiment can certainly be registered by the photon number counting algorithm; even black body radiation may be detected by a TES optimized for the visible regime.

Despite these impressive attributes, TES devices must be operated in very extreme cryogenic conditions, their output signal requires advanced analysis to extract photon number information, and they suffer from high timing jitter (*e.g.*, ~ 100 ns) [364] and long dead times. Nevertheless, TES detectors are desirable for their striking ability to distinguish the number of photons within a detection event—a crucial capability in many quantum optics experiments. TES-based photon-number-resolving detectors (PNRDs) provide a linear response in photon number with a dynamic range of one photon up to about 20-30 photons [366, 367], with the possibility of extending to higher photon number resolution via multiplexing [368] and machine learning techniques [369].

3.3.4 Time-to-digital conversion*

Each of the aforementioned detectors output a stream of electrical pulses corresponding to single photon detection events. In many cases, we are interested in knowing the time at which each detection event occurred (*e.g.*, when using a photon pair source we must herald the detection of one photon with its partner, thus requiring knowledge of the arrival time of both photons in a pair). Such information requires a means of time-to-digital conversion, whereby the analog electrical pulses from the detector are converted into a digital representation of the time at which they occurred. A time-to-digital converter provides us with just this capability, and is a critical component in most quantum optics experiments²⁸.

3.4 Characterizing single-photon sources

Since the perfect “on-demand” single-photon source does not yet exist, we still rely on characterization methods to ensure that our sources of single photons are in fact generating the light we expect. In general, photon statistics can be used to describe and classify different states of light, such as the coherent, thermal, and squeezed light we saw in previous sections²⁹. The question now becomes how to measure the photon

²⁸Many commercial options exist, for example the *TimeTagger* series of time-to-digital converters from *Swabian Instruments*.

²⁹We already introduced the photon statistics for these sources of light when we derived the probability of having n photons present, $P(n)$, in each state of light.

statistics of a given light source. Though it is possible to use PNRDs to simply measure the photon number distribution of a given state of light [370], such detectors are limited in their dynamic range (and not every lab has access to PNRDs). We must therefore resort to other photon characterization metrics from which we can infer the photon statistics, and thus gain an understanding of the type of source we are working with. In addition to photon statistics, there are several other characterization metrics which are often important to consider, such as the mean photon number of a source, or its heralding efficiency (when considering a photon pair source).

3.4.1 Mean photon number

A common characterization metric for low-light sources is the mean photon number per mode, μ . This metric can be used to describe attenuated coherent states, deterministic single-photon sources, and probabilistic single-photon sources. The ideal single-photon source would have $\mu = 1$, exactly, for the mode (or modes) of interest; however, in practical sources—such as the attenuated coherent states or photon pair sources used in this thesis—there is a trade-off between the total number of generated photons and the number of modes containing more than one photon. In general, a given application will have an optimal value of μ , which maximizes count rates while minimizing the probability of a multiphoton event. With knowledge of the system efficiency (including detection efficiency), η_{sys} , and the laser rep rate, the mean photon number per pulse of a given source can be calculated for an attenuated coherent pulse as

$$\mu_{\alpha} = \frac{(\text{counts per sec at detector})}{\eta_{\text{sys}}(\text{rep rate})}. \quad (3.199)$$

The mean photon number per pulse of a photon pair source can, likewise, be calculated as

$$\mu_{s(i)} = \frac{[\text{signal (idler) photon singles per sec at detector}]}{\eta_{\text{sys}, s(i)}(\text{rep rate})}, \quad (3.200)$$

though it should be noted that it is also possible to calculate the mean photon number of a pair source from its coincidence rate and heralding efficiencies for the signal and

idler photons, η_s and η_i , respectively, as

$$\mu_{s(i)} = \frac{(\text{coincidence rate at detectors})}{\eta_s \eta_i (\text{rep rate})}. \quad (3.201)$$

3.4.2 Heralding efficiency

When working with a photon pair source (also called a heralded single-photon source), we generally use a heralded detection event (also referred to as a *coincidence* event) of one photon in a pair (the *idler*) to infer the existence of the other photon in the pair (the *signal*). The heralding efficiency is therefore the conditional probability of detecting a signal photon, given that the idler photon has been collected and detected. The heralding efficiency for the signal (or idler) photon can be expressed as

$$\eta_{s(i)} = P_{s|i(i|s)} = \frac{P_{s,i}}{P_{i(s)}} = \frac{N_C}{N_{i(s)}}, \quad (3.202)$$

where $P_{s|i(i|s)}$ is the probability of detecting a signal (or idler) photon conditioned on detecting an idler (or signal) photon, which can be expressed in terms of $P_{s,i}$, the probability of detecting a coincidence, and $P_{i(s)}$, the probability of detecting an idler (or signal) photon. This can also be expressed in terms of N_C , the number of two-fold coincidence events detected, and $N_{i(s)}$, the total number of idler (or signal) photons detected, within a set time interval. We note that the heralding efficiency may be defined at different points within an optical setup; for example, the heralding efficiency may be defined immediately after the photon pair source, or it could be defined after the signal photon propagates through a complicated optical system. It is also common to see reported values of heralding efficiency in which the detector efficiency (or even the full system efficiency) has been factored out so as to exclude losses and calculate the heralding efficiency of only the source itself.

3.4.3 Quantum coherence and quantum correlation functions*

In quantum optics, correlation functions are often used to characterize the statistical and coherence properties of light. The quantum theory of coherence is directly

inspired by the classical theory of coherence, which is best motivated with a discussion of Young's double-slit experiment [371], depicted in Fig. 3.15.

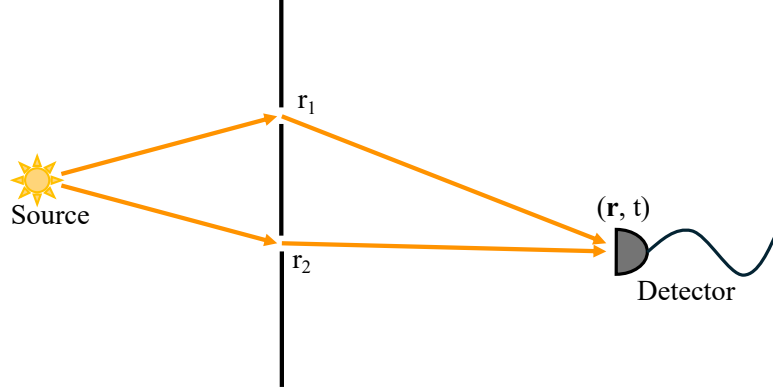


Figure 3.15: **Young's double-slit experiment.** Radiation from a source is directed at two slits. The light emerging from these two slits can interfere at the detector.

Classical first-order coherence

The time-averaged intensity measured at the detector at position r and time t is given by

$$\langle I(\mathbf{r}, t) \rangle = I_1 + I_2 + 2\sqrt{I_1 I_2} |\gamma^{(1)}(x_1, x_2)|^2 \cos[\phi(x_1, x_2)], \quad (3.203)$$

where I_1 and I_2 are the intensities of the fields from slits 1 and 2, respectively, and $\gamma^{(1)}(x_1, x_2)$ is the *first-order normalized correlation function*, given by

$$\gamma^{(1)}(x_1, x_2) = \frac{\langle E^*(x_1)E(x_2) \rangle}{\sqrt{\langle |E(x_1)|^2 \rangle \langle |E(x_2)|^2 \rangle}}, \quad (3.204)$$

where $\phi(x_1, x_2)$ is the phase difference between the two fields that arises from a difference in path length³⁰. Note that we use the notation $x_i = (\mathbf{r}_i, t_i)$. It is convenient

³⁰This path length difference is assumed to be less than the coherence length of the light source, $l_{\text{coh}} = c/\Delta\omega$, where $\Delta\omega$ is the bandwidth of the source.

to introduce Rayleigh's definition of fringe visibility,

$$\mathcal{V} = \frac{I_{\max} - I_{\min}}{I_{\max} + I_{\min}}, \quad (3.205)$$

where

$$I_{\min}^{\max} = I_1 + I_2 \pm 2\sqrt{I_1 I_2} |\gamma^{(1)}(x_1, x_2)|, \quad (3.206)$$

and so

$$\mathcal{V} = \frac{2\sqrt{I_1 I_2} |\gamma^{(1)}(x_1, x_2)|}{I_1 + I_2}. \quad (3.207)$$

There are clearly three notable coherence regimes,

$$\begin{aligned} |\gamma^{(1)}(x_1, x_2)| &= 1 && \text{complete coherence,} \\ 0 < |\gamma^{(1)}(x_1, x_2)| &< 1 && \text{partial coherence,} \\ |\gamma^{(1)}(x_1, x_2)| &= 0 && \text{complete incoherence.} \end{aligned}$$

Young's double slit experiment demonstrates the dependence of *spatial* interference on coherence—specifically, on the first-order correlation function, $\gamma^{(1)}(x_1, x_2)$. It should be noted that coherence can also depend—in the same manner—on *temporal* interference, as seen, for example, in a Mach-Zehnder interferometer³¹.

Quantum first-order coherence

A quantum theory of coherence can be constructed in a manner which closely parallels the classical theory. In the case of quantum coherence, we recognize that detectors measure the intensity of a light source by absorbing its photons. We therefore require a means of quantifying the response of the absorbing detection system. We consider an ideal detector consisting of a single atom, with dimensions smaller than the wavelength of the incident light. The absorption of light will result in the ionization of the atom, where the generation of a photo-electron constitutes the detection

³¹In this case, we now consider timescales that are shorter than the coherence time of the source, $\tau_{\text{coh}} = l_{\text{coh}}/c = 1/\Delta\omega$.

of a photon. By counting these photo-electrons, the statistical properties of the light source can be determined. This single-atom detector couples to the light field through the dipole interaction, described by the Heisenberg-picture Hamiltonian,

$$\hat{\mathcal{H}} = -\hat{\mathbf{d}} \cdot \hat{\mathbf{E}}(\mathbf{r}, t), \quad (3.208)$$

where $\hat{\mathbf{d}}$ is the dipole operator and the electric field operator is defined as in Eq. 3.59. Since the wavelength of light is assumed to be larger than the dimensions of the atom, we can make the dipole approximation, $|\mathbf{k} \cdot \mathbf{r}| \ll 1$. The electric field can be further simplified by considering only the positive-frequency part of the field, since it contains the annihilation operator,

$$\hat{\mathbf{E}}(\mathbf{r}, t) = \hat{\mathbf{E}}^{(+)}(\mathbf{r}, t) = i \sum_{\mathbf{k}, \sigma} \left(\frac{\hbar \omega}{2 \epsilon_0 V} \right)^{1/2} \hat{\mathbf{e}}_{\mathbf{k}, \sigma} \hat{a}_{\mathbf{k}, \sigma}(t). \quad (3.209)$$

Upon absorption of the incident radiation, the atom makes a transition from the ground state, $|g\rangle$, to the ionized state, $|e\rangle$, while the electric field makes a transition from the initial state, $|i\rangle$, to a final state, $|f\rangle$. The matrix element for a transition is therefore

$$\langle e| \otimes \langle f| \hat{\mathcal{H}} |i\rangle \otimes |g\rangle = -\langle e| \hat{\mathbf{d}} |g\rangle \langle f| \hat{\mathbf{E}}^{(+)}(\mathbf{r}, t) |i\rangle, \quad (3.210)$$

and the probability that the field undergoes a transition from the initial to final state is given by

$$w(\mathbf{r}, t) = \left| \langle f| \hat{\mathbf{E}}^{(+)}(\mathbf{r}, t) |i\rangle \right|^2. \quad (3.211)$$

Since the final state of the field is never measured (*i.e.*, we are only interested in the final state of the detector), we must sum over all possible final states of the field,

$$\begin{aligned} \sum_f \left| \langle f| \hat{\mathbf{E}}^{(+)}(\mathbf{r}, t) |i\rangle \right|^2 &= \sum_f \langle i| \hat{\mathbf{E}}^{(-)}(\mathbf{r}, t) |f\rangle \langle f| \hat{\mathbf{E}}^{(+)}(\mathbf{r}, t) |i\rangle \\ &= \langle i| \hat{\mathbf{E}}^{(-)}(\mathbf{r}, t) \cdot \hat{\mathbf{E}}^{(+)}(\mathbf{r}, t) |i\rangle, \end{aligned} \quad (3.212)$$

where we have made use of the fact that this set of final states can be considered complete. In practice, we generally do not know precisely the initial state, $|i\rangle$, nor is it necessarily a pure state. For this reason, we introduce the density operator formalism, where

$$\hat{\rho}_F = \sum_i p_i |i\rangle \langle i|, \quad (3.213)$$

such that the expectation value of $\hat{\mathbf{E}}^{(-)}(\mathbf{r}, t) \cdot \hat{\mathbf{E}}^{(+)}(\mathbf{r}, t)$ is replaced by the ensemble average and we write

$$\begin{aligned} \langle i | \hat{\mathbf{E}}^{(-)}(\mathbf{r}, t) \cdot \hat{\mathbf{E}}^{(+)}(\mathbf{r}, t) | i \rangle &= \text{Tr} \left[\hat{\rho}_F \hat{\mathbf{E}}^{(-)}(\mathbf{r}, t) \cdot \hat{\mathbf{E}}^{(+)}(\mathbf{r}, t) \right] \\ &= \sum_i p_i \langle i | \hat{\mathbf{E}}^{(-)}(\mathbf{r}, t) \cdot \hat{\mathbf{E}}^{(+)}(\mathbf{r}, t) | i \rangle. \end{aligned} \quad (3.214)$$

Using our earlier notation, $x = (\mathbf{r}, t)$, we define

$$G^{(1)}(x, x) = \text{Tr} \left[\hat{\rho}_F \hat{\mathbf{E}}^{(-)}(x) \cdot \hat{\mathbf{E}}^{(+)}(x) \right], \quad (3.215)$$

which is simply the intensity of light at the space-time point $x = (\mathbf{r}, t)$, and so $G^{(1)}(x, x)$ is the quantum mechanical analog of the time-averaged intensity that we defined for classical coherence. We can describe Young's double-slit interference using our quantum coherence picture, where the field at the detector is now defined as

$$\hat{\mathbf{E}}^{(+)}(\mathbf{r}, t) = \hat{\mathbf{E}}^{(+)}(\mathbf{r}_1, t_1) + \hat{\mathbf{E}}^{(+)}(\mathbf{r}_2, t_2). \quad (3.216)$$

We can write the time-averaged intensity as

$$\begin{aligned} \langle I(\mathbf{r}, t) \rangle &= \text{Tr} \left[\hat{\rho}_F \hat{\mathbf{E}}^{(-)}(\mathbf{r}, t) \hat{\mathbf{E}}^{(+)}(\mathbf{r}, t) \right] \\ &= G^{(1)}(x_1, x_1) + G^{(1)}(x_2, x_2) + 2\text{Re} \left[G^{(1)}(x_1, x_2) \right], \end{aligned} \quad (3.217)$$

and, defining the normalized first-order quantum correlation function

$$g^{(1)}(x_1, x_2) = \frac{G^{(1)}(x_1, x_2)}{\sqrt{G^{(1)}(x_1, x_1)G^{(1)}(x_2, x_2)}}, \quad (3.218)$$

we have

$$\langle I(\mathbf{r}, t) \rangle = I_1 + I_2 + 2\sqrt{I_1 I_2} |g^{(1)}(x_1, x_2)| \cos [\phi(x_1, x_2)]. \quad (3.219)$$

As in the classical case, we have

$$\begin{aligned} |g^{(1)}(x_1, x_2)| &= 1 && \text{complete coherence,} \\ 0 < |g^{(1)}(x_1, x_2)| &< 1 && \text{partial coherence,} \\ |g^{(1)}(x_1, x_2)| &= 0 && \text{complete incoherence.} \end{aligned}$$

At a fixed position, we can write the first-order correlation as a function of the time difference, $\tau = t_2 - t_1$,

$$g^{(1)}(\tau) = \frac{\langle \hat{E}^{(-)}(t) \hat{E}^{(+)}(t + \tau) \rangle}{\sqrt{\langle \hat{E}^{(-)}(t) \hat{E}^{(+)}(t) \rangle \langle \hat{E}^{(-)}(t + \tau) \hat{E}^{(+)}(t + \tau) \rangle}}, \quad (3.220)$$

which, for a single-mode field, can be written as

$$g^{(1)}(\tau) = \frac{\langle \hat{a}^\dagger(t) \hat{a}(t + \tau) \rangle}{\langle \hat{a}^\dagger \hat{a} \rangle}. \quad (3.221)$$

The following first-order correlation functions can be found,

$$g^{(1)}(\tau) = \begin{cases} e^{-i\omega_0 \tau} & \text{coherent state} \\ e^{-i\omega_0 \tau - |\tau|/\tau_{\text{coh}}} & \text{collision broadened thermal state} \\ e^{-i\omega_0 \tau - \frac{\pi}{2} \left(\frac{\tau}{\tau_{\text{coh}}} \right)^2} & \text{Doppler broadened thermal state,} \end{cases} \quad (3.222)$$

and are plotted in Fig. 3.16.

First-order coherence is capable of determining the coherence length of a light source, or the degree to which a light source is monochromatic; however, it tells us nothing about the statistical properties of the light. First-order coherence experiments are therefore unable to distinguish between states of light with identical spectral distributions. In other words, such experiments provide us with no information on

the photon number distributions of a state of light. For this, we must consider higher-order coherence experiments.

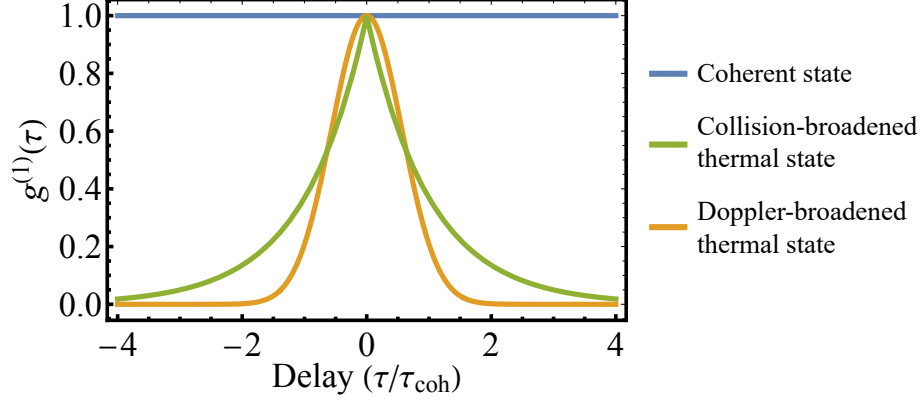


Figure 3.16: **First-order correlation, $g^{(1)}(\tau)$.** Expressions for the first-order correlation, $g^{(1)}(\tau)$, found in Eq. 3.222, are plotted for delay times normalized to the coherence length (τ/τ_{coh}) .

Classical second-order coherence

In the 1950s, a groundbreaking experiment from astronomers Hanbury Brown and Twiss developed a new kind of coherence experiment involving the correlation of intensities rather than of fields [372]. As seen in Fig. 3.17, this experiment splits an incoming light source onto two detectors and measures a delayed coincidence rate. One detector registers a count at time t while the other registers a count at time $t + \tau$. If the time delay, τ , is smaller than the coherence time of the source, information on the statistics of the light beam can be determined.

The coincidence count rate is proportional to the time average, $\langle I(t)I(t + \tau) \rangle$, where $I(t)$ and $I(t + \tau)$ are the instantaneous intensities at the two detectors. Note that here, these are classical quantities. If the average intensity at each detector is $\langle I(t) \rangle$, then the probability of obtaining a coincidence count is

$$\begin{aligned} \gamma^{(2)}(\tau) &= \frac{\langle I(t)I(t + \tau) \rangle}{\langle I(t) \rangle^2} \\ &= \frac{\langle E^*(t)E^*(t + \tau)E(t + \tau)E(t) \rangle}{\langle E^*(t)E(t) \rangle^2}, \end{aligned} \quad (3.223)$$

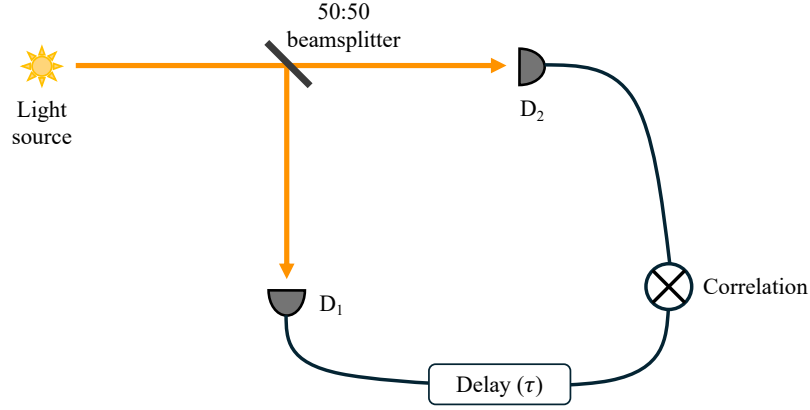


Figure 3.17: **Hanbury Brown-Twiss interferometer.** The radiation from a light source is split into two paths by a 50:50 beam splitter. Each path is directed towards a different detector, D_1 or D_2 . Time-delayed (τ) correlations between detection events at the two detectors are measured.

which is the classical second-order correlation function. Unlike the first-order correlation function, the second-order correlation function is not restricted to be unity or less. This can be seen by considering the second-order correlation for zero time delay,

$$\gamma^{(2)}(0) = \frac{\langle [I(t)]^2 \rangle}{\langle I(t) \rangle^2}. \quad (3.224)$$

It follows from the Cauchy-Bunyakovsky-Schwarz inequality that

$$\langle [I(t)]^2 \rangle \geq \langle I(t) \rangle^2, \quad (3.225)$$

and (with some math³²) it can be shown that there exists a limit for classical fields of

$$\gamma^{(2)}(\tau) \leq \gamma^{(2)}(0), \quad (3.226)$$

for which we will soon see that the quantum mechanical analog of this limit is violated by some quantum states. We also note that the classical second-order coherence can

³²See, *e.g.*, Gerry and Knight's *Introductory quantum optics* [314].

be written as

$$\gamma^{(2)}(\tau) = 1 + |\gamma^{(1)}(\tau)|^2. \quad (3.227)$$

If light incident on one detector is independent of the light incident on the other, there should be a uniform coincidence rate and thus also a uniform $\gamma^{(2)}(\tau)$. This was expected by Hanbury Brown and Twiss; but instead they observed twice the coincidence detection rate for $\tau = 0$ as compared with large τ . Evidently to them, the photons appeared to be arriving in “bunched” pairs. We note here that the light sources used by Hanbury Brown and Twiss included a mercury-vapour lamp [372] and later the light from the star, Sirius [373]—both thermal states of light.

Quantum second-order coherence

We can extend our arguments used in the case of first-order quantum coherence to the second-order by considering the joint counting rate between two detectors at $x_1 = (\mathbf{r}_1, t_1)$ and $x_2 = (\mathbf{r}_2, t_2)$. The transition probability for the absorption of a photon at each of the two detectors is proportional to

$$\left| \langle f | \hat{\mathbf{E}}^{(+)}(x_1) \hat{\mathbf{E}}^{(+)}(x_2) | i \rangle \right|^2, \quad (3.228)$$

which, after summing over all the possible final states, becomes the expectation value,

$$\langle i | \hat{\mathbf{E}}^{(-)}(x_1) \hat{\mathbf{E}}^{(-)}(x_2) \hat{\mathbf{E}}^{(+)}(x_2) \hat{\mathbf{E}}^{(+)}(x_1) | i \rangle. \quad (3.229)$$

This can be generalized to the case of mixed states through the density operator, where we define

$$G^{(2)}(x_1, x_2; x_2, x_1) = \text{Tr} \left[\hat{\rho}_F \hat{\mathbf{E}}^{(-)}(x_1) \hat{\mathbf{E}}^{(-)}(x_2) \hat{\mathbf{E}}^{(+)}(x_2) \hat{\mathbf{E}}^{(+)}(x_1) \right], \quad (3.230)$$

which can be interpreted as the ensemble average of $I(x_1)I(x_2)$. From here, the normalized second-order quantum correlation function can be defined,

$$g^{(2)}(x_1, x_2; x_2, x_1) = \frac{G^{(2)}(x_1, x_2; x_2, x_1)}{G^{(1)}(x_1, x_1)G^{(1)}(x_2, x_2)}, \quad (3.231)$$

which is the joint probability of detecting one photon at x_1 and a second at x_2 . At a fixed position, we can write this second-order correlation as a function of the time difference, $\tau = t_2 - t_1$,

$$g^{(2)}(\tau) = \frac{\langle \hat{E}^{(-)}(t) \hat{E}^{(-)}(t + \tau) \hat{E}^{(+)}(t + \tau) \hat{E}^{(+)}(t) \rangle}{\langle \hat{E}^{(-)}(t) \hat{E}^{(+)}(t) \rangle \langle \hat{E}^{(-)}(t + \tau) \hat{E}^{(+)}(t + \tau) \rangle}, \quad (3.232)$$

which is the probability of detecting a photon at $t + \tau$, conditional on detecting a photon at t . For a single-mode field, the second-order correlation function can be written as

$$\begin{aligned} g^{(2)}(\tau) &= \frac{\langle \hat{a}^\dagger(t) \hat{a}^\dagger(t + \tau) \hat{a}(t + \tau) \hat{a}(t) \rangle}{\langle \hat{a}^\dagger \hat{a} \rangle^2} \\ &= 1 + \frac{\langle (\Delta \hat{n})^2 \rangle - \langle \hat{n} \rangle}{\langle \hat{n} \rangle^2} \end{aligned} \quad (3.233)$$

Unlike the first-order correlation function, the second-order correlation function is able to provide insight into the photon statistics of a given light source, and is thus a useful—and common—characterization metric for quantum light sources. The following second-order correlation functions can be shown (*e.g.*, using the classical relation between the first- and second-order coherence in Eq. 3.227):

$$g^{(2)}(\tau) = \begin{cases} 1 & \text{coherent state} \\ 1 + e^{-2|\tau|/\tau_0} & \text{collision broadened thermal state} \\ 1 + e^{-\pi \left(\frac{\tau}{\tau_{\text{coh}}}\right)^2} & \text{Doppler broadened thermal state.} \end{cases} \quad (3.234)$$

For the photon number state, however, we find a very different second-order correlation function,

$$g^{(2)}(0) = \begin{cases} 0 & n = 0, 1 \\ 1 - \frac{1}{n} & n \geq 2, \end{cases} \quad (3.235)$$

which is outside the allowed range for its classical counterpart, $\gamma^{(2)}(0)$. This result can be interpreted as a quantum mechanical violation of the Cauchy-Bunyakovsky-Schwarz inequality. The second-order correlation for the different states of light we have considered are shown in Fig. 3.18.

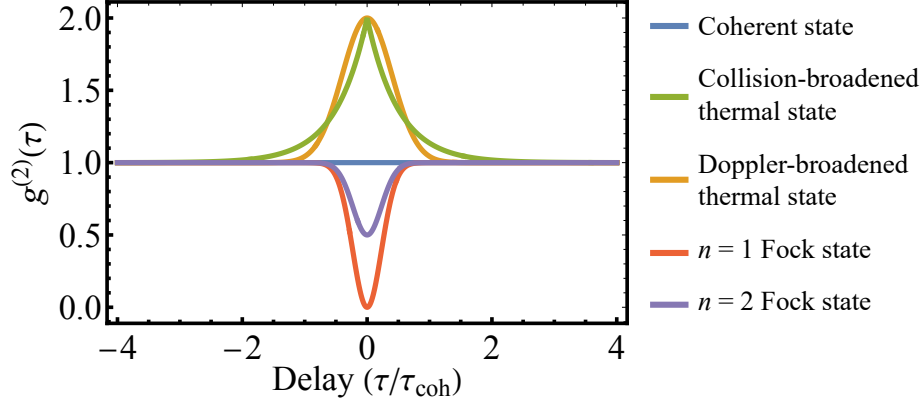


Figure 3.18: **Second-order correlation, $g^{(2)}(\tau)$.** Expressions for the second-order correlation, $g^{(2)}(\tau)$, found in Eqs. 3.234 and 3.235, are plotted for delay times normalized to the coherence length (τ/τ_{coh}).

Through Eq. 3.233, we can characterize the photon number statistics as follows

$$g^{(2)}(0) \begin{cases} > 1 & \text{super-Poissonian} \\ = 1 & \text{Poissonian} \\ < 1 & \text{sub-Poissonian.} \end{cases} \quad (3.236)$$

Classically, $g^{(2)}(\tau) \leq g^{(2)}(0)$, which is indicative of the *photon bunching* effects observed by Hanbury Brown and Twiss; however, some nonclassical states of light can exhibit a violation of this inequality, where $g^{(2)}(\tau) > g^{(2)}(0)$ for $\tau < \tau_{\text{coh}}$, indicating *photon anti-bunching* effects. We summarize the regimes associated with photon

arrival times as follows³³,

$$g^{(2)}(\tau) \begin{cases} \leq g^{(2)}(0) & \text{photon bunching} \\ = g^{(2)}(0) = 1 & \text{random} \\ > g^{(2)}(0) & \text{photon anti-bunching,} \end{cases} \quad (3.237)$$

which are depicted in Fig. 3.19.

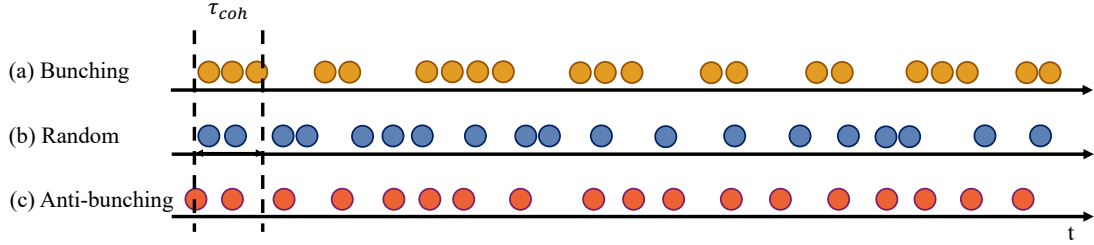


Figure 3.19: **Detection of photons as a function of time.** Effects of (a) photon bunching, (b) random photon statistics, and (c) photon anti-bunching.

3.4.4 Second-order autocorrelation

In the lab, we can use the second-order correlation function defined in the previous section to infer the photon statistics of our light sources. An ideal photon pair source emits a maximum of one photon pair per pump pulse; however, in practice, photon pair sources always have a non-zero probability of emitting multiple photon pairs for a single pump pulse. It should also be noted that noise in the system can lead to false detection events, degrading the purity of the source. A setup similar to that of Hanbury Brown and Twiss [372], as shown in Fig. 3.20, can be used to measure the second-order autocorrelation of photons produced via nonlinear processes, such as SPDC or SFWM.

³³It should be noted that often photon anti-bunching is confused as identical to sub-Poissonian statistics, but these are different effects [314]!

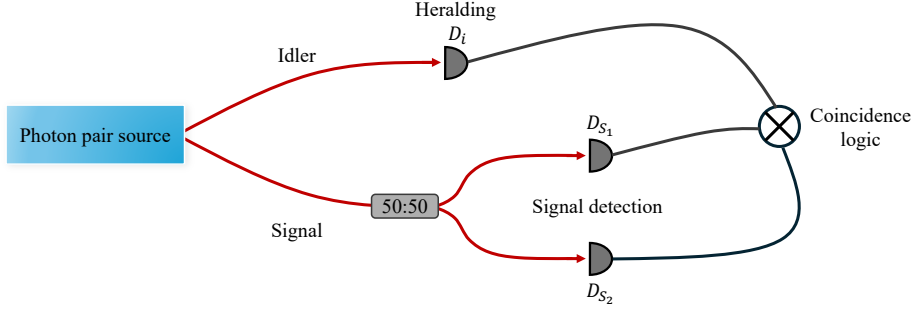


Figure 3.20: **Second-order autocorrelation for a photon pair source.** Idler and signal photons from a photon pair source are split. The idler photons are sent to detector D_i , providing the option of a heralded measurement. Signal photons are split onto two detectors, D_{s_1} and D_{s_2} , by a 50:50 beam splitter. Coincidence logic is used to record the various click patterns needed.

Heralded second-order autocorrelation

The heralded second-order autocorrelation provides a measure of the probability of such multiphoton pair events and false detection events due to noise photons. This measurement is performed by first splitting the generated photon pairs (either using a PBS or DM, depending on the nonlinear process). The idler photon is sent directly to a detector for heralding (D_i), while the signal photon is then split on a 50:50 beam splitter of which the output ports are directed towards two separate detectors (D_{s_1} and D_{s_2}). The heralded second-order autocorrelation is determined as follows,

$$g_H^{(2)}(0) = g_{s_1, s_2 | i}^{(2)}(0) = \frac{P_{s_1, s_2 | i}}{P_{s_1 | i} P_{s_2 | i}} = \frac{N_{s_1, s_2, i} N_i}{N_{s_1, i} N_{s_2, i}}, \quad (3.238)$$

where $P_{s_1, s_2 | i}$ is the probability of detecting a signal photon at detectors D_{s_1} and D_{s_2} conditioned on detecting an idler photon at detector D_i , while $P_{s_1 | i}$ and $P_{s_2 | i}$ correspond to the probabilities of detecting a signal photon at D_{s_1} or D_{s_2} , conditioned on the detection of an idler photon at D_i .

The heralded second-order autocorrelation can also be written in terms of the total numbers of various click patterns measured within a given time interval. Here, $N_{s_1, s_2, i}$ is the number of three-fold detection events measured between D_{s_1} , D_{s_2} , and

D_i , while N_i is the number of idler photons measured at D_i , and the number of two-fold detection events measured between D_{s_1} (or D_{s_2}) and D_i is given by $N_{s_1,i}$ (or $N_{s_2,i}$). An ideal photon pair source would have no possibility of a coincidence event occurring between the two signal detectors, s_1 and s_2 , implying that $g_H^{(2)}(0) = 0$; thus, in practice, a $g_H^{(2)}(0)$ as low as possible is desired. The heralded second-order autocorrelation will generally increase with pump pulse energy, as well as with the number of noise counts in the system.

Unheralded second-order autocorrelation

We can also measure the unheralded second-order autocorrelation for a photon pair source, by ignoring the idler photon. The unheralded second-order autocorrelation can be determined as

$$g_U^{(2)}(0) = g_{s_1,s_2}^{(2)}(0) = \frac{P_{s_1,s_2}}{P_{s_1}P_{s_2}} = \frac{N_{s_1,s_2}N_p}{N_{s_1}N_{s_2}}, \quad (3.239)$$

where we see that we no longer consider the probabilities conditioned on an idler photon detection event. The probability of event x is $P_x = N_x/N_p$, where N_p is the total number of events in a given time interval (*i.e.*, the number of pump pulses). The $g_U^{(2)}(0)$ relates to the purity of a photon pair source, P , and also provides a measure of the Schmidt number, K (the effective number of modes which the photons can be found in), through the relationship [374, 375]

$$g_U^{(2)}(0) = 1 + P = 1 + \frac{1}{K}. \quad (3.240)$$

We note that the unheralded second-order autocorrelation for a single-mode photon pair source ($K = P = 1$) approaches $g_U^{(2)}(0) = 2$, which indicates the same photon statistics as for a thermal state. On the other hand, for a highly mixed state containing many modes ($K \gg 1$ and $P \rightarrow 0$), $g_U^{(2)}(0) = 1$, indicating Poissonian statistics.

3.4.5 Second-order cross-correlation

Photon pair sources can also be characterized by the second-order cross-correlation function, which provides a means of examining correlations in the photon number

degree of freedom between the signal and idler photon. This metric can be determined through the experimental setup depicted in Fig. 3.21, and is calculated as

$$g_X^{(2)}(0) = g_{s,i}^{(2)}(0) = \frac{P_{s,i}}{P_s P_i} = \frac{N_{s,i} N_p}{N_s N_i}, \quad (3.241)$$

where $P_{s,i}$ is the probability of detecting a coincidence event between the signal and idler photon, while P_s and P_i are the probabilities of detecting a signal or idler photon, respectively. Again, we have that $P_x = N_x/N_p$, where N_p is the total number of events in the measurement interval (or rather, the number of pump pulses).

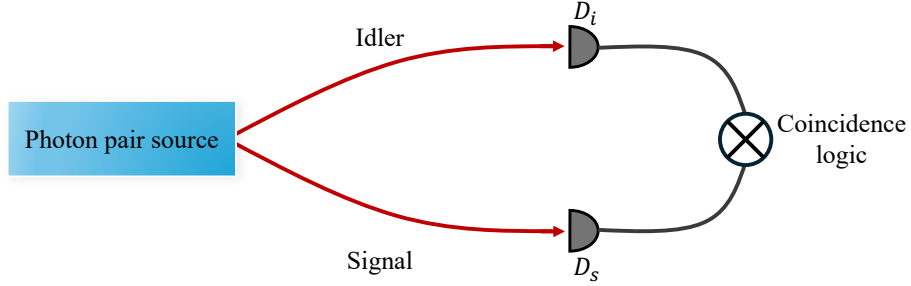


Figure 3.21: **Second-order cross-correlation for a photon pair source.** Idler and signal photons from a photon pair source are split and sent to detectors D_i and D_s , respectively. Coincidence logic is used to record the click patterns needed.

The second-order cross-correlation can be used to show that a photon pair source is indeed nonclassical by demonstration of a violation of the Cauchy-Bunyakovsky-Schwarz inequality [374],

$$\left(g_{s,i}^{(2)}(0)\right)^2 \leq g_{s,s}^{(2)}(0) \cdot g_{i,i}^{(2)}(0), \quad (3.242)$$

which relates the second-order cross-correlation to the unheralded second-order autocorrelation of the signal and idler photons. For classical light, $1 \leq g_{x,x}^{(2)}(0) \leq 2$, so any measurement in which $g_{s,i}^{(2)}(0) > 4$ indicates a nonclassical state.

3.4.6 Coincidence-to-accidental ratio

Another common metric used to characterize photon pair sources is the coincidence-to-accidental ratio (CAR). For a pulsed source, the CAR can be calculated as

$$\text{CAR} = \frac{P_{s,i}(0)}{P_{s,i}(nT)}, \quad (3.243)$$

where $P_{s,i}(0)$ is the probability of a coincidence event between the signal and idler photons at zero relative delay and $P_{s,i}(nT)$ is the probability of a coincidence event between signal and idler photons with a delay of $\tau = nT$ (n is an integer number and T is the period between pump pulses).

Since photon pair sources are probabilistic, the probability of a coincidence between a signal photon from one pump pulse and an idler photon from an adjacent pump pulse is simply the product of the individual detection probabilities, $P_{s,i}(T) = P_s P_i$. In this case, we see that the CAR reduces to Eq. 3.241, and is identical to the second-order cross-correlation at zero time delay.

3.4.7 Spectral and temporal correlations

When working with photon pair sources, an important consideration is whether a measurement of some property of one photon in a pair allows us to infer information about the other photon. Depending on the nonlinear process used to generate photon pairs, correlations may exist for Fourier conjugate variables, such as position and momentum or frequency (energy) and time. Here, we focus on quantifying correlations between photons in the frequency-time degree of freedom—noting that an analogous treatment can be used for other Fourier conjugate pairs. We can quantify the spectral and temporal correlations between photons from a pair source through the joint spectral intensity (JSI) and joint temporal intensity (JTI), respectively—or through the joint spectral amplitude (JSA) and joint temporal amplitude (JTA), respectively.

We first consider frequency correlations, which are quantified by the JSI or JSA. The bi-photon spectral wavefunction of a pair of photons can be written as [376],

$$|\Psi_{\omega_s, \omega_i}\rangle = \iint d\omega_s d\omega_i f(\omega_s, \omega_i) \hat{a}_s^\dagger(\omega_s) \hat{a}_i^\dagger(\omega_i) |0\rangle_s |0\rangle_i, \quad (3.244)$$

where $f(\omega_s, \omega_i)$ is the JSA and $|f(\omega_s, \omega_i)|^2$ is the JSI. The JSA is phase sensitive, while the JSI is not, where both depend on two functions: the phase-matching function, $\phi(\omega_s, \omega_i, \omega_p)$, and the pump spectral amplitude, $\alpha(\omega_p)$. We write the JSA as³⁴

$$f_{\text{SPDC}}(\omega_s, \omega_i) = \int d\omega_p \alpha(\omega_p) \phi(\omega_s, \omega_i, \omega_p), \quad (3.245)$$

$$f_{\text{SFWM}}(\omega_s, \omega_i) = \int d\omega_p \alpha(\omega_p) \alpha(\omega_s + \omega_i - \omega_p) \phi(\omega_s, \omega_i, \omega_p), \quad (3.246)$$

for the SPDC and SFWM processes, where the phase-matching function is given by

$$\phi(\omega_s, \omega_i, \omega_p) = \text{sinc}\left(\frac{\Delta \mathbf{k} L}{2}\right) \exp\left(\frac{i \Delta \mathbf{k} L}{2}\right). \quad (3.247)$$

We previously discussed phase matching in detail in Section 2.2.2, but as a reminder, $\Delta \mathbf{k}$ is referred to as the wavevector mismatch and is given by

$$\Delta \mathbf{k}_{\text{SPDC}} = \mathbf{k}_p - \mathbf{k}_s - \mathbf{k}_i \quad (3.248)$$

$$\Delta \mathbf{k}_{\text{SFWM}} = 2\mathbf{k}_p - \mathbf{k}_s - \mathbf{k}_i, \quad (3.249)$$

for SPDC and SFWM, respectively. The dependence on pump spectral amplitude arises due to the finite pump bandwidth and represents energy conservation, while the phase-matching function represents momentum conservation for a given pair of photon frequencies. The $\text{sinc}\left(\frac{\Delta \mathbf{k} L}{2}\right)$ part of the phase-matching function produces side lobes which are often undesirable as they can limit the purity of the generated photons [377]. In the lab, it is possible to apodize this functional form by spectrally filtering the two generated photons. The effect of spectral filtering can be accounted for by including filter functions, in the JSI expression,

$$f_{\text{SPDC}}(\omega_s, \omega_i) = \int d\omega_p \alpha(\omega_p) \phi(\omega_s, \omega_i, \omega_p) S_s(\omega_s) S_i(\omega_i) \quad (3.250)$$

$$f_{\text{SFWM}}(\omega_s, \omega_i) = \int d\omega_p \alpha(\omega_p) \alpha(\omega_s + \omega_i - \omega_p) \phi(\omega_s, \omega_i, \omega_p) S_s(\omega_s) S_i(\omega_i), \quad (3.251)$$

where $S_s(\omega_s)$ and $S_i(\omega_i)$ correspond to the spectral filtering performed on the signal and idler photons, respectively. Fig. 3.22 illustrates the various factors which influence

³⁴Note that by making use of a linear approximation for the phase mismatch, and modeling the pump as a Gaussian, it is possible to obtain an expression for the JSA in closed analytical form.

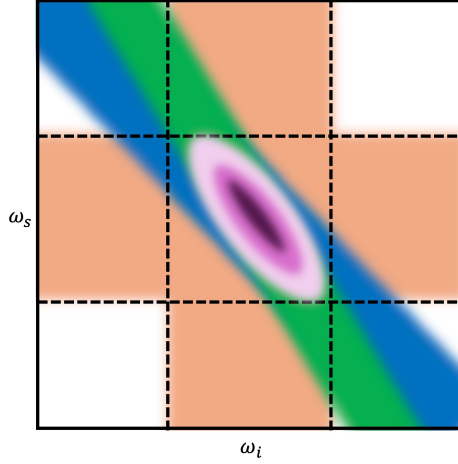


Figure 3.22: **Joint spectral intensity (JSI) illustration.** The pump spectral amplitude, $\alpha(\omega_p)$, is shown in blue, while the phase-matching function, $\phi(\omega_s, \omega_i, \omega_p)$, is shown in green. The spectral filtering functions, $S_s(\omega_s)$ and $S_i(\omega_i)$, are depicted by the larger orange regions with dashed outlines. The JSI, shown in purple, will be non-zero when all four of these functions overlap. Figure adapted from [378].

$f(\omega_s, \omega_i)$. Since it is not phase sensitive, the JSI is generally much easier to measure than the JSA, and so a variety of measurement approaches have been demonstrated for the JSI [379], including with scanning monochromators [300, 380], frequency-to-time mapping [381, 382, 383], Fourier transform spectroscopy [384], and stimulated emission tomography [385, 386, 387]. Stimulated emission tomography has also been used to demonstrate the more difficult phase-sensitive JSA measurement [388].

Photon pairs generated through SPDC or SFWM are naturally anticorrelated in frequency to satisfy energy conservation, while positively correlated in their time-of-arrival. Since SPDC and SFWM are probabilistic processes, there exists an uncertainty in the time at which photon pairs are generated (in other words, where in the nonlinear medium they were generated). But, regardless of when they were generated, the photons in a pair must have been produced simultaneously, leading to positive timing correlations. These temporal correlations can be described by the JTA, $F(t_s, t_i)$, which is given by the Fourier transform of the JSA,

$$F(t_s, t_i) = \iint d\omega_s d\omega_i f(\omega_s, \omega_i) e^{-i\omega_s t_s} e^{-i\omega_i t_i}, \quad (3.252)$$

where the JTI is given by $|F(t_s, t_i)|^2$. The bi-photon temporal wavefunction is then

$$|\Psi_{t_s, t_i}\rangle = \iint dt_s t_i F(t_s, t_i) \hat{a}_s^\dagger(t_s) \hat{a}_i^\dagger(t_i) |0\rangle_s |0\rangle_i. \quad (3.253)$$

The JSI and JTI for frequency-time correlated photon pairs are represented conceptually in Fig. 3.23. We note here that separable states, where $f(\omega_s, \omega_i) = f(\omega_s)f(\omega_i)$ and $F(t_s, t_i) = F(t_s)F(t_i)$, appear circular (or as vertical or horizontal ovals) in the JSI and JTI, as opposed to the correlated ellipses depicted in Fig. 3.23. The widths of these distributions along the $\omega_s = \omega_i$ and $t_s = -t_i$ axes can, together, be measured and used to demonstrate violation of the energy-time uncertainty relation, providing an entanglement witness [265, 300, 389].

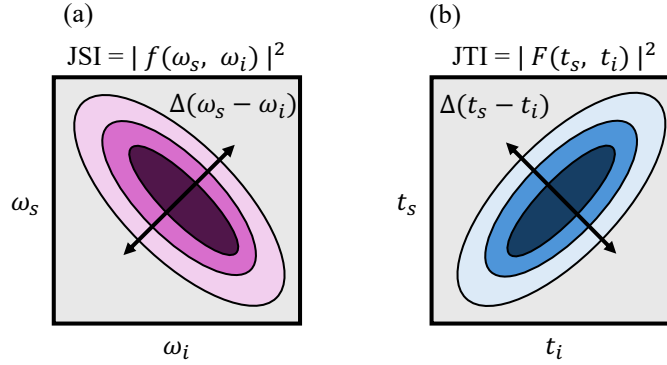


Figure 3.23: **Joint spectral and temporal correlations.** The joint spectral intensity (JSI) and joint temporal intensity (JTI) of frequency-time entangled photons is depicted. The widths of these distributions, indicated by the arrows, can be used to demonstrate violation of the energy-time uncertainty. Figure adapted from [378].

Oftentimes, ultrashort pulses are used to generate photon pairs through SPDC or SFWM, which leads to temporal correlations on ultrafast timescales. As such, measurement of the JTI can be challenging—but, not impossible with optical gating. Measurement of the JTI has been demonstrated using upconversion-based gating techniques [300, 390, 389]. Upconversion, *e.g.*, through sum-frequency generation (SFG), provides high timing resolution, but this technique can suffer from low efficiency. We have demonstrated an alternative approach using the all-optical Kerr gate presented in Chapter 2, which comes with its own set of challenges and advantages.

3.5 Characterization of ultrashort photon pairs

This section is based on the following publication:

A. R. Cameron, **K. L. Fenwick**, S. W. L. Cheng, S. Schwarz, B. MacLellan, P. J. Bustard, D. England, B. Sussman, and K. J. Resch. “Ultrafast measurement of energy-time entanglement with an optical Kerr shutter”, *Physical Review A* **108**(4): L041503 (2023).

DOI: <https://doi.org/10.1103/PhysRevA.108.L041503>

© 2023 American Physical Society

Another interesting “side quest” in our broader research effort towards all-optical quantum computing in the ultrafast regime included using the optical Kerr gate for ultrafast measurement of the JTI of a photon pair source. As discussed in Section 3.4.7, measuring the JTI of a photon pair source, pumped by ultrashort laser pulses, requires optical gating techniques. JTI measurement has been demonstrated through SFG gating [390]. This method was extended—by our collaborators at the *University of Waterloo’s Institute for Quantum Computing (IQC)*—in an impressive demonstration of the direct characterization of both spectral and temporal correlations in a photon pair source [300], and subsequently in the reconstruction of the full two-photon phase-sensitive JSA [389]. Despite the clear successes of SFG gating for measurement of temporal correlations in photon pair sources, this technique still has its limitations. By definition, SFG requires that the photons of interest undergo frequency conversion. Moreover, SFG is often limited in its efficiency, such that a full JTI measurement takes a long period of time. Given previous demonstrations of optical Kerr gating³⁵ with > 97% efficiency [258, 260], we sought to explore the feasibility of this gating technique for the measurement of the JTI of a photon pair source, as depicted in Fig. 3.24.

Unlike in previous demonstrations of the optical Kerr gate, this work requires a gated measurement of the temporal wave packet of each photon in the pair source; we do not wish to gate the entire photon wave packet, but a portion of the wave packet.

³⁵Note that in the publication presented in this chapter, the optical Kerr gate is referred to as an optical Kerr *shutter*. These terms can be taken as equivalent.

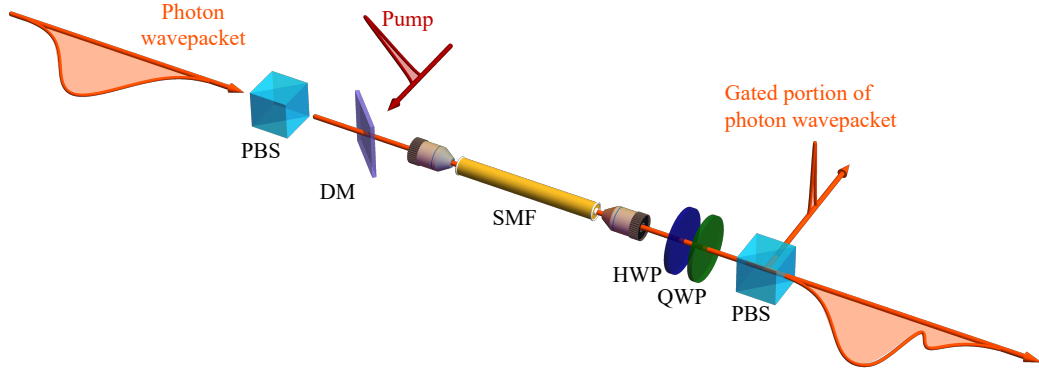


Figure 3.24: **Measurement of the temporal wave packet of a photon with an optical Kerr gate.** The optical Kerr gate is used to measure a portion of the temporal wave packet of a photon. The Kerr medium is a short piece of single-mode fiber (SMF), placed between two crossed polarizers. A strong pump pulse can be combined with the photon wave packet on a dichroic mirror (DM) before both beams are launched into the SMF. The pump pulse induces a birefringence in the SMF which rotates the polarization of a portion of the wave packet. The gated portion of the wave packet is redirected by a polarizing beam splitter (PBS). Note that two such Kerr gates are implemented in the work presented here to measure the joint temporal intensity (JTI) of the photon pair source. HWP: half-wave plate; QWP: quarter-wave plate.

This requires significantly short gate durations—shorter than the picosecond-duration gate demonstrated in the case of near-unit efficiency [258, 260]. In order to achieve a sufficiently short gate duration, we opted to use short lengths of SMF (3.5 cm) as the media for Kerr gating of the two photons. Unlike in SFG gating, where the resolution is ultimately limited by the response time of the nonlinear process, the resolution of an optical Kerr gate in SMF will necessarily be limited by the pump pulse duration and temporal walk-off due to dispersion. Nevertheless, with 3.5 cm SMF, we were able to achieve adequate gate resolutions for JTI measurement of 320 ± 30 fs and 290 ± 30 fs for the signal and idler photons, respectively. Should higher resolution be required, bulk crystals could be employed for optical Kerr gating [257]. Another means of improving the resolution exploits dispersion-engineered photonic crystal fiber (PCF), which has been implemented for walk-off reduction, where the group velocity of the pump and signal are matched [391]; however, such a regime requires that the optical gate be

placed within a Sagnac interferometer, which can be a complicated experimental design to maintain.

While this experiment demonstrated the successful measurement of both the JSI and JTI of ultrashort photon pairs, it certainly has its limitations. Perhaps the biggest limitation we encountered was due to noise induced by parasitic nonlinear processes within the optical Kerr gate. Although we knew beforehand that processes such as self-phase modulation (SPM), Raman scattering, and two-photon absorption could lead to unwanted noise counts in a Kerr-gated measurement, we had yet to operate in the wavelength regime presented here. As it turns out, the wavelength regime presented in this work is a considerably noisy one! For this reason, we were unable to operate the Kerr gates with near-unit efficiency—such pump powers would have generated far too many noise photons. We learned that caution must be taken when operating the Kerr gate in a regime where the pump pulses have higher frequency than the signal; in this case, noise due to Raman scattering plays a large role in the measurement capabilities. We also confirmed our suspicions that, in general, operating in a regime where the signal frequency is further from the pump pulse central frequency is often always better. For the experiment presented here, we were limited by the fact that we only had a single pulsed laser to pump both the photon pair generation process as well as the two optical Kerr gates. It is anticipated that improvements could be made to the signal-to-noise ratio (SNR) with the implementation of multiple, time-locked, wavelength-tunable ultrafast lasers.

Despite the limitations encountered in this experiment, we were still able to demonstrate the measurement of both the JSI and JTI of a photon pair source. Furthermore, these measurements allowed us to demonstrate a violation of the energy-time uncertainty relation, and therefore witness energy-time entanglement. This work provided us with knowledge of the resolutions achievable through optical Kerr gating in SMF, as well as the limitations of this technique—both important insights in the context of photonic quantum computing in the ultrafast regime. Further details on this work can be found in the theses of Sandra Cheng [392] and Andrew Cameron [378].

Ultrafast measurement of energy-time entanglement with an optical Kerr shutter

Andrew R. Cameron^{1,2,*} Kate L. Fenwick^{3,4,†} Sandra W. L. Cheng^{1,2} Sacha Schwarz^{1,2,5} Benjamin MacLellan^{1,2}
Philip J. Bustard³ Duncan England³ Benjamin Sussman^{3,4} and Kevin J. Resch^{1,2}

¹*Institute for Quantum Computing, University of Waterloo, Waterloo, Ontario, Canada N2L 3G1*

²*Department of Physics & Astronomy, University of Waterloo, Waterloo, Ontario, Canada N2L 3G1*

³*National Research Council of Canada, 100 Sussex Drive, Ottawa, Ontario, Canada K1A 0R6*

⁴*Department of Physics, University of Ottawa, Advanced Research Complex, 25 Templeton Street, Ottawa, Ontario, Canada K1N 6N5*

⁵*Infinite Potential Laboratories LP, 485 Wes Graham Way, Waterloo, Ontario, Canada N2L A07*



(Received 23 May 2023; accepted 2 October 2023; published 25 October 2023)

The energy-time degree of freedom has emerged as a promising avenue for photonic quantum technologies due to its intrinsic robustness against decoherence; however, the timescales associated with energy-time entangled photons make for difficult measurement and manipulation. We implement two optical Kerr gates, in short (35 mm) pieces of single-mode fiber, to achieve ultrafast measurement of the time correlations of energy-time entangled photon pairs, with resolutions of 320 ± 30 fs and 290 ± 30 fs for signal and idler photons, respectively. These measurements, in addition to joint spectral measurements of the photon-pair state, are used to verify entanglement by means of the violation of a time-bandwidth inequality. Without the need for an interferometric setup or frequency conversion, the optical Kerr shutter is a valuable addition to the ultrafast quantum optics control and metrology toolbox.

DOI: [10.1103/PhysRevA.108.L041503](https://doi.org/10.1103/PhysRevA.108.L041503)

The energy-time degree of freedom is important for many quantum technologies, including quantum networks [1,2], optical quantum computers [3], and quantum sensing [4]. This degree of freedom is useful due to its intrinsic robustness against decoherence for long-distance transmission of quantum information [5], increasing imaging resolution via interferometric techniques [6], and for realizing high-dimensional entangled quantum states [7]. Energy-time entangled pairs of ultrafast photons (femtosecond-picosecond duration) are challenging to control and measure with sufficient resolution. Measuring in the subpicosecond regime is particularly important because there has yet to be a single photon detector with comparable resolution. The highest detector resolution to date has been demonstrated with superconducting nanowire single-photon detectors (SNSPD), which have seen timing resolution on the order of a few picoseconds to tens of picoseconds, depending on the photon's frequency [8,9].

Fast gating of optical signals is commonly performed electronically by micromechanical switches [10,11] or electrooptic modulators [12] on nanosecond timescales. Optical gating has been used to surpass these timing restrictions, with resolutions of 450 ps in ring resonators [13], 10 ps in nonlinear optical loop mirrors [14], and subpicosecond with sum-frequency generation (SFG) [15]. Optical gating is paramount in the detection and control of subpicosecond energy-time-entangled photon pairs [16]. SFG optical gating has been used for time-resolved detection of energy-time entangled photon pairs [17], which can exhibit correlations in time on the order of a few femtoseconds. SFG temporal

measurements have been taken alongside spectral measurements to completely reconstruct a two-photon joint spectral amplitude [18].

A promising alternative method for ultrafast optical gating is to use an optical Kerr shutter (OKS) [19]. This method relies on the optical Kerr effect which can occur in any material, including those which are centrosymmetric. This makes it suitable for integration in standard single-mode fiber (SMF), where spatial overlap between the signal and pump is easy to achieve. In an OKS, the transient birefringence induced by a strong laser pulse will rotate the polarization of a photon pulse only where the two pulses temporally overlap in the Kerr medium. Picosecond and subpicosecond Kerr gating has been shown for classical applications, such as optical communications [20–22]. Previous demonstrations have shown near-unity efficiency operation of the Kerr shutter in YAG crystal [23] and short (10 cm) pieces of SMF on picosecond timescales [24–27]. In these cases, the group velocity walk-off between the signal and pump in the SMF was exploited to fully rotate the polarization qubit of a single photon for quantum communication and information applications. To measure the temporal waveform of a single photon, the Kerr shutter must be capable of operating at finer resolutions. This requires a unique Kerr shutter operation regime which has not previously been demonstrated with single-photon signals. Finer resolution can be achieved by operating with a signal and pump close in wavelength. This has been demonstrated for a classical signal, where a resolution as low as 305 fs was achieved [28]; however, this operation regime introduces noise from pump-self-phase modulation and cannot easily be operated above 30%. Walk-off reduction has also been implemented using photonic crystal fiber (PCF) as the $\chi^{(3)}$ medium to match the group velocities of both pulses [29]. Alternatively,

short lengths of SMF can be implemented to achieve finer Kerr shutter resolution.

In this work, we implement an OKS with a short 35-mm SMF for each entangled photon and demonstrate fast gated measurements with 320 ± 30 fs and 290 ± 30 fs resolution for the signal and idler photons, respectively. We characterize photon-pair correlations in time without the need for an interferometric setup. Unlike previous experiments using SFG for optical gating, no frequency conversion of the photons is required and the only difference between gated and ungated photons is their polarization. Here, we will be using this polarization rotation to measure time correlations; however, polarizing optics after an OKS could easily be used to reroute photons with subpicosecond resolution for use in other quantum information protocols.

Uncertainty relations can be used to detect energy-time entanglement through temporal and spectral measurements. Two separable photons labeled signal (*s*) and idler (*i*) must satisfy the inequality [30,31]

$$\Delta(\omega_s + \omega_i) \Delta(t_s - t_i) \geq 1, \quad (1)$$

where ω corresponds to frequency, t corresponds to time of arrival, and $\Delta(\omega_s + \omega_i) [\Delta(t_s - t_i)]$ signifies standard deviation in the sum of their frequencies (difference of their detection times) [32]. The quantities in Eq. (1) can be determined experimentally by measuring the joint spectral intensity (JSI) and joint temporal intensity (JTI) of a two-photon system. Violating this inequality is a sufficient condition for witnessing entanglement.

The expected two-photon temporal width can be estimated by the quadrature sum of the pump pulse at each shutter τ_p , the walk-off between the pump and signal (idler) photon, $\Delta\tau_{s(i)}$, and the intrinsic width of the JTI about the $t_s = -t_i$ axis, $\Delta(t_s - t_i)_{\text{Int}}$, as

$$\Delta(t_s - t_i) = \sqrt{\Delta\tau_s^2 + \Delta\tau_i^2 + 2\tau_p^2 + \Delta(t_s - t_i)_{\text{Int}}^2}, \quad (2)$$

where $\Delta\tau_{s(i)} = L(v_{g(s)}^{-1} - v_{g(p)}^{-1})$, L is the length of the Kerr medium, $v_{g(s(i)})$ is the group velocity of the signal (idler) photon, and $v_{g(p)}$ is the group velocity of the pump pulse. Note that the factor of 2 in front of the τ_p^2 term accounts for the pump pulse width at each of the two shutters. Photons from spontaneous parametric downconversion (SPDC) have strong correlations in time of arrival and exhibit an intrinsic width $\Delta(t_s - t_i)_{\text{Int}}$. This width is modeled by first considering the spectral correlations and then using a double Fourier transform to the time domain. The walk-off terms in Eq. (2), arising from the difference in group velocity between the pump and photons, further increase the width of the JTI. The effect of walk-off on temporal resolution is visualized in Fig. 1. This figure also illustrates the photon's temporal distribution, consisting of short photon pulses with different possible arrival times.

An illustration of the experiment is shown in Fig. 2. Optical pulses with 148-fs full-width at half maximum (FWHM) from a titanium sapphire laser with an 80-MHz repetition rate are guided to three parts of the experiment: an energy-time entangled photon source and two Kerr shutters. The pulsed light is first frequency doubled by a 2-mm β -bismuth borate (BiBO) crystal to 387.5 nm, spectrally filtered by a 0.1-nm FWHM bandpass filter, and then downconverted in a 5-mm

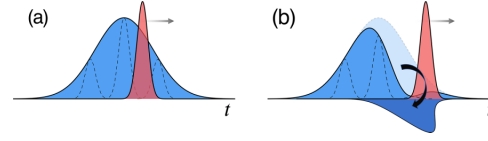


FIG. 1. Visualization of the main operating principle of an OKS. A 775-nm pump pulse walk-off in a 35-mm fiber in the reference frame of a 714-nm photon. (a) Initial temporal representations of the pump pulse relative to the photon. The broader photon distribution, shown in blue, is a combination of shorter photons created at varying times in the crystal, depicted with dotted lines. The pump pulse is shown in red. (b) Final temporal representation after the pump has swept through a section of the photon pulse, rotating its polarization. Delaying the pump pulse relative to the photon allows for gated measurement of different temporal segments of the photon.

BiBO crystal to produce energy-time entangled photon pairs with wavelengths 714 nm and 847 nm, which we refer to as the signal and idler, respectively. Each photon is spectrally narrowed to a bandwidth of 6-nm FWHM, after which we measure a signal count rate of $3.6 \times 10^6 \text{ s}^{-1}$, an idler count rate of $2.6 \times 10^6 \text{ s}^{-1}$, and a signal-idler coincidence count rate of $4 \times 10^5 \text{ s}^{-1}$ using a 3-ns coincidence window. Both photons are fiber-coupled and directed to (a) an OKS, or (b) a scanning monochromator.

The two photons travel through different lengths of fiber to reach their respective OKS. Ideally, our final measurement of the temporal profile of each photon would reveal the true transform-limited widths in time; however, dispersion in each fiber stretches the temporal profile of each photon. The signal photon passes through 50 cm of fiber which applies $10\,910 \text{ fs}^2$ of dispersion. The idler photon passes through 21.2 m of fiber which applies $344\,064 \text{ fs}^2$ of dispersion. One way to compensate for the dispersion in each path would be to build a grating compressor after each fiber to apply negative dispersion to each photon. Optimal gratings with a near-Littrow configuration provide a total compressor transmission efficiency of around 65% which drops further for shorter wavelength light [33–35], so to avoid unnecessary loss in the experiment we build a single grating compressor in the idler path. Due to the photon-pair time-of-arrival correlations, the gratings can locally cancel the dispersion of the idler and non-locally cancel the dispersion of its partner signal photon [36] by applying second order dispersion of $-(344\,064 + 10\,910) \text{ fs}^2$ on the idler photon. Experimentally, we adjust the grating position to minimize the temporal width of the JTI.

The signal and idler photon polarizations are both prepared along the horizontal axis, while the pump polarization is prepared at 45° relative to the photons to maximize Kerr rotation along the interaction region [24]. Each photon is combined with a pump pulse on a dichroic mirror and directed into a 35-mm SMF (Thorlabs S630-HP, cleaved to within 0.001-mm uncertainty). The length of the fiber L was selected to minimize walk-off in each OKS, $\Delta\tau_{s(i)}$ (which is directly proportional to L), while working within the length constraints imposed by the fiber mount. For these fiber lengths, $\Delta\tau_{s(i)}$ is estimated to be 297 fs and 256 fs for the signal and idler photons, respectively. Differences in fiber lengths due to cleaving uncertainty have a negligible impact on the

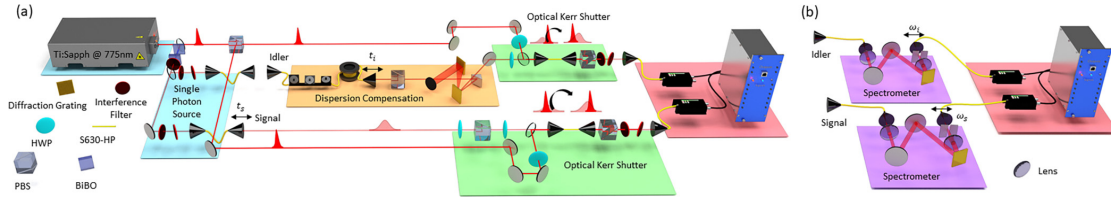


FIG. 2. Schematic of experimental setup. (a) A titanium-sapphire (Ti:Sapph) laser produces 775-nm pulsed light with a repetition rate of 80 MHz. The light is frequency doubled in a 2-mm β -BiBO crystal to 387.5 nm to pump the single-photon source. Energy-time entangled photon pairs are generated by Type-I SPDC in a 5-mm BiBO crystal. The 847-nm idler photon is sent through a 21.2-m fiber and a grating compressor for dispersion control of the idler photon and nonlocal dispersion compensation of the 714-nm signal photon. Each photon is Kerr gated by a strong pulse, picked-off from the output of the Ti:Sapph laser, within a 35-mm piece of SMF (Thorlabs S630-HP). Coincidence detection of the output of each OKS enables measurement of the JTI of the two-photon entangled state. (b) Each photon from the source can alternatively be directed to a scanning monochromator for measurement of the JSI. In this configuration, a grating is used to spread the single photon's optical frequency components spatially, after which a single-photon detector can be spatially scanned to detect a given frequency component.

difference in resolution. In fact, it is the difference in walk-off regimes between the signal and idler OKS which leads to this resolution difference. Operating with a difference in resolution is acceptable, provided each OKS is capable of sampling its photon in time.

Both devices have an equal input pump power of 800 mW. Unlike bulk media nonlinear optical crystals, SMF has the advantage that two modes are confined to a small core on the order of 5 μm in diameter which maintains high intensity to increase the nonlinear effect and also facilitates alignment. Pump and photon pulses were coupled into their respective SMFs, each with 40% coupling efficiency. In each OKS, only the part of the photon pulse that overlaps with the pump pulse in the Kerr medium will have its polarization rotated. This portion of the photon pulse will transmit through the Glan-Taylor polarizing beamsplitter (PBS) at the output of each 35-mm SMF. The pump pulse and photon are then separated by interference filters angle-tuned to pass the 6-nm bandwidth of the gated photons. The pump is 61 nm and 72 nm away from the signal and idler photons in wavelength, respectively, which is an equal 32 THz on either side of the pump; however, unwanted noise processes, such as self-phase modulation and Raman scattering, can generate pump noise in the spectral region of the single photons. As a result, tight spectral filtering is required for lowering background counts.

The light that transmits through the PBS and spectral filters, consisting of pump noise and Kerr-gated light from the single photons, is coupled to SMF, and detected by avalanche photodiodes (APDs). This SMF also acts as a spatial filter for light coupled to the cladding modes of the short 35-mm SMFs. Counts registered by each detector are time-tagged and analyzed with coincidence logic using a 3-ns coincidence window. The relative time of arrival between each photon and its corresponding pump pulse is varied by stepper-motors on the output fiber couplers before each OKS and denoted t_s and t_i in Fig. 2(a). Sweeping both motors in a raster scan pattern and counting coincidences at each position fully maps out the JTI of the two-photon system. As shown in Fig. 2(b), spectral measurements are made directly after the source with each photon fiber-coupled and routed directly to scanning monochromators, as in previous experiments [16,18].

Similarly, raster scanning the motors through frequency measurements on each photon builds up the JSI. We note that it would be possible to work with the energy-time degree of freedom on slower timescales than the requirements of this experimental setup, but only at the cost of requiring higher-frequency resolution in the scanning monochromators instead of high temporal resolution.

We define the maximum achieved gating efficiency to be the fraction of gated photons out of the total number of photons, acknowledging that the pump samples a short window in time and photons have varying arrival times related to the width of the down-conversion crystal and the probabilistic pair-generation. To measure the temporal profile of the single-photon wavepacket, each OKS must be operated such that it only ever samples a portion of the total number of photons; thus, the maximum achieved gating efficiency necessarily cannot be unity with this experimental setup. The maximum achieved gating efficiency is estimated to be approximately 16% at the peak of the JTI, and depends on the pump pulse duration and intensity, group velocity walk-off, and fiber length. Even if the pump could be operated at higher powers and provide a polarization conversion efficiency of 100%, we estimate the maximum possible gating efficiency to be 47% based on walk-off and pulse widths. The total efficiency of the entire temporal coincidence measurement which combines coupling losses from the grating compressor, both OKS coupling losses, both final SMF coupling losses, and gating efficiency is approximately 0.01%.

We present both the raw data and noise from the OKS temporal measurement in Fig. 3. Each pixel of the images in Figs. 3(a) and 3(b) is a coincidence measurement between the signal OKS and idler OKS with relative delays t_s and t_i . Figure 3(a) shows the raw data which includes the two-photon correlations as well as the unwanted background consisting of accidental coincidences and constant pump leakage, while Fig. 3(b) is an estimate of the background. The coincidence rates between the gated signal and idler photons are a function of electronic delay between the two detectors. Accidental coincidence peaks occur every 12.5 ns from the relative zero delay, corresponding to the repetition rate of the laser. The peak directly following the relative zero delay is used to

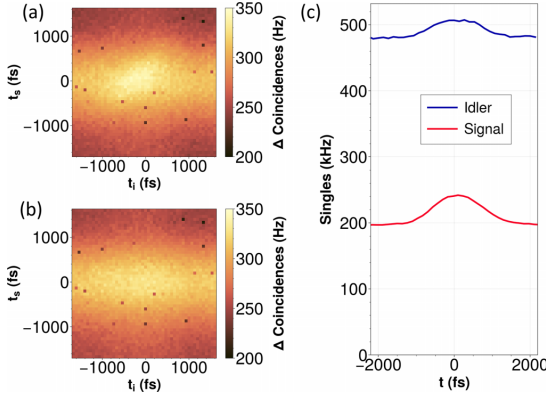


FIG. 3. Background subtraction in postprocessing. (a) Measured coincidences between two Kerr shutters at different relative gate delays t_s and t_i . (b) Estimation of accidental coincidences and constant pump background, measured by adding 12.5 ns of electronic delay between the two photon counting signals (corresponding to the repetition rate of the laser). The small number of artifacts that make a speckle pattern across the image are the result of a momentary fault in time tagging electronics. (c) Single counts at detectors after each OKS.

estimate accidental coincidences plus the constant pump leakage. The background profile in Fig. 3(b) reveals a horizontal “stripe” pattern in the accidental coincidences. The relative noise difference of the signal and idler OKS is apparent in Fig. 3(c). The idler noise rate is 480 kHz counts compared to the 200 kHz counts for the signal channel. This is because Raman scattering is higher on the low-frequency (Stokes) side of the pump pulse.

Both temporal and spectral correlations are presented in Fig. 4. The JTI in Fig. 4(a) is obtained by pixel-wise subtraction of the raw data and background estimation from Figs. 3(a) and 3(b). Note, there are two timescales of importance: A cross-sectional slice of the JTI corresponds to the temporal width of an individual photon with 320 ± 30 fs for the signal, and 290 ± 30 fs for the idler, while the marginals of the JTI correspond to the uncertainty in arrival time which we measure to be 470 ± 30 fs for the signal and 520 ± 30 fs for the idler. This can be visualized in Fig. 1 where the blue photon pulse has a much wider envelope than any given photon, shown as dotted lines, in the temporal distribution. Spectral measurements taken with single-photon scanning monochromators exhibit low noise and therefore do not require the background subtraction procedure discussed for the temporal measurements.

The JTI is measured by subtracting the measured background estimate in Fig. 3(b) from the raw data in Fig. 3(a). Measurement uncertainty takes into account the Poissonian statistics of the photon counting as well as the effect of background subtraction. Because the data and background are very similar in magnitude, the JTI measurement is highly sensitive to this background subtraction. As seen in Fig. 4(b), the baseline of the temporal trace is negative (due to background subtraction), implying that the background estimate introduces some uncertainty. We multiply the estimated

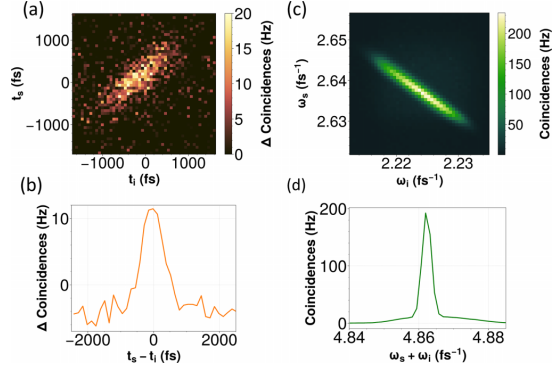


FIG. 4. Experimental characterization of the photon-pair temporal and spectral correlations. (a) Joint temporal intensity, (b) joint spectral intensity, (c) cross-sectional slices of (a) about the $t_s = -t_i$ axis, and (d) cross-sectional slices of (b) about the $\omega_s = \omega_i$ axis. For both (b) and (d), multiple slices were taken through the distributions and averaged together. The time-of-arrival of the two photons are positively correlated, while their frequencies are anticorrelated. Negative coincidence values in (c) are possible only because of the JTI background subtraction demonstrated in Fig. 3. Gaussian fitting to plots (c) and (d) yields $\Delta(t_s - t_i) = (340 \pm 30)$ fs and $\Delta(\omega_s + \omega_i) = (0.00141 \pm 0.00002)$ fs⁻¹. Together, these quantities demonstrate entanglement with $\Delta(t_s - t_i)\Delta(\omega_s + \omega_i) = (0.48 \pm 0.04)$ which is less than 1 by 13 standard deviations.

background by a variable scaling factor to determine the dependence of $\Delta(t_s - t_i)$ on the background level. This variation is included in the reported measurement uncertainty. We note here, for clarity, that this scaling factor is only used to analyze the effect of a sensitive background on the quoted JTI width error, not to adjust the background-subtracted data presented.

The estimate in Eq. (2) gives $\Delta(t_s - t_i) = (430 \pm 30)$ fs. Experimental data reveals a somewhat smaller width of $\Delta(t_s - t_i) = (340 \pm 30)$ fs. Many cross-sectional slices of the JTI about the $t_s = -t_i$ axis and JSI about the $\omega_s = \omega_i$ axis are averaged and shown in Figs. 4(c) and 4(d). Gaussian fits to these curves show an entanglement witness value of $\Delta(t_s - t_i)\Delta(\omega_s + \omega_i) = (340 \pm 30)$ fs (0.00141 ± 0.00002) fs⁻¹ = 0.48 ± 0.04 . This demonstrates a violation of Eq. (1) by 13 standard deviations indicating entanglement in the energy-time degree of freedom.

Our experimental parameters were based on a compromise of the various factors that influence efficiency, noise, and temporal resolution in an OKS. The OKS efficiency increases with pump intensity and fiber length; however, increasing these parameters also increases the generation rate of noise photons by self-phase modulation and spontaneous Raman scattering. In general, a large frequency difference between the pump and photon pulses will limit the generation of these noise photons at the signal and idler frequencies. In our normally dispersive SMF, an increased frequency difference increases the pump-photon group velocity walk-off and thus reduces the temporal resolution. We operated our experiment with a frequency difference $\Delta\omega = 32$ THz, fiber length $L = 35$ mm, and pump power $P = 800$ mW, yielding sufficient efficiency, signal-to-noise ratio, and temporal resolution, to

3.5. CHARACTERIZATION OF ULTRASHORT PHOTON PAIRS 171

ULTRAFAST MEASUREMENT OF ENERGY-TIME ...

PHYSICAL REVIEW A **108**, L041503 (2023)

measure energy-time entanglement. Future OKS pulse metrology experiments might operate with lower Raman scattering noise levels by using a pump pulse frequency lower than both the signal and idler pulses. One option for achieving this could include implementing two time-locked pump lasers: one for each OKS. Improvements in the signal-to-noise ratio (SNR) can be anticipated if the pump wavelength is not as restricted. For example, the OKS has seen near-unit efficiency and an SNR of 790 for a regime with an 800-nm pump and 685-nm signal photons [24]. Longer interaction lengths for improved efficiency could also be used, while still maintaining sufficient temporal resolution, by use of dispersion-engineered photonic crystal fibers for pump-photon group velocity matching [29].

In this work, we used optical Kerr shutters to directly measure the JTI of two energy-time entangled photons. Correlations were measured with a temporal resolution of

320 ± 30 fs and 290 ± 30 fs for signal and idler photons, respectively, which provided a sufficient gating resolution to violate a time-bandwidth inequality and therefore witness energy-time entanglement. With the capability of distinguishing events less than 1 picosecond apart, and therefore outside the temporal resolution of current detector speeds, the optical Kerr shutter is a valuable addition to the available methods in ultrafast quantum optics control and metrology.

The authors would like to thank John Donohue and Jean-Philippe MacLean for helpful discussions and assistance. This research was supported, in part, by the Natural Sciences and Engineering Research Council of Canada (NSERC), National Research Council Quantum Sensors Challenge Program, Canada Research Chairs, Industry Canada, the Canada Foundation for Innovation (CFI), Ontario Research Fund (ORF), and the Canada First Research Excellence Fund (CFREF).

-
- [1] R. Valivarthi, S. I. Davis, C. Pena, S. Xie, N. Lauk, L. Narvaez, J. P. Allmaras, A. D. Beyer, Y. Gim, M. Hussein *et al.*, *PRX Quantum* **1**, 020317 (2020).
 - [2] R. Tannous, W. Wu, S. Vinet, C. Perumangatt, D. Sinar *et al.*, *arXiv:2302.05038*.
 - [3] L. S. Madsen, F. Laudenbach, M. F. Askarani, F. Rortais, T. Vincent *et al.*, *Nature (London)* **606**, 75 (2022).
 - [4] Y. Zhang, D. England, A. Nomerotski, P. Svihra, S. Ferrante, P. Hockett, and B. Sussman, *Phys. Rev. A* **101**, 053808 (2020).
 - [5] A. Cuevas, G. Carvacho, G. Saavedra, J. Cariñe, W. Nogueira *et al.*, *Nat. Commun.* **4**, 2871 (2013).
 - [6] M. R. Brown, M. Allgaier, V. Thiel, J. Monnier, M. G. Raymer *et al.*, *arXiv:2212.07395*.
 - [7] F. Steinlechner, S. Ecker, M. Fink, B. Liu, J. Bavaresco *et al.*, *Nat. Commun.* **8**, 15971 (2017).
 - [8] B. Korzh, Q.-Y. Zhao, J. P. Allmaras, S. Frasca, T. M. Autry *et al.*, *Nat. Photon.* **14**, 250 (2020).
 - [9] G. G. Taylor, E. N. MacKenzie, B. Korzh, D. V. Morozov, B. Bumble *et al.*, *Appl. Phys. Lett.* **121**, 214001 (2022).
 - [10] K. Hogari and T. Matsumoto, *Appl. Opt.* **30**, 1253 (1991).
 - [11] C. Knoernschild, C. Kim, F. P. Lu, and J. Kim, *Opt. Express* **17**, 7233 (2009).
 - [12] P. Kolchin, C. Belthangady, S. Du, G. Y. Yin, and S. E. Harris, *Phys. Rev. Lett.* **101**, 103601 (2008).
 - [13] V. R. Almeida, C. A. Barrios, R. R. Panepucci, M. Lipson, M. A. Foster *et al.*, *Opt. Lett.* **29**, 2867 (2004).
 - [14] M. A. Hall, J. B. Altepeter, and P. Kumar, *Phys. Rev. Lett.* **106**, 053901 (2011).
 - [15] M. Allgaier, G. Vigh, V. Ansari, C. Eigner, V. Quiring *et al.*, *Quantum Sci. Technol.* **2**, 034012 (2017).
 - [16] J.-P. W. MacLean, J. M. Donohue, and K. J. Resch, *Phys. Rev. Lett.* **120**, 053601 (2018).
 - [17] O. Kuzucu, F. N. C. Wong, S. Kurimura, and S. Tsvetkova, *Phys. Rev. Lett.* **101**, 153602 (2008).
 - [18] J.-P. W. MacLean, S. Schwarz, and K. J. Resch, *Phys. Rev. A* **100**, 033834 (2019).
 - [19] D. England, F. Bouchard, K. Fenwick, K. Bonsma-Fisher, Y. Zhang *et al.*, *Appl. Phys. Lett.* **119**, 160501 (2021).
 - [20] N. Whitaker, H. Avramopoulos, P. French, M. C. Gabriel, R. LaMarche *et al.*, *Opt. Lett.* **16**, 1838 (1991).
 - [21] E. Yamada, K. Suzuki, and M. Nakazawa, in *Optical Fiber Communication Conference* (Optica, Washington, DC, 1995), p. ThL5.
 - [22] L. Moller, Y. Su, X. Liu, J. Leuthold, and C. Xie, *IEEE Photon. Technol. Lett.* **15**, 1597 (2003).
 - [23] C. Kupchak, P. J. Bustard, K. Heshami, J. Erskine, M. Spanner, D. G. England, and B. J. Sussman, *Phys. Rev. A* **96**, 053812 (2017).
 - [24] C. Kupchak, J. Erskine, D. England, and B. Sussman, *Opt. Lett.* **44**, 1427 (2019).
 - [25] F. Bouchard, D. England, P. J. Bustard, K. L. Fenwick, E. Karimi, K. Heshami, and B. Sussman, *Phys. Rev. Appl.* **15**, 024027 (2021).
 - [26] F. Bouchard, D. England, P. J. Bustard, K. Heshami, and B. Sussman, *PRX Quantum* **3**, 010332 (2022).
 - [27] F. Bouchard, K. Bonsma-Fisher, K. Heshami, P. J. Bustard, D. England, and B. Sussman, *Phys. Rev. A* **107**, 022618 (2023).
 - [28] K. L. Fenwick, D. G. England, P. J. Bustard, J. M. Fraser, and B. J. Sussman, *Opt. Express* **28**, 24845 (2020).
 - [29] M. Yabuno, T. Takumi, F. China, S. Miki, H. Terai *et al.*, *Opt. Express* **30**, 4999 (2022).
 - [30] L.-M. Duan, G. Giedke, J. I. Cirac, and P. Zoller, *Phys. Rev. Lett.* **84**, 2722 (2000).
 - [31] P. van Loock and A. Furusawa, *Phys. Rev. A* **67**, 052315 (2003).
 - [32] L. K. Shalm, D. R. Hamel, Z. Yan, C. Simon, K. J. Resch *et al.*, *Nat. Phys.* **9**, 19 (2013).
 - [33] J. Lavoie, J. M. Donohue, L. G. Wright, A. Fedrizzi, and K. J. Resch, *Nat. Photon.* **7**, 363 (2013).
 - [34] E. Treacy, *IEEE J. Quantum Electron.* **5**, 454 (1969).
 - [35] I. Walmsley, L. Waxer, and C. Dorrer, *Rev. Sci. Instrum.* **72**, 1 (2001).
 - [36] J. D. Franson, *Phys. Rev. A* **45**, 3126 (1992).

Chapter 4

Quantum information processing with single photons

Equipped with a background in ultrafast, nonlinear, and quantum optics, we can now discuss the details of our all-optical approach to photonic quantum information processing in the ultrafast regime. The computational complexity of photonic quantum information processors increases with the number of photons in use, so we first focus our attention to single-photon implementations in this chapter. Photonic quantum computing schemes all largely follow the same foundational framework of linear optical quantum computing, which is presented in Section 4.1. We then discuss, in Section 4.2, how platforms within this framework can be used to implement quantum algorithms and quantum simulations. The various encoding methods for photonic quantum computing are discussed in Section 4.3, including our ultrafast time-bin encoding scheme. This chapter concludes with two crucial proof-of-concept demonstrations of our ultrafast time-bin encoding scheme in action. In Section 4.4, we demonstrate a quantum computational algorithm (the quantum walk) using our platform. These capabilities are extended in Section 4.5, which presents a demonstration of the programmability of our photonic quantum circuits.

4.1 Linear optical quantum computing

A major advantage of photonic quantum computing is that its information carriers, photons, are exceptionally resilient to decoherence; the quantum information encoded in a photon tends to stay there. Yet this very advantage also has a downside: photons do not naturally interact with each other, making it difficult to construct quantum computational gates. In order to construct a photonic quantum computer, we require some means of introducing an effective interaction between photons. This remained a large obstacle to scalable photonic quantum information processing prior to the 21st century, as it suggested an apparent need for nonlinear optical interactions in the single-photon regime. Achieving the desired nonlinear couplings at sufficient strengths had been demonstrated, but remained technically challenging [393]. As a result, several proposals of linear optical schemes emerged in an attempt to tackle this challenge [394, 395, 396]; however, they required exponentially large physical resources. It was not until 2001, when Knill, Laflamme, and Milburn constructed a breakthrough protocol (called the *KLM protocol*) for efficient¹ photonic quantum computing using only single-photon sources, linear optical elements, and single-photon detectors [76].

The KLM scheme makes use of nonclassical interference to generate a phase shift which is nonlinear with regards to the photon number, conditioned on photon measurements in certain heralding modes. Such operations can be built into probabilistic logical gates, which are to be used in a “repeat-until-success” mode where the operation of a successful gate is then teleported onto the logical qubits. Although the gates themselves are probabilistic, the concatenation of a large number of these gate operations—and many ancilla² qubits—essentially allows for deterministic gate operations. Unlike encoding a single photon across many modes, the KLM proposal

¹In the context of computational theory, *efficient* algorithms require resources (*i.e.*, time, space) that scale no faster than polynomially. The KLM protocol demonstrates a low linear resource scaling.

²Unlike many classical logic gates, quantum logic gates are reversible. There is no way to deterministically put qubits into specific, prescribed states without having access to some qubits whose original state is known *a priori*. These qubits, for which the state is known in advance, are called *ancilla* qubits. Ancilla qubits play a critical role in quantum computing, as they provide a way to control and manipulate the state of the main computational qubits. Nondemolition measurements are possible with ancilla qubits, allowing for error correction.

demonstrated a resource-efficient implementation of two- and multi-qubit gates, albeit with a large overhead of ancilla qubits and their associated circuitry. With its proof of scalability, the KLM protocol led to a new paradigm of quantum computing with photons—referred to as *linear optical quantum computing* (LOQC)—and inspired a worldwide push towards universal quantum computation with photons. The KLM scheme has led to the development and improvement of a variety of photonic encodings, new schemes for quantum gates, and demonstrations of new protocols and algorithms [397, 398, 399, 400, 401]. Circuit-based approaches [244, 402, 403], which the work in this thesis focuses on, have also evolved from the KLM scheme and continue to be a promising path towards both intermediate-scale and universal quantum information processing. Another significant development in photonic quantum computing was the realization that a cluster-state model was well-suited to photonic qubits [77, 404], since large cluster states³ can—at least, in principle—be constructed efficiently using entangled photon sources and the teleportation gates used in the KLM protocol. The different approaches to LOQC are summarized in Table 4.1.

In the effort towards universal photonic quantum computing, many intermediate goals have emerged, providing short- to medium-term targets and opening up new paths towards full-scale devices. These intermediate goals include:

1. Developing individual photonic quantum gates of increasing complexity;
2. Designing universal multiport interferometers as scalable quantum circuits;
3. Constructing cluster-state-based photonic quantum computing schemes;
4. Devising new algorithms for sampling problems;
5. Implementing small-scale quantum algorithms and platforms to support them;
6. Developing scalable methods of encoding information in photons; and
7. Introducing simplifying and supporting techniques for each of the above.

³Cluster states are graphs in which qubits are the nodes and entangling links between qubits are the edges.

This thesis focuses on the development of a new, scalable quantum information encoding mechanism, ultrafast time-bin encoding (UTBE). In this chapter, we will demonstrate photonic quantum circuits in which we implement both a photonic quantum algorithm and universal multiport interferometer based on our UTBE platform.

Linear optical quantum computing (LOQC)

Gate-based approach: As in the KLM scheme, computations are performed by applying a series of gates on photonic qubits. With the ability to precisely control individual gates, this scheme offers high flexibility; however, many ancilla qubits are required which can be a challenge. Furthermore, operations are built into probabilistic logic gates, where success probability can be limited. Note that the gate-based approach and circuit-based approach are often considered to be the same.

Circuit-based approach: Computations are performed by sending photons through a large interferometric network, which applies an arbitrary unitary transformation. In some cases, such networks can be bulky and require many physical resources (though, we overcome this challenge here). Note that the circuit-based approach and gate-based approach are often considered to be the same. Here, we refer to large multimode interferometers when we say “circuit-based”. The boson sampling problem makes use of this type of scheme. Importantly, such multimode interferometers are expected to be useful in cluster-state-based approaches.

Cluster-state-based approach: Complicated entangled states (“cluster states” or “graph states”) are used to perform computations through a sequence of single-qubit measurements. This scheme is less reliant on individual gate success, but is considered *one-way* or *measurement-based* because the resource state is destroyed by the necessary measurements. The outcome of each individual measurement is random, but each measurement is related in such a way that the computation always succeeds. This offers the potential for increased computational efficiency, but requires careful design of the cluster state and precise measurement strategies.

Table 4.1: **Approaches to linear optical quantum computing (LOQC).** A summary of the current approaches towards LOQC.

4.1.1 Linear optical elements

The basic building blocks of LOQC are beam splitters and phase shifters. Following our creation and annihilation operator formalism developed in Chapter 3, we can describe these devices as unitary operators. As depicted in Fig 4.1, a beam splitter consists of a semi-reflective mirror, at which an incident beam of light will be partially transmitted and partially reflected. We denote a photon in either one of the two

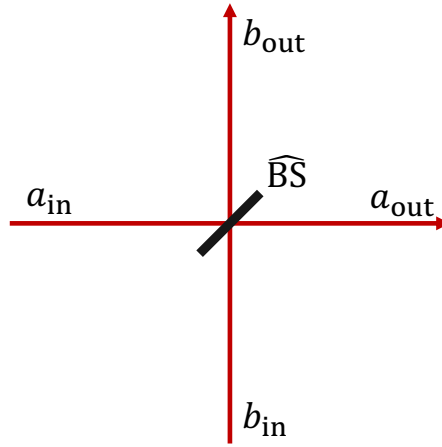


Figure 4.1: **A beam splitter (BS).** Input modes denoted by a_{in} and b_{in} , while output modes denoted by a_{out} and b_{out} .

incoming modes as $\hat{a}_{\text{in}}^\dagger$ or $\hat{b}_{\text{in}}^\dagger$, which are transformed by the beam splitter operator, $\widehat{\text{BS}}$, as⁴

$$\begin{pmatrix} \hat{a}_{\text{out}}^\dagger \\ \hat{b}_{\text{out}}^\dagger \end{pmatrix} = \widehat{\text{BS}} \begin{pmatrix} \hat{a}_{\text{in}}^\dagger \\ \hat{b}_{\text{in}}^\dagger \end{pmatrix} = \begin{pmatrix} \cos(\theta) & -e^{i\phi}\sin(\theta) \\ e^{-i\phi}\sin(\theta) & \cos(\theta) \end{pmatrix} \begin{pmatrix} \hat{a}_{\text{in}}^\dagger \\ \hat{b}_{\text{in}}^\dagger \end{pmatrix}, \quad (4.1)$$

which belongs to the $SU(2)$ group⁵ of rotations in the Bloch sphere. Here, we have parameterized the reflection, R , and transmission, T , of the beam splitter with the parameter θ , where $R = \sin^2(\theta)$ and $T = 1 - R = \cos^2(\theta)$. The relative phase between the two output ports is parameterized by ϕ . For a 50:50 beam splitter, $\theta = \pi/4$, and

⁴Here, we consider a lossless beam splitter [405, 406].

⁵ $SU(2)$ refers to the *Special Unitary Group of degree 2*. It is a group of 2×2 unitary matrices with a determinant of 1. The $SU(2)$ group is a strictly real Lie group, topologically compact and equivalent to a 3-dimensional sphere.

the phase is typically chosen to be either $\phi = 0$ or $\phi = \pi$. Alternatively, a symmetric beam splitter (which does not distinguish between which port a photon may enter) is obtained with $\phi = \pi/2$ ⁶. Mathematically, the parameters, θ and ϕ , represent the angles of rotation about two orthogonal axes in the Bloch sphere⁷, depicted in Fig 4.2. A mirror is a special case of a beam splitter, where $\theta = \pi/2$ ($R = 1$ and $T = 0$).

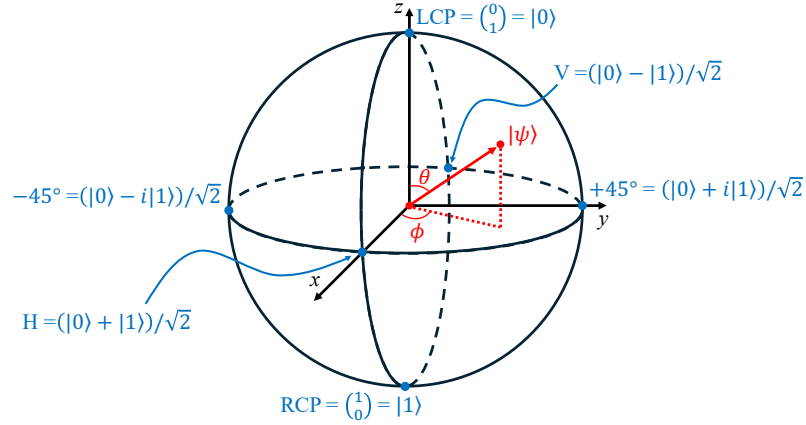


Figure 4.2: **The Bloch (or Poincaré) sphere.** Different polarization states and quantum mechanical states are depicted on the unit sphere.

An $SU(2)$ rotation can also be achieved with a combination of linear phase retarders (LPRs), *i.e.*, quarter- and half-wave plates. Wave plates are constructed from birefringent material, for which the index of refraction is different for light polarized along one axis relative to light polarized along the other (perpendicular) axis. The unitary operator, $\hat{P}(\delta, \psi)$, which describes a general LPR (with an optical retardation of δ and its optical axis at an angle of ψ with respect to the horizontal axis) is given by the Jones matrix⁸, acting on two input modes as

$$\begin{pmatrix} \hat{a}_{\text{out}}^\dagger \\ \hat{b}_{\text{out}}^\dagger \end{pmatrix} = \hat{P} \begin{pmatrix} \hat{a}_{\text{in}}^\dagger \\ \hat{b}_{\text{in}}^\dagger \end{pmatrix} = \begin{pmatrix} \cos^2(\psi) + e^{i\delta} \sin^2(\psi) & (1 - e^{i\delta})\cos(\psi)\sin(\psi) \\ (1 - e^{i\delta})\cos(\psi)\sin(\psi) & \sin^2(\psi) + e^{i\delta} \cos^2(\psi) \end{pmatrix} \begin{pmatrix} \hat{a}_{\text{in}}^\dagger \\ \hat{b}_{\text{in}}^\dagger \end{pmatrix}. \quad (4.2)$$

⁶Operationally, these two beam splitter descriptions are equivalent since there does not exist an absolute reference with which to measure the phase of the beam splitter. Choosing between the two often simply comes down to a matter of personal preference!

⁷Note that the Bloch sphere [407] is mathematically equivalent to the Poincaré sphere [408]. The Bloch sphere is more commonly used to define quantum mechanical states, while the Poincaré sphere is more commonly used to define polarization states in optics.

⁸The Jones matrix is used to describe how the polarization state of light changes as it passes through an optical element (*e.g.*, a lens, polarizer, or wave plate). It is part of the Jones calculus, which is a tool for analyzing polarized light and its interactions with optical systems.

A general $SU(2)$ unitary transformation can be constructed from a sequence of LPRs; for example, two variable optical retarders with retardation δ_1 and δ_2 , with optical axes at $\psi = 0$ and $\psi = \pi/4$, respectively:

$$\begin{aligned}\hat{P}(\delta_1, 0) \cdot \hat{P}(\delta_1, \pi/4) &= \begin{pmatrix} \cos(\delta_2/2) & -i\sin(\delta_2/2) \\ -i\sin(\delta_2/2) & \cos(\delta_2/2) \end{pmatrix} \begin{pmatrix} e^{i\delta_1} & 0 \\ 0 & 1 \end{pmatrix} \\ &= \begin{pmatrix} e^{i\delta_1}\cos(\delta_2/2) & -i\sin(\delta_2/2) \\ -ie^{i\delta_1}\sin(\delta_2/2) & \cos(\delta_2/2) \end{pmatrix}\end{aligned}\quad (4.3)$$

which is equivalent to a symmetric beam splitter with $R = \sin^2(\delta_2/2)$, and a phase shift of δ_1 in one arm. Since any two $SU(2)$ rotations along orthogonal axes can generate arbitrary rotations in the Bloch sphere, the above transformations are sufficient to implement any photonic single-qubit operation. With these linear optical elements, we can construct arbitrary linear optical circuits—this is at the heart of photonic quantum computing.

4.1.2 Circuit-based approach

The circuit-based approach to photonic quantum computing combines linear optical elements, as presented in the previous section, into multiport interferometers which are capable of implementing any arbitrary linear transformation between several optical channels. We reiterate here that one major technical challenge with this approach is getting photons to interact with one another; it is not impossible, but difficult. Since this thesis aims to introduce a new photonic quantum information encoding scheme, we first focus our attention here to photonic quantum circuits acting on only a single photon. The challenge of demonstrating multiphoton interactions is tackled next, in Chapter 5.

The remaining difficulties associated with constructing photonic quantum computational circuits arise from currently imperfect photon generation and detection, as well as from loss mechanisms (*e.g.*, scattering or absorption) in optical components

which make up the circuits. As such, it is crucial to minimize the optical depth⁹ necessary to construct multiport interferometers capable of performing any unitary transformation between a set of optical channels, as seen in Fig. 4.3(a). While useful

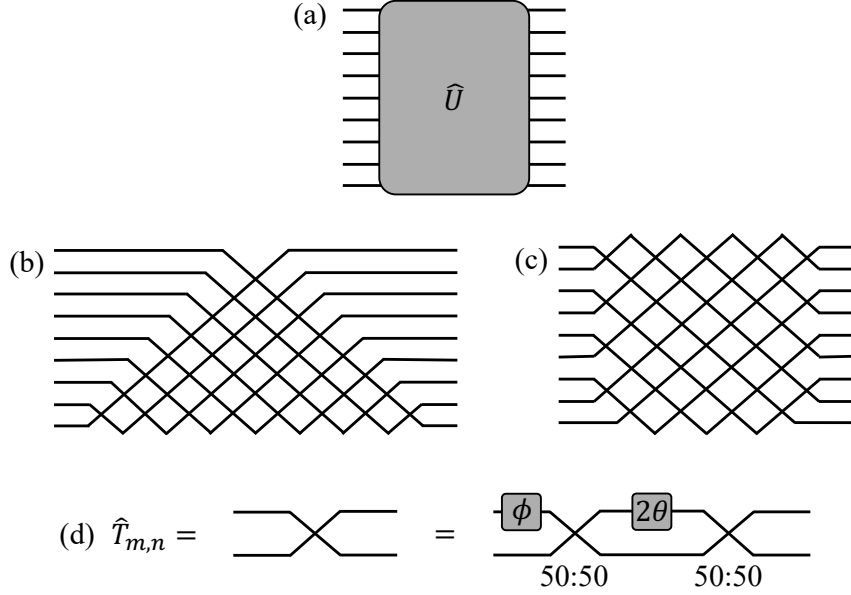


Figure 4.3: **Universal N -port interferometer.** The desired universal unitary transformation shown in (a) can be implemented using either (b) the triangular mesh of Reck *et. al.* [402] or (c) the rectangular mesh of Clements *et. al.* [403]. Each line corresponds to an optical channel (or mode), while crossings between channels correspond to a building block of the interferometer. As outlined in (d), these building blocks can be implemented physically using a single-mode phase shifter, ϕ , followed by a Mach-Zehnder interferometer, consisting of 50:50 beam splitters.

operations can be performed by large, yet nonuniversal, meshes of beam splitters and phase shifters [409], it was not until the seminal work of Reck *et. al.* in 1994 [402] that universal programmability was realized in these devices using the triangular mesh depicted in Fig. 4.3(b). This in turn led to an increased interest in the design of universal multiport interferometers [244, 410, 411], and in 2016, a new optimal design—based on the rectangular mesh shown in Fig. 4.3(c)—was realized by Clements *et. al.* [403].

⁹The optical depth is defined as the longest path through the multiport interferometer, corresponding to the number of optical elements (beam splitters, phase shifters) traversed by that path.

Both the triangular and rectangular mesh designs for a universal N -port interferometer minimize the total number of beam splitters required to $N(N-1)/2$. Although the total number of beam splitters is minimized in both mesh designs, the scheme of Clements *et. al.* reduces the optical depth by roughly a factor of two, which is crucial for minimizing optical losses and reducing the necessary resources (material, space).

In this thesis, we follow the rectangular mesh design of Clements *et. al.* [403]. Their ideal, lossless multiport interferometer can be described by an $N \times N$ unitary transformation, $\hat{U}$, capable of transforming a set of N input channels (modes) into any arbitrary set of N output channels (modes), such that $\hat{a}_{\text{in}}^\dagger \xrightarrow{\hat{U}} \hat{a}_{\text{out}}^\dagger$. The trick to the designs of both Reck *et. al.* [402] and Clements *et. al.* [403] comes from decomposing $\hat{U}$ into a product of matrices, $\hat{T}_{m,n}$, corresponding to the building blocks which make up the mesh (phase shifters and beam splitters). These building blocks should be capable of any transformation in $SU(2)$, which can be accomplished with a beam splitter of arbitrary reflectivity, preceded by a phase shift at one input port [405, 412]. A beam splitter with variable reflectivity can be substituted by a Mach-Zehnder interferometer [413, 414] comprising symmetric 50:50 beam splitters [402]. The building blocks of the universal multiport interferometer, depicted in Fig. 4.3(d), take the form

$$\hat{T}_{m,n} = \begin{pmatrix} 1 & 0 & \cdots & \cdots & \cdots & \cdots & \cdots & 0 \\ 0 & 1 & & & & & & \vdots \\ \vdots & & \ddots & & & & & \vdots \\ \vdots & & & e^{i\phi}\cos\theta & -\sin\theta & & & \vdots \\ \vdots & & & e^{i\phi}\sin\theta & \cos\theta & & & \vdots \\ \vdots & & & & & \ddots & & \vdots \\ \vdots & & & & & & 1 & 0 \\ 0 & \cdots & \cdots & \cdots & \cdots & \cdots & 0 & 1 \end{pmatrix}, \quad (4.4)$$

which is equivalent to the matrix in Eq. 4.3, with $\theta = \delta_1/2$ and $\phi = \delta_2$, acting on channels $\hat{a}_{m=n-1}^\dagger$ and $\hat{a}_n^\dagger$.

Both mesh schemes provide an explicit algorithm for writing any arbitrary unitary

matrix, $\hat{U}$, as

$$\hat{U} = \hat{D} \prod_{(m,n) \in S} \hat{T}_{m,n}, \quad (4.5)$$

where S denotes a specific ordered sequence of two-mode transformations and $\hat{D}$ is a diagonal matrix—equivalent to a set of phase shifts on the individual output channels—which is physically irrelevant for most physical applications. As shown by Clements *et. al.* [403], a physical multiport interferometer, composed of phase shifters and Mach-Zehnder interferometers in the rectangular mesh configuration of Fig. 4.3(c), with programmable parameters, θ and ϕ , in the $\hat{T}_{m,n}$ matrices, allows us to implement a universal transformation with $\hat{U}$.

Photonic quantum circuits built from such universal multiport interferometers allow for any arbitrary transformation on a set of input modes, making these devices a promising avenue for photonic quantum computing. Such circuits can support nonuniversal protocols, such as the boson sampling protocol discussed in the next section, as well as full-fledged LOQC. For the latter, it may be preferable to build smaller interferometers with a higher number of photons that can, for instance, perform programmable gates and support feed-forward operations¹⁰. Nevertheless, the implementation of a large, universal multiport interferometer is often an important step in demonstrating the feasibility of a particular photonic quantum information processing scheme or platform—as we focus on in this thesis.

4.1.3 Boson sampling protocol

A highly topical intermediate photonic quantum computing task is that of *boson sampling*, proposed by Aaronson and Arkhipov in 2011 [139] (which was based on the work of Troyansky and Tishby [415] in 1996). Although nonuniversal¹¹, there is strong theoretical evidence that a quantum advantage exists for the boson sampling protocol, thus making it a candidate for demonstrating the computational power of

¹⁰This is largely due to the fact that the computational complexity scales faster with multiple photons across a few modes, compared to one photon across many modes.

¹¹The boson sampling problem itself is nonuniversal; however, *tunable* boson sampling devices could, in theory, be used for universal quantum computation.

a given (bosonic) quantum computational platform¹².

The protocol implements a random N -mode universal interferometer, such as the one in Fig. 4.3(a), and requires the user to sample from the probability distribution of identical bosons (photons) at the output. It is difficult to obtain samples from this output probability distribution through classical computation, but computationally easy with a bosonic quantum circuit¹³. As its name suggests, boson sampling is well-defined for a system of any bosonic particles; however, photonic platforms are currently the leading candidates for implementing this sampling problem. Interestingly, the same computational advantage exists even when the photons arrive at random input modes of the circuit [417].

Quantum computational advantage has been demonstrated using photonic quantum circuits with 50 indistinguishable single-mode squeezed states in a 100-mode interferometer [18], and more recently, with 216 squeezed modes entangled with three-dimensional connectivity [19]. The latter makes use of photon-number-resolving detectors (PNRDs), as described in Section 3.3.3, in order to sample events with more than one photon. Furthermore, time-bin encoding is employed in the latter demonstration to access more squeezed modes without increasing the physical extent or complexity of their system. This shows the promise of time-bin encoding schemes—such as the one presented in this thesis—for photonic quantum computation.

Boson sampling does not require any ancilla qubits, adaptive feed-forward measurements, nor entangling operations. Although this makes the boson sampling protocol more straightforward to implement than the universal KLM scheme, it also means that boson sampling is ultimately a nonuniversal model of quantum computation. It is therefore important to recognize the possible limitations of systems that function as boson samplers. We note that the UTBE photonic quantum computing platform demonstrated in this thesis is more akin to that of a boson sampling protocol than the fully universal KLM scheme. Nevertheless, with a demonstration of promise in this more intermediate-scale, it is not unreasonable to anticipate that our encoding scheme could be extended to more general LOQC.

¹²It is worth mentioning here that boson sampling itself does not have much practical use (at least, that we know of) aside from approximating the permanent of a complex matrix [416] or demonstrating that a platform is capable of achieving a quantum computational advantage.

¹³Here, the circuit is essentially simulating itself!

4.1.4 Cluster-state-based photonic quantum computing

While conventional photonic quantum processors operate on initially uncorrelated input qubits in a gate-based architecture, cluster-state-based photonic quantum computing takes a different approach. The philosophy behind cluster-state (also called *one-way* or *measurement-based*) quantum computing [77, 404, 418] is to swap the “easy” and “hard” parts of the computation. Rather than send many uncorrelated photonic qubits (“easy” to generate) into a complex circuit (the “hard” part), a cluster state can be built up using nondeterministic interactions (“hard”), after which the computation is performed using deterministic single-qubit operations (“easy”). Since the first experimental demonstration of the essentials of one-way quantum computing [419], steps have been taken towards generating larger cluster states [116, 420], constructing larger computational networks [421], demonstrating new methods [422], and improving feed-forward designs [423]. Increasingly realistic schemes for large-scale photonic quantum computing have emerged, largely been driven by theoretical developments in one-way quantum computing [401].

In comparison to the KLM protocol, one-way photonic quantum computing is also more loss-tolerant [424]. Indeed, theoretical and experimental developments in this realm suggest that cluster-state-based photonic quantum computation may be a more realistic approach towards a full-scale photonic quantum computer. One advantage to using cluster states is that they can be generated and processed on-the-fly, without the need for quantum memories [425]. Another advantage is that cluster states can be built through nondeterministic fusion operations which merge two smaller entangled states into one [426, 427]. In the fusion-based protocol, the nondeterministic operation slightly reduces the size of the entangled states, but importantly, does not destroy them. Furthermore, missing links and nodes due to fusion failure or system losses are not necessarily detrimental to this protocol; error correction is possible, below a certain threshold [201, 428, 429, 430]. Evidently, these emerging cluster-state-based schemes are a promising approach to realizing universal photonic quantum computing. The fact that the cluster-state approach requires deterministic single-qubit operations makes this an attractive future avenue for application of our UTBE approach.

4.2 Photonic quantum algorithms

While it is undoubtedly important¹⁴ that efforts are made towards the development and implementation of quantum computational platforms for solving difficult problems, it is just as important to devise ways in which these systems can be used to solve such problems. In other words, we also need to develop quantum algorithms which can be implemented on quantum information processors. A classical algorithm is a finite set of instructions, providing a step-by-step procedure, fed into a classical computer in order to solve some problem; quantum algorithms are no different, but the step-by-step procedure makes use of quantum mechanical properties (*e.g.*, superposition, entanglement, or quantum interference) and, instead, can only be fed into a quantum processor. Perhaps the most widely known quantum algorithms are Shor’s algorithm for factoring [40] and Grover’s algorithm for searching unstructured databases or unordered lists [48], but many other algorithms exist.

In some cases, quantum algorithms are inspired by classical algorithms that have proven invaluable; for example, the *quantum walk*. The quantum walk is a quantum algorithm based on its classical counterpart, the *random walk*. quantum walks provide polynomial speedup over classical algorithms for many practical problems, including the element distinctness problem [432], the triangle-finding problem [433], and formula evaluation [434]. The boson sampling problem outlined in the previous section can be isomorphic to a multiphoton quantum walk when uncorrelated photons are input to the device [138]. Even the Grover search algorithm can be viewed as a quantum walk algorithm. Given its widespread applicability, this thesis demonstrates the implementation of a quantum walk—in addition to showcasing programmable photonic quantum circuits—using our UTBE approach.

4.2.1 Random walks: the inspiration for quantum walks

Random processes are fundamental in nature and ubiquitous in our lives. The random walk¹⁵, one of the most important stochastic processes in probability theory,

¹⁴Of course, some skepticism remains as to whether this field of research will deliver on all its promises [431]. What the future will look like, I cannot say; however, I do think it is important that efforts are made, because *what if it works?*

¹⁵Also sometimes called the *drunkard’s walk*.

provides a means of emulating the evolution of random processes [435]. Random walks have been used to describe a wide range of phenomena including, for example, Brownian motion [436], genetic drift in populations [437], stock share prices [438], and fixational eye movements [439]. They have also been used in applications such as image processing [440], and sampling from a large state space (*i.e.*, in Markov Chain Monte Carlo simulations) [441].

The basic principle of a random walk is nicely visualized by the *Galton board* shown in Fig. 4.4(a). A classical *walker* is depicted as a red bead, which traverses through a matrix of black pegs. Each time the walker hits a peg, it will have a 50:50 chance of being deflected to the left or right. For many trials of beads, a binomial probability distribution of beads in the buckets at the bottom of the board will emerge. The probabilities of the bead arriving at each peg are shown in Fig. 4.4(b). Note that Fig. 4.4 depicts a one-dimensional random walk. This algorithmic model can be extended to a quantum walker rather than the classical red bead. As we will see in the next section, the behavior of a quantum walker is remarkably different.

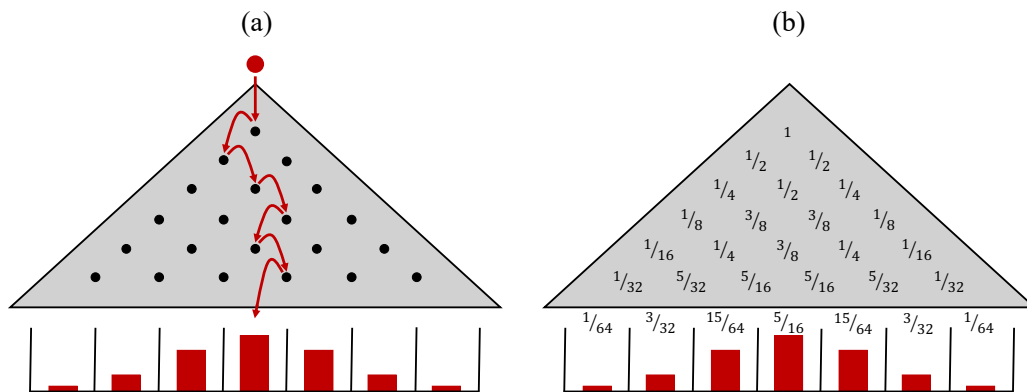


Figure 4.4: **Depiction of a discrete one-dimensional random walk.** The triangular Galton board depicted here helps visualize the random walk. A red bead (the *walker*) is dropped at the top of the board in (a) onto the first black peg. It has a 50:50 chance of being directed to the left or right. Here, it is first deflected to the left. The bead continues to traverse the matrix of pegs, each time having a 50:50 chance of being deflected left or right. If this experiment is run many times (*i.e.*, for many trials of red beads), a binomial probability distribution will emerge in the distribution of beads in the buckets at the bottom. The probabilities associated with arriving at each peg are shown in (b).

4.2.2 The quantum walk (QW)

Given the widespread applicability of the random walk, Aharonov, Davidovich, and Zagury introduced a quantum mechanical analog: the quantum walk (QW) [442]. In a QW, the walker is described by a quantum mechanical wave function that spreads across the different possible outcomes. The quantum walker remains in a coherent superposition of all possible paths through the QW, until detection at one of the output ports. As such, the probability amplitudes of the various trajectories can interfere, leading to a fundamentally different behavior in the QW compared to in the stochastic random walk. The output probability distribution is measured by repeating the QW many times. QWs can be either continuous [138, 443] or discrete [444]; in this thesis, we focus on discrete QWs.

Depicted in Fig. 4.5(a), a single photon (the quantum mechanical “bead”) traverses a matrix of beam splitters (the quantum mechanical “Galton board”). As the photon propagates through the QW, its wave function will evolve, according to the unitary operations applied by each beam splitter. Since the photon wavefunction remains in

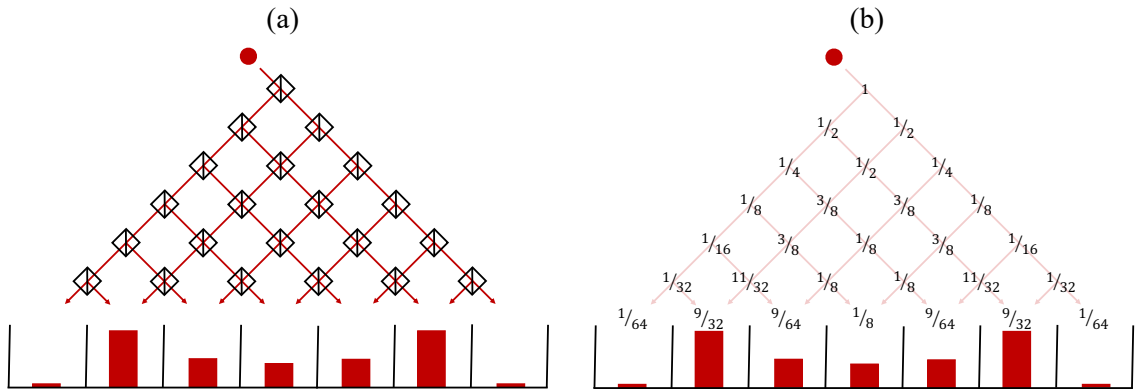


Figure 4.5: **Depiction of a discrete one-dimensional quantum walk (QW).** A photon (the walker) enters the QW at the top-most beam splitter in (a). Its wave function spreads across all possible paths as it propagates through the matrix of beam splitters comprising the QW, until it is detected at one of the possible output ports. If this experiment is run many times, the probability distribution at the bottom—strikingly different from the binomial distribution of the random walk—will emerge. The probabilities associated with each path are shown in (b).

a superposition of all possible paths throughout the QW, interference can occur. This interference leads to a remarkably different output distribution than in the stochastic random walk. Since the photon will only ever arrive at one of the detectors at the various output ports, multiple trials must be performed to observe the output probability distribution. Fig. 4.4(b) shows the probabilities associated with each path¹⁶. We note here that the QW can be viewed as a specific implementation of the photonic quantum circuits we discussed in the previous section.

The QW has become an invaluable tool, with applications in quantum simulation [445, 446, 447, 448, 449, 450], quantum search algorithms [451, 452, 453], quantum transport [454], and machine learning [455]. It has also been proven that QWs can be universal for quantum computing when encoded into qubits [444, 456] or when multiple interacting particles traverse the walk together [150]. Given this broad application space, the QW is both a promising quantum computational algorithm and also a useful testbed for current quantum information processing platforms. In this thesis, the QW algorithm is used to test and demonstrate the capabilities of our UTBE technique for quantum information processing.

4.3 Encoding methods in photonic quantum information processing

Our discussion of LOQC has so far largely focused on the action of beam splitters and phase shifters combined into photonic quantum circuits. It is natural to envision photonic quantum circuits which manipulate the path of a photon, but it is important to remember that encoding need not be limited to this degree of freedom. In fact, quantum information can be encoded in the many different photonic degrees of freedom, including path [18], polarization [237], frequency [180, 238, 457], temporal mode [458, 459, 460], orbital angular momentum (OAM) [240, 241, 461], or time of arrival [242, 249, 462]. Fig. 4.6 depicts each of these different encoding schemes, while Table 4.2 summarizes the various advantages and challenges associated with these encoding methods.

¹⁶These probabilities are calculated from the complex probability amplitudes.

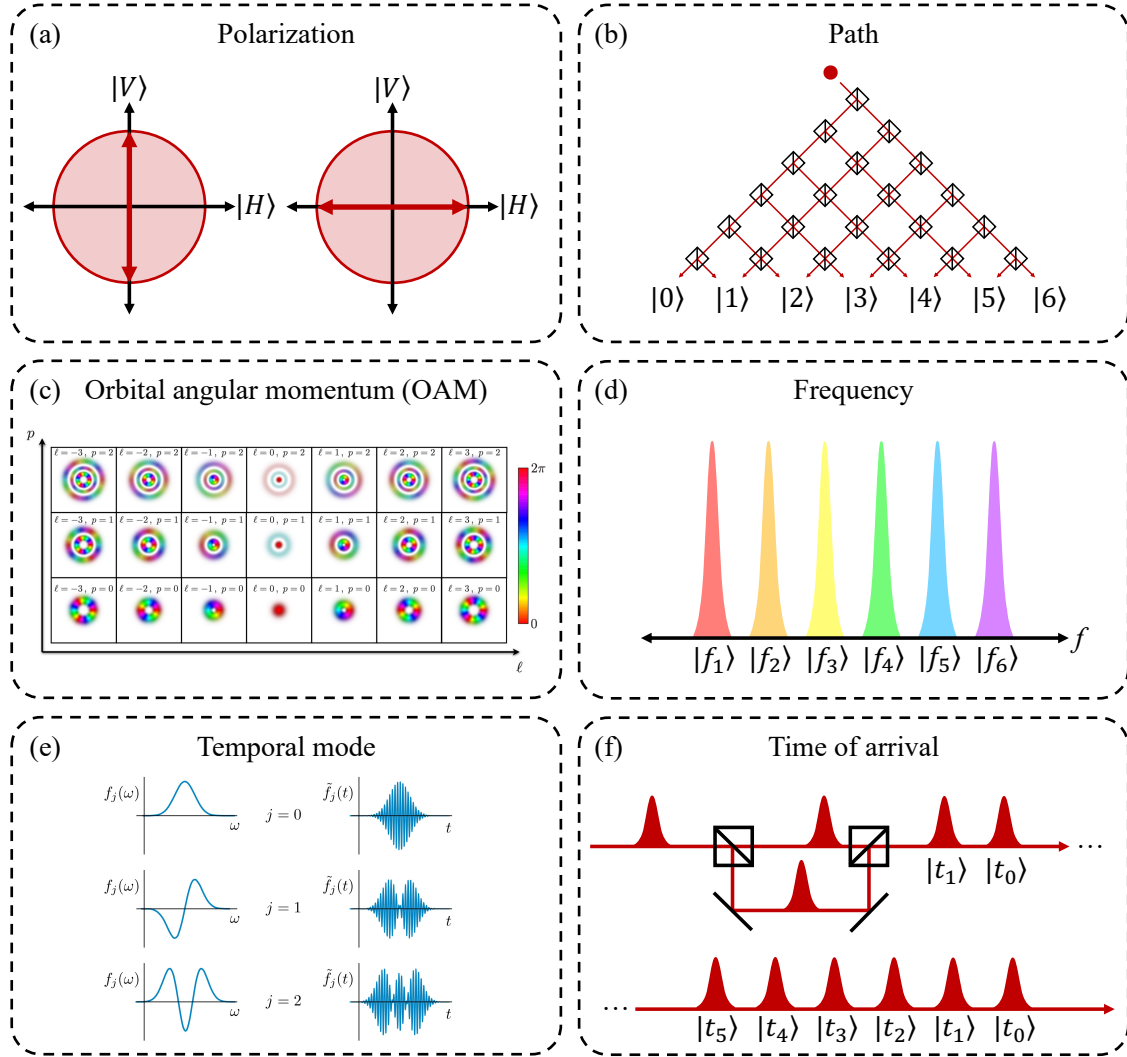


Figure 4.6: **Encoding methods in photonic quantum information processing.** (a) Polarization encoding; (b) path encoding; (c) orbital angular momentum (OAM) encoding, figure reproduced from [463]; (d) frequency-bin encoding; (e) temporal mode encoding, figure reproduced from [458]; (f) time-bin encoding. Details on these methods provided in Table 4.2.

Photonic degrees of freedom for encoding quantum information

Polarization (or spin angular momentum): A common, practical, and straightforward encoding method, given that linear optical components are capable of high-fidelity polarization preparation, transformation, and measurement. Although limited to a Hilbert space of two dimensions, it is becoming increasingly common to couple polarization to other degrees of freedom of single photons.

Path (or propagation direction): Quantum information is stored in the distinct paths a photon can take, *e.g.*, through a high-dimensional network of beam splitters. Although the technology requirements are low, accurate control can be challenging with free-space optics. Nevertheless, this method is well-suited for photonic integrated circuits, which provide accurate control and programmability.

Orbital angular momentum (OAM): Associated with the spatial structure of a photon’s wavefront, the OAM degree of freedom provides access to a high-dimensional Hilbert space—limited only by the accuracy and resolution of devices for preparation and manipulation. This method can be sensitive to decoherence due to turbulence in free-space, and challenging to propagate in optical fiber.

Frequency: Photons are prepared in equispaced and nominally identical *frequency bins*. This method is naturally high-dimensional, robust against decoherence over long-distance fiber networks and in free-space, and allows for propagation along a single optical path; however, manipulation requires quantum frequency conversion, which can be challenging to achieve.

Temporal mode: Associated with the temporal shape of a photon’s wave packet, temporal mode encoding spans a high-dimensional Hilbert space and is naturally suited for integration into existing single-mode fiber communication networks. Although robust against decoherence, this method requires coherent manipulation, *e.g.*, with a quantum pulse gate [464], which can be challenging to operate efficiently.

Time of arrival: This high-dimensional method prepares photons in equispaced *time bins*, along a single optical path. Only linear optical elements are required, but active stabilization of unbalanced interferometers may be required. Nevertheless, this method is suitable in fiber, integrated photonics, and free space. Time bin spacing must be sufficiently larger than detector jitter time.

Table 4.2: **Photonic quantum information encoding methods.** A summary of the advantages and challenges of each method is provided.

It should be noted that the number of photons in a given mode can also be considered as a degree of freedom in quantum information processing [465]; however, this requires PNRDs, which many laboratories do not currently have access to¹⁷. One quality that is often considered when choosing an encoding method is the dimensionality of the Hilbert space. The KLM scheme for LOQC requires *qubits*, where a photon is prepared in a superposition of two possible states associated with a particular photonic degree of freedom. This method is often referred to as *dual-rail encoding*. Yet, one major advantage of photonic quantum computing is that it need not necessarily be confined to qubits; many of the degrees of freedom which we have just discussed allow for the encoding of multilevel *qudits* (d -dimensional bits). Furthermore, it is possible to use several of the photonic degrees of freedom simultaneously [466, 467, 468, 469, 470], which provides a natural advantage to photonic quantum information processing schemes. The photonic quantum circuit schemes of Reck *et. al.* [402] and Clements *et. al.* [403], which we discussed in Section 4.1.2, provide a means of realizing such d -dimensional encoding with linear optics. In principle, these photonic quantum circuits could provide universal processing with a single photon over many modes, but one must be careful as these generally lead to exponential scaling of the number of optical components; however, with a measurement-based approach, *e.g.*, using cluster states with feed-forward capabilities as discussed in Section 4.1.4, these d -dimensional encoding mechanisms can indeed be quite powerful [471].

Other important considerations when choosing an encoding method for quantum information processing are: stability, ease of manipulation and programmability, system loss, and bandwidth. Each of these capabilities are crucial for scalability of quantum information processing systems. In general, the aforementioned encoding methods are each capable of achieving a subset of these capabilities; however, it is difficult to achieve all capabilities at once—especially without the need for complex technological requirements or feedback controls. This thesis presents a new encoding method, UTBE, which aims to achieve the key ingredients for scalability in quantum information processing.

¹⁷We have recently installed TES detectors with this capability at the *National Research Council (NRC) of Canada*, which opens up the possibility of encoding quantum information in the photon number degree of freedom!

4.3.1 Ultrafast time-bin encoding (UTBE)

Given the promise of measurement-based photonic quantum computing [206, 471], the ability to scale photonic circuits to a large number of modes is becoming increasingly crucial. It is therefore important that encoding methods can support scalability to many modes—while also maintaining low loss, high stability, straightforward programmability, and high bandwidth. Originally devised for quantum communication and networking purposes [249, 250, 251], *time-bin encoding* has recently emerged as a promising candidate for achieving the capabilities necessary for quantum information processing [19]. Unlike path encoding, time-bin encoding occurs along a single optical path, which simplifies alignment while also reducing the total number of optical elements and detectors required. It can be performed in free space, on-chip, or in optical fiber without the risk of decoherence. Moreover, it is straightforward to implement in that it only requires phase shifters and unbalanced Mach-Zehnder (or fiber-loop) interferometers, as shown in Fig. 4.7; yet, such interferometers must be phase stable.

To scale quantum processors based on the straightforward time-bin encoding method, a critical requirement will be maintaining interferometric phase stability across the entire device for extended periods of time. Oftentimes, active phase stabilization is required, which may be undesirable. Another alternative is to significantly reduce the path length difference within the interferometers in a quantum photonic circuit based on time-bin encoding—this is the underlying idea behind our introduction of UTBE to the landscape of quantum information encoding methods.

In UTBE, information is encoded in the arrival time of photons, on ultrafast timescales. This scheme makes use of ultrashort pulses to prepare single photons with hundreds-of-femtoseconds (10^{-13} s) duration, encoded in time bins that are separated by several picoseconds (10^{-12} s). Far from the timescales often associated with mechanical fluctuations and instability, this operation regime offers high phase stability without the need for active stabilization. As depicted in Fig. 4.7(a), with all-optical phase shifters (*i.e.*, optical Kerr gates) and mode couplers (*i.e.*, birefringent crystals), it is possible to construct the universal multiport interferometer of Clements *et. al.* [403] within an N -dimensional UTBE space. Each physical circuit element consists of an optical Kerr gate, followed by a birefringent α -barium borate (α -BBO) crystal, as shown in Fig. 4.7(b).

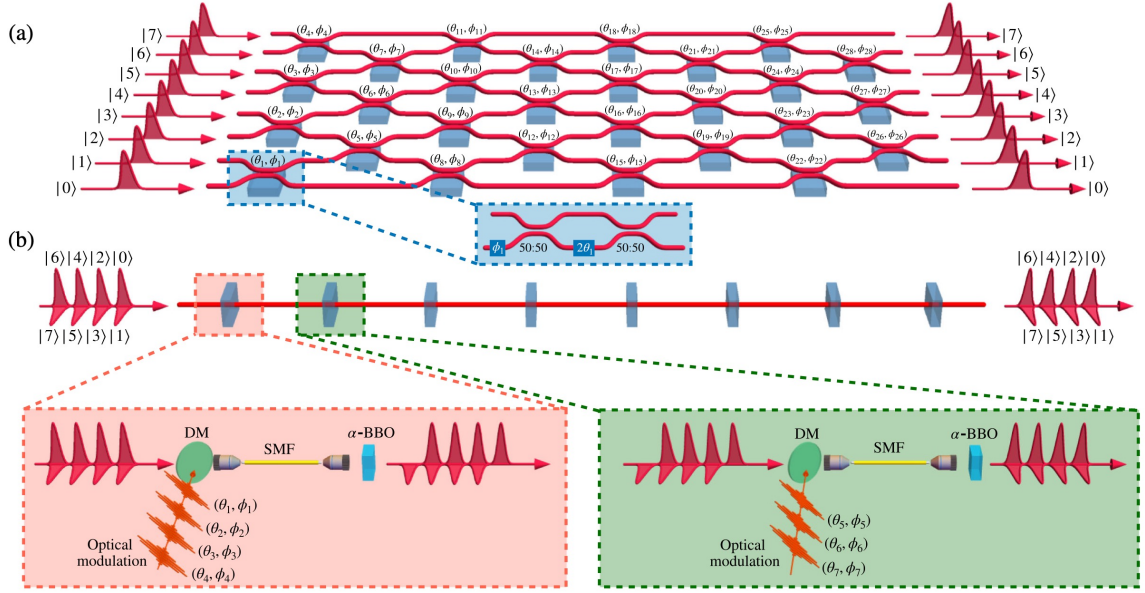


Figure 4.7: **Photonic quantum circuits with ultrafast time-bin encoding (UTBE).** (a) Photons traverse the N -dimensional UTBE space, which is depicted in terms of the familiar rectangular mesh of Clements *et al.* [403]. (b) In UTBE, a train of ultrafast time-bin states—each consisting of two possible polarization states—is processed by a programmable optical ultrafast temporal interferometric network element (POUTINE), depicted in the orange and green insets. The overall unitary transformation of the universal multimode interferometer is achieved by tuning the sequence of mode couplers (optical Kerr gates) and time-bin interferometers (birefringent α -barium borate (α -BBO) crystals). DM, dichroic mirror; SMF, single-mode fiber. Figure reproduced from [267].

Here, the N -dimensional encoding space is defined by N creation operators,

$$\{\hat{a}_{H,t_0}^\dagger, \hat{a}_{V,t_0}^\dagger, \hat{a}_{H,t_1}^\dagger, \hat{a}_{V,t_1}^\dagger, \dots, \hat{a}_{H,t_{N/2}}^\dagger, \hat{a}_{V,t_{N/2}}^\dagger\}, \quad (4.6)$$

associated with the horizontal (H) and vertical (V) polarization modes and ultrafast time bins, t_i , where $i \in 0, \dots, N/2$. Single-photon states can be written in the

computational basis¹⁸ as:

$$\{|\psi_i^N\rangle\} = \{|H\rangle \otimes |t_0\rangle, |V\rangle \otimes |t_0\rangle, \dots, |H\rangle \otimes |t_{N/2}\rangle, |V\rangle \otimes |t_{N/2}\rangle\}, \quad (4.7)$$

$$\text{where we use the convention } |H\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \text{ and } |V\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}, \quad (4.8)$$

where N is an even number. The overall universal unitary transformation of the multimode interferometer can be achieved through a sequence of $N(N-1)/2$ programmable optical ultrafast temporal interferometric network elements (POUTINES), which act between adjacent modes in the network. Each POUTINE consists of an optical Kerr gate pumped by a sequence of control pulses, followed by a birefringent α -BBO crystal.

The phase shift, ϕ_m , or coupling ratio, θ_m , of each POUTINE (labeled with index m) can be programmed by control pulses within an optical Kerr gate. In the work presented in this thesis, each control pulse manipulates the polarization within a specific time bin by applying a phase shift, thus coupling the two polarization modes within a time bin. This is described by the unitary transformation,

$$\hat{U}_{\text{Kerr}}^{(k)}(\theta_m, \phi_m) = \sum_j \left[\begin{pmatrix} e^{i\phi_{m(j,k)}} \cos \theta_{m(j,k)} & -i \sin \theta_{m(j,k)} \\ -ie^{i\phi_{m(j,k)}} \sin \theta_{m(j,k)} & \cos \theta_{m(j,k)} \end{pmatrix} \otimes |t_j\rangle \langle t_j| \right], \quad (4.9)$$

where $k \in \{0, \dots, N-1\}$ defines the circuit depth, for which we must sum over the operations across all modes, $j \in \{0, \dots, [N + (-1)^k - 1]/2\}$. It should be noted that two $N/2$ trains of control pulses are required for full programmability. The first train, consisting of control pulses that are co- or cross-polarized with the incoming photon, can be used to tune ϕ_m . The second train, consisting of control pulses that are diagonally polarized with respect to the incoming photon, can be used to tune θ_m . Nevertheless, for a large subset of transformations, a single train of control pulses can be used to manipulate ϕ_m and θ_m , by tuning the intensity and polarization of control pulses within each optical Kerr gate, which we demonstrate in this thesis.

¹⁸The computational basis simply refers to the basis that is most natural in a given context, in other words, its physical state space.

The Kerr gate unitary operation directly achieves the desired coupling between modes within a given time bin, but not for modes associated with different time bins. In order to couple the adjacent modes corresponding to different time bins, a birefringent α -BBO crystal is used, described by the unitary operation,

$$\hat{U}_{\text{Bir}} = \sum_{j=0}^{N/2} \left[\begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \otimes |t_j\rangle \langle t_j| + \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \otimes |t_{j+1}\rangle \langle t_j| \right]. \quad (4.10)$$

The birefringent crystal here acts as an unbalanced Mach-Zehnder interferometer in the time domain on ultrafast timescales. The thickness of the crystal provides the path length difference of the two arms in the interferometer. In order to achieve time bins of equal separation throughout the circuit, birefringent crystals of the same thickness must be used. It should be noted that these birefringent crystals will necessarily rotate the basis of the polarization components; a simple polarization rotation can be used to compensate for this, described by the unitary operation,

$$\hat{R}(\Theta) = \begin{pmatrix} \cos\Theta & -\sin\Theta \\ \sin\Theta & \cos\Theta \end{pmatrix} \otimes \sum_{j=0}^{N/2} |t_j\rangle \langle t_j|. \quad (4.11)$$

In practice, this polarization rotation can be achieved either using wave plates or by rotating the subsequent birefringent crystal.

The overall unitary operation associated with a single POUTINE is thus given by

$$\hat{\mathcal{U}}^{(k)}(\theta_m, \phi_m) = \hat{R}(\Theta) \cdot \hat{U}_{\text{Bir}} \cdot \hat{U}_{\text{Kerr}}^{(k)}(\theta_m, \phi_m), \quad (4.12)$$

and so the overall universal unitary transformation of the entire photonic quantum circuit is given by

$$\hat{U}^{(N)}(\theta_m, \phi_m) = \prod_{k=0}^{N-1} \hat{\mathcal{U}}^{(k)}(\theta_m, \phi_m). \quad (4.13)$$

Photonic quantum algorithms, such as the QW discussed in Section 4.2.2, can also be implemented using this UTBE encoding scheme. In the case of a “standard” QW,

in which each time bin is addressed in the same manner at a given circuit depth, one can employ a passive photonic circuit using UTBE, in which the Kerr gate operation is not required. We demonstrate our UTBE encoding scheme for such a QW in this thesis, as shown in Fig. 4.8.

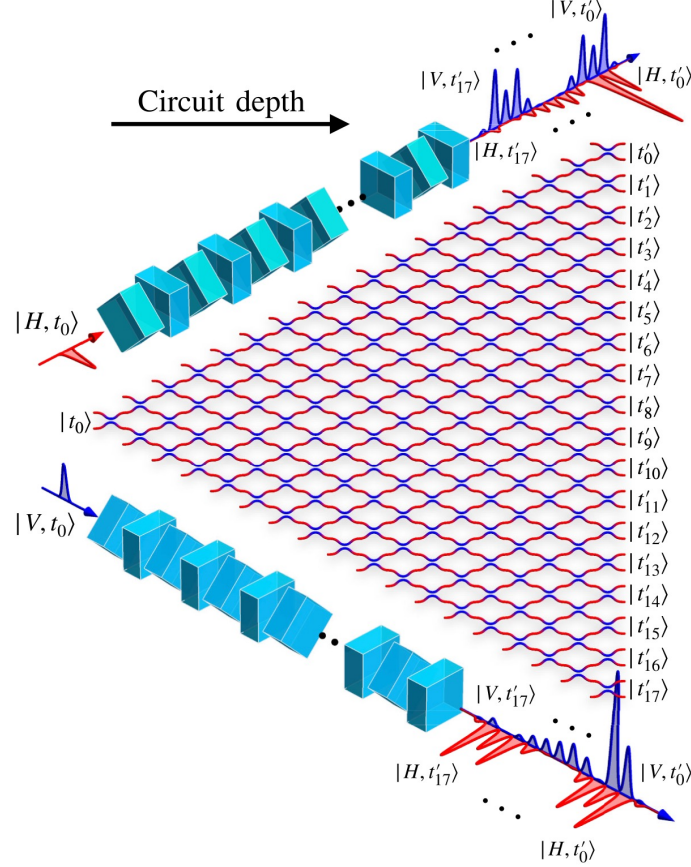


Figure 4.8: **Photonic quantum walk with ultrafast time-bin encoding (UTBE)**. Photons traverse the N -dimensional UTBE space, which consists of a passive photonic quantum circuit built with birefringent α -barium borate (α -BBO) crystals. Figure reproduced from [267].

The UTBE scheme demonstrates exceptional phase stability over remarkably long periods of time, consists of low-loss optical elements operating along a single optical path, and is capable of storing many modes within an ultrashort time frame. Each of these characteristics are key ingredients for scalable photonic quantum information processing.

4.4 Implementing a quantum algorithm

This section is based on the following publication:

K. L. Fenwick, F. Bouchard, G. S. Thekkadath, D. England, P. J. Bustard, K. Heshami, and B. Sussman. “Photonic quantum walk with ultrafast time-bin encoding”, *Optica* **11**(7): 1017–1023 (2024).

DOI: <https://doi.org/10.1364/OPTICA.510312>

© 2024 Optica Publishing Group

Previous demonstrations of qubits [261] and qudits [262] encoded in the ultrafast time-bin degree of freedom outline the promise of this method for quantum information processing. In this chapter, we expand upon these previous works and implement single-photon quantum information processors based on a UTBE scheme. Operating our photonic quantum information processors in the single-photon regime is an important step in developing our technique, allowing us to characterize key metrics such as: loss, stability, fidelity, and programmability. In this section, we focus on characterizing these parameters for a passive photonic quantum circuit by implementing the QW algorithm using our UTBE technique.

As discussed in Section 4.2.2, the QW is both a promising quantum algorithm for quantum simulation and a valuable testbed for characterizing quantum information processing platforms. Our primary aim here was to test the circuit depth limits of UTBE. The QW based on UTBE comprises a line of N birefringent α -BBO crystals, each of which acts as a Mach-Zehnder interferometer in the ultrafast time domain. A single QW step can be achieved with a single α -BBO crystal, which reduces the total number of optical components required in comparison to schemes which rely on path encoding. The first two steps of a QW with UTBE are shown in Fig. 4.9, to help visualize the interferometric action and timescales involved. We note here that each α -BBO crystal is placed in a kinematic rotation mount to allow full control over the three rotational degrees of freedom. Full control over the orientation of each crystal allows us to tune the splitting ratio of each interferometer and relative phase it imparts on the incoming photon wave function, without the need for additional wave plates. This further minimizes the total number of optical elements required.

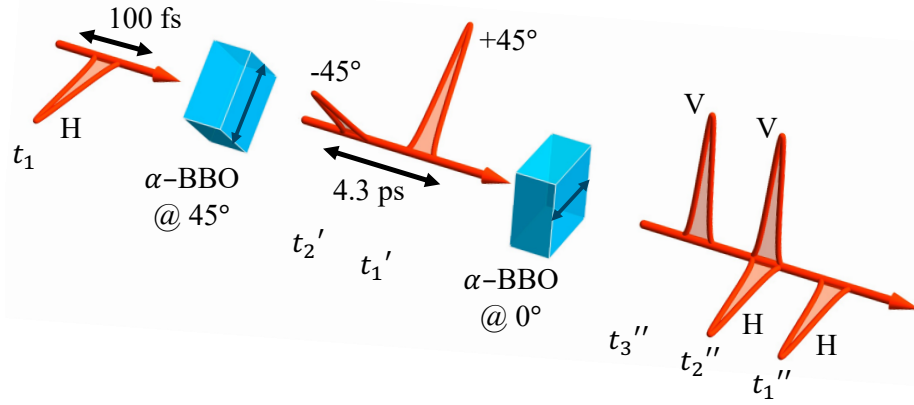


Figure 4.9: **Passive interferometers used in ultrafast time-bin encoding (UTBE).** A horizontally-polarized (H) single photon, with a 100 fs pulse duration, enters a birefringent α -barium borate (α -BBO) crystal in time bin t_1 . The fast axis of the crystal is set such that it is 45° relative to the polarization of the input photon. The input photon can then arrive in either the early time bin, t_1' , or late time bin, t_2' , each with a 50% probability. A second α -BBO crystal with its fast axis at 0° splits each of these time bins into two, leading to a train of three time bins, where interference occurs in the middle time bin, t_2'' . Depicted here, time bins are separated by 4.3 ps, which is determined by the crystal thickness and photon wavelength.

In addition to minimizing the total number of optical elements required for a passive QW, a key feature of this design is the remarkably low loss of each crystal. With the proper anti-reflection (AR) coating, the loss of a single crystal is < -0.045 dB; this is an important characteristic in the context of scalability and allows us to demonstrate here a photonic QW with up to 18 steps, which maintains an overall fidelity $> 95\%$. Here, our circuit depth of 18 was not limited by the system itself, but rather by the total number of α -BBO crystals found in our lab. It should be noted that some of these crystals did not have the proper AR coating and contributed closer to 0.1 dB of loss. Thankfully, the fix to this is straightforward with a bit of purchasing power. Even with this loss, the 38-mode interferometer was still capable of high-fidelity performance for a variety of input states.

Of course, working at the timescales outlined in Fig. 4.9 poses a challenge for most detectors. Although SNSPDs have seen remarkably low (3 ps) timing jitter in

recent years [11], such specialized designs are not currently commercially available and detector dead time can still be an issue. If you have somehow managed to stick with me through the lengthy previous chapters, you might now be thinking: why not use the optical Kerr gate for measurement? Indeed, this is what we do! Here, we implement our optical Kerr gate after the QW to measure its output. Notably, we work within a wavelength regime where we are capable of Kerr gating with efficiency of $> 99\%$. It is important to note that this value refers only to the efficiency of the Kerr gate operation; the efficiency of these measurement devices are still ultimately limited by coupling efficiency to the SMF used as the Kerr medium ($\sim 80\%$).

High-efficiency operation is crucial for scalability of quantum information processors; without a high-efficiency system, extension to the multiphoton regime will be limited. We note here that QWs based on UTBE have been demonstrated using an SFG gate for time-resolved detection [472, 473, 474]; however, such a gated measurement is ultimately limited in detection efficiency and indeed will be difficult to scale to the multiphoton regime. Although our implementation of the optical Kerr gate for measurement is currently a scanning-based method which necessarily takes time, one might consider implementing multiple optical Kerr gates in series to achieve simultaneous detection via time-division multiplexing. Of course, this will be ultimately limited by the total pulse energy available for optical Kerr gating. If using an 80 MHz titanium-doped sapphire (Ti:Sapph) laser as in this thesis, pulse energy will impose an upper limit on the total number of optical Kerr gates achievable; however, utilizing a lower repetition rate laser system is anticipated to allow for a significantly larger number of optical Kerr gates.

Perhaps the key attribute of our approach is the inherent phase stability at the timescales we operate at. The active phase locking in previous demonstrations of time-bin interferometers, with path differences reaching several kilometers [19], is a considerable achievement; however, this requirement can be eliminated altogether with our UTBE scheme. The passive circuit design used here allowed us to explore the stability of a large multimode interferometer (with up to 38 modes) based on UTBE. We found that the system remained phase-stable for over 50 hours, without the need for any active stabilization. Based on the flat trend observed in fidelity as a function of time, it is anticipated that the system will remain phase stable over

an even longer period of time. Further anecdotal evidence of the system stability is provided in our alignment protocol, which required each α -BB0 crystal to be mounted on a magnetic base. This allowed us to align the crystals comprising the QW in a pairwise manner: each additional crystal was aligned to its preceding crystal, with no other crystals present in the QW. Furthermore, these magnetic bases provided a straightforward means of measuring the QW output for an increasing number of steps. Notably, we found that it was possible to add and remove crystals from the QW setup over the course of many days and still preserve the interference.

One limitation of the passive design implemented here is that full programmability, where each time bin is addressed individually at each circuit depth, is not possible; however, a great deal of programmability is still present in this passive design. The full control over the three rotational degrees of freedom of each α -BBO crystal allows the user to arbitrarily program each step in the QW. In the case of quantum simulation using QWs, this is often all that is required in terms of programmability. Yet, we note here that full programmability could be achieved by implementing optical Kerr gates within the QW itself. Indeed, we will see such an extension in the next section of this chapter.

In demonstrating a QW using UTBE, we have shown that our platform is capable of performing a photonic quantum algorithm with high fidelity ($> 95\%$) and low loss (< -0.045 dB per step) for a large circuit depth (of up to 18 steps), for various input states (6 different input states) with remarkably high stability (demonstrated for > 50 hours). Additionally, we demonstrate some preliminary results for operation with two-photon input states—an important capability in the context of scalability. These capabilities will be extended upon in Chapter 5.

Photonic quantum walk with ultrafast time-bin encoding

KATE L. FENWICK,^{1,2}  FRÉDÉRIC BOUCHARD,^{1,*} GUILLAUME S. THEKKADATH,¹ 
DUNCAN ENGLAND,¹  PHILIP J. BUSTARD,¹  KHABAT HESHAMI,^{1,2} AND BENJAMIN SUSSMAN^{1,2}

¹National Research Council of Canada, 100 Sussex Drive, Ottawa, Ontario K1A 0R6, Canada

²Department of Physics, University of Ottawa, Advanced Research Complex, 25 Templeton Street, Ottawa, Ontario K1N 6N5, Canada

Received 24 October 2023; revised 3 July 2024; accepted 6 July 2024; published 18 July 2024

The quantum walk (QW) has proven to be a valuable testbed for fundamental inquiries in quantum technology applications such as quantum simulation and quantum search algorithms. Many benefits have been found by exploring implementations of QWs in various physical systems, including photonic platforms. Here, we propose a platform to perform quantum walks based on ultrafast time-bin encoding (UTBE) and all-optical Kerr gating. This platform supports the scalability of quantum walks to a large number of steps and walkers while retaining a significant degree of programmability. More importantly, ultrafast time bins are encoded at the picosecond time scale, far away from mechanical fluctuations. This enables the scalability of our platform to many modes while preserving excellent interferometric phase stability over extremely long periods of time without requiring active phase stabilization. Our 18-step QW is shown to preserve interferometric phase stability over a period of 50 h, with an overall walk fidelity maintained above 95%. © 2024 Optica Publishing Group under the terms of the [Optica Open Access Publishing Agreement](#)

<https://doi.org/10.1364/OPTICA.510312>

1. INTRODUCTION

Random processes are fundamental in nature and ubiquitous in life. The random walk (RW), one of the most important stochastic processes in probability theory, provides a means of emulating the evolution of random processes [1]. The quantum mechanical analog of the classical RW is the quantum walk (QW) [2,3]. Unlike its classical counterpart, the QW is described by a quantum mechanical wave function, which spreads across the different possible outcomes. Quantum interference effects in the wave function lead to drastically different outcomes than those observed in a classical RW, making the QW a powerful tool for observing the manifestation of non-classical effects. Beyond its fundamental importance in observing quantum mechanical wave function evolution, the QW has become an invaluable tool with applications in quantum simulations [4–8], quantum search algorithms [9–11], quantum transport [12], universal quantum computations [13], and machine learning [14,15]. Various other physical systems have been used to perform QWs, such as atoms [16], trapped ions [17,18], nuclear magnetic resonance [19], and superconducting qubits [20], but, due to the interference present in QWs, photonic systems lend themselves naturally to the task.

Experimental realizations of photonic QWs have been reported with walkers in both the spatial and temporal degrees of freedom using different implementations, such as bulk optics [21–32], fiber loops [33–40], fiber cavities [41], integrated photonics [42–47], and measurement-based approaches [48]. Nevertheless, practical limitations including the physical dimensions, the overall phase

stability, the loss per step, and the degree of control over individual QW steps, pose challenges to scaling the size and complexity of these systems. Time bin encoding [49,50] is attractive because all steps share a common spatial mode and, in the special case where every step is identical, this offers good phase stability. However, for a more general walk, multiple imbalanced interferometers with active stabilization are required. These challenges motivate the development of ultrafast time-bin encoding (UTBE) [7,8,15,51–54]. In the UTBE scheme, information is encoded in photon arrival time, with femtosecond-duration pulses spaced by several picoseconds. Ultrafast time bins can be manipulated using birefringent crystals [55,56], which, with appropriate coating, can be extremely low loss and remain phase-stable for days [57].

Readout of ultrafast time bins is unattainable by most single photon detectors; however, it can be achieved through optical gating. UTBE has previously been demonstrated for QWs with an upconversion detection scheme [7]; however, upconversion efficiency is often very low, which limits scalability to a larger number of walkers (photons). In comparison, a particularly promising method for measuring ultrafast time bins uses an all-optical Kerr gate [58], which has previously been demonstrated to achieve near-unit efficiency gating on picosecond timescales [59,60]. The Kerr gate has also been implemented at sub-picosecond timescales, on orders of hundreds-of-femtoseconds [61]. In this article, we propose and experimentally demonstrate an UTBE scheme for QWs using birefringent α -barium borate (α -BBO) crystals, which is read out by an all-optical ultrafast Kerr gate. We measure the step-by-step evolution of the QW for single-photon input states

with varied polarization and time-bin distributions. The QW is demonstrated to achieve high fidelity for up to 18 steps and remarkably high stability over long periods of time. The inherent phase stability of our UTBE scheme paves the way for scaling up experimental QW implementations to a larger numbers of steps while maintaining low loss at each step. Moreover, each step can be addressed separately, providing greater control over the QW parameters and potential for step-by-step programmability. This flexibility, combined with high detection efficiency, low loss per step, and inherent phase stability of our UTBE scheme, paves the way for more complex QW implementations with a large number of steps and multi-photon interference. Finally, we note that our UTBE approach is fundamentally platform-agnostic and can be implemented in a wide range of architectures. In particular, the small time-bin separation opens up new avenues for time-bin approaches with integrated photonics architectures.

2. EXPERIMENTAL IMPLEMENTATION

A QW acts on a quantum particle represented by a tensor product of two different basis states. One basis will define the space of the “walker,” and the other basis will define the space of the “coin.” As the quantum particle “walks through” each step of the QW, its probability distribution in the walker space is changed depending on its input coin state. Here, we use a single-photon-level pulse as the quantum particle in the photonic QW. Ultrafast time bins, with basis states $|t_i\rangle \in \{|t_0\rangle, |t_1\rangle, |t_2\rangle, \dots\}$, define the space of the walker. The polarization state of the photon takes the coin’s role, with basis states $|H\rangle$ and $|V\rangle$, corresponding to horizontally and vertically polarized light, respectively.

A single step, n , of our photonic QW, is given by the unitary step operator,

$$\hat{U}_n = \hat{S}_n \cdot \hat{C}_n. \quad (1)$$

The shift operator, $\hat{S}_n$, raises or maintains the time-bin state of the single photon depending on its polarization state,

$$\hat{S}_n = \sum_m (|H\rangle\langle H| \otimes |t_m\rangle\langle t_m| + |V\rangle\langle V| \otimes |t_{m+1}\rangle\langle t_m|), \quad (2)$$

and the quantum mechanical analog of the coin flip is given by the coin operator, $\hat{C}_n$, acting on the coin subspace such that

$$\begin{aligned} \hat{C}_n = \sum_m & \left(\cos\left(\frac{\Omega_n}{2}\right) |H\rangle\langle H| + e^{i\gamma_n} \sin\left(\frac{\Omega_n}{2}\right) |H\rangle\langle V| \right. \\ & \left. + e^{-i\gamma_n} \sin\left(\frac{\Omega_n}{2}\right) |V\rangle\langle H| - \cos\left(\frac{\Omega_n}{2}\right) |V\rangle\langle V| \right) \\ & \otimes |t_m\rangle\langle t_m|, \end{aligned} \quad (3)$$

where Ω_n and γ_n are parameters that can be adjusted to tune the probability amplitude and phase of the state, respectively. Different types of walks can be realized by varying the parameters of the coin operator, for example biased [62] and step-dependent coin [63] walks.

To achieve the step operator in Eq. (1), we use a birefringent crystal, α -BBO, with a very low loss per step of < -0.045 dB. The thickness of the α -BBO crystals sets the temporal separation between time bins. In this work, we use α -BBO crystals each with a thickness of 10 mm, providing a temporal separation of 4.3 ps at the signal wavelength. The orientation of each α -BBO crystal sets the coin operator, $\hat{C}_n$. As seen in Fig. 1, each step in the QW we present here is individually programmable, consisting of a single α -BBO crystal with access to three rotational degrees of freedom. In this work, we set $\Omega_n = \pi/2$ by rotating the fast axis of each α -BBO to alternating values of 45° and 0° , for odd and even steps, respectively. We also tune the tip and tilt of each α -BBO to set the phase to $\gamma_n = 0$. We note here for clarity that the α -BBO crystals, in their own crystal basis, implement only the shifting operator, $\hat{S}_n$, while the coin operator, $\hat{C}_n$, is implemented by the relative-basis rotations achieved by rotating and tilting each crystal about the optical axis, relative to the previous one. By exploiting the rotational degrees of freedom of each α -BBO, our QW scheme can be operated without the need for quarter- and half-waveplates (QWPs and HWPs), making it quite compact.

The QW comprises a line of N birefringent α -BBO crystals, each of which performs one step in the walk corresponding to the operator $\hat{U}_n$. The total evolution of an N -step QW is therefore given by

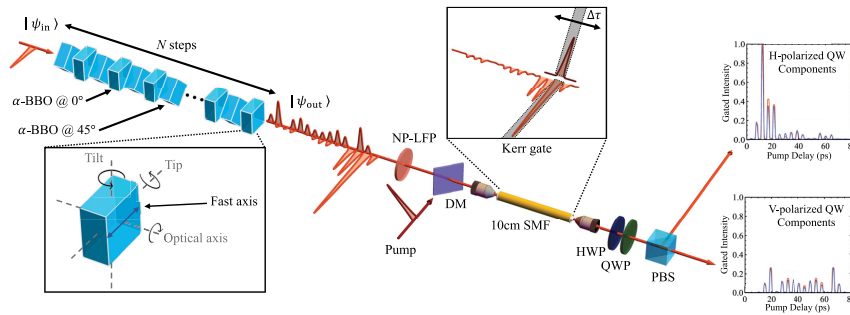


Fig. 1. Conceptual experimental setup. Simplified experimental setup of the ultrafast time-bin QW. The input state, $|\psi_{in}\rangle$, propagates through an N -step QW. Each step in the QW consists of a birefringent α -BBO crystal, which has three rotational degrees of freedom (shown in the bottom inset). The output state, $|\psi_{out}\rangle$, is sampled by an optical Kerr gate, where a strong pump pulse induces polarization rotation only where it overlaps with $|\psi_{out}\rangle$ (shown in the top inset). Prior to recombination with the pump on a dichroic mirror (DM), the QW output state is sent through a nanoparticle linear film polarizer (NP-LFP), which can be rotated to measure either its H- or V-polarized components. Scanning the pump, $\Delta\tau$, allows for readout of the entire QW output state. Plots on the right show experimental data (blue) collected for the H- and V-polarized QW components, compared with model (red). PBS, polarizing beam-splitter, HWP, half-waveplate, QWP, quarter-waveplate, SMF, single mode fiber. Further experimental details are provided in Supplement 1, Section 1.

$$|\psi_{\text{out}}^{(N)}\rangle = \prod_{n=1}^N \hat{U}_n |\psi_{\text{in}}\rangle. \quad (4)$$

In general, the single photon input state can be written as

$$|\psi_{\text{in}}\rangle = \sum_m (\alpha_m |H\rangle + \beta_m |V\rangle) \otimes a_m |t_m\rangle, \quad (5)$$

where α_m , β_m , and a_m are complex numbers with $\sum_m (|\alpha_m|^2 + |\beta_m|^2) |a_m|^2 = 1$. Control over the input state is also necessary in order to program the QW for quantum simulations. The input state can be prepared in different polarization states, using QWPs and HWPs, and across multiple time bins using an appropriate sequence of α -BBO crystals.

The result of the QW is obtained by measuring the probability distribution of $|\psi_{\text{out}}^{(N)}\rangle$ for each polarization component. As these time bins occur on ultrafast timescales, an all-optical Kerr gate is necessary to measure the output state. The Kerr gate relies on cross-phase modulation via the optical Kerr effect, a third-order ($\chi^{(3)}$) optical nonlinearity where a strong pulse of light generates a localized intensity-dependent refractive index modulation in the medium through which it propagates. By doing so, the strong

pulse induces a birefringence that can rotate the polarization of a secondary optical signal. In this work, the Kerr gate medium is a short (10 cm) piece of single-mode fiber (SMF). Depicted in Fig. 1, when crossed-polarizers are placed along the signal beam path before and after the SMF, the signal that is temporally overlapped with the pump will undergo polarization rotation. The entire QW output state is measured by scanning pump delay, $\Delta\tau$, and detecting Kerr-gated intensity. We achieve a Kerr-gate efficiency of $> 99\%$, for which a mathematical description is provided in Supplement 1, Section 2. An overall system efficiency is outlined in Supplement 1, Section 3.

3. RESULTS

Changing the input state of the QW changes its evolution. We thus examine the step-by-step evolution of the QW for various input states, shown in Fig. 2, in order to demonstrate the robustness of our protocol. A PBS, HWP, and QWP are used to generate different input polarization states. Input states with more than one time bin can be prepared using the α -BBO crystals from the first steps of the QW. For the results presented here, single time-bin input state

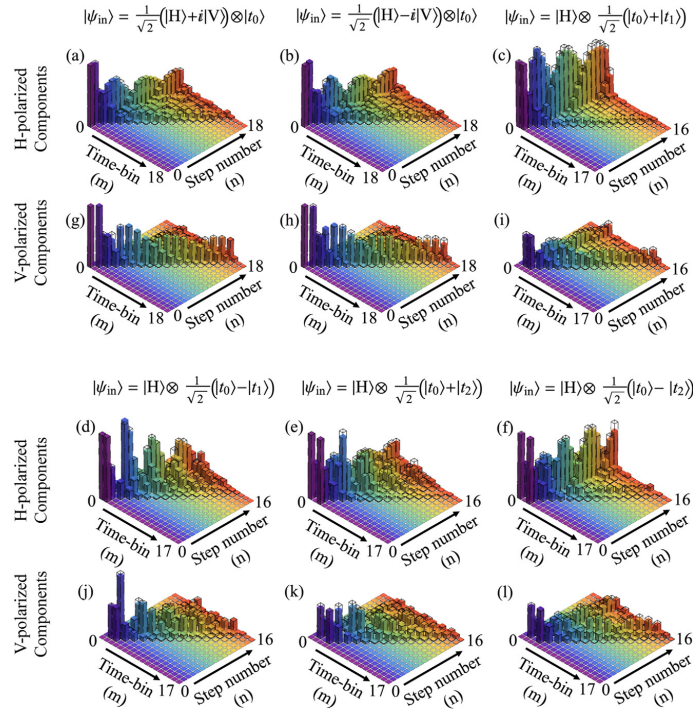


Fig. 2. Discretized step-by-step evolution of the QW for different input states. The H-polarized and V-polarized components for six different input states (defined above each dataset) are shown. The top rows (a)–(f) show the H-polarized components of the output state. The bottom rows (g)–(l) show the V-polarized components of the output state. The color indicates the step-by-step evolution (purple to red) of the experimental QW data, which is compared with the model (black outlines). Here, we can observe the emerging two-peaked distribution expected from quantum interference in the QW [3]. When the input state is polarized along one of the α -BBO crystal axes, such as $|\psi_{\text{in}}\rangle = |H\rangle \otimes 1/\sqrt{2}(|t_0\rangle + |t_1\rangle)$ or $|\psi_{\text{in}}\rangle = |H\rangle \otimes 1/\sqrt{2}(|t_0\rangle - |t_2\rangle)$, we observe an asymmetry in the two-peaked distribution. When the input state is in a superposition of these two polarizations, such as $|\psi_{\text{in}}\rangle = 1/\sqrt{2}(|H\rangle \pm i|V\rangle) \otimes |t_0\rangle$, the two-peaked output distribution will be symmetric. For the input states $|\psi_{\text{in}}\rangle = |H\rangle \otimes 1/\sqrt{2}(|t_0\rangle - |t_1\rangle)$ and $|\psi_{\text{in}}\rangle = |V\rangle \otimes 1/\sqrt{2}(|t_0\rangle + |t_2\rangle)$, we do not observe as strong a splitting of the distribution due to the phase of the input state. Details on data collection and processing are provided in Supplement 1, Section 4.

evolution is shown up to 18 steps. For the two time-bin input state evolution, the first two α -BBO crystals in the QW are removed and repurposed as needed for input state preparation, so a 16-step QW is performed.

The measured evolution compares well to theoretical evolution, which is indicated by the transparent black outlines. The agreement between experiment and theory is quantified by a fidelity calculation,

$$F_n(X, Y) = \left(\sum_{i=0}^n \sum_{j=1}^2 \sqrt{p_{i,j} q_{i,j}} \right)^2, \quad (6)$$

where $F_n(X, Y)$ describes the overlap between the experimental state, X , and theoretical state, Y , for a given number of steps, n , in the QW. The index i sums over the time bins and runs from the input time bin, $i = 0$, to the total number of time bins present after n steps. The index j sums over the two polarization components (H and V). The probabilities of the experimental and theoretical QW outputs of being in a given polarization and time bin are denoted by $p_{i,j}$ and $q_{i,j}$, respectively. The fidelity calculated for the six different input states is shown in Fig. 3. The experimental data are in good agreement with the theory, with fidelities remaining above 95% for all input states.

A key feature of the QW is that its probability distribution spreads considerably faster than that of a classical random walk. This can lead to exponentially faster hitting times when traversing a graph [9], an extremely useful attribute for quantum search algorithms. The standard deviation or variance of the QW output can be calculated to show this feature [23]. We calculate the step-by-step variance of our QW, shown in Fig. 4, for the different input states investigated in this work. We observe that the QW variance grows exponentially with the step number, as opposed to the linear growth in variance expected for a classical random walk.

4. DISCUSSION

Not only does our QW protocol demonstrate high fidelity for a large number of steps, it is also capable of preserving the interferometric phase stability for prolonged periods. We verified the stability over a 50 h period (see Supplement 1, Section 5). With this high stability, and a low loss of < -0.045 dB per step, our QW platform is expected to be easily scalable to a larger number of steps. To visualize where our platform fits into the current QW landscape with these attributes, it is compared to a sample of other platforms reported in the literature, on the basis of number of steps achieved, loss per step, and overall fidelity in Fig. 5. The sample of references included does not constitute an exhaustive list, but does provide an overall picture of the current experimental capabilities of discrete photonic QWs.

The combined robustness, programmability, and stability of our QW platform make it a particularly promising solution for scalability to a larger number of output modes while remaining space-efficient. These attributes are crucial for realizing QWs in regimes where classical verification is not possible or debilitatingly slow, for example efficiently simulating Hamiltonian evolution [64,65]. QWs have already been demonstrated for simulation purposes such as in topological investigations [24,26,27,52]. In these cases, fewer steps than we report here were required. It is anticipated that our QW could be extended into even more complex regimes in a variety of ways, depending on the problem at

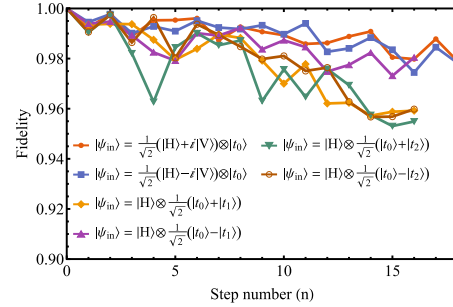


Fig. 3. Step-by-step fidelity of the QW for different input states. Calculated fidelity after each step for the six different input states measured. Fidelities remain $> 97\%$ and $> 95\%$ over the entire evolution of each 18-step QW and 16-step QW, respectively.

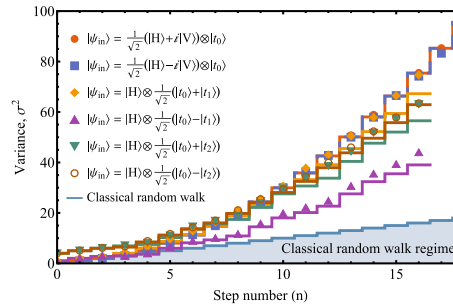


Fig. 4. Step-by-step variance of the QW. Experimental QW variances (data points) are compared with their respective theoretical variances (step-like lines) for various input states. The variance of a classical RW (for the case of a single classical particle that begins the walk in a single “time-bin”) is shaded at the bottom (light blue) for reference. For some input states, experimental values are higher than theoretical values due to background noise across all time bins in the measurement.

hand. Increasing the number of steps in the QW, thereby allowing access to more modes, is one option. This can be done by simply adding more α -BBO crystals to the QW signal beam path. With only 31 steps it is expected that the QW could be used in simulating parton showers [66], cascades of radiation produced in high-energy particle collisions. Another feasible near-term option requires 40 linear QW steps for the simulation of symmetries, topological phases, and bound states [67]. QW platforms can also borrow inspiration from experiments which use two-dimensional arrays of quantum particles for quantum simulation, such as Rydberg atom arrays [16,68] or trapped ion arrays [17,18,69]. Extending the space of the QW to two dimensions [34,70,71] can be used to simulate quantum systems with a natural mapping to two dimensions, such as in topological systems [72]. Beyond this, some QW schemes have even been demonstrated up to three- [73] and four-dimensions [36,74]. It would be straightforward to increase the dimensionality of the ultrafast time-bin QW platform, where multiple time-bin bases could be achieved using birefringent crystals of different thicknesses.

Since our QW platform is programmable at each step, it is realistic to consider the implications of implementing a time-dependent coin operator, where the coin depends on the temporal

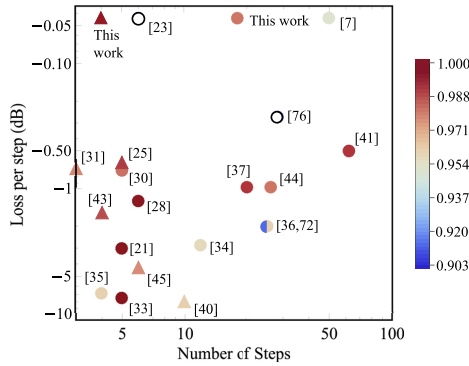


Fig. 5. Current QW Landscape. The loss per step is plotted against the number of steps for the experimental discrete photonic QWs that are cited. The color of each point corresponds to the reported fidelity, a scale for which is shown to the right of the plot. Circles and triangles correspond to QWs with one-photon and two-photon inputs, respectively. For references in which fidelity was not reported, the data points are white with black outlines. Note that this plot does not constitute an exhaustive list of all QW studies in the literature, and works that did not report on all three attributes (loss per step, number of steps, and fidelity) were not included. See Table S1 in [Supplement 1](#) for a more comprehensive list. Here, we include our 18-step, one-photon QW with 98% fidelity and our 4-step, two-photon QW with > 99% fidelity. Although the QW reported in [7], also based on UTBE, has a low loss per step and high number of steps, this implementation is ultimately limited by its detection efficiency, as elaborated on in [Supplement 1](#), Section 6.

variable [75,76]. Time-dependent coin operators have been used to demonstrate periodic revivals of a QW distribution [63] and can demonstrate emergence of a Parrondo paradox in QWs without resorting to intricate high-dimensional coins [53,77]. A time-dependent coin could be implemented in our ultrafast time-bin QW by adding a Kerr gate in the QW itself. Step-dependent coin operation could also be achieved, without active Kerr gates, by simply rotating and tilting the α -BBO crystals. These capabilities would allow for walking along any arbitrary graph, thereby extending the QW protocol for use in quantum search algorithms [9–11,78]. One could even envision implementing machine learning to program the QW [79,80], or using the QW for machine learning [14].

Although increasing the number of steps or dimensionality of the QW may be adequate in some cases, multi-photon input states are required if one is to simulate more complex, multi-particle quantum dynamics. In order to outline the promise of our platform in handling multi-photon input states, we performed a two-photon QW up to four steps. In this case, the attenuated coherent state (V-polarized) used for the one-photon QW was combined with a heralded single photon (H-polarized), after which both sources entered the QW. Results for the two-photon QW are presented in Fig. 6 and maintain a fidelity of > 99%, calculated using Eq. (6), in comparison to a simulation performed using the open-source Python library from Xanadu, *Strawberry Fields* [81,82]. See [Supplement 1](#), Section 7 for more details on the experimental setup used for the two-photon QW demonstration and the simulation.

Two-photon QWs have previously been demonstrated in various platforms [42,83–85] and have even been used to reveal

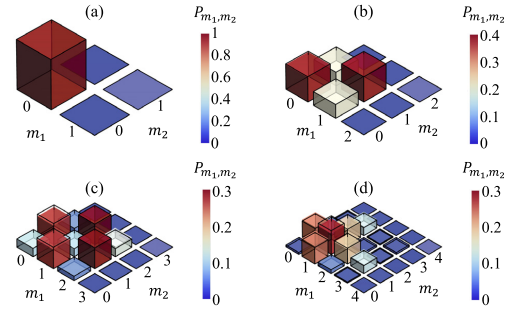


Fig. 6. Two-photon quantum walk. The H-polarized components for a two-photon QW are shown for (a) 1 step, (b) 2 steps, (c) 3 steps, and (d) 4 steps. Here, the attenuated coherent state is combined with a heralded single photon. The probability, P_{m_1, m_2} , of measuring a coincidence between time bin m_1 and time bin m_2 is shown for the different possible two-photon output states, where its values are indicated by the color bar. For $m_1 = m_2 = m$, P_{m_1, m_2} is the probability of measuring two photons in time bin m . For $m_1 \neq m_2$, P_{m_1, m_2} is the probability of measuring one photon in time bin m_1 and one photon in time bin m_2 , where we note that P_{m_1, m_2} and P_{m_2, m_1} are equivalent and refer to the same data. Experimental data (colored) are compared with a simulation (black outlines). For a more detailed description of the experimental setup and simulation, see [Supplement 1](#), Section 7.

previously unknown two-photon effects [86] and perform two-photon tomography [87]. Multi-photon entangled states are extremely sensitive to interferometric noise, as is demonstrated by the N00N state [88] and other highly entangled multi-photon states [89]. A crucial requirement in the scaling of QWs to multi-photon states is, therefore, maintaining interferometric stability across the entire QW over extended periods of time. The prolonged phase stability of our QW platform makes it a strong candidate for this task. Extra caution must be taken with multi-photon input states, since particle losses can drastically alter the output [40]; however, the high efficiency of our scheme makes it realistic to move to multi-photon input states. In order to measure n -fold coincidences, where n is the number of input photons, more Kerr gates would be required. It is possible to implement multiple Kerr-gates for such a task [90]; however, another solution could be to implement fast detectors, such as superconducting nanowire single-photon detectors (SNSPDs), which can achieve timing jitter as low as 3 ps [91]. In this case, thicker α -BBO crystals could be used to achieve ultrafast time-bins with larger temporal separation. It should be noted that measuring n -fold coincidences with SNSPDs is not trivial and would require multiplexing across $\sim n^2$ fast detectors. Furthermore, careful consideration of detector dead time would be required.

Our proposed UTBE scheme for discrete photonic QW demonstrates the key ingredients for scalability, including high interferometric phase stability for a large number of modes, programmability at each step, and low loss per step. With these combined capabilities we offer a pathway to scale programmable QWs with simple experimental tools, opening up new avenues for exploring problems that are intractable by classical computers.

Funding. Natural Sciences and Engineering Research Council of Canada, Vanier Canada Graduate Scholarships (Vanier CGS) Program.

Acknowledgment. We thank Aaron Goldberg, Andrew Proppe, Brayden Freitas, Denis Guay, Doug Moffatt, Kent Bonsma-Fisher, Rune Lausten, and Yingwen Zhang for their support and insightful discussions.

Disclosures. The authors declare no conflicts of interest.

Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

Supplemental document. See Supplement 1 for supporting content.

REFERENCES

1. N. Masuda, M. A. Porter, and R. Lambiotte, "Random walks and diffusion on networks," *Phys. Rep.* **716**, 1–58 (2017).
2. Y. Aharonov, L. Davidovich, and N. Zagury, "Quantum random walks," *Phys. Rev. A* **48**, 1687–1690 (1993).
3. J. Kempe, "Quantum random walks: an introductory overview," *Contemp. Phys.* **44**, 307–327 (2003).
4. F. W. Strauch, "Relativistic quantum walks," *Phys. Rev. A* **73**, 054302 (2006).
5. D. Witthaut, "Quantum walks and quantum simulations with Bloch-oscillating spinor atoms," *Phys. Rev. A* **82**, 033602 (2010).
6. C.-W. Lee, P. Kurzyński, and H. Nha, "Quantum walk as a simulator of nonlinear dynamics: nonlinear Dirac equation and solitons," *Phys. Rev. A* **92**, 052336 (2015).
7. X.-Y. Xu, Q.-Q. Wang, W.-W. Pan, *et al.*, "Measuring the winding number in a large-scale chiral quantum walk," *Phys. Rev. Lett.* **120**, 260501 (2018).
8. Q.-Q. Wang, X.-Y. Xu, W.-W. Pan, *et al.*, "Dynamic-disorder-induced enhancement of entanglement in photonic quantum walks," *Optica* **5**, 1136–1140 (2018).
9. N. Shenvi, J. Kempe, and K. B. Whaley, "Quantum random-walk search algorithm," *Phys. Rev. A* **67**, 052307 (2003).
10. A. M. Childs and J. Goldstone, "Spatial search by quantum walk," *Phys. Rev. A* **70**, 022314 (2004).
11. D. Qu, S. Marsh, K. Wang, *et al.*, "Deterministic search on star graphs via quantum walks," *Phys. Rev. Lett.* **128**, 050501 (2022).
12. O. Mülken and A. Blumen, "Continuous-time quantum walks: models for coherent transport on complex networks," *Phys. Rep.* **502**, 37–87 (2011).
13. A. M. Childs, "Universal computation by quantum walk," *Phys. Rev. Lett.* **102**, 180501 (2009).
14. F. Flamini, M. Krumm, L. J. Fiderer, *et al.*, "Reinforcement learning and decision making via single-photon quantum walks," *arXiv* (2023).
15. Q.-Q. Wang, S. Dong, X.-W. Li, *et al.*, "Efficient learning of mixed-state tomography for photonic quantum walk," *Sci. Adv.* **10**, ead4871 (2024).
16. M. Karski, L. Förster, J.-M. Choi, *et al.*, "Quantum walk in position space with single optically trapped atoms," *Science* **325**, 174–177 (2009).
17. H. Schmitz, R. Matjeschk, C. Schneider, *et al.*, "Quantum walk of a trapped ion in phase space," *Phys. Rev. Lett.* **103**, 090504 (2009).
18. F. Zähringer, G. Kirchmair, R. Gerritsma, *et al.*, "Realization of a quantum walk with one and two trapped ions," *Phys. Rev. Lett.* **104**, 100503 (2010).
19. C. A. Ryan, M. Laforest, J.-C. Boileau, *et al.*, "Experimental implementation of a discrete-time quantum random walk on an NMR quantum-information processor," *Phys. Rev. A* **72**, 062317 (2005).
20. M. Gong, S. Wang, C. Zha, *et al.*, "Quantum walks on a programmable two-dimensional 62-qubit superconducting processor," *Science* **372**, 948–952 (2021).
21. B. Do, M. L. Stohler, S. Balasubramanian, *et al.*, "Experimental realization of a quantum quincunx by use of linear optical elements," *J. Opt. Soc. Am. B* **22**, 499–504 (2005).
22. D. Bouwmeester, I. Marzoli, G. P. Karman, *et al.*, "Optical Galton board," *Phys. Rev. A* **61**, 013410 (1999).
23. M. A. Broome, A. Fedrizzi, B. P. Lanyon, *et al.*, "Discrete single-photon quantum walks with tunable decoherence," *Phys. Rev. Lett.* **104**, 153602 (2010).
24. T. Kitagawa, M. A. Broome, A. Fedrizzi, *et al.*, "Observation of topologically protected bound states in photonic quantum walks," *Nat. Commun.* **3**, 882 (2012).
25. F. Cardano, F. Massa, H. Qassim, *et al.*, "Quantum walks and wavepacket dynamics on a lattice with twisted photons," *Sci. Adv.* **1**, e1500087 (2015).
26. F. Cardano, M. Maffei, F. Massa, *et al.*, "Statistical moments of quantum-walk dynamics reveal topological quantum transitions," *Nat. Commun.* **7**, 11439 (2016).
27. F. Cardano, A. D'Errico, A. Dauphin, *et al.*, "Detection of Zak phases and topological invariants in a chiral quantum walk of twisted photons," *Nat. Commun.* **8**, 15516 (2017).
28. F. Nejdassattari, Y. Zhang, F. Bouchard, *et al.*, "Experimental realization of wave-packet dynamics in cyclic quantum walks," *Optica* **6**, 174–180 (2019).
29. A. Gerdali, A. Laneve, L. D. Bonavena, *et al.*, "Experimental investigation of superdiffusion via coherent disordered quantum walks," *Phys. Rev. Lett.* **123**, 140501 (2019).
30. A. D'Errico, F. Cardano, M. Maffei, *et al.*, "Two-dimensional topological quantum walks in the momentum space of structured light," *Optica* **7**, 108–114 (2020).
31. C. Esposito, M. R. Barros, A. D. Hernández, *et al.*, "Quantum walks of two correlated photons in a 2D synthetic lattice," *npj Quantum Inf.* **8**, 34 (2022).
32. F. Di Colandrea, A. Babazadeh, A. Dauphin, *et al.*, "Ultra-long quantum walks via spin-orbit photonics," *Optica* **10**, 324–331 (2023).
33. A. Schreiber, K. N. Cassemiro, V. Potoček, *et al.*, "Photons walking the line: a quantum walk with adjustable coin operations," *Phys. Rev. Lett.* **104**, 050502 (2010).
34. A. Schreiber, A. Gábris, P. P. Rohde, *et al.*, "A 2D quantum walk simulation of two-particle dynamics," *Science* **336**, 55–58 (2012).
35. Y.-C. Jeong, C. Di Franco, H.-T. Lim, *et al.*, "Experimental realization of a delayed-choice quantum walk," *Nat. Commun.* **4**, 2471 (2013).
36. L. Lorz, E. Meyer-Scott, T. Nitsche, *et al.*, "Photonic quantum walks with four-dimensional coins," *Phys. Rev. Res.* **1**, 033036 (2019).
37. A. Gerdali, S. De, A. Laneve, *et al.*, "Transient subdiffusion via disordered quantum walks," *Phys. Rev. Res.* **3**, 023052 (2021).
38. D. Bagrets, K. W. Kim, S. Barkhofen, *et al.*, "Probing the topological Anderson transition with quantum walks," *Phys. Rev. Res.* **3**, 023183 (2021).
39. P. Held, M. Engelkemeier, S. De, *et al.*, "Driven Gaussian quantum walks," *Phys. Rev. A* **105**, 042210 (2022).
40. F. Pegoraro, P. Held, S. Barkhofen, *et al.*, "Dynamic conditioning of two particle discrete-time quantum walks," *Phys. Scripta* **98**, 034005 (2023).
41. J. Boutari, A. Feizpour, S. Barz, *et al.*, "Large scale quantum walks by means of optical fiber cavities," *J. Opt.* **18**, 094007 (2016).
42. A. Peruzzo, M. Lobino, J. C. Matthews, *et al.*, "Quantum walks of correlated photons," *Science* **329**, 1500–1503 (2010).
43. L. Sansoni, F. Sciarrino, G. Vallone, *et al.*, "Two-particle bosonic-fermionic quantum walk via integrated photonics," *Phys. Rev. Lett.* **108**, 010502 (2012).
44. N. C. Harris, G. R. Steinbrecher, M. Prabhu, *et al.*, "Quantum transport simulations in a programmable nanophotonic processor," *Nat. Photonics* **11**, 447–452 (2017).
45. I. Pitsios, L. Bianchi, A. S. Rab, *et al.*, "Photonic simulation of entanglement growth and engineering after a spin chain quench," *Nat. Commun.* **8**, 1569 (2017).
46. T. Meany, M. Gräfe, R. Heilmann, *et al.*, "Laser written circuits for quantum photonics," *Laser Photon. Rev.* **9**, 363–384 (2015).
47. M. Gräfe, R. Heilmann, M. Lebugle, *et al.*, "Integrated photonic quantum walks," *J. Opt.* **18**, 103002 (2016).
48. S. De, V. Ansari, J. Sperling, *et al.*, "Measurement-based quantum walks on high-dimensional graphs," *arXiv* (2022).
49. J. Brendel, N. Gisin, W. Tittel, *et al.*, "Pulsed energy-time entangled twin-photon source for quantum communication," *Phys. Rev. Lett.* **82**, 2594 (1999).
50. M. Lubasch, A. A. Valido, J. J. Renema, *et al.*, "Tensor network states in time-bin quantum optics," *Phys. Rev. A* **97**, 062304 (2018).
51. X.-Y. Xu, Q.-Q. Wang, S.-J. Tao, *et al.*, "Experimental classification of quenched quantum walks by dynamical Chern number," *Phys. Rev. Res.* **1**, 033039 (2019).
52. X.-Y. Xu, Q.-Q. Wang, M. Heyl, *et al.*, "Measuring a dynamical topological order parameter in quantum walks," *Light Sci. Appl.* **9**, 7 (2020).
53. M. Jan, Q.-Q. Wang, X.-Y. Xu, *et al.*, "Experimental realization of Parrondo's paradox in 1D quantum walks," *Adv. Quantum Technol.* **3**, 1900127 (2020).

54. S.-J. Tao, Q.-Q. Wang, Z. Chen, *et al.*, "Experimental optimal generation of hybrid entangled states in photonic quantum walks," *Opt. Lett.* **46**, 1868–1871 (2021).
55. C. Kupchak, P. J. Bustard, K. Heshami, *et al.*, "Time-bin-to-polarization conversion of ultrafast photonic qubits," *Phys. Rev. A* **96**, 053812 (2017).
56. J. M. Donohue, M. Agnew, J. Lavoie, *et al.*, "Coherent ultrafast measurement of time-bin encoded photons," *Phys. Rev. Lett.* **111**, 153602 (2013).
57. F. Bouchard, D. England, P. J. Bustard, *et al.*, "Quantum communication with ultrafast time-bin qubits," *PRX Quantum* **3**, 010332 (2022).
58. D. England, F. Bouchard, K. Fenwick, *et al.*, "Perspectives on all-optical Kerr switching for quantum optical applications," *Appl. Phys. Lett.* **119**, 160501 (2021).
59. C. Kupchak, J. Erskine, D. England, *et al.*, "Terahertz-bandwidth switching of heralded single photons," *Opt. Lett.* **44**, 1427–1430 (2019).
60. F. Bouchard, D. England, P. J. Bustard, *et al.*, "Achieving ultimate noise tolerance in quantum communication," *Phys. Rev. Appl.* **15**, 024027 (2021).
61. K. L. Fenwick, D. G. England, P. J. Bustard, *et al.*, "Carving out configurable ultrafast pulses from a continuous wave source via the optical Kerr effect," *Opt. Express* **28**, 24845–24853 (2020).
62. M. Štefáňák, T. Kiss, and I. Jex, "Recurrence of biased quantum walks on a line," *New J. Phys.* **11**, 043027 (2009).
63. P. Xue, R. Zhang, H. Qin, *et al.*, "Experimental quantum-walk revival with a time-dependent coin," *Phys. Rev. Lett.* **114**, 140502 (2015).
64. G. H. Low and I. L. Chuang, "Optimal Hamiltonian simulation by quantum signal processing," *Phys. Rev. Lett.* **118**, 010501 (2017).
65. A. M. Childs, R. Cleve, E. Deotto, *et al.*, "Exponential algorithmic speedup by a quantum walk," in *Proceedings of the Thirty-fifth Annual ACM Symposium on Theory of Computing* (2003), pp. 59–68.
66. K. Bepari, S. Malik, M. Spannowsky, *et al.*, "Quantum walk approach to simulating parton showers," *Phys. Rev. D* **106**, 056002 (2022).
67. J. K. Asbóth, "Symmetries, topological phases, and bound states in the one-dimensional quantum walk," *Phys. Rev. B* **86**, 195414 (2012).
68. M. Kalinowski, R. Samajdar, R. G. Melko, *et al.*, "Bulk and boundary quantum phase transitions in a square Rydberg atom array," *Phys. Rev. B* **105**, 174417 (2022).
69. R. J. MacDonell, T. Navickas, T. F. Wohlers-Reichel, *et al.*, "Predicting molecular vibronic spectra using time-domain analog quantum simulation," *arXiv* (2022).
70. H. Tang, X.-F. Lin, Z. Feng, *et al.*, "Experimental two-dimensional quantum walk on a photonic chip," *Sci. Adv.* **4**, eaat3174 (2018).
71. A. Oliveira, R. Portugal, and R. Donangelo, "Decoherence in two-dimensional quantum walks," *Phys. Rev. A* **74**, 012312 (2006).
72. C. Chen, X. Ding, J. Qin, *et al.*, "Observation of topologically protected edge states in a photonic two-dimensional quantum walk," *Phys. Rev. Lett.* **121**, 100502 (2018).
73. M. Ehrhardt, R. Keil, L. J. Maczewsky, *et al.*, "Exploring complex graphs using three-dimensional quantum walks of correlated photons," *Sci. Adv.* **7**, eabc5266 (2021).
74. C. S. Hamilton, A. Gábris, I. Jex, *et al.*, "Quantum walk with a four-dimensional coin," *New J. Phys.* **13**, 013015 (2011).
75. M.-C. Banuls, C. Navarrete, A. Pérez, *et al.*, "Quantum walk with a time-dependent coin," *Phys. Rev. A* **73**, 062304 (2006).
76. A. Schreiber, K. Cassemiro, V. Potoček, *et al.*, "Decoherence and disorder in quantum walks: from ballistic spread to localization," *Phys. Rev. Lett.* **106**, 180403 (2011).
77. M. A. Pires and S. M. D. Queirós, "Parrondo's paradox in quantum walks with time-dependent coin operators," *Phys. Rev. E* **102**, 042124 (2020).
78. D. L. Feder, "Perfect quantum state transfer with spinor bosons on weighted graphs," *Phys. Rev. Lett.* **97**, 180502 (2006).
79. R. S. Mathew, R. O'Donnell, D. Pizzey, *et al.*, "The Raspberry Pi auto-aligner: machine learning for automated alignment of laser beams," *Rev. Sci. Instrum.* **92**, 015117 (2021).
80. K. M. Hinrichs and J. J. Piotrowski, "Neural networks for faster optical alignment," *Opt. Eng.* **59**, 074107 (2020).
81. N. Killoran, J. Izaac, N. Quesada, *et al.*, "Strawberry fields: a software platform for photonic quantum computing," *Quantum* **3**, 129 (2019).
82. T. R. Bromley, J. M. Arrazola, S. Jahangiri, *et al.*, "Applications of near-term photonic quantum computers: software and algorithms," *Quantum Sci. Technol.* **5**, 034010 (2020).
83. H. Defienne, M. Barbieri, I. A. Walmsley, *et al.*, "Two-photon quantum walk in a multimode fiber," *Sci. Adv.* **2**, e1501054 (2016).
84. J. O. Owens, M. A. Broome, D. N. Biggerstaff, *et al.*, "Two-photon quantum walks in an elliptical direct-write waveguide array," *New J. Phys.* **13**, 075003 (2011).
85. F. Klauck, M. Heinrich, and A. Szameit, "Photonic two-particle quantum walks in Su-Schrieffer-Heeger lattices," *Photon. Res.* **9**, A1–A7 (2021).
86. D. S. Simon, S. Osawa, and A. V. Sergienko, "Quantum-clustered two-photon walks," *Phys. Rev. A* **101**, 032118 (2020).
87. J. G. Titchener, A. S. Soltsev, and A. A. Sukhorukov, "Two-photon tomography using on-chip quantum walks," *Opt. Lett.* **41**, 4079–4082 (2016).
88. I. Afek, O. Ambar, and Y. Silberberg, "High-noon states by mixing quantum and classical light," *Science* **328**, 879–881 (2010).
89. X. Guo, C. R. Breum, J. Borregaard, *et al.*, "Distributed quantum sensing in a continuous-variable entangled network," *Nat. Phys.* **16**, 281–284 (2020).
90. F. Bouchard, K. Bonsma-Fisher, K. Heshami, *et al.*, "Measuring ultrafast time-bin qubits," *Phys. Rev. A* **107**, 022618 (2023).
91. B. Korzh, Q.-Y. Zhao, J. P. Allmaras, *et al.*, "Demonstration of sub-3 ps temporal resolution with a superconducting nanowire single-photon detector," *Nat. Photonics* **14**, 250–255 (2020).

Supplemental Document

OPTICA

Photonic quantum walk with ultrafast time-bin encoding: supplement

KATE L. FENWICK,^{1,2}  FRÉDÉRIC BOUCHARD,^{1,*} GUILLAUME S. THEKKADATH,¹  DUNCAN ENGLAND,¹  PHILIP J. BUSTARD,¹  Khabat HESHAMI,^{1,2} AND BENJAMIN SUSSMAN^{1,2}

¹National Research Council of Canada, 100 Sussex Drive, Ottawa, Ontario K1A 0R6, Canada

²Department of Physics, University of Ottawa, Advanced Research Complex, 25 Templeton Street, Ottawa, Ontario K1N 6N5, Canada

This supplement published with Optica Publishing Group on 18 July 2024 by The Authors under the terms of the [Creative Commons Attribution 4.0 License](https://creativecommons.org/licenses/by/4.0/) in the format provided by the authors and unedited. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

Supplement DOI: <https://doi.org/10.6084/m9.figshare.26206898>

Parent Article DOI: <https://doi.org/10.1364/OPTICA.510312>

Photonic quantum walk with ultrafast time-bin encoding: supplemental document

1. EXPERIMENTAL DETAILS

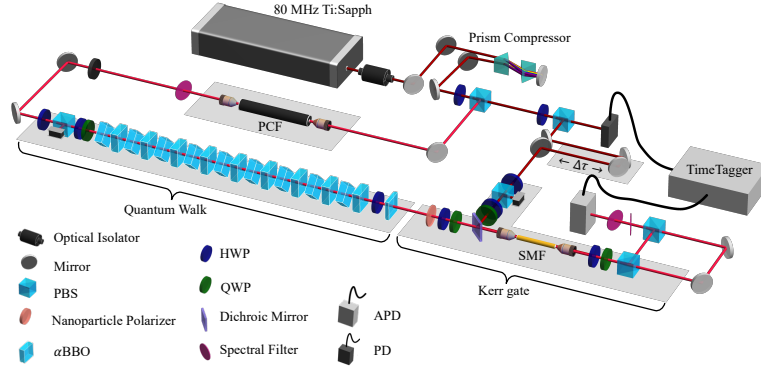


Fig. S1. Experimental Setup. Detailed experimental setup of the ultrafast time-bin QW. In the above configuration, all 18 α -BBO crystals are used in the QW (*i.e.*, none are used for input state generation). PBS: polarizing beamsplitter, HWP: half-waveplate, QWP: quarter-waveplate, APD: avalanche photodiode, PD: photodiode, SMF: single-mode fiber; PCF: photonic crystal fiber.

The full experimental setup in this work is shown in Fig. S1. Both the signal and pump originate from an 80 MHz titanium sapphire (Ti:Sapph) laser, centered at a wavelength of $\lambda_{\text{pump}} = 800$ nm with a spectral bandwidth of $\Delta\lambda_{\text{pump}} = 11$ nm. The output pulses from the Ti:Sapph laser are first sent through an optical isolator to avoid unwanted backreflected light re-entering the laser cavity. After the optical isolator, a prism compressor is used for dispersion compensation. The pulses are then split into two paths (one for the signal and one for the pump) by a HWP and a polarizing beamsplitter (PBS), which provide control over the pulse energy in the two paths.

Supercontinuum generation (SCG) in a 12 cm photonic crystal fiber (PCF) generates signal pulses [1]. When pumped with pulses from the Ti:sapph laser, the PCF (NKT Photonics, FemtoWHITE CARS) generates a dual peak spectrum which is tunable with input pulse energy. As the pump pulse energy is increased, the two SCG peaks will shift further away from the pump central wavelength and increase in bandwidth. In the sub-nJ pump pulse energy regime, SCG in the PCF provides access to a signal wavelength range of 600 to 1100 nm. Input pulses with energy ~ 0.25 nJ generate a spectrum with one of the dual peaks centered at 720 nm, which is spectrally-filtered to $\Delta\lambda_{\text{signal}} = 5$ nm using a band-pass filter. Neutral density (ND) filters are then used to attenuate the signal pulses to a mean photon number of $\mu \approx 0.8$ photons per pulse. The input photon polarization state is prepared using a PBS, QWP, and a HWP, after which it enters the quantum walk (QW). It should be noted that, for the configuration shown in Fig. S1, the signal pulses are prepared in a single time bin, $|\psi_{\text{in}}\rangle = (\alpha|H\rangle + \beta|V\rangle) \otimes |t_0\rangle$.

Input states with more than one time bin can be prepared using the α -BBO crystals from the first steps of the QW. Here, we prepare two time-bin states given by

$$|\psi_{\text{in}}\rangle = |H\rangle \otimes \frac{1}{\sqrt{2}}(|t_0\rangle + e^{i\nu}|t_k\rangle), \quad (\text{S1})$$

where the phase, ν , and the arrival of the t_k time bin are varied. The position, k , of the t_k time bin depends on the number of α -BBO crystals used to generate the input state, whereas the tip and tilt of the crystals sets ν . In the data presented here, $k = 1, 2$ and $\nu = 0, \pi$. The first two α -BBO crystals in the QW are removed and repurposed as needed for input state preparation, so a 16-step QW is performed. Note that for all input states, a PBS is added after the crystal(s) used for input state generation, to set the input polarization state to $|H\rangle$.

The α -BBO crystals comprising the QW are housed in kinematic rotation mounts, providing control over all three rotational degrees of freedom. By controlling each crystal's rotational degree of freedom, $\hat{C}_n$ can be set arbitrarily for each step of the QW. Alignment of the entire QW is done in a cascading manner, where each crystal is tuned relative to the previous one.

The QW output is recombined with the pump pulses on a dichroic mirror, so that the Kerr-gated measurement can be made. Prior to recombination, the pump is prepared as followed. The pump pulse energy is first tuned, using a HWP and PBS combination, to maximize the gating efficiency. A PBS, QWP, and HWP are used to prepare the pump polarization so that it is 45° relative to the signal polarization at the 10 cm SMF input to maximize gate efficiency. Finally the pump pulses are sent through a variable delay line, $\Delta\tau$, to delay the pump relative to the output of the QW. Scanning $\Delta\tau$ allows for temporal traces of the QW signal to be read-out by the Kerr gate at the picosecond timescale. The pump and signal are both coupled to the 10 cm SMF with 80% efficiency.

The QW signal is Kerr-gated, with $> 99\%$ internal efficiency, by the pump pulse within the 10 cm SMF (Thorlabs, S630-HP). After the Kerr gate, a QWP and HWP compensate for any intrinsic polarization rotation or birefringence within the 10 cm SMF. A PBS then splits the gated and non-gated QW signal along two different optical path lengths (separated on the nanosecond timescale). Spectral filters attenuate the pump and any noise generated by the pump inside the 10 cm SMF. The gated and non-gated signals are both detected by an avalanche photodiode (APD) and processed using a time-to-digital converter.

2. KERR GATE EFFICIENCY

The Kerr gate efficiency, η , is given by [2]

$$\eta(T) = \sin^2(2\theta)\sin^2\left(\frac{\Delta\phi(T)}{2}\right), \quad (\text{S2})$$

where θ is the angle between the signal and pump polarization, and $\Delta\phi(T)$ is the induced nonlinear birefringence in the SMF due to the presence of the pump pulse. A unit gating efficiency is achieved when $\theta = \pi/4$ and $\Delta\phi = \pi$. The nonlinear phase shift is given by [3]

$$\Delta\phi(T) = \frac{8\pi n_2}{3\lambda_s} \int_0^L I_p(T - d_w z) dz, \quad (\text{S3})$$

where n_2 is the nonlinear refractive index of the SMF, λ_s is the signal wavelength, and z is the propagation distance within the SMF of length L . As the SMF is a dispersive medium, the pump and signal will experience temporal walkoff, which is defined by $d_w = v_{gp}^{-1} - v_{gs}^{-1}$, where v_{gp} and v_{gs} are the group velocities of the pump and signal pulses, respectively. The intensity profile of the pump, I_p , is expressed in a frame that moves with the signal [4], $T = t - z/v_{gs}$, where t is the time in the laboratory frame.

3. OVERALL SYSTEM EFFICIENCY

The efficiency of the overall QW setup is defined by each optical element it comprises. Here, we quote the loss, in dB, for the different optical elements used. Silver mirrors: -0.088 dB; HWP: -0.223 dB; QWP: -0.223 dB; nanoparticle thin-film polarizer (NP-TFP): -0.044 dB; telescope: -0.094 dB; dichroic mirror: -0.044 dB; Kerr gate (includes aspheric lenses and 10 cm SMF): -1.192 dB; PBS: -0.706 dB; spectral filters: -1.871 dB; 1 m SMF to detector: -0.706 dB; APD: -2.218 dB. The majority of the α -BBO crystals used in the QW have an AR coating which leads to -0.044 dB loss (Newlight Photonics, ABT5100-A-P, a-cut); however, some crystals have an AR coating which leads to -0.269 dB loss (Newlight Photonics, ABT5100-A-AR750, a-cut). Combining the various experimental losses (including those of optical elements not depicted in Fig. S1), we find the efficiency of the walk to be $\sim 23\%$, and the detection efficiency (including Kerr gating, fiber coupling and APD efficiency) to be $\sim 21\%$. This returns an overall system efficiency of $\sim 5\%$.

4. DATA COLLECTION AND PROCESSING

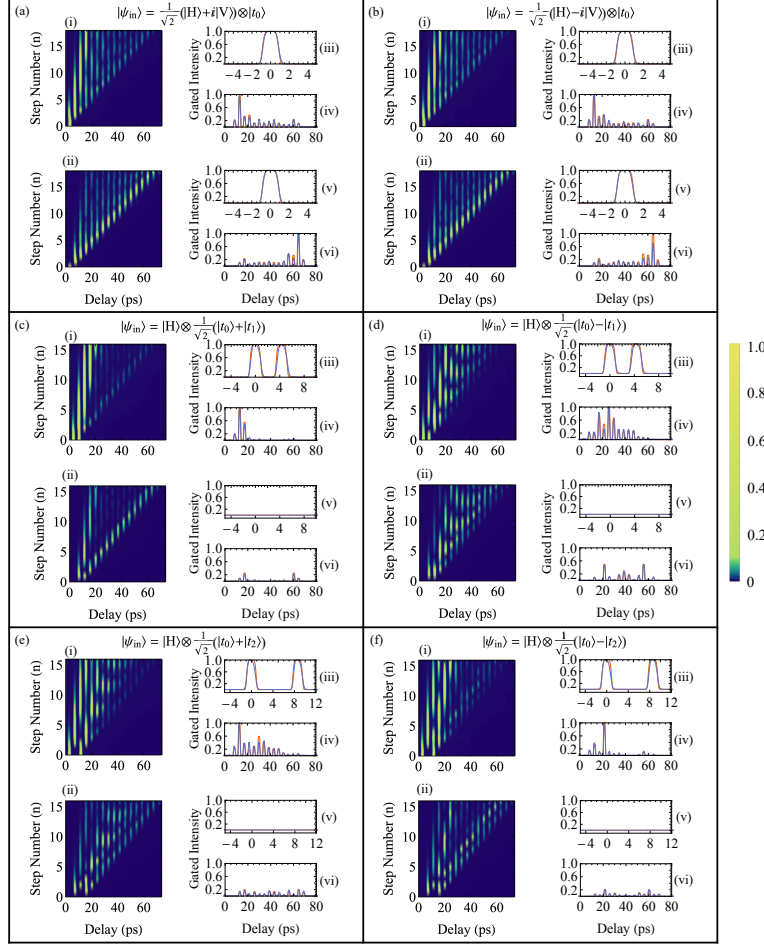


Fig. S2. Measured data for the UTBE QW for various input states. Boxes (a)-(f) correspond to various input states (indicated at the top of each box). In each box, the step-by-step evolution of the H- and V-polarized components of the QW are shown in (i) and (ii), respectively. A color scale for these is shown to the right of all the plots. Each box compares the experimental traces (blue) with the theoretical traces (red) for: the H-polarized input state in (iii), the H-polarized output state in (iv), the V-polarized input state in (v), and the V-polarized output state in (vi). Experimental traces agree well with theoretical traces.

The QW output state is sampled by the Kerr gate after each step is added. Fig. S2 shows the temporal traces of the six different input states (see subsets (iii) for H-polarized components and (v) for V-polarized components) and their corresponding outputs (see subsets (iv) for H-polarized components and (vi) for V-polarized components) after the 18- or 16-step QWs. The input and output experimental traces, shown in blue, compare well to the theory, shown in red. The experimental step-by-step evolution of the QW for each input state is also shown in Fig. S2 (see subsets (i) for H-polarized components and (ii) for V-polarized components). We note here

that the time bin, $|t_0\rangle$, occurs at a delay of 0 ps. After each step of the QW, one new time bin is added to the output state at a delay of $4.3n$ ps, where n is the step number. At the last step in the QW, the time bins span a range of 0 to $4.3N$ ps, where N is the total number of steps. This leads to a step-by-step evolution that expands along the delay axis as the QW evolves. The QW output traces are discretized by taking the peak value at each time-bin delay, $\Delta\tau = 4.3n$, where n is the step number.

5. INTERFEROMETRIC PHASE STABILITY

The advantage of using ultrafast time bins is highlighted in Fig. S3, where we see high stability for the 19-mode interferometric output associated with our 18-step QW over a 50 hour continuous measurement window.

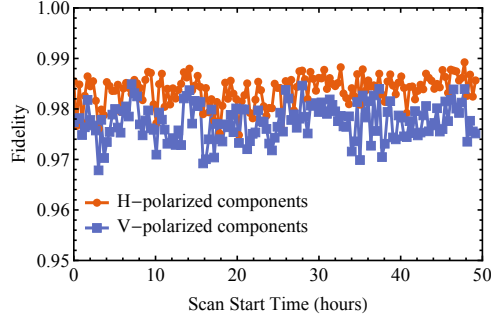


Fig. S3. Interferometric phase stability of UTBE QW. Calculated fidelity for the H- and V-polarized components of an 18-step QW with an input state of: $|\psi_{\text{in}}\rangle = |H\rangle \otimes |t_0\rangle$. Note that the overall fidelity combines both the H- and V-polarized components, but here we opt to show that they both have interferometric phase stability. The fidelity of the V-polarized components is slightly less than that of the H-polarized components, as there is less V-polarized light for this configuration. The data presented here was collected continuously over 50 hours to showcase the stability of the QW platform.

6. QW LANDSCAPE

A summary of experimental implementations of discrete-time photonic QWs is provided in Table S1. Although efforts were made to ensure a comprehensive list, it is not exhaustive. For studies presented in Table S1, experiments performed with an attenuated laser are considered to be using a 1 photon source. Note that two references quantify the quality of their results by the distance, $D = \frac{1}{2} \sum_i |p_i^{\text{exp}} - p_i^{\text{th}}|$, between the experimental, p_i^{exp} , and theoretical, p_i^{th} , probability distributions, rather than fidelity.

We observe that our QW platform is capable of achieving a larger number of steps than many other bulk optics implementations. Although the ultrafast time-bin encoding scheme presented in [5] can achieve up to 50 steps with a loss per calcite crystal (each QW step) of -0.044 dB, it implements an upconversion detection scheme which ultimately limits the total detection efficiency (upconversion process and measurement by a photomultiplier tube) to $\leq 0.5\%$. Such detection loss would make it difficult to perform QWs with more than one walker. Integrated photonics and fiber/optical loop schematics achieve a large number of steps, but they have a higher loss per step. Finally, we note two promising QW implementations, which differ from the others reported in Table S1. In [6], measurements on rotated bases implement the targeted unitary for a 400-step QW. In [7], 320-step QWs are simulated by a single large “step”: 3 liquid-crystal metasurfaces, encoded with the appropriate unitary. Such a scheme reports loss for a single liquid-crystal metasurface as -0.706 dB. This does not directly translate to a loss per step, because the liquid crystal could be programmed for many different sizes of QWs, with each limited by the same total loss; however, for the 320-step QWs reported, this corresponds to a very low “loss per step” of -0.007 dB. We do not include this metric in Table S1, as it is not directly comparable to the loss per step of the other discrete-time photonic QWs reported.

Year	Ref.	N_{steps}	Loss per step [dB]	Fidelity	N_{photons}	Platform
2005	[8]	5	-3.010	0.9984	1 photon	Bulk optics
2008	[9]	1	–	–	1 photon	Bulk optics
2010	[10]	5	-7.447	0.999	1 photon	Fiber loop
2010	[11]	6	-0.044	$D = 0.031$	1 photon	Bulk optics
2011	[12]	12	–	–	1 photon	Optical loop
2011	[13]	28	-0.269	$D = 0.052$	1 photon	Fiber loop
2012	[14]	7	–	–	1 photon	Bulk optics
2012	[15]	4	-1.600	0.988	2 photons	Integrated photonics
2012	[16]	12	-2.840	0.957	1 photon	Fiber network
2013	[17]	8	–	0.951	2 photons	Integrated photonics
2013	[18]	4	-6.840	0.96	1 photon	Optical loop
2014	[19]	16	–	0.918	1 photon	Integrated photonics
2015	[20]	5	-0.655	0.991	2 photons	Orbital angular momentum
2015	[21]	16	–	0.81	1 photon	Bulk optics
2016	[22]	62	-0.500	0.99	1 photon	Optical ring cavities
2016	[23]	6	–	–	1 photon	Orbital angular momentum
2017	[24]	7	–	–	1 photon	Orbital angular momentum
2017	[25]	6	-4.437	0.977	2 photons	Integrated photonics
2017	[26]	26	-0.969	0.98	1 photon	Integrated photonics
2018	[5]	50	-0.044	0.948	1 photon	Bulk optics (UTBE)
2018	[27]	25	-2.007	0.96	1 photon	Optical loop
2019	[28]	6	-1.249	0.9982	1 photon	Orbital angular momentum
2019	[29]	7	–	0.999	2 photons	Bulk optics
2019	[30]	25	-2.007	0.912	1 photon	Optical loop
2020	[31]	5	-0.706	0.982	1 photon	Transverse momentum
2021	[32]	20	-0.969	0.99	1 photon	Optical loop
2022	[33]	12	–	0.992	1 photon	Bulk optics
2022	[34]	3	-0.706	0.9773	2 photons	Transverse momentum
2022	[6]	400	–	0.98	1 photon	Measurement-based
2023	[7]	320	-0.007	0.77	1 photon	Liquid-crystal metasurfaces
2023	[35]	10	-8.239	0.961	2 photons	Optical loop
2023	This work	18	-0.044	0.98	1 photon	Bulk optics (UTBE)

Table S1. Current QW landscape. A sample of photonic discrete-time QWs to-date.

7. TWO-PHOTON QW

Two key changes are made to the experimental setup from Fig. S1 in order to demonstrate a two-photon QW. First, the input to the PCF is split into two paths, one of which continues to propagate through the PCF for supercontinuum generation as before, providing an attenuated coherent state. In the other, new path, spontaneous four-wave mixing (SFWM) in a 2.5 cm polarization-maintaining fiber (PMF) generates a photon pair. The PMF is pumped with pulses from the Ti:Sapph laser which are centered at 795 nm, spectrally-filtered to a bandwidth of 3 nm, and have an average power of 100 mW. This generates a photon pair at wavelengths of $\lambda_i = 953.5$ nm and $\lambda_s = 681.6$ nm for the idler and signal, respectively. The signal and idler photons are split on a dichroic mirror, and the idler photon is then spectrally-filtered and sent directly to an APD for detection. The signal photon is combined with the attenuated coherent state on a PBS, after which both are spectrally-filtered. Here, the attenuated coherent state is vertically polarized while the heralded signal photon is horizontally polarized. A delay line on the attenuated coherent state is adjusted such that it arrives at the same time as the heralded single photon at the input of the QW. For a mean photon number of $\mu \approx 0.24$ photons per pulse for the attenuated coherent state and a coincidence-to-accidentals ratio of 22 for the heralded photon, we observe a Hong-Ou-Mandel interference between the attenuated coherent state and heralded single photon of 70%. The two photons then propagate through up to four steps of the QW. In this case, only the horizontally-polarized components of the QW output were measured.

Second, another Kerr gate is added to the experimental setup after the first Kerr gate (in series). This is necessary to observe the two-photon output states in which the photons exit the walk in two different time bins. Specifically, the PBS after the Kerr gate depicted in Fig. S1 directs vertically-polarized light towards an APD and horizontally-polarized light towards an additional Kerr gate. After the additional Kerr gate, another PBS directs the horizontally- (H) and vertically- (V)-polarized light towards two separate APDs. The setup is operated in two different configurations, to measure the two-photon states where the photons are either in the same time bin or in different time bins.

For the two-photon output states where the photons are in the same time bin, the H-polarized input is rotated by 90° , so that all time bins in the first switch are now V-polarized. The first Kerr gate is turned “on” (*i.e.*, the strong pump pulse for Kerr gating is present) and the second is turned “off” (*i.e.*, the strong pump pulse for Kerr gating is not present). The first Kerr gate rotates the polarization of the single time bin of interest (from V to H), such that only this time bin will be allowed to transmit through the PBS and subsequently through the second Kerr gate SMF. After the second Kerr gate, a HWP and QWP rotate the polarization of the time bin of interest by 45° and a PBS splits the two photons between its H and V output ports, with 50% probability. Coincidences between the APDs at these two output ports and the heralding APD are measured, and multiplied by two to account for the 50% probability of this type of event occurring.

For the two-photon output states where the photons are in different time bins, both Kerr gates are turned “on”. In this case, the input to the first Kerr gate remains H-polarized. Each Kerr gate rotates the polarization of a different time bin (from H to V), and these time bins are directed to the APDs at the V-polarized output ports of the PBS immediately after their respective Kerr gate. Coincidences between the APDs at the two V-polarized output ports and the heralding APD are measured.

The experimental data from this two-photon QW is compared to a simulation which was performed using the open-source Python library from Xanadu, *Strawberry Fields* [36, 37]. The simulation propagates a heralded photon from a two-mode squeezed vacuum state and an attenuated coherent state through an interferometric network corresponding to our QW [38]. Here, we use the Gaussian numeric backend. Parameters such as the coherent state mean photon number, μ , squeezing parameter, r , system efficiency, and mode-matching between the attenuated coherent state and heralded photon can be adjusted. Here, we perform the simulation using parameters obtained from the characterization of our experimental setup: $\mu = 0.24$, $r = 0.12$, system efficiency = 0.05, and mode-matching = 70%. The fidelity between experimental measurements and the simulation is calculated using Eq. 6 in the manuscript.

REFERENCES

1. J. M. Dudley, G. Genty, and S. Coen, “Supercontinuum generation in photonic crystal fiber,” *Rev. Mod. Phys.* **78**, 1135 (2006).
2. H. Kanbara, H. Kobayashi, T. Kaino, *et al.*, “Highly efficient ultrafast optical kerr shutters

- with the use of organic nonlinear materials," *JOSA B* **11**, 2216–2223 (1994).
3. G. Agrawal, *Applications of nonlinear fiber optics* (Elsevier, 2001).
 4. C. Kupchak, J. Erskine, D. England, and B. Sussman, "Terahertz-bandwidth switching of heralded single photons," *Opt. letters* **44**, 1427–1430 (2019).
 5. X.-Y. Xu, Q.-Q. Wang, W.-W. Pan, *et al.*, "Measuring the winding number in a large-scale chiral quantum walk," *Phys. Rev. Lett.* **120**, 260501 (2018).
 6. S. De, V. Ansari, J. Sperling, *et al.*, "Measurement-based quantum walks on high-dimensional graphs," *arXiv preprint arXiv:2206.06059* (2022).
 7. F. Di Colandrea, A. Babazadeh, A. Dauphin, *et al.*, "Ultra-long quantum walks via spin-orbit photonics," *Optica* **10**, 324–331 (2023).
 8. B. Do, M. L. Stohler, S. Balasubramanian, *et al.*, "Experimental realization of a quantum quincunx by use of linear optical elements," *JOSA B* **22**, 499–504 (2005).
 9. P. S. Ribeiro, S. P. Walborn, C. Raitz Jr, *et al.*, "Quantum random walks and wave-packet reshaping at the single-photon level," *Phys. Rev. A* **78**, 012326 (2008).
 10. A. Schreiber, K. N. Cassemiro, V. Potoček, *et al.*, "Photons walking the line: a quantum walk with adjustable coin operations," *Phys. Rev. Lett.* **104**, 050502 (2010).
 11. M. A. Broome, A. Fedrizzi, B. P. Lanyon, *et al.*, "Discrete single-photon quantum walks with tunable decoherence," *Phys. Rev. Lett.* **104**, 153602 (2010).
 12. C. S. Hamilton, A. Gábris, I. Jex, and S. M. Barnett, "Quantum walk with a four-dimensional coin," *New J. Phys.* **13**, 013015 (2011).
 13. A. Schreiber, K. Cassemiro, V. Potoček, *et al.*, "Decoherence and disorder in quantum walks: from ballistic spread to localization," *Phys. Rev. Lett.* **106**, 180403 (2011).
 14. T. Kitagawa, M. A. Broome, A. Fedrizzi, *et al.*, "Observation of topologically protected bound states in photonic quantum walks," *Nat. Commun.* **3**, 1–7 (2012).
 15. L. Sansoni, F. Sciarrino, G. Vallone, *et al.*, "Two-particle bosonic-fermionic quantum walk via integrated photonics," *Phys. Rev. Lett.* **108**, 010502 (2012).
 16. A. Schreiber, A. Gábris, P. P. Rohde, *et al.*, "A 2d quantum walk simulation of two-particle dynamics," *Science* **336**, 55–58 (2012).
 17. A. Crespi, R. Osellame, R. Ramponi, *et al.*, "Anderson localization of entangled photons in an integrated quantum walk," *Nat. Photonics* **7**, 322–328 (2013).
 18. Y.-C. Jeong, C. Di Franco, H.-T. Lim, *et al.*, "Experimental realization of a delayed-choice quantum walk," *Nat. Commun.* **4**, 1–7 (2013).
 19. M. Gräfe, R. Heilmann, A. Perez-Leija, *et al.*, "On-chip generation of high-order single-photon w-states," *Nat. Photonics* **8**, 791–795 (2014).
 20. F. Cardano, F. Massa, H. Qassim, *et al.*, "Quantum walks and wavepacket dynamics on a lattice with twisted photons," *Sci. Adv.* **1**, e1500087 (2015).
 21. P. Xue, R. Zhang, H. Qin, *et al.*, "Experimental quantum-walk revival with a time-dependent coin," *Phys. Rev. Lett.* **114**, 140502 (2015).
 22. J. Boutari, A. Feizpour, S. Barz, *et al.*, "Large scale quantum walks by means of optical fiber cavities," *J. Opt.* **18**, 094007 (2016).
 23. F. Cardano, M. Maffei, F. Massa, *et al.*, "Statistical moments of quantum-walk dynamics reveal topological quantum transitions," *Nat. Commun.* **7**, 11439 (2016).
 24. F. Cardano, A. D'Errico, A. Dauphin, *et al.*, "Detection of zak phases and topological invariants in a chiral quantum walk of twisted photons," *Nat. Commun.* **8**, 15516 (2017).
 25. I. Pitsios, L. Banchi, A. S. Rab, *et al.*, "Photonic simulation of entanglement growth and engineering after a spin chain quench," *Nat. Commun.* **8**, 1569 (2017).
 26. N. C. Harris, G. R. Steinbrecher, M. Prabhu, *et al.*, "Quantum transport simulations in a programmable nanophotonic processor," *Nat. Photonics* **11**, 447–452 (2017).
 27. C. Chen, X. Ding, J. Qin, *et al.*, "Observation of topologically protected edge states in a photonic two-dimensional quantum walk," *Phys. Rev. Lett.* **121**, 100502 (2018).
 28. F. Nejdassattari, Y. Zhang, F. Bouchard, *et al.*, "Experimental realization of wave-packet dynamics in cyclic quantum walks," *Optica* **6**, 174–180 (2019).
 29. A. Gerdali, A. Laneve, L. D. Bonavena, *et al.*, "Experimental investigation of superdiffusion via coherent disordered quantum walks," *Phys. Rev. Lett.* **123**, 140501 (2019).
 30. L. Lorz, E. Meyer-Scott, T. Nitsche, *et al.*, "Photonic quantum walks with four-dimensional coins," *Phys. Rev. Res.* **1**, 033036 (2019).
 31. A. D'Errico, F. Cardano, M. Maffei, *et al.*, "Two-dimensional topological quantum walks in the momentum space of structured light," *Optica* **7**, 108–114 (2020).
 32. A. Gerdali, S. De, A. Laneve, *et al.*, "Transient subdiffusion via disordered quantum walks,"

- Phys. Rev. Res. **3**, 023052 (2021).
33. D. Qu, S. Marsh, K. Wang, *et al.*, “Deterministic search on star graphs via quantum walks,” Phys. Rev. Lett. **128**, 050501 (2022).
 34. C. Esposito, M. R. Barros, A. Durán Hernández, *et al.*, “Quantum walks of two correlated photons in a 2d synthetic lattice,” npj Quantum Inf. **8**, 34 (2022).
 35. F. Pegoraro, P. Held, S. Barkhofen, *et al.*, “Dynamic conditioning of two particle discrete-time quantum walks,” Phys. Scripta **98**, 034005 (2023).
 36. N. Killoran, J. Izaac, N. Quesada, *et al.*, “Strawberry fields: A software platform for photonic quantum computing,” Quantum **3**, 129 (2019).
 37. T. R. Bromley, J. M. Arrazola, S. Jahangiri, *et al.*, “Applications of near-term photonic quantum computers: software and algorithms,” Quantum Sci. Technol. **5**, 034010 (2020).
 38. G. Thekkadath, K. Fenwick, F. Bouchard, *et al.*, “Utbequantumwalk,” <https://github.com/tguillaume/UTBEQuantumWalk> (2024).

4.5 Implementing programmable circuits

This section is based on the following publication:

F. Bouchard, **K. Fenwick**, K. Bonsma-Fisher, D. England, P. J. Bustard, K. Hesami, and B. Sussman. “Programmable Photonic Quantum Circuits with Ultrafast Time-Bin Encoding”, *Physical Review Letters* **133**(9): 090601 (2024).

DOI: <https://doi.org/10.1103/PhysRevLett.133.090601>

© 2024 American Physical Society

The previous section demonstrated the remarkable stability, robustness, and low-loss of our UTBE scheme by implementing a large, multimode interferometer to perform the QW algorithm; however, this interferometer was not fully programmable across each mode at any given circuit depth. Here, we extend the programmability of our photonic quantum circuits based on UTBE, by adding optical Kerr gates within the circuit itself. This is a major step towards full programmability, which is crucial in the context of developing a full-scale universal quantum information processor based on UTBE.

In this implementation, we make use of the same birefringent α -BBO crystals to act as time-bin interferometers on ultrafast timescales. Each of these crystals is preceded by an optical Kerr gate, pumped by a sequence of control pulses. Combining these two physical elements allows us to achieve a sequence of POUTINEs, as described in Section 4.3.1. The programmability arises from the fact that the intensity of each individual control pulse can be tuned, subsequently allowing continuous adjustment of the optical Kerr gate efficiency at a given time bin. This provides full control over the coupling ratio between the polarization components within a given time bin. It should be noted that, for complete universality, two trains of control pulses per POUTINE, *i.e.*, $O(N^2)$ in total, would be required. Nevertheless, with a single train of control pulses per POUTINE, our circuit is capable of achieving a large subset of unitary transformations. Furthermore, only $O(N)$ optical components are required by our architecture of POUTINEs cascaded in a single beam line.

In this work, 362 different unitary transformations are programmed in up to eight dimensions in a temporal circuit to demonstrate the high degree of programmability

achievable by our UTBE scheme. Included in these unitary transformations is the four-dimensional Hadamard gate and an eight-dimensional permutation transformation. In addition to demonstrating the programmability potential of UTBE using optical Kerr gates and birefringent crystal, we also demonstrate the stability of our system over > 100 hours. Small fluctuations in fidelity are observed over time, which can be attributed to fluctuations in the control pulse energy in the optical Kerr gates. We reiterate here that this ultrastable interferometric phase arises from the fact that the phase of each arm in a given multiport time-bin interferometer (α -BBO crystal) is set by tilting the birefringent crystals, which can be kept fixed with high precision. Although this stability measurement was ended after 108 hours, from the trend, we expect the temporal circuit to remain phase stable for much longer.

The programmable nature of our UTBE scheme supports a range of photonic quantum computing approaches, from nonuniversal approaches like Gaussian boson sampling to full-fledged LOQC. We note here that our implementation does not really follow the KLM scheme devised in 2001 for LOQC; however, nowadays, measurement-based photonic quantum computing [206, 471] is anticipated to be the way forward for full-scale quantum information processing. Given this, it is important to develop large, multimode interferometers, *e.g.*, based on the universal design of Clements *et al.* [403]. Of course, achieving full-scale measurement-based quantum computing will require the generation of cluster states and incorporate feed-forward capabilities into the design.

This work is a major step towards photonic quantum information processing using UTBE. The photonic quantum circuit demonstrated here consists of a nontrivial number of modes which can be implemented without the need for complex fabrication processes; however, we note that our UTBE method is agnostic of the physical platform and could be implemented on-chip if desired. The implementation here is achieved using readily available experimental components, *e.g.*, ultrafast lasers, optical fibers, and birefringent crystals, which provides a versatile set of tools—available to many researchers in quantum optics—to design and program quantum circuits.

Programmable Photonic Quantum Circuits with Ultrafast Time-Bin EncodingFrédéric Bouchard^{1,*}, Kate Fenwick^{1,2}, Kent Bonsma-Fisher¹, Duncan England¹, Philip J. Bustard¹, Khabat Heshami^{1,2} and Benjamin Sussman^{1,2}¹*National Research Council of Canada, 100 Sussex Drive, Ottawa, Ontario K1A 0R6, Canada*²*Department of Physics, University of Ottawa, Advanced Research Complex, 25 Templeton Street, Ottawa, Ontario K1N 6N5, Canada*

(Received 17 November 2023; accepted 9 July 2024; published 26 August 2024)

We propose a quantum information processing platform that utilizes the ultrafast time-bin encoding of photons. This approach offers a pathway to scalability by leveraging the inherent phase stability of collinear temporal interferometric networks at the femtosecond-to-picosecond timescale. The proposed architecture encodes information in ultrafast temporal bins processed using optically induced nonlinearities and birefringent materials while keeping photons in a single spatial mode. We demonstrate the potential for scalable photonic quantum information processing through two independent experiments that showcase the platform's programmability and scalability, respectively. The scheme's programmability is demonstrated in the first experiment, where we successfully program 362 different unitary transformations in up to eight dimensions in a temporal circuit. In the second experiment, we show the scalability of ultrafast time-bin encoding by building a passive optical network, with increasing circuit depth, of up to 36 optical modes. In each experiment, fidelities exceed 97%, while the interferometric phase remains passively stable for several days.

DOI: [10.1103/PhysRevLett.133.090601](https://doi.org/10.1103/PhysRevLett.133.090601)

Coherence lies at the heart of the nonclassical quantum phenomena—entanglement and superposition—that underpin quantum technologies in computing, sensing, and communications. Decoherence, in which information leaks from a target system to its environment, is a persistent obstacle to the practical realization of these transformative capabilities. Realizing states that exhibit sustained quantum coherence necessitates a careful selection of both the physical system and encoding scheme. In comparison to many systems, photons exhibit a unique resistance to decoherence, maintaining their quantum state over extended periods of time and distances, even in ambient conditions. This robustness, coupled with their relative ease of control and detection, renders them exceptional candidates for foundational investigations of quantum mechanics [1,2] and quantum technology applications [3]. A particularly promising strategy lies in the development of photonic quantum systems that operate on ultrafast timescales, as short as femtoseconds. Such a rapid operational timeline ensures that quantum information processing tasks are completed before decoherence can significantly disrupt the system, thereby preserving the integrity of the quantum

information. However, maintaining coherence between different photonic states is nontrivial, owing to the challenges of maintaining phase stability between distinct paths. Here, we present a platform that makes use of ultrafast time-bin encoded photons, propagating on a collinear path, to perform quantum processing.

Generating and manipulating quantum states of light, from single photons to complex multiphoton states, remains a significant challenge. To harness the full potential of photonic platforms, photonic quantum processors must be capable of generating and controlling these states across a large number of optical modes. Recent advances in integrated programmable circuits have enabled universal programmability in up to 20 modes [4–9]. Although significant progress has been made in the development of fully programmable universal photonic quantum processors, scaling these technologies to accommodate and manipulate a larger number of modes remains an open challenge. Spatial encoding in integrated and bulk photonic platforms currently serve as the backbone of most quantum photonic architectures; however, another approach using time-bin encoding, or time multiplexing, is emerging with the potential for low loss and programmability [10–16]. One of the critical requirements in scaling quantum photonic platforms will be maintaining interferometric phase stability across the entirety of the quantum device and over extended periods of time. This challenge is particularly important when dealing with multiphoton states, given the heightened phase sensitivity of highly entangled states with large photon numbers. This is

Published by the American Physical Society under the terms of the *Creative Commons Attribution 4.0 International* license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

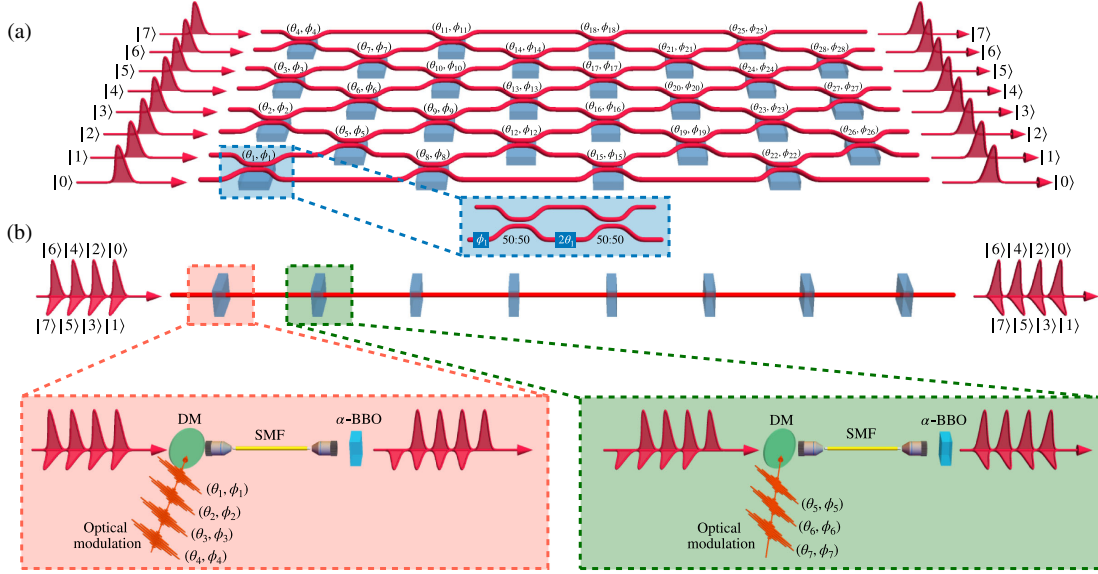


FIG. 1. Quantum circuits comparison. (a) Typically, an array of photonic quantum states is processed by a linear optical network in a waveguide configuration. The unitary transformation is programmed by tuning a sequence of beam splitters and phase shifters or by tuning two independent phase shifters in a balanced Mach-Zehnder interferometer. The output of the quantum circuit is then directed to the detection stage. (b) Here, an ultrafast pulse train of photonic quantum states is processed by a temporal linear optical network in a single spatial mode. The optical Kerr effect modulation, accomplished by overlapping bright control pulses with the quantum signals inside a single-mode fiber, applies phase shifts and couples modes. Birefringent α -BBO crystals act as time-bin interferometers. The output of the ultrafast temporal quantum circuit is then directed to the detection stage. DM, dichroic mirror; SMF, single-mode fiber.

famously demonstrated by NOON states [17] and other highly entangled multiphoton states [18]. In addition to phase stability, other key requirements for developing photonic quantum processors include high modulation speeds, high efficiencies, high fidelities, low noise levels, and seamless integration with the source and detectors. Our approach, as we will demonstrate, offers a promising path to meeting all of these requirements.

This work proposes and demonstrates an experimental platform to universal photonic quantum information processing through ultrafast time-bin encoding (UTBE). The platform's building blocks are a series of programmable optical ultrafast temporal interferometric network elements (POUTINES) that can be cascaded to form a fully connected temporal quantum circuit. Time-bin photonic states are prepared, and evolve, in a single beam. We program phase shifting and mode coupling operations by a combination of passive birefringent elements and active modulation through nonlinear optical Kerr effect interactions with intense ultrafast control pulses in optical fiber [19]. The fiber-based architecture of our platform offers compatibility with a wide range of quantum sources and new detector technologies, such as superconducting nanowire detectors and photon-number resolving detectors.

Perhaps the key attribute of our approach is the inherent phase stability of temporal modes at this timescale [20–22], a significant benefit when seeking to upscale processing to larger circuits. The ultrafast time-bin interferometers are extremely compact, robust, and can operate in a single spatial mode. The birefringent elements required to create interferometric pathways between distinct time bins provide excellent interferometric phase stability. This raises the possibility of upscaling critical quantum technologies such as quantum computing, quantum simulation, quantum communication, and quantum sensing. In what follows, we showcase the versatility and potential of our platform through two separate experiments that highlight its programmability, scalability, and stability.

Our goal is to build a universal linear optical network, described by an $N \times N$ unitary matrix, capable of transforming a set of N input time-bin modes into any arbitrary set of N output modes [23,24]. With integrated waveguides [7,25,26] or bulk optical devices [27,28], such a linear optical network consists of a large multiport interferometer with $O(N^2)$ points of interference that must be carefully tuned and stabilized; see Fig. 1(a). To retain the full programmability of our circuits, $O(N^2)$ control pulses are required. Nevertheless, the proposed ultrafast time-bin

architecture requires only $O(N)$ components, i.e., POUTINES, cascaded in a single beam line; see Fig. 1(b). The N -dimensional encoding space is defined using N creation operators, $\{\hat{a}_{H,t_0}^\dagger, \hat{a}_{V,t_0}^\dagger, \hat{a}_{H,t_1}^\dagger, \hat{a}_{V,t_1}^\dagger, \dots, \hat{a}_{H,t_{N/2}}^\dagger, \hat{a}_{V,t_{N/2}}^\dagger\}$, associated with horizontal (H) and vertical (V) polarization modes and ultrafast time bins, t_i , where $i \in 0, \dots, N/2$. In the computational basis, single-photon states are represented by $\{|\psi_i^N\rangle\} = \{(10)^T \otimes |t_0\rangle, (01)^T \otimes |t_0\rangle, \dots, (10)^T \otimes |t_{N/2}\rangle, (01)^T \otimes |t_{N/2}\rangle\}$, for even N , where $(10)^T$ and $(01)^T$ correspond to horizontally and vertically polarized states, respectively. The overall unitary transformation is achieved through a sequence of $N(N-1)/2$ programmable mode couplers, analogous to beam splitters and phase shifters, acting on adjacent modes in the network.

Our approach utilizes mode couplers that manipulate polarization at a single time bin, implemented via the optical Kerr effect, a third-order nonlinearity that induces local birefringence through cross-phase modulation with a bright pulse of light [29],

$$U_{\text{Kerr}}^{(k)}(\theta_m, \phi_m) = \sum_j \begin{pmatrix} e^{i\phi_{m(j,k)}} \cos \theta_{m(j,k)} & -i \sin \theta_{m(j,k)} \\ -ie^{i\phi_{m(j,k)}} \sin \theta_{m(j,k)} & \cos \theta_{m(j,k)} \end{pmatrix} \otimes |t_j\rangle\langle t_j|, \quad (1)$$

where θ_m and ϕ_m are the coupling and phase shifting parameters, respectively, $k \in \{0, \dots, N-1\}$, $j \in \{0, \dots, [N+(-1)^k-1]/2\}$, and $m \in \{1, \dots, N(N-1)/2\}$, for even N . For adjacent modes within a time bin, i.e., $\hat{a}_{H,t_j}^\dagger$ and $\hat{a}_{V,t_j}^\dagger$, this sequence of induced polarization rotations, $U_{\text{Kerr}}(\theta_m, \phi_m)$, directly achieves the required mode coupling. However, for adjacent modes belonging to different time bins, i.e., $\hat{a}_{V,t_j}^\dagger$ and $\hat{a}_{H,t_{j+1}}^\dagger$, a birefringent crystal is required to bring both modes back to a single time bin, i.e.,

$$U_{\text{Bir}} = \sum_{j=0}^{N/2} \left[\begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \otimes |t_j\rangle\langle t_j| + \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \otimes |t_{j+1}\rangle\langle t_j| \right]. \quad (2)$$

The birefringent crystal used in our scheme acts as a time-bin interferometer, similar to unbalanced Mach-Zehnder or fiber loop interferometers, where the path difference between both arms of the interferometer matches the time-bin separation. This is achieved by using birefringent crystals of the same length as those used in the generation of the ultrafast time-bin states. In order to bring adjacent modes within a time bin back together, we employ another transformation, namely a polarization rotation given by

$$R(\Theta) = \begin{pmatrix} \cos \Theta & -\sin \Theta \\ \sin \Theta & \cos \Theta \end{pmatrix} \otimes \sum_{j=0}^{N/2} |t_j\rangle\langle t_j|, \quad (3)$$

which can be achieved with wave plates or by rotating the subsequent birefringent crystal. A single POUTINE is thus given by

$$\mathcal{U}^{(k)}(\theta_m, \phi_m) = R(\pi/2) \cdot U_{\text{Bir}} \cdot U_{\text{Kerr}}^{(k)}(\theta_m, \phi_m). \quad (4)$$

Finally, the overall unitary transformation can then be constructed from a sequence of N POUTINES, i.e., $U^{(N)}(\theta_m, \phi_m) = \prod_{k=0}^{N-1} \mathcal{U}^{(k)}(\theta_m, \phi_m)$. The transformation in Eq. (1) can be achieved directly using two trains of control pulses, where the intensity of each pulse in a first train controls the phase parameters, ϕ_m , and the intensity of each pulse in a second diagonally polarized train of pulses determines the coupling parameters, θ_m . Nevertheless, for a large subset of transformations, the parameters θ_m and ϕ_m can be implemented with a single train of control pulses with their intensity and polarization carefully chosen; see Supplemental Material [30].

The use of picosecond time bins is a crucial advantage in our approach as it enables the implementation of highly compact time-bin interferometers with submillimeter path difference. This can be achieved by collinearly propagating two orthogonal linear polarization modes through a few millimeters of birefringent crystal. The compact size of these interferometers represents a significant improvement over alternative methods and reduces the experimental overhead. For instance, in previous demonstrations, time-bin interferometers have been realized with path differences reaching several kilometers, making the phase locking of such interferometers a significant achievement [15]. However, the use of ultrafast time bins in our platform eliminates this requirement altogether.

To assess the performance of our platform, we began by designing a simple temporal circuit with $N=4$ optical modes composed of a single optically induced mode coupler, as depicted in Fig. 2(a). Using a Ti:Sa laser to pump an optical parametric oscillator at a rate of 80 MHz, we generate single-photon-level signal pulses and introduce them into the temporal circuit. Here, we use α -BBO crystals with a thickness of 10 mm as our birefringent elements to create the appropriate ultrafast time bins with a corresponding temporal separation of 4.3 ps. Details about the generation and detection of ultrafast time-bin states can be found in the Supplemental Material [30]. Control pulses are then prepared from the pump laser to program the circuit, while taking care to select nonoverlapping wavelengths for the signal and control pulses, i.e., $\lambda_{\text{signal}} = 715$ nm and $\lambda_{\text{control}} = 790$ nm. The average efficiency of a single POUTINE is given by -1 dB of loss and is currently limited by the coupling efficiency to the single-mode fiber. The mode coupling parameter, θ , is controlled by varying the energy of the control pulse, allowing us to continuously tune the circuit from the identity transformation, $U_0^{N=4}$, to a SWAP-like gate, $U_4^{N=4}$, as shown in Fig. 2(b). To assess the quality and stability of the circuit, we measure the output

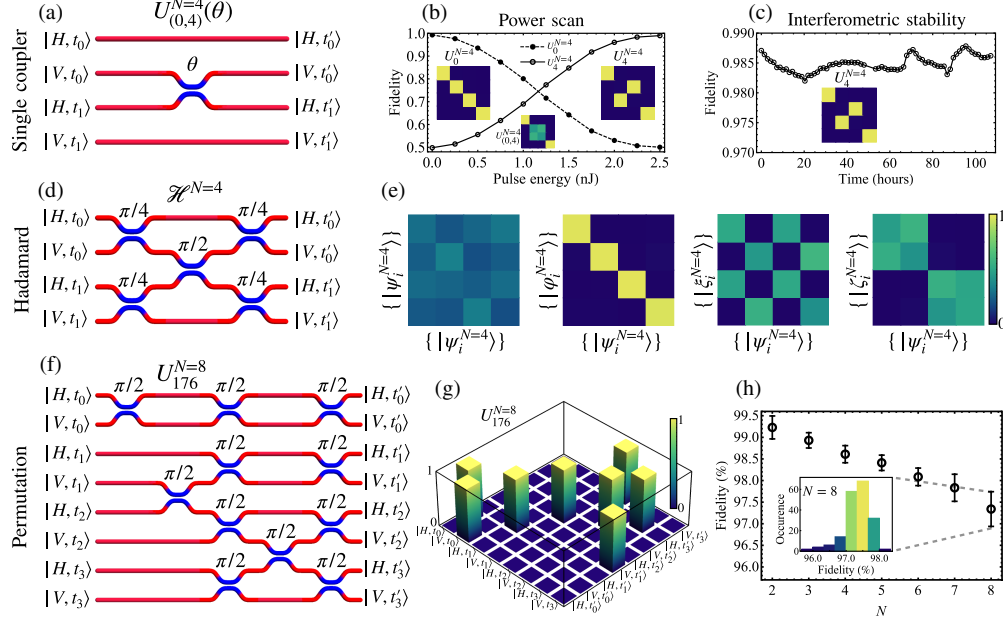


FIG. 2. Experimental demonstration of the programmable quantum temporal circuit. This figure presents a range of experimental results that showcase the programmability, robustness, and scalability of our UTBE platform for linear optical quantum computing. (a) Shows the circuit diagram of a four-dimensional temporal circuit with a mode coupler, where the coupling parameter θ controls the transformation from the identity $U_0^{N=4}$ to a SWAP-like gate $U_4^{N=4}$. This transformation is achieved with an overall system loss, including detector efficiency, of -5.2 dB. (b) Depicts the experimental measurement of the temporal circuit's evolution and fidelity with respect to θ , while (c) shows the long-term robustness of the SWAP-like gate without active stabilization or any other intervention. (d) Illustrates the four-dimensional Hadamard gate with the corresponding θ values, while (e) shows the measured probability of detection matrices for different input bases. This transformation is achieved with an overall system loss, including detector efficiency, of -7.2 dB. (f) Displays an example of an $N=8$ permutation transformation ($U_{176}^{N=8}$) with each mode coupler's coupling parameter. The experimentally measured detection probability matrix is shown in (g). This transformation is achieved with an overall system loss, including detector efficiency, of -7.2 dB. (h) Demonstrates the mean fidelity of all 362 measured permutation transformations from $N=2$ to $N=8$. For $N=8$, the fidelity distribution is shown explicitly in the inset.

number of counts distribution for each input mode and compute the fidelity, $F = (1/N)\text{Tr}(|U_{\text{target}}^\dagger \cdot |U_{\text{exp}}|)$, where U_{target} and U_{exp} are the target and experimentally measured probability of detection matrices, respectively. Although interferometric phase stability can be a challenge for linear optical networks, even for those as small as $N=4$, we found no degradation or significant drift over several days; see Fig. 2(c). The small variations in fidelity we observed were likely due to fluctuations in the control pulse energy. Our ultrastable interferometric phase arises from the fact that the phase of each arm of the multiport time-bin interferometer is determined by the tilting angle of the birefringent crystals, which can be kept fixed with high precision. We ended our stability measurements after 108 h, but expect the temporal circuit to remain phase stable for much longer.

Our exploration of temporal quantum circuits next progresses to a more complex network, the high-dimensional Hadamard gate. Although experimentally

challenging to implement, high-dimensional Hadamard gates are an important component of qudit-based quantum computing approaches [31–33]. To the best of our knowledge, this is the first experimental demonstration of a high-dimensional Hadamard gate implemented in the temporal degree of freedom of photons. The circuit diagram for the four-dimensional Hadamard gate, $\mathcal{H}^{N=4}$, is depicted in Fig. 2(d), which can be realized with two control pulses and half-wave plates acting uniformly across all time bins in the temporal network. We characterized the quality of the Hadamard circuit by measuring the probability of detection matrices for input quantum signals prepared in different bases, including the computational basis, $\{|\psi_i^{N=4}\rangle\} = \{|H, t_0\rangle, |V, t_0\rangle, |H, t_1\rangle, |V, t_1\rangle\}$, the four-dimensional discrete Fourier transform basis, $\{|\phi_i^{N=4}\rangle\} = \{(|H\rangle \pm |V\rangle) \otimes (|t_0\rangle \pm |t_1\rangle)/\sqrt{2}\}$, and other superposition bases, such as $\{|\xi_i^{N=4}\rangle\} = \{(|H\rangle \pm |V\rangle) \otimes |t_0\rangle/\sqrt{2}, (|H\rangle \pm |V\rangle) \otimes |t_1\rangle/\sqrt{2}\}$ and $\{|\zeta_i^{N=4}\rangle\} = \{|H\rangle \otimes (|t_0\rangle \pm |t_1\rangle)/\sqrt{2}, |V\rangle \otimes (|t_0\rangle \pm |t_1\rangle)/\sqrt{2}\}$.

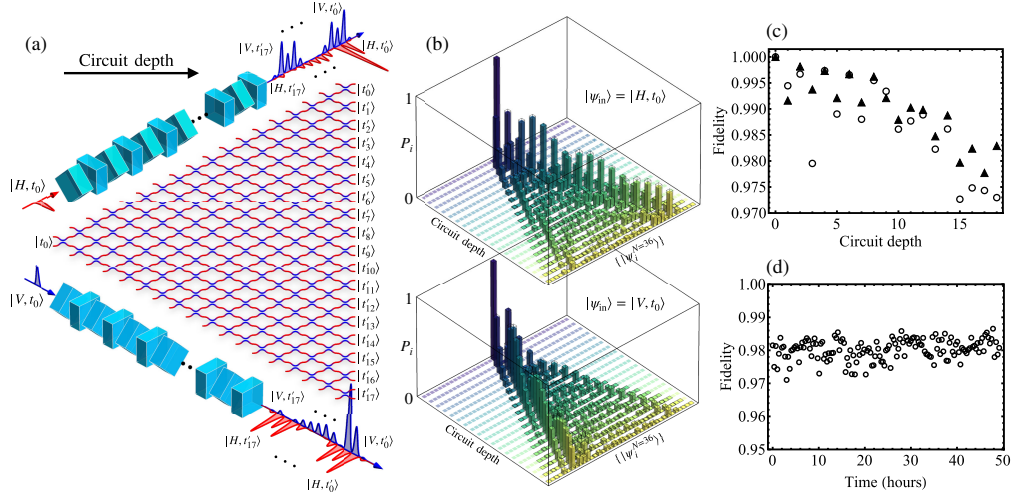


FIG. 3. Scalability demonstration of the programmable quantum temporal circuit. (a) Shows the theoretical output probability distribution of a fixed temporal circuit with 171 couplers and varied circuit depth for different input states, $|\psi_{\text{in}}\rangle = |H, t_0\rangle$ (top) and $|\psi_{\text{in}}\rangle = |V, t_0\rangle$ (bottom). (b) Displays the measured output state probability compared to the target state (black outline) for both input states as a function of circuit depth. (c) Shows the fidelity as a function of circuit depth for the two input states (triangles and circles corresponding to horizontal and vertical input polarization states, respectively). (d) Shows the long-term stability of the $N = 36$ temporal circuit with a circuit depth of 18, with no significant decrease in output state fidelity observed over a period of 50 h. This transformation is achieved with an overall system loss, including detector efficiency, of -13 dB.

To further demonstrate the versatility and controllability of our temporal quantum processor, we perform a set of high-dimensional permutation transformations, which are essential for quantum information processing. The number of distinct permutation transformations for a linear optical network with N modes is $N!$. In our experiments, we were able to implement and measure all 32 permutation matrices for N up to 4, while for larger dimensions, we sampled a subset of permutations due to their overwhelming number. In total, we have experimentally implemented and measured 362 permutation transformations for dimensions ranging from $N = 2$ to $N = 8$. As an illustration, Figs. 2(f) and 2(g), respectively, show the circuit diagram and the probability of detection matrix obtained experimentally for the $N = 8$ permutation transformation $U_{176}^{N=8}$. We achieved an average transformation fidelity above 97% for all measured permutation matrices for all values of N (Fig. 2(h)). The small decrease in fidelity can be attributed to an increase in total noise when considering a larger number of modes.

Having demonstrated the programmability of our platform by achieving high-dimensional unitary transformations in the temporal degree of freedom, we next investigate phase stability when scaling temporal circuits to a large number of modes, as illustrated in Fig. 3(a). For this purpose, we consider a fixed circuit with each coupler set to $\theta = \pi/4$ across all time bins, and measure the output distribution of the circuit when fed with a single time bin for increasing circuit depths, as shown in Fig. 3(b).

Remarkably, even for the largest circuit depth ($N = 36$) with a total of 171 mode couplers involved in the transformation, the same prolonged phase stability observed in the previous demonstrations ($N = 2, \dots, 8$) is still maintained over an extended period of time, i.e., over 50 h for $N = 36$, as seen in Fig. 3(c). Moreover, the transformation fidelity remains excellent even as we increase the circuit depth, as depicted in Fig. 3(d). Notably, since the 36-mode temporal interferometric setup occurs collinearly in a single spatial mode, the number of physical elements scales linearly with circuit depth, and only 18 birefringent crystals are required to construct this large-scale circuit.

Our results demonstrate the significant potential of the UTBE platform for advancing quantum technologies. Although our platform is based on polarization to create the temporal circuit, other degrees of freedom such as frequency bins or path encoding can be used in parallel with the ultrafast time bins. Frequency conversion of quantum signals based on third-order nonlinearities, such as cross-phase modulation [34] and four-wave mixing [35], have recently seen remarkable progress, enabling further development of frequency-bin encoding [36]. Compared to polarization, a hybrid time-bin and frequency-bin encoding can offer a much larger encoding space, extending to multiple dimensions with fewer experimental resources. In this scheme, time-bin interferometers can be realized by replacing the birefringent crystals with dispersion-engineered fibers, which provide greater control and programmability of ultrafast temporal quantum processors.

Furthermore, integrating the UTBE architecture into a waveguide architecture with ultrafast control could leverage past investments in integrated photonics and exploit key features of our ultrafast platform. Finally, recent advances in superconducting nanowire single-photon detectors, where a sub-3 ps temporal resolution has been achieved [37], enable efficient measurements of ultrafast time-bin encoded photons.

In conclusion, the proposed platform presents a promising scalability strategy that prioritizes phase stability as a critical feature to develop future implementations with larger mode and photon numbers. We have demonstrated that a programmable quantum circuit with a nontrivial number of modes can be implemented without the need for complex fabrication processes. Leveraging readily available experimental components in quantum optics labs, e.g., ultrafast lasers, optical fibers, and birefringent crystals, we have provided a versatile set of tools to design and program quantum circuits, enabling researchers to explore a wide range of quantum processing applications. We anticipate that our platform will be a fertile ground for exploring novel sources, implementations, detection schemes, and other applications, such as quantum neural networks [38], quantum metrology [39], and quantum networks [40]. UTBE is a powerful addition to current approaches as it offers a single underlying platform for generating non-classical light and conducting quantum processing. Notably, our platform has tremendous potential for enabling various applications in multiphoton entangled state generation. Moreover, the universal nature of our UTBE scheme supports a range of photonic quantum computing approaches, from nonuniversal approaches like Gaussian boson sampling to full-fledged linear optical quantum computing.

Acknowledgments—We thank Guillaume Thekkadath, Aaron Goldberg, Yingwen Zhang, Andrew Proppe, Rune Lausten, Denis Guay, and Doug Moffatt for support and insightful discussions.

-
- [1] L. K. Shalm, E. Meyer-Scott, B. G. Christensen, P. Bierhorst, M. A. Wayne, M. J. Stevens, T. Gerrits, S. Glancy, D. R. Hamel, M. S. Allman *et al.*, *Phys. Rev. Lett.* **115**, 250402 (2015).
 - [2] M. Giustina, M. A. M. Versteegh, S. Wengerowsky, J. Handsteiner, A. Hochrainer, K. Phelan, F. Steinlechner, J. Kofler, J.-Å. Larsson, C. Abellán *et al.*, *Phys. Rev. Lett.* **115**, 250401 (2015).
 - [3] E. Knill, R. Laflamme, and G. J. Milburn, *Nature (London)* **409**, 46 (2001).
 - [4] J. Carolan, C. Harrold, C. Sparrow, E. Martín-López, N. J. Russell, J. W. Silverstone, P. J. Shadbolt, N. Matsuda, M. Oguma, M. Itoh *et al.*, *Science* **349**, 711 (2015).
 - [5] N. C. Harris, G. R. Steinbrecher, M. Prabhu, Y. Lahini, J. Mower, D. Bunandar, C. Chen, F. N. Wong, T. Baehr-Jones, M. Hochberg *et al.*, *Nat. Photonics* **11**, 447 (2017).
 - [6] C. Taballione, T. A. Wolterink, J. Lugani, A. Eckstein, B. A. Bell, R. Grootjans, I. Visscher, D. Geskus, C. G. Roeloffzen, J. J. Renema *et al.*, *Opt. Express* **27**, 26842 (2019).
 - [7] C. Taballione, R. van der Meer, H. J. Snijders, P. Hooijschuur, J. P. Epping, M. de Goede, B. Kassenberg, P. Venderbosch, C. Toebes, H. van den Vlekert *et al.*, *Mater. Quantum Technol.* **1**, 035002 (2021).
 - [8] J. M. Arrazola, V. Bergholm, K. Brádler, T. R. Bromley, M. J. Collins, I. Dhand, A. Fumagalli, T. Gerrits, A. Goussev, L. G. Helt *et al.*, *Nature (London)* **591**, 54 (2021).
 - [9] C. Taballione, M. C. Anguita, M. de Goede, P. Venderbosch, B. Kassenberg, H. Snijders, D. Smith, J. P. Epping, R. van der Meer, P. W. Pinkse *et al.*, *Quantum* **7**, 1071 (2023).
 - [10] S. Yokoyama, R. Ukai, S. C. Armstrong, C. Sornphiphatphong, T. Kaji, S. Suzuki, J.-i. Yoshikawa, H. Yonezawa, N. C. Menicucci, and A. Furusawa, *Nat. Photonics* **7**, 982 (2013).
 - [11] P. C. Humphreys, B. J. Metcalf, J. B. Spring, M. Moore, X.-M. Jin, M. Barbieri, W. S. Kolthammer, and I. A. Walmsley, *Phys. Rev. Lett.* **111**, 150501 (2013).
 - [12] K. R. Motes, A. Gilchrist, J. P. Dowling, and P. P. Rohde, *Phys. Rev. Lett.* **113**, 120501 (2014).
 - [13] Y. He, X. Ding, Z.-E. Su, H.-L. Huang, J. Qin, C. Wang, S. Unsleber, C. Chen, H. Wang, Y.-M. He *et al.*, *Phys. Rev. Lett.* **118**, 190501 (2017).
 - [14] W. Asavanant, Y. Shiozawa, S. Yokoyama, B. Charoensombutamon, H. Emura, R. N. Alexander, S. Takeda, J.-i. Yoshikawa, N. C. Menicucci, H. Yonezawa *et al.*, *Science* **366**, 373 (2019).
 - [15] L. S. Madsen, F. Laudenbach, M. F. Askarani, F. Rortais, T. Vincent, J. F. Bulmer, F. M. Miatto, L. Neuhaus, L. G. Helt, M. J. Collins *et al.*, *Nature (London)* **606**, 75 (2022).
 - [16] S. Sempere-Llagostera, R. B. Patel, I. A. Walmsley, and W. S. Kolthammer, *Phys. Rev. X* **12**, 031045 (2022).
 - [17] I. Afek, O. Ambar, and Y. Silberberg, *Science* **328**, 879 (2010).
 - [18] X. Guo, C. R. Breum, J. Borregaard, S. Izumi, M. V. Larsen, T. Gehring, M. Christandl, J. S. Neergaard-Nielsen, and U. L. Andersen, *Nat. Phys.* **16**, 281 (2020).
 - [19] D. England, F. Bouchard, K. Fenwick, K. Bonsma-Fisher, Y. Zhang, P. J. Bustard, and B. J. Sussman, *Appl. Phys. Lett.* **119**, 160501 (2021).
 - [20] J. M. Donohue, M. Agnew, J. Lavoie, and K. J. Resch, *Phys. Rev. Lett.* **111**, 153602 (2013).
 - [21] F. Bouchard, D. England, P. J. Bustard, K. Heshami, and B. Sussman, *PRX Quantum* **3**, 010332 (2022).
 - [22] F. Bouchard, K. Bonsma-Fisher, K. Heshami, P. J. Bustard, D. England, and B. Sussman, *Phys. Rev. A* **107**, 022618 (2023).
 - [23] M. Reck, A. Zeilinger, H. J. Bernstein, and P. Bertani, *Phys. Rev. Lett.* **73**, 58 (1994).
 - [24] W. R. Clements, P. C. Humphreys, B. J. Metcalf, W. S. Kolthammer, and I. A. Walmsley, *Optica* **3**, 1460 (2016).
 - [25] J. B. Spring, B. J. Metcalf, P. C. Humphreys, W. S. Kolthammer, X.-M. Jin, M. Barbieri, A. Datta, N. Thomas-Peter, N. K. Langford, D. Kundys *et al.*, *Science* **339**, 798 (2013).

-
- [26] C. Antón, J. C. Loredó, G. Coppola, H. Ollivier, N. Viggianiello, A. Harouri, N. Somaschi, A. Crespi, I. Sagnes, A. Lemaitre *et al.*, *Optica* **6**, 1471 (2019).
- [27] H. Wang, J. Qin, X. Ding, M.-C. Chen, S. Chen, X. You, Y.-M. He, X. Jiang, L. You, Z. Wang *et al.*, *Phys. Rev. Lett.* **123**, 250503 (2019).
- [28] H.-S. Zhong, Y.-H. Deng, J. Qin, H. Wang, M.-C. Chen, L.-C. Peng, Y.-H. Luo, D. Wu, S.-Q. Gong, H. Su *et al.*, *Phys. Rev. Lett.* **127**, 180502 (2021).
- [29] C. Kupchak, J. Erskine, D. England, and B. Sussman, *Opt. Lett.* **44**, 1427 (2019).
- [30] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevLett.133.090601> for additional experimental details.
- [31] F. Brandt, M. Hiekkamäki, F. Bouchard, M. Huber, and R. Fickler, *Optica* **7**, 98 (2020).
- [32] Y. Chi, J. Huang, Z. Zhang, J. Mao, Z. Zhou, X. Chen, C. Zhai, J. Bao, T. Dai, H. Yuan *et al.*, *Nat. Commun.* **13**, 1166 (2022).
- [33] M. Ringbauer, M. Meth, L. Postler, R. Stricker, R. Blatt, P. Schindler, and T. Monz, *Nat. Phys.* **18**, 1053 (2022).
- [34] N. Matsuda, *Sci. Adv.* **2**, e1501223 (2016).
- [35] H. J. McGuinness, M. G. Raymer, C. J. McKinstrie, and S. Radic, *Phys. Rev. Lett.* **105**, 093604 (2010).
- [36] C. Joshi, A. Farsi, S. Clemmen, S. Ramelow, and A. L. Gaeta, *Nat. Commun.* **9**, 847 (2018).
- [37] B. Korzh, Q.-Y. Zhao, J. P. Allmaras, S. Frasca, T. M. Autry, E. A. Bersin, A. D. Beyer, R. M. Briggs, B. Bumble, M. Colangelo *et al.*, *Nat. Photonics* **14**, 250 (2020).
- [38] G. R. Steinbrecher, J. P. Olson, D. Englund, and J. Carolan, *npj Quantum Inf.* **5**, 60 (2019).
- [39] J. C. Matthews, X.-Q. Zhou, H. Cable, P. J. Shadbolt, D. J. Saunders, G. A. Durkin, G. J. Pryde, and J. L. O’Brien, *npj Quantum Inf.* **2**, 16023 (2016).
- [40] A. S. Cacciapuoti, M. Caleffi, F. Tafuri, F. S. Cataliotti, S. Gherardini, and G. Bianchi, *IEEE Network* **34**, 137 (2019).

Supplemental Material: Programmable Photonic Quantum Circuits with Ultrafast Time-bin Encoding

Frédéric Bouchard,^{1,*} Kate Fenwick,^{1,2} Kent Bonsma-Fisher,¹ Duncan England,¹ Philip J. Bustard,¹ Khabat Heshami,^{1,2} and Benjamin Sussman^{1,2}

¹*National Research Council of Canada, 100 Sussex Drive, Ottawa, Ontario K1A 0R6, Canada*

²*Department of Physics, University of Ottawa, Advanced Research Complex,
25 Templeton Street, Ottawa ON Canada, K1N 6N5*

The supplementary material presents additional experimental details and results.

I. METHODS

To thoroughly explore various aspects of our platform, we conducted two separate experiments, each focusing on different features of our scheme. Our experimental setup revolves around the utilization of ultrafast time-bin encoding for short sub-picosecond pulses, as depicted in Figure S1. In both experiments, we employed a Ti:sapphire (Ti:Sa) laser that generates pulses with a repetition rate of $f_{\text{rep}} = 80$ MHz. In the first experiment, we generated pulses with a center wavelength of $\lambda_{\text{control}} = 790$ nm. These pulses were split into two paths using a combination of a half-wave plate (HWP) and a polarizing beam splitter (PBS). In one path, an optical parametric oscillator (OPO) was employed to pump and generate frequency-doubled signal pulses with a center wavelength of $\lambda_{\text{signal}} = 715$ nm. Meanwhile, in the other path, the control pulses were prepared and directed into the temporal circuit for further processing.

For the second experiment, we generate pulses with a center wavelength of $\lambda_{\text{control}} = 800$ nm. Similar to the first experiment, these pulses were split into two paths. In one path, the pulses were coupled to a photonic crystal fiber, enabling the generation of signal pulses at a center wavelength of $\lambda_{\text{signal}} = 720$ nm through supercontinuum generation and spectral filtering. In the second path, the control pulses were sent to the output detection stage. It is worth noting that although the control and signal wavelengths in each experiment were close, they were not identical. This discrepancy arises due to the optimization process for generating signal pulses using different techniques such as optical parametric oscillation and supercontinuum generation. However, regardless of the small wavelength offset, we observed similar performance in both experiments. Moreover, we note that our platform is largely wavelength-agnostic and can be operated at wavelengths ranging from visible to telecom-band wavelengths.

A. Input state preparation

The signal pulses, denoted by $|\psi_{\text{in},0}\rangle = |H, t_0\rangle$, are directed to the state preparation stage. In the second experiment, input states consisted of a single time-bin state with varying polarization states, i.e.

$$|\psi_{\text{in}}\rangle = (\cos\theta|H\rangle + \sin\theta|V\rangle) \otimes |t_0\rangle. \quad (1)$$

On the other hand, in the first experiment, by employing a series of optical elements including a HWP, PBS, and α -BBO crystals, we enable the generation of a diverse range of 8-dimensional ultrafast time-bin states. In the initial phase of the state preparation, a 10-mm α -BBO crystal produces two adjacent time bins: $|t_0\rangle$ and $|t_1\rangle$. By adjusting the angle of the HWP preceding the crystal, we can create an intermediate state represented as

$$|\psi_{\text{in},1}\rangle = \cos\theta_1|H, t_0\rangle + \sin\theta_1|V, t_1\rangle. \quad (2)$$

Our objective here is to generate an arbitrary time-bin state, independent of polarization. To achieve this, we project the overall state onto a single polarization, resulting in the following expression:

$$|\psi_{\text{in},2}\rangle = |H\rangle \otimes (\cos\theta_1|t_0\rangle + \sin\theta_1|t_1\rangle). \quad (3)$$

It is worth noting that the introduction of an additional quarter-wave plate (QWP) allows the generation of a more general state: $|\psi_{\text{in},2}\rangle = |H\rangle \otimes (\cos\theta_1|t_0\rangle + \exp(i\phi_1)\sin\theta_1|t_1\rangle)$. The subsequent stage of the state preparation involves a 20-mm α -BBO crystal (or a sequence of two 10-mm α -BBO crystals) to generate the subsequent time bins, specifically $|t_2\rangle$ and $|t_3\rangle$. By adjusting the optical axis of a second HWP, we gain control over the intermediate input state, resulting in

$$|\psi_{\text{in},3}\rangle = \cos\theta_2|H\rangle \otimes (\cos\theta_1|t_0\rangle + \sin\theta_1|t_1\rangle) + \sin\theta_2|V\rangle \otimes (\cos\theta_1|t_2\rangle + \sin\theta_1|t_3\rangle). \quad (4)$$

Projecting onto a specific polarization state once again, we obtain the following intermediate state:

$$|\psi_{\text{in},4}\rangle = |H\rangle \otimes (\cos\theta_1\cos\theta_2|t_0\rangle + \sin\theta_1\cos\theta_2|t_1\rangle + \cos\theta_1\sin\theta_2|t_2\rangle + \sin\theta_1\sin\theta_2|t_3\rangle). \quad (5)$$

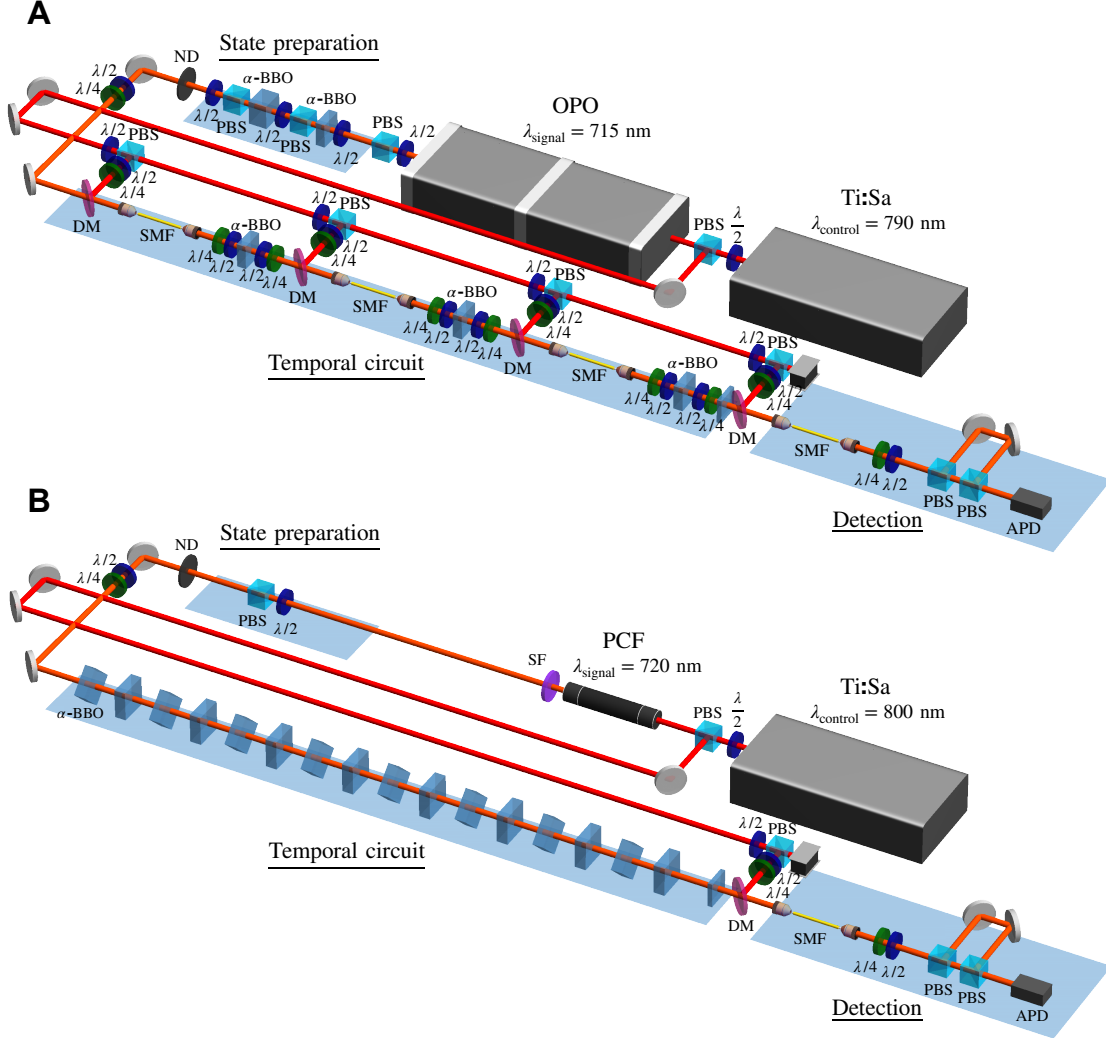


FIG. S1. **Experimental setup.** **A** Detailed experimental setup for the first demonstration investigating the programmability of the ultrafast time-bin encoding platform. **B** Detailed experimental setup for the second demonstration investigating the scalability of the ultrafast time-bin encoding platform.

Finally, by adjusting a final HWP, the polarization can be tuned to generate the ultimate input state:

$$\begin{aligned}
 |\psi_{\text{in}}\rangle = & \cos \theta_1 \cos \theta_2 \cos \theta_3 |H, t_0\rangle + \cos \theta_1 \cos \theta_2 \sin \theta_3 |V, t_0\rangle \\
 & + \sin \theta_1 \cos \theta_2 \cos \theta_3 |H, t_1\rangle + \sin \theta_1 \cos \theta_2 \sin \theta_3 |V, t_1\rangle \\
 & + \cos \theta_1 \sin \theta_2 \cos \theta_3 |H, t_2\rangle + \cos \theta_1 \sin \theta_2 \sin \theta_3 |V, t_2\rangle \\
 & + \sin \theta_1 \sin \theta_2 \cos \theta_3 |H, t_3\rangle + \sin \theta_1 \sin \theta_2 \sin \theta_3 |V, t_3\rangle. \quad (6)
 \end{aligned}$$

It is important to note that the addition of QWPs at each stage or the removal of some PBS elements can allow for

the preparation of more general states while preserving the coupling between polarization and time bins. However, in the current configuration, we are able to sample a wide array of input states that are particularly useful for transformations such as high-dimensional Hadamard gates. Finally, the signal is attenuated to the single photon level using a neutral density filter, achieving an average mean photon number per pulse of $\mu = 0.18$ and $\mu = 0.8$ for the first and second experiment, respectively.

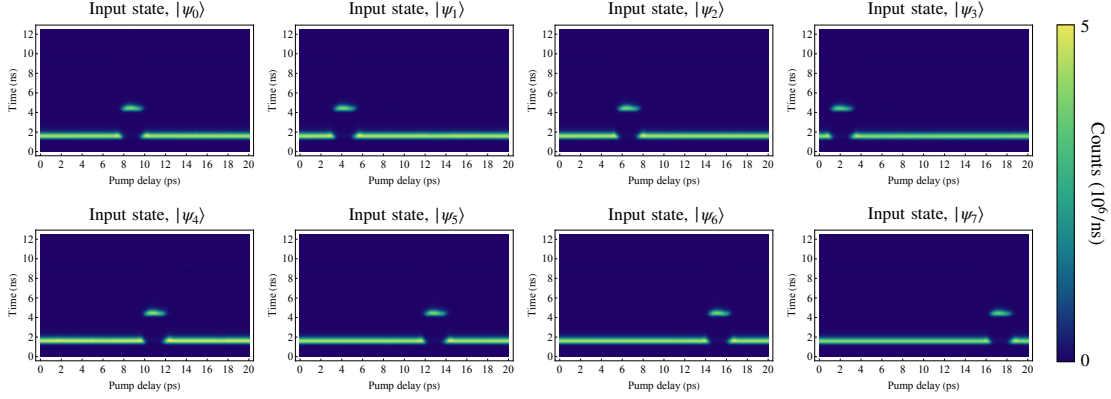


FIG. S2. **Measured counts.** The measured number of counts is shown as a function of arrival time on the APD for varying pump delay. The counts are shown for each input state. By scanning the pump delay with respect to the output signal, we can determine the output probability distribution of the ultrafast time-bin states exiting the temporal circuit.

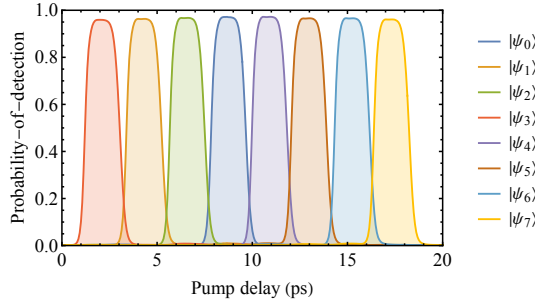


FIG. S3. **Probability-of-detection trace.** The ratio of counts in each polarization state, determined by the appropriate coincidence window, is shown as a function of pump delay, for each input state.

B. Temporal circuit

In the first experiment, the input state is directed into the temporal circuit for further processing. This is accomplished by coupling the input signal into a 14-cm single-mode fiber (SMF). To program the temporal circuit, the control beam is also coupled to the single-mode fibers. We achieved an average coupling efficiency of 85% and 80% for the signal and control pulses, respectively, across all SMFs in the temporal circuit. Although not depicted in Figure S1, lens systems are employed to prevent spatial divergence of the control beam throughout the setup, ensuring higher coupling efficiency for the control beam. Efficient coupling of the signal pulses between consecutive SMFs is facilitated by utilizing 3-axis fiber coupling stages at both the input and output stages. Furthermore, we estimate that the signal coupling effi-

ciency could be further enhanced by incorporating anti-reflection coated fibers.

At each SMF stage, the signal and control beams are initially overlapped using a dichroic mirror (DM). To compensate for polarization rotations induced by the DM, a combination of half-wave plates (HWP) and quarter-wave plates (QWP) is employed for both the signal and control beams. The relative time of arrival of the control pulse is independently adjusted for each step of the temporal circuit using motorized translational stages. By varying the time of arrival of the control pulses, the energy of the control pulses, and the polarization of the control pulses, we can program the temporal circuit to operate in a wide range of configurations. It is worth mentioning that the control pulses can also be prepared in a superposition of ultrafast time-bins using appropriate thicknesses of α -BBO crystals, although not shown in Figure S1. Additionally, HWP and QWP combinations are placed after each SMF within the temporal circuit to compensate for any polarization rotation of the signal pulses occurring inside the SMFs. Finally, due to limitations in pulse energy from the Ti:Sa oscillator, we restrict our temporal circuit to include only three SMFs, with an additional SMF dedicated to measuring the final output. However, by utilizing laser systems with lower repetition rates, we anticipate the availability of a significantly larger number of independently controllable control pulses without power restrictions.

In the second experiment, we deliberately limit the degree of programmability to investigate the scalability of our platform. In this context, we focus on a passive temporal circuit without active optical nonlinear phase modulation. In the circuit diagram, this corresponds to fixed values of the coupling and phase parameters within a single Programmable Optical Ultrafast Temporal Interferometric Network Element (POUTINE). However, it is important to note that each POUTINE can still be inde-

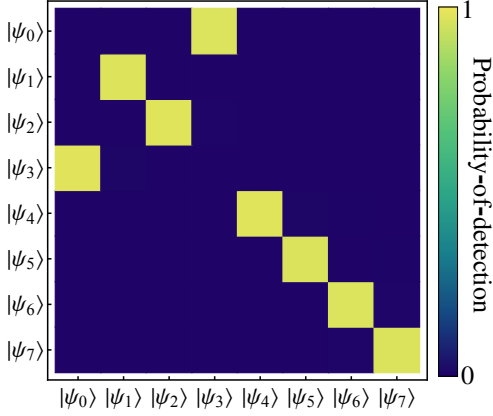


FIG. S4. **Probability-of-detection matrix for $U_{13}^{N=8}$.** The input and output ultrafast time-bin states are measured in the computational basis, i.e. $|\psi_i\rangle$.

pendently programmed by adjusting the rotation and tilt of the α -BBO crystal in each layer of the temporal circuit. By sacrificing some configurability, we can explore the performance of our platform when increasing the circuit depth up to 18 POUTINES, corresponding to a total number of modes of $N = 36$ in the temporal circuit. This circuit is achieved via a sequence of 18 α -BBO crystals. On average, each crystal contribute to -0.044 dB of loss.

C. Detection

The output of the temporal circuit is directed to a final SMF, where the signal beam is overlapped with a control pulse. Prior to coupling to the SMF, the signal passes through a 5-mm α -BBO crystal, enabling the conversion of the final polarization information into an additional time-bin degree of freedom. As a result, we obtain an 8-dimensional time-bin-only state. In our demonstration, we use a polarizer rotated by 45 degrees, to co-polarized all time bins exiting the 5-mm α -BBO crystal, thus simplifying the presentation of our measurement for this demonstration. By doing so, we measure only half of the photons coming out of the crystal. However, both polarization states can be analyzed in parallel using two separate optical switch to measure the final output time-bin states. The co-polarized time-bin state is then efficiently coupled to the final SMF. Prior to the final SMF, appropriate spectral filtering is required to filter out the control pulses. This is achieved with appropriate short-pass and band-pass filters. Moreover, the dichroic mirrors used to combine the control and signal pulses in the temporal circuits also help in filtering out the control pulses throughout the circuit. Within the fiber, the control pulse is scanned across the signal state, leading to

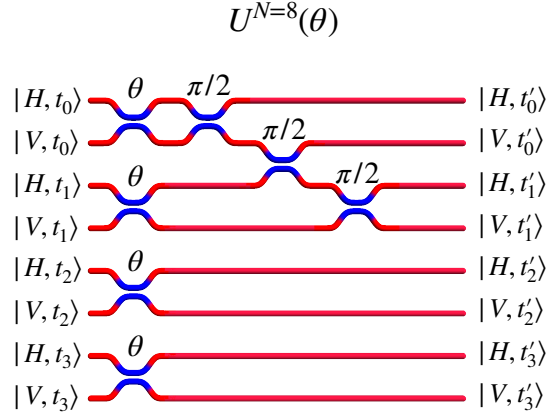


FIG. S5. **Passive programming quantum circuit.** Circuit diagram of a transformation that is passively programmed between the $N = 8$ permutation gates $U_{98}^{N=8}$ and $U_{99}^{N=8}$. The uniform coupling parameter θ across a full layer of the circuit is achieved experimentally via a half-wave plate rotated at the appropriate angle.

polarization rotation of specific time-bin output states. To reconstruct the ultrafast time-bin state at the output of the temporal circuit with sub-picosecond resolution, we employ two polarizing beam splitters (PBS) to measure the ratio of horizontal-to-vertical polarization at the SMF output. Each polarization is made to propagate through different path lengths, thus resulting in different nanosecond scale time-of-arrival at the APD. This measurement provides us with the necessary information for accurate reconstruction of the final ultrafast time-bin state. We note that in a typical experiment, one can use a series of switch to demultiplex each polarization and time-bin states to a single APD, resulting in 8 detectors passively recording the output of each mode of the temporal circuit.

For photon detection, we utilize an avalanche photodiode (APD). To ensure precise temporal synchronization, the APD is electronically gated by referencing the pump laser's pulses, which are measured by a separate photodiode. The signals detected by the APD are then transmitted to a time-to-digital converter (TDC) (Swabian Instrument, Time Tagger Ultra) for further analysis. The TDC exhibits a timing jitter of 22 ps (full width at half maximum). However, since the maximum data-transfer rate of the TDC is 65 MHz, which is lower than the repetition rate of the pump laser, we implement a conditional filter. This filter effectively discards all clicks from the fast photodiode that do not coincide with a detection event from the APD, directly at the TDC. To define the coincidence window, we rely on the timing jitter of the APD. In this case, we set the coincidence time win-

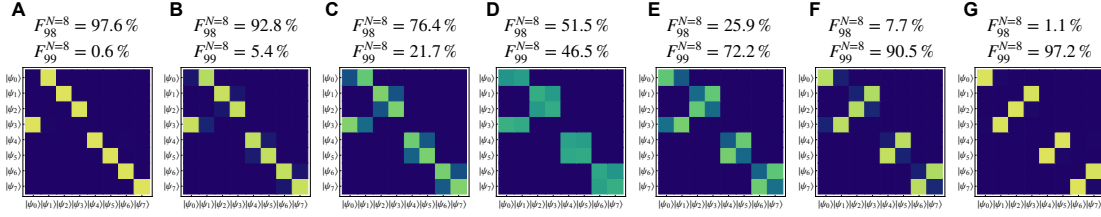


FIG. S6. **Passive programming.** Experimentally measured probability-of-detection matrices corresponding to passively programmed transformations between the $U_{98}^{N=8}$ and $U_{99}^{N=8}$ permutation gates. The programming parameter θ is varied from **A** $\theta = 0$, to **G** $\theta = \pi/2$ in increments of $\pi/12$. Experimental gate fidelities for the permutation gates $U_{98}^{N=8}$ and $U_{99}^{N=8}$ are shown for each transformation.

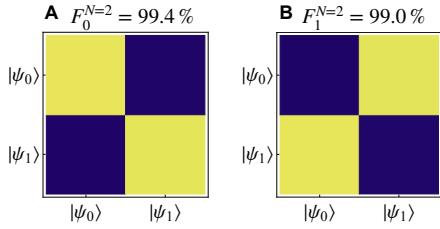


FIG. S7. **Permutation gates for $N = 2$.** Experimentally measured probability-of-detection matrices for the 2 permutation gates in dimension $N = 2$. Corresponding experimental gate fidelities are shown for each transformation.

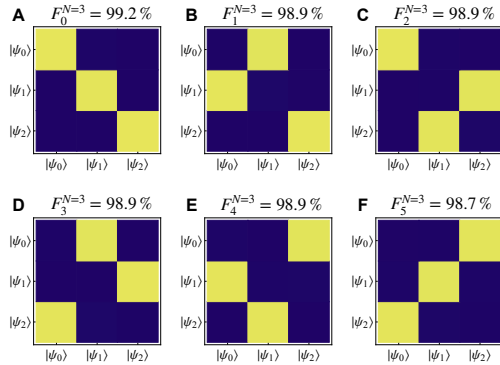


FIG. S8. **Permutation gates for $N = 3$.** Experimentally measured probability-of-detection matrices for the 6 permutation gates in dimension $N = 3$. Corresponding experimental gate fidelities are shown for each transformation.

dow to $\Delta t_{\text{coinc}} = 2$ ns, allowing for accurate detection events. From the measured data, we extract the probability of detection in each output time-bin state, enabling further analysis and characterization of the system's performance.

D. Mode couplers

A building block of our programmable quantum temporal circuit is the ability to couple adjacent modes in the interferometric network. For two adjacent modes, represented by the creation operators $\hat{a}_{H,t_j}^\dagger$ and $\hat{a}_{V,t_j}^\dagger$, an arbitrary mode coupler can be represented by the 2-dimensional unitary transformation,

$$U_{\text{Kerr}}(\theta_j, \phi_j) = \begin{pmatrix} e^{i\phi_j} \cos \theta_j & -i \sin \theta_j \\ -ie^{i\phi_j} \sin \theta_j & \cos \theta_j \end{pmatrix} \otimes |t_j\rangle\langle t_j|$$

analogous to a symmetric beam splitter with a phase shifter in one of the input ports. The parameter θ_j controls the strength of the coupling between the two modes and ϕ_j controls the relative phase between the two output modes. This transformation amounts to a general polarization transformation that can be achieved by a birefringent element with the appropriate birefringence and orientation of the optical axis. However, we note that this birefringence must be applied only onto the time-bin t_j and turned off for subsequent time bins. We achieve this ultrafast birefringence optically via the nonlinear optical Kerr effect inside the SMF. More precisely, the strong control pulse induces a nonlinear phase shift

$$\Delta\Phi = 8\pi n_2 L_{\text{eff}} I_{\text{pump}} / 3\lambda_{\text{signal}}, \quad (8)$$

between the orthogonal polarization states of a signal pulse, where n_2 is the nonlinear refractive index of the SMF, L_{eff} is the effective length of the nonlinear medium, I_{pump} is the intensity of the control pulse, and λ_{signal} is the center wavelength of the signal pulse. Moreover, we take advantage of the group velocity difference between the signal and pump wavelength in order to achieve a uniform nonlinear phase across the temporal profile of the signal pulse.

The local polarization transformation induced by a control pulse is described by the Jones matrix associated to a linear phase retarder (LPR) with an optical retardation of δ and its optical axis at an angle of ψ with respect to the horizontal axis, i.e.,

$$U_{\text{LPR}}(\delta, \psi) = e^{-i\frac{\delta}{2}} \begin{pmatrix} \cos^2 \psi + e^{i\delta} \sin^2 \psi & (1 - e^{i\delta}) \cos \psi \sin \psi \\ (1 - e^{i\delta}) \cos \psi \sin \psi & \sin^2 \psi + e^{i\delta} \cos^2 \psi \end{pmatrix}. \quad (9)$$

Here, the parameters δ and ψ correspond to the nonlinear phase shift shown in Eq. 8, where $\delta(I_{\text{pump}}) = \Delta\Phi(I_{\text{pump}})$ is set by varying the intensity of the control pulse, and the polarization angle of the control pulse, respectively. For a general unitary transformations, a sequence of two control pulses with variable optical retardations δ_1 and δ_2 with their optical axis at $\psi_1 = 0^\circ$ and $\psi_2 = 45^\circ$, respectively, maps directly to the desired 2x2 unitary transformation from Eq. 7. The corresponding Jones matrix is given by,

$$\begin{aligned} U_{2 \times 2}(\delta_1, \delta_2) &= U_{\text{LPR}}(\delta_2, \pi/4) \cdot U_{\text{LPR}}(\delta_1, 0) \\ &= \begin{pmatrix} \cos \delta_2/2 & -i \sin \delta_2/2 \\ -i \sin \delta_2/2 & \cos \delta_2/2 \end{pmatrix} \begin{pmatrix} e^{i\delta_1} & 0 \\ 0 & 1 \end{pmatrix} \\ &= \begin{pmatrix} e^{i\delta_1} \cos \delta_2/2 & -i \sin \delta_2/2 \\ -i e^{i\delta_1} \sin \delta_2/2 & \cos \delta_2/2 \end{pmatrix}, \quad (10) \end{aligned}$$

where we obtain the transformation for a symmetric beam splitter with reflectivity $\cos \delta_2$ and phase shift δ_1 . Thus the mode coupler operation in Eq. 7 is achieved with a sequence of two control pulses with,

$$\theta_j(I_j^{(2)}) = \Delta\Phi_j^{(2)}/2 = 4\pi n_2 L_{\text{eff}} I_j^{(2)}/3\lambda_{\text{signal}} \quad (11)$$

$$\phi_j(I_j^{(1)}) = \Delta\Phi_j^{(1)} = 8\pi n_2 L_{\text{eff}} I_j^{(1)}/3\lambda_{\text{signal}}, \quad (12)$$

where $I_j^{(1)}$ and $I_j^{(2)}$ correspond to the intensity of the first and second control pulses, respectively. We note that achieving the two-pulse configuration can be approached in two distinct manners. Firstly, it can be realized by employing two separate SMFs, effectively doubling the total number of required SMFs. Alternatively, it can be executed within a single longer SMF by operating each control pulse within the time-bin separation of the quantum signals. The latter approach necessitates fewer SMFs resulting in lower loss due to imperfect coupling efficiency, but leads to more noise since each control pulse must propagate through a larger length of fiber. Moreover, the sequence of co-polarized and diagonally polarized control pulses for each POUTINE can be meticulously prepared with independently adjustable intensity using various recombining schemes. For instance, employing a sequence of birefringent crystals with precisely selected optical axis angles, followed by a final polarizer, allows for the generation of a controlled train of intensity-adjusted control pulses. Furthermore, adding fast electro-optic modulators between the birefringent crystals can achieve the same result with large reconfiguration speeds.

Furthermore, we note that for a large subset of unitary transformations U_{Kerr} with mode coupling parameters, θ_j

and ϕ_j , a single control pulse can be enough to perform the desired transformation. This can be seen by comparing Eq. 7 with the Jones matrix associated with a single control pulse, see Eq. 9.

The fully connected quantum temporal circuit can then be constructed by applying independent coupling, $\theta_{m(j,k)}$, and phase shifting, $\phi_{m(j,k)}$, parameters for each time bins, t_j , and each layer labeled by the index $k \in \{0, \dots, N-1\}$. The over programming of the circuit can be described by,

$$\begin{aligned} U_{\text{Kerr}}^{(k)}(\theta_m, \phi_m) &= \sum_j \begin{pmatrix} e^{i\phi_{m(j,k)}} \cos \theta_{m(j,k)} & -\sin \theta_{m(j,k)} \\ e^{i\phi_{m(j,k)}} \sin \theta_{m(j,k)} & \cos \theta_{m(j,k)} \end{pmatrix} \otimes |t_j\rangle\langle t_j|, \quad (13) \end{aligned}$$

where a total of $m \in \{1, \dots, N(N-1)/2\}$ coupling and phase shifting parameters can be independently tuned via the optical Kerr effect.

For example, in the case of the 4-dimensional Hadamard gate, a control pulse polarized at an angle of 45° with respect to the horizontal polarization direction of the signal pulse achieves the $\theta = \pi/2$ mode coupling, see Fig. 2-D from the main text. However, the nonlinear polarization rotation achieved by the control pulse also leads to an additional phase shift between the horizontal and vertical polarization states of the signal pulses. The additional phase can be undone to realize a $\phi = 0$ mode coupler with a second control pulse polarized at a fixed angle of 0° with respect to the horizontal polarization of the signal pulses.

II. SUPPLEMENTARY EXPERIMENTAL RESULTS

A. Data processing

Here we outline the process by which the output time-bin states from the temporal circuit are measured and processed to obtain the final probability-of-detection matrix. To do so, we choose a specific temporal circuit and show each step of the data processing.

The unitary transformation that we consider here is the 8-dimensional permutation gate,

$$U_{13}^{N=8} = \begin{pmatrix} 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 \end{pmatrix}. \quad (14)$$

For each input time-bin state in the temporal circuit, the output time-bin state is sent through the detection stage comprised of a Kerr switch where the relative delay of the control pulse with respect to the output time-bin

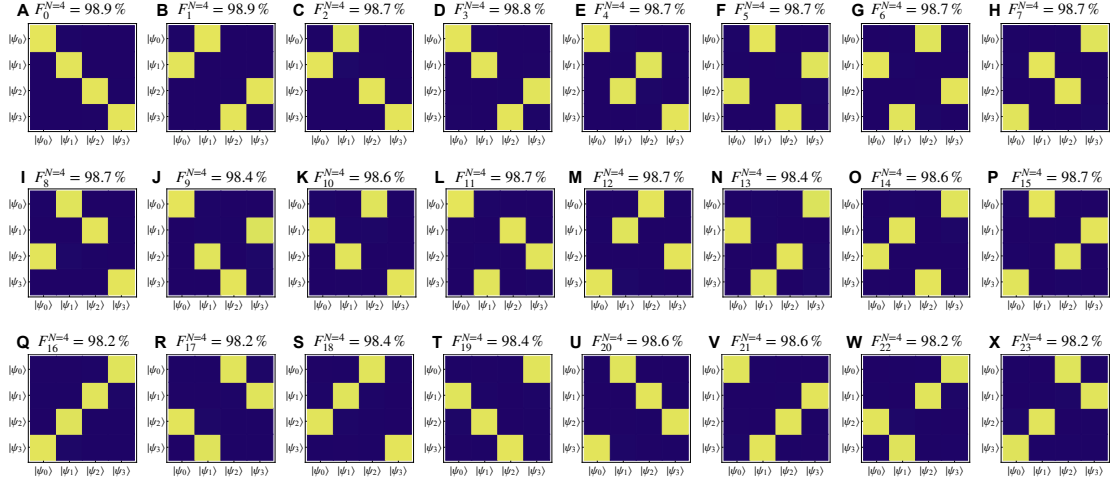


FIG. S9. **Permutation gates for $N = 4$.** Experimentally measured probability-of-detection matrices for the 24 permutation gates in dimension $N = 4$. Corresponding experimental gate fidelities are shown for each transformation.

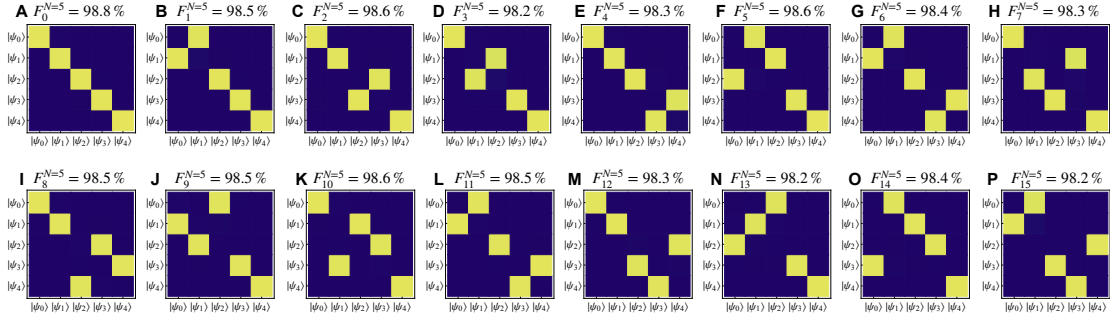


FIG. S10. **Permutation gates for $N = 5$.** Experimentally measured probability-of-detection matrices for the first 16 permutation gates in dimension $N = 5$. Corresponding experimental gate fidelities are shown for each transformation.

state is varied. By varying the control pulse delay, we can scan the temporal profile of the temporal circuit output. The output of the Kerr switch is sent to a PBS where a polarization-dependent delay allows us to resolve different polarization states temporally using a single APD, see Fig. S2.

The counts are acquired by integrating the signals within two coincidence windows corresponding to horizontal and vertical polarizations. The ratio of these counts is calculated to generate probability-of-detection trace, as illustrated in Figure S3. These matrices serve to define the probability of measuring photons in different ultrafast time-bin states at the output of the temporal circuit. The shape of the probability-of-detection trace, observed as a function of pump delay, is determined by several factors. These factors include the temporal profiles of the control pulse and the signal pulse,

the difference in group velocity between the control and signal pulses within the SMF, and the length of the SMF itself. The presence of a flat-top characteristic in the probability-of-detection trace signifies that the Kerr switch covers a longer time duration compared to the duration of the signal pulse. This aspect ensures that the detection efficiencies remain close to unity. However, it is worth noting that this arrangement may introduce additional noise stemming from the control pulse within the SMF, which can be considered as unnecessary noise for the desired signal.

To obtain a comprehensive understanding of the system, we calculate a probability-of-detection matrix based on the probability-of-detection trace as a function of pump delay. For all output ultrafast time-bin state of the temporal circuit, a corresponding set of pump delay values is determined. The probability-of-detection

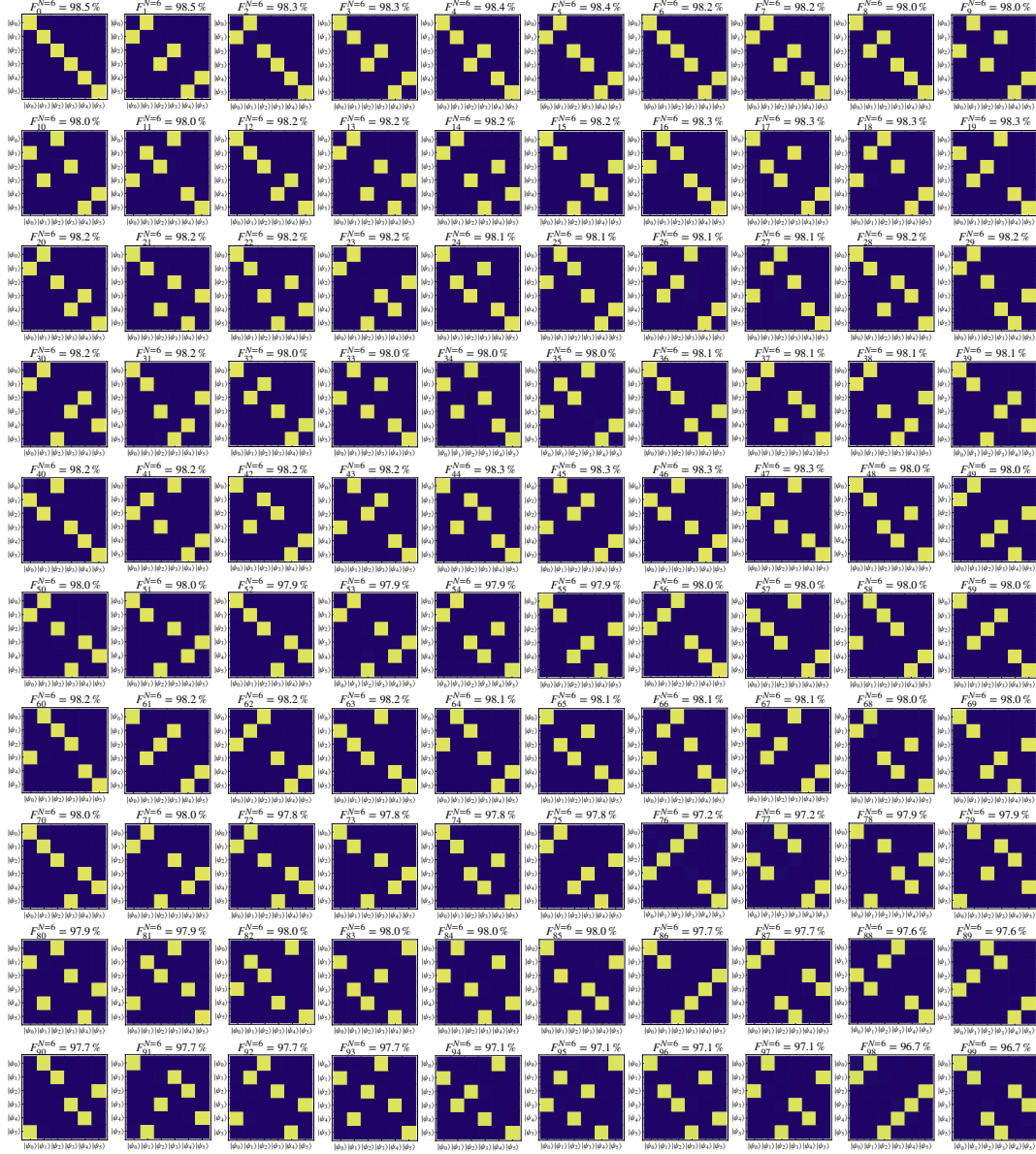


FIG. S11. **Permutation gates for $N = 6$.** Experimentally measured probability-of-detection matrices for the first 100 permutation gates in dimension $N = 6$. Corresponding experimental gate fidelities are shown for each transformation.

matrix is then constructed by considering the ratio of counts in each polarization at those specific pump delays for each input state, as depicted in Figure S4. This matrix provides valuable insights into the measurement probabilities associated with different experimentally realized unitary transformations.

B. Passive programming

In our temporal circuit architecture, parameters in the network can be tuned optically by varying the polarization and the pulse energy of the control pulses. Moreover, for uniform mode parameters across a layer of the

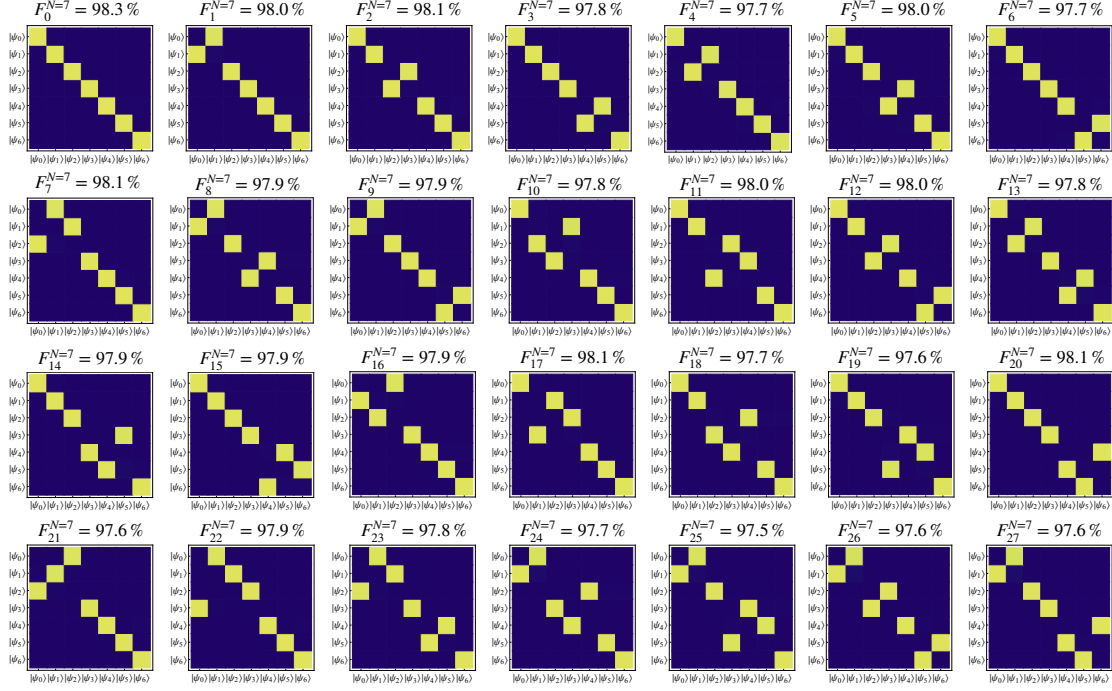


FIG. S12. **Permutation gates for $N = 7$.** Experimentally measured probability-of-detection matrices for the first 28 permutation gates in dimension $N = 7$. Corresponding experimental gate fidelities are shown for each transformation.

interferometric network, it is possible to program sections of the circuits using passive polarization optics, see Fig. S5. As an example, we implement an 8-dimensional permutation gate consisting of 3 optically programmed mode couplers. A half-wave plate is then placed at the input of the circuit and rotated to tune the coupling parameter θ of each mode couplers in the first layer of the circuit. By doing so, we continuously transition from the 8-dimensional permutation gates $U_{98}^{N=8}$ to $U_{99}^{N=8}$, see Fig. S6.

C. Permutation gates

We explore the programmability of our platform by realizing an important set of transformations, namely permutation gates. For lower dimensions, permutation gates corresponds to well know transformations such as Pauli

gates, cyclic gates, and a wide range of controlled-gates. Here, we perform an exhaustive experimental investigation of the implementation of permutation gates in dimensions ranging from $N = 2$ to $N = 8$. For a given dimension, or number of modes, N , there exist $N!$ corresponding permutation transformations. For lower dimensions, we have experimentally implemented and measured all permutation transformations, i.e. 2, 6, and 24 for dimensions $N = 2, 3$, and 4, respectively. However, for larger dimensions, such as $N = 8$, where the total number of permutation gates is larger than 40,000, we limit ourselves to the first 186 permutation gates. For dimensions $N = 5, 6$, and 7, we measured a total of 16, 100, and 28 permutation transformations, respectively. This corresponds to having implemented a total of 362 permutation transformations. Experimentally measured probability-of-detection matrices for $N = 2, 3, 4, 8$ are shown in Fig S7-S14.

10

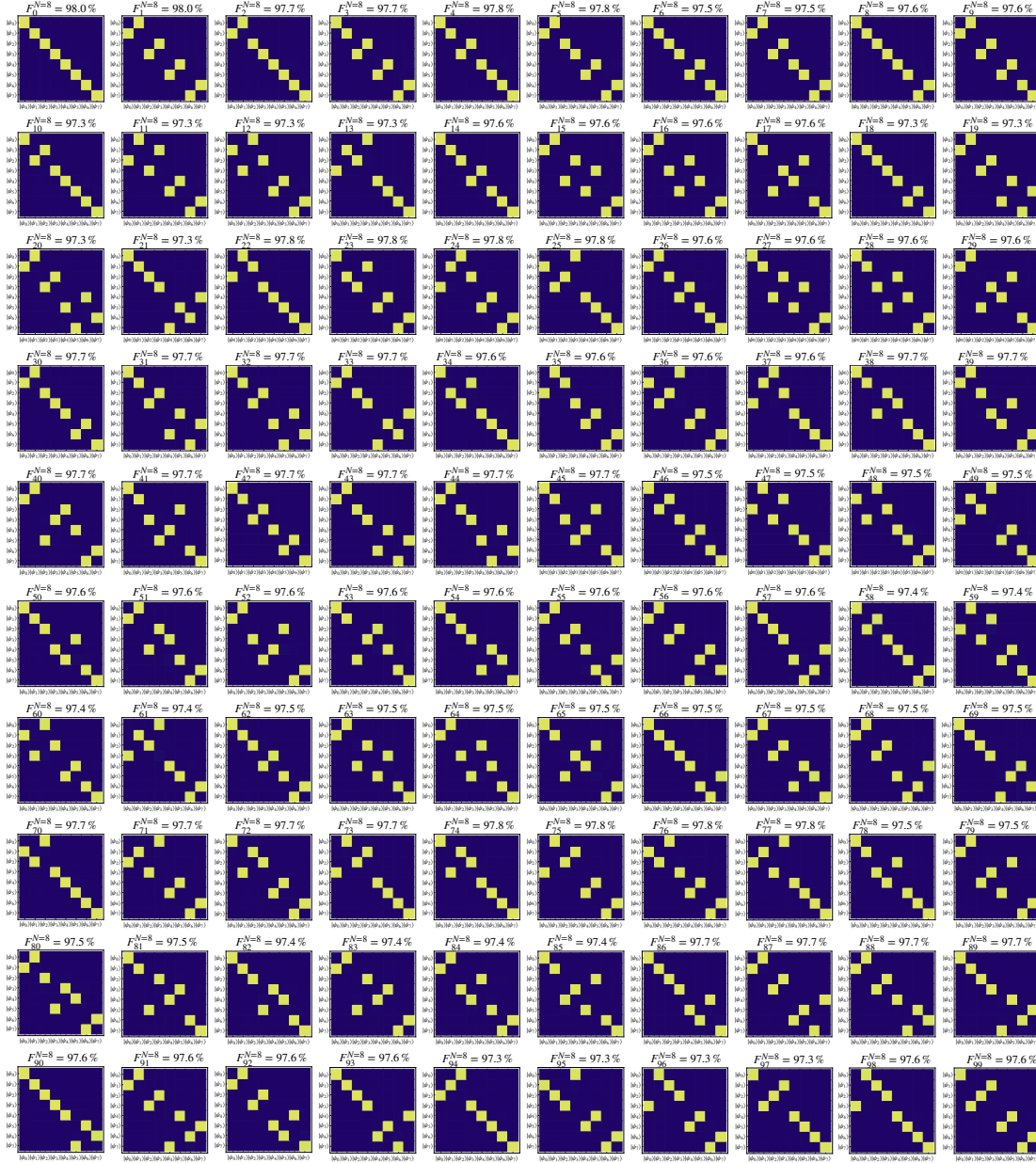


FIG. S13. **Permutation gates for $N = 8$.** Experimentally measured probability-of-detection matrices for permutation gates $U_0^{N=8}$ to $U_{99}^{N=8}$ in dimension $N = 8$. Corresponding experimental gate fidelities are shown for each transformation.

11

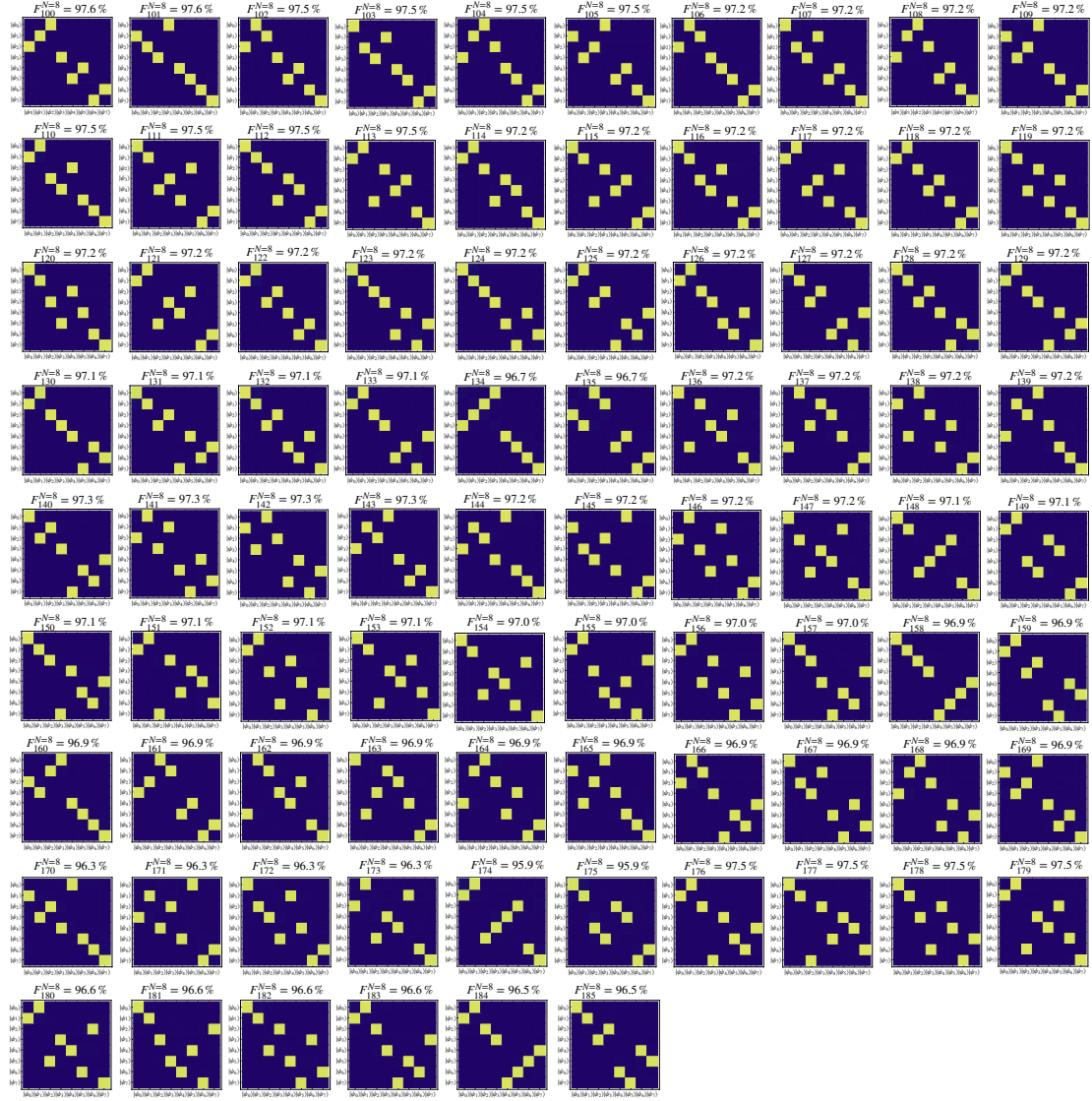


FIG. S14. **Permutation gates for $N = 8$.** Experimentally measured probability-of-detection matrices for permutation gates $U_{100}^{N=8}$ to $U_{185}^{N=8}$ in dimension $N = 8$. Corresponding experimental gate fidelities are shown for each transformation.

Chapter 5

Multiphoton quantum information processing

Ultrafast time-bin encoding (UTBE) demonstrates the key ingredients for scalability in quantum information processing, including high interferometric phase stability for a large number of modes, programmability, and low loss. With these combined capabilities, UTBE offers a promising pathway to scaling up photonic quantum information processors. As we have seen, UTBE works well in the single-photon regime; however, we must still confirm that it is capable of operation with multiple photons. In fact, computational complexity grows exponentially with the number of photons present in a quantum information processor. Multiphoton capabilities are thus often regarded as what gives these devices an edge over classical computers. We first summarize the requirements for scalable quantum information processing in Section 5.1. Next, in Section 5.2, we consider multiphoton interference effects, focusing on the hallmark Hong-Ou-Mandel interference which forms the basis for a large portion of quantum information processing and communication schemes. This chapter concludes with a demonstration of up to three-photon interference in a quantum walk with UTBE, in Section 5.3.

5.1 Requirements for scalable quantum information processing

It is generally accepted that a quantum computer must meet the criteria set by DiVincenzo in 2000 [69]. Our UTBE scheme is examined in the context of DiVincenzo’s criteria in Table 5.1. In an ideal scenario, a quantum information processor would

DiVincenzo’s criteria for quantum computing
Well-defined state space of qubits: The state space defined by UTBE is experimentally accessible with high accuracy. Furthermore, UTBE provides access to robust, high-dimensional qudits. We must now work to create a system with an arbitrarily high number of well-characterized qudits. Ability to initialize the state of the qubits: Photons can be easily initialized to a desired input state with UTBE using birefringent crystals and wave plates. The challenge lies in creating multiphoton input states, which rely on non-deterministic (low efficiency) processes. Long relevant decoherence times: Photons naturally have long decoherence times; yet, factors such as loss and instability can lead to dephasing. UTBE provides low loss and high stability, but there is still room for improvement. A universal set of quantum gates: Rather than implement qubit operations, UTBE uses qudits to achieve programmable unitary transformations, which has seen success operating on single photons. Deterministic multiphoton operations remain a challenge; however, measurement-based schemes present a promising avenue. A qubit-specific measurement capability: Projective measurements of a UTBE quantum state can be performed with high efficiency through optical Kerr gating, but this is still limited by fiber-coupling losses. Multiphoton operation will require as low loss as possible.

Table 5.1: **DiVincenzo’s criteria for quantum computing.** A summary of the criteria necessary for quantum computing, set by DiVincenzo in 2000 [69], including an evaluation of how our ultrafast time-bin encoding (UTBE) scheme fares against them.

have zero loss, absolutely no decoherence, complete stability, and perfect fidelity of all operations; however, in practice, systems will always have some loss and decoherence, may not be perfectly stable, and operation fidelity will not be 100%. The goal is to get as close as possible to these characteristics, such that error correction can be used to compensate for any remaining imperfections.

We see in Table 5.1 that UTBE generally meets DiVincenzo’s criteria for quantum computing; yet, the need for multiphoton operations is a recurring theme. A logical next step for us is therefore moving into the multiphoton quantum information processing regime. We extend the capabilities of UTBE to the multiphoton regime in this chapter, where we have elected to showcase the quantum walk (QW) algorithm operating on up to three photons¹.

5.2 Multiphoton quantum interference

Wave interference is ubiquitous in the physical world. Although it is often associated with signal degradation, interference is also of considerable utility; *e.g.*, in tuning musical instruments [475], optimized performance in concert halls [476], and in reaching exquisite levels of measurement sensitivity, such as for measurement of gravitational waves [477]. The “waves” of quantum mechanics can also interfere mathematically in Hilbert space. Quantum interference is differentiated from classical interference by its unusual scaling, making it a key ingredient for quantum computational speedup [478].

Having no classical analogue, multiphoton interference is fundamental to photonic quantum information processing and provides a distinct advantage over classical computation. In 1987, Hong, Ou, and Mandel published their seminal demonstration of quantum interference between two photons (usually referred to as *HOM interference*) [479], which has since formed the basis for many quantum photonic applications, including quantum communication [480] and quantum computation [76]. In dual-rail photonic quantum computing, two-photon HOM interference is heavily relied upon to perform the two-qubit gates necessary for universal computation [76]. In

¹It should be noted that we infer the presence of up to three photons using various coincidence click patterns at our detectors, described in more detail in [268].

measurement-based schemes, multiphoton interference in large multiport interferometers is crucial [481].

5.2.1 Two-photon Hong-Ou-Mandel (HOM) interference

In order to better understand why multiphoton interference is so important for quantum computation, we turn our attention towards two-photon Hong-Ou-Mandel (HOM) interference². The action of a beam splitter is at the heart of HOM interference. Previously, we described the unitary action of a beam splitter in Section 4.1.1. We use this formalism to describe how a beam splitter can be used to mix two input modes, and demonstrate the outcome when these two input modes are indistinguishable photons. Although simple, we have seen previously that the beam splitter is an essential building block of any LOQC system.

We follow our description of a beam splitter, as in Fig. 4.1 and Eq. 4.1. For the HOM effect, we consider a 50:50 beam splitter, depicted in Fig. 5.1(a), where $\theta = \pi/4$ and we take $\phi = 0$. The beam splitter transformation can then be written as

$$\begin{aligned}\hat{a}_{\text{in}}^\dagger &\xrightarrow{\widehat{\text{BS}}} \frac{1}{\sqrt{2}} \left(\hat{a}_{\text{out}}^\dagger + \hat{b}_{\text{out}}^\dagger \right) \\ \hat{b}_{\text{in}}^\dagger &\xrightarrow{\widehat{\text{BS}}} \frac{1}{\sqrt{2}} \left(\hat{a}_{\text{out}}^\dagger - \hat{b}_{\text{out}}^\dagger \right),\end{aligned}\tag{5.1}$$

where we have written the transformation on the input creation operators, $\hat{a}_{\text{in}}^\dagger$ and $\hat{b}_{\text{in}}^\dagger$, in terms of the output creation operators, $\hat{a}_{\text{out}}^\dagger$ and $\hat{b}_{\text{out}}^\dagger$. We will see momentarily why this is a useful representation. In HOM, two photons are incident on the beam splitter, one at each input port. This can be written in terms of creation operators, in the beam splitter input modes, acting on the multimode vacuum state, $|0\rangle$, as

$$|\psi\rangle_{\text{in}} = \hat{a}_{\text{in},m}^\dagger \hat{b}_{\text{in},n}^\dagger |0\rangle,\tag{5.2}$$

where, for now, we allow for each photon to be distinguished based on an additional degree of freedom (*i.e.*, polarization, timing, frequency, spatial mode, ...), labeled

²For further reading on the HOM effect, I recommend this review article [482].

here by m and n . The beam splitter, $\widehat{\text{BS}}$, transforms this input state as

$$\begin{aligned} \hat{a}_{\text{in},m}^\dagger \hat{b}_{\text{in},n}^\dagger |0\rangle &\xrightarrow{\widehat{\text{BS}}} \frac{1}{2} \left(\hat{a}_{\text{out},m}^\dagger + \hat{b}_{\text{out},m}^\dagger \right) \left(\hat{a}_{\text{out},n}^\dagger - \hat{b}_{\text{out},n}^\dagger \right) |0\rangle \\ &= \frac{1}{2} \left(\hat{a}_{\text{out},m}^\dagger \hat{a}_{\text{out},n}^\dagger - \hat{a}_{\text{out},m}^\dagger \hat{b}_{\text{out},n}^\dagger + \hat{b}_{\text{out},m}^\dagger \hat{a}_{\text{out},n}^\dagger - \hat{b}_{\text{out},m}^\dagger \hat{b}_{\text{out},n}^\dagger \right) |0\rangle. \end{aligned} \quad (5.3)$$

These four terms correspond to the four scenarios depicted in Fig 5.1(b). The hall-

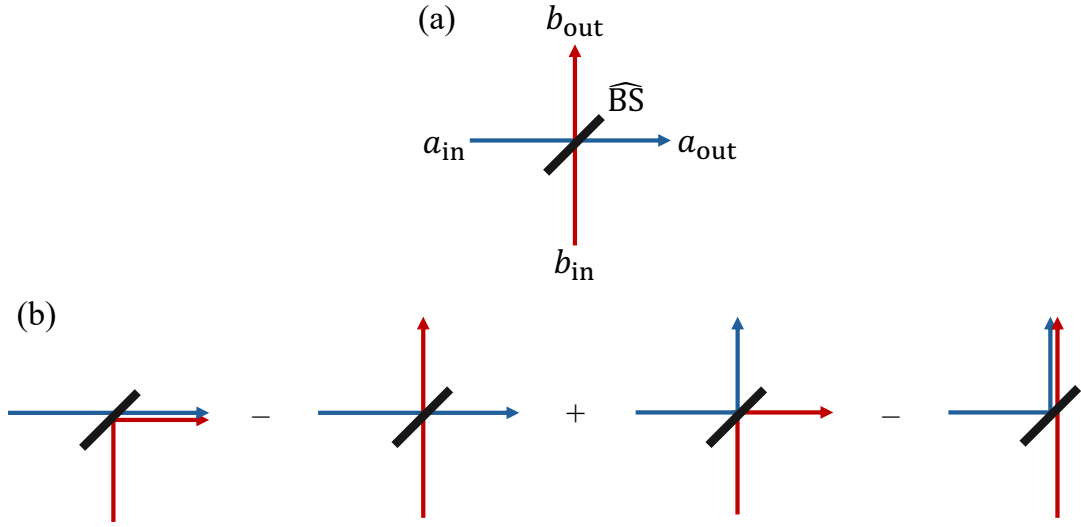


Figure 5.1: **Two-photon interference at a beam splitter.** A beam splitter is depicted in (a), with input ports, a_{in} and b_{in} , and output ports, a_{out} and b_{out} . The four possible ways in which the two photons can traverse the beam splitter are shown in (b), corresponding to the four terms in Eq. 5.3. Note that the degrees of freedom m and n are depicted in blue and red, respectively, to better visualize the different paths each photon can traverse.

mark HOM two-photon interference emerges when we consider indistinguishable photons at the two input ports (*i.e.*, $m = n$), such that the output state is now

$$\begin{aligned} \hat{a}_{\text{in}}^\dagger \hat{b}_{\text{in}}^\dagger |0\rangle &\xrightarrow{\widehat{\text{BS}}} \frac{1}{2} \left(\hat{a}_{\text{out}}^\dagger \hat{a}_{\text{out}}^\dagger - \hat{a}_{\text{out}}^\dagger \hat{b}_{\text{out}}^\dagger + \hat{b}_{\text{out}}^\dagger \hat{a}_{\text{out}}^\dagger - \hat{b}_{\text{out}}^\dagger \hat{b}_{\text{out}}^\dagger \right) |0\rangle \\ &= \frac{1}{2} \left[\left(\hat{a}_{\text{out}}^\dagger \right)^2 - \left(\hat{b}_{\text{out}}^\dagger \right)^2 \right] |0\rangle \\ &= \frac{1}{\sqrt{2}} \left[|2\rangle_{a_{\text{out}}} - |2\rangle_{b_{\text{out}}} \right], \end{aligned} \quad (5.4)$$

where we have removed the $m = n$ subscripts for simplicity. We see an entirely new interference effect—physically distinct from the interference of a single photon or between classical fields—that arises only in the quantum mechanical picture. This is an important result; such two-photon HOM interference is a purely quantum mechanical effect, and provides a means of controlling photons.

In placing detectors at each of the two output ports and performing a coincidence-type measurement, as in the experimental configuration shown in Fig. 5.2(a), one can expect to measure zero coincidence events. The characteristic HOM “dip”, as seen

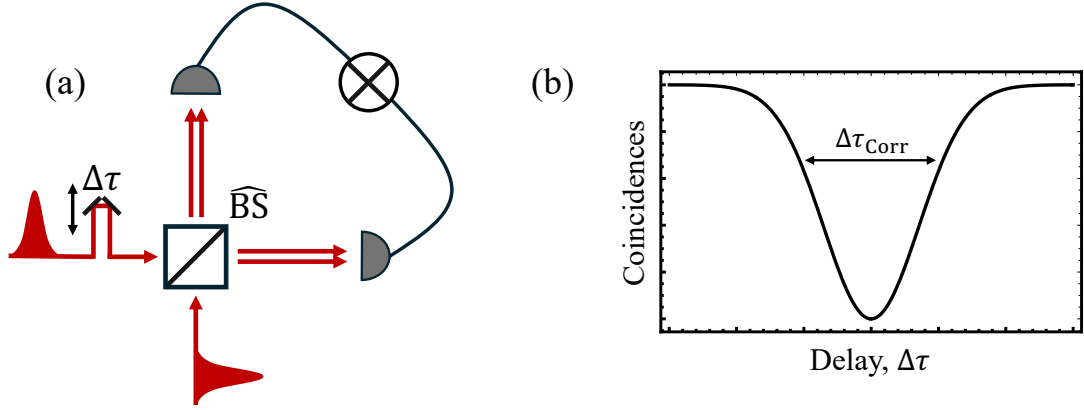


Figure 5.2: **A typical HOM experiment.** The typical experimental configuration to achieve HOM two-photon interference is depicted in (a), while its characteristic “dip” is depicted in (b). Here, $\Delta\tau_{\text{Corr}}$ refers to the correlation time of the incident photons, and is related to their bandwidth, $\Delta\omega$, through: $\Delta\tau_{\text{Corr}} = 1/\Delta\omega$. For perfectly indistinguishable photons, the visibility of the HOM dip will be 100%.

in Fig. 5.2(b), can be observed by adding a delay line to one of the input ports, and scanning the delay of one input photon relative to the other. Only when the two photons overlap in time will they interfere destructively. The visibility of this interference is related to the mode overlap between the two photons, calculated as

$$\mathcal{V} = \frac{C_{\text{max}} - C_{\text{min}}}{C_{\text{max}}}, \quad (5.5)$$

where C_{max} is the maximum coincidence rate, and C_{min} is the minimum coincidence rate as measured at $\Delta\tau$. It should be noted that this destructive and constructive

interference can be adjusted by tailoring the beam splitter operator or the symmetry of the two-photon input state, making this a critical tool in the photonic quantum computing toolbox.

5.2.2 Multiphoton interference

Thus far, we have considered only two-photon HOM interference at a beam splitter; however, interference of photons need not be limited to this simple, yet powerful, case. Indeed, it is possible to extend the capabilities of interference by introducing more photons within more complicated multiport interferometers. As evidenced by boson sampling demonstrations, the complexity of such a scheme quickly extends beyond the simulation capabilities of classical computers, providing a promising candidate for the demonstration of quantum advantage [18, 19].

The increased complexity offered by multiphoton interference in multiport interferometers opens the door to many other new effects, including in quantum teleportation schemes [483], generating multipartite entanglement [484], and most relevant to this thesis, in QWs. In sending just two photons into a multiport interferometer, their propagation can be described by a complex bipartite QW, which gives rise to interesting correlation patterns [138, 485]. Such capabilities are expected to be crucial in quantum simulation using QWs [486, 487]. Given the necessity of multiphoton operation for photonic quantum information processing, it is crucial that our UTBE scheme is capable of operation in the multiphoton regime.

5.3 Implementing multiphoton capabilities

This section is based on the following preprint:

K. L. Fenwick, J. Baker, G. S. Thekkadath, A. Z. Goldberg, K. Heshami, P. J. Bustard, D. England, F. Bouchard, and B. Sussman. “Multiphoton interference in a single-spatial-mode quantum walk”, *arXiv preprint arXiv:2409.11483* (2024).

DOI: <https://doi.org/10.48550/arXiv.2409.11483>

Multiphoton interference is crucial to many photonic quantum technologies, including especially quantum information processing. It is therefore important that our UTBE scheme is capable of operating in the multiphoton regime, as we demonstrate here. We elect to operate our UTBE scheme in a passive configuration, demonstrating a multiphoton QW algorithm that consists only of birefringent crystals (*i.e.*, no active Kerr gates are included within the interferometer). Although this limits programmability, the focus of this demonstration is on successful multiphoton interference, for which the QW provides an excellent testbed.

Here, we extend our QW from Section 4.4 to the multiphoton regime. We operate the QW up to 11 steps, providing access to as many as 12 ultrafast time bins (a total of 24 modes). The key difference here lies in the input state to the QW, which now consists of an attenuated coherent state and one photon from a pair source. We observe a HOM visibility of up to 70% between the attenuated coherent state and heralded single photon at the first step of the QW. This, in itself, is rather interesting—we overcome the classical limit of 50% HOM visibility, and thus observe quantum interference between two photons which are generated from two independent processes. One might still be inclined to ask why we do not opt for two photon pair sources for this demonstration. Although a higher HOM visibility can be expected from two heralded photons at the input to the QW, our choice of input states provides more flexibility in observing various combinations of different input states.

Originally, we set out to demonstrate a two-photon QW, but found that we were also capable of observing some coincidence patterns consistent with three-photon propagation within the QW. As such, we measure different one-, two-, and three-fold coincidence patterns at the output of the QW to infer the propagation of one,

two, and three photons through the multiport interferometer. In order to measure these coincidence patterns, we require two Kerr gates at the output of the QW for time-demultiplexing. It should be noted that we measure the QW output in a single polarization basis. Both output polarizations could be observed with the addition of a second line of two Kerr gates, in parallel. Alternatively, the QW output distribution could be measured in the superposition basis of its two output polarizations. Furthermore, with two Kerr gates in our detection scheme, we can measure only a subset of three-fold coincidence patterns. With a third Kerr gate, the full three-fold coincidence pattern could be measured. Nevertheless, we still observe intriguing results. In the various one-, two-, and three-fold coincidence measurements, we can analyze the output probability distributions when a photon from the pair source is both heralded (*i.e.*, arising from a quantum two-mode squeezed vacuum state) and unheralded (*i.e.*, arising from a classical thermal state). Notably, we observe stronger clustering of probability outcomes when heralded—a signature of bosonic quantum interference, which can only arise from nonclassical input states.

Our experimental results are verified with a simulation performed using the open-source quantum optics simulation package, *Strawberry Fields*, from *Xanadu* [488, 489]. Our simulation propagates a two-mode squeezed vacuum (TMSV) state and an attenuated coherent state through a circuit which corresponds to our QW. In addition to verifying our results, our simulation provides us with another means of investigating nonclassicality in our demonstration. By replacing the TMSV state with its closest classical approximation, namely the two-mode squeezed thermal (TMST) state, we can demonstrate that the quantum model outperforms the classical model [490]. We find significantly better agreement between the experimental data and model when the simulation includes the quantum TMSV state rather than a TMST state.

Despite its limitations, this demonstration is an important step in outlining the promise of UTBE for photonic quantum information processing. Our scheme exhibits the same desirable low loss, high fidelity, and stable operation as seen in the previous chapter—only now, extended to the multiphoton regime. The low event detection rates arising from the use of probabilistic single-photon sources still remains an obstacle; however, with improvements in source technology (or perhaps with photon-number-resolving detectors), our UTBE scheme is anticipated to provide a promising pathway towards scalable photonic quantum information processing.

Multiphoton interference in a single-spatial-mode quantum walk

Kate L. Fenwick,^{1,2} Jonathan Baker,¹ Guillaume S. Thekkadath,² Aaron Z. Goldberg,² Khabat Heshami,^{1,2} Philip J. Bustard,² Duncan England,² Frédéric Bouchard,^{2,*} and Benjamin Sussman^{1,2}

¹*Department of Physics, University of Ottawa, Advanced Research Complex,
25 Templeton Street, Ottawa ON Canada, K1N 6N5*

²*National Research Council of Canada, 100 Sussex Drive, Ottawa, Ontario K1A 0R6, Canada*

Multiphoton interference is crucial to many photonic quantum technologies. In particular, interference forms the basis of optical quantum information processing platforms and can lead to significant computational advantages. It is therefore interesting to study the interference arising from various states of light in large interferometric networks. Here, we implement a quantum walk in a highly stable, low-loss, multiport interferometer with up to 12 ultrafast time bins. This time-bin interferometer comprises a sequence of birefringent crystals which produce pulses separated by 4.3 ps, all along a single optical axis. Ultrafast Kerr gating in an optical fiber is employed to time-demultiplex the output from the quantum walk. We measure one-, two-, and three-photon interference arising from various input state combinations, including a heralded single-photon state, a thermal state, and an attenuated coherent state at one or more input ports. Our results demonstrate that ultrafast time bins are a promising platform to observe large-scale multiphoton interference.

INTRODUCTION

Wave interference is ubiquitous in the physical world. While often associated with signal degradation, interference is also of considerable utility. For example, controlled interference of acoustic waves is critical for instrument tuning [1] and optimized performance in concert halls [2], while metrology frequently relies on interferometry and can reach exquisite levels of sensitivity, *e.g.*, in measuring gravitational waves [3]. By contrast, the “waves” of quantum mechanics interfere mathematically in Hilbert space. Quantum interference arises from the fact that quantum systems can remain in a superposition of many possible states, only until measurement. The unusual scaling of quantum interference differentiates it from classical interference, making it a key ingredient for quantum computational speedup [4] and the distribution of quantum walks, as demonstrated here.

Quantum interference can be exploited in many platforms, including trapped atoms [5] or ions [6, 7], nuclear magnetic resonance [8], and superconducting qubits [9]; however, photonic platforms are naturally suited to this task, offering advantages such as the ability to operate at room temperature and exceptional resilience to decoherence [10–12]. Amplitude-splitting interferometers [13] remain the tool nonpareil for the generation and manipulation of optical interference. They rely on the action of a beam splitter, and are also well-suited to measure quantum interference: when two indistinguishable single photons are each incident at one of the two input ports of a beam splitter, the hallmark “Hong-Ou-Mandel” interference dip can be observed in coincident detection at the output [14, 15]. This effect is the foundation of logic gates in linear optical quantum computing (LOQC) [16]. Large scale implementations of LOQC will require the propaga-

tion of multiphoton input states in reconfigurable interferometric networks [17]. In this context, experimental investigations of quantum interference in large interferometric networks become increasingly relevant and valuable.

The quantum walk (QW) [18, 19], the quantum mechanical analog of the random walk, provides an excellent platform for generating, manipulating, and observing quantum interference. Here, we use a photonic QW to manipulate and observe multiphoton interference for a variety of input states. Photonic QWs have been demonstrated experimentally, with interference in both the path and time degrees of freedom, in many different platforms, including bulk optics [20–33], fiber loops [34–41], fiber cavities [42], integrated photonics [43–48], and measurement-based approaches [49]. Nevertheless, certain limitations must be taken into account when selecting a platform to study multiphoton interference in a QW. The overall phase stability of the multi-mode interferometer must be high, the loss per step will necessarily be low, and each individual layer of the interferometric network should be readily programmable.

One promising QW platform [52] which addresses these concerns uses ultrafast time-bin encoding (UTBE) [32, 53–55], where the beam splitter action occurs in the time domain, on ultrafast timescales. Sub-picosecond-duration photons enter and exit the time-domain beam splitter in time-bin modes, separated by several picoseconds. These time bins can be achieved, manipulated, and detected using birefringent crystals and ultrashort pulses [56–58]; this enables operation in a single spatial mode, which offers high phase stability in transmission and compatibility with single-mode fibers. Direct measurement of the multi-mode time-bin signal at these time scales is not yet possible with fast photodetection and electronics; however, demultiplexing is possible with ultrafast optical Kerr gating [59].

In this work, we implement a QW in a single spatial mode, using UTBE, to investigate one-, two-, and

*                                        

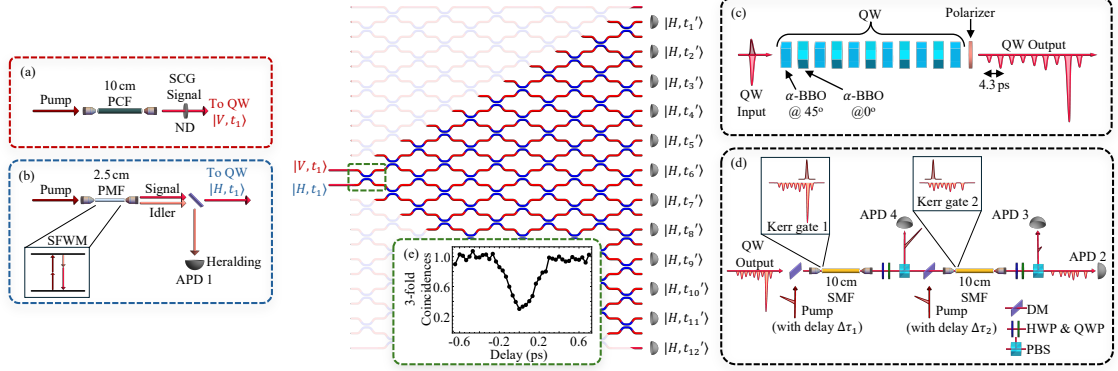


FIG. 1. **Simplified experimental setup.** Photons from (a) an attenuated coherent state and (b) a heralded single-photon source enter (c) the QW in modes $|V, t_1\rangle$ and $|H, t_1\rangle$, respectively. The analogous planar circuit for (c) is shown in the centre background. Shown in inset (a), the attenuated coherent state is achieved through supercontinuum generation (SCG) in a 10 cm photonic crystal fiber (PCF), which is spectrally filtered and attenuated to the single-photon level with neutral density (ND) filtering. Inset (b) depicts the heralded single-photon source, where photon pairs are generated through spontaneous four-wave mixing (SFWM) in 2.5 cm of polarization-maintaining fiber (PMF) [50, 51]. The idler photon ($\lambda_i = 954$ nm) is measured by the first avalanche photodiode (APD 1) and heralds the arrival of the signal photon ($\lambda_s = 682$ nm) at the output of the QW. Photons from these two sources traverse the N -layer QW ($N = 1 \dots 11$), where each layer consists of a birefringent α -BBO crystal, as seen in inset (c), with three rotational degrees of freedom. The multi-mode probability distribution across the possible horizontally polarized output states, $|H, t_m'\rangle$, is demultiplexed via optical Kerr gating, as shown in inset (d). In each optical Kerr gate, a strong pump pulse induces polarization rotation only where it overlaps in time with the QW output signal. Scanning the pump delay, $\Delta\tau_1$ and $\Delta\tau_2$, allows for readout of the entire horizontally polarized QW output state. Various coincidence patterns, measured by APDs 1–4 and processed using a time-to-digital converter, are analyzed to extract one-, two-, and three-photon interference. For example, detecting coincidences between APD 1, 2 and 4 (for a single interferometric layer, $N = 1$, and when both Kerr gates are turned off) allows us to measure the Hong-Ou-Mandel (HOM) two-photon interference between the two different input states, as seen in inset (e), where coincidence counts are normalized. We observe up to 70% HOM interference visibility between the two independent sources. DM: dichroic mirror, PBS: polarizing beam splitter, HWP: half-waveplate, QWP: quarter-waveplate, SMF: single mode fiber. See Fig. S1 in the Supplementary Materials for further experimental details.

three-photon interference. Optical Kerr gating, with efficiency $> 97\%$, is used to demultiplex the time-bin encoded probability distribution at the output of the QW. After demultiplexing, various click patterns at the detectors are recorded, allowing for the measurement of one-, two-, and three-photon probability distributions. Experimental measurements are compared to a simulation [60] of the QW, which uses the open source quantum optics simulation package, Strawberry Fields [61, 62]. Results demonstrate the versatility of the QW for generating, manipulating, and observing interference in a large interferometric network—a crucial capability in the context of photonic quantum information processing. Moreover, this experimental demonstration of multiphoton interference in a QW is consistent with requirements for quantum state generation [63], quantum state detection [64], and quantum simulation [65].

EXPERIMENTAL IMPLEMENTATION

The interferometric network, depicted in Fig. 1, is based on a QW scheme consisting of layers of beam split-

ters. In general, a QW involves some quantum particle (or particles) with two degrees of freedom, represented by a tensor product of two Hilbert spaces. One Hilbert space is used to describe the walker, while the other is used to describe a coin. At each beam splitter layer, the particle(s) will have some probability of exiting in each of the possible output walker states, depending on how the coin portion of its state is prepared. Here, the quantum particles traversing the QW are photons from either an attenuated ultrafast laser pulse, depicted in Fig. 1(a), or from a heralded single-photon source, depicted in Fig. 1(b). Ultrafast time bins form the basis states of the walker, $|t_m\rangle \in \{|t_1\rangle, |t_2\rangle, |t_3\rangle, \dots\}$, while the polarization state of each photon defines its coin, with basis states $|H\rangle$ and $|V\rangle$, corresponding to horizontal and vertical polarization, respectively. The state of each photon is given by a tensor product of basis states from each of the walker and coin Hilbert spaces such that there are no initial correlations or entanglement between the degrees of freedom.

Here, each beam splitter layer of the interferometric network corresponds to a single step of the photonic QW. The n^{th} step in the QW is described by the unitary step

operator,

$$\hat{U}_n = \hat{S}_n \cdot \hat{C}_n. \quad (1)$$

The shift operator, $\hat{S}_n$, raises or maintains the time-bin state of the photon, depending on its polarization state, and is given by

$$\hat{S}_n = \sum_m |H\rangle \langle H| \otimes |t_m\rangle \langle t_m| + |V\rangle \langle V| \otimes |t_{m+1}\rangle \langle t_m|. \quad (2)$$

The coin operator is given by

$$\hat{C}_n = \sum_m \left(\cos \frac{\Omega_n}{2} |H\rangle \langle H| + e^{i\gamma_n} \sin \frac{\Omega_n}{2} |H\rangle \langle V| + e^{-i\gamma_n} \sin \frac{\Omega_n}{2} |V\rangle \langle H| - \cos \frac{\Omega_n}{2} |V\rangle \langle V| \right) \otimes |t_m\rangle \langle t_m|, \quad (3)$$

where the parameters, Ω_n and γ_n , can be adjusted to tune the probability amplitude and phase, respectively, of the n^{th} step in the QW.

In this work, the unitary step operator in Eq. 1 is achieved using a single birefringent α -barium borate (α -BBO) crystal. With the proper anti-reflection coating, the loss through each crystal is < -0.045 dB. The temporal separation between ultrafast time bins is set by the thickness of the α -BBO crystals; here, a crystal thickness of 10 mm provides a temporal separation of ~ 4.3 ps at the signal wavelength. Each α -BBO crystal can be rotated along three axes, providing individual programmability of each layer in the QW. Rotating the n^{th} α -BBO crystal about the optical axis sets Ω_n , while adjusting its tip and tilt sets γ_n .

Shown in Fig. 1(c), the QW comprises a sequence of N birefringent α -BBO crystals, each performing one step of the walk, $\hat{U}_n$. The N -step QW can then be described by the unitary operator,

$$\hat{\mathcal{U}} = \prod_{n=1}^N \hat{U}_n, \quad (4)$$

which acts on an initial probability amplitude vector. When more than one photon is input to the walk, it becomes easier to consider the QW evolution using the second quantization formalism. Here, we can count the number of photons in the i^{th} mode, which is defined by a specific polarization (H or V) and time bin (t_m). In this picture, $\hat{\mathcal{U}}$ still provides all the information required to predict multiphoton evolution, but it now acts on each specific mode. For example, the case of a separable multiphoton input state with a total of K photons can be written as

$$|\psi_{\text{in}}\rangle = \prod_{i=1}^M \frac{1}{\sqrt{k_i!}} (\hat{a}_i^\dagger)^{k_i} |0\rangle, \quad (5)$$

where $|0\rangle$ is the vacuum state, $(\hat{a}_i^\dagger)$ is the creation operator for a photon in mode i , and the total number of photons satisfies $K = \sum_{i=1}^M k_i$. The QW acts on the multi-mode creation operator such that it maps $\hat{a}_i^\dagger \rightarrow \hat{\mathcal{U}} \hat{a}_i^\dagger \hat{\mathcal{U}}^\dagger$.

The total evolution of the N -step walk is then given by

$$|\psi_{\text{out}}^{(N)}\rangle = \prod_{i=1}^M \frac{1}{\sqrt{k_i!}} (\hat{\mathcal{U}} \hat{a}_i^\dagger \hat{\mathcal{U}}^\dagger)^{k_i} |0\rangle. \quad (6)$$

Multiphoton interference will influence the measured probability distribution for a given output state, $|\psi_{\text{out}}^{(N)}\rangle$. Since the time bins defining the position of each walker occur on ultrafast timescales, ultrafast optical gating is necessary for output signal demultiplexing. QWs based on UTBE have been demonstrated with upconversion schemes for detection [32, 53]; however, upconversion efficiency is often very low. Instead, we implement all-optical Kerr gating [59, 66], depicted in Fig. 1(d), for measurement. Kerr gating relies on a third-order ($\chi^{(3)}$) optical nonlinearity, by which a strong pulse of light is used to generate an intensity dependent refractive index modulation in the medium through which it propagates. This induces a birefringence, localized to the timing of the strong pulse, which can be used to rotate the polarization of a secondary optical signal, when the two are overlapped in the Kerr medium. Here, we use short (10 cm) pieces of single-mode fiber (SMF) as the Kerr medium. Each Kerr gate is operated with a high efficiency of $> 97\%$, which is crucial for observing multiphoton interference.

An ideal measurement would simultaneously detect the photon number content of each time bin at the output of the multiphoton QW [67]; however, this is not possible with available technologies. Instead, we are able to extract information by gating out time bins individually (*e.g.*, a one-fold measurement of time bin t_{m_1} at APD 3) and pair-wise (*e.g.*, a two-fold measurement of time bins t_{m_1} and t_{m_2} at APDs 3 and 4) with two Kerr gates. Time-resolved detection events in the gated time bins can also be measured in coincidence with detection events in the remaining time bins (*e.g.*, a three-fold measurement of time bins t_{m_1} , t_{m_2} , and $[t_{m_3}]$, at APDs 3, 4, and 2, where $[t_{m_3}]$ refers to all remaining time bins, $[t_{m_3}] = \{t_m \mid m \neq m_1 \wedge m \neq m_2\}$). We compare our measured detection probability patterns for heralded (conditional on an event at APD 1) and unheralded one, two, and three-fold events with those simulated using Strawberry Fields software.

RESULTS

In order to observe the effect of multiphoton quantum interference in the QW, we use two distinct categories of input state to the network. The signal mode from the two-mode squeezed vacuum (TMSV) source (Fig. 1(b)) is input to the $|H, t_1\rangle$ bin for all measurements. This acts as a nonclassical source of heralded single photons when APD 1 detects an idler photon. In the unheralded case, when all laser shots are included, the input is a single-mode thermal state, with mean photon number $\mu_\xi \approx 0.026$. Either of these two input states is in-

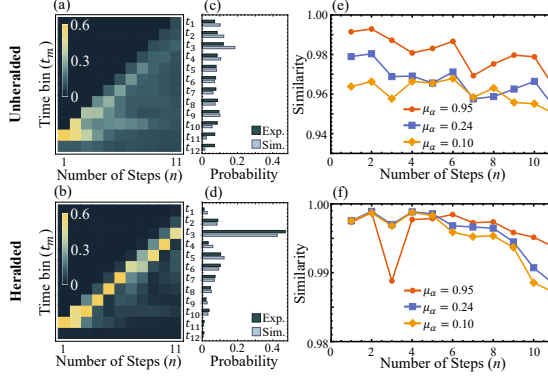


FIG. 2. **One-fold measurements.** Unheralded and heralded one-fold measurements presented in the top and bottom rows, respectively. The step-by-step QW output evolution (for $\mu_\alpha = 0.1$) is plotted in the first column, (a) and (b), with the corresponding 11-step probability distribution shown in the middle column, (c) and (d). Experimental data is compared with a simulation through a similarity calculation, plotted in the last column, (e) and (f), for the three different values of μ_α .

put to the QW along with an attenuated coherent state (ACS), with mean photon number μ_α , always input to $|V, t_1\rangle$. We measure the output QW probability distribution across the possible horizontally polarized output time bins and examine one-, two-, and three-fold coincidences for both the heralded (single photon input) and unheralded (thermal state input) cases. See Table S1 in the Supplementary Materials for details on the various coincidence patterns considered. Data is collected for three different values of μ_α in order to observe its effects.

Quantum interference requires that the photons in the walk are indistinguishable (*i.e.*, ideally, with perfect mode overlap). We quantify this by measuring Hong-Ou-Mandel (HOM) interference between the ACS and the heralded single photon, for a single QW layer. For a given squeezing parameter of the heralded single-photon source, the HOM visibility will also depend on the mean photon number of the ACS [68, 69]. We measure a maximum HOM visibility of 70%, which is used as an estimate for the mode overlap between the two input sources in the Strawberry Fields simulations.

We first consider the heralded and unheralded one-fold measurements of the QW output, as shown in Fig. 2. Here, only one Kerr gate and APD 4 is required to make the measurement. The unheralded and heralded distributions are shown in Figs. 2(a) and (b), respectively. The QW output after 11 steps is plotted in Figs. 2(c) and (d), next to the corresponding step-by-step QW evolution from (a) and (b), respectively. Here, experimental data is plotted next to the Strawberry Fields simulation [60], for the corresponding photonic circuit. Each step-by-step QW output probability distribution is mea-

sured for three values of μ_α , and a similarity calculation between experiment and simulation is made using the Bhattacharyya coefficient. Similarity calculations are presented in Figs. 2(c) and (d), and correspond to the data presented in (a) and (b), respectively. In all data presented, the similarity remains above 95%. The qualitative difference between the heralded and unheralded distributions is determined by the relative brightness of the light at the two input ports [52]. For the unheralded case, the output distribution is dominated by the ACS V -polarized input ($\mu_\alpha \gg \mu_\xi$); for the heralded case, the output distribution is dominated by the H -polarized single photon input ($\mu_\alpha \ll 1$).

Next, heralded and unheralded two-fold measurements are presented in Fig. 3. Here, two Kerr gates and APDs 3 and 4 are required to measure the complete probability distribution. The unheralded and heralded two-fold measurements are presented in the top and bottom rows, respectively. Measurements are taken for $\mu_\alpha = 0.1, 0.24$, and 0.95 . Each experimental dataset is compared with the simulation, through a similarity calculation, as shown in the respective insets in Fig. 3. This simulation propagates an ACS and the signal mode of a TMSV state through N steps of the QW unitary in Eq. 1. Details on the simulation are provided in the Supplementary Materials (see Fig. S5).

We see that the probability distributions in Figs. 3(d)-(f), for which the two-fold measurements are heralded, show stronger clustering than the distributions in Figs. 3(a)-(c), for which the two-fold measurements are not heralded. For example, the largest probabilities measured in (d), (e), and (f) are 3.0, 2.4, and 1.9 times larger than the largest probabilities measured in (a), (b), and (c), respectively. The probability distributions in (d)-(f) also show detection outcomes which are more suppressed overall. Both of these effects are signatures of bosonic quantum interference, caused by non-classical input states. Another way to investigate this nonclassicality is by testing the accuracy of an optimized classical model. This model simulates the experiment using a two-mode squeezed thermal (TMST) source, also referred to as a “squashed state”, rather than the TMSV source. For low enough squeezing, the TMST source provides the closest classical approximation to a photon pair source after loss [70]. We find significantly better similarities when experimental data is compared with the quantum simulation using a TMSV source, rather than a TMST source. See Fig. S6 in the Supplementary Materials for details on this comparison.

Finally, we present heralded and unheralded three-fold measurements in Fig. 4. Due to the fact that only two Kerr gates are implemented in this work, we are limited to measurements where one detector measures an event in time bin t_{m_1} , another detector measures an event in time bin t_{m_2} , and the remaining detector measures an event in time bin $[t_{m_3}]$, where $[t_{m_3}] = \{t_m \mid m \neq m_1 \wedge m \neq m_2\}$. This approach is possible only because the Kerr gates are operated with such high efficiency, ensuring that photons

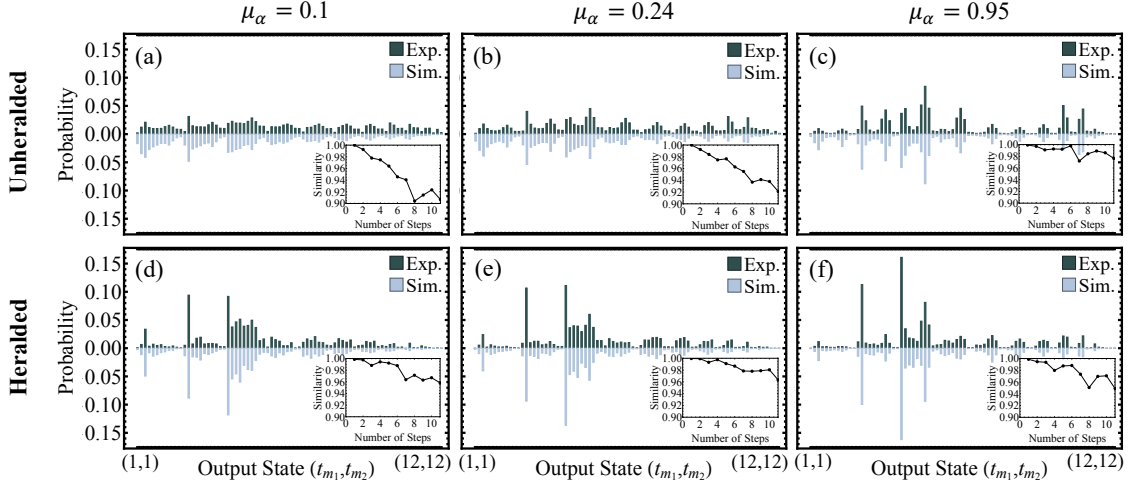


FIG. 3. **Two-fold measurements.** Unheralded and heralded measurements of the QW output distributions from the 11-step QW are shown in the top, (a)-(c), and bottom, (d)-(f), rows, respectively. Data is collected for $\mu_\alpha = 0.1$ (first column), 0.24 (second column), and 0.95 (third column). The step-by-step similarities between each experimental dataset and its corresponding simulation are shown in the respective insets.

in time bins t_{m_1} and t_{m_2} have a very low probability of arriving at the detector which measures $[t_{m_3}]$. See Fig. S7 for a depiction of this experimental configuration. We note here that with a third Kerr gate, the full three-fold probability distribution could be measured directly. Nevertheless, we present a subset of the unheralded and heralded three-fold measurements at the output of a 9-step QW in Fig. 4(a) and (b), respectively. The similarities between experiment and simulation are 95% and 89% for the unheralded and heralded three-fold measurements, respectively. We note that all measurements in Fig. 4 correspond to a value of $\mu_\alpha = 0.95$, which provides the highest coincidence rates. A similarity calculation for the step-by-step evolution of the three-fold measurements is provided in Fig. S8.

DISCUSSION AND OUTLOOK

The QW model, which is essentially a large, multi-mode interferometric network, provides a convenient testbed for interference-based quantum platforms (*e.g.*, Gaussian boson sampling [71]), for example in quantum information processing and quantum metrology. By implementing a QW here, we are able to demonstrate the potential of UTBE for the generation, manipulation, and measurement of multi-mode interference between up to three photons in a single spatial mode. This is a key step towards scalable quantum information processing platforms, which require high-dimensional multi-mode interference of many photons in order to push beyond the current limitations of classical computation, entering the

realm of “useful” quantum computation.

In practice, many-photon operation still poses a large challenge. One of the main limitations for multi-photon operation in many-mode interferometric networks is that the loss scales with the circuit depth. As such, achieving low loss per circuit depth, as demonstrated here, is crucial to scale these photonic circuits to the multi-photon regime. Single-photon sources pose another limitation to the scalability of photonic quantum information processing schemes. Deterministic sources, such as quantum dots [72] or defect centers [73] are promising for on-demand single photons with high rates; however, achieving high interference visibility is still difficult. Probabilistic sources, generated through nonlinear processes such as the SFWM source in this work or spontaneous parametric downconversion [14] can be used to achieve high interference visibility, but due to their probabilistic nature pose constraints on count rates; however, with photon number resolving detectors, probabilistic sources can be operated at higher rates, making them a promising avenue for photonic quantum information processing [74]. This would be compatible with continuous-variable (CV) quantum information processing schemes, which consist of deterministic sources of squeezed light [75, 76].

Not only useful for information processing, quantum interference holds implications for metrology where the large interferometric network presented here could be extended for parameter estimation [77-79]. Most proposals of parameter estimation with QWs consider the case where the parameters of the walk itself are being estimated. This is useful in the context of implementing QWs with low experimental error; parameter estimation

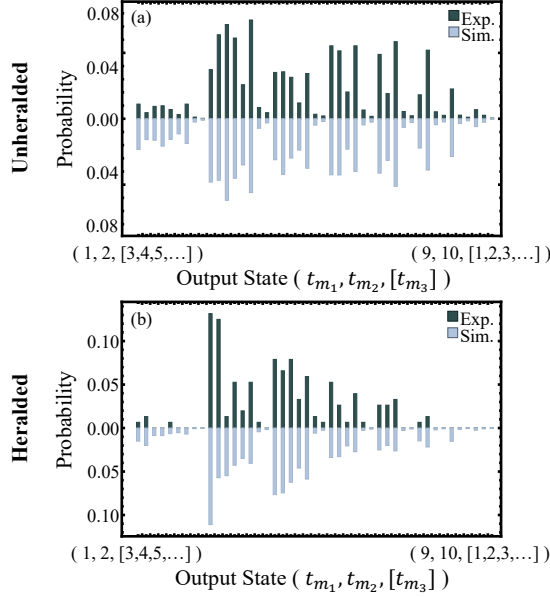


FIG. 4. **Partial three-fold measurements.** Output distributions from a 9-step QW are shown when three-fold measurements are (a) unheralded and (b) heralded. Data is shown for $\mu_\alpha = 0.95$. Experimental data is compared to simulation, with similarities of (a) 95% and (b) 89%, respectively.

could be used to determine which, if any, of the many QW parameters require adjustment. Parameter estimation in a QW may also be extended to other applications including, for example, the analysis of gravitational wave measurements [80].

With the demonstrated ability to handle interference of up to three photons across 24 modes, our QW with UTBE holds potential in interference-based quantum platforms. We anticipate that the QW will be a useful tool in quantum metrology, and, with improvement to multiphoton event detection rates, will be capable of manipulating interference between > 3 photons, providing a pathway for scalable photonic quantum information processing.

ACKNOWLEDGMENTS

This work is supported by the Vanier Canada Graduate Scholarships program. We thank Alicia Sit, Andrew Proppe, Brayden Freitas, Denis Guay, Doug Moffatt, Milica Banic, Nicolas Couture, Noah Lupu-Gladstein, Ramy Tannous, Rune Lausten, and Yingwen Zhang for their support and insightful discussions.

-
- [1] H. L. Helmholtz, *On the Sensations of Tone as a Physiological Basis for the Theory of Music* (Cambridge University Press, 2009).
 - [2] J. J. Dammerud, The Journal of the Acoustical Society of America **128**, 506 (2010).
 - [3] B. P. Abbott, R. Abbott, T. Abbott, M. Abernathy, F. Acernese, K. Ackley, C. Adams, T. Adams, P. Addesso, R. X. Adhikari, *et al.*, Physical review letters **116**, 061102 (2016).
 - [4] D. Stahlke, Physical Review A **90**, 022302 (2014).
 - [5] T. Graham, Y. Song, J. Scott, C. Poole, L. Phuttitarn, K. Jooya, P. Eichler, X. Jiang, A. Marra, B. Grinkemeyer, *et al.*, Nature **604**, 457 (2022).
 - [6] J. I. Cirac and P. Zoller, Nature **404**, 579 (2000).
 - [7] D. Kielpinski, C. Monroe, and D. J. Wineland, Nature **417**, 709 (2002).
 - [8] J. A. Jones, V. Vedral, A. Ekert, and G. Castagnoli, Nature **403**, 869 (2000).
 - [9] J. M. Gambetta, J. M. Chow, and M. Steffen, npj quantum information **3**, 2 (2017).
 - [10] F. Arute, K. Arya, R. Babbush, D. Bacon, J. C. Bardin, R. Barends, R. Biswas, S. Boixo, F. G. Brandao, D. A. Buell, *et al.*, Nature **574**, 505 (2019).
 - [11] H.-S. Zhong, H. Wang, Y.-H. Deng, M.-C. Chen, L.-C. Peng, Y.-H. Luo, J. Qin, D. Wu, X. Ding, Y. Hu, *et al.*, Science **370**, 1460 (2020).
 - [12] L. S. Madsen, F. Laudenbach, M. F. Askarani, F. Rortais, T. Vincent, J. F. Bulmer, F. M. Miatto, L. Neuhaus, L. G. Helt, M. J. Collins, *et al.*, Nature **606**, 75 (2022).
 - [13] A. A. Michelson and E. W. Morley, American journal of science **3**, 333 (1887).
 - [14] C.-K. Hong, Z.-Y. Ou, and L. Mandel, Physical review letters **59**, 2044 (1987).
 - [15] F. Bouchard, A. Sit, Y. Zhang, R. Fickler, F. M. Miatto, Y. Yao, F. Sciarrino, and E. Karimi, Reports on Progress in Physics **84**, 012402 (2020).
 - [16] E. Knill, R. Laflamme, and G. J. Milburn, nature **409**, 46 (2001).
 - [17] L. Carosini, V. Oddi, F. Giorgino, L. M. Hansen, B. Seron, S. Piacentini, T. Guggemos, I. Agresti, J. C. Lored, and P. Walther, Science Advances **10**, ead90993 (2024).
 - [18] Y. Aharonov, L. Davidovich, and N. Zagury, Physical Review A **48**, 1687 (1993).
 - [19] J. Kempe, Contemporary Physics **44**, 307 (2003).
 - [20] B. Do, M. L. Stohler, S. Balasubramanian, D. S. Elliott, C. Eash, E. Fischbach, M. A. Fischbach, A. Mills, and B. Zwickl, JOSA B **22**, 499 (2005).
 - [21] D. Bouwmeester, I. Marzoli, G. P. Karman, W. Schleich, and J. Woerdman, Physical Review A **61**, 013410 (1999).
 - [22] M. A. Broome, A. Fedrizzi, B. P. Lanyon, I. Kassal, A. Aspuru-Guzik, and A. G. White, Physical Review Letters **104**, 153602 (2010).

- [23] T. Kitagawa, M. A. Broome, A. Fedrizzi, M. S. Rudner, E. Berg, I. Kassal, A. Aspuru-Guzik, E. Demler, and A. G. White, *Nature Communications* **3**, 1 (2012).
- [24] F. Cardano, F. Massa, H. Qassim, E. Karimi, S. Slusarenko, D. Paparo, C. de Lisio, F. Sciarrino, E. Santamato, R. W. Boyd, *et al.*, *Science Advances* **1**, e1500087 (2015).
- [25] F. Cardano, M. Maffei, F. Massa, B. Piccirillo, C. De Lisio, G. De Filippis, V. Cataudella, E. Santamato, and L. Marrucci, *Nature Communications* **7**, 11439 (2016).
- [26] F. Cardano, A. D’Errico, A. Dauphin, M. Maffei, B. Piccirillo, C. de Lisio, G. De Filippis, V. Cataudella, E. Santamato, L. Marrucci, *et al.*, *Nature Communications* **8**, 15516 (2017).
- [27] F. Nejadsattari, Y. Zhang, F. Bouchard, H. Larocque, A. Sit, E. Cohen, R. Fickler, and E. Karimi, *Optica* **6**, 174 (2019).
- [28] A. Gherardi, A. Laneve, L. D. Bonavena, L. Sansoni, J. Ferraz, A. Fratalocchi, F. Sciarrino, Á. Cuevas, and P. Mataloni, *Physical Review Letters* **123**, 140501 (2019).
- [29] A. D’Errico, F. Cardano, M. Maffei, A. Dauphin, R. Barboza, C. Esposito, B. Piccirillo, M. Lewenstein, P. Massignan, and L. Marrucci, *Optica* **7**, 108 (2020).
- [30] C. Esposito, M. R. Barros, A. Durán Hernández, G. Carvacho, F. Di Colandrea, R. Barboza, F. Cardano, N. Spagnolo, L. Marrucci, and F. Sciarrino, *npj Quantum Information* **8**, 34 (2022).
- [31] F. Di Colandrea, A. Babazadeh, A. Dauphin, P. Massignan, L. Marrucci, and F. Cardano, *Optica* **10**, 324 (2023).
- [32] Q.-Q. Wang, S. Dong, X.-W. Li, X.-Y. Xu, C. Wang, S. Han, M.-H. Yung, Y.-J. Han, C.-F. Li, and G.-C. Guo, *Science Advances* **10**, ead14871 (2024).
- [33] F. Di Colandrea, N. Dehghan, F. Cardano, A. D’Errico, and E. Karimi, *Communications Physics* **7**, 265 (2024).
- [34] A. Schreiber, K. N. Cassemiro, V. Potoček, A. Gábris, P. J. Mosley, E. Andersson, I. Jex, and C. Silberhorn, *Physical Review Letters* **104**, 050502 (2010).
- [35] A. Schreiber, A. Gábris, P. P. Rohde, K. Laiho, M. Štefaňák, V. Potoček, C. Hamilton, I. Jex, and C. Silberhorn, *Science* **336**, 55 (2012).
- [36] Y.-C. Jeong, C. Di Franco, H.-T. Lim, M. Kim, and Y.-H. Kim, *Nature Communications* **4**, 1 (2013).
- [37] L. Lorz, E. Meyer-Scott, T. Nitsche, V. Potoček, A. Gábris, S. Barkhofen, I. Jex, and C. Silberhorn, *Physical Review Research* **1**, 033036 (2019).
- [38] A. Gherardi, S. De, A. Laneve, S. Barkhofen, J. Sperling, P. Mataloni, and C. Silberhorn, *Physical Review Research* **3**, 023052 (2021).
- [39] D. Bagrets, K. W. Kim, S. Barkhofen, S. De, J. Sperling, C. Silberhorn, A. Altland, and T. Micklitz, *Physical Review Research* **3**, 023183 (2021).
- [40] P. Held, M. Engelkemeier, S. De, S. Barkhofen, J. Sperling, and C. Silberhorn, *Physical Review A* **105**, 042210 (2022).
- [41] F. Pegoraro, P. Held, S. Barkhofen, B. Brecht, and C. Silberhorn, *Physica Scripta* **98**, 034005 (2023).
- [42] J. Boutari, A. Feizpour, S. Barz, C. Di Franco, M. Kim, W. Kolthammer, and I. Walmsley, *Journal of Optics* **18**, 094007 (2016).
- [43] A. Peruzzo, M. Lobino, J. C. Matthews, N. Matsuda, A. Politi, K. Poulios, X.-Q. Zhou, Y. Lahini, N. Ismail, K. Wörhoff, *et al.*, *Science* **329**, 1500 (2010).
- [44] L. Sansoni, F. Sciarrino, G. Vallone, P. Mataloni, A. Crespi, R. Ramponi, and R. Osellame, *Physical Review Letters* **108**, 010502 (2012).
- [45] N. C. Harris, G. R. Steinbrecher, M. Prabhu, Y. Lahini, J. Mower, D. Bunandar, C. Chen, F. N. Wong, T. Baehr-Jones, M. Hochberg, *et al.*, *Nature Photonics* **11**, 447 (2017).
- [46] I. Pitsios, L. Banchi, A. S. Rab, M. Bentivegna, D. Caprara, A. Crespi, N. Spagnolo, S. Bose, P. Mataloni, R. Osellame, *et al.*, *Nature Communications* **8**, 1569 (2017).
- [47] T. Meany, M. Gräfe, R. Heilmann, A. Perez-Leija, S. Gross, M. J. Steel, M. J. Withford, and A. Szameit, *Laser & Photonics Reviews* **9**, 363 (2015).
- [48] M. Gräfe, R. Heilmann, M. Lebugle, D. Guzman-Silva, A. Perez-Leija, and A. Szameit, *Journal of Optics* **18**, 103002 (2016).
- [49] S. De, V. Ansari, J. Sperling, S. Barkhofen, B. Brecht, and C. Silberhorn, *Physical Review Research* **6**, L022040 (2024).
- [50] B. J. Smith, P. Mahou, O. Cohen, J. Lundeen, and I. Walmsley, *Optics express* **17**, 23589 (2009).
- [51] J. Erskine, D. England, C. Kupchak, and B. Sussman, *Optics letters* **43**, 907 (2018).
- [52] K. L. Fenwick, F. Bouchard, G. S. Thekkadath, D. England, P. J. Bustard, K. Heshami, and B. Sussman, *Optica* **11**, 1017 (2024).
- [53] X.-Y. Xu, Q.-Q. Wang, W.-W. Pan, K. Sun, J.-S. Xu, G. Chen, J.-S. Tang, M. Gong, Y.-J. Han, C.-F. Li, *et al.*, *Physical Review Letters* **120**, 260501 (2018).
- [54] F. Bouchard, K. Bonsma-Fisher, K. Heshami, P. J. Bustard, D. England, and B. Sussman, *Physical Review A* **107**, 022618 (2023).
- [55] F. Bouchard, K. Fenwick, K. Bonsma-Fisher, D. England, P. J. Bustard, K. Heshami, and B. Sussman, *arXiv preprint arXiv:2404.17657* (2024).
- [56] C. Kupchak, P. J. Bustard, K. Heshami, J. Erskine, M. Spanner, D. G. England, and B. J. Sussman, *Physical Review A* **96**, 053812 (2017).
- [57] F. Bouchard, D. England, P. J. Bustard, K. Heshami, and B. Sussman, *PRX Quantum* **3**, 010332 (2022).
- [58] J. M. Donohue, M. Agnew, J. Lavoie, and K. J. Resch, *Physical Review Letters* **111**, 153602 (2013).
- [59] D. England, F. Bouchard, K. Fenwick, K. Bonsma-Fisher, Y. Zhang, P. J. Bustard, and B. J. Sussman, *Applied Physics Letters* **119**, 160501 (2021).
- [60] G. Thekkadath, K. Fenwick, F. Bouchard, D. England, P. Bustard, K. Heshami, and B. Sussman, “Utbequantumwalk,” <https://github.com/tguillaume/UTBEQuantumWalk> (2024).
- [61] N. Killoran, J. Izaac, N. Quesada, V. Bergholm, M. Amy, and C. Weedbrook, *Quantum* **3**, 129 (2019).
- [62] T. R. Bromley, J. M. Arrazola, S. Jahangiri, J. Izaac, N. Quesada, A. D. Gran, M. Schuld, J. Swinerton, Z. Zabaneh, and N. Killoran, *Quantum Science and Technology* **5**, 034010 (2020).
- [63] S. Konno, W. Asavanant, F. Hanamura, H. Nagayoshi, K. Fukui, A. Sakaguchi, R. Ide, F. China, M. Yabuno, S. Miki, *et al.*, *Science* **383**, 289 (2024).
- [64] L. Pezzé, A. Smerzi, G. Khoury, J. Hodelin, and D. Bouwmeester, *Physical review letters* **99**, 223602 (2007).
- [65] F. H. Somhorst, R. van der Meer, M. Correa Anguita, R. Schadow, H. J. Snijders, M. de Goede, B. Kassenberg,

- P. Venderbosch, C. Taballione, J. Epping, *et al.*, Nature communications **14**, 3895 (2023).
- [66] C. Kupchak, J. Erskine, D. England, and B. Sussman, Optics letters **44**, 1427 (2019).
- [67] A. Z. Goldberg, Optica Quantum **2**, 14 (2024).
- [68] J. Rarity, P. Tapster, and R. Loudon, Journal of Optics B: Quantum and Semiclassical Optics **7**, S171 (2005).
- [69] A. Xu, L. Duan, L. Wang, and Y. Zhang, Optics Express **31**, 5662 (2023).
- [70] H. Qi, D. J. Brod, N. Quesada, and R. García-Patrón, Physical review letters **124**, 100502 (2020).
- [71] G. Thekkadath, S. Sempere-Llagostera, B. Bell, R. Patel, M. Kim, and I. Walmsley, PRX Quantum **3**, 020336 (2022).
- [72] P. Laferrière, E. Yeung, I. Miron, D. B. Northeast, S. Haffouz, J. Lapointe, M. Korkusinski, P. J. Poole, R. L. Williams, and D. Dalacu, Scientific Reports **12**, 6376 (2022).
- [73] N. Mizuochi, T. Makino, H. Kato, D. Takeuchi, M. Ogura, H. Okushi, M. Nothaft, P. Neumann, A. Gali, F. Jelezko, *et al.*, Nature photonics **6**, 299 (2012).
- [74] Y.-H. Deng, Y.-C. Gu, H.-L. Liu, S.-Q. Gong, H. Su, Z.-J. Zhang, H.-Y. Tang, M.-H. Jia, J.-M. Xu, M.-C. Chen, *et al.*, Physical review letters **131**, 150601 (2023).
- [75] U. L. Andersen, G. Leuchs, and C. Silberhorn, Laser & Photonics Reviews **4**, 337 (2010).
- [76] H. Yonezawa and A. Furusawa, Optics and Spectroscopy **108**, 288 (2010).
- [77] D. Ellinas, P. D. Jarvis, and M. Pearce, Physica Scripta **99**, 025107 (2024).
- [78] P. Rajauria, P. Chawla, and C. Chandrashekar, arXiv preprint [arXiv:2007.04572](https://arxiv.org/abs/2007.04572) (2020).
- [79] M. Annabestani, M. Hassani, D. Tamascelli, and M. G. Paris, Physical Review A **105**, 062411 (2022).
- [80] G. Escrig, R. Campos, P. A. Casares, and M. Martin-Delgado, Classical and Quantum Gravity **40**, 045001 (2023).

Supplementary Material: Multiphoton interference in a single-spatial-mode quantum walk

Kate L. Fenwick,^{1,2} Jonathan Baker,¹ Guillaume S. Thekkadath,² Aaron Z. Goldberg,² Khabat Heshami,^{1,2} Philip J. Bustard,² Duncan England,² Frédéric Bouchard,^{2,*} and Benjamin Sussman^{1,2}

¹*Department of Physics, University of Ottawa, Advanced Research Complex,
25 Templeton Street, Ottawa ON Canada, K1N 6N5*

²*National Research Council of Canada, 100 Sussex Drive, Ottawa, Ontario K1A 0R6, Canada*

1. EXPERIMENTAL DETAILS

The full experimental setup for the multiphoton quantum walk (QW) is shown in Fig. S1. The output pulses (80 MHz repetition rate, central wavelength: $\lambda_p = 795$ nm, bandwidth: $\Delta\lambda = 11.4$ nm) from the titanium sapphire (Ti:Sapph) laser are first sent through an optical isolator to avoid unwanted backreflected light re-entering the laser cavity. After the optical isolator, a prism compressor provides dispersion compensation, which is necessary for stable operation of supercontinuum generation (SCG) in the photonic crystal fiber (PCF). The pulses are then split into two paths by a HWP and a polarizing beam splitter (PBS), which provide control over the pulse energy for both QW input state generation and optical Kerr gating.

A. Input state generation

Pump pulses from the Ti:Sapph are further split into two paths by a HWP and PBS for the pumping of two independent photon sources which will provide the input state to the QW. Spontaneous four-wave mixing (SFWM) in polarization-maintaining fiber (PMF) is used to generate heralded single photons, while SCG in PCF is attenuated to the single-photon level to provide an attenuated coherent state. Photons from the attenuated coherent state and heralded single-photon source are recombined on a PBS and then spectrally filtered to a bandwidth of ~ 3 nm before entering the QW.

Heralded single-photon source

SFWM in a 2.5 cm PMF produces photon pairs where the central wavelengths of the herald and signal photons are $\lambda_i = 954$ nm and $\lambda_s = 682$ nm, respectively. The idler photon is detected at the first avalanche photodiode (APD 1), which heralds the detection of the signal photon after traversing the QW. The pulses pumping the SFWM process are spectrally filtered to a bandwidth of about 4 nm, so as to achieve a joint spectral intensity (JSI) [1] as round as possible, and have energy ~ 1.25 nJ.

Attenuated coherent state generation

SCG in a 12 cm PCF generates signal pulses [2]. When pumped with pulses from the Ti:Sapph laser, the PCF (NKT Photonics, FemtoWHITE CARS) generates a dual peak spectrum which is tunable with input pulse energy. As the pump pulse energy is increased, the two SCG peaks shift further away from the central wavelength of the pump. In the sub-nJ pump pulse energy regime, SCG in the PCF provides access to a signal wavelength range of 600 to 1100 nm. Pump pulses with energy ~ 0.18 nJ generate a spectrum with one of the dual peaks centered at 682 nm. Neutral density (ND) filters are then used to attenuate the signal pulses to single-photon-level, after which a HWP is used for continuous tuning of the mean photon number of the coherent state, μ_α , prior to its recombination with the heralded single photon at the PBS. The single-photon-level pulses are sent through an optical delay line, $\Delta\tau_{\text{HOM}}$, which allows for measurement of Hong-Ou-Mandel (HOM) two-photon interference [3] between the attenuated coherent state and heralded single-photon source. After a HOM interference measurement is made, $\Delta\tau_{\text{HOM}}$ can be set at the optimal position to maximize quantum interference between the two sources within the QW.

B. Quantum walk (QW)

Photons from the heralded single-photon source enter the quantum walk in the $|H, t_1\rangle$ mode, while photons from the attenuated coherent state enter the walk in the $|V, t_1\rangle$ mode. Photons which traverse the QW will exit in one of the 24 output modes, $|V, t'_1\rangle, |H, t'_1\rangle, \dots, |V, t'_{12}\rangle, |H, t'_{12}\rangle$. We elect to measure only the H-polarized output modes for simplicity, which is done by sending the QW output signal through a nanoparticle linear film polarizer (N-LFP). The horizontally polarized (H-polarized) QW output is then sent into the two Kerr gates for demultiplexing and measurement.

C. Demultiplexing via optical Kerr gating

The QW output signal is a train of time bins, separated by about 4.3 ps, each of which has a horizontally-polarized and vertically-polarized component. Demulti-

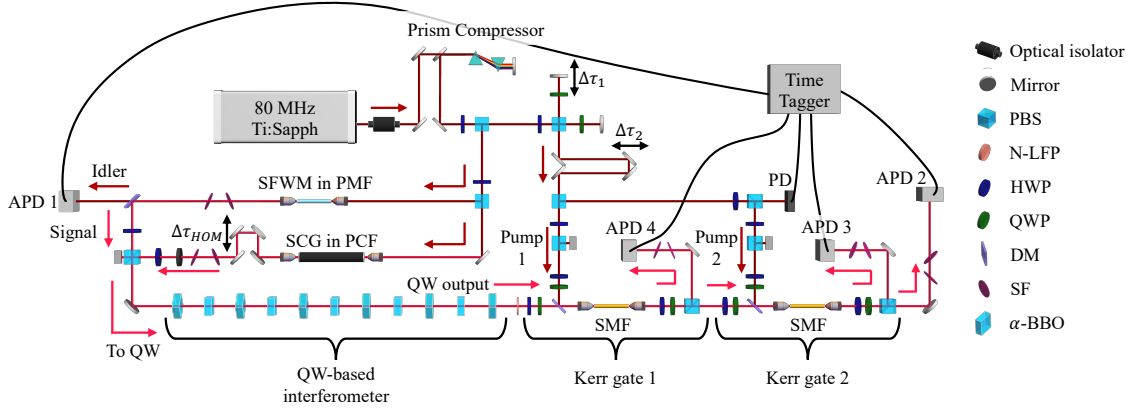


FIG. S1. **Detailed experimental setup of the multiphoton QW.** Ti:Sapph: titanium sapphire; SFWM: spontaneous four-wave mixing; PMF: polarization-maintaining fiber; SCG: supercontinuum generation; PCF: photonic crystal fiber; $\Delta\tau_{\text{HOM}}$: optical delay for Hong-Ou-Mandel measurement and interference optimization; QW: quantum walk; $\Delta\tau_1$: optical delay of pump pulse 1; $\Delta\tau_2$: optical delay of pump pulse 2; SMF: single-mode fiber; PBS: polarizing beam splitter, N-LFP: nanoparticle linear film polarizer; HWP: half-waveplate, QWP: quarter-waveplate, DM: dichroic mirror; SF: spectral filter; α -BBO: α -barium borate crystal; PD: photodiode; APD: avalanche photodiode.

plexing in the time domain is required so that specific time bins can be selected out from the train of time bins for measurement. This is achieved via optical Kerr gating, with $> 97\%$ internal efficiency in Kerr gates 1 and 2, in short 10 cm pieces of SMF (Thorlabs, 780-HP). We note here that although the signal wavelength is below the single-mode cutoff for this particular type of SMF, we are still able to operate with both the pump and signal in the single-mode regime, due to the fact that the short SMF is not bent.

The internal efficiency of each Kerr gate, η , is given by [4]

$$\eta(T) = \sin^2(2\theta) \sin^2\left(\frac{\Delta\phi(T)}{2}\right), \quad (1)$$

where θ is the angle between the signal and pump polarization, and $\Delta\phi(T)$ is the induced nonlinear birefringence in the SMF due to the presence of the pump pulse. Gating efficiency is maximized when $\theta = \pi/4$ and $\Delta\phi = \pi$. The nonlinear phase shift is given by [5]

$$\Delta\phi(T) = \frac{8\pi n_2}{3\lambda_s} \int_0^L I_p(T - d_w z) dz, \quad (2)$$

where n_2 is the nonlinear refractive index of the SMF, λ_s is the signal wavelength, and z is the propagation distance within the SMF of length L . The pump and signal will experience temporal walkoff in each Kerr gate, $d_w = v_{gp}^{-1} - v_{gs}^{-1}$, where v_{gp} and v_{gs} are the group velocities of the pump and signal pulses, respectively. The intensity profile of the pump, I_p is expressed in a frame that moves with the signal [6], $T = t - z/v_{gs}$, where t is the time in the laboratory frame.

The output signal from the QW is recombined with pump pulses 1 and 2 from the Ti:Sapph on dichroic mirrors (DMs) for demultiplexing in Kerr gates 1 and 2, respectively. Prior to the DMs, the pump pulses 1 and 2 are each prepared as followed. First, the pump pulses enter a Michelson interferometer in which one arm has variable delay, $\Delta\tau_1$, to control the relative delay between pump pulses 1 and 2. The absolute delay of both pump pulses, relative to the QW output signal, is controlled by the delay, $\Delta\tau_2$. Scanning $\Delta\tau_1$ and $\Delta\tau_2$ allows for demultiplexing of the ultrafast time-bin output signal from the QW. The energy of both pump pulses is first tuned, using a HWP and PBS combination, to maximize the gating efficiency. A PBS, QWP, and HWP are then used to prepare the polarization of each pump pulse (45° relative to the signal polarization at the 10 cm single-mode fiber (SMF) input) in order to maximize gate efficiency. The QW output signal is coupled to the SMF in Kerr gates 1 and 2 with efficiencies of 75% and 85%, respectively, while pump pulses 1 and 2 are coupled to Kerr gates 1 and 2 with efficiencies of 80% and 70%, respectively. The signal coupling efficiency to the second Kerr gate is accounted for in measurements by dividing the total events measured after this Kerr gate by 0.85.

After each Kerr gate, a QWP and HWP compensate for any intrinsic polarization rotation or birefringence within the 10 cm SMF. A PBS then splits the gated and non-gated QW signal, where the gated signal is sent to an APD for detection. Prior to each APD, spectral filters attenuate the pump and any noise generated by the pump within the 10 cm SMF. The non-gated signal is also sent to an APD for detection. All APDs are connected to a time-to-digital converter (TimeTagger Ultra,

Swabian Instruments) for one-, two-, and three-fold measurements. Section 4 discusses the different Kerr gate demultiplexing configurations required for measurement. The transmission efficiency of photons through the full experimental system, from the QW input through to detection, is $\sim 8\%$.

2. CHARACTERIZATION OF THE HERALDED SINGLE-PHOTON SOURCE

We first characterize the heralded single-photon source, before it is propagated through the QW. To do so, we use a flip mirror (after the DM used to split the signal (s) and idler (i) photons, not depicted in Fig. S1) to couple the signal photons directly to a 50:50 fiber beam splitter which then directs the two signal photon paths to two APDs (s_1 and s_2). Shown in Fig. S2(a), the second order auto-correlation between the two signal photon paths conditioned on the arrival of an idler photon, $g_{s_1, s_2|i}^{(2)}(0)$, clearly demonstrates the nonclassicality of the

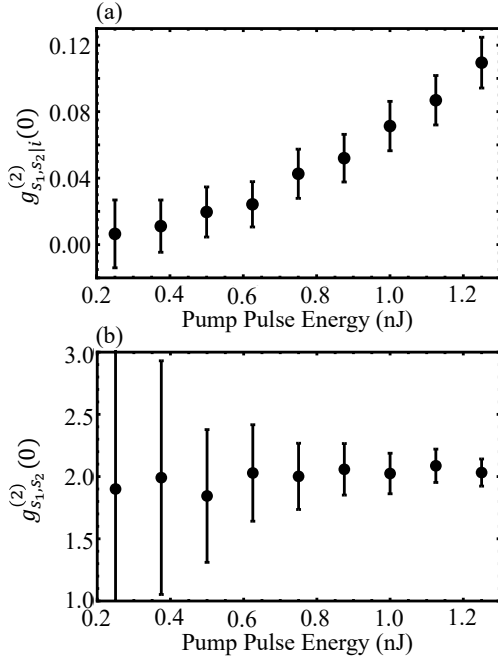


FIG. S2. **Second order auto-correlation function of heralded single-photon source.** The second order auto-correlation of the signal photons from the heralded single-photon source (a) conditioned on the detection of an idler photon and (b) not conditioned on the detection of an idler photon, as a function of increasing pulse energy used to pump the SFWM process is shown. Note that we pump the source with ~ 1.25 nJ pulses for all measurements.

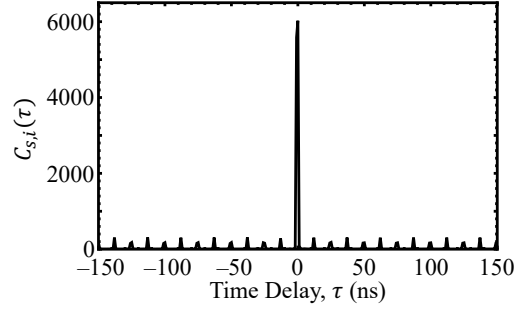


FIG. S3. **Coincidence rate between signal and idler photons.** The delay-dependent rate of coincidences between the signal and idler photons, $C_{s,i}(\tau)$, from the photon pair source is shown for pump pulses with energy ~ 1.25 nJ. The coincidence-to-accidental (CAR) ratio extracted from this measurement is ~ 35 .

photon pairs produced through SFWM ($g_{s_1, s_2}^{(2)}(0) < 1$). For an ideal two-mode squeezed state, the unconditional auto-correlation between the two signal photon paths, $g_{s_1, s_2}^{(2)}(0) = 2$, which is seen in Fig. S2(b).

When the signal photons are propagated through the full experimental setup, we measure a total of $\sim 12,000$ heralded photons per second when the pulses pumping the SFWM process have ~ 1.25 nJ pulse energy. Under these conditions, we measure the delay-dependent coincidence rate between the signal and idler photons, $C_{s,i}(\tau)$, as shown in Fig. S3. From this measurement, we can extract a coincidence-to-accidentals ratio of ~ 35 for the operating pump pulse energy of ~ 1.25 nJ.

We opt for narrow spectral filtering of the signal photons so as to ensure good spectral overlap between the heralded photons and attenuated coherent state. This allows us to achieve high HOM visibility, but limits our heralding efficiency to about 7%. We expect that any limitations to the mode overlap between the two sources most likely arises due to temporal mode overlap, since photons from the two sources are generated through entirely different processes and also propagate through different lengths of various material. Nevertheless, we are able to achieve HOM visibility as high as 70%, as demonstrated in the following section.

3. HOM INTERFERENCE BETWEEN THE HERALDED SINGLE-PHOTON SOURCE AND ATTENUATED COHERENT STATE

The HOM two-photon interference defines the extent of the mode overlap (or indistinguishability) between the heralded single-photon source and attenuated coherent state. We note here that, since the herald photons are also spectrally filtered, this overlap will not necessarily be the same between the attenuated coherent state and the

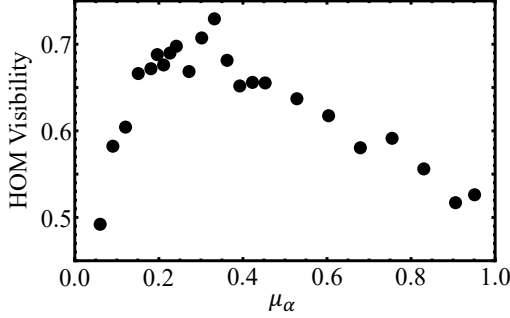


FIG. S4. **HOM two-photon interference visibility as a function of mean photon number of the attenuated coherent state.** The HOM interference visibility is plotted as a function of increasing mean photon number of the attenuated coherent state, μ_α . We see a peak in the interference visibility at $\mu_\alpha \approx 0.24$ of about 0.7. We present measurements of the QW output for $\mu_\alpha \approx 0.1, 0.24$, and 0.95 in the manuscript.

thermal state (photons arising from the SFWM process which are not heralded). The level of indistinguishability will influence the multiphoton interference observed.

We interfere photons from the heralded single-photon source and attenuated coherent state at the first step of the QW (*i.e.*, the first ultrafast time-bin beam splitter). Rotating the fast axis of the first birefringent α -BBO crystal of the QW to 45° provides 50:50 beam splitter action on photons from both sources. Each photon can exit the time-bin beam splitter in either the $|H, t_1\rangle$ state or the $|V, t_2\rangle$ state. For perfectly indistinguishable photons, HOM interference will reduce the probability of one photon exiting in the $|H, t_1\rangle$ state and one photon exiting in the $|V, t_2\rangle$ state to zero. We use the PBS after the SMF in Kerr gate 1, see Fig. S1, to split the two output ports of the time-bin beam splitter for measurement at different detectors. Measuring coincidences, C_{124} , between APDs 1, 2, and 4 while scanning the optical delay of the attenuated coherent state relative to the heralded single-photon source, τ_{HOM} , allows for the observation of HOM two-photon interference.

This HOM interference is measured for increasing mean photon number of the attenuated coherent state, and the visibility,

$$V = \frac{C_{124, \text{MAX}} - C_{124, \text{MIN}}}{C_{124, \text{MAX}}}, \quad (3)$$

is calculated and plotted in Fig. S4. We observe a maximum HOM visibility of about 70% for $\mu_\alpha = 0.24$. For lower values of μ_α , the reduced visibility is mainly due to system noise, while for higher values of μ_α , the reduced visibility is mainly due to contributions from events with more than one photon in the attenuated coherent state [7]. In order to observe the effect of μ_α on multiphoton interference, we measured the one-, two-,

and three-fold probability distributions for three different values of μ_α : 0.1, 0.24, and 0.95.

4. DATA COLLECTION AND PROCESSING

Data for the one-, two-, and three-fold measurements is collected simultaneously and extracted from the various detection event patterns measured at the four APDs. We measure these detection event patterns for three different values of the mean photon number of the attenuated coherent state, $\mu_\alpha = 0.1, 0.24$, and 0.95, for an increasing number of QW steps, $N = 1$ to 11.

For each dataset collected (*i.e.*, for a given value of N and μ_α), we measure the QW output probability distribution in two different Kerr gating regimes. First, we measure in the regime where two different time bins are Kerr gated. Here, both Kerr gates are on (*i.e.*, both pump 1 and 2 are present in their respective Kerr gates), and the two H-polarized input time bins, which are rotated by Kerr gates 1 and 2, are directed towards APD 4 and 3, respectively. This regime allows us to measure the two- and three-fold detection events associated with states in which photons exit the QW in different time bins.

In the second regime, a single time bin is Kerr gated. This regime allows for straightforward measurement of the one-fold detection events, but requires some modification in order to measure two-fold events in which the photons exit the QW in the same time bin. Here, the polarization of the QW output is rotated from H to V, prior to entering Kerr gate 1. Kerr gate 1 is turned on (*i.e.*, pump 1 is present), and rotates the polarization of the time bin of interest, from V to H. The non-gated time bins will be directed towards APD 4, while the Kerr-gated, H-polarized time bin then propagates through the optics of Kerr gate 2; however, Kerr gate 2 is inactive (*i.e.*, pump 2 is not present). The HWP after Kerr gate 2 is used to rotate the H-polarized time bin of interest by 45° , such that the PBS will have a 50% chance of splitting the two-photon state along its two output ports, towards APD 2 and 3. This regime allows us to record one-fold measurements, by combining events at APD 2 and 3, as well as two-fold measurements associated with states in which two photons exit the QW in the same time bin, by measuring coincidences between APD 2 and 3 (and APD 1, if heralding is included). Note that in this regime, we multiply two-fold events associated with photons in the same time by a factor of two, in order to account for the 50% probability of this measurement. We also divide these events by 0.85 to account for the $\sim 85\%$ coupling to the second Kerr gate SMF.

The heralded and unheralded one-, two-, and three-fold measurements can be extracted from the various coincidence patterns measured at the four APDs in the experimental setup. We consider cases in which measurements are unheralded or heralded, in order to demonstrate the interference arising from nonclassical and classical pho-

Measurement		Coincidence pattern at APDs
One-folds	Unheralded	$S_2 + S_3$
	Heralded	$C_{12} + C_{13}$
Two-folds	Unheralded	Same: $\frac{2}{0.85}C_{23}$
		Diff: C_{34}
	Heralded	Same: $\frac{2}{0.85}C_{123}$
		Diff: C_{134}
Three-folds	Unheralded	C_{234}
	Heralded	C_{1234}

TABLE SI. **Detection event patterns used for analysis of one-, two-, and three-fold measurements.** For each measurement type, both unheralded and heralded measurements are considered. The coincidence patterns used to calculate the various measurements are provided in the last column, where S_i refers to the detection of a single event at detector i , C_{ij} refers to the detection of a coincidence between detectors i and j , C_{ijk} refers to the detection of a coincidence between detectors i , j , and k , and finally C_{1234} refers to the detection of a coincidence between all four detectors.

ton statistics, respectively. The relevant coincidence patterns are presented in Table SI, where S_i ($i \in 1, 2, 3, 4$) are the detection events at APDs 1, 2, 3, and 4, respectively; C_{ij} ($i, j \in 1, 2, 3, 4$; $i \neq j$) are coincidence events between two APDs; C_{ijk} ($i, j, k \in 1, 2, 3, 4$; $i \neq j \neq k$) are coincidence events between three APDs; and C_{1234} are coincidence events between all four APDs.

A. One-fold measurements

For one-fold measurements, we operate the Kerr gate demultiplexing in only the second regime, which measures photons in a single time bin, t_m . In this case, only the Kerr-gated time bin is directed towards APDs 2 and 3, from which detected events are added together. In referring to the one-fold measurements, we really mean that one detection event is being measured at the output of the Kerr gate demultiplexing. These measurements can be unheralded or heralded, to elucidate the difference between an input state consisting of an attenuated coherent state and either a thermal state or two-mode squeezed vacuum state (TMSV), respectively.

B. Two-fold measurements

In order to collect two-fold measurements, we operate the Kerr gate demultiplexing in both regimes, measuring the case where photons can arrive in two different time

bins ($t_{m_1} \neq t_{m_2}$) or the same time bin ($t_{m_1} = t_{m_2}$). Here, we refer to the case where two events are detected after Kerr gate demultiplexing as two-fold measurements. These measurements can also be unheralded or heralded, to elucidate the difference which arises when the SFWM source provides either a thermal state or TMSV state.

C. Three-fold measurements

The partial subset of three-fold measurements is collected when operating the Kerr gate demultiplexing in only the first regime, in which all three photons arrive in different time bins, t_{m_1}, t_{m_2} , or t_{m_3} . Here, we note that the third time bin, t_{m_3} , can be in any one of the remaining time bins, where $t_{m_1} \neq t_{m_2} \neq t_{m_3}$. Here, we refer to the case where detection events occur at all three detectors after Kerr gating as three-fold measurements. These measurements can be unheralded or heralded, again to demonstrate the effect of either a thermal state or TMSV state, along with an attenuated coherent state, propagating through the QW.

5. SIMULATION OF THE QW

Experimental measurements are compared to a simulation [8] of the multi-mode interferometric network using the open source quantum optics simulation package from *Xanadu*, Strawberry Fields [9, 10]. The code used to run our simulation, and details about its implementation, can be found on GitHub [8].

An example QW circuit for $N = 2$ steps is shown in Fig. S5. Here, we use the Gaussian numeric backend. The simulation takes a TMSV state input to modes 0 and 1, and a coherent state input to mode 2. Mode 0 is the heralding mode and does not propagate through

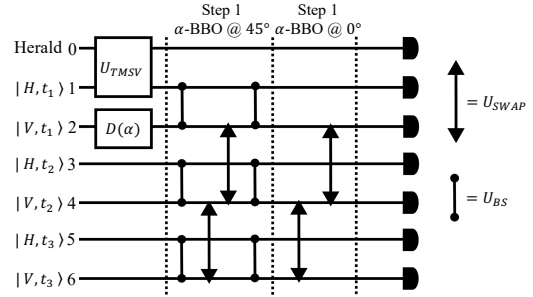


FIG. S5. **Example of the simulated quantum photonic circuit for two QW steps.** The QW takes a two-mode squeezed vacuum (TMSV) state at input modes 0 and 1, where mode 0 is the herald which does not traverse the QW. Input to mode 2 is a weak coherent state, $D(\alpha)$. Each step of the QW consists of a unitary beam splitter operator, U_{BS} , and a unitary SWAP operator, U_{SWAP} .

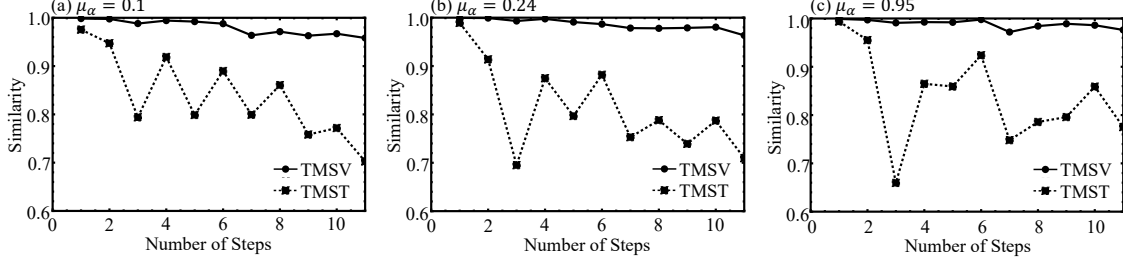


FIG. S6. **Similarity between experiment and simulation with a TMSV and TMST input state.** The similarity between experimental two-fold measurements and simulation with either a TMSV or TMST input to modes 0 and 1, for (a) $\mu_\alpha = 0.1$, (b) $\mu_\alpha = 0.24$, and (c) $\mu_\alpha = 0.95$.

the QW. Each step of the walk is performed by a 50:50 beam splitter gate (BS), U_{BS} , and a SWAP gate, U_{SWAP} (*i.e.*, a beam splitter with unit reflectivity). The SWAP gates are applied on the later time bins first to prevent propagation of light backwards in time. The simulation accounts for various experimental imperfections such as dark counts (*i.e.*, noise photons), optical losses, and mode mismatch between the TMSV and attenuated coherent state. Mode mismatch is accounted for by simulating the QW twice. First, the probabilities are calculated for a heralded photon (TMSV) and an attenuated coherent state, as in the experiment. Second, the probabilities are calculated for a vacuum input in place of the heralded photon and an attenuated coherent state. Note that here, the idler photon still heralds detection events, but the signal photon is effectively removed from the calculation by introducing 100% loss in its mode, prior to entering the QW. It is assumed that the detectors cannot deduce from which of these two possible QWs the photons came from, and so we convolve their photon statistics (with weights according to the estimated mode mismatch).

It is important to note that all simulations performed in this work take real experimental values as inputs (*i.e.*, no fitting is performed). Here, we perform the simulation using parameters obtained from the characterization of our experimental setup: $\mu_\alpha = 0.1, 0.24$, or 0.95 ; squeezing parameter $r = 0.12$; system efficiency = 8%, and mode-matching = 70%. The similarity between experimental measurements and simulation results is calculated as

$$S_n(X, Y) = \left(\sum_{i=1}^{n+1} \sqrt{p_i q_i} \right)^2, \quad (4)$$

where $S_n(X, Y)$ describes the overlap between the experimental state, X , and simulation state, Y , for a given number of steps, n , in the QW. The index i sums over the time bins and runs from the input time bin, $i = 1$, to the total number of time bins present after n steps. The probabilities of the experimental and theoretical QW outputs of being in a given time-bin are denoted by p_i and q_i , respectively.

A. Simulating the heralded single-photon source as a “squashed state”

In order to demonstrate nonclassicality of the two-fold measurements, we can compare them with a simulation using classical states to emulate the QW. To do so, we modify the simulation by replacing the TMSV state with a “squashed state”. The squashed state is a two-mode squeezed thermal state (TMST) which is input to modes 0 and 1, for heralding and QW propagation, respectively. We constrain the amount of squeezing in the squashed state to ensure it has a classical quasiprobability distribution. This provides the best possible classical state approximation to the lossy TMSV state [11]. In Fig. S6, we compare the similarity between experimental data and the simulation, for $\mu_\alpha = 0.1, 0.24$, and 0.95 , when either a TMSV or TMST state is input to modes 0 and 1. We see that the simulation with a TMSV input outperforms the simulation with a TMST input, which demonstrates the nonclassicality of the interference.

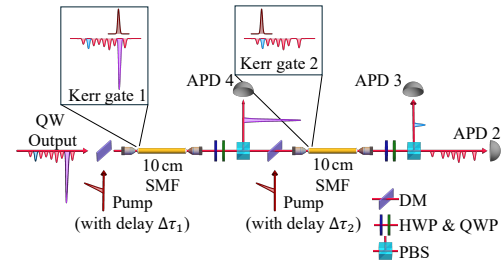


FIG. S7. **Conceptual diagram for three-fold measurements.** We use two Kerr gates to measure a subset of the three-fold detection events. Kerr gate 1 separates time bin t_{m1} (purple) from the QW output signal, to be measured by APD 2. Kerr gate 2 separates time bin t_{m2} (blue) from the QW output signal, to be measured by APD 3. All remaining time bins, $[t_{m3}]$, are measured by APD 4.

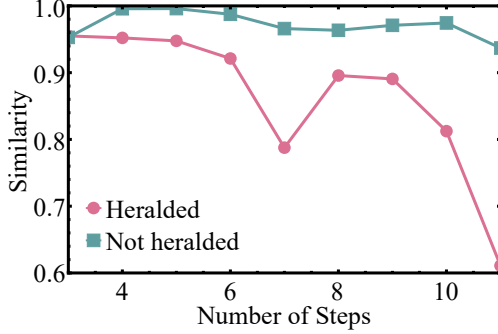


FIG. S8. **Similarity between experiment and simulation for three-fold measurements.** Heralded (pink circles) and unheralded (blue squares) three-fold experimental measurements are compared with simulation through a similarity calculation.

6. NOTES ON THREE-FOLD MEASUREMENTS

Shown in Fig. S7, three-fold measurements are performed by Kerr gating two specific time bins, t_{m_1} and t_{m_2} , and sending the remaining, non-gated time bins, $[t_{m_3}]$, to a third detector. With only two Kerr gates,

we are limited to three-fold probability distribution measurements, $P_{(t_{m_1}, t_{m_2}, [t_{m_3}])}$, where $[t_{m_3}] = \{t_m \mid m \neq m_1 \wedge m \neq m_2\}$.

We calculate the similarity between the subset of three-fold measurements, $P_{(t_{m_1}, t_{m_2}, [t_{m_3}])}$, and the corresponding simulations, shown in Fig. S8,. Since the three-fold measurements here require at least three time bins, we measure only for QW steps 3 to 11. We can attribute the lower similarity of heralded three-fold measurements to the fact that these measurements require a coincidence between all four APDs (heralded three-fold rates will be less than the unheralded three-fold rates). The rapid decrease in similarity for steps 9 to 11 can be attributed to insufficient integration time leading to very few coincidence events, and thus a large error. We also note that we calculate these coincidence events from the expressions for three-fold measurements given in Table SI, which further increases the error; this could have been mitigated by measuring the raw time tags from the time-to-digital converter directly, rather than performing coincidence logic with the time-to-digital converter. All measurements described here consider only the case where all three photons arrive in different time bins; measurement of probabilities where two or three photons can arrive in the same time bin would require three Kerr gates and additional 50:50 beam splitters.

-
- [1] K. Zielnicki, K. Garay-Palmett, D. Cruz-Delgado, H. Cruz-Ramirez, M. F. O'Boyle, B. Fang, V. O. Lorenz, A. B. U'Ren, and P. G. Kwiat, *Journal of Modern Optics* **65**, 1141 (2018).
 - [2] J. M. Dudley, G. Genty, and S. Coen, *Reviews of Modern Physics* **78**, 1135 (2006).
 - [3] C.-K. Hong, Z.-Y. Ou, and L. Mandel, *Physical review letters* **59**, 2044 (1987).
 - [4] H. Kanbara, H. Kobayashi, T. Kaino, T. Kurihara, N. Ooba, and K. Kubodera, *JOSA B* **11**, 2216 (1994).
 - [5] G. Agrawal, *Applications of nonlinear fiber optics* (Elsevier, 2001).
 - [6] C. Kupchak, J. Erskine, D. England, and B. Sussman, *Optics letters* **44**, 1427 (2019).
 - [7] A. Xu, L. Duan, L. Wang, and Y. Zhang, *Optics Express* **31**, 5662 (2023).
 - [8] G. Thekkadath, K. Fenwick, F. Bouchard, D. England, P. Bustard, K. Heshami, and B. Sussman, "Utbe-quantumwalk," <https://github.com/tguillaume/UTBEQuantumWalk> (2024).
 - [9] N. Killoran, J. Izaac, N. Quesada, V. Bergholm, M. Amy, and C. Weedbrook, *Quantum* **3**, 129 (2019).
 - [10] T. R. Bromley, J. M. Arrazola, S. Jahangiri, J. Izaac, N. Quesada, A. D. Gran, M. Schuld, J. Swinerton, Z. Zabaneh, and N. Killoran, *Quantum Science and Technology* **5**, 034010 (2020).
 - [11] H. Qi, D. J. Brod, N. Quesada, and R. García-Patrón, *Physical review letters* **124**, 100502 (2020).

Chapter 6

Concluding remarks

6.1 Summary and conclusions

This thesis has taken us on a journey through the development of a promising new technique for quantum information processing: *ultrafast time-bin encoding (UTBE)*. This journey can be divided into the six different projects summarized in this thesis, each of which are a crucial step towards scalable photonic quantum computing.

We began with an overview of quantum information processing in Chapter 1, with the goal of better understanding the current state of this field. This chapter also briefly discussed where our UTBE scheme fits into the broader picture of quantum information processing.

The remainder of this thesis continues in a way that parallels my learning experience throughout my PhD. In Chapter 2, we established an essential foundation in ultrafast and nonlinear optics, with a focus on the development and characterization of optical Kerr gating—a key component of our UTBE technique. This chapter concluded by presenting two implementations of the optical Kerr gate in the classical regime. The first implementation [263] allowed us to characterize the intrinsic Kerr gate resolution for a given set of experimental parameters and explore its configurability. Carving pulses from a continuous wave (CW) laser, we saw Kerr gate speeds as low as 305 fs. In the second implementation [264], we deepened our understanding of the optical Kerr gate’s capabilities, through a fun “side quest” in which the high temporal resolution of the Kerr gate was translated to the frequency domain.

We extended our foundational knowledge to include quantum optics in Chapter 3, where we focused on the generation, detection, and characterization of the most important aspect of any photonic quantum information processor: single photons. This chapter concluded by demonstrating that optical Kerr gating is not only capable of manipulating single photons on ultrafast timescales, but can also be used to measure the temporal wave packet of energy-time entangled photon pairs [265]. In this work, two Kerr gates were used to measure the joint temporal intensity of a photon pair source. Although ultimately limited by noise, the temporal resolution of each Kerr gate still allowed for the verification of entanglement through a violation of the time-bandwidth inequality.

Having built an extensive background in ultrafast, nonlinear, and quantum optics—and having demonstrated the versatility and efficacy of optical Kerr gating—we proceeded to implement our UTBE scheme for photonic quantum information processing in Chapter 4. This chapter presented two key demonstrations of UTBE, each operating on single photons. First, we demonstrated a well-known quantum algorithm—the quantum walk (QW)—using our UTBE technique [266]. We then explored the programmability of our UTBE scheme in the second demonstration [267]. Within the large multimode interferometers presented in each of these experiments, we saw remarkable phase stability over extended periods of time, high fidelity operation, programmability, and low loss—all key ingredients for the scalability of quantum information processors.

Given the promising capabilities of UTBE seen in the single-photon operation regime, we extended our investigations to include the last key ingredient for scalability: operation in the multiphoton regime. Chapter 5 presented a QW algorithm using our UTBE technique, operating with up to three photons [268]. Even in the multiphoton regime, our UTBE scheme saw the same desirable low loss, high fidelity, and stable operation as seen in our single-photon demonstrations.

In light of these promising demonstrations of UTBE for photonic quantum information processing, one question remains: where do we go from here?

6.2 Outlook

While the ultimate goal of this work is scaling our ultrafast approach towards full-fledged quantum information processing, this remains a long-term pursuit. Thankfully, this overarching goal can be broken down into several more manageable milestones, in order to address the current challenges.

Perhaps the largest challenge to overcome is pushing the capabilities of UTBE to include many more photons. Despite having demonstrated UTBE with up to three photons in Chapter 5, it is important to recognize that every additional photon will dramatically increase experimental acquisition times. This limitation is imposed by current single-photon sources, which rely on probabilistic nonlinear optical processes. While significant progress has been made in the fabrication of deterministic (“on-demand”) single-photon sources [7, 344], these sources are currently characterized by very long lifetimes¹, and are thus not compatible with the ultrafast timescales of our approach. Deterministic sources of nonclassical light do, however, exist in the ultrafast regime, in the form of squeezed states. Each time a nonlinear crystal is pumped by a strong pulse of light, the same squeezed state will be generated. Working with such deterministic squeezed states would require a paradigm shift towards continuous-variable (CV) encoding with homodyne measurement, which is non-trivial to implement. Nevertheless, these nonclassical states are crucial deterministic resources, *e.g.*, in scaling up cluster-state-based photonic quantum computing.

There are several promising avenues towards increased efficiency when working with probabilistic sources. For example, one could employ time-multiplexing with a switchable storage cavity to turn probabilistic sources into pseudo-deterministic sources [491, 492]. Time-multiplexing offers a strong advantage over space-multiplexing in that only a single nonlinear crystal is required. We anticipate our UTBE scheme to be readily compatible with such a time-multiplexed source. Another means of improving the rates of heralded single-photon sources employs photon-number-resolving detectors (PNRDs) [493, 494]. Sources could be pumped with higher power to increase the number of photon pairs produced, after which PNRDs would allow for

¹Note that long lifetimes are generally considered a good thing, as they indicate that these sources have a very narrow transition linewidth.

post-selection onto only single-photon events. This scheme mitigates the risk of detecting undesirable multiphoton pairs. PNRDs also open the door to combining the photon number degree of freedom with the other degrees of freedom used in UTBE (ultrafast time-bin and polarization), which could provide expanded information processing capabilities.

In addition to expanding our multiphoton capabilities, there is still room to improve the overall efficiency of our UTBE systems. Although our UTBE schemes have already seen an impressively low loss per circuit depth, there is always room for improvement. In particular, we are currently most limited by our coupling efficiency ($\sim 70\text{--}85\%$) to the single-mode fiber (SMF) in which optical Kerr gating is performed. Many factors contribute to the proper coupling of light into a SMF core [495], which could be better optimized for improved coupling efficiency. One may even consider using tapered fiber [496]. Yet another option includes foregoing the SMF altogether and instead using a crystal, with a high third-order nonlinear susceptibility, as the Kerr medium in our ultrafast all-optical gates [257].

Another limitation faced by our UTBE protocol is the pump energy requirements of each optical Kerr gate. To operate a single Kerr gate with near-unit efficiency, we require about 260 mW average power at a repetition rate of 80 MHz (3.25 nJ pulse energy). To put this into context, a Ti:Sapph with total output power of around 2.5 W would be able to pump around nine Kerr gates (with approximately 160 mW buffer left to pump single-photon sources and to account for losses along the pump beamline). Given that one Kerr gate per input photon is required for measurement—and full circuit programmability requires multiple optical Kerr gates—this limits the total possible circuit complexity; however, lower repetition-rate pulsed laser systems could be used instead to provide significantly higher pulse energy, thus mitigating restrictions imposed by total available pump power.

In light of recent demonstrations of superconducting nanowire single-photon detectors (SNSPDs) with timing jitter as low as 3 ps [11], one may even consider removing the Kerr gated measurement requirement at the output of our UTBE photonic circuits. In this case, thicker birefringent crystals could be used to achieve ultrafast time-bins with larger temporal separation than demonstrated in this thesis. Measuring n -fold coincidences with SNSPDs is not trivial and would require multiplexing

across $\sim n^2$ fast detectors; however, fast enough SNSPDs do have some photon-number-resolving capabilities [355, 356, 357], which could be leveraged. It should be noted that careful consideration of detector dead time would be required if using SNSPDs with low timing jitter. Nevertheless, this is an intriguing possibility.

By further improving our system efficiency and scaling up the number of photons our UTBE scheme is capable of handling, we can begin implementing more complicated unitary transformations, thus accessing more complex computational protocols. Two interesting avenues include the implementation of multiphoton gates (as in the KLM-based approach) or the generation of complex input states, *e.g.*, cluster states [254, 497] (as in the measurement-based approach). Another interesting direction could include investigating the quantum simulation potential of QWs using UTBE, *e.g.*, in high-energy physics [498] or in condensed matter and materials physics [447]. Of course, for full-fledged quantum computing, it will also be important to consider feed-forward measurement capabilities and error correction schemes. Another intriguing possibility in the context of noisy intermediate-scale quantum (NISQ) devices—which are likely to lack full-scale error correction—is the implementation of machine learning protocols to assist in performing quantum computation by controlling noise [499, 500].

Indeed, given the promising demonstrations of UTBE in this thesis, there are now many interesting avenues to explore with our ultrafast all-optical approach to quantum information processing. It is certainly an exciting time in the field of quantum computing and I am eager to see where this work, along with the broader efforts of many others in the field, will lead. I hope that, in reading this thesis, you have shared in some of my excitement and enthusiasm for this field.

Bibliography

- [1] R. P. Feynman. “Simulating physics with computers”. *International Journal of Theoretical Physics*, **21**(6/7):467–488, (1982).
- [2] P. W. Shor. “Fault-tolerant quantum computation”. In *Proceedings of 37th Conference on Foundations of Computer Science*, pages 56–65. IEEE, (1996).
- [3] D. J. Egger, C. Gambella, J. Marecek, S. McFaddin, M. Mevissen, R. Raymond, A. Simonetto, S. Woerner, and E. Yndurain. “Quantum computing for finance: State-of-the-art and future prospects”. *IEEE Transactions on Quantum Engineering*, **1**:1–24, (2020).
- [4] C. W. Bauer, B. Nachman, and M. Freytsis. “Simulating collider physics on quantum computers using effective field theories”. *Physical Review Letters*, **127**(21):212001, (2021).
- [5] A. O. Sushkov, I. Lovchinsky, N. Chisholm, R. L. Walsworth, H. Park, and M. D. Lukin. “Magnetic resonance detection of individual proton spins using quantum reporters”. *Physical Review Letters*, **113**(19):197601, (2014).
- [6] H. Bernien, S. Schwartz, A. Keesling, H. Levine, A. Omran, H. Pichler, S. Choi, A. S. Zibrov, M. Endres, M. Greiner, *et al.* “Probing many-body dynamics on a 51-atom quantum simulator”. *Nature*, **551**:579–584, (2017).
- [7] P. Laferriere, E. Yeung, L. Giner, S. Haffouz, J. Lapointe, G. C. Aers, P. J. Poole, R. L. Williams, and D. Dalacu. “Multiplexed single-photon source based on multiple quantum dots embedded within a single nanowire”. *Nano Letters*, **20**(5):3688–3693, (2020).

- [8] B. P. Lanyon, T. J. Weinhold, N. K. Langford, J. L. O’Brien, K. J. Resch, A. Gilchrist, and A. White. “Manipulating biphotonic qutrits”. *Physical Review Letters*, **100**(6):060504, (2008).
- [9] A. Divochiy, F. Marsili, D. Bitauld, A. Gaggero, R. Leoni, F. Mattioli, A. Korneev, V. Seleznev, N. Kaurova, O. Minaeva, *et al.* “Superconducting nanowire photon-number-resolving detector at telecommunication wavelengths”. *Nature Photonics*, **2**(5):302–306, (2008).
- [10] D. V. Reddy, R. R. Nerem, S. W. Nam, R. P. Mirin, and V. B. Verma. “Superconducting nanowire single-photon detectors with 98% system detection efficiency at 1550 nm”. *Optica*, **7**(12):1649–1653, (2020).
- [11] B. Korzh, Q.-Y. Zhao, J. P. Allmaras, S. Frasca, T. M. Autry, E. A. Bersin, A. D. Beyer, R. M. Briggs, B. Bumble, M. Colangelo, *et al.* “Demonstration of sub-3 ps temporal resolution with a superconducting nanowire single-photon detector”. *Nature Photonics*, **14**(4):250–255, (2020).
- [12] T. Graham, Y. Song, J. Scott, C. Poole, L. Phuttitarn, K. Jooya, P. Eichler, X. Jiang, A. Marra, B. Grinkemeyer, *et al.* “Multi-qubit entanglement and algorithms on a neutral-atom quantum computer”. *Nature*, **604**:457–462, (2022).
- [13] J. I. Cirac and P. Zoller. “A scalable quantum computer with ions in an array of microtraps”. *Nature*, **404**:579–581, (2000).
- [14] D. Kielpinski, C. Monroe, and D. J. Wineland. “Architecture for a large-scale ion-trap quantum computer”. *Nature*, **417**:709–711, (2002).
- [15] J. A. Jones, V. Vedral, A. Ekert, and G. Castagnoli. “Geometric quantum computation using nuclear magnetic resonance”. *Nature*, **403**:869–871, (2000).
- [16] J. M. Gambetta, J. M. Chow, and M. Steffen. “Building logical qubits in a superconducting quantum computing system”. *npj Quantum Information*, **3**(1):2, (2017).

- [17] F. Arute, K. Arya, R. Babbush, D. Bacon, J. C. Bardin, R. Barends, R. Biswas, S. Boixo, F. G. Brandao, D. A. Buell, *et al.* “Quantum supremacy using a programmable superconducting processor”. *Nature*, **574**:505–510, (2019).
- [18] H.-S. Zhong, H. Wang, Y.-H. Deng, M.-C. Chen, L.-C. Peng, Y.-H. Luo, J. Qin, D. Wu, X. Ding, Y. Hu, *et al.* “Quantum computational advantage using photons”. *Science*, **370**(6523):1460–1463, (2020).
- [19] L. S. Madsen, F. Laudenbach, M. F. Askarani, F. Rortais, T. Vincent, J. F. Bulmer, F. M. Miatto, L. Neuhaus, L. G. Helt, M. J. Collins, *et al.* “Quantum computational advantage with a programmable photonic processor”. *Nature*, **606**:75–81, (2022).
- [20] L. Szilard. “Über die entropieverminderung in einem thermodynamischen system bei eingriffen intelligenter wesen”. *Zeitschrift für Physik*, **53**(11):840–856, (1929).
- [21] J. V. Neumann. “Thermodynamik quantummechanischer gesamheiten”. *Nachrichten von der Gesellschaft der Wissenschaften zu Göttingen, Mathematisch-Physikalische Klasse*, **1**:273–291, (1927).
- [22] L. Boltzmann. “Über die beziehung zwischen dem zweiten hauptsatze des mechanischen wärmetheorie und der wahrscheinlichkeitsrechnung, respective den sätzen über das wärmegleichgewicht”. Kk Hof-und Staatsdruckerei, (1877).
- [23] H. Nyquist. “Certain factors affecting telegraph speed”. *Transactions of the American Institute of Electrical Engineers*, **43**:412–422, (1924).
- [24] R. V. Hartley. “Transmission of information”. *The Bell System Technical Journal*, **7**(3):535–563, (1928).
- [25] R. Landauer. “Irreversibility and heat generation in the computing process”. *IBM Journal of Research and Development*, **5**(3):183–191, (1961).
- [26] C. H. Bennett. “Logical reversibility of computation”. *IBM Journal of Research and Development*, **17**(6):525–532, (1973).

- [27] A. K. Fedorov, N. Gisin, S. M. Beloussov, and A. I. Lvovsky. “Quantum computing at the quantum advantage threshold: a down-to-business review”. *arXiv preprint arXiv:2203.17181*, (2022).
- [28] A. S. Holevo. “Bounds for the quantity of information transmitted by a quantum communication channel”. *Problemy Peredachi Informatsii*, **9**(3):3–11, (1973).
- [29] R. Poplavskii. “Thermodynamic models of information processes”. *Soviet Physics Uspekhi*, **18**(3):222, (1975).
- [30] R. S. Ingarden. “Quantum information theory”. *Reports on Mathematical Physics*, **10**(1):43–72, (1976).
- [31] C. E. Shannon. “A mathematical theory of communication”. *The Bell System Technical Journal*, **27**(3):379–423, (1948).
- [32] Y. I. Manin. “Vychislimoe i nevychislimoe (computable and noncomputable)”, (1980). [*Sovetskoe Radio, Moscow*].
- [33] P. Benioff. “The computer as a physical system: A microscopic quantum mechanical Hamiltonian model of computers as represented by Turing machines”. *Journal of statistical physics*, **22**:563–591, (1980).
- [34] D. Deutsch. “Quantum theory, the Church–Turing principle and the universal quantum computer”. *Proceedings of the Royal Society of London. A. Mathematical and Physical Sciences*, **400**(1818):97–117, (1985).
- [35] D. Deutsch and R. Jozsa. “Rapid solution of problems by quantum computation”. *Proceedings of the Royal Society of London. Series A: Mathematical and Physical Sciences*, **439**(1907):553–558, (1992).
- [36] T. Toffoli. “Reversible computing”. In *International Colloquium on Automata, Languages, and Programming*, pages 632–644. Springer, (1980).
- [37] E. Fredkin and T. Toffoli. “Conservative logic”. *International Journal of Theoretical Physics*, **21**(3):219–253, (1982).

- [38] K. Igeta and Y. Yamamoto. “Quantum mechanical computers with single atom and photon fields”. In *International Quantum Electronics Conference*, page TuI4. Optica Publishing Group, (1988).
- [39] G. J. Milburn. “Quantum optical Fredkin gate”. *Physical Review Letters*, **62**(18):2124, (1989).
- [40] P. W. Shor. “Algorithms for quantum computation: discrete logarithms and factoring”. In *Proceedings 35th Annual Symposium on Foundations of Computer Science*, pages 124–134. IEEE, (1994).
- [41] R. L. Rivest, A. Shamir, and L. Adleman. “A method for obtaining digital signatures and public-key cryptosystems”. *Communications of the ACM*, **21**(2):120–126, (1978).
- [42] P. W. Shor. “Scheme for reducing decoherence in quantum computer memory”. *Physical Review A*, **52**(4):R2493, (1995).
- [43] A. Peres. “Reversible logic and quantum computers”. *Physical Review A*, **32**(6):3266, (1985).
- [44] D. Aharonov and M. Ben-Or. “Fault-tolerant quantum computation with constant error”. In *Proceedings of the Twenty-Ninth Annual ACM Symposium on Theory of Computing*, pages 176–188, (1997).
- [45] E. Knill, R. Laflamme, and W. Zurek. “Threshold accuracy for quantum computation”. *arXiv preprint quant-ph/9610011*, (1996).
- [46] A. Y. Kitaev. “Quantum measurements and the Abelian stabilizer problem”. *arXiv preprint quant-ph/9511026*, (1995).
- [47] D. Coppersmith. “An approximate Fourier transform useful in quantum factoring”. *arXiv preprint quant-ph/0201067*, (2002).
- [48] L. K. Grover. “A fast quantum mechanical algorithm for database search”. In *Proceedings of the Twenty-Eighth Annual ACM Symposium on Theory of Computing*, pages 212–219, (1996).

- [49] D. R. Simon. “On the power of quantum computation”. *SIAM Journal on Computing*, **26**(5):1474–1483, (1997).
- [50] E. Bernstein and U. Vazirani. “Quantum complexity theory”. *SIAM Journal on Computing*, **26**(5):1411–1473, (1997).
- [51] C. Monroe, D. M. Meekhof, B. E. King, W. M. Itano, and D. J. Wineland. “Demonstration of a fundamental quantum logic gate”. *Physical Review Letters*, **75**(25):4714, (1995).
- [52] I. Waki, S. Kassner, G. Birkel, and H. Walther. “Observation of ordered structures of laser-cooled ions in a quadrupole storage ring”. *Physical Review Letters*, **68**(13):2007, (1992).
- [53] G. Birkel, S. Kassner, and H. Walther. “Multiple-shell structures of laser-cooled $^{24}\text{Mg}^+$ ions in a quadrupole storage ring”. *Nature*, **357**:310–313, (1992).
- [54] M. Raizen, J. Gilligan, J. C. Bergquist, W. M. Itano, and D. J. Wineland. “Ionic crystals in a linear paul trap”. *Physical Review A*, **45**(9):6493, (1992).
- [55] J. I. Cirac and P. Zoller. “Quantum computations with cold trapped ions”. *Physical Review Letters*, **74**(20):4091, (1995).
- [56] S. Lloyd. “A potentially realizable quantum computer”. *Science*, **261**(5128):1569–1571, (1993).
- [57] D. P. DiVincenzo. “Two-bit gates are universal for quantum computation”. *Physical Review A*, **51**(2):1015, (1995).
- [58] D. P. DiVincenzo. “Quantum computation”. *Science*, **270**(5234):255–261, (1995).
- [59] D. G. Cory, A. F. Fahmy, and T. F. Havel. “Ensemble quantum computing by NMR spectroscopy”. *Proceedings of the National Academy of Sciences*, **94**(5):1634–1639, (1997).
- [60] N. A. Gershenfeld and I. L. Chuang. “Bulk spin-resonance quantum computation”. *Science*, **275**(5298):350–356, (1997).

- [61] J. A. Jones and M. Mosca. “Implementation of a quantum algorithm on a nuclear magnetic resonance quantum computer”. *The Journal of Chemical Physics*, **109**(5):1648–1653, (1998).
- [62] I. L. Chuang, N. Gershenfeld, and M. Kubinec. “Experimental implementation of fast quantum searching”. *Physical Review Letters*, **80**(15):3408, (1998).
- [63] D. Loss and D. P. DiVincenzo. “Quantum computation with quantum dots”. *Physical Review A*, **57**(1):120, (1998).
- [64] B. E. Kane. “A silicon-based nuclear spin quantum computer”. *Nature*, **393**:133–137, (1998).
- [65] D. Gottesman. “The Heisenberg representation of quantum computers”. *arXiv preprint quant-ph/9807006*, (1998).
- [66] S. L. Braunstein, C. M. Caves, R. Jozsa, N. Linden, S. Popescu, and R. Schack. “Separability of very noisy mixed states and implications for NMR quantum computing”. *Physical Review Letters*, **83**(5):1054, (1999).
- [67] J. Brooke, D. Bitko, Rosenbaum, and G. Aeppli. “Quantum annealing of a disordered magnet”. *Science*, **284**(5415):779–781, (1999).
- [68] Y. Nakamura, Y. A. Pashkin, and J. Tsai. “Coherent control of macroscopic quantum states in a single-Cooper-pair box”. *Nature*, **398**:786–788, (1999).
- [69] D. P. DiVincenzo. “The physical implementation of quantum computation”. *Fortschritte der Physik: Progress of Physics*, **48**(9-11):771–783, (2000).
- [70] R. Marx, A. Fahmy, J. M. Myers, W. Bermel, and S. Glaser. “Approaching five-bit NMR quantum computing”. *Physical Review A*, **62**(1):012310, (2000).
- [71] E. Knill, R. Laflamme, R. Martinez, and C.-H. Tseng. “An algorithmic benchmark for quantum information processing”. *Nature*, **404**:368–370, (2000).
- [72] E. Farhi, J. Goldstone, S. Gutmann, and M. Sipser. “Quantum computation by adiabatic evolution”. *arXiv preprint quant-ph/0001106*, (2000).

- [73] L. M. Vandersypen, M. Steffen, G. Breyta, C. S. Yannoni, R. Cleve, and I. L. Chuang. “Experimental realization of an order-finding algorithm with an NMR quantum computer”. *Physical Review Letters*, **85**(25):5452, (2000).
- [74] L. M. Vandersypen, M. Steffen, G. Breyta, C. S. Yannoni, M. H. Sherwood, and I. L. Chuang. “Experimental realization of Shor’s quantum factoring algorithm using nuclear magnetic resonance”. *Nature*, **414**:883–887, (2001).
- [75] N. Linden and S. Popescu. “Good dynamics versus bad kinematics: is entanglement needed for quantum computation?”. *Physical Review Letters*, **87**(4):047901, (2001).
- [76] E. Knill, R. Laflamme, and G. J. Milburn. “A scheme for efficient quantum computation with linear optics”. *Nature*, **409**:46–52, (2001).
- [77] R. Raussendorf and H. J. Briegel. “A one-way quantum computer”. *Physical Review Letters*, **86**(22):5188, (2001).
- [78] R. Dumke, M. Volk, T. Müther, F. Buchkremer, G. Birkel, and W. Ertmer. “Micro-optical realization of arrays of selectively addressable dipole traps: A scalable configuration for quantum computation with atomic qubits”. *Physical Review Letters*, **89**(9):097903, (2002).
- [79] S. Gulde, M. Riebe, G. P. Lancaster, C. Becher, J. Eschner, H. Häffner, F. Schmidt-Kaler, I. L. Chuang, and R. Blatt. “Implementation of the Deutsch–Jozsa algorithm on an ion-trap quantum computer”. *Nature*, **421**:48–50, (2003).
- [80] F. Schmidt-Kaler, H. Häffner, M. Riebe, S. Gulde, G. P. Lancaster, T. Deuschle, C. Becher, C. F. Roos, J. Eschner, and R. Blatt. “Realization of the Cirac–Zoller controlled-NOT quantum gate”. *Nature*, **422**:408–411, (2003).
- [81] T. Pittman, M. Fitch, B. Jacobs, and J. Franson. “Experimental controlled-NOT logic gate for single photons in the coincidence basis”. *Physical Review A*, **68**(3):032316, (2003).

- [82] J. L. O’Brien, G. J. Pryde, A. G. White, T. C. Ralph, and D. Branning. “Demonstration of an all-optical quantum controlled-NOT gate”. *Nature*, **426**:264–267, (2003).
- [83] Z. Zhao, Y.-A. Chen, A.-N. Zhang, T. Yang, H. J. Briegel, and J.-W. Pan. “Experimental demonstration of five-photon entanglement and open-destination teleportation”. *Nature*, **430**:54–58, (2004).
- [84] M. S. Anwar, D. Blazina, H. A. Carteret, S. B. Duckett, T. Halstead, J. Jones, C. Kozak, and R. Taylor. “Preparing high purity initial states for nuclear magnetic resonance quantum computing”. *Physical Review Letters*, **93**(4):040501, (2004).
- [85] B. D. Josephson. “Possible new effects in superconductive tunnelling”. *Physics letters*, **1**(7):251–253, (1962).
- [86] T. Duty, G. Johansson, K. Bladh, D. Gunnarsson, C. Wilson, and P. Delsing. “Observation of quantum capacitance in the Cooper-pair transistor”. *Physical Review Letters*, **95**(20):206807, (2005).
- [87] M. A. Sillanpää, T. Lehtinen, A. Paila, Y. Makhlin, L. Roschier, and P. J. Hakonen. “Direct observation of Josephson capacitance”. *Physical Review Letters*, **95**(20):206806, (2005).
- [88] H. Häffner, W. Hänsel, C. Roos, J. Benhelm, D. Chek-al Kar, M. Chwalla, T. Körber, U. Rapol, M. Riebe, P. Schmidt, *et al.* “Scalable multiparticle entanglement of trapped ions”. *Nature*, **438**:643–646, (2005).
- [89] Y. Miroshnychenko, W. Alt, I. Dotsenko, L. Förster, M. Khudaverdyan, D. Meschede, D. Schrader, and A. Rauschenbeutel. “An atom-sorting machine”. *Nature*, **442**:151–151, (2006).
- [90] S. Seidelin, J. Chiaverini, R. Reichle, J. J. Bollinger, D. Leibfried, J. Britton, J. Wesenberg, R. Blakestad, R. Epstein, D. Hume, *et al.* “Microfabricated surface-electrode ion trap for scalable quantum information processing”. *Physical Review Letters*, **96**(25):253003, (2006).

- [91] O. Hosten, M. T. Rakher, J. T. Barreiro, N. A. Peters, and P. G. Kwiat. “Counterfactual quantum computation through quantum interrogation”. *Nature*, **439**:949–952, (2006).
- [92] F. H. Koppens, C. Buizert, K.-J. Tielrooij, I. T. Vink, K. C. Nowack, T. Meunier, L. Kouwenhoven, and L. Vandersypen. “Driven coherent oscillations of a single electron spin in a quantum dot”. *Nature*, **442**:766–771, (2006).
- [93] A. R. Stegner, C. Boehme, H. Huebl, M. Stutzmann, K. Lips, and M. S. Brandt. “Electrical detection of coherent ^{31}P spin quantum states”. *Nature Physics*, **2**(12):835–838, (2006).
- [94] C. Negrevergne, T. Mahesh, C. Ryan, M. Ditty, F. Cyr-Racine, W. Power, N. Boulant, T. Havel, D. Cory, and R. Laflamme. “Benchmarking quantum control methods on a 12-qubit system”. *Physical Review Letters*, **96**(17):170501, (2006).
- [95] N. C. Menicucci, S. T. Flammia, and O. Pfister. “One-way quantum computing in the optical frequency comb”. *Physical Review Letters*, **101**(13):130501, (2008).
- [96] M. Lobino, D. Korystov, C. Kupchak, E. Figueroa, B. C. Sanders, and A. Lvovsky. “Complete characterization of quantum-optical processes”. *Science*, **322**(5901):563–566, (2008).
- [97] J. Chen, J. B. Altepeter, M. Medic, K. F. Lee, B. Gokden, R. H. Hadfield, S. W. Nam, and P. Kumar. “Demonstration of a quantum controlled-NOT gate in the telecommunications band”. *Physical Review Letters*, **100**(13):133603, (2008).
- [98] C.-Y. Lu, W.-B. Gao, O. Gühne, X.-Q. Zhou, Z.-B. Chen, and J.-W. Pan. “Demonstrating anyonic fractional statistics with a six-qubit quantum simulator”. *Physical Review Letters*, **102**(3):030502, (2009).
- [99] N. H. Lindner and T. Rudolph. “Proposal for pulsed on-demand sources of photonic cluster state strings”. *Physical Review Letters*, **103**(11):113602, (2009).

- [100] M. Anderlini, P. J. Lee, B. L. Brown, J. Sebby-Strabley, W. D. Phillips, and J. V. Porto. “Controlled exchange interaction between pairs of neutral atoms in an optical lattice”. *Nature*, **448**:452–456, (2007).
- [101] K. D. Nelson, X. Li, and D. S. Weiss. “Imaging single atoms in a three-dimensional array”. *Nature Physics*, **3**(8):556–560, (2007).
- [102] J. D. Jost, J. Home, J. M. Amini, D. Hanneke, R. Ozeri, C. Langer, J. J. Bollinger, D. Leibfried, and D. J. Wineland. “Entangled mechanical oscillators”. *Nature*, **459**:683–685, (2009).
- [103] T. Monz, K. Kim, A. Villar, P. Schindler, M. Chwalla, M. Riebe, C. F. Roos, H. Häffner, W. Hänsel, M. Hennrich, *et al.* “Realization of universal ion-trap quantum computation with decoherence-free qubits”. *Physical Review Letters*, **103**(20):200503, (2009).
- [104] T. Monz, K. Kim, W. Hänsel, M. Riebe, A. Villar, P. Schindler, M. Chwalla, M. Hennrich, and R. Blatt. “Realization of the quantum Toffoli gate with trapped ions”. *Physical Review Letters*, **102**(4):040501, (2009).
- [105] A. Niskanen, K. Harrabi, F. Yoshihara, Y. Nakamura, S. Lloyd, and J. S. Tsai. “Quantum coherent tunable coupling of superconducting qubits”. *Science*, **316**(5825):723–726, (2007).
- [106] J. Plantenberg, P. De Groot, C. Harmans, and J. Mooij. “Demonstration of controlled-NOT quantum gates on a pair of superconducting quantum bits”. *Nature*, **447**:836–839, (2007).
- [107] J. Majer, J. Chow, J. Gambetta, J. Koch, B. Johnson, J. Schreier, L. Frunzio, D. Schuster, A. A. Houck, A. Wallraff, *et al.* “Coupling superconducting qubits via a cavity bus”. *Nature*, **449**:443–447, (2007).
- [108] M. A. Sillanpää, J. I. Park, and R. W. Simmonds. “Coherent quantum state storage and transfer between two phase qubits via a resonant cavity”. *Nature*, **449**:438–442, (2007).

- [109] M. Neeley, M. Ansmann, R. C. Bialczak, M. Hofheinz, N. Katz, E. Lucero, A. O’Connell, H. Wang, A. N. Cleland, and J. M. Martinis. “Process tomography of quantum memory in a Josephson-phase qubit coupled to a two-level state”. *Nature Physics*, **4**(7):523–526, (2008).
- [110] M. G. Dutt, L. Childress, L. Jiang, E. Togan, J. Maze, F. Jelezko, A. Zibrov, P. Hemmer, and M. Lukin. “Quantum register based on individual electronic and nuclear spin qubits in diamond”. *Science*, **316**(5829):1312–1316, (2007).
- [111] I. Appelbaum, B. Huang, and D. J. Monsma. “Electronic measurement and control of spin transport in silicon”. *Nature*, **447**:295–298, (2007).
- [112] B. Trauzettel, D. V. Bulaev, D. Loss, and G. Burkard. “Spin qubits in graphene quantum dots”. *Nature Physics*, **3**(3):192–196, (2007).
- [113] D. Hsieh, D. Qian, L. Wray, Y. Xia, Y. S. Hor, R. J. Cava, and M. Z. Hasan. “A topological Dirac insulator in a quantum spin Hall phase”. *Nature*, **452**:970–974, (2008).
- [114] S. Bertaina, S. Gambarelli, T. Mitra, B. Tsukerblat, A. Müller, and B. Barbara. “Quantum oscillations in a molecular magnet”. *Nature*, **453**:203–206, (2008).
- [115] G. Fuchs, V. Dobrovitski, D. Toyli, F. Heremans, and D. Awschalom. “Gigahertz dynamics of a strongly driven single quantum spin”. *Science*, **326**(5959):1520–1522, (2009).
- [116] C.-Y. Lu, X.-Q. Zhou, O. Gühne, W.-B. Gao, J. Zhang, Z.-S. Yuan, A. Goebel, T. Yang, and J.-W. Pan. “Experimental entanglement of six photons in graph states”. *Nature Physics*, **3**(2):91–95, (2007).
- [117] B. P. Lanyon, T. J. Weinhold, N. K. Langford, M. Barbieri, D. F. James, A. Gilchrist, and A. G. White. “Experimental demonstration of a compiled version of Shor’s algorithm with quantum entanglement”. *Physical Review Letters*, **99**(25):250505, (2007).

- [118] C.-Y. Lu, D. E. Browne, T. Yang, and J.-W. Pan. “Demonstration of a compiled version of Shor’s quantum factoring algorithm using photonic qubits”. *Physical Review Letters*, **99**(25):250504, (2007).
- [119] A. Politi, M. J. Cryan, J. G. Rarity, S. Yu, and J. L. O’Brien. “Silica-on-silicon waveguide quantum circuits”. *Science*, **320**(5876):646–649, (2008).
- [120] A. Politi, J. C. Matthews, and J. L. O’Brien. “Shor’s quantum factoring algorithm on a photonic chip”. *Science*, **325**(5945):1221–1221, (2009).
- [121] B. P. Lanyon, M. Barbieri, M. P. Almeida, T. Jennewein, T. C. Ralph, K. J. Resch, G. J. Pryde, J. L. O’Brien, A. Gilchrist, and A. G. White. “Simplifying quantum logic using higher-dimensional Hilbert spaces”. *Nature Physics*, **5**(2):134–140, (2009).
- [122] M. Hofheinz, H. Wang, M. Ansmann, R. C. Bialczak, E. Lucero, M. Neeley, A. O’Connell, D. Sank, J. Wenner, J. M. Martinis, *et al.* “Synthesizing arbitrary quantum states in a superconducting resonator”. *Nature*, **459**:546–549, (2009).
- [123] L. DiCarlo, J. M. Chow, J. M. Gambetta, L. S. Bishop, B. R. Johnson, D. Schuster, J. Majer, A. Blais, L. Frunzio, S. Girvin, *et al.* “Demonstration of two-qubit algorithms with a superconducting quantum processor”. *Nature*, **460**:240–244, (2009).
- [124] A. D. Boozer, A. Boca, R. Miller, T. E. Northup, and H. J. Kimble. “Reversible state transfer between light and a single trapped atom”. *Physical Review Letters*, **98**(19):193601, (2007).
- [125] K. S. Choi, H. Deng, J. Laurat, and H. Kimble. “Mapping photonic entanglement into and out of a quantum memory”. *Nature*, **452**:67–71, (2008).
- [126] D. E. Chang, A. S. Sørensen, E. A. Demler, and M. D. Lukin. “A single-photon transistor using nanoscale surface plasmons”. *Nature Physics*, **3**(11):807–812, (2007).

- [127] J. J. Morton, A. M. Tyryshkin, R. M. Brown, S. Shankar, B. W. Lovett, A. Ardavan, T. Schenkel, E. E. Haller, J. W. Ager, and S. Lyon. “Solid-state quantum memory using the ^{31}P nuclear spin”. *Nature*, **455**:1085–1088, (2008).
- [128] J. Hwang, M. Pototschnig, R. Lettow, G. Zumofen, A. Renn, S. Götzinger, and V. Sandoghdar. “A single-molecule optical transistor”. *Nature*, **460**:76–80, (2009).
- [129] M. J. Biercuk, H. Uys, A. P. VanDevender, N. Shiga, W. M. Itano, and J. J. Bollinger. “Optimized dynamical decoupling in a model quantum memory”. *Nature*, **458**:996–1000, (2009).
- [130] B. P. Lanyon, M. Barbieri, M. P. Almeida, and A. G. White. “Experimental quantum computing without entanglement”. *Physical Review Letters*, **101**(20):200501, (2008).
- [131] M. Dolev, M. Heiblum, V. Umansky, A. Stern, and D. Mahalu. “Observation of a quarter of an electron charge at the $\nu = 5/2$ quantum Hall state”. *Nature*, **452**:829–834, (2008).
- [132] S. J. Devitt, A. G. Fowler, A. M. Stephens, A. D. Greentree, L. C. Hollenberg, W. J. Munro, and K. Nemoto. “Architectural design for a topological cluster state quantum computer”. *New Journal of Physics*, **11**(8):083032, (2009).
- [133] P. Marks. “Google demonstrates quantum computer image search”. *New Scientist*, **11**, (2009).
- [134] J. P. Home, D. Hanneke, J. D. Jost, J. M. Amini, D. Leibfried, and D. J. Wineland. “Complete methods set for scalable ion trap quantum information processing”. *Science*, **325**(5945):1227–1230, (2009).
- [135] A. W. Harrow, A. Hassidim, and S. Lloyd. “Quantum algorithm for linear systems of equations”. *Physical Review Letters*, **103**(15):150502, (2009).
- [136] D. Hanneke, J. Home, J. D. Jost, J. M. Amini, D. Leibfried, and D. J. Wineland. “Realization of a programmable two-qubit quantum processor”. *Nature Physics*, **6**(1):13–16, (2010).

- [137] B. P. Lanyon, J. D. Whitfield, G. G. Gillett, M. E. Goggin, M. P. Almeida, I. Kassal, J. D. Biamonte, M. Mohseni, B. J. Powell, M. Barbieri, *et al.* “Towards quantum chemistry on a quantum computer”. *Nature Chemistry*, **2**(2):106–111, (2010).
- [138] A. Peruzzo, M. Lobino, J. C. Matthews, N. Matsuda, A. Politi, K. Poulios, X.-Q. Zhou, Y. Lahini, N. Ismail, K. Wörhoff, *et al.* “Quantum walks of correlated photons”. *Science*, **329**(5998):1500–1503, (2010).
- [139] S. Aaronson and A. Arkhipov. “The computational complexity of linear optics”. In *Proceedings of the Forty-Third Annual ACM Symposium on Theory of Computing*, pages 333–342, (2011).
- [140] M. W. Johnson, M. H. Amin, S. Gildert, T. Lanting, F. Hamze, N. Dickson, R. Harris, A. J. Berkley, J. Johansson, P. Bunyk, *et al.* “Quantum annealing with manufactured spins”. *Nature*, **473**:194–198, (2011).
- [141] T. Monz, P. Schindler, J. T. Barreiro, M. Chwalla, D. Nigg, W. A. Coish, M. Harlander, W. Hänsel, M. Hennrich, and R. Blatt. “14-qubit entanglement: Creation and coherence”. *Physical Review Letters*, **106**(13):130506, (2011).
- [142] P. Schindler, J. T. Barreiro, T. Monz, V. Nebendahl, D. Nigg, M. Chwalla, M. Hennrich, and R. Blatt. “Experimental repetitive quantum error correction”. *Science*, **332**(6033):1059–1061, (2011).
- [143] K. R. Brown, A. C. Wilson, Y. Colombe, C. Ospelkaus, A. M. Meier, E. Knill, D. Leibfried, and D. J. Wineland. “Single-qubit-gate error below 10^{-4} in a trapped ion”. *Physical Review A*, **84**(3):030303, (2011).
- [144] M. Mariantoni, H. Wang, T. Yamamoto, M. Neeley, R. C. Bialczak, Y. Chen, M. Lenander, E. Lucero, A. D. O’Connell, D. Sank, *et al.* “Implementing the quantum von Neumann architecture with superconducting circuits”. *Science*, **334**(6052):61–65, (2011).
- [145] G. Fuchs, G. Burkard, P. Klimov, and D. Awschalom. “A quantum memory intrinsic to single nitrogen–vacancy centres in diamond”. *Nature Physics*, **7**(10):789–793, (2011).

- [146] K. C. Lee, M. R. Sprague, B. J. Sussman, J. Nunn, N. K. Langford, X.-M. Jin, T. Champion, P. Michelberger, K. F. Reim, D. England, *et al.* “Entangling macroscopic diamonds at room temperature”. *Science*, **334**(6060):1253–1256, (2011).
- [147] D. Horsman, A. G. Fowler, S. Devitt, and R. Van Meter. “Surface code quantum computing by lattice surgery”. *New Journal of Physics*, **14**(12):123011, (2012).
- [148] P. C. Maurer, G. Kucsko, C. Latta, L. Jiang, N. Y. Yao, S. D. Bennett, F. Pastawski, D. Hunger, N. Chisholm, M. Markham, *et al.* “Room-temperature quantum bit memory exceeding one second”. *Science*, **336**(6086):1283–1286, (2012).
- [149] K. Saeedi, S. Simmons, J. Z. Salvail, P. Dluhy, H. Riemann, N. V. Abrosimov, P. Becker, H.-J. Pohl, J. J. Morton, and M. L. Thewalt. “Room-temperature quantum bit storage exceeding 39 minutes using ionized donors in silicon-28”. *Science*, **342**(6160):830–833, (2013).
- [150] A. M. Childs, D. Gosset, and Z. Webb. “Universal computation by multiparticle quantum walk”. *Science*, **339**(6121):791–794, (2013).
- [151] C. Schneider, M. Enderlein, T. Huber, and T. Schätz. “Optical trapping of an ion”. *Nature Photonics*, **4**(11):772–775, (2010).
- [152] C. Zipkes, S. Palzer, C. Sias, and M. Köhl. “A trapped single ion inside a Bose–Einstein condensate”. *Nature*, **464**:388–391, (2010).
- [153] J. M. Amini, H. Uys, J. H. Wesenberg, S. Seidelin, J. Britton, J. J. Bollinger, D. Leibfried, C. Ospelkaus, A. P. VanDevender, and D. J. Wineland. “Toward scalable ion traps for quantum information processing”. *New Journal of Physics*, **12**(3):033031, (2010).
- [154] C. Ospelkaus, U. Warring, Y. Colombe, K. Brown, J. Amini, D. Leibfried, and D. J. Wineland. “Microwave quantum logic gates for trapped ions”. *Nature*, **476**:181–184, (2011).

- [155] J. W. Britton, B. C. Sawyer, A. C. Keith, C.-C. J. Wang, J. K. Freericks, H. Uys, M. J. Biercuk, and J. J. Bollinger. “Engineered two-dimensional Ising interactions in a trapped-ion quantum simulator with hundreds of spins”. *Nature*, **484**:489–492, (2012).
- [156] J. Petta, H. Lu, and A. Gossard. “A coherent beam splitter for electronic spin states”. *Science*, **327**(5966):669–672, (2010).
- [157] S. Nadj-Perge, S. Frolov, E. Bakkers, and L. P. Kouwenhoven. “Spin–orbit qubit in a semiconductor nanowire”. *Nature*, **468**:1084–1087, (2010).
- [158] S. Simmons, R. M. Brown, H. Riemann, N. V. Abrosimov, P. Becker, H.-J. Pohl, M. L. Thewalt, K. M. Itoh, and J. J. Morton. “Entanglement in a solid-state spin ensemble”. *Nature*, **470**:69–72, (2011).
- [159] M. Grinolds, P. Maletinsky, S. Hong, M. Lukin, R. Walsworth, and A. Yacoby. “Quantum control of proximal spins using nanoscale magnetic resonance imaging”. *Nature Physics*, **7**(9):687–692, (2011).
- [160] S. Takahashi, I. Tupitsyn, J. Van Tol, C. Beedle, D. Hendrickson, and P. Stamp. “Decoherence in crystals of quantum molecular magnets”. *Nature*, **476**:76–79, (2011).
- [161] M. Fuechsle, J. A. Miwa, S. Mahapatra, H. Ryu, S. Lee, O. Warschkow, L. C. Hollenberg, G. Klimeck, and M. Y. Simmons. “A single-atom transistor”. *Nature Nanotechnology*, **7**(4):242–246, (2012).
- [162] K. Nemoto, M. Trupke, S. J. Devitt, A. M. Stephens, B. Scharfenberger, K. Buczak, T. Nöbauer, M. S. Everitt, J. Schmiedmayer, and W. J. Munro. “Photonic architecture for scalable quantum information processing in diamond”. *Physical Review X*, **4**(3):031022, (2014).
- [163] A. Laucht, J. T. Muhonen, F. A. Mohiyaddin, R. Kalra, J. P. Dehollain, S. Freer, F. E. Hudson, M. Veldhorst, R. Rahman, G. Klimeck, *et al.* “Electrically controlling single-spin qubits in a continuous microwave field”. *Science Advances*, **1**(3):e1500022, (2015).

- [164] Z. Bian, F. Chudak, W. G. Macready, L. Clark, and F. Gaitan. “Experimental determination of Ramsey numbers”. *Physical Review Letters*, **111**(13):130505, (2013).
- [165] C. Salter, R. Stevenson, I. Farrer, C. Nicoll, D. Ritchie, and A. Shields. “An entangled-light-emitting diode”. *Nature*, **465**:594–597, (2010).
- [166] H. Wang, M. Mariani, R. C. Bialczak, M. Lenander, E. Lucero, M. Neeley, A. O’Connell, D. Sank, M. Weides, J. Wenner, *et al.* “Deterministic entanglement of photons in two superconducting microwave resonators”. *Physical Review Letters*, **106**(6):060401, (2011).
- [167] A. Peruzzo, A. Laing, A. Politi, T. Rudolph, and J. L. O’Brien. “Multimode quantum interference of photons in multiport integrated devices”. *Nature Communications*, **2**(1):224, (2011).
- [168] M. Pysher, Y. Miwa, R. Shahrokhshahi, R. Bloomer, and O. Pfister. “Parallel generation of quadripartite cluster entanglement in the optical frequency comb”. *Physical Review Letters*, **107**(3):030505, (2011).
- [169] X.-Q. Zhou, T. C. Ralph, P. Kalasuwan, M. Zhang, A. Peruzzo, B. P. Lanyon, and J. L. O’Brien. “Adding control to arbitrary unknown quantum operations”. *Nature Communications*, **2**(1):413, (2011).
- [170] X.-C. Yao, T.-X. Wang, H.-Z. Chen, W.-B. Gao, A. G. Fowler, R. Raussendorf, Z.-B. Chen, N.-L. Liu, C.-Y. Lu, Y.-J. Deng, *et al.* “Experimental demonstration of topological error correction”. *Nature*, **482**:489–494, (2012).
- [171] D. Nigg, M. Mueller, E. A. Martinez, P. Schindler, M. Hennrich, T. Monz, M. A. Martin-Delgado, and R. Blatt. “Quantum computations on a topologically encoded qubit”. *Science*, **345**(6194):302–305, (2014).
- [172] M. Zhong, M. P. Hedges, R. L. Ahlefeldt, J. G. Bartholomew, S. E. Beavan, S. M. Wittig, J. J. Longdell, and M. J. Sellars. “Optically addressable nuclear spins in a solid with a six-hour coherence time”. *Nature*, **517**:177–180, (2015).

- [173] A. D. Córcoles, E. Magesan, S. J. Srinivasan, A. W. Cross, M. Steffen, J. M. Gambetta, and J. M. Chow. “Demonstration of a quantum error detection code using a square lattice of four superconducting qubits”. *Nature Communications*, **6**(1):6979, (2015).
- [174] M. Veldhorst, C. Yang, J. Hwang, W. Huang, J. Dehollain, J. Muhonen, S. Simmons, A. Laucht, F. Hudson, K. M. Itoh, *et al.* “A two-qubit logic gate in silicon”. *Nature*, **526**:410–414, (2015).
- [175] S. J. Devitt. “Performing quantum computing experiments in the cloud”. *Physical Review A*, **94**(3):032329, (2016).
- [176] D. Alsina and J. I. Latorre. “Experimental test of Mermin inequalities on a five-qubit quantum computer”. *Physical Review A*, **94**(1):012314, (2016).
- [177] P. J. O’Malley, R. Babbush, I. D. Kivlichan, J. Romero, J. R. McClean, R. Barends, J. Kelly, P. Roushan, A. Tranter, N. Ding, *et al.* “Scalable quantum simulation of molecular energies”. *Physical Review X*, **6**(3):031007, (2016).
- [178] T. Monz, D. Nigg, E. A. Martinez, M. F. Brandl, P. Schindler, R. Rines, S. X. Wang, I. L. Chuang, and R. Blatt. “Realization of a scalable Shor algorithm”. *Science*, **351**(6277):1068–1070, (2016).
- [179] C. J. Ballance, T. P. Harty, N. M. Linke, M. A. Sepiol, and D. M. Lucas. “High-fidelity quantum logic gates using trapped-ion hyperfine qubits”. *Physical Review Letters*, **117**(6):060504, (2016).
- [180] M. Kues, C. Reimer, P. Roztock, L. R. Cortés, S. Sciara, B. Wetz, Y. Zhang, A. Cino, S. T. Chu, B. E. Little, *et al.* “On-chip generation of high-dimensional entangled quantum states and their coherent control”. *Nature*, **546**:622–626, (2017).
- [181] J. Preskill. “Quantum computing in the NISQ era and beyond”. *Quantum*, **2**:79, (2018).

- [182] R. Li, L. Petit, D. P. Franke, J. P. Dehollain, J. Helsen, M. Steudtner, N. K. Thomas, Z. R. Yoscovits, K. J. Singh, S. Wehner, *et al.* “A crossbar network for silicon quantum dot qubits”. *Science Advances*, **4**(7):eaar3960, (2018).
- [183] J. Kelly. “Engineering superconducting qubit arrays for quantum supremacy”. In *APS March Meeting Abstracts*, pages A33–001, (2018).
- [184] J. Hsu. “Ces 2018: Intel’s 49-qubit chip shoots for quantum supremacy”, (2018).
- [185] K. Wright, K. M. Beck, S. Debnath, J. Amini, Y. Nam, N. Grzesiak, J.-S. Chen, N. Pienti, M. Chmielewski, C. Collins, *et al.* “Benchmarking an 11-qubit quantum computer”. *Nature Communications*, **10**(1):5464, (2019).
- [186] B. Lekitsch, S. Weidt, A. G. Fowler, K. Mølmer, S. J. Devitt, C. Wunderlich, and W. K. Hensinger. “Blueprint for a microwave trapped ion quantum computer”. *Science Advances*, **3**(2):e1601540, (2017).
- [187] C. Kokail, C. Maier, R. van Bijnen, T. Brydges, M. K. Joshi, P. Jurcevic, C. A. Muschik, P. Silvi, R. Blatt, C. F. Roos, *et al.* “Self-verifying variational quantum simulation of lattice models”. *Nature*, **569**:355–360, (2019).
- [188] Q.-Y. Liang, A. V. Venkatramani, S. H. Cantu, T. L. Nicholson, M. J. Gullans, A. V. Gorshkov, J. D. Thompson, C. Chin, M. D. Lukin, and V. Vuletić. “Observation of three-photon bound states in a quantum nonlinear medium”. *Science*, **359**(6377):783–786, (2018).
- [189] F. Lenzini, J. Janousek, O. Thearle, M. Villa, B. Haylock, S. Kasture, L. Cui, H.-P. Phan, D. V. Dao, H. Yonezawa, *et al.* “Integrated photonic platform for quantum information with continuous variables”. *Science Advances*, **4**(12):eaat9331, (2018).
- [190] H. Wang, J. Qin, X. Ding, M.-C. Chen, S. Chen, X. You, Y.-M. He, X. Jiang, L. You, Z. Wang, *et al.* “Boson sampling with 20 input photons and a 60-mode interferometer in a 10^{14} -dimensional Hilbert space”. *Physical Review Letters*, **123**(25):250503, (2019).

- [191] A. P. Vepsäläinen, A. H. Karamlou, J. L. Orrell, A. S. Dogra, B. Loer, F. Vasconcelos, D. K. Kim, A. J. Melville, B. M. Niedzielski, J. L. Yoder, *et al.* “Impact of ionizing radiation on superconducting qubit coherence”. *Nature*, **584**:551–556, (2020).
- [192] Google AI Quantum and Collaborators, F. Arute, K. Arya, R. Babbush, D. Bacon, J. C. Bardin, R. Barends, S. Boixo, M. Broughton, B. B. Buckley, *et al.* “Hartree-fock on a superconducting qubit quantum computer”. *Science*, **369**(6507):1084–1089, (2020).
- [193] D. Maslov, J.-S. Kim, S. Bravyi, T. J. Yoder, and S. Sheldon. “Quantum advantage for computations with limited space”. *Nature Physics*, **17**(8):894–897, (2021).
- [194] S. Krastanov, H. Raniwala, J. Holzgrafe, K. Jacobs, M. Lončar, M. J. Reagor, and D. R. Englund. “Optically heralded entanglement of superconducting systems in quantum networks”. *Physical Review Letters*, **127**(4):040503, (2021).
- [195] S. Daiss, S. Langenfeld, S. Welte, E. Distant, P. Thomas, L. Hartung, O. Morin, and G. Rempe. “A quantum-logic gate between distant quantum-network modules”. *Science*, **371**(6529):614–617, (2021).
- [196] I. Pogorelov, T. Feldker, C. D. Marciniak, L. Postler, G. Jacob, O. Kriegelsteiner, V. Podlesnic, M. Meth, V. Negnevitsky, M. Stadler, *et al.* “Compact ion-trap quantum computing demonstrator”. *PRX Quantum*, **2**(2):020343, (2021).
- [197] S. Ebadi, T. T. Wang, H. Levine, A. Keesling, G. Semeghini, A. Omran, D. Bluvstein, R. Samajdar, H. Pichler, W. W. Ho, *et al.* “Quantum phases of matter on a 256-atom programmable quantum simulator”. *Nature*, **595**:227–232, (2021).
- [198] P. Scholl, M. Schuler, H. J. Williams, A. A. Eberharter, D. Barredo, K.-N. Schymik, V. Lienhard, L.-P. Henry, T. C. Lang, T. Lahaye, *et al.* “Quantum simulation of 2D antiferromagnets with hundreds of Rydberg atoms”. *Nature*, **595**:233–238, (2021).

- [199] Y. Wu, W.-S. Bao, S. Cao, F. Chen, M.-C. Chen, X. Chen, T.-H. Chung, H. Deng, Y. Du, D. Fan, *et al.* “Strong quantum computational advantage using a superconducting quantum processor”. *Physical Review Letters*, **127**(18):180501, (2021).
- [200] C. Taballione, M. C. Anguita, M. de Goede, P. Venderbosch, B. Kassenberg, H. Snijders, N. Kannan, W. L. Vleeshouwers, D. Smith, J. P. Epping, *et al.* “20-mode universal quantum photonic processor”. *Quantum*, **7**:1071, (2023).
- [201] S. Sara Bartolucci, P. Birchall, H. Bombín, H. Cable, C. Dawson, M. Gimeno-Segovia, E. Johnston, K. Kieling, N. Nickerson, M. Pant, F. Pastawski, T. Rudolph, and C. Sparrow. “Fusion-based quantum computation”. *Nature Communications*, **14**(1):912, (2023).
- [202] Y. Kim, A. Eddins, S. Anand, K. X. Wei, E. Van Den Berg, S. Rosenblatt, H. Nayfeh, Y. Wu, M. Zaletel, K. Temme, *et al.* “Evidence for the utility of quantum computing before fault tolerance”. *Nature*, **618**:500–505, (2023).
- [203] C. Oh, M. Liu, Y. Alexeev, B. Fefferman, and L. Jiang. “Classical algorithm for simulating experimental Gaussian boson sampling”. *Nature Physics*, **20**:1461–1468, (2024).
- [204] S. Simmons. “Scalable fault-tolerant quantum technologies with silicon color centers”. *PRX Quantum*, **5**(1):010102, (2024).
- [205] M. DeCross, R. Haghshenas, M. Liu, Y. Alexeev, C. H. Baldwin, J. P. Bartolotta, M. Bohn, E. Chertkov, J. Colina, D. DelVento, *et al.* “The computational power of random quantum circuits in arbitrary geometries”. *arXiv preprint arXiv:2406.02501*, (2024).
- [206] K. Alexander, A. Bahgat, A. Benyamini, D. Black, D. Bonneau, S. Burgos, B. BurrIDGE, G. Campbell, G. Catalano, A. Ceballos, *et al.* “A manufacturable platform for photonic quantum computing”. *arXiv preprint arXiv:2404.17570*, (2024).
- [207] Google Quantum AI and Collaborators. “Quantum error correction below the surface code threshold”. *Nature*, (2024).

- [208] Y. Y. Atas, J. Zhang, R. Lewis, A. Jahanpour, J. F. Haase, and C. A. Muschik. “SU(2) hadrons on a quantum computer via a variational approach”. *Nature Communications*, **12**(1):6499, (2021).
- [209] C. H. Yang, R. Leon, J. Hwang, A. Saraiva, T. Tanttu, W. Huang, J. Camirand Lemyre, K. W. Chan, K. Tan, F. E. Hudson, *et al.* “Operation of a silicon quantum processor unit cell above one Kelvin”. *Nature*, **580**:350–354, (2020).
- [210] R. Leon, C. H. Yang, J. Hwang, J. C. Lemyre, T. Tanttu, W. Huang, K. W. Chan, K. Tan, F. Hudson, K. Itoh, *et al.* “Coherent spin control of s-, p-, d- and f-electrons in a silicon quantum dot”. *Nature Communications*, **11**(1):797, (2020).
- [211] J. Yoneda, K. Takeda, A. Noiri, T. Nakajima, S. Li, J. Kamioka, T. Koder, and S. Tarucha. “Quantum non-demolition readout of an electron spin in silicon”. *Nature Communications*, **11**(1):1144, (2020).
- [212] S. Asaad, V. Mourik, B. Joecker, M. A. Johnson, A. D. Baczewski, H. R. Firgau, M. T. Mądzik, V. Schmitt, J. J. Pla, F. E. Hudson, *et al.* “Coherent electrical control of a single high-spin nucleus in silicon”. *Nature*, **579**:205–209, (2020).
- [213] K. C. Miao, J. P. Blanton, C. P. Anderson, A. Bourassa, A. L. Crook, G. Wolfowicz, H. Abe, T. Ohshima, and D. D. Awschalom. “Universal coherence protection in a solid-state spin qubit”. *Science*, **369**(6510):1493–1497, (2020).
- [214] R. A. Thomas, M. Parniak, C. Østfeldt, C. B. Møller, C. Bærentsen, Y. Tsaturyan, A. Schliesser, J. Appel, E. Zeuthen, and E. S. Polzik. “Entanglement between distant macroscopic mechanical and spin systems”. *Nature Physics*, **17**(2):228–233, (2021).
- [215] P. Thomas, L. Ruscio, O. Morin, and G. Rempe. “Efficient generation of entangled multiphoton graph states from a single atom”. *Nature*, **608**:677–681, (2022).
- [216] A. Erhard, H. Poulsen Nautrup, M. Meth, L. Postler, R. Stricker, M. Stadler, V. Negnevitsky, M. Ringbauer, P. Schindler, H. J. Briegel, *et al.* “Entangling logical qubits with lattice surgery”. *Nature*, **589**:220–224, (2021).

- [217] L. Postler, S. Heußen, I. Pogorelov, M. Rispler, T. Feldker, M. Meth, C. D. Marciniak, R. Stricker, M. Ringbauer, R. Blatt, *et al.* “Demonstration of fault-tolerant universal quantum gate operations”. *Nature*, **605**:675–680, (2022).
- [218] M. Ringbauer, M. Meth, L. Postler, R. Stricker, R. Blatt, P. Schindler, and T. Monz. “A universal qudit quantum processor with trapped ions”. *Nature Physics*, **18**(9):1053–1057, (2022).
- [219] D. Bluvstein, S. J. Evered, A. A. Geim, S. H. Li, H. Zhou, T. Manovitz, S. Ebadi, M. Cain, M. Kalinowski, D. Hangleiter, *et al.* “Logical quantum processor based on reconfigurable atom arrays”. *Nature*, **626**:58–65, (2024).
- [220] L. Pause, L. Sturm, M. Mittenbühler, S. Amann, T. Preuschoff, D. Schäffner, M. Schlosser, and G. Birkel. “Supercharged two-dimensional tweezer array with more than 1000 atomic qubits”. *Optica*, **11**(2):222–226, (2024).
- [221] S. Paesani, M. Borghi, S. Signorini, A. Mainos, L. Pavesi, and A. Laing. “Near-ideal spontaneous photon sources in silicon quantum photonics”. *Nature Communications*, **11**(1):2505, (2020).
- [222] N. Tömm, A. Javadi, N. O. Antoniadis, D. Najer, M. C. Löbl, A. R. Korsch, R. Schott, S. R. Valentin, A. D. Wieck, A. Ludwig, *et al.* “A bright and fast source of coherent single photons”. *Nature Nanotechnology*, **16**(4):399–403, (2021).
- [223] Y.-H. Deng, Y.-C. Gu, H.-L. Liu, S.-Q. Gong, H. Su, Z.-J. Zhang, H.-Y. Tang, M.-H. Jia, J.-M. Xu, M.-C. Chen, *et al.* “Gaussian boson sampling with pseudo-photon-number-resolving detectors and quantum computational advantage”. *Physical Review Letters*, **131**(15):150601, (2023).
- [224] A. D. King, J. Raymond, T. Lanting, S. V. Isakov, M. Mohseni, G. Poulin-Lamarre, S. Ejtemaee, W. Bernoudy, I. Ozfidan, A. Y. Smirnov, *et al.* “Scaling advantage over path-integral Monte Carlo in quantum simulation of geometrically frustrated magnets”. *Nature Communications*, **12**(1):1113, (2021).

- [225] X. Xue, M. Russ, N. Samkharadze, B. Undseth, A. Sammak, G. Scappucci, and L. M. Vandersypen. “Quantum logic with spin qubits crossing the surface code threshold”. *Nature*, **601**:343–347, (2022).
- [226] M. T. Mądzik, S. Asaad, A. Youssry, B. Joecker, K. M. Rudinger, E. Nielsen, K. C. Young, T. J. Proctor, A. D. Baczewski, A. Laucht, *et al.* “Precision tomography of a three-qubit donor quantum processor in silicon”. *Nature*, **601**:348–353, (2022).
- [227] A. Noiri, K. Takeda, T. Nakajima, T. Kobayashi, A. Sammak, G. Scappucci, and S. Tarucha. “Fast universal quantum gate above the fault-tolerance threshold in silicon”. *Nature*, **601**:338–342, (2022).
- [228] L. Petit, H. Eenink, M. Russ, W. Lawrie, N. Hendrickx, S. Philips, J. Clarke, L. Vandersypen, and M. Veldhorst. “Universal quantum logic in hot silicon qubits”. *Nature*, **580**:355–359, (2020).
- [229] N. W. Hendrickx, W. I. Lawrie, M. Russ, F. van Riggelen, S. L. de Snoo, R. N. Schouten, A. Sammak, G. Scappucci, and M. Veldhorst. “A four-qubit germanium quantum processor”. *Nature*, **591**:580–585, (2021).
- [230] J. M. Pino, J. M. Dreiling, C. Figgatt, J. P. Gaebler, S. A. Moses, M. Allman, C. Baldwin, M. Foss-Feig, D. Hayes, K. Mayer, *et al.* “Demonstration of the trapped-ion quantum CCD computer architecture”. *Nature*, **592**:209–213, (2021).
- [231] Y. Nam, J.-S. Chen, N. C. Pienti, K. Wright, C. Delaney, D. Maslov, K. R. Brown, S. Allen, J. M. Amini, J. Apisdorf, *et al.* “Ground-state energy estimation of the water molecule on a trapped-ion quantum computer”. *npj Quantum Information*, **6**(1):33, (2020).
- [232] A. Browaeys and T. Lahaye. “Many-body physics with individually controlled Rydberg atoms”. *Nature Physics*, **16**(2):132–142, (2020).
- [233] T. Đorđević, P. Samutpraphoot, P. L. Ocola, H. Bernien, B. Grinkemeyer, I. Dimitrova, V. Vuletić, and M. D. Lukin. “Entanglement transport and a

- nanophotonic interface for atoms in optical tweezers”. *Science*, **373**(6562):1511–1514, (2021).
- [234] L. Henriët, L. Beguin, A. Signoles, T. Lahaye, A. Browaeys, G.-O. Reymond, and C. Jurczak. “Quantum computing with neutral atoms”. *Quantum*, **4**:327, (2020).
- [235] N. Gisin and R. Thew. “Quantum communication”. *Nature Photonics*, **1**(3):165–171, (2007).
- [236] H. J. Kimble. “The quantum internet”. *Nature*, **453**:1023–1030, (2008).
- [237] A. Crespi, R. Ramponi, R. Osellame, L. Sansoni, I. Bongioanni, F. Sciarrino, G. Vallone, and P. Mataloni. “Integrated photonic quantum gates for polarization qubits”. *Nature Communications*, **2**(1):566, (2011).
- [238] J. M. Lukens and P. Lougovski. “Frequency-encoded photonic qubits for scalable quantum information processing”. *Optica*, **4**(1):8–16, (2016).
- [239] X. Gao, Y. Zhang, A. D’Errico, K. Heshami, and E. Karimi. “High-speed imaging of spatiotemporal correlations in Hong-Ou-Mandel interference”. *Optics Express*, **30**(11):19456–19464, (2022).
- [240] A. D’Errico, F. Cardano, M. Maffei, A. Dauphin, R. Barboza, C. Esposito, B. Piccirillo, M. Lewenstein, P. Massignan, and L. Marrucci. “Two-dimensional topological quantum walks in the momentum space of structured light”. *Optica*, **7**(2):108–114, (2020).
- [241] F. Brandt, M. Hiekkamäki, F. Bouchard, M. Huber, and R. Fickler. “High-dimensional quantum gates using full-field spatial modes of photons”. *Optica*, **7**(2):98–107, (2020).
- [242] P. C. Humphreys, B. J. Metcalf, J. B. Spring, M. Moore, X.-M. Jin, M. Barbieri, W. S. Kolthammer, and I. A. Walmsley. “Linear optical quantum computing in a single spatial mode”. *Physical Review Letters*, **111**(15):150501, (2013).
- [243] I. L. Chuang and Y. Yamamoto. “Simple quantum computer”. *Physical Review A*, **52**(5):3489, (1995).

- [244] J. Carolan, C. Harrold, C. Sparrow, E. Martín-López, N. J. Russell, J. W. Silverstone, P. J. Shadbolt, N. Matsuda, M. Oguma, M. Itoh, *et al.* “Universal linear optics”. *Science*, **349**(6249):711–716, (2015).
- [245] N. C. Harris, G. R. Steinbrecher, M. Prabhu, Y. Lahini, J. Mower, D. Bunandar, C. Chen, F. N. Wong, T. Baehr-Jones, M. Hochberg, *et al.* “Quantum transport simulations in a programmable nanophotonic processor”. *Nature Photonics*, **11**(7):447–452, (2017).
- [246] C. Taballione, T. A. Wolterink, J. Lugani, A. Eckstein, B. A. Bell, R. Grootjans, I. Visscher, D. Geskus, C. G. Roeloffzen, J. J. Renema, *et al.* “ 8×8 reconfigurable quantum photonic processor based on silicon nitride waveguides”. *Optics Express*, **27**(19):26842–26857, (2019).
- [247] C. Taballione, R. van der Meer, H. J. Snijders, P. Hooijschuur, J. P. Epping, M. de Goede, B. Kassenberg, P. Venderbosch, C. Toebes, H. van den Vlekkert, *et al.* “A universal fully reconfigurable 12-mode quantum photonic processor”. *Materials for Quantum Technology*, **1**(3):035002, (2021).
- [248] J. M. Arrazola, V. Bergholm, K. Brádler, T. R. Bromley, M. J. Collins, I. Dhand, A. Fumagalli, T. Gerrits, A. Goussev, L. G. Helt, *et al.* “Quantum circuits with many photons on a programmable nanophotonic chip”. *Nature*, **591**:54–60, (2021).
- [249] J. Brendel, N. Gisin, W. Tittel, and H. Zbinden. “Pulsed energy-time entangled twin-photon source for quantum communication”. *Physical Review Letters*, **82**(12):2594, (1999).
- [250] I. Marcikic, H. de Riedmatten, W. Tittel, V. Scarani, H. Zbinden, and N. Gisin. “Time-bin entangled qubits for quantum communication created by femtosecond pulses”. *Physical Review A*, **66**(6):062308, (2002).
- [251] I. Marcikic, H. De Riedmatten, W. Tittel, H. Zbinden, M. Legré, and N. Gisin. “Distribution of time-bin entangled qubits over 50 km of optical fiber”. *Physical Review Letters*, **93**(18):180502, (2004).

- [252] J. M. Donohue, M. Agnew, J. Lavoie, and K. J. Resch. “Coherent ultrafast measurement of time-bin encoded photons”. *Physical Review Letters*, **111**(15):153602, (2013).
- [253] K. R. Motes, A. Gilchrist, J. P. Dowling, and P. P. Rohde. “Scalable boson sampling with time-bin encoding using a loop-based architecture”. *Physical Review Letters*, **113**(12):120501, (2014).
- [254] S. Yokoyama, R. Ukai, S. C. Armstrong, C. Sornphiphatphong, T. Kaji, S. Suzuki, J.-i. Yoshikawa, H. Yonezawa, N. C. Menicucci, and A. Furusawa. “Ultra-large-scale continuous-variable cluster states multiplexed in the time domain”. *Nature Photonics*, **7**(12):982–986, (2013).
- [255] Y. He, X. Ding, Z.-E. Su, H.-L. Huang, J. Qin, C. Wang, S. Unsleber, C. Chen, H. Wang, Y.-M. He, *et al.* “Time-bin-encoded boson sampling with a single-photon device”. *Physical Review Letters*, **118**(19):190501, (2017).
- [256] W. Asavanant, Y. Shiozawa, S. Yokoyama, B. Charoensombutamon, H. Emura, R. N. Alexander, S. Takeda, J.-i. Yoshikawa, N. C. Menicucci, H. Yonezawa, *et al.* “Generation of time-domain-multiplexed two-dimensional cluster state”. *Science*, **366**(6463):373–376, (2019).
- [257] C. Kupchak, P. J. Bustard, K. Heshami, J. Erskine, M. Spanner, D. G. England, and B. J. Sussman. “Time-bin-to-polarization conversion of ultrafast photonic qubits”. *Physical Review A*, **96**(5):053812, (2017).
- [258] C. Kupchak, J. Erskine, D. England, and B. Sussman. “Terahertz-bandwidth switching of heralded single photons”. *Optics Letters*, **44**(6):1427–1430, (2019).
- [259] D. England, F. Bouchard, K. Fenwick, K. Bonsma-Fisher, Y. Zhang, P. J. Bustard, and B. J. Sussman. “Perspectives on all-optical Kerr switching for quantum optical applications”. *Applied Physics Letters*, **119**(16), (2021).
- [260] F. Bouchard, D. England, P. J. Bustard, K. L. Fenwick, E. Karimi, K. Heshami, and B. Sussman. “Achieving ultimate noise tolerance in quantum communication”. *Physical Review Applied*, **15**(2):024027, (2021).

- [261] F. Bouchard, D. England, P. J. Bustard, K. Heshami, and B. Sussman. “Quantum communication with ultrafast time-bin qubits”. *PRX Quantum*, **3**(1):010332, (2022).
- [262] F. Bouchard, K. Bonsma-Fisher, K. Heshami, P. J. Bustard, D. England, and B. Sussman. “Measuring ultrafast time-bin qubits”. *Physical Review A*, **107**(2):022618, (2023).
- [263] K. L. Fenwick, D. G. England, P. J. Bustard, J. M. Fraser, and B. J. Sussman. “Carving out configurable ultrafast pulses from a continuous wave source via the optical Kerr effect”. *Optics Express*, **28**(17):24845–24853, (2020).
- [264] K. L. Fenwick, G. S. Thekkadath, P. J. Bustard, D. England, F. Bouchard, and B. Sussman. “Time-of-flight spectroscopy with ultrafast all-optical gating”. *Optics Express*, **33**(3):5914–5922, (2025).
- [265] A. R. Cameron, K. L. Fenwick, S. W. Cheng, S. Schwarz, B. MacLellan, P. J. Bustard, D. England, B. Sussman, and K. J. Resch. “Ultrafast measurement of energy-time entanglement with an optical Kerr shutter”. *Physical Review A*, **108**(4):L041503, (2023).
- [266] K. L. Fenwick, F. Bouchard, G. S. Thekkadath, D. England, P. J. Bustard, K. Heshami, and B. Sussman. “Photonic quantum walk with ultrafast time-bin encoding”. *Optica*, **11**(7):1017–1023, (2024).
- [267] F. Bouchard, K. Fenwick, K. Bonsma-Fisher, D. England, P. J. Bustard, K. Heshami, and B. Sussman. “Programmable photonic quantum circuits with ultrafast time-bin encoding”. *Physical Review Letters*, **133**(9):090601, (2024).
- [268] K. L. Fenwick, J. Baker, G. S. Thekkadath, A. Z. Goldberg, K. Heshami, P. J. Bustard, D. England, F. Bouchard, and B. Sussman. “Multiphoton interference in a single-spatial-mode quantum walk”. *arXiv preprint arXiv:2409.11483*, (2024).
- [269] A. M. Weiner. “Ultrafast optics”. John Wiley & Sons, (2011).

- [270] V. Blanchet, M. Z. Zgierski, T. Seideman, and A. Stolow. “Discerning vibronic molecular dynamics using time-resolved photoelectron spectroscopy”. *Nature*, **401**:52–54, (1999).
- [271] A. E. Boguslavskiy, J. Mikosch, A. Gijsbertsen, M. Spanner, S. Patchkovskii, N. Gador, M. J. Vrakking, and A. Stolow. “The multielectron ionization dynamics underlying attosecond strong-field spectroscopies”. *Science*, **335**(6074):1336–1340, (2012).
- [272] M. Takamoto, F.-L. Hong, R. Higashi, and H. Katori. “An optical lattice clock”. *Nature*, **435**:321–324, (2005).
- [273] M. Malinauskas, A. Žukauskas, S. Hasegawa, Y. Hayasaki, V. Mizeikis, R. Buividas, and S. Juodkazis. “Ultrafast laser processing of materials: from science to industry”. *Light: Science & Applications*, **5**(8):e16133–e16133, (2016).
- [274] A. F. Pegoraro, A. Ridsdale, D. J. Moffatt, Y. Jia, J. P. Pezacki, and A. Stolow. “Optimally chirped multimodal CARS microscopy based on a single Ti:sapphire oscillator”. *Optics Express*, **17**(4):2984–2996, (2009).
- [275] W. H. Knox. “Ultrafast technology in telecommunications”. *IEEE Journal of Selected Topics in Quantum Electronics*, **6**(6):1273–1278, (2000).
- [276] G. Hardy. “A theorem concerning Fourier transforms”. *Journal of the London Mathematical Society*, **1**(3):227–231, (1933).
- [277] W. Heisenberg. “Über den anschaulichen inhalt der quantentheoretischen kinematik und mechanik”. *Zeitschrift für Physik*, **43**(3):172–198, (1927).
- [278] D. Gabor. “Theory of communication.”. *Journal of the Institution of Electrical Engineers-part III: Radio and Communication Engineering*, **93**(26):429–457, (1946).
- [279] P. W. Milonni and J. H. Eberly. “Laser physics”. John Wiley & Sons, (2010).
- [280] H. A. Haus, J. G. Fujimoto, and E. P. Ippen. “Structures for additive pulse mode locking”. *JOSA B*, **8**(10):2068–2076, (1991).

- [281] R. P. Feynman, R. B. Leighton, and M. Sands. “The Feynman Lectures on Physics: Mainly Mechanics, Radiation, and Heat”, volume I. Basic Books, (2015).
- [282] G. Diener. “Superluminal group velocities and information transfer”. *Physics Letters A*, **223**(5):327–331, (1996).
- [283] M. W. Mitchell and R. Y. Chiao. “Causality and negative group delays in a simple bandpass amplifier”. *American Journal of Physics*, **66**(1):14–19, (1998).
- [284] “Fourier transform”. <https://mathworld.wolfram.com/FourierTransform.html>. Accessed: 2024-11-01.
- [285] D. Strickland and G. Mourou. “Compression of amplified chirped optical pulses”. *Optics Communications*, **55**(6):447–449, (1985).
- [286] D. Fabris, W. Holgado, F. Silva, T. Witting, J. W. Tisch, and H. Crespo. “Single-shot implementation of dispersion-scan for the characterization of ultra-short laser pulses”. *Optics Express*, **23**(25):32803–32808, (2015).
- [287] A. Assion, T. Baumert, M. Bergt, T. Brixner, B. Kiefer, V. Seyfried, M. Strehle, and G. Gerber. “Control of chemical reactions by feedback-optimized phase-shaped femtosecond laser pulses”. *Science*, **282**(5390):919–922, (1998).
- [288] H. Sardesai, C.-C. Chang, and A. Weiner. “A femtosecond code-division multiple-access communication system test bed”. *Journal of Lightwave Technology*, **16**(11):1953, (1998).
- [289] R. W. Boyd. “Nonlinear optics”. Elsevier, (2008).
- [290] P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich. “Generation of optical harmonics”. *Physical Review Letters*, **7**(4):118, (1961).
- [291] T. H. Maiman. “Stimulated optical radiation in ruby”. *Nature*, **187**:493–494, (1960).
- [292] N. Couture. “Enhancing time-resolved THz systems through the integration of optical fibers”. PhD thesis, Université d’Ottawa/University of Ottawa, (2023).

- [293] J. Midwinter and J. Warner. “The effects of phase matching method and of uniaxial crystal symmetry on the polar distribution of second-order non-linear optical polarization”. *British Journal of Applied Physics*, **16**(8):1135, (1965).
- [294] M. Hobden and J. Warner. “The temperature dependence of the refractive indices of pure lithium niobate”. *Physics Letters*, **22**(3):243–244, (1966).
- [295] A. Sit, F. Bouchard, R. Fickler, J. Gagnon-Bischoff, H. Larocque, K. Hesami, D. Elser, C. Peuntinger, K. Günthner, B. Heim, *et al.* “High-dimensional intracity quantum cryptography with structured photons”. *Optica*, **4**(9):1006–1010, (2017).
- [296] G. Thekkadath, D. England, F. Bouchard, Y. Zhang, M. Kim, and B. Sussman. “Intensity interferometry for holography with quantum and classical light”. *Science Advances*, **9**(27):eadh1439, (2023).
- [297] V. R. Almeida, C. A. Barrios, R. R. Panepucci, M. Lipson, M. A. Foster, D. G. Ouzounov, and A. L. Gaeta. “All-optical switching on a silicon chip”. *Optics Letters*, **29**(24):2867–2869, (2004).
- [298] M. A. Hall, J. B. Altepeter, and P. Kumar. “Ultrafast switching of photonic entanglement”. *Physical Review Letters*, **106**(5):053901, (2011).
- [299] B. Dromey, S. Kar, M. Zepf, and P. Foster. “The plasma mirror—a subpicosecond optical switch for ultrahigh power lasers”. *Review of Scientific Instruments*, **75**(3):645–649, (2004).
- [300] J.-P. W. MacLean, J. M. Donohue, and K. J. Resch. “Direct characterization of ultrafast energy-time entangled photon pairs”. *Physical Review Letters*, **120**(5):053601, (2018).
- [301] Q. Guo, R. Sekine, L. Ledezma, R. Nehra, D. J. Dean, A. Roy, R. M. Gray, S. Jahani, and A. Marandi. “Femtojoule femtosecond all-optical switching in lithium niobate nanophotonics”. *Nature Photonics*, **16**(9):625–631, (2022).

- [302] K. L. Fenwick. “Exploiting ultrafast optical pulses: probing excitonic properties of 2D materials and utilizing the Kerr effect for all-optical switching”. Master’s thesis, Queen’s University (Canada), (2019).
- [303] A. Mahjoubfar, D. V. Churkin, S. Barland, N. Broderick, S. K. Turitsyn, and B. Jalali. “Time stretch and its applications”. *Nature Photonics*, **11**(6):341–351, (2017).
- [304] V. Torres-Company, J. Lancis, and P. Andres. “Space-time analogies in optics”. *Progress in Optics*, **56**:1–80, (2011).
- [305] D. Solli, J. Chou, and B. Jalali. “Amplified wavelength–time transformation for real-time spectroscopy”. *Nature Photonics*, **2**(1):48–51, (2008).
- [306] K. Goda and B. Jalali. “Dispersive Fourier transformation for fast continuous single-shot measurements”. *Nature Photonics*, **7**(2):102–112, (2013).
- [307] I. Esmaeil Zadeh, J. W. Los, R. B. Gourgues, J. Chang, A. W. Elshaari, J. R. Zichi, Y. J. Van Staaden, J. P. Swens, N. Kalhor, A. Guardiani, *et al.* “Efficient single-photon detection with 7.7 ps time resolution for photon-correlation measurements”. *ACS Photonics*, **7**(7):1780–1787, (2020).
- [308] Schott. “Tie-31: Mechanical and thermal properties of optical glass”, (2010).
- [309] M. Planck. “On the law of distribution of energy in the normal spectrum”. *Annalen der Physik*, **4**(553):1, (1901).
- [310] A. Einstein. “Über einen die erzeugung und verwandlung des lichtes betreffenden heuristischen gesichtspunkt”, (1905).
- [311] N. Bohr. “On the constitution of atoms and molecules”. *The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science*, **26**(151):1–25, (1913).
- [312] P. A. M. Dirac. “The quantum theory of the emission and absorption of radiation”. *Proceedings of the Royal Society of London. Series A, Containing Papers of a Mathematical and Physical Character*, **114**(767):243–265, (1927).

- [313] R. Loudon. “The quantum theory of light”. OUP Oxford, (2000).
- [314] C. C. Gerry and P. L. Knight. “Introductory quantum optics”. Cambridge university press, (2023).
- [315] M. O. Scully and M. S. Zubairy. “Quantum optics”. Cambridge university press, (1997).
- [316] N. Bohr. “Über die serienspektra der elemente”. *Zeitschrift für Physik*, **2**(5):423–469, (1920).
- [317] A.-L. Cauchy. “Sur les formules qui résultent de l’emploi du signe et sur $>$ ou $<$, et sur les moyennes entre plusieurs quantités”. *Cours d’Analyse, 1er Partie: Analyse Algebrique*, pages 373–377, (1821).
- [318] V. Bouniakowsky. “Sur quelques inégalités concernant les intégrales ordinaires et les intégrales aux différences finies”. Commissionnaires de l’Académie Impériale des Sciences, St. Petersburg, (1859).
- [319] H. A. Schwarz and H. Schwarz. “Ueber ein die flächen kleinsten flächeninhalts betreffendes problem der variationsrechnung: Festschrift zum siebenzigsten geburtstage des herrn karl weierstrass”. Springer, (1890).
- [320] C. Fabre and N. Treps. “Modes and states in quantum optics”. *Reviews of Modern Physics*, **92**(3):035005, (2020).
- [321] E. Schrödinger. “Der stetige übergang von der mikro-zur makromechanik”. *Naturwissenschaften*, **14**:664–666, (1926).
- [322] M. Tse, H. Yu, N. Kijbunchoo, A. Fernandez-Galiana, P. Dupej, L. Barsotti, C. Blair, D. Brown, S. Dwyer, A. Effler, *et al.* “Quantum-enhanced advanced LIGO detectors in the era of gravitational-wave astronomy”. *Physical Review Letters*, **123**(23):231107, (2019).
- [323] L. Boltzmann. “Studien uber das gleichgewicht der lebenden kraft”. *Wissenschaftliche Abhandlungen*, **1**:49–96, (1868).

- [324] M. D. Eisaman, J. Fan, A. Migdall, and S. V. Polyakov. “Invited review article: Single-photon sources and detectors”. *Review of Scientific Instruments*, **82**(7), (2011).
- [325] G. Brassard, N. Lütkenhaus, T. Mor, and B. C. Sanders. “Limitations on practical quantum cryptography”. *Physical Review Letters*, **85**(6):1330, (2000).
- [326] F. Hufnagel, A. Sit, F. Bouchard, Y. Zhang, D. England, K. Heshami, B. J. Sussman, and E. Karimi. “Investigation of underwater quantum channels in a 30 meter flume tank using structured photons”. *New Journal of Physics*, **22**(9):093074, (2020).
- [327] T. Schmitt-Manderbach, H. Weier, M. Fürst, R. Ursin, F. Tiefenbacher, T. Scheidl, J. Perdigues, Z. Sodnik, C. Kurtsiefer, J. G. Rarity, *et al.* “Experimental demonstration of free-space decoy-state quantum key distribution over 144 km”. *Physical Review Letters*, **98**(1):010504, (2007).
- [328] P. Michler, A. Kiraz, C. Becher, W. Schoenfeld, P. Petroff, L. Zhang, E. Hu, and A. Imamoglu. “A quantum dot single-photon turnstile device”. *Science*, **290**(5500):2282–2285, (2000).
- [329] V. Zwiller, T. Aichele, W. Seifert, J. Persson, and O. Benson. “Generating visible single photons on demand with single InP quantum dots”. *Applied Physics Letters*, **82**(10):1509–1511, (2003).
- [330] C. Santori, D. Fattal, J. Vučković, G. S. Solomon, and Y. Yamamoto. “Indistinguishable photons from a single-photon device”. *Nature*, **419**:594–597, (2002).
- [331] J. Kim, O. Benson, H. Kan, and Y. Yamamoto. “A single-photon turnstile device”. *Nature*, **397**:500–503, (1999).
- [332] F. De Martini, G. Di Giuseppe, and M. Marrocco. “Single-mode generation of quantum photon states by excited single molecules in a microcavity trap”. *Physical Review Letters*, **76**(6):900, (1996).

- [333] C. Brunel, B. Lounis, P. Tamarat, and M. Orrit. “Triggered source of single photons based on controlled single molecule fluorescence”. *Physical Review Letters*, **83**(14):2722, (1999).
- [334] A. Kuhn, M. Hennrich, and G. Rempe. “Deterministic single-photon source for distributed quantum networking”. *Physical Review Letters*, **89**(6):067901, (2002).
- [335] J. McKeever, A. Boca, A. Boozer, R. Miller, J. Buck, A. Kuzmich, and H. Kimble. “Deterministic generation of single photons from one atom trapped in a cavity”. *Science*, **303**(5666):1992–1994, (2004).
- [336] C. Maurer, C. Becher, C. Russo, J. Eschner, and R. Blatt. “A single-photon source based on a single Ca^+ ion”. *New Journal of Physics*, **6**(1):94, (2004).
- [337] B. B. Blinov, D. L. Moehring, L.-M. Duan, and C. Monroe. “Observation of entanglement between a single trapped atom and a single photon”. *Nature*, **428**:153–157, (2004).
- [338] M. Keller, B. Lange, K. Hayasaka, W. Lange, and H. Walther. “Continuous generation of single photons with controlled waveform in an ion-trap cavity system”. *Nature*, **431**:1075–1078, (2004).
- [339] C. Kurtsiefer, S. Mayer, P. Zarda, and H. Weinfurter. “Stable solid-state source of single photons”. *Physical Review Letters*, **85**(2):290, (2000).
- [340] A. Beveratos, R. Brouri, T. Gacoin, J.-P. Poizat, and P. Grangier. “Nonclassical radiation from diamond nanocrystals”. *Physical Review A*, **64**(6):061802, (2001).
- [341] A. Kuzmich, W. Bowen, A. Boozer, A. Boca, C. Chou, L.-M. Duan, and H. Kimble. “Generation of nonclassical photon pairs for scalable quantum communication with atomic ensembles”. *Nature*, **423**:731–734, (2003).
- [342] C. H. van der Wal, M. D. Eisaman, A. André, R. L. Walsworth, D. F. Phillips, A. S. Zibrov, and M. D. Lukin. “Atomic memory for correlated photon states”. *Science*, **301**(5630):196–200, (2003).

- [343] J. Benedikter, H. Kaupp, T. Hümmer, Y. Liang, A. Bommer, C. Becher, A. Krueger, J. M. Smith, T. W. Hänsch, and D. Hunger. “Cavity-enhanced single-photon source based on the silicon-vacancy center in diamond”. *Physical Review Applied*, **7**(2):024031, (2017).
- [344] S. Haffouz, K. D. Zeuner, D. Dalacu, P. J. Poole, J. Lapointe, D. Poitras, K. Mnaymneh, X. Wu, M. Couillard, M. Korkusinski, *et al.* “Bright single InAsP quantum dots at telecom wavelengths in position-controlled InP nanowires: the role of the photonic waveguide”. *Nano Letters*, **18**(5):3047–3052, (2018).
- [345] B. Y. Zel’Dovich and D. Klyshko. “Field statistics in parametric luminescence”. *Soviet Journal of Experimental and Theoretical Physics Letters*, **9**:40, (1969).
- [346] D. C. Burnham and D. L. Weinberg. “Observation of simultaneity in parametric production of optical photon pairs”. *Physical Review Letters*, **25**(2):84, (1970).
- [347] J. Rarity, P. Tapster, and E. Jakeman. “Observation of sub-Poissonian light in parametric downconversion”. *Optics Communications*, **62**(3):201–206, (1987).
- [348] B. J. Smith, P. Mahou, O. Cohen, J. Lundeen, and I. Walmsley. “Photon pair generation in birefringent optical fibers”. *Optics Express*, **17**(26):23589–23602, (2009).
- [349] X.-C. Yao, T.-X. Wang, P. Xu, H. Lu, G.-S. Pan, X.-H. Bao, C.-Z. Peng, C.-Y. Lu, Y.-A. Chen, and J.-W. Pan. “Observation of eight-photon entanglement”. *Nature Photonics*, **6**(4):225–228, (2012).
- [350] A. D. Semenov, G. N. Gol’tsman, and A. A. Korneev. “Quantum detection by current carrying superconducting film”. *Physica C: Superconductivity*, **351**(4):349–356, (2001).
- [351] G. Gol’tsman, O. Okunev, G. Chulkova, A. Lipatov, A. Semenov, K. Smirnov, B. Voronov, A. Dzardanov, C. Williams, and R. Sobolewski. “Picosecond superconducting single-photon optical detector”. *Applied Physics Letters*, **79**(6):705–707, (2001).

- [352] J. A. Lau, V. B. Verma, D. Schwarzer, and A. M. Wodtke. “Superconducting single-photon detectors in the mid-infrared for physical chemistry and spectroscopy”. *Chemical Society Reviews*, **52**(3):921–941, (2023).
- [353] C. M. Natarajan, M. G. Tanner, and R. H. Hadfield. “Superconducting nanowire single-photon detectors: physics and applications”. *Superconductor Science and Technology*, **25**(6):063001, (2012).
- [354] A. J. Kerman, E. A. Dauler, W. E. Keicher, J. K. Yang, K. K. Berggren, G. Gol’tsman, and B. Voronov. “Kinetic-inductance-limited reset time of superconducting nanowire photon counters”. *Applied Physics Letters*, **88**(11), (2006).
- [355] C. M. Natarajan, L. Zhang, H. Coldenstrodt-Ronge, G. Donati, S. N. Dorenbos, V. Zwiller, I. A. Walmsley, and R. H. Hadfield. “Quantum detector tomography of a time-multiplexed superconducting nanowire single-photon detector at telecom wavelengths”. *Optics Express*, **21**(1):893–902, (2013).
- [356] J. Los, M. Sidorova, B. Lopez-Rodriguez, P. Qualm, J. Chang, S. Steinhauer, V. Zwiller, and I. E. Zadeh. “High-performance photon number resolving detectors for 850–950 nm wavelength range”. *APL Photonics*, **9**(6), (2024).
- [357] D. Zhu, M. Colangelo, C. Chen, B. A. Korzh, F. N. Wong, M. D. Shaw, and K. K. Berggren. “Resolving photon numbers using a superconducting nanowire with impedance-matching taper”. *Nano Letters*, **20**(5):3858–3863, (2020).
- [358] S. W. Nam, A. Lita, D. Rosenberg, and A. J. Miller. “Optical and near-infrared photon detection with superconducting transition-edge sensors”. In *2006 Digest of the LEOS Summer Topical Meetings*, pages 17–18. IEEE, (2006).
- [359] J. Clarke and A. I. Braginski. “Fundamentals and technology of SQUIDS and SQUID systems”. Wiley-VCH, (2004).
- [360] B. Cabrera, R. Clarke, P. Colling, A. Miller, S. Nam, and R. Romani. “Detection of single infrared, optical, and ultraviolet photons using superconducting transition edge sensors”. *Applied Physics Letters*, **73**(6):735–737, (1998).

- [361] T. Gerrits, N. Thomas-Peter, J. C. Gates, A. E. Lita, B. J. Metcalf, B. Calkins, N. A. Tomlin, A. E. Fox, A. L. Linares, J. B. Spring, *et al.* “On-chip, photon-number-resolving, telecommunication-band detectors for scalable photonic information processing”. *Physical Review A*, **84**(6):060301, (2011).
- [362] A. E. Lita, D. Rosenberg, S. Nam, A. J. Miller, D. Balzar, L. Kaatz, and R. Schwall. “Tuning of tungsten thin film superconducting transition temperature for fabrication of photon number resolving detectors”. *IEEE Transactions on Applied Superconductivity*, **15**(2):3528–3531, (2005).
- [363] K. D. Irwin and G. C. Hilton. “Cryogenic particle detection”, pages 63–150. Springer Berlin Heidelberg, (2005).
- [364] A. E. Lita, A. J. Miller, and S. W. Nam. “Counting near-infrared single-photons with 95% efficiency”. *Optics Express*, **16**(5):3032–3040, (2008).
- [365] A. J. Miller, S. W. Nam, J. M. Martinis, and A. V. Sergienko. “Demonstration of a low-noise near-infrared photon counter with multiphoton discrimination”. *Applied Physics Letters*, **83**(4):791–793, (2003).
- [366] Z. H. Levine, T. Gerrits, A. L. Migdall, D. V. Samarov, B. Calkins, A. E. Lita, and S. W. Nam. “Algorithm for finding clusters with a known distribution and its application to photon-number resolution using a superconducting transition-edge sensor”. *JOSA B*, **29**(8):2066–2073, (2012).
- [367] L. Lolli, E. Taralli, and M. Rajteri. “Ti/Au TES to discriminate single photons”. *Journal of Low Temperature Physics*, **167**:803–808, (2012).
- [368] T. Gerrits, B. Calkins, N. Tomlin, A. E. Lita, A. Migdall, R. Mirin, and S. W. Nam. “Extending single-photon optimized superconducting transition edge sensors beyond the single-photon counting regime”. *Optics Express*, **20**(21):23798–23810, (2012).
- [369] N. Dalbec-Constant, G. Thekkadath, D. England, B. Sussman, T. Gerrits, and N. Quesada. “Accurate unsupervised photon counting from transition edge sensor signals”. *arXiv preprint arXiv:2411.05737*, (2024).

- [370] K. Wakui, Y. Eto, H. Benichi, S. Izumi, T. Yanagida, K. Ema, T. Numata, D. Fukuda, M. Takeoka, and M. Sasaki. “Ultrabroadband direct detection of nonclassical photon statistics at telecom wavelength”. *Scientific Reports*, **4**(1):4535, (2014).
- [371] T. Young. “I. The Bakerian Lecture. Experiments and calculations relative to physical optics”. *Philosophical Transactions of the Royal Society of London*, **94**:1–16, (1804).
- [372] R. H. Brown and R. Q. Twiss. “Correlation between photons in two coherent beams of light”. *Nature*, **177**:27–29, (1956).
- [373] R. Hanbury Brown and R. Q. Twiss. “A test of a new type of stellar interferometer on sirius”. *Nature*, **178**:1046–1048, (1956).
- [374] J. B. Spring, P. S. Salter, B. J. Metcalf, P. C. Humphreys, M. Moore, N. Thomas-Peter, M. Barbieri, X.-M. Jin, N. K. Langford, W. S. Kolthammer, *et al.* “On-chip low loss heralded source of pure single photons”. *Optics Express*, **21**(11):13522–13532, (2013).
- [375] W. Mauerer, M. Avenhaus, W. Helwig, and C. Silberhorn. “How colors influence numbers: Photon statistics of parametric down-conversion”. *Physical Review A*, **80**(5):053815, (2009).
- [376] K. Garay-Palmett, H. McGuinness, O. Cohen, J. Lundeen, R. Rangel-Rojo, A. U’ren, M. Raymer, C. McKinstrie, S. Radic, and I. Walmsley. “Photon pair-state preparation with tailored spectral properties by spontaneous four-wave mixing in photonic-crystal fiber”. *Optics Express*, **15**(22):14870–14886, (2007).
- [377] A. Christ and C. Silberhorn. “Limits on the deterministic creation of pure single-photon states using parametric down-conversion”. *Physical Review A*, **85**(2):023829, (2012).
- [378] A. Cameron. “Measuring quantum correlations of polarization, spatial mode, and energy-time entangled photon pairs”. PhD thesis, University of Waterloo, (2023).

- [379] K. Zielnicki, K. Garay-Palmett, D. Cruz-Delgado, H. Cruz-Ramirez, M. F. O’Boyle, B. Fang, V. O. Lorenz, A. B. U’Ren, and P. G. Kwiat. “Joint spectral characterization of photon-pair sources”. *Journal of Modern Optics*, **65**(10):1141–1160, (2018).
- [380] A. Eckstein, G. Boucher, A. Lemaître, P. Filloux, I. Favero, G. Leo, J. E. Sipe, M. Liscidini, and S. Ducci. “High-resolution spectral characterization of two photon states via classical measurements”. *Laser & Photonics Reviews*, **8**(5):L76–L80, (2014).
- [381] A. O. Davis, P. M. Saulnier, M. Karpiński, and B. J. Smith. “Pulsed single-photon spectrometer by frequency-to-time mapping using chirped fiber Bragg gratings”. *Optics Express*, **25**(11):12804–12811, (2017).
- [382] M. Avenhaus, A. Eckstein, P. J. Mosley, and C. Silberhorn. “Fiber-assisted single-photon spectrograph”. *Optics Letters*, **34**(18):2873–2875, (2009).
- [383] G. Thekkadath, M. Houde, D. England, P. Bustard, F. Bouchard, N. Quesada, and B. Sussman. “Gain-induced group delay in spontaneous parametric down-conversion”. *arXiv preprint arXiv:2405.07909*, (2024).
- [384] W. Wasilewski, P. Wasylczyk, P. Kolenderski, K. Banaszek, and C. Radzewicz. “Joint spectrum of photon pairs measured by coincidence Fourier spectroscopy”. *Optics Letters*, **31**(8):1130–1132, (2006).
- [385] M. Liscidini and J. Sipe. “Stimulated emission tomography”. *Physical Review Letters*, **111**(19):193602, (2013).
- [386] B. Fang, O. Cohen, M. Liscidini, J. E. Sipe, and V. O. Lorenz. “Fast and highly resolved capture of the joint spectral density of photon pairs”. *Optica*, **1**(5):281–284, (2014).
- [387] J. Erskine, D. England, C. Kupchak, and B. Sussman. “Real-time spectral characterization of a photon pair source using a chirped supercontinuum seed”. *Optics Letters*, **43**(4):907–910, (2018).

- [388] I. Jizan, B. Bell, L. G. Helt, A. C. Bedoya, C. Xiong, and B. J. Eggleton. “Phase-sensitive tomography of the joint spectral amplitude of photon pair sources”. *Optics Letters*, **41**(20):4803–4806, (2016).
- [389] J.-P. W. MacLean, S. Schwarz, and K. J. Resch. “Reconstructing ultrafast energy-time-entangled two-photon pulses”. *Physical Review A*, **100**(3):033834, (2019).
- [390] O. Kuzucu, F. N. Wong, S. Kurimura, and S. Tovstonog. “Joint temporal density measurements for two-photon state characterization”. *Physical Review Letters*, **101**(15):153602, (2008).
- [391] M. Yabuno, T. Takumi, F. China, S. Miki, H. Terai, P. J. Mosley, R.-B. Jin, and R. Shimizu. “Ultrafast measurement of a single-photon wave packet using an optical Kerr gate”. *Optics Express*, **30**(4):4999–5007, (2022).
- [392] S. Cheng. “Single photon measurements: from spin-orbit lattices to ultrafast pulses”. Master’s thesis, University of Waterloo, (2021).
- [393] Q. A. Turchette, C. J. Hood, W. Lange, H. Mabuchi, and H. J. Kimble. “Measurement of conditional phase shifts for quantum logic”. *Physical Review Letters*, **75**(25):4710, (1995).
- [394] N. J. Cerf, C. Adami, and P. G. Kwiat. “Optical simulation of quantum logic”. *Physical Review A*, **57**(3):R1477, (1998).
- [395] J. C. Howell and J. A. Yeazell. “Reducing the complexity of linear optics quantum circuits”. *Physical Review A*, **61**(5):052303, (2000).
- [396] P. Kwiat, J. Mitchell, P. Schwindt, and A. White. “Grover’s search algorithm: an optical approach”. *Journal of Modern Optics*, **47**(2-3):257–266, (2000).
- [397] T. C. Ralph and G. J. Pryde. “Optical quantum computation”. In *Progress in Optics*, volume 54, pages 209–269. Elsevier, (2010).
- [398] P. Kok, W. J. Munro, K. Nemoto, T. C. Ralph, J. P. Dowling, and G. J. Milburn. “Linear optical quantum computing with photonic qubits”. *Reviews of Modern Physics*, **79**(1):135–174, (2007).

- [399] J. L. O’Brien, A. Furusawa, and J. Vučković. “Photonic quantum technologies”. *Nature Photonics*, **3**(12):687–695, (2009).
- [400] T. D. Ladd, F. Jelezko, R. Laflamme, Y. Nakamura, C. Monroe, and J. L. O’Brien. “Quantum computers”. *Nature*, **464**:45–53, (2010).
- [401] S. Slussarenko and G. J. Pryde. “Photonic quantum information processing: A concise review”. *Applied Physics Reviews*, **6**(4), (2019).
- [402] M. Reck, A. Zeilinger, H. J. Bernstein, and P. Bertani. “Experimental realization of any discrete unitary operator”. *Physical Review Letters*, **73**(1):58, (1994).
- [403] W. R. Clements, P. C. Humphreys, B. J. Metcalf, W. S. Kolthammer, and I. A. Walmsley. “Optimal design for universal multiport interferometers”. *Optica*, **3**(12):1460–1465, (2016).
- [404] M. A. Nielsen. “Optical quantum computation using cluster states”. *Physical Review Letters*, **93**(4):040503, (2004).
- [405] A. Zeilinger. “General properties of lossless beam splitters in interferometry”. *American Journal of Physics*, **49**(9):882–883, (1981).
- [406] H. Fearn and R. Loudon. “Quantum theory of the lossless beam splitter”. *Optics Communications*, **64**(6):485–490, (1987).
- [407] F. Bloch. “Nuclear induction”. *Physical Review*, **70**(7-8):460, (1946).
- [408] H. Poincaré. “Théorie mathématique de la lumière”. *Paris*, **2**:275–285, (1892).
- [409] M. M. Mirsalehi, T. K. Gaylord, and E. I. Verriest. “Integrated-optical Givens rotation device”. *Applied Optics*, **25**(10):1608–1614, (1986).
- [410] D. A. Miller. “Self-configuring universal linear optical component”. *Photonics Research*, **1**(1):1–15, (2013).
- [411] D. A. Miller. “Perfect optics with imperfect components”. *Optica*, **2**(8):747–750, (2015).

- [412] B. Yurke, S. L. McCall, and J. R. Klauder. “SU(2) and SU(1,1) interferometers”. *Physical Review A*, **33**(6):4033, (1986).
- [413] L. Zehnder. “Ein neuer interferenzrefraktor”, (1891).
- [414] L. Mach. “Ueber einen interferenzrefraktor”. *Zeitschrift für Instrumentenkunde*, **12**(3):89, (1892).
- [415] L. Troyansky and N. Tishby. “On the quantum evaluation of the determinant and the permanent of a matrix”. In *Proceedings of the 4th Workshop on Physics and Computation*, volume 96. Citeseer, (1996).
- [416] C. S. Hamilton, R. Kruse, L. Sansoni, S. Barkhofen, C. Silberhorn, and I. Jex. “Gaussian boson sampling”. *Physical Review Letters*, **119**(17):170501, (2017).
- [417] A. P. Lund, A. Laing, S. Rahimi-Keshari, T. Rudolph, J. L. O’Brien, and T. C. Ralph. “Boson sampling from a Gaussian state”. *Physical Review Letters*, **113**(10):100502, (2014).
- [418] R. Raussendorf, D. E. Browne, and H. J. Briegel. “Measurement-based quantum computation on cluster states”. *Physical Review A*, **68**(2):022312, (2003).
- [419] P. Walther, K. J. Resch, T. Rudolph, E. Schenck, H. Weinfurter, V. Vedral, M. Aspelmeyer, and A. Zeilinger. “Experimental one-way quantum computing”. *Nature*, **434**:169–176, (2005).
- [420] H.-S. Zhong, Y. Li, W. Li, L.-C. Peng, Z.-E. Su, Y. Hu, Y.-M. He, X. Ding, W. Zhang, H. Li, *et al.* “12-photon entanglement and scalable scattershot boson sampling with optimal entangled-photon pairs from parametric down-conversion”. *Physical Review Letters*, **121**(25):250505, (2018).
- [421] C. Greganti, M.-C. Roehsner, S. Barz, T. Morimae, and P. Walther. “Demonstration of measurement-only blind quantum computing”. *New Journal of Physics*, **18**(1):013020, (2016).
- [422] S. Barz, E. Kashefi, A. Broadbent, J. F. Fitzsimons, A. Zeilinger, and P. Walther. “Demonstration of blind quantum computing”. *Science*, **335**(6066):303–308, (2012).

- [423] R. Prevedel, P. Walther, F. Tiefenbacher, P. Böhi, R. Kaltenbaek, T. Jennewein, and A. Zeilinger. “High-speed linear optics quantum computing using active feed-forward”. *Nature*, **445**:65–69, (2007).
- [424] M. Varnava, D. E. Browne, and T. Rudolph. “How good must single photon sources and detectors be for efficient linear optical quantum computation?”. *Physical Review Letters*, **100**(6):060502, (2008).
- [425] M. Gimeno-Segovia, P. Shadbolt, D. E. Browne, and T. Rudolph. “From three-photon Greenberger-Horne-Zeilinger states to ballistic universal quantum computation”. *Physical Review Letters*, **115**(2):020502, (2015).
- [426] D. E. Browne and T. Rudolph. “Resource-efficient linear optical quantum computation”. *Physical Review Letters*, **95**(1):010501, (2005).
- [427] F. Ewert and P. van Loock. “3/4-efficient bell measurement with passive linear optics and unentangled ancillae”. *Physical Review Letters*, **113**(14):140403, (2014).
- [428] M. Varnava, D. E. Browne, and T. Rudolph. “Loss tolerance in one-way quantum computation via counterfactual error correction”. *Physical Review Letters*, **97**(12):120501, (2006).
- [429] T. Rudolph. “Why I am optimistic about the silicon-photonics route to quantum computing”. *APL Photonics*, **2**(3), (2017).
- [430] S. Morley-Short, M. Gimeno-Segovia, T. Rudolph, and H. Cable. “Loss-tolerant teleportation on large stabilizer states”. *Quantum Science and Technology*, **4**(2):025014, (2019).
- [431] M. I. Dyakonov. “Will we ever have a quantum computer?”, volume 22. Springer, (2020).
- [432] A. Ambainis. “Quantum walk algorithm for element distinctness”. *SIAM Journal on Computing*, **37**(1):210–239, (2007).
- [433] F. Magniez, M. Santha, and M. Szegedy. “Quantum algorithms for the triangle problem”. *SIAM Journal on Computing*, **37**(2):413–424, (2007).

- [434] E. Farhi, J. Goldstone, and S. Gutmann. “A quantum algorithm for the Hamiltonian NAND tree”. *arXiv preprint quant-ph/0702144*, (2007).
- [435] N. Masuda, M. A. Porter, and R. Lambiotte. “Random walks and diffusion on networks”. *Physics Reports*, **716**:1–58, (2017).
- [436] M. Kac. “Random walk and the theory of brownian motion”. *The American Mathematical Monthly*, **54**(7P1):369–391, (1947).
- [437] J. E. Neigel and J. C. Avise. “Application of a random walk model to geographic distributions of animal mitochondrial DNA variation.”. *Genetics*, **135**(4):1209–1220, (1993).
- [438] M. D. Godfrey, C. W. Granger, and O. Morgenstern. “The random-walk hypothesis of stock market behavior”. *Kyklos*, **17**(1):1–30, (1964).
- [439] R. Engbert, K. Mergenthaler, P. Sinn, and A. Pikovsky. “An integrated model of fixational eye movements and microsaccades”. *Proceedings of the National Academy of Sciences*, **108**(39):E765–E770, (2011).
- [440] L. Grady. “Random walks for image segmentation”. *IEEE Transactions on Pattern Analysis and Machine Intelligence*, **28**(11):1768–1783, (2006).
- [441] S. Brooks. “Markov chain Monte Carlo method and its application”. *Journal of the Royal Statistical Society: Series D (the Statistician)*, **47**(1):69–100, (1998).
- [442] Y. Aharonov, L. Davidovich, and N. Zagury. “Quantum random walks”. *Physical Review A*, **48**(2):1687, (1993).
- [443] E. Farhi and S. Gutmann. “Quantum computation and decision trees”. *Physical Review A*, **58**(2):915, (1998).
- [444] N. B. Lovett, S. Cooper, M. Everitt, M. Trevers, and V. Kendon. “Universal quantum computation using the discrete-time quantum walk”. *Physical Review A*, **81**(4):042330, (2010).
- [445] D. Witthaut. “Quantum walks and quantum simulations with Bloch-oscillating spinor atoms”. *Physical Review A*, **82**(3):033602, (2010).

- [446] T. Kitagawa, M. A. Broome, A. Fedrizzi, M. S. Rudner, E. Berg, I. Kassal, A. Aspuru-Guzik, E. Demler, and A. G. White. “Observation of topologically protected bound states in photonic quantum walks”. *Nature Communications*, **3**(1):882, (2012).
- [447] J. K. Asbóth. “Symmetries, topological phases, and bound states in the one-dimensional quantum walk”. *Physical Review B*, **86**(19):195414, (2012).
- [448] C.-W. Lee, P. Kurzyński, and H. Nha. “Quantum walk as a simulator of nonlinear dynamics: Nonlinear Dirac equation and solitons”. *Physical Review A*, **92**(5):052336, (2015).
- [449] F. Cardano, M. Maffei, F. Massa, B. Piccirillo, C. De Lisio, G. De Filipis, V. Cataudella, E. Santamato, and L. Marrucci. “Statistical moments of quantum-walk dynamics reveal topological quantum transitions”. *Nature Communications*, **7**(1):11439, (2016).
- [450] K. Bepari, S. Malik, M. Spannowsky, and S. Williams. “Quantum walk approach to simulating parton showers”. *Physical Review D*, **106**(5):056002, (2022).
- [451] N. Shenvi, J. Kempe, and K. B. Whaley. “Quantum random-walk search algorithm”. *Physical Review A*, **67**(5):052307, (2003).
- [452] A. M. Childs and J. Goldstone. “Spatial search by quantum walk”. *Physical Review A*, **70**(2):022314, (2004).
- [453] D. Qu, S. Marsh, K. Wang, L. Xiao, J. Wang, and P. Xue. “Deterministic search on star graphs via quantum walks”. *Physical Review Letters*, **128**(5):050501, (2022).
- [454] O. Mülken and A. Blumen. “Continuous-time quantum walks: Models for coherent transport on complex networks”. *Physics Reports*, **502**(2-3):37–87, (2011).
- [455] F. Flamini, M. Krumm, L. J. Fiderer, T. Müller, and H. J. Briegel. “Towards interpretable quantum machine learning via single-photon quantum walks”. *Quantum Science and Technology*, (2023).

- [456] A. M. Childs. “Universal computation by quantum walk”. *Physical Review Letters*, **102**(18):180501, (2009).
- [457] Y. Cai, J. Roslund, G. Ferrini, F. Arzani, X. Xu, C. Fabre, and N. Treps. “Multimode entanglement in reconfigurable graph states using optical frequency combs”. *Nature Communications*, **8**(1):15645, (2017).
- [458] B. Brecht, D. V. Reddy, C. Silberhorn, and M. G. Raymer. “Photon temporal modes: a complete framework for quantum information science”. *Physical Review X*, **5**(4):041017, (2015).
- [459] V. Averchenko, D. Sych, G. Schunk, U. Vogl, C. Marquardt, and G. Leuchs. “Temporal shaping of single photons enabled by entanglement”. *Physical Review A*, **96**(4):043822, (2017).
- [460] V. Ansari, E. Roccia, M. Santandrea, M. Doostdar, C. Eigner, L. Padberg, I. Gianani, M. Sbroscia, J. M. Donohue, L. Mancino, *et al.* “Heralded generation of high-purity ultrashort single photons in programmable temporal shapes”. *Optics Express*, **26**(3):2764–2774, (2018).
- [461] M. Erhard, R. Fickler, M. Krenn, and A. Zeilinger. “Twisted photons: new quantum perspectives in high dimensions”. *Light: Science & Applications*, **7**(3):17146–17146, (2018).
- [462] H. Jayakumar, A. Predojević, T. Kauten, T. Huber, G. S. Solomon, and G. Weihs. “Time-bin entangled photons from a quantum dot”. *Nature Communications*, **5**(1):4251, (2014).
- [463] F. Bouchard. “Quantum cryptography beyond qubits”. PhD thesis, Université d’Ottawa/University of Ottawa, (2019).
- [464] A. Eckstein, B. Brecht, and C. Silberhorn. “A quantum pulse gate based on spectrally engineered sum frequency generation”. *Optics Express*, **19**(15):13770–13778, (2011).

- [465] D. A. Vajner, N. D. Kewitz, M. von Helversen, S. C. Wein, Y. Karli, F. Kappe, V. Remesh, S. F. C. da Silva, A. Rastelli, G. Weihs, *et al.* “Towards photon-number-encoded high-dimensional entanglement from a sequentially excited quantum three-level system”. *arXiv preprint arXiv:2407.05902*, (2024).
- [466] W.-B. Gao, C.-Y. Lu, X.-C. Yao, P. Xu, O. Gühne, A. Goebel, Y.-A. Chen, C.-Z. Peng, Z.-B. Chen, and J.-W. Pan. “Experimental demonstration of a hyper-entangled ten-qubit Schrödinger cat state”. *Nature Physics*, **6**(5):331–335, (2010).
- [467] T. M. Graham, H. J. Bernstein, T.-C. Wei, M. Junge, and P. G. Kwiat. “Superdense teleportation using hyperentangled photons”. *Nature Communications*, **6**(1):7185, (2015).
- [468] X.-L. Wang, X.-D. Cai, Z.-E. Su, M.-C. Chen, D. Wu, L. Li, N.-L. Liu, C.-Y. Lu, and J.-W. Pan. “Quantum teleportation of multiple degrees of freedom of a single photon”. *Nature*, **518**:516–519, (2015).
- [469] M. Malik, M. Erhard, M. Huber, M. Krenn, R. Fickler, and A. Zeilinger. “Multi-photon entanglement in high dimensions”. *Nature Photonics*, **10**(4):248–252, (2016).
- [470] X.-L. Wang, Y.-H. Luo, H.-L. Huang, M.-C. Chen, Z.-E. Su, C. Liu, C. Chen, W. Li, Y.-Q. Fang, X. Jiang, *et al.* “18-qubit entanglement with six photons’ three degrees of freedom”. *Physical Review Letters*, **120**(26):260502, (2018).
- [471] H. J. Briegel, D. E. Browne, W. Dür, R. Raussendorf, and M. Van den Nest. “Measurement-based quantum computation”. *Nature Physics*, **5**(1):19–26, (2009).
- [472] X.-Y. Xu, Q.-Q. Wang, W.-W. Pan, K. Sun, J.-S. Xu, G. Chen, J.-S. Tang, M. Gong, Y.-J. Han, C.-F. Li, *et al.* “Measuring the winding number in a large-scale chiral quantum walk”. *Physical Review Letters*, **120**(26):260501, (2018).
- [473] Q.-Q. Wang, X.-Y. Xu, W.-W. Pan, K. Sun, J.-S. Xu, G. Chen, Y.-J. Han, C.-F. Li, and G.-C. Guo. “Dynamic-disorder-induced enhancement of entanglement in photonic quantum walks”. *Optica*, **5**(9):1136–1140, (2018).

- [474] Q.-Q. Wang, S. Dong, X.-W. Li, X.-Y. Xu, C. Wang, S. Han, M.-H. Yung, Y.-J. Han, C.-F. Li, and G.-C. Guo. “Efficient learning of mixed-state tomography for photonic quantum walk”. *Science Advances*, **10**(11):eadl4871, (2024).
- [475] H. L. Helmholtz. “On the sensations of tone as a physiological basis for the theory of music”. Cambridge University Press, (2009).
- [476] J. J. Dammerud. “Stage acoustics for symphony orchestras in concert halls”. *The Journal of the Acoustical Society of America*, **128**(1):506–506, (2010).
- [477] B. P. Abbott, R. Abbott, T. Abbott, M. Abernathy, F. Acernese, K. Ackley, C. Adams, T. Adams, P. Addesso, R. X. Adhikari, *et al.* “Observation of gravitational waves from a binary black hole merger”. *Physical Review Letters*, **116**(6):061102, (2016).
- [478] D. Stahlke. “Quantum interference as a resource for quantum speedup”. *Physical Review A*, **90**(2):022302, (2014).
- [479] C.-K. Hong, Z.-Y. Ou, and L. Mandel. “Measurement of subpicosecond time intervals between two photons by interference”. *Physical Review Letters*, **59**(18):2044, (1987).
- [480] M. Żukowski, A. Zeilinger, M. A. Horne, and A. K. Ekert. “ “Event-ready-detectors” Bell experiment via entanglement swapping”. *Physical Review Letters*, **71**(26):4287, (1993).
- [481] Z.-Y. J. Ou. “Multi-photon quantum interference”, volume 43. Springer, (2007).
- [482] F. Bouchard, A. Sit, Y. Zhang, R. Fickler, F. M. Miatto, Y. Yao, F. Sciarrino, and E. Karimi. “Two-photon interference: the Hong-Ou-Mandel effect”. *Reports on Progress in Physics*, **84**(1):012402, (2020).
- [483] Y.-H. Luo, H.-S. Zhong, M. Erhard, X.-L. Wang, L.-C. Peng, M. Krenn, X. Jiang, L. Li, N.-L. Liu, C.-Y. Lu, *et al.* “Quantum teleportation in high dimensions”. *Physical Review Letters*, **123**(7):070505, (2019).

- [484] M. C. Tichy, F. Mintert, and A. Buchleitner. “Limits to multipartite entanglement generation with bosons and fermions”. *Physical Review A*, **87**(2):022319, (2013).
- [485] K. Poullos, R. Keil, D. Fry, J. D. Meinecke, J. C. Matthews, A. Politi, M. Lobino, M. Gräfe, M. Heinrich, S. Nolte, *et al.* “Quantum walks of correlated photon pairs in two-dimensional waveguide arrays”. *Physical Review Letters*, **112**(14):143604, (2014).
- [486] J. O. Owens, M. A. Broome, D. N. Biggerstaff, M. E. Goggin, A. Fedrizzi, T. Linjordet, M. Ams, G. D. Marshall, J. Twamley, M. J. Withford, *et al.* “Two-photon quantum walks in an elliptical direct-write waveguide array”. *New Journal of Physics*, **13**(7):075003, (2011).
- [487] A. Crespi, R. Osellame, R. Ramponi, V. Giovannetti, R. Fazio, L. Sansoni, F. De Nicola, F. Sciarrino, and P. Mataloni. “Anderson localization of entangled photons in an integrated quantum walk”. *Nature Photonics*, **7**(4):322–328, (2013).
- [488] N. Killoran, J. Izaac, N. Quesada, V. Bergholm, M. Amy, and C. Weedbrook. “Strawberry fields: A software platform for photonic quantum computing”. *Quantum*, **3**:129, (2019).
- [489] T. R. Bromley, J. M. Arrazola, S. Jahangiri, J. Izaac, N. Quesada, A. D. Gran, M. Schuld, J. Swinarton, Z. Zabaneh, and N. Killoran. “Applications of near-term photonic quantum computers: software and algorithms”. *Quantum Science and Technology*, **5**(3):034010, (2020).
- [490] H. Qi, D. J. Brod, N. Quesada, and R. García-Patrón. “Regimes of classical simulability for noisy gaussian boson sampling”. *Physical Review Letters*, **124**(10):100502, (2020).
- [491] T. Pittman, B. Jacobs, and J. Franson. “Single photons on pseudodemand from stored parametric down-conversion”. *Physical Review A*, **66**(4):042303, (2002).

- [492] F. Kaneda, B. G. Christensen, J. J. Wong, H. S. Park, K. T. McCusker, and P. G. Kwiat. “Time-multiplexed heralded single-photon source”. *Optica*, **2**(12):1010–1013, (2015).
- [493] S. I. Davis, A. Mueller, R. Valivarthi, N. Lauk, L. Narvaez, B. Korzh, A. D. Beyer, O. Cerri, M. Colangelo, K. K. Berggren, *et al.* “Improved heralded single-photon source with a photon-number-resolving superconducting nanowire detector”. *Physical Review Applied*, **18**(6):064007, (2022).
- [494] L. Stasi, P. Caspar, T. Brydges, H. Zbinden, F. Bussi eres, and R. Thew. “High-efficiency photon-number-resolving detector for improving heralded single-photon sources”. *Quantum Science and Technology*, **8**(4):045006, (2023).
- [495] R. E. Wagner and W. J. Tomlinson. “Coupling efficiency of optics in single-mode fiber components”. *Applied Optics*, **21**(15):2671–2688, (1982).
- [496] S. Perumal Sankar, N. Hariharan, and R. Varatharajan. “A novel method to increase the coupling efficiency of laser to single mode fibre”. *Wireless Personal Communications*, **87**(2):419–430, (2016).
- [497] D. Bauch, N. K ocher, N. Heinisch, and S. Schumacher. “Time-bin entanglement in the deterministic generation of linear photonic cluster states”. *arXiv preprint arXiv:2403.08527*, (2024).
- [498] G. Gustafson, S. Prestel, M. Spannowsky, and S. Williams. “Collider events on a quantum computer”. *Journal of High Energy Physics*, **2022**(11):1–27, (2022).
- [499] M. Y. Niu, S. Boixo, V. N. Smelyanskiy, and H. Neven. “Universal quantum control through deep reinforcement learning”. *npj Quantum Information*, **5**(1):33, (2019).
- [500] S. Mavadia, V. Frey, J. Sastrawan, S. Dona, and M. J. Biercuk. “Prediction and real-time compensation of qubit decoherence via machine learning”. *Nature Communications*, **8**(1):14106, (2017).