

PROBING COLLECTIVE MULTI-ELECTRON EFFECTS WITH FEW CYCLE LASER PULSES

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1. Please continue.
2. The experiment requires that you continue.
3. It is absolutely essential that you continue.
4. You have no other choice, you must go on.

Stanley Milgram, 1961.

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1.e4 e5 2.Nf3 Nc6 3.Bb5 a6 4.Ba4 Nf6 5.0-0 Be7 6.Re1 b5 7.Bb3 d6 8.c3 0-0 9.h3 h6
10.d4 Re8 11.Nbd2 Bf8 12.Nf1 Bd7 13.Ng3 Na5 14.Bc2 c5 15.b3 Nc6 16.d5 Ne7 17.Be3
Ng6 18.Qd2 Nh7 19.a4 Nh4 20.Nxh4 Qxh4 21.Qe2 Qd8 22.b4 Qc7 23.Rec1 c4 24.Ra3 Rec8
25.Rca1 Qd8 26.f4 Nf6 27.fxe5 dxe5 28.Qf1 Ne8 29.Qf2 Nd6 30.Bb6 Qe8 31.R3a2 Be7
32.Bc5 Bf8 33.Nf5 Bxf5 34.exf5 f6 35.Bxd6 Bxd6 36.axb5 axb5 37.Be4 Rxa2 38.Qxa2
Qd7 39.Qa7 Rc7 40.Qb6 Rb7 41.Ra8+ Kf7 42.Qa6 Qc7 43.Qc6 Qb6+ 44.Kf1 Rb8 45.Ra6 10

Abstract

High Harmonic Generation (HHG) enables the production of bursts of coherent soft x-rays with attosecond pulse duration. This process arises from the nonlinear interaction between intense infrared laser pulses and an ionizing gas medium. Soft x-ray photons are used for spectroscopy of inner-shell electron correlation and exchange processes, and the availability of attosecond pulse durations will enable these processes to be resolved on their natural time scales. The maximum or *cutoff* photon energy in HHG increases with both the intensity as well as the wavelength of the driving laser. It is highly desirable to increase the harmonic cutoff as this will allow for the generation of shorter attosecond pulses, as well as HHG spectroscopy of increasingly energetic electronic transitions.

While the harmonic cutoff increases with laser wavelength, there is a corresponding decrease in harmonic yield. The first part of this thesis describes the experimental measurement of the wavelength scaling of HHG efficiency, which we report as $\lambda^{-6.3 \pm 1.1}$ in xenon, and $\lambda^{-6.5 \pm 1.1}$ in krypton.

To increase the HHG cutoff, we have developed a $1.8 \mu\text{m}$ source, with stable carrier envelope phase and a pulse duration of < 2 optical cycles. The $1.8 \mu\text{m}$ wavelength allowed for a significant increase in the harmonic cutoff compared to equivalent 800 nm sources, while still maintaining reasonable harmonic yield. By focusing this source into neon we have produced 400 eV harmonics that extend into the x-ray water window.

In addition to providing a source of photons for a secondary target, the HHG spectrum carries the signature of the electronic structure of the generating medium. In krypton we observed a Cooper minimum at 85 eV, showing that photoionization cross sections can be measured with HHG. Measurements in xenon lead to the first clear observation of electron correlation effects during HHG, which manifest as a broad peak in the HHG spectrum centred at 100 eV.

This thesis also describes several improvements to the HHG experiment including the development of an ionization detector for measuring laser intensity, as well

as an investigation into the role of laser mode quality on HHG phase matching and efficiency.

Résumé

La génération d'harmoniques élevées (GHE) est un processus créant des rayons X mous sous forme d'impulsions attosecondes cohérentes. Ces impulsions sont créées à partir de l'interaction non-linéaire entre d'intenses impulsions laser infrarouge et un médium gazeux ionisant. Les rayons X sont utilisés en spectroscopie pour étudier les corrélations et processus d'échange des électrons de coeur et la disponibilité d'une source attoseconde rend possible la résolution temporelle de ces phénomènes. L'énergie maximale ou *seuil* des photons produits par GHE augmente avec l'intensité et la longueur d'onde des impulsions laser infrarouge. Augmenter ce seuil représente un intérêt capital car les impulsions attosecondes créées sont alors plus brèves et des transitions spectroscopiques plus énergétiques peuvent être observées.

Bien que l'énergie seuil augmente avec la longueur d'onde du laser infrarouge, l'intensité des harmoniques générées va en diminuant. La première partie de cette thèse consiste à décrire expérimentalement la dépendance en longueur d'onde de l'efficacité de GHE, que nous trouvons être $\lambda^{-6.3 \pm 1.1}$ dans le xénon et $\lambda^{-6.5 \pm 1.1}$ dans le krypton.

Pour augmenter le seuil, nous avons développé une source infrarouge de $1.8 \mu\text{m}$ de phase porteuse-enveloppe stable et de durée d'impulsions correspondant à deux cycles optiques. Une source de $1.8 \mu\text{m}$ permet une augmentation significative du seuil par rapport aux sources de 800nm tout en conservant une efficacité de génération raisonnable. En focalisant cette source dans du néon, nous avons produit des harmoniques de 400 eV s'étendant jusque dans la fenêtre de l'eau des rayons X.

En plus de fournir une source de photons utilisée pour étudier un deuxième médium, le spectre de GHE contient la signature de la structure électronique du premier médium, où la génération a eu lieu. Dans le krypton, nous avons observé un minimum de Cooper à 85 eV, démontrant ainsi que les sections efficaces de photoionisation peuvent être mesurées avec la GHE. Des mesures prises dans le xénon ont mené à la première observation claire d'effets de corrélations d'électrons durant la

GHE, qui se manifestent par un large pic du spectre de GHE centré à 100 eV.

Cette thèse décrit également diverses améliorations aux expériences de GHE, dont le développement d'un détecteur d'ions pour mesurer l'intensité du laser, ainsi qu'une étude sur les effets de la qualité du mode laser sur l'efficacité et l'accord de phase dans la GHE.

Acknowledgements

You do not need to look very far in the world's universities, or in Canadian industry, to find people who have spent time working at the Canadian National Research Council (NRC). Many whom I have met describe their time at the NRC as among the happiest and most professionally satisfying of their lives. I have had a long relationship with the NRC starting with several undergraduate summer student positions, followed by a Master's degree at the Institute for National Measurement Standards and now PhD work at the Steacie Institute for Molecular Sciences. I feel incredibly lucky to have grown up in this organization where I have benefited from inspired leadership, a diversity of good advice and the freedom to try my ideas and explore. I would especially like to thank my supervisor David Villeneuve for his patience and support. It was great to find a supervisor who also likes to do physics (and answer emails) on Christmas Eve, and who despite spending most days in his office is still happiest in the lab. I have also benefited greatly from the advice and guidance of Paul Corkum whose enthusiasm for physics is infectious. The combination of Paul and Dave is an amazing team that never runs short of ideas and with whom every year seems to be more exciting than the last. I will truly miss working with them. Much of the experimental work for this PhD was conducted at the Advanced Laser Light Source (ALLS) in Verennes Quebec. At ALLS I collaborated closely with the team lead by Francois Légaré, who is a former student of Paul's. I first met Francois during a brief car ride near the end of his PhD when he was excitedly telling me all about how to compress 800 nm pulses. Several years later his team is compressing high energy infrared pulses and most conversations still end with 'we will build a bigger laser!' (Not bad for a chemist). Along the way I have worked with a number of great researchers whom I wish to thank, these include Nathaniel Kajumba, Heide Bandulet, Daniel Comtois, Mathieu Gigèure, Eric Bisson, Philippe Lassonde, Julien Bertrand, Andre Staudte, Kevin Lee, Genevieve Gariepy, Noah Puskas, Chris Smeenk and Giulio Vampa. I would like to give special mention to the two post doc-

toral fellows that I worked most closely with. For the wavelength scaling experiments I worked with Carlos Trallero for countless days and nights over several beam times spanning a period of years. The wavelength scaling experiments were by far the most difficult of this PhD and Carlos' ability to maintain a positive attitude and to truly work hard in the lab made this experiment possible. It is remarkable how almost anything that can happen in the lab has an analogous, and *hilarious*, story from Carlos' childhood in Cuba. Pulse compression at $1.8\text{ }\mu\text{m}$ was lead by Bruno Schmidt. We were all set to make harmonics at $1.4\text{ }\mu\text{m}$ when he fortunately ignored my advice to 'get going' with the experiment and decided to try to compress the idler with bulk fused silica. Bruno's willingness to try new things, relentless drive and proven ability with nonlinear optics has enabled him to build one of the best laser sources in the world for high harmonic generation. I would also like to thank Kyung Taec Kim who I started to work with toward the end of my PhD. Kyung Taec taught me about phase matching and the role of perturbative fields which really changed my view of high harmonic generation.

At the NRC I benefited from technical assistance from Bert Avery, David Crane and Ryan Kroeker. Most of the experimental work for this PhD was conducted at ALLS which benefits tremendously from a highly dedicated and talented technical staff. In particular I would like to thank François Poitras and Antoine Laramée for their work on the the laser system as well as Guylaine Lavallée for his engineering support.

At the University of Ottawa I had the opportunity to take a number of courses offered by Thomas Brabec. Thomas is an inspired and patient teacher who brings a rare combination of deep theoretical knowledge coupled with a practical understanding of experimental realities to his teaching. I really enjoyed our talks.

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Finally, I would like to thank my parents Arlene and Ted Shiner for their unwavering support. At last I must continue search for ways to adequately thank my wife Antonia for standing by me throughout this endeavour. This work involved a lot of travel and many promises that it will not always be like this. I am looking forward to our next great adventure together.

Dedication

I dedicate this thesis to my loving patient wife Antonia, who makes great lunches and helps me find my socks.

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Publications:

1. K. T. Kim, C. Zhang, **A. D. Shiner**, S. E. Kirkwood, E. Frumker, G. Gariepy, A. Naumov, D. M. Villeneuve, and P. B. Corkum, “Manipulating quantum paths for space-time characterization of attosecond pulses,” *Nature*, 2012. (Manuscript ID: 2012-08-09966, Submitted August, 2012)
2. **A. D. Shiner**, C. Trallero-Herrero, N. Kajumba, B. E. Schmidt, Kyung Taec Kim, H.-C. Bandulet, D. Comtois, F. Légaré, J.-C. Kieffer, D. M. Rayner, P. B. Corkum, and D. M. Villeneuve, “Characterization of ionization and intensity during high harmonic generation,” 2012. (In Preparation)
3. **A. D. Shiner**, B. E. Schmidt, C. Trallero-Herrero, P. B. Corkum, J.-C. Kieffer, F. Légaré, and D. M. Villeneuve, “Observation of cooper minimum in krypton using high harmonic spectroscopy,” *Journal of Physics B: Atomic, Molecular and Optical Physics*, vol. 45, no. 7, p. 074010, 2012
4. C. Trallero-Herrero, C. Jin, B. E. Schmidt, **A. D. Shiner**, J.-C. Kieffer, P. B. Corkum, D. M. Villeneuve, C. D. Lin, F. Légaré, and A. T. Le, “Generation of broad xuv continuous high harmonic spectra and isolated attosecond pulses with intense mid-infrared lasers,” *Journal of Physics B: Atomic, Molecular and Optical Physics*, vol. 45, no. 1, p. 011001, 2012
5. **A. D. Shiner**, B. E. Schmidt, C. Trallero-Herrero, H. J. Wörner, S. Patchkovskii, P. B. Corkum, J.-C. Kieffer, F. Légaré, and D. M. Villeneuve, “Probing collective multi-electron dynamics in xenon with high-harmonic spectroscopy,” *Nature Physics*, vol. 7, p. 464467, Mar. 2011
6. B. E. Schmidt, **A. D. Shiner**, P. Lassonde, J.-C. Kieffer, P. B. Corkum, D. M. Villeneuve, and F. Légaré, “CEP stable 1.6 cycle laser pulses at 1.8 μm ,” *Optics Express*, vol. 19, no. 7, pp. 6858–6864, 2011
7. B. E. Schmidt, P. Béjot, M. Giguère, **A. D. Shiner**, C. Trallero-Herrero, É. Bisson, J. Kasparian, J. Wolf, D. M. Villeneuve, J. Kieffer, P. B. Corkum, and

- F. Légaré, “Compression of 1.8 μm laser pulses to sub two optical cycles with bulk material,” *Applied Physics Letters*, vol. 96, p. 121109, Mar. 2010
8. C. Trallero-Herrero, B. Schmidt, **A. D. Shiner**, P. Lassonde, É. Bisson, J.-C. Kieffer, P. Corkum, D. Villeneuve, and F. Légaré, “High harmonic generation in ethylene with infrared pulses,” *Chemical Physics*, vol. 366, no. 1-3, pp. 33 – 36, 2009
 9. **A. D. Shiner**, C. Trallero-Herrero, N. Kajumba, H.-C. Bandulet, D. Comtois, F. Légaré, M. Giguère, J.-C. Kieffer, P. B. Corkum, and D. M. Villeneuve, “Wavelength scaling of high harmonic generation efficiency,” *Physical Review Letters*, vol. 103, no. 7, p. 073902, 2009
 10. M. Giguère, B. E. Schmidt, **A. D. Shiner**, M.-A. Houle, H. C. Bandulet, G. Tempea, D. M. Villeneuve, J.-C. Kieffer, and F. Légaré, “Pulse compression of submillijoule few-optical-cycle infrared laser pulses using chirped mirrors,” *Opt. Lett.*, vol. 34, no. 12, pp. 1894–1896, 2009
 11. H.-C. Bandulet, D. Comtois, **A. D. Shiner**, C. Trallero-Herrero, N. Kajumba, T. Ozaki, P. B. Corkum, D. M. Villeneuve, J.-C. Kieffer, and F. Légaré, “High harmonic generation with a spatially filtered optical parametric amplifier,” *Journal of Physics B: Atomic, Molecular and Optical Physics*, vol. 41, no. 24, p. 245602 (7pp), 2008
 12. B. E. Schmidt, **A. D. Shiner**, M. Giguère, P. Lassonde, C. Trallero-Herrero, J.-C. Kieffer, P. B. Corkum, D. M. Villeneuve, and F. Légaré, “High harmonic generation with long wavelength few-cycle laser pulses,” *Journal of Physics B: Atomic, Molecular and Optical Physics*, vol. 45, no. 7, p. 074008, 2012

Additional Works:

Publications that are not directly related to work presented in this thesis.

1. H. Akagi, T. Otobe, A. Staudte, **A. D. Shiner**, F. Turner, R. Dörner, D. M. Villeneuve, and P. B. Corkum, “Laser Tunnel Ionization from Multiple Orbitals in HCl,” *Science*, vol. 325, no. 5946, pp. 1364–1367, 2009
2. P. Dubé, A. A. Madej, J. E. Bernard, L. Marmet, and **A. D. Shiner**, “A narrow linewidth and frequency-stable probe laser source for the $^{88}\text{Sr}^+$ single ion optical frequency standard,” *Applied Physics B: Lasers and Optics*, pp. 40–+, Feb. 2009
3. **A. D. Shiner**, A. A. Madej, P. Dubé, and J. E. Bernard, “Absolute optical frequency measurement of saturated absorption lines in Rb near 422 nm,” *Applied Physics B: Lasers and Optics*, vol. 89, pp. 595–601, Dec. 2007

Non-Refereed Conference Proceedings

1. **A. D. Shiner**, B. E. Schmidt, C. Trallero-Herrero, H. J. Wörner, S. Patchkovskii, J.-C. Kieffer, P. B. Corkum, F. Légaré, and D. M. Villeneuve, “Probing collective multi-electron dynamics with high harmonic spectroscopy - the giant resonance in xenon,” in *The 3rd International Conference on Attosecond Physics*, (Hokkaido University, Sapporo, Japan), 2011. (Oral Presentation)
2. **A. D. Shiner**, B. E. Schmidt, C. Trallero-Herrero, H. J. Wörner, S. Patchkovskii, P. B. Corkum, J.-C. Kieffer, F. Légaré, and D. M. Villeneuve, “Probing collective multi-electron dynamics with high harmonic spectroscopy - the giant resonance in xenon,” in *Quantum Electronics and Laser Science Conference*, (Baltimore, Maryland, USA), p. QMC1, Optical Society of America, 2011. (Oral Presentation)
3. F. Legare, B. E. Schmidt, P. Béejot, M. Giguère, **A. D. Shiner**, C. Trallero-Herrero, É. Bisson, J. Kasparian, J.-P. Wolf, D. M. Villeneuve, J.-C. Kieffer, and P. B. Corkum, “Intense sub-2 optical cycle laser pulses at 1.8 micron for high harmonic generation,” *APS Meeting Abstracts*, p. L6007, Mar. 2010
4. C. Trallero-Herrero, B. Schmidt, **A. D. Shiner**, J.-C. Kieffer, P. Corkum, F. Legare, and D. Villeneuve, “Time Reversing High Harmonic Generation: Towards table top XUV spectroscopy,” *APS Meeting Abstracts*, p. 3006, Mar. 2010
5. **A. D. Shiner**, C. Trallero-Herrero, N. Kajumba, H.-C. Bandulet, D. Comtois, F. Légaré, M. Giguère, J.-C. Kieffer, P. B. Corkum, and D. M. Villeneuve, “Scaling of high harmonic generation efficiency with wavelength,” in *Attosecond Physics 2009*, (Manhattan Kansas), 2009. (Poster Presentation)
6. F. Légaré, B. E. Schmidt, **A. D. Shiner**, C. Trallero-Herrero, H. J. Wörner, M. Giguère, P. B. Corkum, J.-C. Kieffer, and D. M. Villeneuve, “High harmonic generation using 1425 nm sub-3 cycles laser pulses,” in *Attosecond Physics 2009*, (Manhattan Kansas), 2009. (Oral Presentation)
7. **A. D. Shiner**, C. Trallero-Herrero, N. Kajumba, H.-C. Bandulet, D. Comtois, F. Légaré, J.-C. Kieffer, P. B. Corkum, and D. M. Villeneuve, “Scaling of high harmonic generation efficiency with wavelength,” in *Ultra-Fast Dynamic Imaging of Molecules II*, (Ischia Island (Naples), Italy), 2009. (Poster Presentation)
8. **A. D. Shiner**, C. Trallero-Herrero, N. Kajumba, P. B. Corkum, D. Villeneuve, B. Heidi-C, C. Daniel, F. Légaré, and J.-C. Kieffer, “Scaling of high harmonic generation efficiency with wavelength,” in *2008 Gordon Research Conference on Multiphoton Processes*, 2008. (Poster Presentation)

9. **A. D. Shiner**, A. A. Madej, P. Dubé, and J. E. Bernard, “Absolute measurement of Rb transition frequencies at 422-nm using a femtosecond optical frequency comb,” in *Deutsche Physikalische Gesellschaft Summer School on Attosecond Physics, Bad Honnef, Germany.*, 2007. (Poster Presentation)
10. H.-C. Bandulet, D. Comtois, T. Ozaki, J.-C. Kieffer, and F. Légaré, “High harmonic generation with a spatially-filtered optical parametric amplifier,” in *2008 Gordon Research Conference on Multiphoton Processes*, 2008. (Poster Presentation)
11. A. Staudte, H. Akagi, D. Pavicic, **A. D. Shiner**, F. Turner, R. Doerner, D. M. Villeneuve, M. Y. Ivanov, and P. B. Corkum, “Alignment-free molecular frame tunneling probabilities,” in *39th Annual Meeting of the APS Division of Atomic, Molecular, and Optical Physics*, 2008
12. C. Trallero-Herrero, **A. D. Shiner**, N. Kajumba, C. Bandulet, Heidi, D. Comtois, F. Légaré, J.-C. Kieffer, P. Corkum, and D. Villeneuve, “Scaling of high harmonic generation efficiency with wavelength,” in *39th Annual Meeting of the APS Division of Atomic, Molecular, and Optical Physics*, 2008
13. H. Akagi, A. Staudte, **A. D. Shiner**, F. Turner, D. M. Villeneuve, M. Y. Ivanov, and P. B. Corkum, “Molecular frame angular distribution of tunnel ionization probability of HCL molecule determined by correlation measurement between electron and fragment ion,” in *Second Annual Meeting of the Japan Society of Molecular Science*, 2008

Patents Awarded

1. **A. D. Shiner**, C. D. Schaaff, R. C. Madter, R. S. Mathew, M. F. Habicher, and Q. A. Luong, “Over-voltage protection circuit.” US Patent Number 20030076642, Filed April 24, 2003, Accepted April 25, 2006

Statement of originality and collaborative contributions

The author states that to the best of his knowledge this body of work constitutes original research in the field of physics. The publications related to this thesis are listed in the author's CV starting on page 1 and will be referred to by number in round brackets. The primary achievements of this thesis are described in the publications on the wavelength scaling of high harmonic generation efficiency (9), the development of an ion detector (2), and the discovery of multi-electron correlation effect in the HHG spectra of xenon (5). All publications presented in this thesis resulted from the collaborative efforts of the author, post doctoral fellows from the National Research Council of Canada, as well as researchers from the Advanced Laser Light Source at INRS EMT. The author participated in all experimental campaigns that led to publications presented in this thesis. For the publications listed above, the author played a leadership role in all aspects of experimental design, execution and in particular in the analysis of the results. He also wrote major sections of the published manuscripts. The xenon result was enabled by the development of pulse compression technology in the infrared. Source development was led by Mathieu Giguère at $1.4\text{ }\mu\text{m}$ (10) and by Bruno Schmidt at $1.8\text{ }\mu\text{m}$ (7). The development of the OPA as a carrier envelope phase (CEP) stable source at $1.8\text{ }\mu\text{m}$ (6) was also led by Bruno Schmidt. For this project the author was involved in developing the f-2f interferometer for CEP measurement, and led the development of the CEP control actuator as well as the system for active stabilization of the CEP. The computer simulation results presented in (6,7) and (12) were provided by Pierre Béjot and co-workers [30]. The simulation results presented in the introduction of Ch.6 (Figs. 6.1, 6.5, 6.6 and 6.8) were calculated by the author using a code that he independently developed. A review paper was written by Bruno Schmit that summarized the pulse compression efforts in the infrared

(12). The author provided the high harmonic generation results and intensity measurements for this paper. The ionization results presented in publications throughout this thesis were produced by the author using a code that he developed. Chapter 5, which describes the role of spacial filtering for HHG in the infrared, contains the results of free-space HHG simulations from a code that was developed by the author. The functions for calculating the nonlinear source term in the propagation code were supplied by David Villeneuve. For the krypton Cooper minimum paper (3), the author participated in the experiment and analyzed the results. For the C₂H₄ paper (8) the author participated in the experiments but the analysis was led by Carlos Trallero.

In addition to the scientific contributions listed above the author is responsible for a number of technical improvements not listed in the thesis. These include:

1. Designing a new XUV spectrometer for the detection of 1 keV photons.
2. Redesigning the HHG target chamber to use reflective optics and a computer controlled 3-axis translation of the gas jet in vacuo.
3. Designing the control system and analysis tools for the HHG experiment.
4. Calibrating the spectral response of visible and infrared spectrometers for the pulse measurement apparatus using radiometric standards at the Canadian Institute for National Measurement Standards.
5. Rebuilding a 500 Hz, 0.5 mJ regenerative amplifier.

French translation of the thesis abstract was provided by Genevieve Gariépy.

Chapter 1

Introduction

Much of what we know about matter comes from observing its interaction with light. As our ability to generate and control light improves we discover previously unknown physical processes. In spectroscopy we can detect the presence of a gas by passing light through a sample and watching for absorption with a spectrometer. By looking more closely we find that the wavelengths that are absorbed are specific to the gas and they tell us not only which gas is present but also about the change in state that took place when the photons were absorbed. For example, changes in how fast a molecule is rotating are usually associated with the absorption of microwave photons. Vibrations between atoms that make up a molecule are coupled to transitions in the infrared. Visible and ultraviolet photons excite valance electrons, whereas extreme ultraviolet (XUV) and soft x-ray photons excite the inner shell electrons which are close to the core of each atom in a molecule. The study of these processes requires a high degree of control over the wavelength and the spectral purity of the light sources that are used to probe the sample.

While we can learn a lot by observing that a microwave photon was absorbed or by noting the presence of the reaction products of a photochemical reaction, what is really interesting is the ability to watch the rotation in time or to follow a photochemical reaction as it happens. In our macroscopic world, fast cameras and electronic circuits are sufficient to resolve most motion. Speeding bullets and shattering windows are all easily recorded with cameras. Recording processes with faster time scales, such as picoseconds for molecular rotation or femtoseconds for molecular vibration, requires us to use the properties of light itself to initiate (pump), and then probe the motion. To study dynamics it is not sufficient to simply take a snapshot of a molecule,

we need to be able to initiate a process and then to probe it at later times. Our timing resolution will be limited by the duration of our pulses, our ability to control the timing between them and the degree to which samples respond to the pump pulses in a reproducible way. The use of femtosecond laser pulses to study chemical reactions is termed femto-chemistry which saw explosive growth in the 1990's ultimately leading to the award of the 1999 Nobel Prize in Chemistry to Ahmed H. Zewail [31].

While femtosecond laser sources have made it possible to control and follow chemical reactions in time, the advent of attosecond pulses introduces the opportunity to follow the correlated motion of electrons on their natural femtosecond to attosecond time scale [32]. The development of table-top sources of XUV radiation with attosecond pulse duration will enable researchers to start addressing fundamental questions such as:

1. If we remove an electron from one end of a molecule, how long does it take for the positive charge to re-distribute?
2. Is it possible to measure x-ray photoionization cross-sections in a table-top experiment?
3. Can we time resolve core-electron excitation and exchange during electron re-scattering?

Most physics students learning quantum mechanics will begin by solving for the energy levels and transition probabilities of a hydrogen atom. The atom's single electron moving in the field of a proton represents one of the few analytically solvable problems in quantum mechanics. Most real systems have more than one electron and the interaction between those electrons is responsible for many important processes in nature. When a photon is absorbed promoting one of the electrons in an atom into the continuum, the other electrons respond to this and their motion can affect the ejected electron on its way into the continuum. Electron correlation effects can be seen in the transition strength and angular distributions of photo-electrons produced with XUV and soft x-ray photons. These correlations are responsible for configuration interaction in chemical reactions [33], cooperative phenomena in solids such as super conductivity [34], as well as for charge transport in biological systems [35].

So far, most of our knowledge of electron correlation effects resulted from the development of synchrotron light sources, which provide the XUV photons needed to

ionize inner shell electrons, where most of this electronic structure originates. The studies to date have primarily focused on the spectral (energy), domain where the signature of correlation effects is visible but where phase and timing information are usually not available [32]. Attosecond pulses produced from high harmonic generation offer the unique opportunity to study electron correlation effects in time.

This thesis describes the development of shorter femtosecond laser pulses in the infrared with precisely controlled electric fields, and the use of these pulses to produce XUV bursts with photon energies extending to hundreds of electron volts. In doing so we produce the bandwidth needed for shorter attosecond pulses and make the first observation of electron correlation and exchange processes during high harmonic generation.

This introduction will start with a discussion of how short pulses are produced. It will then discuss the interaction between atoms and intense laser fields that leads to HHG, and finishes with some proposed applications.

1.1 Generating femtosecond laser pulses

On the femtosecond timescale there is no electronic trigger or shutter that can directly gate an optical pulse. Short pulses have to be produced using properties of the light itself. If we look at the spectrum of a pulse we find that it is comprised of sinusoidal waves at many different frequencies as shown in Fig. 1.1. When the waves are all in phase at one instant in time they add constructively producing a very high peak field. At other times they destructively interfere, and the fields cancel. The duration of the pulse is limited by the range of frequency components (bandwidth), that can be brought into phase. The main tool for producing femtosecond pulses is the Titanium Sapphire laser (Ti:Saph), which has the ability to simultaneously operate at wavelengths from 700-1000 nm. Ti:Saph lasers can produce over one million frequency components, which can all be brought into phase resulting in pulse durations of tens of femtoseconds (10^{-15} s) [36]. In addition to producing very short pulses, femtosecond lasers are also capable of reaching extremely high peak intensities. For example, a typical Ti:Saph oscillator producing 30 fs pulses at a repetition rate of 80 Mhz has an average output power of 400 mW, or 5 nJ per pulse. Because this laser is only on for 0.00024% of the time all of this energy is concentrated in a very short interval and the instantaneous power is on the order of 166 kW! For the experiments

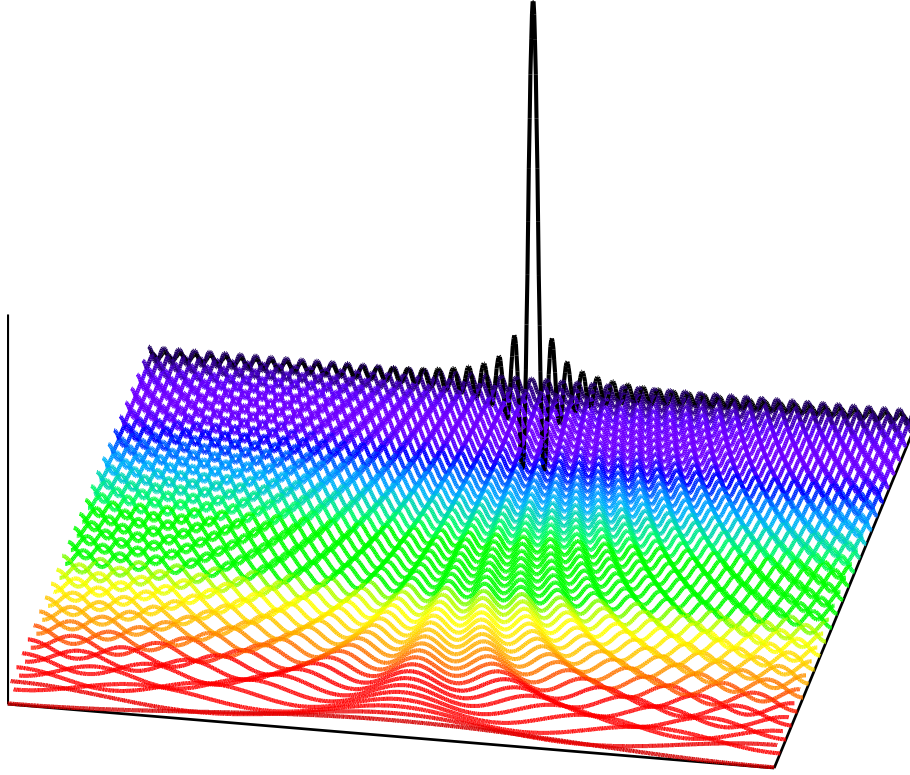


Figure 1.1: The short pulse which is plotted in black in the background is produced by coherently summing many frequency components (plotted in the foreground), which are all in phase at one instant.

that will be presented later in this thesis this pulse energy is amplified more than 2 million times, so that when focused onto atoms produces intensities that are high enough to overwhelm the coulomb force that binds the electrons.

1.2 How short can the pulse be?

As shown in Fig. 1.1 a pulse is constructed by coherently summing together the contributions from sine-waves at many different frequencies. By including more wavelengths we can produce shorter pulses. The shortest pulse that can be produced for a given spectral bandwidth is given by the Fourier relation $\Delta\tau \Delta\nu \geq C_B$, where $\Delta\nu$ is the spectral width, $\Delta\tau$ is the pulse duration and for Gaussian pulses $C_B = 0.441$ [37].

Note that this provides a lower limit for the pulse duration for a given bandwidth, achieving this limit requires all of the frequency components to be perfectly in phase. Combining spectral components with the correct spectral phase represents one of the main challenges when producing short pulses. Sunlight has an enormous bandwidth, but the random phase between the different colours means that a transform limited pulse can never be produced from this light.

By the end of the 20th century scientists were reaching the limits of the achievable pulse duration with visible light. Figure 1.2 shows the evolution of pulse duration with time. Pulses had been compressed down to a few femtoseconds, which was

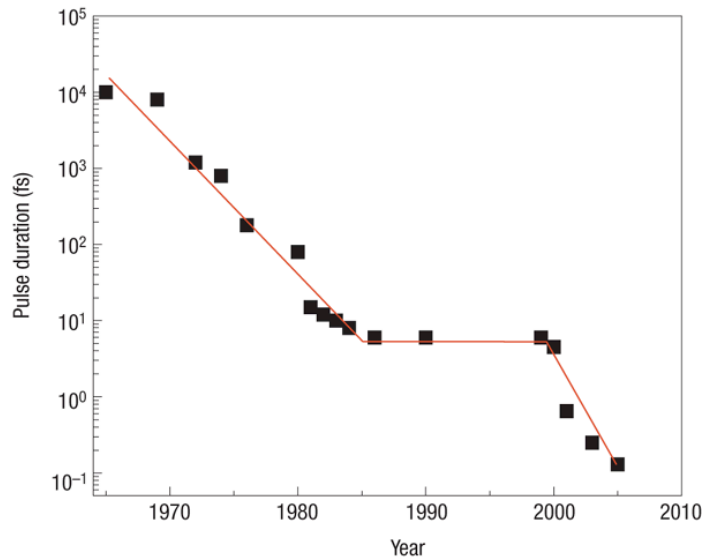


Figure 1.2: Timeline for the shortest optical pulses that could be produced in a given year. (Figure from: [38])

sufficient to see molecular dynamics, but not short enough to resolve most electronic motion. Producing the bandwidth needed for even shorter pulses presented a serious challenge because the minimum duration of a pulse is limited to around one cycle of the shortest wavelength used to produce it. For visible light the shortest wavelength is ~ 400 nm, which limits the pulse duration to ~ 1.3 fs. Generating shorter pulses necessitated developing sources in the extreme ultraviolet (XUV), which is a region of the spectrum where most materials are no longer transparent, and where normal lasers do not work. In 2001 the paradigm shift that enabled short pulse generation in the attosecond regime was the discovery of high harmonic generation (HHG) [39, 40].

1.3 High Harmonic Generation

In High Harmonic Generation an intense laser pulse is focused into a gas target, where a nonlinear interaction results in the generation of a coherent beam of XUV radiation at odd multiples of the fundamental laser photon energy. Figure 1.3 shows an artistic rendering of the HHG spectrometer at ALLS that which was used throughout this thesis. The laser enters the vacuum chamber from the foreground of the image and is

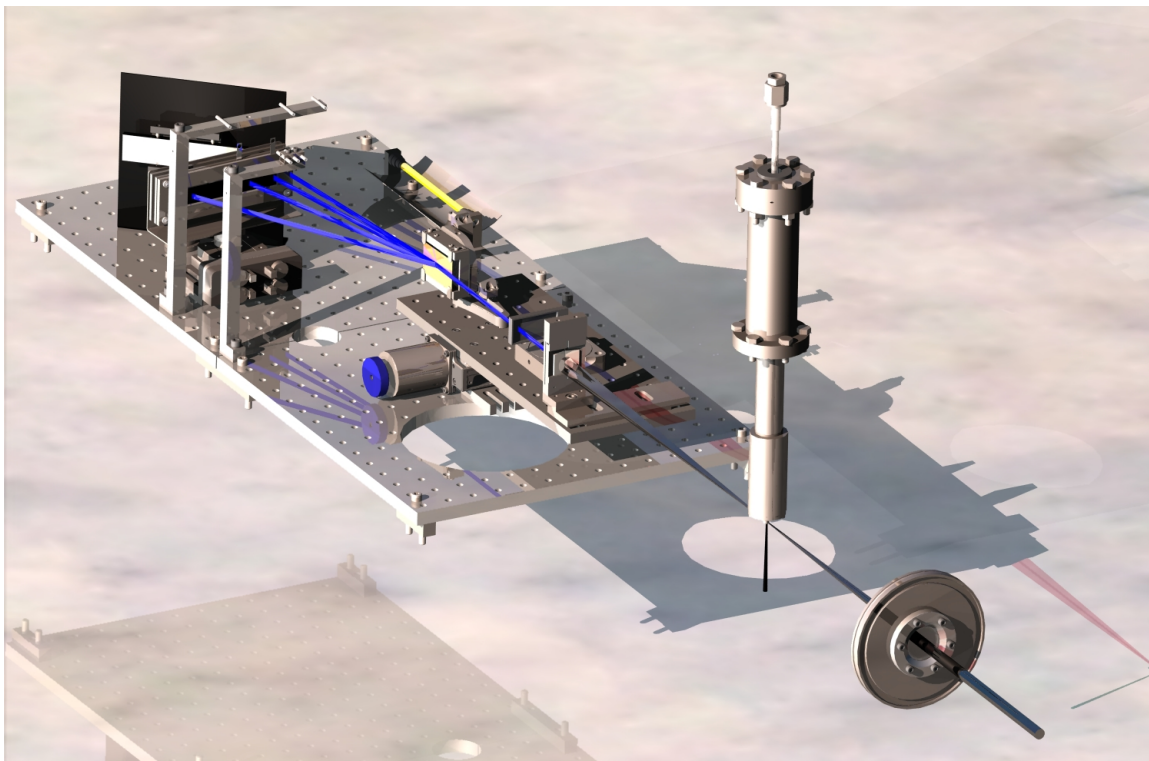


Figure 1.3: Rendering of HHG spectrometer at ALLS. The laser is shown entering the spectrometer from the foreground in the image and is focused into a gas jet that enters from the top. High harmonic radiation is generated at the laser focus, and co-propagate with the laser into the spectrometer. The beam passes through a slit and is spectrally dispersed by a grating onto an MCP image intensifier. The MCP enhances the spectrally resolved XUV spectrum and converts it into visible light, which is then photographed on the back side of the MCP with a digital camera.

focused under a gas jet, shown entering from the top. The resulting XUV radiation co-propagates with the laser field, passes through a slit, and is spectrally dispersed by a diffraction grating. The high harmonic radiation then fans out across the surface of

a microchannel plate detector, allowing it to be detected as visible light by a camera (not shown). An image of the HHG spectrum from xenon is shown in Fig. 1.4. A photograph of the HHG spectrometer at ALLS that was taken during the final

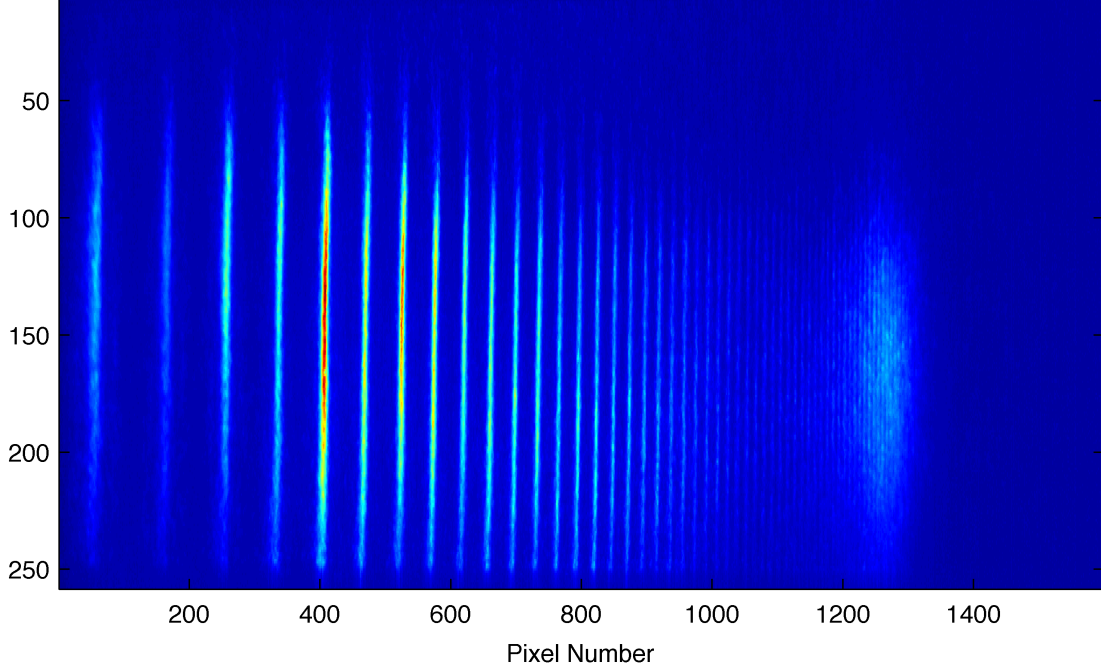


Figure 1.4: False colour image of the high harmonics spectrum detected by the micro channel plate image intensifier.

experimental campaign of this PhD is shown in Fig. 1.5. The laser enters from the right side of the image and the MCP detector is imaged by the camera in the lower left corner. The keyboard in the lower right corner of the photo give a sense of scale. The inset shows the target chamber in operation, where the jet enters from the side and the ionizing neon atoms emit an orange plume.

1.4 Applications

The first half of this thesis discusses the development of techniques to produce HHG photons that extend further into the x-ray regime by increasing the laser wavelength and by decreasing the pulse duration. By using a short-pulse, long-wavelength laser source we have dramatically increased the harmonic cutoff in xenon compared to what was previously possible with harmonic radiation produced using conventional

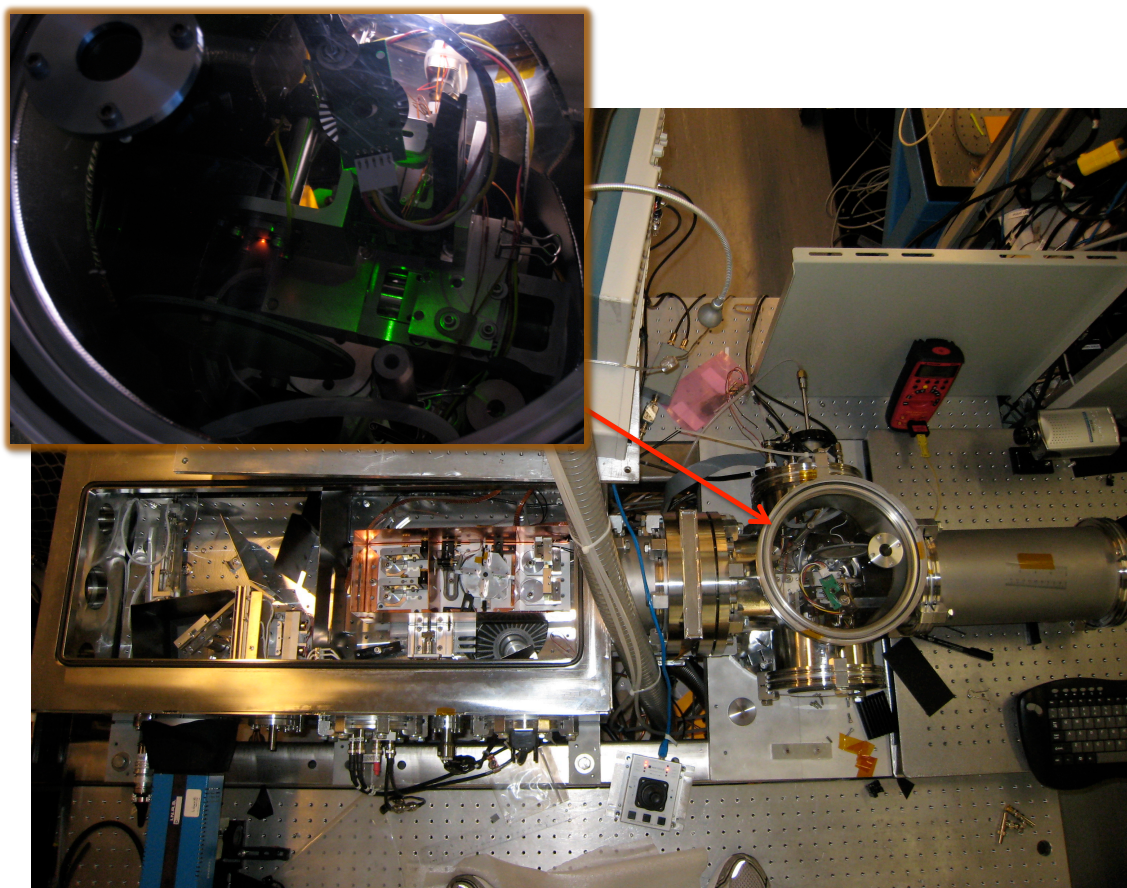


Figure 1.5: Photograph of HHG spectrometer at ALLS. The laser enters from the right and is focused into the gas target in the round ‘Source’ chamber. The resulting beam of XUV radiation passes into the diagnostic chamber on the left side of the image where it is spectrally dispersed by a spectrometer and imaged onto a micro channel plate detector. The detector amplifies the image of the XUV radiation and converts it to visible light, which is photographed by a digital camera. The inset shows neon atoms ionizing at the laser focus. The expanding plume of excited neon ions radiate orange light as they recombine with free electrons in the plasma.

800 nm sources. We have gone on to produce harmonics in neon that extend into the water window between 284 eV and 543 eV (Fig. 6.10). The technological implications of this are two fold. First, HHG provides a coherent source of soft x-rays which are useful for applications such as conventional microscopy and spectroscopy. Second, the enormous bandwidth from HHG can support attosecond pulses which can be used to probe electronic motion in the time domain. Today, most experiments that require

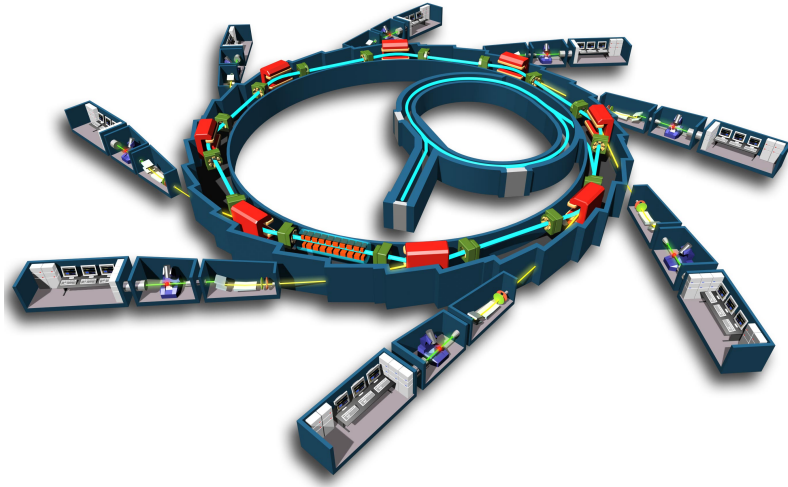


Figure 1.6: Illustration of the SOLEIL French national synchrotron facility near Paris France. (Figure from: [41])

a tuneable source of x-rays are conducted at synchrotrons facilities such as SOLEIL, shown in Fig 1.6. Synchrotrons provide a bright source of soft x-rays but require tens to hundreds of millions of dollars of investment due to their complexity and size (SOLEIL accelerator ring: 354 m circumference). The high cost limits availability and scientific progress.

1.4.1 Microscopy

Soft x-ray sources, even without attosecond timing resolution, have a number of important scientific and industrial applications. One of the most immediate is for microscopy of biological samples in the water window [42]. Imaging biological samples such as proteins and DNA requires spatial resolution of a few tens of nanometers, which implies a light source with a wavelength that is comparable to or shorter than this dimension. However, resolution is determined not only by the size of the features that can be identified, but also by the contrast between the specimen and its surroundings. The advantage to microscopy in the water window is that carbon rich tissue such as proteins and lipids absorb strongly whereas the water that the samples are suspended (or living), in is transparent. A number of sources are available for x-ray microscopy including incoherent plasma sources, synchrotrons and Free Electron Lasers (FEL) [43, 42]. Recently, HHG has been demonstrated as a table-top source for

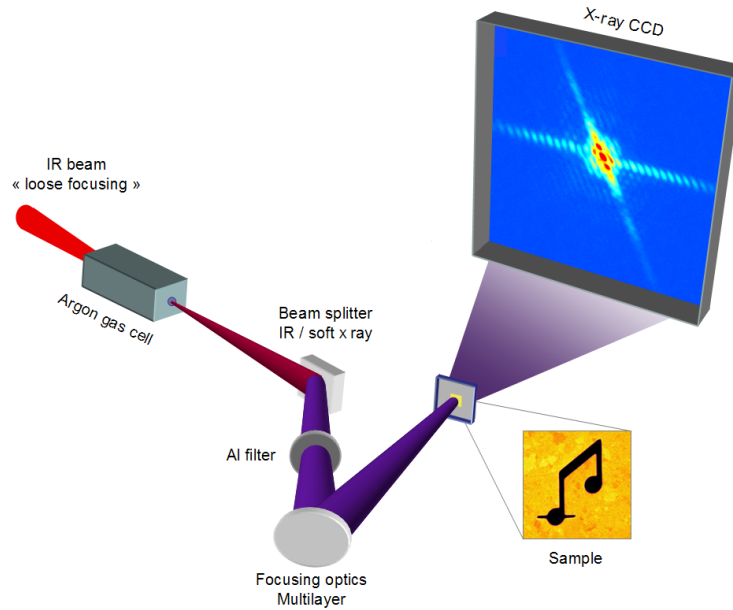


Figure 1.7: In lensless imaging a coherent beam of light passes through a sample and the resulting diffraction pattern is measured in the far field. A phase retrieval algorithm solves for the near field pattern that corresponds to the measured far field image. (Figure from: [45])

microscopy with several advantages [44, 45, 46]. First, HHG sources are much more compact and less expensive than synchrotrons and FELs. Pulses from HHG sources provide attosecond timing resolution, and the synchronized femtosecond laser pulses used for HHG can also be used to initiate dynamics in the sample. Third, the spatial resolution of a conventional x-ray microscope is usually limited by the quality of the x-ray focusing optics. The high spatial coherence of HHG sources enables ‘*lensless*’ imaging techniques. In lensless imaging, the sample is illuminated with a coherent source and the resulting diffraction pattern is detected in the far field as an intensity distribution (Fig. 1.7). A computer algorithm is used to iteratively solve for the near field pattern that corresponds to the measured far field image. This technique avoids the absorption and loss of x-ray optics and offers spacial and temporal resolution limited by the attosecond pulse. To date, single shot imaging has been demonstrated with 119 nm spacial resolution [45] and 25 nm resolution has been achieved with a 30 second exposure time [44].

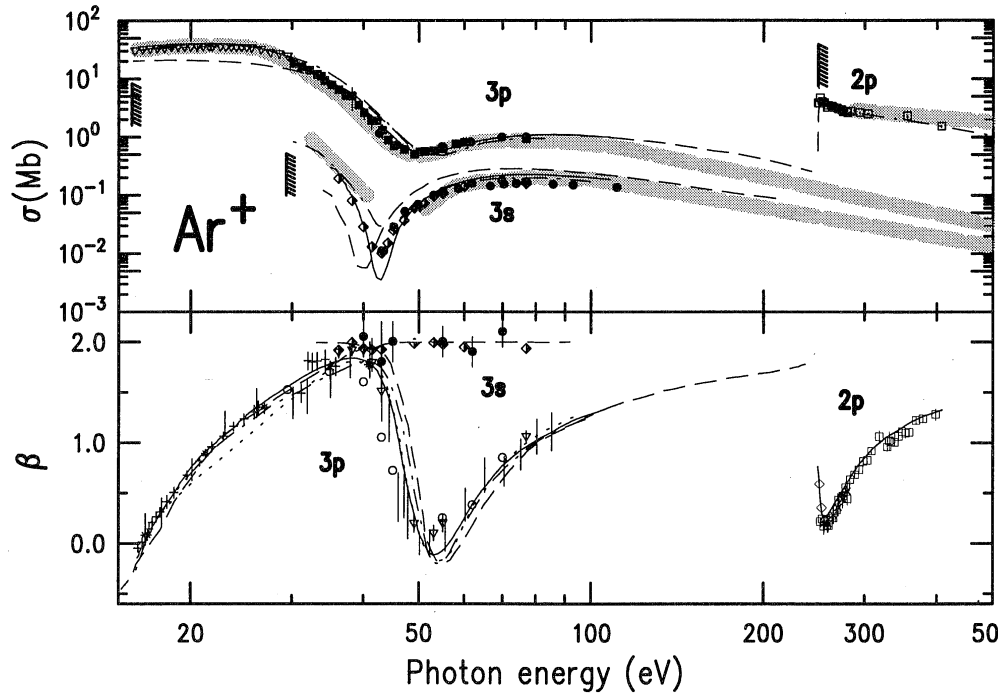


Figure 1.8: Argon photoionization cross-section σ and asymmetry parameter β (for definitions see Ch. 8). The symbols represent measurements from synchrotron sources and the solid curves are theoretical calculations. (Figure from: [47])

1.4.2 Spectroscopy

Measuring the cross-section for photoionization from different energy levels of an atom, plus the angle of the ejected electron with respect to the incident photon, is one of the primary methods used by researchers to study the electronic structure of matter [47]. An example of the state- and angle-resolved photoionization cross-section for argon is shown in Fig. 1.8. Measurements such as this are normally only possible at synchrotron facilities. The second part of this thesis shows that by analyzing the spectra produced through HHG it is possible to extract the photoionization cross-section from the generating medium.

1.4.3 Probing Electron Correlation

The interaction between the electrons in an atom or molecule is responsible for charge transfer in biological systems [32]. An example of charge transfer is the landmark ex-

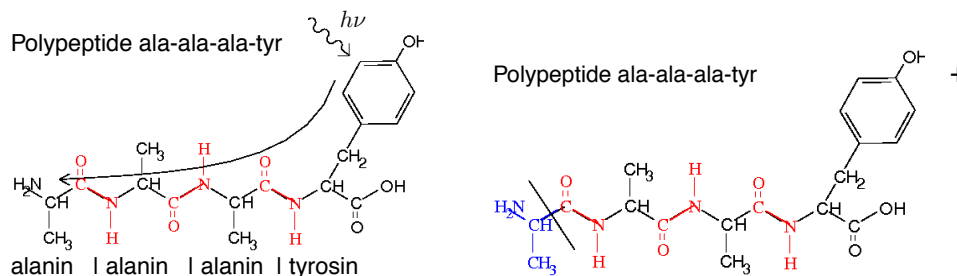


Figure 1.9: Example of charge transfer along the peptide chain Ala-Ala-Ala-Tyr. Ultraviolet photons preferentially ionize the achromatic chromophore end of the molecule. The resulting hole migrates down the backbone of the peptide chain and initiates fragmentation at the opposite end [49]. (Figure from: [50])

periment by R. Weinkauff and co-workers who showed that site specific ionization from one part of a molecule can trigger bond breaking and fragmentation at a different site [48, 49]. A peptide chain was synthesized with a chromophore at one end (Fig. 1.9 A), that is preferentially ionized by absorbing UV photons. Immediately following ionization a ‘hole’ is left at the ionization site, which then travels down the peptide chain and initiates bond breaking at the opposite end (Fig 1.9 B). Theoretical modelling in smaller molecules has shown the time for charge migration along the length of a molecule is on the order of a few femtoseconds [51, 32]. The development of shorter femtosecond and attosecond pulses will enable a new class of experiments where charge transfer and electron correlation effects are probed on their natural time scales.

The use of HHG to probe electron correlation effects is demonstrated in chapter 10. During the recombination step of HHG the continuum electron excites a core 4d electron. This excitation is responsible for a large enhancement of the photo-recombination cross-section to the valance 5p state, which is observed as a ten-fold increase in HHG yield at 100 eV.

Chapter 2

Theoretical Background

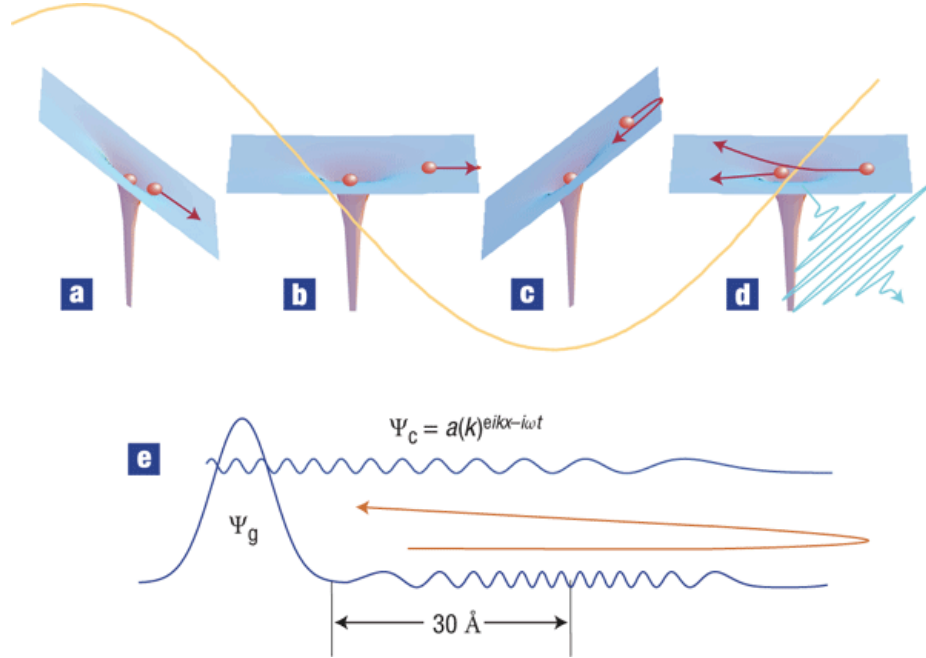


Figure 2.1: In HHG an atom tunnel ionizes near the peak of a laser field, freeing an electron, which is accelerated away from the parent ion (a-b). When the field reverses direction the electron is driven back (c) and may re-encounter the parent ion leading to the emission of an XUV photon (d). Harmonic emission arises from the interference between the continuum electron and the ground state wavefunction of the atom (e). (Figure from [38])

The high harmonic generation process for producing attosecond pulses begins with a femtosecond laser, which when focused reaches peak intensities on the order

of $10^{14} - 10^{15} \text{ W/cm}^2$. This intensity is sufficient to overwhelm the coulomb potential that binds the valence electrons in an atom. Near the peak of each half cycle of the laser field the coulomb barrier is suppressed and there is an opportunity for an electron to **tunnel ionize**, passing through the barrier, into the continuum (Fig. 2.1 (a)). At the moment of ionization an electron appears in the continuum where it is immediately accelerated away by the strong laser field (Fig. 2.1 (b)). The continuum electron can gain hundreds of electron volts of kinetic energy accelerating in the field which, upon changing sign, drives the electron back toward the ion. In most cases the continuum electron will continue to oscillate in the field until the end of the pulse and never re-encounters the ion. There are however, some trajectories where the electron returns to the parent ion and recombine through a radiative transition, releasing the energy gained accelerating in the continuum in the form of an XUV photon (Fig. 2.1 (d)). Figure 2.2 shows a calculation of the bursts of XUV emission during a laser pulse and Figure 1.4 shows an example of a HHG spectrum from xenon. Because harmonic generation is periodic in time, repeating every half cycle of the laser field, we observe periodicity in frequency with the XUV spectrum consisting of odd multiples (harmonics), of the laser photon energy. The three steps for high harmonic

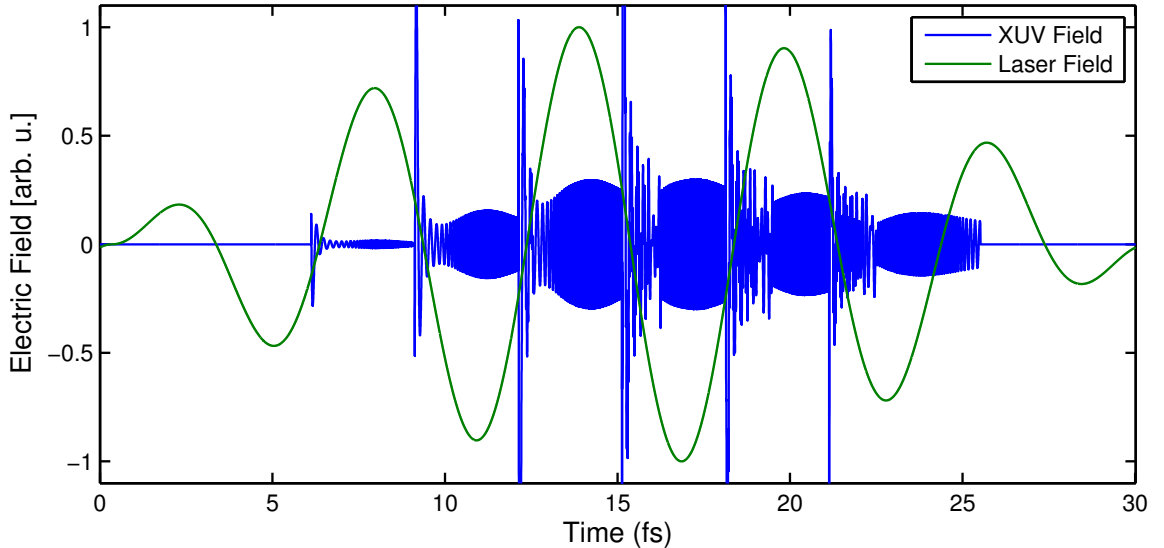


Figure 2.2: Calculated HHG emission for a hydrogen atom with a flat photo recombination cross-section in a 5-cycle (full width to zero), $1.8 \mu\text{m}$, $2.0 \cdot 10^{14} \text{ W/cm}^2$ laser field. Both long and short trajectory components are included.

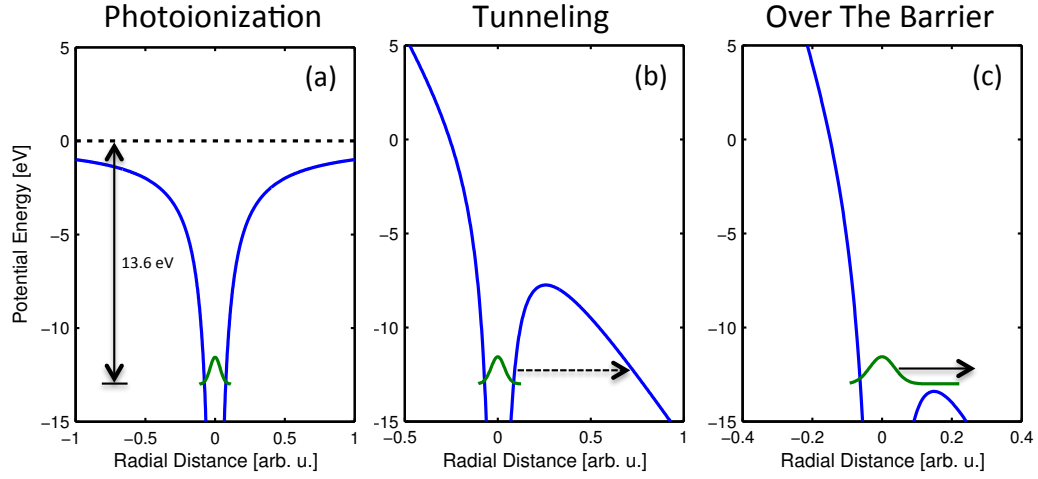


Figure 2.3: Ionization pathways. The bound electron wavefunction is shown in green and the potential well is plotted in blue. Ionization pathways: (a) In the perturbative limit one photon is absorbed promoting the electron (green), into the continuum. (b) In the tunneling regime a strong laser field tips the coulomb potential allowing the electron to tunnel into the continuum. (c) A very intense laser field can overwhelm the coulomb potential and the electron is no longer bound to the nucleus. In this over the barrier regime the electron is free to leave.

generation of strong field ionization, acceleration of the continuum electron in the laser field, and photo-recombination are each described in the following sections of this chapter.

2.1 Ionization

In a hydrogen atom the electron sits in a r^{-1} coulomb potential well as shown in Fig. 2.3 (a)¹. The ionization potential (I_p), is the amount of energy that the electron would need to gain in order to escape. In the traditional perturbative view of photoionization, we consider the interaction between an atom and a photon with energy $>I_p$ and look at the dipole transition rate between the ground state of the atom and the continuum. For the case where one photon has sufficient energy to promote the bound state electron into the continuum, the transition rate will be proportional to the laser intensity. If the photon energy is less than the ionization potential it is still possible to ionize the atom through the absorption of two or more photons at the

¹Atomic units are used throughout ($m_e = e = \hbar = 1$)

same time. If q photons are absorbed the ionization rate scales with laser intensity I as I^q [52]. The I^q scaling assumes intensities in the perturbative regime, where only a few photons are absorbed, which implies that the atomic coulomb potential well is not significantly distorted by the laser field. It is also possible to ionize an atom by placing it in an extremely strong DC electric field [53]. In this case the field competes with the coulomb potential, causing the well to tip over as shown in Fig. 2.3 (b). The bound electron energy is still less than the height of the barrier, but there is some probability for the electron *tunneling* under the barrier into the continuum. The tunneling rate depends critically on the height and width of the barrier, which is determined by the instantaneous electric field strength and direction. For ionization with intense laser pulses, there are conditions where ionization is well described by the tunneling picture. This is the case when the laser field is intense enough to significantly distort the coulomb potential and the modified potential well does not change during the time it takes the electron to tunnel under the barrier into the continuum. The question of whether it is meaningful to talk about an actual tunneling time has been the subject of lively debate in the attosecond community [54, 55, 56]. It is described here because it provides an intuitive picture that is helpful for understanding the behaviour of ionization in low and high field regimes.

To calculate the tunneling time we consider an electron of mass ($m_e = 1$) that is sitting in a zero range potential well whose depth is equal to the ionization potential I_p of the atom. A laser field interacting with this system tips the potential well creating a triangular barrier region as shown in Fig. 2.4. The electron enters this barrier region with initial velocity v_0 given by $\frac{1}{2}m_e v_0^2 = I_p$, and is assumed to exit with zero velocity giving an average of $\bar{v} = \sqrt{I_p/2}$. The laser electric field F causes the electric potential to drop by I_p over a distance of x_0 so $x_0 = I_p/F$. The tunneling time τ is then given by:

$$\tau = \frac{x_0}{\bar{v}} = \frac{\sqrt{2I_p}}{F}. \quad (2.1.1)$$

In the quasi-static regime the laser field changes slowly compared to the tunneling time. To describe the transition between the tunneling and multi-photon regimes we introduce the dimensionless Keldysh parameter γ :

$$\gamma = \omega_L \tau = \frac{\omega_L}{F} \sqrt{2I_p}. \quad (2.1.2)$$

When $\gamma \ll 1$ the field evolves slowly compared with the tunneling time, and ionization

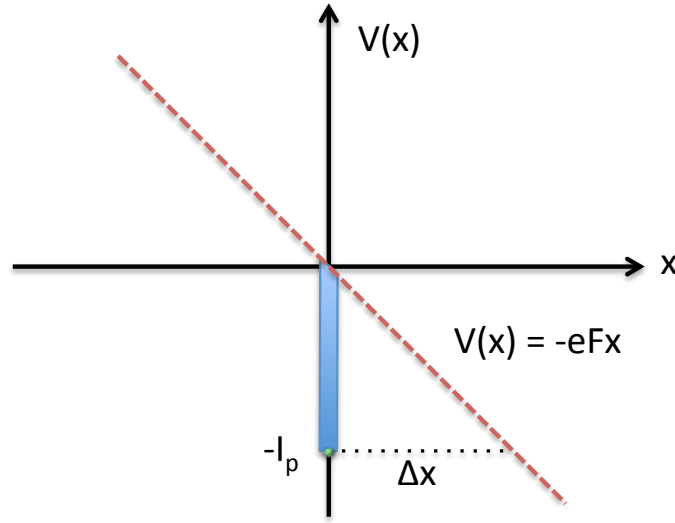


Figure 2.4: A simplified model for tunnel ionization starts with an electron sitting in a zero range potential with energy $(-I_p)$ that is distorted by the laser electric field F . The field tips the potential well creating a triangular barrier region of width Δx . Ionization is well-described by the strong field approximation if the time required for an electron to travel through this barrier region is small relative to period the laser field.

is well described by tunneling in the instantaneous electric field. When $\gamma \gg 1$ there will be virtually no tunneling and ionization requires many cycles of the laser field. In this *multi-photon* regime the ionization rate is mostly constant during a given half-cycle. In extreme cases the laser field can completely overwhelm the coulomb potential and the electron is free to leave as shown in Fig. 2.3 (c). This is called *over the barrier ionization*.

2.2 Non-Adiabatic Ionization Model

Throughout this thesis the Yudin-Ivanov (YI) model [57] is used to calculate ionization rates. The YI model is designed to give accurate ionization rates in the tunneling regime ($\gamma \ll 1$) as well as in the region where $\gamma \approx 1$. This is important because many HHG experiments are done with 800 nm lasers under conditions where γ is around one and pure tunneling models, such as the commonly used Ammosov, Delone and Krainov (ADK) [58, 59] model, underestimate the ionization rate at low intensities.

The YI model returns the instantaneous ionization rate $\Gamma(t)$ given by:

$$\Gamma(t) = n(t) \exp \left(-\frac{\mathcal{E}^2 A^2(t)}{\omega_L^3} \Phi(\gamma(t), \theta(t)) \right) \quad (2.2.1)$$

where $n(t)$ is the ground state population, $\mathcal{E}$ is the peak field strength, $A(t)$ is the field envelope and $\theta(t)$ is the instantaneous phase of the electric field. The overall phase is chosen such that $\theta(t) = 0$ corresponds to the peak of a given oscillation of the optical field. The YI model requires the following conditions to be satisfied:

$$\omega_L \ll I_p \quad (2.2.2)$$

$$\mathcal{E} \ll (2I_p)^{3/2} \quad (2.2.3)$$

Equation (2.2.2) requires there to be enough time for at least some tunneling. The YI model is not applicable to single photon ionization with high energy photons. The second condition requires that the field does not completely overwhelm the coulomb potential. If Eq. 2.2.3 is not satisfied, the atom is in the over the barrier ionization regime and the probability of tunneling is unity.

The calculated sub-cycle ionization rate for xenon in laser wavelengths ranging from 400 nm to 1.8 μm is shown in Fig. 2.5. In all cases the peak laser intensity was chosen so that 43% of the atoms are ionized by the end of the pulse. When ions are produced with a short 11 fs, 1.8 μm pulse ($\gamma = 0.36$), the ionization is concentrated around the peaks of the most intense half cycles of the laser field, and goes to zero at the zero crossings of the field. The situation is different for a 400 nm 20 fs pulse, where the value of the Keldysh parameter ($\gamma = 2.47$) would suggest that multi-photon ionization should play a role. What we find is that at 400 nm ionization is still concentrated around the peaks of the most intense half cycles. In addition to this, there is significant ionization at the zero crossings of the laser field. Ionization at the zero crossing of the field cannot be explained with tunneling and requires us to consider the interaction between the atom and the laser at times prior to the moment of ionization. Moving to shorter wavelengths or lower intensities, the ionization rate continues to lose its sub-cycle dependence on the laser field. In the limit of one photon absorption the ionization rate depends only on the cycle averaged laser intensity, and ionization is equally probable at all times along a given half cycle. For xenon atoms in an 800 nm, 20 fs pulse the Keldysh parameter is ($\gamma \approx 1$), which means that for

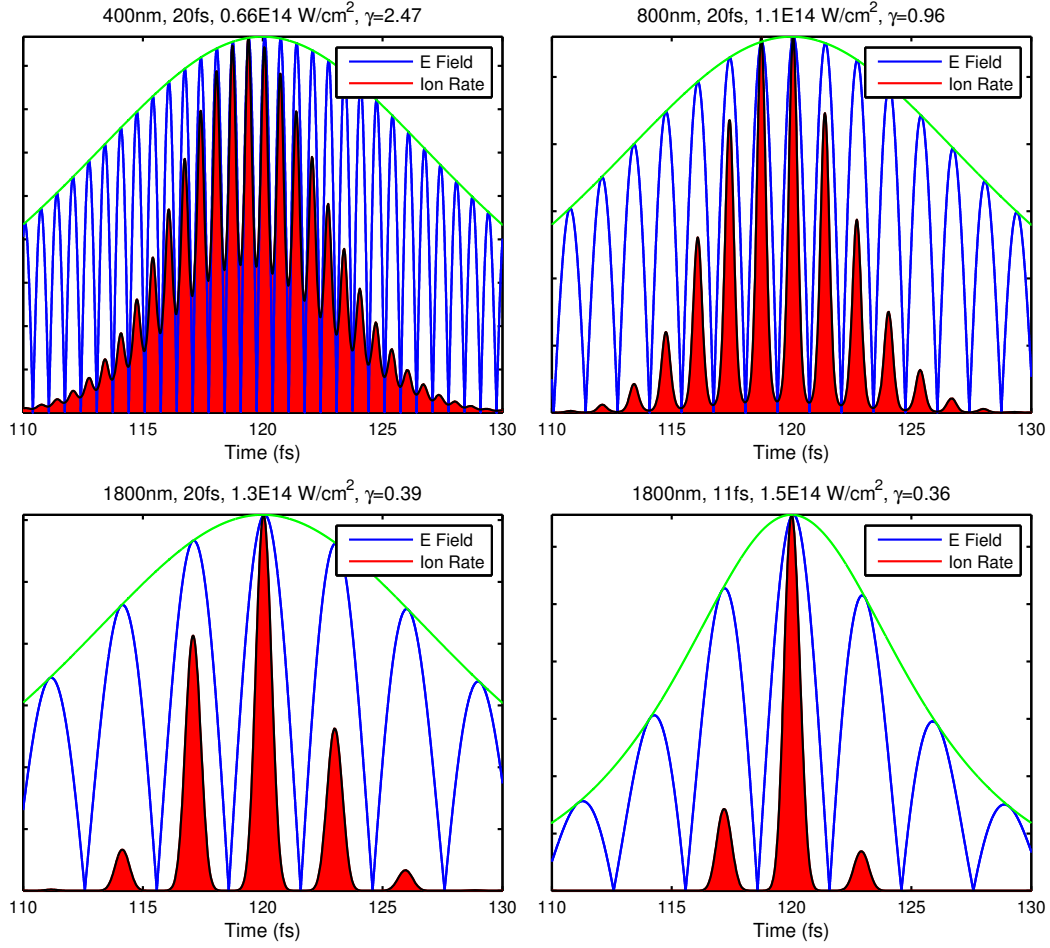


Figure 2.5: Sub-cycle ionization rate for xenon under various laser conditions, calculated with the Yudin Ivanov ionization model [57]. The magnitude of the electric field is plotted in blue and the instantaneous ionization rate is shown in red. Both are plotted in arbitrary units. In all cases the intensity is chosen so that 43% of the atoms are ionized by the end of the laser pulse. At 400 nm the Keldysh parameter is $\gamma = 2.47$ and there is significant ionization at the zero crossings of the electric field, indicating a large multi-photon contribution to ionization. At $1.8 \mu\text{m}$, $\gamma = 0.36$ and the ionization rate goes to zero at the zero crossings of the laser electric field. Ionization in xenon with a $1.8 \mu\text{m}$ short pulse is well described by quasi-static tunneling.

laser conditions which are commonly used in HHG experiments the assumption of pure quasi-static tunneling is not fully justified.

2.3 High Harmonic Generation

To understand high harmonic generation, it is helpful to consider the motion of a classical electron that suddenly appears in a laser field given by $F(t) = \mathcal{E} \cos(\omega t)$ at some birth time t_b with zero initial velocity. The wavelength of a visible or infrared laser is long compared the atomic length scale ($\sim \text{\AA}$), so spatial variation of the field can be ignored. The potential energy V of the free electron is modelled with a zero range potential $V = -e \mathbf{F} \cdot \mathbf{x}$, where e is the electron charge, $\mathbf{F}$ is the electric field and $\mathbf{x}$ is the position of the electron. As the electron accelerates in the field, we can solve for its trajectory with the classical equations of motion ($m_e = e = 1$):

$$v(t, t_b) = \frac{\mathcal{E}}{\omega} (\sin(\omega t) - \sin(\omega t_b)) \quad (2.3.1)$$

$$x(t, t_b) = \frac{\mathcal{E}}{\omega^2} (\cos(\omega t_b) - \cos(\omega t)) - \frac{\mathcal{E}}{\omega} \sin(\omega t_b)(t - t_b) \quad (2.3.2)$$

Using the initial conditions $x(t_b) = v(t_b) = 0$ we find is that there is a range of birth times where the electron is initially driven away from the ion but is then driven back when the laser field changes sign. Recollision occurs if the electron returns to the origin ($x(t, t_b) = 0$). A plot of the allowed combinations of birth and return times within one optical cycle of the laser is shown in Fig. 2.6. None of the electrons born when the laser phase is between -90 and zero degrees return to the parent ion. From zero degrees on the electron *can* return, and for each return time t there are two possible birth times. These are referred to as the long and the short trajectories.

The upper panel in Fig. 2.6 shows the recollision energy $v^2(t)/2$ for a given trajectory. When the continuum electron re-collides it can be re-absorbed through a radiative transition releasing the energy gained while accelerating in the continuum (plus the ionization potential of the atom), in the form of an XUV photon. This emission is caused by the induced dipole between the re-scattering electron and the ground state of the target atom or molecule. Experiments typically measure the HHG power spectrum $P(\omega)$, which is given by Fourier components of the acceleration of the *induced dipole moment* $\mathbf{D}(t)$:

$$P(\omega) \propto \left| \int \frac{d^2 D(t)}{dt^2} e^{i\omega t} dt \right|^2 \approx \omega^4 |D(\omega)|^2 \quad (2.3.3)$$

Calculating the induced dipole through direct solution of the time dependent Schrödinger

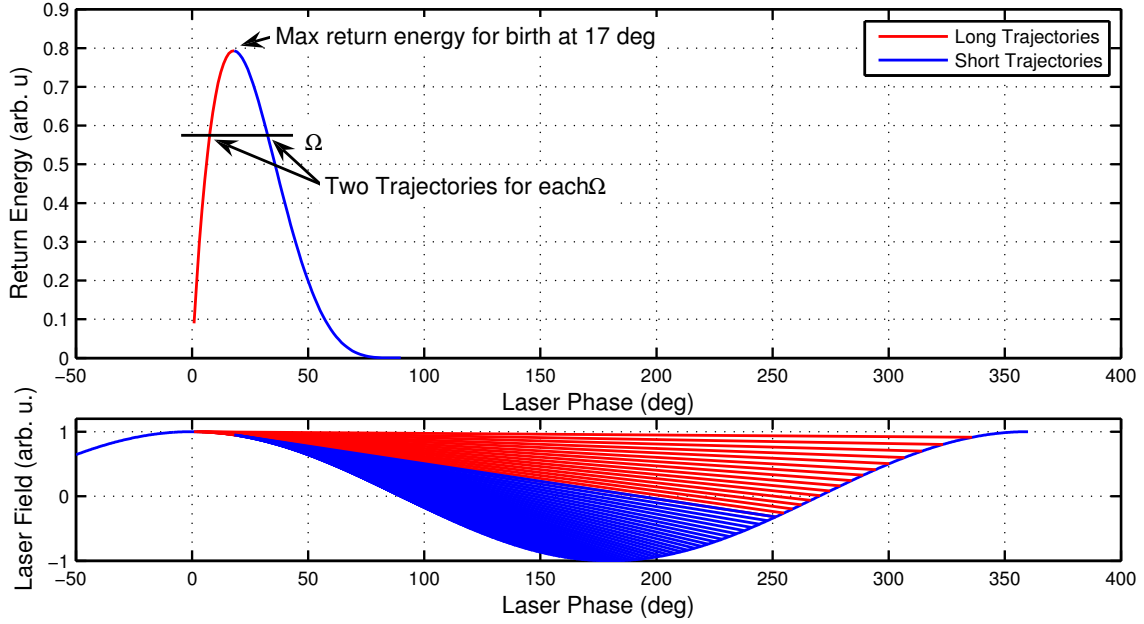


Figure 2.6: Classical trajectories for an electron born in a linearly polarized laser field. The lower panel shows one cycle of the laser field with lines connecting combinations of birth and return times that satisfy the conditions for high harmonic generation (see text). The blue lines correspond to the ‘short’ trajectories and the red lines are the ‘long’ trajectories. For each of these trajectories the electron energy at the time of re-collision is plotted in the upper panel.

equation is very demanding numerically. Fortunately, most of the key features of the HHG spectrum can be modelled using the strong field approximation as described in the following section.

2.4 Strong Field Approximation

In quantum mechanics if the wavefunction $|\psi(t = 0)\rangle$ is known at some initial time $t = 0$, you can calculate the wavefunction at later times using the propagator [60]:

$$|\psi(t)\rangle = \exp\left(-i \int_0^t \hat{H}(\tau) d\tau\right) |\psi(t = 0)\rangle \quad (2.4.1)$$

Normally we cannot simply replace the Hamiltonian $\hat{H}$ with the energy E because the system is not in an eigen state of $\hat{H}$. Solving for the time evolution requires direct evaluation of the integral in Eq. 2.4.1, which often cannot be solved analytically and difficult to solve numerically. The Strong Field Approximation (SFA) [61] was developed to model strong field processes such as HHG [62] and above threshold ionization ATI [63]. The basic idea of the SFA for calculating HHG spectra is that the HHG process can be broken into the three steps corresponding to tunnel ionization, propagation of the free electron in the continuum, and photo-recombination. To a surprisingly good approximation the three steps can be treated separately, and most importantly the time evolution of the continuum electronic wavefunction can be solved while completely neglecting the presence of the ion. These steps are depicted graphically in Fig. 2.7 and are described as follows:

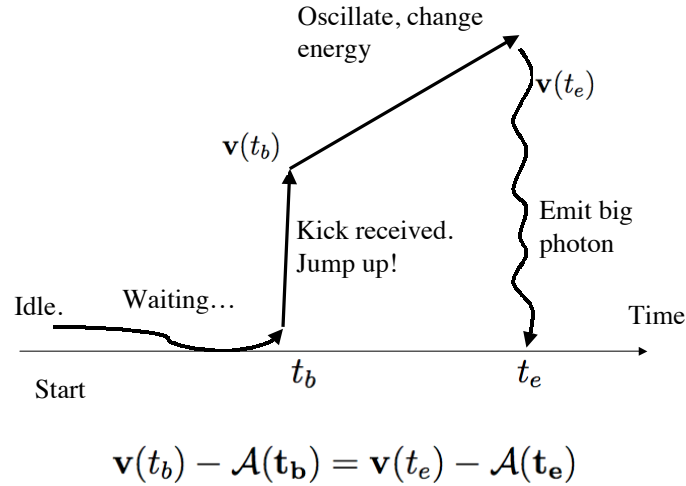


Figure 2.7: Three step model for HHG. The atom waits for some amount of time for its electron to be ionized by the laser field. The electron accelerates in the continuum before recombining in the third step of the process. (Figure from: [64])

STEP 1: First the atom waits in its ground state $|\psi_N\rangle$ until at some time t_b the laser ionizes the atom projecting an electron into the continuum state $|\chi_1(t)\rangle$ and leaving behind the ion in state $|\psi_1^+\rangle$.

$$|\chi_1(t_b), \psi_1^+\rangle \langle \chi_1(t_b), \psi_1^+ | \hat{V}_L(t_b) | \psi_N \rangle e^{-iE_N t_b} \quad (2.4.2)$$

Here, $\hat{V}_L(t) = -e \mathbf{F}(t) \cdot \mathbf{r}$ is the interaction hamiltonian due to the laser field and the term $e^{-iE_N t_b}$ accounts for the phase evolution of the ground state, with energy E_N from $t = 0$ to the ionization time t_b .

STEP 2: Next, the evolution of the continuum electron, which is initially in state $|\chi_1(t_b)\rangle$, is treated as that of a free particle in a laser field. This evolution can be calculated analytically using the Volkov propagator, taking advantage of the fact that the *Canonical Momentum* $\mathbf{k}$ is a conserved quantity for a free electron in a laser field with vector potential $\mathcal{A}(t) = \int_{-\infty}^t \mathbf{E}(t') dt'$. The canonical momentum is defined as $\mathbf{k} = \mathbf{v}(t) - e\mathcal{A}(t)$, where in atomic units the electron charge is ($e=1$). Because the canonical momentum is conserved, we can use the value of the vector potential to calculate the electron velocity at any time t once the initial values of the velocity and vector potential are known. Mathematically this is expressed as:

$$\mathbf{k} = \mathbf{v}(t) - \mathcal{A}(\mathbf{t}) = \mathbf{v}(t') - \mathcal{A}(\mathbf{t}'), \quad (2.4.3)$$

and

$$\mathbf{v}(t) = \mathbf{v}(t') - \mathcal{A}(\mathbf{t}') + \mathcal{A}(\mathbf{t}), \quad (2.4.4)$$

The energy of the electron at any other time is $E(t) = \frac{1}{2}m_e \mathbf{v}(t)^2 = \frac{1}{2}[\mathbf{v}(t') - \mathcal{A}(\mathbf{t}') + \mathcal{A}(\mathbf{t})]^2$. Since the continuum electron is in an energy eigenstate, we can substitute this energy in for the continuum electron Hamiltonian:

$$|\chi_1(t_e)\rangle = \exp\left(-i \int_{t_b}^{t_e} \hat{H}(t') dt'\right) |\chi_1(t_b)\rangle \quad (2.4.5)$$

$$= \exp\left(-i \int_{t_b}^{t_e} E(t') dt'\right) |\chi_1(t_b)\rangle \quad (2.4.6)$$

$$= \exp\left(-i \int_{t_b}^{t_e} \frac{1}{2} [\mathbf{k} + \mathcal{A}(t')]^2 dt'\right) |\chi_1(t_b)\rangle \quad (2.4.7)$$

$$= \exp(-i S_{\nu 1}(t_b, t_e)) |\chi_1(t_b)\rangle \quad (2.4.8)$$

The continuum electron wavefunction $|\chi_1(t)\rangle$ is propagated forward in time from the moment of ionization (t_b) to the time of recombination (t_e). The integral in Eq. 2.4.7

is recognized as the classical action $S_{\nu 1}$ which is defined as,

$$S_{\nu 1}(t_b, t_e) = \frac{1}{2} \int_{t_b}^{t_e} [\mathbf{k} - \mathcal{A}(t')]^2 dt'. \quad (2.4.9)$$

Since the ion is assumed to be in an eigenstate $|\psi_1^+\rangle$, its evolution between the time of birth t_b and a later time t_e is simply given by the energy of the state E_1 as:

$$|\psi_1^+(t_e)\rangle = \exp(-iE_1(t_e - t_b)) |\psi_1^+(t_b)\rangle \quad (2.4.10)$$

Putting these pieces together, the total wavefunction at t_e is:

$$|\chi_1(t_e), \psi_1^+(t_e)\rangle = e^{-iS_{\nu 1}(t_e, t_b)} e^{-iE_1(t_e - t_b)} |\chi_1(t_b), \psi_1^+\rangle \quad (2.4.11)$$

STEP 3: At time t_e the electron re-encounters the parent ion and the dipole transition between the continuum electron $|\chi_1(t_e), \psi_1^+(t_e)\rangle$ and the ground state $|\psi_N\rangle$ leads to the emission of a photon. Note that from time $t=0$ to t the phase of the neutral has also evolved as:

$$|\psi_N(t)\rangle = \exp(-iE_N t) |\psi_N(t=0)\rangle \quad (2.4.12)$$

$$\langle \psi_N(t) | = \exp(+iE_N t) \langle \psi_N(t=0) | \quad (2.4.13)$$

The matrix element for an electron born at time t_b and re-combining at time t is:

$$\langle \psi_N(t) | \hat{\mathbf{r}} | \chi_2(t), \psi_2^+ \rangle = \langle \psi_N(t=0) | \hat{\mathbf{r}} | e^{+iE_N(t-t_b)} e^{-iS_{\nu 1}(t, t_b)} e^{-iE_1(t-t_b)} |\chi_1(t_b), \psi_1^+ \rangle \quad (2.4.14)$$

The total time dependent dipole at time t is given by the integral over all possible birth times between $t=0$ and t , and canonical momenta $\mathbf{k}$:

$$\begin{aligned} \mathbf{D}(t) &= \int d^3\mathbf{k} \int_0^t dt_b \langle \psi_N(t) | \hat{\mathbf{r}} | e^{-iS_{\nu 1}(t_b, t)} |\chi_1(t_b), \psi_1^+ \rangle \\ &\quad \times e^{+iE_N t} e^{-iE_1(t-t_b)} e^{-iE_N t_b} \\ &\quad \times \langle \chi_1(t_b), \psi_1^+ | \hat{V}_L(t_b) | \psi_N \rangle. \end{aligned} \quad (2.4.15)$$

The acceleration of the induced dipole $\mathbf{D}(t)$ leads to HHG emission. In this derivation, continuum-continuum transitions have been ignored because they do not contribute to HHG. The four-dimensional integral for $\mathbf{D}(t)$ is still expensive to solve numerically.

Fortunately, the laser field and initial velocity of the electron defines the canonical momentum, and for a given emission time t there are only a few birth times (t_b) where the electron returns to the parent ion leading to the emission of an XUV photon. Because of this, the integration can be reduced to a sum over the trajectories that return to the ion at time t by using saddle point methods [65, 61]. Figure 2.6 shows that the *long* and *short* trajectories can be found by solving for the motion of a charged particle that appears in the laser field at birth time t_b and is required to return to the its starting point at some later time t .

Lewenstein et al. [61] performed a saddle point analysis and gave the conditions for an electron to return to the parent ion with the resulting emission of an XUV photon:

1. The electron is born with zero initial velocity.
2. The electron must return to the parent ion.
3. Energy must be conserved such that the $(2M+1)$ -th harmonic is emitted only at times when the electron returns with sufficient energy to generate that photon. This requires that $\frac{1}{2}(\mathbf{k}_{\text{st}}(t, t_b) - \mathcal{A}(t))^2 + I_p = (2M + 1)\omega_L$, where $\mathbf{k}_{\text{st}}(t, t_b)$ is the canonical momentum at the stationary phase point for t_b and t .

For the saddle point solution of Eq. 2.4.15 we usually use the formulation by Yakovlev et al. [66] where:

$$\mathbf{D}(t) = \Re \left[e^{-i\pi/4} \sum_{\text{trajectories}} a_{\text{ion}}(t) a_{\text{pr}}(t) a_{\text{rec}}(t) \right] \quad (2.4.16)$$

Here each step is described with trajectory specific probability amplitudes, $a_{\text{ion}}(t)$, $a_{\text{pr}}(t)$ and $a_{\text{rec}}(t)$.

The instantaneous ionization rate $|a_{\text{ion}}(t)|^2$ is:

$$a_{\text{ion}} = \left(\frac{dn(t_b)}{dt} \right)^{1/2} \quad (2.4.17)$$

where $n(t)$ is the ground state population (Sec. 2.2). The amplitude for an electron born at time t_b returning to the parent ion at time t is:

$$a_{\text{pr}}(t) = \left(\frac{2\pi}{t - t_b} \right)^{3/2} \frac{(2I_p)^{1/4}}{|F_L(t_b)|} \exp[-i(t - t_b)I_p - iS(t)] \quad (2.4.18)$$

where $S(t)$ is the classical action for the electron accelerating in the continuum (See Eq. 2.4.9). The recombination dipole for a returning electron with momentum $\mathbf{v}(t) = \mathbf{A}_L(t') - \mathbf{A}_L(t)$ is:

$$\mathbf{a}_{\text{rec}}(t) = \sqrt{1 - n(t)} \cdot \mathbf{d}(\mathbf{v}) \quad (2.4.19)$$

where $\mathbf{d}(\mathbf{v})$ is the recombination dipole matrix element for the specific atom or molecule under study and the $\sqrt{1 - n(t)}$ term accounts for the blue shift caused by depletion of the ground state while the electron is in the continuum. (Note that [66] uses $\sqrt{n(t)}$). For the case of transition between plane wave continuum states and the hydrogen 1s ground state, the recombination dipole has a simple analytic form:

$$\mathbf{d}(\mathbf{v}) = \langle \Psi_{H_{1s}} | \mathbf{r} | \mathbf{v} \rangle = \frac{\mathbf{v}}{(\mathbf{v}^2 + 2I_p)^3}. \quad (2.4.20)$$

Figure. 2.8 shows an HHG spectrum that was calculated using the SFA and the recombination matrix element for a hydrogen atom. The simulation used a $1.8 \mu\text{m}$ 10 cycle pulse which produced 152 odd harmonics of the 0.69 eV laser photon energy.

2.5 Retrieving Photo-Recombination Cross-Sections from HHG spectra

The preceding section introduced the SFA and showed that the induced dipole $\mathbf{D}(t)$ can be expressed as the sum over all trajectories of the product of terms corresponding to ionization, propagation, and re-combination. The sum over the first two terms defines the returning electron wavepacket that probes the recombination matrix element. The matrix element is assumed to be time independent (unless the target system is undergoing dynamics). The idea of expressing the HHG spectrum as the product of a wavepacket $W(E)$ and the field-free recombination matrix element $d(\omega)$ has been used by a number of authors [67, 68, 69], and was recently developed into the Quantitative Rescattering (QRS) model by the group of C. D. Lin [70]. In QRS the induced dipole acceleration $D(\omega)$ in HHG is written as the product of two terms:

$$D(\omega) = W(E)d(\omega) \quad (2.5.1)$$

where each term in the above expression is a *complex* number with a corresponding amplitude and phase. The quantity $|W(E)|^2$ is the probability distribution, or flux

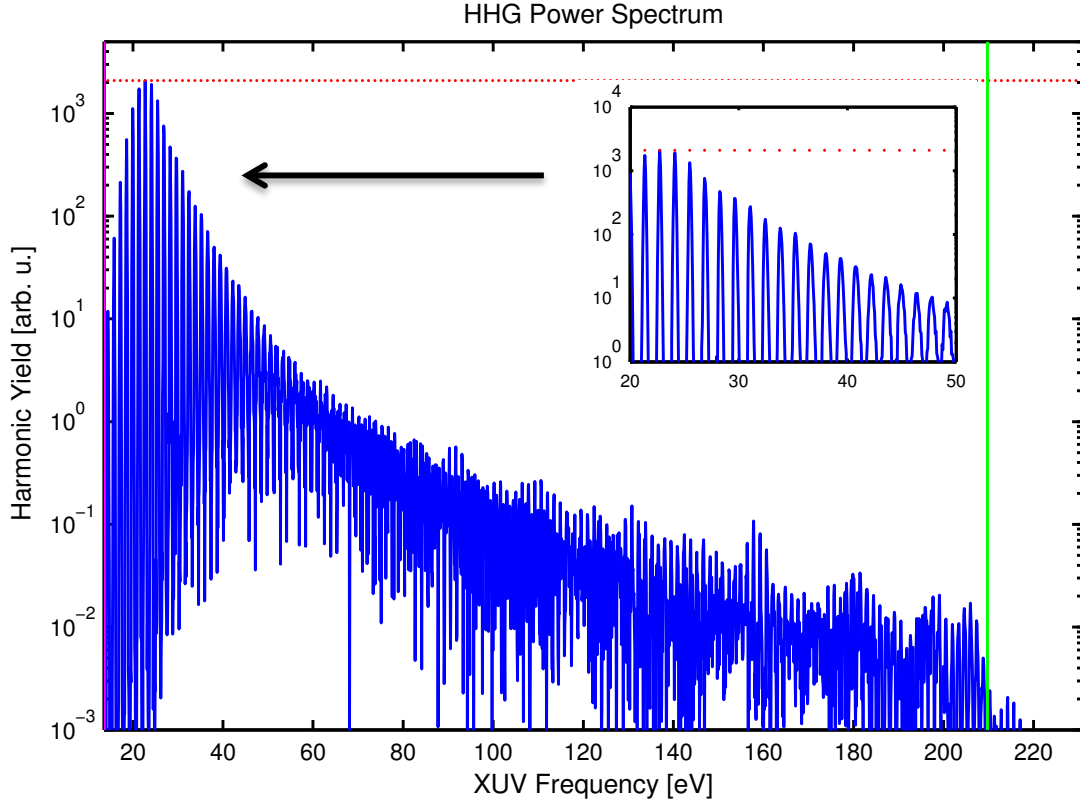


Figure 2.8: HHG spectrum for atomic hydrogen calculated using the strong field approximation for a $1.8\,\mu\text{m}$, 10 cycle pulse. The predicted cutoff based on the scaling law ($E_c = 210\,\text{eV}$), is indicated with the green line and the inset shows an expanded view of the region between 20 and 50 eV. Discrete harmonics at odd multiples of the laser photon energy ($E_{\text{photon}} = 0.69\,\text{eV}$), are visible.

of electrons of a given energy E returning to the parent ion. The returning electron energy is related to the emitted photon energy by $E = \omega - I_p$. The key finding of QRS is that the wavepacket depends on the laser parameters, but is largely independent of the details of the structure of the atom or molecule that is being used to produce the high harmonics [70, 71]. The wavepacket can be determined from a reference HHG spectrum $P^{\text{ref}}(\omega)$ that is either measured or calculated (for instance using the SFA), from a system whose recombination matrix element $d^{\text{ref}}(\omega)$ is known. This calibration can be used to analyze a second HHG spectrum $P(\omega)$ that is obtained under the same laser conditions. The returning electron wavepacket measured with the reference HHG spectrum is factored out of the spectrum under study, leaving behind the

recombination dipole or equivalently its photo-recombination cross-section.

$$P(\omega) = \left| \frac{d(\omega)}{d^{\text{ref}}(\omega)} \right|^2 P^{\text{ref}}(\omega) \quad (2.5.2)$$

$$= \frac{\sigma(\omega)}{\sigma^{\text{ref}}(\omega)} P^{\text{ref}}(\omega) \quad (2.5.3)$$

In the above expression σ and σ^{ref} are short-hand notations for unknown and ‘reference’ differential photo-recombination cross-sections respectively. The calculation of field-free cross-sections is discussed in more detail in Ch. 8.

Chapter 3

Wavelength Scaling of High Harmonic Generation

This chapter is based on the following paper:

A. D. Shiner, C. Trallero-Herrero, N. Kajumba, H.-C. Bandulet, D. Comtois, F. Légaré, M. Giguère, J.-C. Kieffer, P. B. Corkum, and D. M. Villeneuve, “Wavelength scaling of high harmonic generation efficiency,” *Physical Review Letters*, vol. 103, no. 7, p. 073902, 2009

Section 2.4 introduced the SFA and showed that the most energetic (cutoff), harmonic scales as $E_c = I_p + 3.17 U_p$ where $U_p \propto I \lambda^2$. Clearly, by increasing the laser wavelength we can significantly increase the energy of the cutoff harmonic. At present the vast majority of HHG experiments use 800 nm Ti:Sa lasers, which can achieve pulse energies on the order of Joules, and pulse durations of less than 5 femtoseconds. This technology has reached maturity, and scientists are now looking to increase the HHG cutoff by increasing the laser wavelength (Fig. 3.1). One topic in the HHG community which has gained considerable interest in the past few years has been the scaling of HHG yield with laser wavelength. Theoretically this yield was expected to drop as roughly λ^{-5} [72, 73, 74] but experimental proof of this remained elusive. The reason for this dramatic decrease can be understood by referring back to the SFA. From Eq. 2.4.18 we note that during the time the electron spends accelerating in the continuum its wavepacket is spreading. This decreases the probability of the electron successfully re-encountering the parent ion and lowers the harmonic yield.

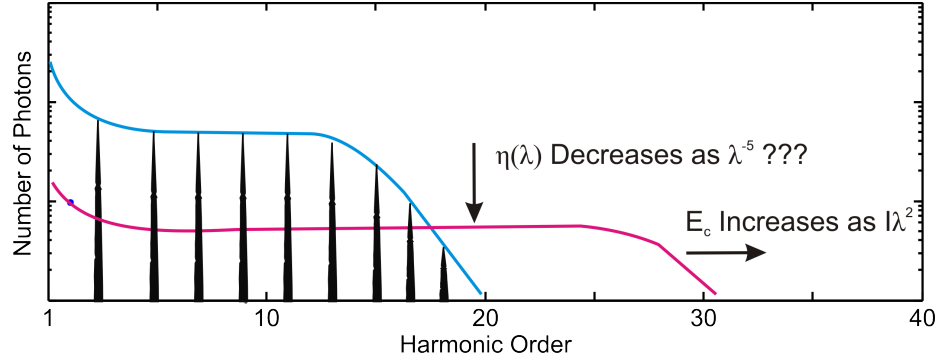


Figure 3.1: In HHG the cutoff is proportional to the laser intensity times the square of the laser wavelength. However, as the wavelength increases the harmonic yield decreases.

The probability for the electron re-encountering the parent ion $|a_{\text{pr}}(t)|^2$ is:

$$|a_{\text{pr}}(t)|^2 \propto \left(\frac{2\pi}{t - t_b} \right)^3. \quad (3.0.1)$$

For a given harmonic, the time the electron spends in the continuum $(t - t_b)$ is proportional to λ , so the three dimensional spread of the wavepacket decreases the harmonic yield as λ^3 [61].

The $I\lambda^2$ scaling of the cutoff also decreases the yield in a given spectral interval. This is because the total energy in the xuv beam is spread across its spectral bandwidth. If the bandwidth increases, with all other things being equal, the yield in a given region of that spectrum will decrease. Because the bandwidth increases as λ^2 the harmonic yield integrated over a given interval of the spectrum is expected to decrease with λ^2 .

Combining the contributions of wavepacket spreading and increased spectral bandwidth the harmonic yield is expected to fall as λ^{-5} [72]. In the study that is presented in the following paper we find that the yield in fact decreases as $\lambda^{-6.3 \pm 1.1}$ in xenon and $\lambda^{-6.5 \pm 1.1}$ in krypton, slightly worse than theory predicts. This means that from a practical standpoint you cannot increase the cutoff indefinitely by simply increasing the laser wavelength. Greater gas density or improved phase matching will be required in order to compensate for the decrease in XUV flux.

Wavelength Scaling of High Harmonic Generation Efficiency

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Using longer wavelength laser drivers for high harmonic generation is desirable because the highest extreme ultraviolet frequency scales as the square of the wavelength. Recent numerical studies predict that high harmonic efficiency falls dramatically with increasing wavelength, with a very unfavorable $\lambda^{-(5-6)}$ scaling. We performed an experimental study of the high harmonic yield over a wavelength range of 800–1850 nm. A thin gas jet was employed to minimize phase matching effects, and the laser intensity and focal spot size were kept constant as the wavelength was changed. Ion yield was simultaneously measured so that the total number of emitting atoms was known. We found that the scaling at constant laser intensity is $\lambda^{-6.3 \pm 1.1}$ in Xe and $\lambda^{-6.5 \pm 1.1}$ in Kr over the wavelength range of 800–1850 nm, somewhat worse than the theoretical predictions.

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PACS numbers: 42.65.Ky, 32.80.Wr, 42.65.Re

High harmonic generation (HHG) is a three-step process [1,2] in which an intense laser removes an electron from an atom or molecule, accelerates the electron, and then recombines the electron with the parent ion. The resulting emission extends to the extreme ultraviolet (XUV) and is composed of odd harmonics of the driving laser frequency. HHG provides a source of attosecond pulses [3–5]. The recombination of the recolliding electron also provides a measure of the electronic structure of molecules [6]. Increasing the extent of the XUV spectrum can lead to shorter attosecond pulses [5] and will give better resolution for molecular orbital imaging [6] by reducing the electron's de Broglie wavelength [7].

The highest frequency produced [8], for a given ionization state, is approximately $\Omega_{\max} = 3.17U_p + I_p$, where $U_p = F^2/(4\omega^2)$ is the ponderomotive energy, F is the peak laser electric field, ω is the laser angular frequency, and I_p is the ionization potential of that state (atomic units are used). U_p scales as $I\lambda^2$, where I is the peak laser intensity and λ is the laser wavelength. One approach to extend the cutoff frequency $\Omega_{\max}$ is to increase the laser intensity. Unfortunately, there is a practical limit beyond which the intensity cannot be increased—the point at which the state is fully ionized.

An alternate approach is to use longer laser wavelengths [9,10]. The λ^2 scaling of $\Omega_{\max}$ is unfortunately predicted to be offset by an unfavorable scaling of the conversion efficiency from laser energy into XUV energy due to the spreading of the continuum electron wave packet in both space and time. This was first explored numerically by Tate *et al.* [11] using time-dependent Schrödinger and strong field approximation calculations to show that the single-atom HHG efficiency $\eta(\lambda) \propto \lambda^{-(5-6)}$ at constant intensity when integrated over a few tens of eV in the XUV spectrum. This power law has been verified by further theoretical studies [12,13].

There have been several experimental studies on the dependence of different properties of HHG on laser wavelength. These studies include a demonstration that the cutoff frequency is extended with longer wavelength drivers but with a loss of efficiency [10,14], and measurements of the phase matching conditions that optimize the XUV cutoff for different wavelengths [15–17]. There has also been a recent report of the wavelength dependence of the attosecond chirp [18]. Despite strong interest in the wavelength scaling of HHG efficiency by the attosecond community, there are very few quantitative studies reported in the literature. Shan and Chang [10] compared the intensity of the 37th harmonic in Xe for three wavelengths from 1.22 to 1.51 μm . They found that from the first to the second wavelength the harmonic intensity decreased as roughly the cube of the free electron transit time as predicted by Lewenstein *et al.* [8]. The measurement for the third wavelength deviated from this trend by nearly a factor of 2. Colosimo *et al.* [19] reported measurements at two wavelengths. Their experiment was not designed to be sensitive to the single atoms response, and the large decrease in HHG efficiency with increasing wavelength was shown to be sensitive to the gas density and hence to phase matching. Thus, experimental confirmation of the wavelength scaling at the single-atom level has remained elusive.

It is difficult to design an experiment in which the laser wavelength is the only parameter that changes. We wish to provide experimental confirmation of the single-atom calculations, and so we must avoid collective effects such as phase mismatch, differing focal spot diameters, and different intensities. We have carefully arranged the experiment so that all of the important parameters are controlled. (1) The laser beam is spatially filtered through a hollow core fiber capillary at all wavelengths. (2) The output of the capillary is relay-imaged into the gas jet in which harmonics are generated, providing the same focal spot diameter at

all wavelengths. (3) The gas jet is only 0.5 mm thick, to minimize the possibility of phase mismatch and reabsorption. (4) The number of ions produced is independently measured. (5) The intensity in the gas jet is determined at each wavelength by measuring the ion yield versus intensity. (6) As a final correction, the high harmonic yield is normalized by the ion yield squared so that the emission per atom is measured.

The experimental arrangement is shown in Fig. 1. The 800 nm radiation was produced directly by a Ti:sapphire laser (Thales, 100 Hz, 80 mJ, 30 fs). Longer wavelengths were produced by directing 7 mJ of the 800 nm radiation into an optical parametric amplifier (OPA) (Light Conversion TOPAS-HE) which output 2.3 mJ (signal + idler). From this source we selected three wavelengths: 1300, 1400 (signal), and 1850 nm (idler). Further details can be found in Ref. [20].

Each source was coupled into a 400 μm -diameter hollow core fiber which performs spatial filtering and provides a well-defined spatial mode for harmonic generation regardless of wavelength [20]. The mode exiting the fiber was imaged into the gas jet with the $4f$ image relay geometry shown in Fig. 1. The distance between the collimating and focusing lenses was equal to the sum of the focal lengths of each lens. The $4f$ geometry ensures that the spatial mode in the gas jet will have the same 33 μm spot size, regardless of wavelength.

The gas jet was operated with a constant backing pressure of ~ 3 bar of xenon or krypton. The thickness of the gas sample was 0.5 mm, in all cases much shorter than the Rayleigh range of the laser focus. Gas density in the focus was estimated to be 10^{17} cm^{-3} .

Harmonic radiation produced in the gas jet passed into an XUV spectrometer where the beam was spectrally dispersed by a spherical grating (Hitachi 001-0266) and then imaged onto a microchannel plate (MCP) backed by a phosphor screen (Burle APD 3115 32/25/8 I EDR MgF2 P20). The resulting spectrum was recorded with a high dynamic range digital camera. In all cases we averaged the observed spectra over several laser shots and applied corrections for the wavelength-dependent efficiency of the MCP [21] as well as the spectrometer grating [22].

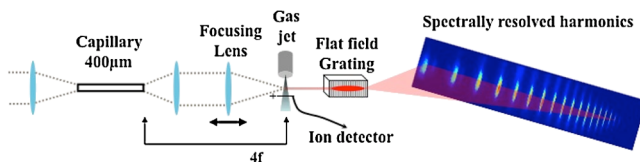


FIG. 1 (color online). Optical layout. Various laser sources were focused into a hollow core waveguide to improve the spatial mode quality and to produce the same beam diameter. The output of the waveguide was imaged into the pulsed valve gas jet in which the high harmonics were produced. The $4f$ imaging system ensured that the output of the waveguide was imaged into the gas jet.

An ion detector consisting of a wire mesh biased at -500 V was located below the pulsed jet to measure the number of ions produced during harmonic generation. To maintain the same focus conditions, the laser beam was focused directly beneath the pulsed jet by finding the lens position that maximized the ion yield. The resulting harmonics propagated as a low divergence beam with no visible annular structure, consistent with short trajectory only.

To verify our detection system, we measured the dependence of the ion signal and the integrated harmonic energy $E(\lambda)$ on pulsed valve gas backing pressure, as shown in Fig. 2. We find that the ion signal increases linearly with backing pressure, confirming that the current is proportional to the number of ions produced in the interaction volume. The total harmonic energy is seen to increase quadratically with pressure. This is to be expected since HHG is a phase-matched process in which the electric fields from individual atoms add coherently [23–25], provided that there is no significant phase mismatch. Calculations of the phase mismatch due to ionization of the sample show that it is negligible with our short 0.5 mm jet. Reabsorption of the XUV is also calculated to be negligible but in any case is the same for all laser wavelengths for each atom. The observed quadratic increase in harmonic yield with pressure proves that we are operating in a regime that is free of significant phase mismatch or reabsorption. By dividing the harmonic energy by the square of the ion yield, we can remove the measurement's sensitivity to variations in gas pressure, as shown in the lower panel of Fig. 2.

As HHG is a highly nonlinear process, we must be able to accurately control and measure the laser intensity in the

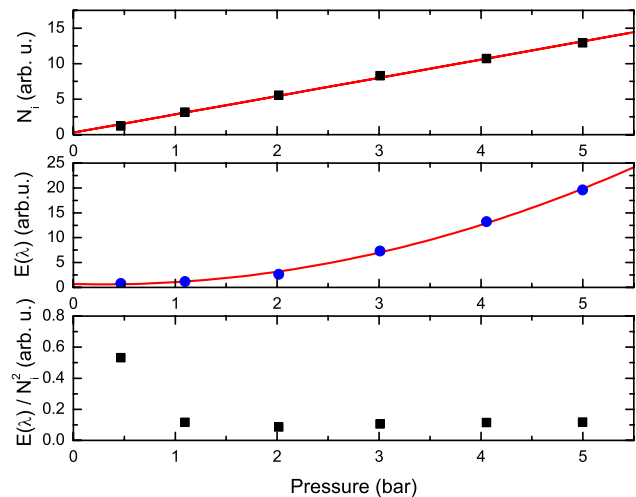


FIG. 2 (color online). Measure ion signal (top) and integrated harmonic yield (middle) from Xe as a function of the pulsed valve backing pressure. The ion signal is proportional to the backing pressure, and the harmonic yield increases quadratically with pressure, confirming that there is no significant phase mismatch. Dividing the integrated harmonic yield by the square of the ion yield (bottom), we effectively remove the pressure dependence of the measurement.

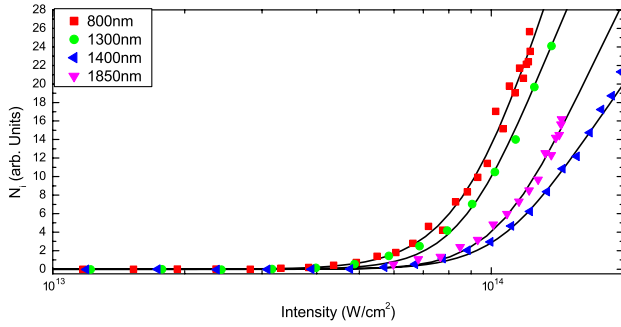


FIG. 3 (color online). Points are the experimentally measured ion signal from Xe recorded for each wavelength and a range of laser pulse energies. The solid lines show the Yudin-Ivanov ionization rate integrated over the focal volume.

interaction volume for all wavelengths. In order to calibrate the peak intensity at each wavelength, the ion yield N_i was recorded as a function of pulse energy, as shown by the symbols in Fig. 3. The solid curves were calculated using the Yudin-Ivanov nonadiabatic ionization model [26] integrated over a Gaussian spatial intensity distribution. This model includes both quasistatic tunneling as well as the wavelength-dependent multiphoton contribution to the ionization probability. We minimize the square error between theory and experiment by scaling the horizontal (intensity) axis of the measured data and the vertical (ionization yield) axis of the theoretical curve. This fitting procedure determines the absolute intensity of the laser in the interaction volume for a given pulse energy. The saturation intensity I_{sat} was determined from the semilog plot of Fig. 3 by extrapolating the asymptotic part of each theoretical curve to zero signal [27]. At saturation, 43% of the atoms in the most intense region of the focus will have ionized. On such a plot the limiting slope is proportional to the focal volume and gas density. For all wavelengths the limiting slopes agree to within $\pm 32\%$ (1σ) indicating that the $4f$ focusing geometry effectively maintains a constant interaction volume.

To facilitate direct measurement of the total XUV energy, the flat field spectrometer was operated without an entrance slit. Because of the beam's small transverse dimension, discrete harmonics are still observed. A representative HHG spectrum is shown in the inset in Fig. 1. By removing the entrance slit, we are able to directly determine the XUV energy by integrating the pixel values from the spectral image on the MCP that correspond to a given range of photon energies. The XUV detection system and this procedure are independent of laser wavelength.

Because of the large range of HHG amplitudes as the laser wavelength is changed, care has to be exercised to maintain linearity of the MCP. To ensure linearity, the MCP voltage was adjusted for each laser condition to ensure that the MCP was not saturating. We separately characterized the voltage-dependent gain of the MCP.

HHG yield as a function of laser intensity was recorded at four wavelengths. We chose two conditions to compare

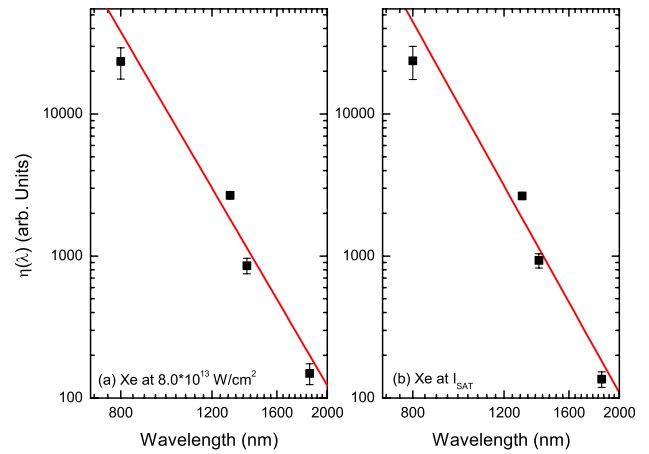


FIG. 4 (color online). Scaling of harmonic yield in Xe in the range 16–32 eV, corrected for the single-atom ionization rate (see text). Results shown at a constant intensity of 8.0×10^{13} W/cm² (a) and at saturation intensity I_{sat} (b). The scaling law is $\eta(\lambda) \propto \lambda^x$, where $x = -6.3 \pm 1.1$ at 8.0×10^{13} W/cm² and $x = -6.6 \pm 1.0$ at I_{sat} .

the HHG efficiency versus wavelength—constant intensity and constant ionized fraction. Figure 4(a) shows the experimental results in Xe at a constant intensity of 8.0×10^{13} W/cm². Figure 4(b) shows the experimental results where the ionized fraction was kept constant by setting the laser to the saturation intensity I_{sat} at each wavelength, as shown in Table I. Experimental results for Kr gas at a constant intensity of 1.3×10^{14} W/cm² are shown in Fig. 5(a) as well as at a constant fraction of the saturation intensity $0.8I_{\text{sat}}$ in Fig. 5(b).

Our goal is to determine the single-atom scaling, to enable comparison with recent theoretical predictions. To make this comparison, we must convert the coherent emission from N_0 neutral atoms by dividing $E(\lambda)$ by N_0^2 . We directly measure the ion yield $N_i = N_0 P_{\text{YI}}$, and we determine the ionization probability P_{YI} from the Yudin-Ivanov model fit shown in Fig. 3. We have thus plotted $\eta(\lambda) = E(\lambda)/N_0^2$ in Figs. 4 and 5. The exponent x for the deduced scaling law $\eta(\lambda) \propto \lambda^{-|x|}$ is shown in Table II.

It was proposed by Yakovlev, Ivanov, and Krausz [28] that it is possible to overcome the single-atom response associated with longer driver wavelengths by reducing the laser intensity. The idea is that you want to produce emission at a particular frequency, and since the cutoff scales as $I\lambda^2$, one can reduce I . Lower intensity produces fewer

TABLE I. Laser parameters used for the study.

λ (nm)	Source	τ (fs)	I_{sat} (10^{14} W/cm ²)	
			Xe	Kr
800	Ti:sapph.	62	0.84	1.50
1300	OPA sig.	130	0.84	1.44
1400	OPA sig.	55	1.01	1.73
1850	OPA idl.	64	1.02	1.72

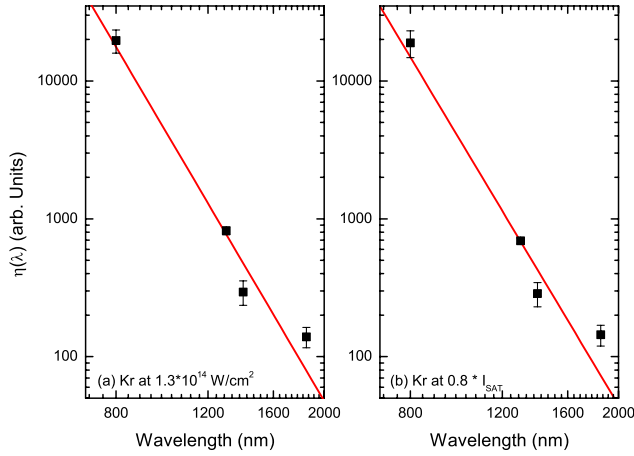


FIG. 5 (color online). Scaling of harmonic yield in Kr in the range 16–32 eV corrected for the single-atom ionization rate (see text). Results shown are for a constant intensity 1.3×10^{14} W/cm² (a) as well as at a constant fraction of the saturation intensity $0.8I_{\text{sat}}$ (b). The scaling law is $\eta(\lambda) \propto \lambda^x$, where $x = -6.5 \pm 1.1$ at 1.3×10^{14} W/cm² and $x = -6.4 \pm 1.3$ at $0.8I_{\text{sat}}$.

electrons and thus less phase mismatch. It is not possible for us to test this conjecture, since we intentionally used a very short gas jet to avoid phase mismatch. We also cannot test the scaling at constant $I\lambda^2$ because that implies a very low intensity at the longest wavelength, an intensity at which we could not detect any harmonics. Our results suggest just the opposite, that the XUV emission from a short medium in the 16–32 eV range goes down even more rapidly than λ^{-5} if the intensity is adjusted as $I(\lambda) \propto \lambda^{-2}$. To achieve Yakovlev's goal, the medium must also be extended or the focusing geometry changed.

In summary, we measured the high harmonic efficiency versus wavelength at constant intensity and at constant ionization fraction, using a short gas jet where phase mismatch is not an issue. By simultaneously measuring the ionization, we normalize to the number of atoms and isolate the single-atom response. We observed a scaling law of $\eta(\lambda) \propto \lambda^{-6.3 \pm 1.1}$ in Xe and $\lambda^{-6.5 \pm 1.1}$ in Kr (see Table II). The dramatic decrease in HHG efficiency predicted by Tate *et al.* [11] came as a surprise to the attosecond community and suggests that there will be significant challenges for experiments employing longer wavelength sources. Our finding that the single-atom HHG

TABLE II. Value of the efficiency scaling exponents obtained for $\eta(\lambda) \propto \lambda^{-|x|}$ integrated in the 16–32 eV range.

Gas	Intensity	Exponent
Xe	8.0×10^{13} W/cm ²	-6.3 ± 1.1
Xe	I_{sat}	-6.6 ± 1.0
Kr	1.3×10^{14} W/cm ²	-6.5 ± 1.1
Kr	$0.8I_{\text{sat}}$	-6.4 ± 1.3

yield decreases even more rapidly than predicted will be an important consideration in the design of future experiments.

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- [1] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
- [2] K. J. Schafer, B. Yang, L. F. DiMauro, and K. C. Kulander, Phys. Rev. Lett. **70**, 1599 (1993).
- [3] A. Scrinzi, M. Y. Ivanov, R. Kienberger, and D. M. Villeneuve, J. Phys. B **39**, R1 (2006).
- [4] P. B. Corkum and F. Krausz, Nature Phys. **3**, 381 (2007).
- [5] G. Sansone *et al.*, Science **314**, 443 (2006).
- [6] J. Itatani *et al.*, Nature (London) **432**, 867 (2004).
- [7] V.-H. Le, A.-T. Le, R.-H. Xie, and C. D. Lin, Phys. Rev. A **76**, 013414 (2007).
- [8] M. Lewenstein, P. Balcou, M. Y. Ivanov, A. L'Huillier, and P. B. Corkum, Phys. Rev. A **49**, 2117 (1994).
- [9] B. Sheehy *et al.*, Phys. Rev. Lett. **83**, 5270 (1999).
- [10] B. Shan and Z. Chang, Phys. Rev. A **65**, 011804 (2001).
- [11] J. Tate, T. Augustine, H. G. Muller, P. Salières, P. Agostini, and L. F. DiMauro, Phys. Rev. Lett. **98**, 013901 (2007).
- [12] K. Schiessl, K. L. Ishikawa, E. Persson, and J. Burgdörfer, Phys. Rev. Lett. **99**, 253903 (2007).
- [13] M. V. Frolov, N. L. Manakov, and A. F. Starace, Phys. Rev. Lett. **100**, 173001 (2008).
- [14] B. Shan, A. Cavalieri, and Z. Chang, Appl. Phys. B **74**, s23 (2002).
- [15] T. Popmintchev, M.-C. Chen, O. Cohen, M. E. Grisham, J. J. Rocca, M. M. Murnane, and H. C. Kapteyn, Opt. Lett. **33**, 2128 (2008).
- [16] E. J. Takahashi, T. Kanai, K. L. Ishikawa, Y. Nabekawa, and K. Midorikawa, Phys. Rev. Lett. **101**, 253901 (2008).
- [17] Y. Fu *et al.*, Phys. Rev. A **79**, 013802 (2009).
- [18] G. Doumy, J. Wheeler, C. Roedig, R. Chirla, P. Agostini, and L. F. DiMauro, Phys. Rev. Lett. **102**, 093002 (2009).
- [19] P. Colosimo *et al.*, Nature Phys. **4**, 386 (2008).
- [20] H.-C. Bandulet *et al.*, J. Phys. B **41**, 245602 (2008).
- [21] R. Hemphill, J. Edelstein, and D. Rogers, Appl. Opt. **36**, 1421 (1997).
- [22] J. Edelstein, M. C. Hettrick, S. Mrowka, P. Jelinsky, and C. Martin, Appl. Opt. **23**, 3267 (1984).
- [23] P. Balcou, P. Salières, A. L'Huillier, and M. Lewenstein, Phys. Rev. A **55**, 3204 (1997).
- [24] E. Constant *et al.*, Phys. Rev. Lett. **82**, 1668 (1999).
- [25] M. B. Gaarde, J. L. Tate, and K. J. Schafer, J. Phys. B **41**, 132001 (2008).
- [26] G. L. Yudin and M. Y. Ivanov, Phys. Rev. A **64**, 013409 (2001).
- [27] S. M. Hankin, D. M. Villeneuve, P. B. Corkum, and D. M. Rayner, Phys. Rev. A **64**, 013405 (2001).
- [28] V. S. Yakovlev, M. Y. Ivanov, and F. Krausz, Opt. Express **15**, 15 351 (2007).

Chapter 4

Characterization of Ionization and Intensity During High Harmonic Generation

This chapter is based on the following papers:

1. **A. D. Shiner**, C. Trallero-Herrero, N. Kajumba, B. E. Schmidt, Kyung Taec Kim, H.-C. Bandulet, D. Comtois, F. Légaré, J.-C. Kieffer, D. M. Rayner, P. B. Corkum, and D. M. Villeneuve, “Characterization of ionization and intensity during high harmonic generation,” 2012. (In Preparation)

This chapter shows that a simple ion detector in the HHG spectrometer target chamber measures a signal that is proportional to the number of ions produced in the laser focus, and does so over the range of pressures that are typically used for HHG experiments. By measuring the dependence of the ion signal on pulse energy and focal position it is possible to determine the laser intensity in the gas medium as well as the laser focal parameters such as spot size and Rayleigh range. The main results of this chapter have been prepared for publication. This chapter starts by providing some additional background for the paper. First it describes the intensity dependence of the single atom ionization yield, and then go on to explain how this affects the mode-integrated ion yield that we measure when varying the laser parameters such as intensity or focal spot size. Following this introduction the main ion detector paper is given.

4.1 Intensity Dependent Ionization Yield

Section 2.2 introduced strong field ionization, and the Yudin Ivanov [57] non-adiabatic ionization model, which was used throughout this thesis. The following analysis assumes a random carrier-envelope phase, and parameterizes the instantaneous ionization rate Γ with the laser electric field envelope $A(t)$, and the peak intensity at a given radius I_r . The probability that a neutral atom at radius r is ionized by the end of a laser pulse is:

$$P(r)_{\text{ion}} = \left(1 - e^{-\int_{-\infty}^{\infty} \Gamma(I_r, A(t)) dt}\right) \quad (4.1.1)$$

So far the effect of spatial averaging has been ignored and I have only considered the single atom response. A plot of the probability for ionizing a xenon atom with a 1.8 μm , 45fs laser pulse is plotted in Fig. 4.1 for a range of intensities. At an intensity

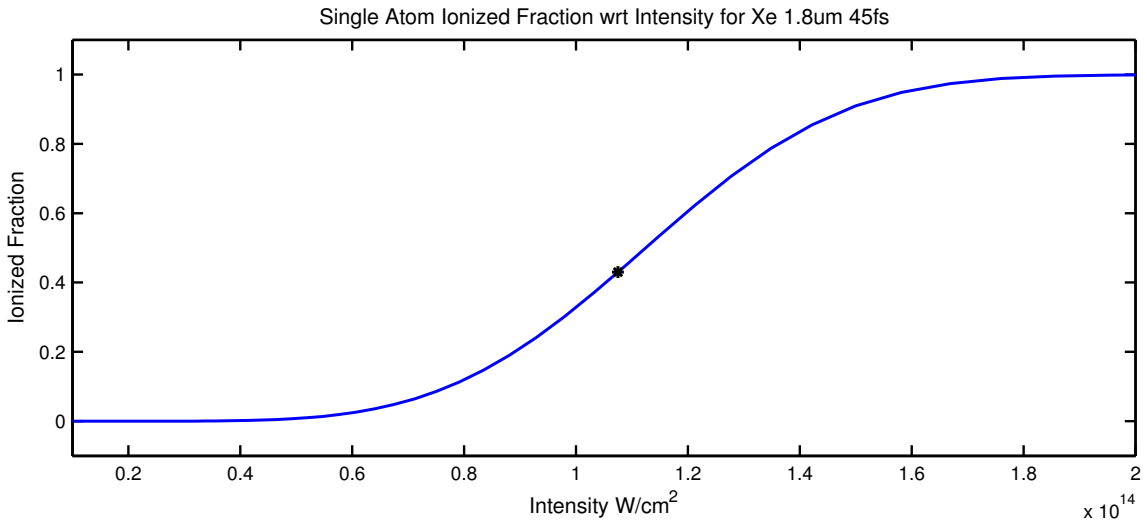


Figure 4.1: Calculated single atom ionization yield for xenon in a 1.8 μm , 45 fs pulse.

of $1.08 \cdot 10^{14} \text{ W}/\text{cm}^2$, 43% of the sample is ionized. In most cases, a laser is focused into a gas sample and the ion yield that we measure is integrated over the intensity distribution in the focus. For all of the measurements presented in this thesis, the laser was focused into a thin gas jet which was typically $\sim 0.5 \text{ mm}$ in diameter. This is much shorter than the Rayleigh length of the laser focus $\sim 10 \text{ mm}$, so we can assume that the intensity only depends on the radial distance from the axis and does not vary in the propagation direction as the beam passes through the jet. The single

atom ionization yield for a range of intensities is plotted in Fig. 4.2 as a function of radial position from the focus. At high intensity, the atoms on-axis ($r = 0$), are fully ionized. Following the analysis of Hankin et. al. [75], the ion signal that we would

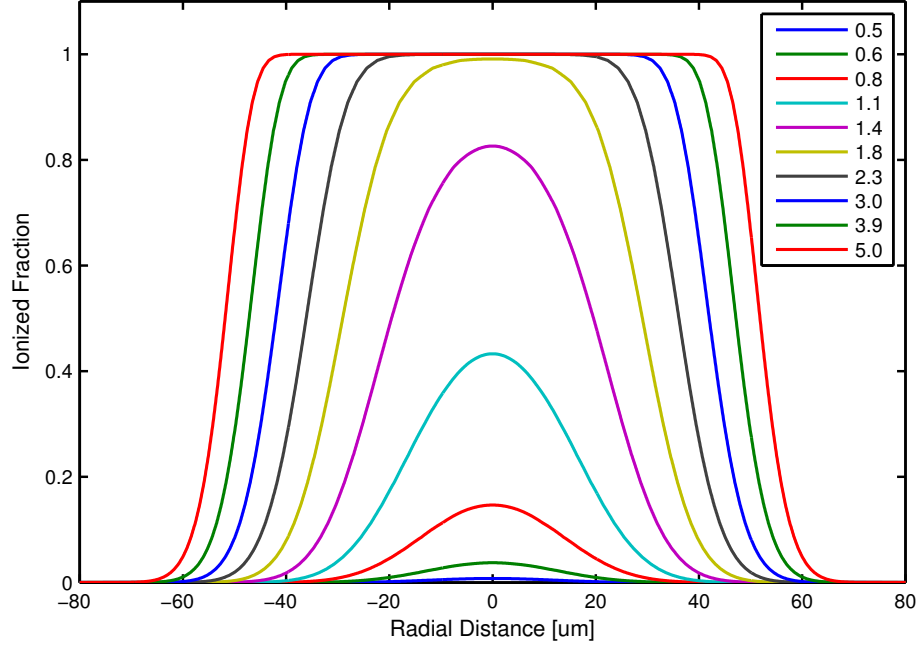


Figure 4.2: Radial dependence of the single atom ion yield for xenon in a $1.8 \mu\text{m}$, 45 fs pulse with focal spot size $w_0 = 60 \mu\text{m}$. Peak on-axis intensities are given in the legend [$\times 10^{14} \text{ W/cm}^2$].

detect from a volume element at radius r is:

$$dS = \alpha 2\pi r dr cl \left(1 - e^{-\int_{-\infty}^{\infty} \Gamma(I_r, A(t)) dt} \right) \quad (4.1.2)$$

where α is the ion detector sensitivity, c is the neutral atom concentration and l is the thickness of the sample. If we integrate over a Gaussian intensity distribution of the form $I(r) = I_r = I_0 \exp(-r^2/R^2)$ and make a change of variables from dr to dI_r

then we get:

$$dS = \frac{\alpha \pi c l R^2 dI_r}{I_r} \left(1 - e^{-\int_{-\infty}^{\infty} \Gamma(I_r, A(t)) dt} \right) \quad (4.1.3)$$

$$S = \alpha \pi c l R^2 \int_0^{I_0} \frac{\left(1 - e^{-\int_{-\infty}^{\infty} \Gamma(I_r, A(t)) dt} \right) dI_r}{I_r} \quad (4.1.4)$$

$$(4.1.5)$$

Differentiating with respect to $\ln(I_0)$ gives:

$$\frac{dS}{d \ln(I_0)} = \alpha \pi c l R^2 \left(1 - e^{-\int_{-\infty}^{\infty} \Gamma(I_r, A(t)) dt} \right) \quad (4.1.6)$$

At low intensities, the rate at which the ion yield increases with intensity will depend on the details of the single atom ionization rate $\Gamma(I_r, A(t))$. Once the atoms in the most intense part of the beam are fully ionized, the term $\left(1 - e^{-\int_{-\infty}^{\infty} \Gamma(I_r, A(t)) dt} \right)$ goes to one and the limiting slope on a semilog plot is: $\alpha \pi c l R^2$. This asymptotic behaviour is clearly seen in Fig. 4.3. Extrapolating the asymptotic slope back to the intensity axis gives the saturation intensity I_{sat} , which is the intensity where 43% of the sample in the most intense (on axis), part of the beam is ionized by the end of the pulse. This is indicated with the black triangle in Fig. 4.3.

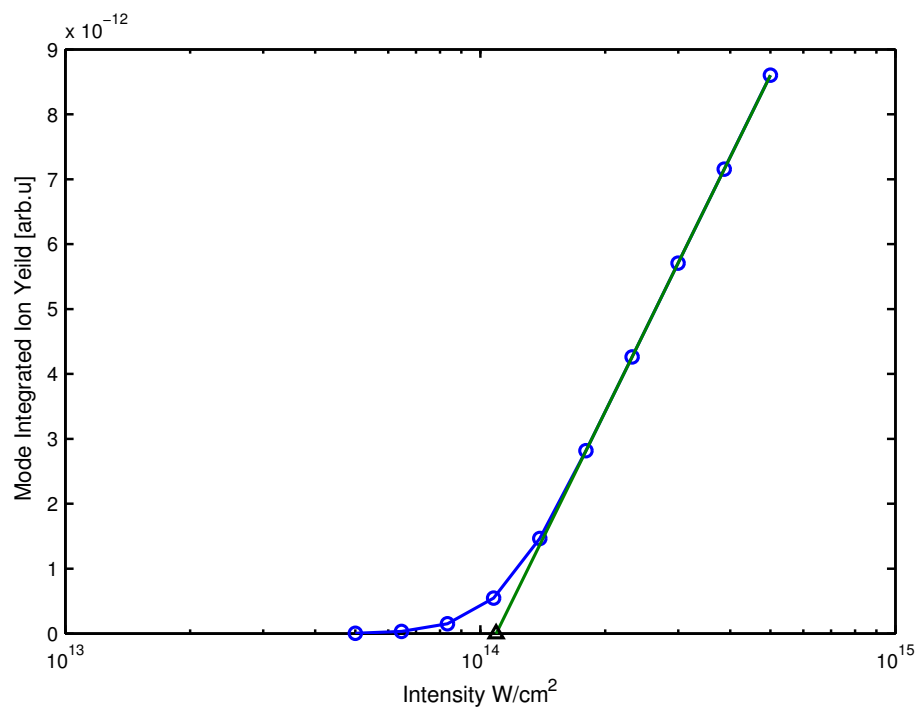


Figure 4.3: Single atom ionization yield for Xe in a 1.8 μm 45 fs pulse. The saturation intensity is indicated with a black triangle.

Characterization of Ionization and Intensity During High Harmonic Generation

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(Dated: September 19, 2012)

High harmonic generation in gas targets leads to the production of attosecond pulses. The process of high harmonic generation requires that the gas be ionized by an intense femtosecond laser field. We show that the ability to measure the ionization process in the gas target is an important tool in understanding and optimizing high harmonic generation. We show that a simple ion detector can determine the laser intensity in the gas jet, find the location of the laser focus as well as the focal parameters, optimize pulsed valve timing, verify phase-matched generation, and predict the harmonic cutoff. We demonstrate these measurements using an 1800 nm laser source and xenon target, with a cutoff reaching a photon energy of 100 eV.

PACS 42.65.Ky, 42.65.Re, 32.80.Rm

I. INTRODUCTION

In high harmonic generation (HHG), an intense ($> 10^{14}$ W/cm²) femtosecond laser field ionizes an atom. The free electron is accelerated in the continuum by the laser field, and then recombines with the parent ion, leading to the emission of an extreme ultraviolet photon [1, 2]. The acceleration of the electron in the field can be substantial, resulting in recollision energies of hundreds of electron volts and the production of soft x-ray photons [3–5]. This process is periodic, repeating every half-cycle of the laser field and this periodicity is observed in the spectrum as *harmonics* at odd multiples of the fundamental laser photon energy. The process is locked to the optical cycle of the driving laser, and the emission from all atoms in the focal volume adds coherently. There are various geometries used for the gas target – tube [6], pulsed valve [7], gas cell [8] or capillary [5]. Pulsed valve gas sources are used in high harmonic spectroscopy applications [9, 10] because the high pressure expansion produces rotationally cold molecules that can be aligned in space using impulsive alignment techniques [11, 12]. Because the gas from such sources expands into the vacuum chamber, we have experimental access to the ions that are produced in the laser focus. Since ionization is the first step in HHG, experimental access to the ions can give insight into the HHG process.

In this paper we will show that a simple ion detector can be coupled with a pulsed valve gas jet to characterize the ionization step of the harmonic generation process. This detector has proven to be a very useful diagnostic tool because the measured ion signal is proportional to the number of ions produced for the range of gas densities normally used in HHG experiments. Measuring the

dependence of the ion signal on the pulsed jet position and timing allows the jet to be reliably positioned and synchronized with respect to the laser, and gives a measure of the spatial and temporal profile of the gas pulse. The laser intensity can be calibrated in situ by recording the dependence of the ion signal on pulse energy. Finally, the harmonic yield can be normalized to the ion signal which allows for a measurement of the single-atom harmonic yield.

Ionization detectors have been used by other groups to detect rotational revivals during HHG experiments with aligned molecules [13, 14]. They have also been used to optimize harmonic generation when harmonics are produced in femtosecond enhancement cavities [15] as well as to detect ions resulting from photoionization with xuv photons [16].

This paper is organized as follows. In Sec. II we give an overview of the experimental setup used for HHG experiments and then describe the ion detector in detail. Section III demonstrates that the ion signal scales linearly with gas pressure in the laser focus over the range of pressures commonly used for HHG experiments. We then show that, because HHG is a phase matched process, the harmonic yield increases quadratically with pressure and that by normalizing to the ion yield it is possible to normalize to the number of emitters in the focus. In Sec. IV we take advantage of the strong dependence of the ionization rate on laser intensity. By measuring the ion signal as a function of pulse energy and fitting this data to an ionization model we calibrate the laser intensity in the interaction volume. In Section V we compare the observed HHG cutoff to the cutoff that would be expected based on the intensity measured with the ion detector. Section VI shows that, by varying the timing between the pulsed gas jet and the laser pulse, we can measure

the temporal profile of the gas pulse. In Section VII a measurement of the ion signal as a function of jet position is used to locate the focus and reliably position the jet a known distance from the focus. Section VIII shows that the ion signal can be used to determine the laser focal spot size and Rayleigh length and in Sec. IX we use the ion signal to determine the spatial profile of the gas expansion. Finally we conclude in Sec. X.

II. ION DETECTOR

Experiments were performed at the Advanced Laser Light Source (ALLS) at INRS-EMT using a Ti:Sapphire laser (Thales, 100 Hz, 80 mJ, 30 fs), which is described in [17] and [18]. The wavelength of the laser can be shifted with an optical parametric amplifier (Light Conversion TOPAS-HE), broadened in a gas filled hollow fiber and then compressed with either chirped mirrors at $1.4\text{ }\mu\text{m}$ [19] or anomalous dispersion in fused silica glass at $1.8\text{ }\mu\text{m}$ [20]. In the HHG source chamber, gas is emitted from a pulsed gas jet (Parker Series 9, $500\text{ }\mu\text{m}$ orifice) operating at a repetition rate of 50 Hz with a nominal backing pressure of 3 bar. With the jet operating, the source chamber pressure rises to a typical operating pressure of 10^{-4} Torr. The laser is focused a few millimeters below the jet orifice with an estimated gas density on the order of 10^{17} cm^{-3} .

The ion detector consists of a 102 mm diameter stainless steel mesh sandwiched between two fiberglass support rings and is positioned roughly 7 cm downstream from the jet. An illustration of the detector mounted below the pulsed jet is shown in Fig. 1 and the key components are labeled in Fig. 2. The mesh is connected to a high voltage bias and decoupling circuit which is also shown in Fig. 2. The bias is usually on the order of $V_{\text{bias}} = -500\text{ V}$ and is connected to the mesh through bias resistor $R_1 = 1\text{ M}\Omega$. The value of R_1 can be reduced if a lower detector sensitivity is desired. When a pulse of ions reaches the detector the transient signal passes through decoupling capacitor $C_1 = 2\text{ nF}$ and is measured using the high impedance input of a digitizing oscilloscope (Tektronix TDS3054B). The $R_2 = 1\text{ M}\Omega$ resistor to ground prevents the output of the decoupling circuit from floating to V_{bias} when the oscilloscope is not connected.

The oscilloscope is triggered at the same time as the pulsed jet and is set to automatically acquire and average over a predefined number of laser shots (typically 32) before sending the averaged ionization trace to the computer. The response of the system is slower than the actual duration of the ion pulse, typically 150 microseconds. In the sections that follow, we define “ion yield” to be the peak value of the averaged trace. We also compared the peak value with the integrated trace, and found that both measures gave comparable scaling with intensity and pressure.

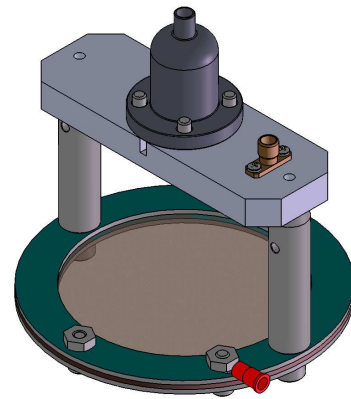


FIG. 1. Illustration of the ion detector mounted below Parker pulsed gas jet. The pulsed valve is the cylindrical structure at the top which emits pulses of gas in the downward direction. The ion detector is a simple wire mesh that is biased at -500 V . The mesh design permits the gas to pass through it without reflecting back to the pulsed valve.

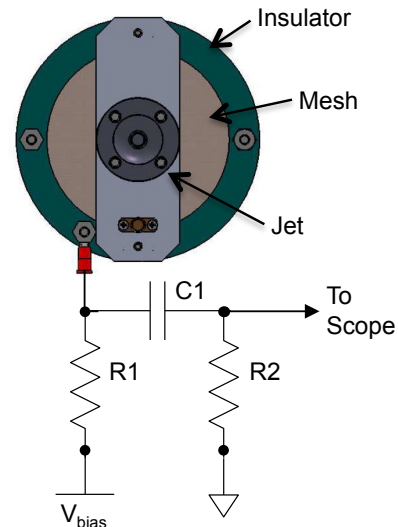


FIG. 2. Overhead view of the ion detector along with the schematic for the bias and decoupling circuit. Typical component values are $V_{\text{bias}} = -500\text{ V}$, $R_1 = 1\text{ M}\Omega$, $R_2 = 1\text{ M}\Omega$ and $C_1 = 2\text{ nF}$. The oscilloscope has a $1\text{ M}\Omega$ input impedance.

III. LINEARITY WITH PRESSURE

In order for this ion detector to provide a useful measure of the total number of ions produced in the gas jet, we must verify that its signal scales linearly with the number of ions produced by the laser over the range of pressures that are relevant to most HHG experiments. We verified this by measuring the dependence of the ion signal on pulsed jet backing pressure, with the laser intensity held constant. The density of atoms under the jet scales linearly with jet backing pressure [21] so varying

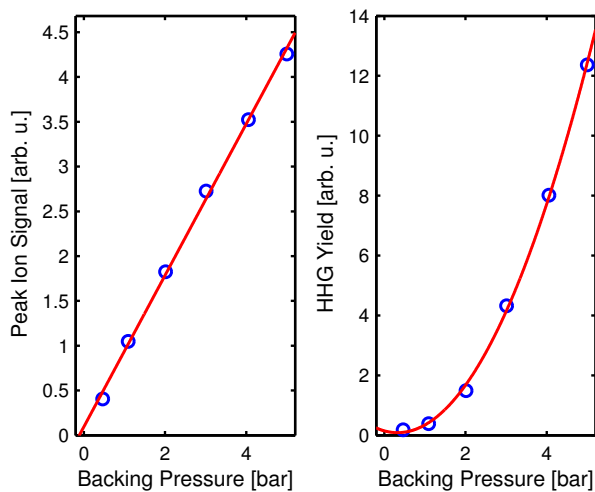


FIG. 3. Measured ion signal from xenon as a function of pulsed valve backing pressure produced with a $1.3\ \mu\text{m}$, 130 fs pulse (left). The ion signal is proportional to the backing pressure, indicating that the ion detector signal is linear with the number of ions in the interaction volume. The harmonic yield (right) scales quadratically with backing pressure, indicating that the harmonic generation process was well phase matched. By dividing the harmonic yield by the square of the ion signal we get the harmonic yield per emitter.

the pressure provides a convenient means of changing the number of ions produced in the focus. The results of this scan are shown in Fig. 3 where we observe a clear linear relation between ion signal and pressure. Because the ion detector does not interfere with HHG or the xuv spectrometer, we are able to record ions and HHG photons simultaneously. The right panel of Fig. 3 shows that the xuv photon yield increases quadratically with jet backing pressure. Quadratic scaling is expected as HHG is a phase matched process in which the emitted electric field of each atom adds coherently, resulting in a quadratic intensity dependence on the number of atoms. By normalizing the harmonic yield to the square of the ion signal we are able to determine the harmonic yield per emitter, which was used for our recent HHG wavelength scaling experiments [17].

When using this type of detector in a given application, it is important to first verify that the detected signal scales linearly with pressure. When producing harmonics using very high backing pressures or with the focus very close to the jet, it is possible to saturate the detector.

IV. INTENSITY CALIBRATION

One of the most useful applications of this detector is for determining the laser intensity at the position where harmonics are produced. One approach for intensity calibration is to measure the spot size of the laser at the focus and to estimate the intensity based on the pulse energy, spot size and pulse duration. In practice this ap-

proach is tedious and not very accurate as it is difficult to accurately measure the spot size, and to account for variations in mode quality. Furthermore, measurements of the pulse energy are sensitive to any long range tail or pedestal that may be present in the pulse. These features may have virtually no effect on strong field processes but if the energy they contribute is erroneously attributed to the main peak of the short pulse they can result in a large error in the estimated peak intensity. We now show that we can accurately determine the laser intensity in the gas jet using the ion detector. Because the measured ion signal scales linearly with the number of ions in the laser focus we can use the ion detector to measure the dependence of the ionization rate on pulse energy. We then apply a single-atom ionization rate model. The fitting procedure described below gives the proportionality factor that relates the laser pulse energy, which is easily measured outside the vacuum chamber, to the intensity in the interaction volume [22].

Accurate models are available for calculating the dependence of the field-ionization rate on laser intensity, for example the ADK model [23]. A more general version was developed by Yudin and Ivanov [24]. This non-adiabatic model has been extensively tested, and it has been shown that it gives compatible results to the ADK theory for intensities that are well described by the tunneling regime (with Keldysh parameter $\gamma \ll 1$) and remains accurate for intensities where there is a substantial multi-photon contribution to ionization ($\gamma > 1$). The validity of the Yudin-Ivanov model for $\gamma > 1$ has been verified experimentally by Uiberacker *et al.* [25] who also compared the model to 3D TDSE results and showed good agreement for $\gamma = 20$ and $\gamma = 1$ (see the supplementary information of Ref. [25]).

The model assumes that the radial profile of the laser beam is Gaussian, and that the gas jet is short in comparison to the confocal parameter. The single-atom ionization rate is temporally integrated over the pulse duration, and spatially integrated over the radial distribution. Calculations for the mode-integrated ionization yield are shown as the solid (blue) curves in the semilogy plots in Fig. 4. This type of plot has a characteristic shape given by an exponential increase in the ionization yield as the intensity becomes high enough to ionize atoms in the most intense part of the focus. As the intensity increases the neutral atoms on axis are depleted and additional ions are produced as the wings of the Gaussian intensity distribution become intense enough to ionize the sample. The intensity dependence of the ionization rate once the on-axis atoms are fully ionized scales as the area of a disk. For our thin jet the intensity dependent ionization yield scales with the square of the spot size (area of a disk). We define the *saturation intensity* as the intensity at which 43% of the sample in the most intense, on axis, part of the focus is ionized. Extrapolating the asymptotic part of Fig. 4 to the horizontal axis gives the saturation intensity [22]. Table I lists the calculated saturation intensities for a range of laser parameters and

λ (nm)	cycles	τ (fs)	$I_{\text{sat}} (10^{14} \text{ W/cm}^2)$				
			Xe	Kr	Ar	Ne	He
800	2	5	1.60	2.85	4.51	14.4	23.0
800	10	27	1.01	1.81	2.86	9.10	14.5
800	20	53	0.86	1.54	2.44	7.75	12.3
1400	2	9	1.52	2.62	4.06	12.5	19.8
1400	10	47	1.03	1.77	2.72	8.28	13.0
1400	20	93	0.89	1.53	2.35	7.13	11.2
1800	2	12	1.45	2.47	3.81	11.6	18.4
1800	10	60	1.01	1.71	2.62	7.90	12.4
1800	20	120	0.88	1.49	2.27	6.82	10.7

TABLE I. Calculated saturation intensities for the noble gases using the Yudin-Ivanov nonadiabatic ionization model [24]. At the saturation intensity (I_{sat}), 43% of the sample on axis is ionized by the end of the pulse.

target atoms.

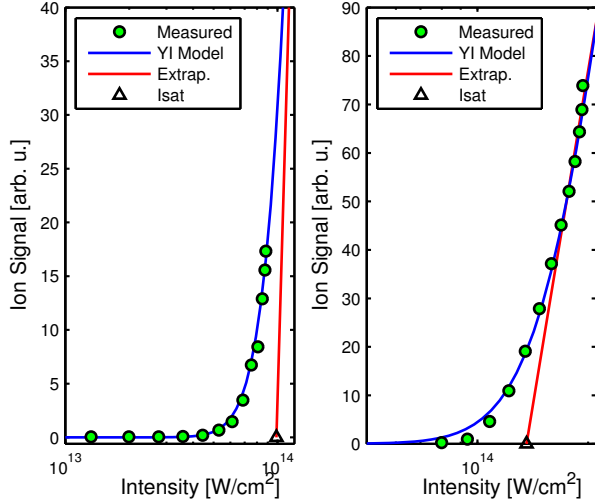


FIG. 4. Measured intensity dependent ion signal in xenon (circles) fit to the mode integrated ionization yields calculated with the Yudin-Ivanov model (blue curves) for a $1.8 \mu\text{m}$ 73 fs pulse (left) and a $1.8 \mu\text{m}$ 11 fs pulse (right). The asymptotic intensity dependence extrapolated to the intensity axis gives the saturation intensities (I_{sat}).

To find the proportionality factor that relates laser intensity to pulse energy we measure the ion signal for a range of pulse energies. We then minimize the square error between theory and experiment by scaling the horizontal (intensity) axis of the measured data and the vertical (ionization yield) axis of the theoretical curve. This fitting procedure determines the absolute intensity of the laser in the interaction volume for a given pulse energy [22]. If the intensity is high enough to strongly saturate the sample, as was the case in the right panel of Fig. 4, the proportionality factor can be estimated with a semilog plot of the ion signal with respect to pulse energy. Extrapolating the asymptotic part of the curve back to the

horizontal axis gives the pulse energy that saturated the sample with the corresponding saturation intensity, I_{sat} , given in Table. I. The proportionality factor between the saturation intensity and energy can then be used to calculate the intensity for all other pulse energies. If the peak intensity was not high enough to strongly saturate the sample such as in the left panel of Fig. 4 the intensity axis can still be calibrated by using the fitting procedure described above. However, care should be taken to ensure that measurements come close enough to saturation in order to uniquely define the curve. In practice this can be checked by repeating the fitting procedure with a number of different initial seed values and ensuring that the fit always converges to the same value.

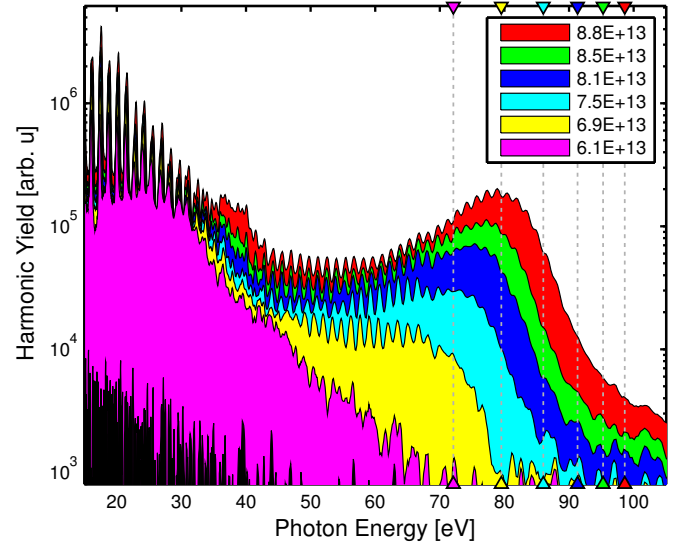


FIG. 5. High harmonic spectra in xenon produced with a $1.82 \mu\text{m}$, 73 fs pulse. The legend shows the intensity for each spectrum that was inferred using the ion detector (cf. Fig. 4). The predicted cutoff energy for each spectrum, based on the inferred laser intensity is shown by the triangles on the horizontal axis.

V. HHG CUTOFF

Since the ion detector does not interfere with the HHG measurement, it is possible to simultaneously record the ion yield and the HHG spectrum. In Fig. 5 we show the HHG spectra that were recorded along with the ionization measurements shown in the left panel of Fig. 4. The peak around 80 eV is associated with the giant resonance in xenon, which we discuss elsewhere [26]. As the laser intensity increases, this peak becomes evident. The intensity calibration described above allows us to estimate the peak laser intensity for each spectrum, which is shown in the figure legend. Based on our calibration of the intensity, we plot the expected cutoff $\Omega_{\text{max}} = I\lambda^2 + I_p$ for

each spectrum with the triangles and vertical lines shown in Fig. 5, using the inferred experimental intensity. Extrapolating the rapid decrease in harmonic yield past the plateau of each spectrum to the horizontal axis gives an estimate of the HHG cutoff.

The intensities inferred from these cutoffs agree to better than 3% with the intensities measured with the ionization method. This excellent agreement tells us that the HHG process for these conditions is almost perfectly phase matched over the entire spectrum. This highly phase matched process is surprising, considering that the highest intensity shown in Fig. 5 (8.8×10^{13} W/cm²) is very close to the saturation intensity of (9.74×10^{13} W/cm²) for the given laser conditions. At this intensity 29% of the atoms along the laser axis are ionized. Such a high degree of ionization causes plasma dispersion that will strongly affect the phase velocity at the $1.8 \mu\text{m}$ laser wavelength. Under these conditions we would expect poor phase matching. The fact that we observe excellent phase matching suggests that another mechanism is acting to compensate for the effect of plasma dispersion.

VI. PULSED JET TIMING

The timing of the pulse jet is chosen so that the laser interacts with the gas when the jet density is the highest. Ideally the jet will produce a gas pulse with very high density while the laser is present and turn off completely at all other times. In practice the ~ 1 ms gas pulses are dramatically longer than the < 100 fs pulses produced by our lasers system. In order to measure the temporal profile of the gas pulses exiting a pulsed jet, dedicated instruments have been developed such as fast ionization detectors [27]. These are often used because the temporal profile of the gas pulse is an important diagnostic for correct operation of the jet. Here we show that we can directly measure the distribution of the gas pulse in time by varying the delay between the laser pulse and the electronic trigger for the pulsed jet controller. This is done with a Stanford Research Digital Delay Generator (DG535) that is controlled with a GPIB interface. In Fig. 6 we plot the measured gas pulse for two different jet configurations. First, we show the pulse from a $\phi = 250 \mu\text{m}$ jet that expands directly into the target chamber, and then, we show the pulse from a $\phi = 500 \mu\text{m}$ jet with a small 14 mm long, $\phi = 350 \mu\text{m}$ diameter tube mounted under the jet. The tube limits the flow of the gas and acts as a reservoir supplying gas for a few milliseconds after the jet solenoid has closed. We clearly see this behavior in the ion signal. Note that these datasets were recorded on different days and with slightly different laser conditions. They have been normalized to give the same peak ion signal.

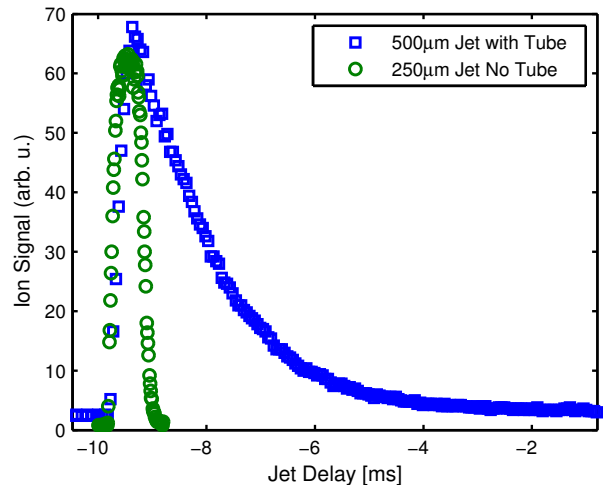


FIG. 6. Ion signal as a function of the delay between the laser pulse and the gas jet trigger for two different jet configurations. The gas pulse from a $250 \mu\text{m}$ jet orifice expanding directly into the target chamber is shown as the green circles and the pulse from a $500 \mu\text{m}$ jet orifice with a small 14 mm long, $350 \mu\text{m}$ diameter tube mounted under the jet is shown in blue. The data is plotted so that the gas density at later times relative to the jet trigger signal are shown on the right side of the graph. Data were normalized so both curves have the same peak value. Conditions were 800 nm, 45 fs, 2-3 bar N_2 backing pressure.

VII. GAS JET POSITION

High harmonic generation is a phase matched process and the position of the jet relative to the laser focus is a key parameter used to control phase matching [28]. It is generally known that placing the gas jet after the laser focus, where the pump beam is diverging, results in good phase matching for the short electron trajectories and poor phase matching for the long trajectories [29]. When the gas jet is moved towards the laser focus, the long trajectories become more collimated and can be measured separately from the short trajectories [30]. It is common for researchers working in this field to infer the relative jet position based on the divergence of HHG radiation. It was recently shown [18, 31, 32], that there are conditions where a low divergence xuv beam can be produced from focal positions where we would normally expect divergent *long trajectory* harmonics. Moving forward, it will be important to have a clear measurement of the position of the jet with respect to the focus.

While it is possible to estimate the jet position relative to the laser focus using a ruler when the vacuum chamber is open to air, we find that a far better approach is to scan the jet through the focus and locate the position where the ionization signal is maximized. This is not necessarily the same as the position at which the HHG signal maximizes. Figure 7 shows the dependence of ion signal on jet position. The position of the gas jet at which the ion signal peaks is where the laser beam

waist is located directly under the gas jet. This method of locating the laser focus position is independent of the HHG process, since it only detects the amount of ionization. Therefore it is independent of any phase matching process. Since the ion signal is proportional to the gas density integrated along the laser axis, Fig. 7 is also a map of the gas density below the gas jet.

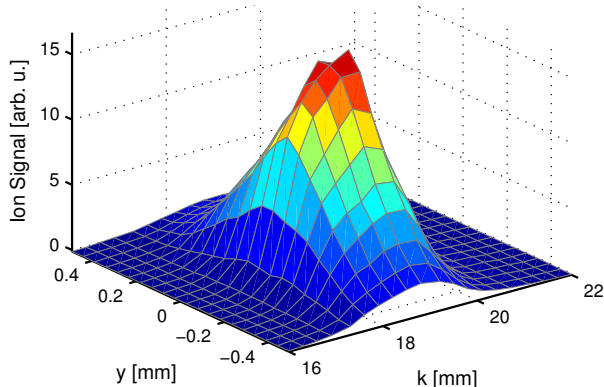


FIG. 7. Ion signal as a function of jet position. The laser focus was held constant, while the gas jet was translated in two dimensions: parallel to the laser propagation direction, k , and perpendicular to both the laser propagation direction and the gas expansion direction y . Conditions were 800 nm, 45 fs, 3 bar N_2 , jet orifice $\phi = 500 \mu\text{m}$.

VIII. FOCAL PARAMETERS

Assuming Gaussian focusing, the laser spot size at the focus is defined by its $1/e$ field radius w_0 [33]. The beam diverges as we move away from the focus with the spot size $w(z)$ increases as:

$$w(z)^2 = w_0^2 \left[1 + \left(\frac{\lambda_0 z}{\pi w_0^2} \right)^2 \right] \quad (1)$$

where λ_0 is the laser wavelength and z is the distance from the focus in the propagation direction. As the spot size increases, the on-axis intensity I_0 decreases as:

$$I_0(z) = I_0(z=0) \frac{w_0^2}{w(z)^2}. \quad (2)$$

By translating our thin jet along z , we measure an ionization signal that is characteristic of the beam divergence and intensity. Starting with a measurement of the laser intensity at the focus using the method described in Sec. IV, we use the Yudin-Ivanov model [24] to calculate the z dependence of the ionization yield as a function of the beam waist w_0 at the focus. We then fit this result to the measured data and find the spot size that gives the best agreement. Figure 8 shows the result of this where we find satisfactory agreement for $w_0 = 64 \mu\text{m}$. Given w_0

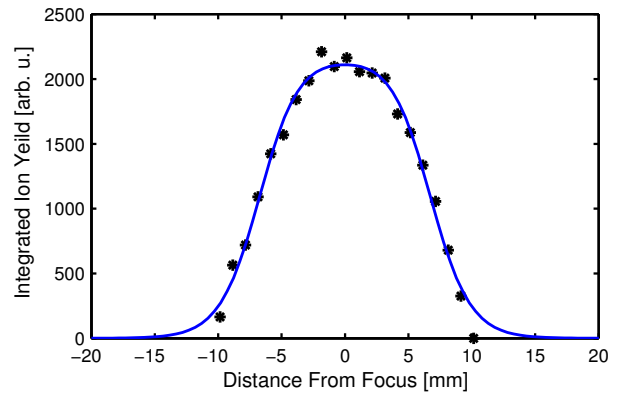


FIG. 8. Measured dependence of ionization yield on jet position in the laser propagation direction (symbols). The solid curve shows the z dependence of the calculated mode integrated ionization yield for a thin sample in a laser field that is focused to a $64 \mu\text{m}$ spot. Conditions were $1.8 \mu\text{m}$, 45fs, $I(z=0) = 2.44 \cdot 10^{14} \text{ W/cm}^2$, xenon.

the Rayleigh length z_0 which is defined as the distance from the focus where the spot size increases to $\sqrt{2} w_0$ is:

$$z_0 = \frac{\pi w_0^2}{\lambda_0}. \quad (3)$$

For our $1.8 \mu\text{m}$ laser with $w_0 = 64 \mu\text{m}$ we find that $z_0 = 7.1 \text{ mm}$. Examining Fig. 8, we see that this distance corresponds to the position at which the ion yield decreases by about a factor of 2. Therefore we can accurately estimate the beam waist and the confocal parameter of the laser focus.

IX. GAS JET DENSITY PROFILE

The ion signal profile shown in Fig. 7 reveals the density profile of the gas jet. The gas is emitted from the gas jet in a direction perpendicular to the orifice, a direction that we call the x -axis. The gas density profile will be cylindrically symmetric about the x -axis, $\rho(r, x)$, where r is the radial coordinate around x . We take the scan in the lateral direction y across the gas jet in Fig. 7, at the position in the focal direction corresponding to the best focus. The experimentally measured ionization signal versus y can be Abel-inverted to give the actual radial density profile. Instead, we model the radial density profile as a Gaussian shape with width r_0 , namely $\rho(r, x) = \rho_0 \exp(-(r/r_0)^2)$. We then integrate the predicted ionization signal along a line of constant y through the gas jet. The experimental signal is compared with the predicted signal, and the fitting parameters ρ_0 and r_0 are adjusted for the best fit, shown in Fig. 9. The fit to the Gaussian profile is sufficiently good that it is not necessary to perform an Abel inversion. The inferred $1/e$ half-width of gas density is 0.3 mm , giving a FWHM of 0.5 mm .

The actual shape of the density profile of the gas jet is important when modeling the HHG process. In most cases the actual profile is not known. Here we show that the profile can be accurately measured. The actual Gaussian profile can be input into HHG propagation codes to predict the phase-matched HHG output.

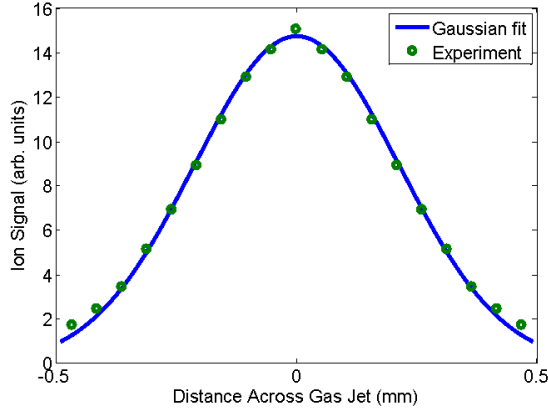


FIG. 9. Measured lateral width of the pulsed valve gas jet density. The gas density is modeled as a Gaussian, $\rho(r, x) = \rho_0 \exp(-(r/r_0)^2)$. Line integrals through the model density profile are taken, yielding the solid line. The circles represent the measured ion signal as the gas jet is traversed across the laser beam at the position of best focus. The fitted $1/e$ half-width of density is 0.3 mm.

X. SUMMARY AND CONCLUSIONS

We have shown that a simple ion detector is a useful diagnostic tool for HHG experiments in which a free gas expansion is used. The measured ion signal scales linearly with the number of ions in the interaction volume and this linearity can be easily verified by measuring the dependence of the ion signal on the jet backing pressure. The ion signal can be used to characterize the temporal profile of the gas pulse from the jet and to locate the position of the jet relative to the laser focus. By measuring the dependence of the ion signal on pulse energy and fitting the data to an ionization model we can determine the laser intensity in the interaction volume. We show that the measured laser intensity in the gas jet and the cutoff of the simultaneously measured HHG spectrum match within 3%. The good match between measured intensity and cutoff suggest that the phase matching of HHG at $1.8 \mu\text{m}$ is better than can be expected, based on conventional phase mismatch arguments.

ACKNOWLEDGEMENTS

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- [1] P. B. Corkum, Phys. Rev. Lett., **71**, 1994 (1993).
 - [2] K. J. Schafer, B. Yang, L. F. DiMauro, and K. C. Kulander, Phys. Rev. Lett., **70**, 1599 (1993).
 - [3] B. Shan and Z. Chang, Phys. Rev. A, **65**, 011804 (2001).
 - [4] E. J. Takahashi, T. Kanai, K. L. Ishikawa, Y. Nabekawa, and K. Midorikawa, Phys. Rev. Lett., **101**, 253901 (2008).
 - [5] M.-C. Chen, P. Arpin, T. Popmintchev, M. Gerrity, B. Zhang, M. Seaberg, D. Popmintchev, M. M. Murnane, and H. C. Kapteyn, Phys. Rev. Lett., **105**, 173901 (2010).
 - [6] M. Drescher, M. Hentschel, R. Kienberger, G. Tempea, C. Spielmann, G. A. Reider, P. B. Corkum, and F. Krausz, Science, **291**, 1923 (2001).
 - [7] P. Salieres, T. Ditmire, K. S. Budil, M. D. Perry, and A. L'Huillier, Journal of Physics B: Atomic, Molecular and Optical Physics, **27**, L217 (1994).
 - [8] Y. Tamaki, J. Itatani, Y. Nagata, M. Obara, and K. Midorikawa, Phys. Rev. Lett., **82**, 1422 (1999).
 - [9] H. J. Wörner, J. B. Bertrand, D. V. Kartashov, P. B. Corkum, and D. M. Villeneuve, Nature, **466**, 604 (2010), ISSN 0028-0836.
 - [10] H. J. Wörner, J. B. Bertrand, B. Fabre, J. Higuier, H. Ruf, A. Dubrouil, S. Patchkovskii, M. Spanner, Y. Mairesse, V. Blanchet, E. Mével, E. Constant, P. B. Corkum, and D. M. Villeneuve, Science, **334**, 208 (2011).
 - [11] F. Rosca-Pruna and M. J. J. Vrakking, Phys. Rev. Lett., **87**, 153902 (2001).
 - [12] H. Stapelfeldt and T. Seideman, Rev. Mod. Phys., **75**, 543 (2003).
 - [13] M. Kaku, K. Masuda, and K. Miyazaki, Japanese Journal of Applied Physics, **43**, L591 (2004).
 - [14] T. Kanai, S. Minemoto, and H. Sakai, Nature, **435**, 470 (2005).
 - [15] R. J. Jones, K. D. Moll, M. J. Thorpe, and J. Ye, Phys. Rev. Lett., **94**, 193201 (2005).
 - [16] J.-P. Brichta, M. C. H. Wong, J. B. Bertrand, H.-C. Bandulet, D. M. Rayner, and V. R. Bhardwaj, Phys. Rev. A, **79**, 033404 (2009).
 - [17] A. D. Shiner, C. Trallero-Herrero, N. Kajumba, H.-C. Bandulet, D. Comtois, F. Légaré, M. Giguère, J.-C. Kieffer, P. B. Corkum, and D. M. Villeneuve, Physical Review Letters, **103**, 073902 (2009).
 - [18] H.-C. Bandulet, D. Comtois, A. D. Shiner, C. Trallero-Herrero, N. Kajumba, T. Ozaki, P. B. Corkum, D. M. Villeneuve, J.-C. Kieffer, and F. Légaré, Journal of Physics B: Atomic, Molecular and Optical Physics, **41**, 245602 (7pp) (2008).
 - [19] M. Giguère, B. E. Schmidt, A. D. Shiner, M.-A. Houle, H. C. Bandulet, G. Tempea, D. M. Villeneuve, J.-C. Kieffer, and F. Légaré, Opt. Lett., **34**, 1894 (2009).
 - [20] B. E. Schmidt, P. Béjot, M. Giguère, A. D. Shiner, C. Trallero-Herrero, É. Bisson, J. Kasparian, J. Wolf, D. M. Villeneuve, J. Kieffer, P. B. Corkum, and F. Légaré, Applied Physics Letters, **96**, 121109 (2010).

- [21] H. W. Liepmann and A. Roshko, *Elements of Gasdynamics* (Wiley, 1957).
- [22] S. M. Hankin, D. M. Villeneuve, P. B. Corkum, and D. M. Rayner, Phys. Rev. A, **64**, 013405 (2001).
- [23] N. B. Delone and V. P. Krainov, Uspekhi Fizicheskikh Nauk, **41**, 469 (1998).
- [24] G. L. Yudin and M. Y. Ivanov, Phys. Rev. A, **64**, 013409 (2001).
- [25] M. Uiberacker, T. Uphues, M. Schultze, A. J. Verhoef, V. Yakovlev, M. F. Kling, J. Rauschenberger, N. M. Kabachnik, H. Schroder, M. Lezius, K. L. Kompa, H.-G. Muller, M. J. J. Vrakking, S. Hendel, U. Kleineberg, U. Heinzmann, M. Drescher, and F. Krausz, Nature, **446**, 627 (2007).
- [26] A. D. Shiner, B. Schmidt, C. Trallero-Herrero, Wörner, S. Patchkovskii, P. B. Corkum, J.-C. Kieffer, F. Légaré, and D. M. Villeneuve, Nature Physics, **7**, 464 (2011).
- [27] W. R. Gentry and C. F. Giese, Review of Scientific Instruments, **49**, 595 (1978).
- [28] P. Salières, A. L’Huillier, and M. Lewenstein, Phys. Rev. Lett., **74**, 3776 (1995).
- [29] P. Balcou, P. Salières, A. L’Huillier, and M. Lewenstein, Phys. Rev. A, **55**, 3204 (1997).
- [30] N. Dudovich, O. Smirnova, J. Levesque, M. Ivanov, D. M. Villeneuve, and P. B. Corkum, Nature Physics, **2**, 781 (2006).
- [31] H. Kim, I. Kim, V. Tosa, C. Kim, J. Park, Y. Lee, A. Bartnik, H. Fiedorowicz, and C. Nam, Selected Topics in Quantum Electronics, IEEE Journal of, **10**, 1329 (2004).
- [32] C. Vozzi, M. Negro, F. Calegari, S. Stagira, K. Kovács, and V. Tosa, New Journal of Physics, **13**, 073003 (2011).
- [33] J. Verdeyen, *Laser Electronics*, 2nd ed. (Prentice-Hall, London, 1989).

Chapter 5

Phase Matched HHG and Spatial Filtering

This chapter is based on the following paper:

1. H.-C. Bandulet, D. Comtois, **A. D. Shiner**, C. Trallero-Herrero, N. Kajumba, T. Ozaki, P. B. Corkum, D. M. Villeneuve, J.-C. Kieffer, and F. Légaré, “High harmonic generation with a spatially filtered optical parametric amplifier,” *Journal of Physics B: Atomic, Molecular and Optical Physics*, vol. 41, no. 24, p. 245602 (7pp), 2008

One of the key challenges in the wavelength scaling experiments of Chapter 3 was finding a way to maintain the same focal volume and mode quality for all wavelengths. This became particularly important when producing harmonics at infrared wavelengths using the output of our OPA, whose mode quality at the time would be well described as ‘*terrible*’. Our solution was to spatially filter the OPA beam by passing it through a hollow core fiber, and then to re-image the output of the fiber into the gas target in the HHG experiment. We were surprised to find that even when controlling for intensity and focal volume, the spatially filtered beam produced nearly one order of magnitude greater harmonic yield on-axis, compared to the unfiltered case. This chapter begins with a very brief introduction to HHG phase matching mechanisms. It goes on to show the result of free-space HHG propagation calculations, which help to explain the origin of the enhancement. It concludes with our publication on the benefit of spatial filtering before harmonic generation with OPA-based laser sources.

5.1 Properties of HHG Emission

The Strong Field Approximation (SFA), introduced in Ch. 2, can be used to calculate the induced dipole that is responsible for high harmonic emission from a single atom in a laser field. In practice, harmonics are produced by focusing a laser into a gas target where billions of atoms all participate in HHG. The development of the SFA showed that harmonic emission is intrinsically synchronized with the laser field at the position and moment in time when it takes place, and thus the contribution from the atoms in the ensemble adds coherently. Conditions are usually chosen so that the emission on-axis adds constructively and remains in phase with the laser field as the fundamental and XUV beams propagate through the sample. To the extent that this is successful, harmonics radiation produced as the fundamental beam reaches the end of the sample will be in phase with harmonic radiation that was produced earlier. Under ideal phase matching conditions the electric fields add coherently and the XUV power scales with the square of the number of atoms in the focal volume.

5.2 Phase Matching of HHG

The coupling of energy between the fundamental and harmonic fields is described by the following wave equations [76]:

$$\Delta_{\perp} \mathbf{A}_1(\mathbf{r}, t) + 2ik_1(\mathbf{r}, t) \frac{\partial \mathbf{A}_1(\mathbf{r}, t)}{\partial z} = 0 \quad (5.2.1)$$

$$\Delta_{\perp} \mathbf{A}_q(\mathbf{r}, t) + 2ik_q(\mathbf{r}, t) \frac{\partial \mathbf{A}_q(\mathbf{r}, t)}{\partial z} = -\frac{q^2 \omega_1^2}{\epsilon_0 c^2} P_q^{NL}(\mathbf{r}, t) e^{i(qk_1 - kq)z} \quad (5.2.2)$$

where $\mathbf{A}_1(\mathbf{r}, t)$ and $\mathbf{A}_q(\mathbf{r}, t)$ are the slowly evolving envelope functions for the fundamental and q^{th} harmonic fields respectively. P_q^{NL} is the nonlinear polarization of the medium, which is related to the induced dipole $\mathbf{D}_q(\mathbf{r}, t)$ by $P_q^{NL} = n_a(\mathbf{r}, t) \mathbf{D}_q(\mathbf{r}, t)$, where n_a is the atomic density. In this treatment, the SFA is used to calculate the induced dipole $\mathbf{D}_q(\mathbf{r}, t)$ at a given point in space and time starting from the description of the laser electric field (Eq. 5.2.1). The second time derivative of the polarization acts as the nonlinear source term, coupling energy from the fundamental field into the q^{th} harmonic field. The wavevectors $\mathbf{k}_1$ and $\mathbf{k}_q$ define the propagation of the fundamental and harmonic phase fronts. For energy to transfer efficiently from the fundamental to the harmonic field, the induced dipole at a given position must be in

phase with the harmonic radiation reaching that position that was generated at an earlier time.

For loose focusing and low order processes, such as second harmonic generation (see section 6.2) ($q=2$), the phase of the induced dipole is constant, relative to the phase of the fundamental, and the phase matching condition simply requires the fundamental and harmonic beams to travel with the same phase velocity, $\Delta\mathbf{k} = q\mathbf{k}_1 - \mathbf{k}_q \approx 0$. Several factors can contribute to phase mismatch, such as the wavelength dependent index of refraction of the gas sample $\Delta\mathbf{k}_{q,atom}$, or plasma from the ionizing medium $\Delta\mathbf{k}_{q,el}$. The fundamental and XUV beams each experience phase shifts due to diffraction as they propagate, the difference of which leads to geometric phase mismatch $\Delta\mathbf{k}_{q,geo}$. Finally, in HHG the phase between the fundamental field and the induced dipole is not constant, leading to an intensity dependent dipole phase $\Phi_{q,at}(z, r)$ that varies throughout the focal volume. Balancing all of these factors leads to the following generalized phase matching condition for Eq. 5.2.2 [76]:

$$\Delta\mathbf{k} = (q\mathbf{k}_1^0 - \mathbf{k}_q^0) + \Delta\mathbf{k}_{q,atom} + \Delta\mathbf{k}_{q,el} + \Delta\mathbf{k}_{q,geo} + \nabla\Phi_{q,at} \approx 0 \quad (5.2.3)$$

Where $|\mathbf{k}_q^0| = q|\mathbf{k}_1^0| = 2\pi q/\lambda_1$. The contributions to phase matching are briefly discussed in the following sections.

5.2.1 Atomic Dispersion

The neutral gas medium has a wavelength dependent refractive index and this will, to some extent, affect the phase velocities of the XUV and fundamental beams, resulting in atomic phase mismatch $\Delta\mathbf{k}_{q,atom}$. This effect is usually small compared to other sources of phase mismatch.

5.2.2 Electronic Dispersion

Harmonic generation is necessarily associated with ionization of the medium. As the fundamental and XUV beams propagate through the ionizing medium, there is a large phase shift between the fundamental and XUV beam as plasma is highly dispersive at laser wavelengths and has very low dispersion in the XUV [77]. The phase mismatch

due to free electrons is given by [78]:

$$\Delta k_{q,el}(r, z, t) \approx \frac{e^2 n_e(r, z, t)}{4 \pi \epsilon_0 m_e c^2} q \lambda_1 \quad (5.2.4)$$

where $n_e(r, z, t)$ is the electron density, e is the electron charge, and m_e is the electron mass. Free electron dispersion is one of the most important sources of phase mismatch in HHG. This is particularly true when producing harmonics with IR sources because plasma dispersion is proportional to the laser wavelength.

5.2.3 Geometric Phase

As a laser beam focuses, its phase shifts in both the radial and longitudinal directions. For low order processes, where the material response is assumed to be instantaneous, the induced dipole phase will be q times that of the fundamental [79], resulting in a harmonic beam with the same confocal parameter as the fundamental [80]. This confocal parameter is associated with an effective beam waist that is $q^{1/2}$ times smaller than that of the fundamental and far-field divergence $\theta_{ff} = \lambda/(\pi w_0)$ that is $q^{1/2}$ times smaller. For a Gaussian beam propagating in the positive $\hat{z}$ direction with a focus located at $z=0$, the geometric phase is given by [80]:

$$\Phi_{\text{geo},q}(r, z) = -q \arctan\left(\frac{2z}{b}\right) + \frac{2qz}{b} \left(\frac{r}{w(z)}\right)^2 \quad (5.2.5)$$

where $w(z) = w_0 \sqrt{1 + 4z^2/b^2}$ is the spot size and w_0 is the beam waist, which is related to the confocal parameter by $b = 2\pi w_0^2/\lambda$. The first term describes the Gouy phase shift, which is the backward shift by π radians that a pulse phasefront experiences as it passes through a focus. A plot of the geometric phase on-axis ($r=0$), is shown in Fig. 5.1. The second term describes the curvature of the phase front, which goes from curving in toward the focus at positions where z is negative, to flat at the focus, and then curving back toward the focus as the beam diverges and z becomes positive. This radial phase dependence will be discussed in subsequent sections.

The wavevector associated with the geometric phase shift is $\mathbf{k}_{q,\text{geo}} = \nabla \Phi(r, z)_q$ which always points backward on-axis as the slope of the Gouy phase shift is negative. The harmonic field also diffracts and experiences its own geometric phase shift, which

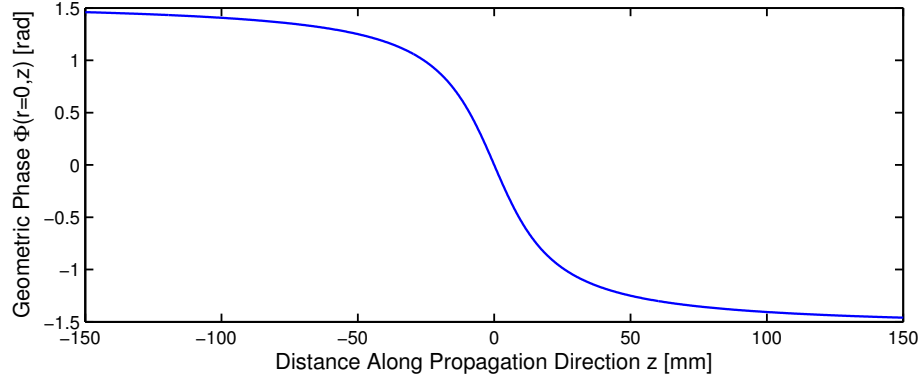


Figure 5.1: Gouy phase shift: $\Phi_{\text{foc},q}(r = 0, z)$

has a similar order of magnitude to that of the fundamental. Because the induced dipole phase shift is q times that of the fundamental it will recede q times faster than the fundamental leading to a wave vector mismatch of:

$$\Delta \mathbf{k}_{\text{geo}} = q \delta \mathbf{k}_{1,\text{geo}} - \delta \mathbf{k}_{q,\text{geo}} \approx q \delta \mathbf{k}_{1,\text{geo}} \approx -2q/b \hat{z} \quad (5.2.6)$$

The consequence of this is that without some other mechanism to compensate for the geometric phase shift, such as dispersion or the intensity dependent dipole phase, it would not be possible to efficiently phase match in a tightly focused geometry [81].

5.2.4 Dipole Phase

In the strong field regime the phase relation between the induced dipole moment and the fundamental field is not constant. The phase of the induced dipole depends on laser intensity, with the result that the laser intensity distribution throughout the focal volume plays a key roll in phase matching. Section 2.4 showed that there are specific electron trajectories associated with the generation of a given harmonic. The intrinsic dipole phase depends on the classical action $S(t_i, t_e)$ and is given by:

$$\Phi_{\text{at},q} = q \omega_0 t_e - S(t_i, t_e) \quad (5.2.7)$$

where the combination of birth time t_i and return time t_e correspond to an allowed trajectory for the generation of harmonic q (see Sec. 2.4)[82]. For a given quantum path, the action is approximately given by $-U_p \cdot (t_e - t_i)$, where the ponderomotive

potential U_p is proportional to the laser intensity. The emission time is nearly constant in the cutoff region and switches abruptly to another, nearly constant, value in the plateau region of the harmonic spectrum [82]. The result of this is that the intrinsic dipole phase is piecewise linear in intensity (see Fig. 5.2). Expressed in terms of

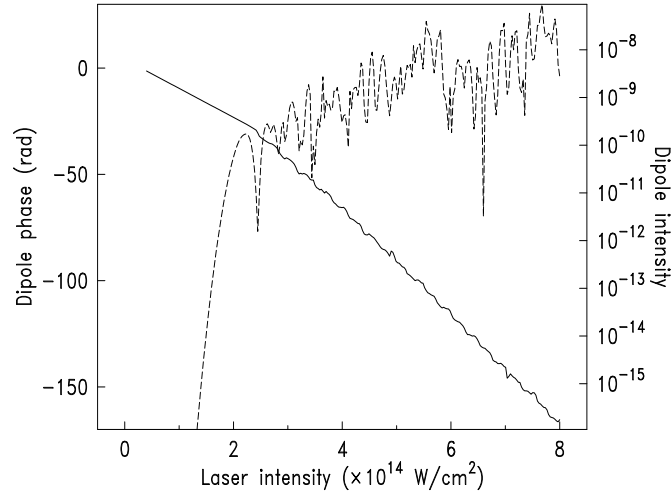


Figure 5.2: Dependence of the 45th harmonic of neon intensity (dashed line) and phase (solid line) on laser intensity. (Figure from: [82])

laser intensity, the induced dipole phase is $\Phi_q \approx -\theta I$. For harmonics produced with 800nm radiation, the slope θ is given by $\theta_{\text{cutoff}} \approx 13.7 \cdot 10^{-14} \text{ rad cm}^2/\text{W}$ in the cutoff region and in the plateau is given by $\theta_{\text{long}} \approx 25.7 \cdot 10^{-14} \text{ rad cm}^2/\text{W}$ and $\theta_{\text{short}} \approx 1.4 \cdot 10^{-14} \text{ rad cm}^2/\text{W}$ for the long and short trajectories respectively[76].

5.2.5 Focal Position for Optimal Phase Matching

Optimum phase matching on-axis is achieved when the phase mismatch between the fundamental and XUV fields is minimized. In many cases high harmonics are produced in a thin gas medium, and the freedom to position that medium relative to the laser focus provides an important parameter that is used to optimize phase matching. As a thin gas jet is translated along the laser propagation direction through a focus, the phase of the fundamental wavefronts vary in both the radial and transverse directions. This phase, multiplied by the harmonic order, is added to the phase of harmonic emission. The wave vector for the fundamental field traveling primarily in

the $\hat{z}$ direction is given by $\mathbf{k}_1(r, z) = k_1^0 \hat{z} + \nabla \Phi_{\text{geo}}(r, z)$, where the phase $\Phi_{\text{geo}}(r, z)$ is defined in Sec. 5.2.3. A plot of the laser wavevector distribution $\mathbf{k}_1(r, z)$ is shown in the left panel of Fig. 5.3 [82]. As the beam approaches the focus the wave vectors tilt in toward the propagation axis. As the beam passes the focus they tilt away. At the focus, as well as at relatively large distances on either side of the focus, the wavefronts are nearly flat and the vector field points parallel to the axis. Changing the position

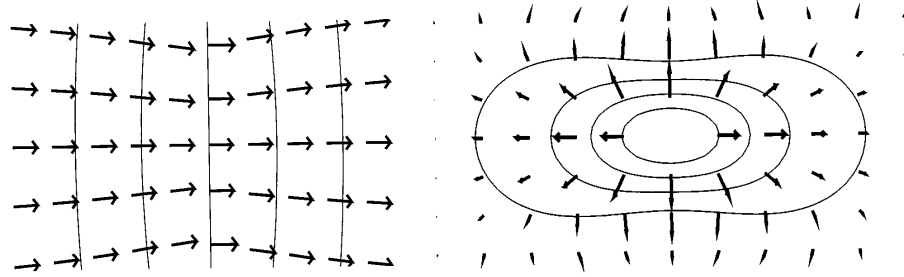


Figure 5.3: (Left) Wavevector distribution ($k_0 \hat{z} + \mathbf{k}_{\text{geo}}$) for a focusing laser beam. The principle propagation direction is along $+\hat{z}$ axis (drawn horizontally), the geometric phase shift near the focus points in the $-\hat{z}$ direction on-axis and tilts toward the focus at small z . (Right) The atomic wave vector points along the intensity gradient and is always directed away from the focus in the radial and longitudinal directions. (Figure from: [82])

of the generating medium allows for control over the geometric phase of the induced dipole, whose phase will include an offset that is q times that of the fundamental field. In addition to the geometric phase, Sec. 5.2.4 showed that the dipole phase depends on the laser intensity, which is associated with the atomic phase term $\Phi_{\text{at}}(\mathbf{r}, z)$. The laser intensity gradient always slopes away from a focus, causing the atomic wave vector $\mathbf{k}_{q,\text{at}}$ to radiate outward from the focus. The right panel of Fig. 5.3 shows an example of the vector field for $\mathbf{k}_{q,\text{at}}$. The different (and in some cases opposite), behaviour of the atomic and geometric phase terms allows the experimentalist to balance the contributions to phase mismatch (Eq. 5.2.3) by selecting an optimal jet position.

The example that follows neglects the role of atomic and electronic dispersion which could be included by adding their corresponding wave vectors. When the jet is located at the focus ($z=0$), the negative geometric phase shift from the Gouy phase leads to a net phase mismatch $\Delta \mathbf{k}$ preventing phase matching on-axis as shown in Fig. 5.4(a). When the jet is positioned after the focus (positive z), the intensity gradient moving along the propagation direction causes the induced dipole phase

wave vector $\nabla\Phi_{at}$ to point in the positive $\hat{z}$ direction, compensating for the geometric phase shift and enabling efficient phase matching on-axis Fig. 5.4(c). When the jet is located before the focus, the geometric phase shift continues to point in the negative z direction as does the on-axis intensity gradient. The geometric and dipole phase shifts do not cancel, and phase matching on-axis is not achieved. Moving off-axis in the radial direction, the intensity gradient points along the radial direction and facilitates phase matching off-axis as shown in Fig. 5.4(d). For off-axis phase matching, the XUV beam emerges as a series of annular rings centered around the propagation axis. Figure. 5.5 shows an example of phase matching with the jet located before or

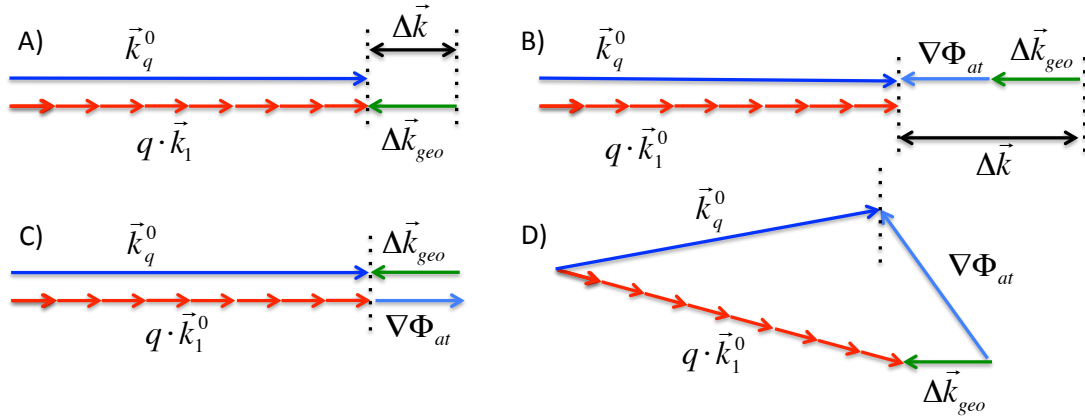


Figure 5.4: Phase matching diagrams for HHG with the gas jet located at various positions. A) On-axis with the jet located at the focus. B) On-axis with the jet before the focus. C) On-axis with the jet after the focus. D) Off-axis phase with the jet before the focus. (Figure adapted from: [82])

after the focus[76]. All results presented in this thesis are for conditions where the jet was positioned for optimum phase matching on-axis, and a collimated beam of XUV radiation is observed in the far-field of the spectrometer.

5.3 Modeling Free Space Propagation

Within the paraxial approximation, a forward propagating beam is fully described by its amplitude and phase at one position along the propagation axis ($z=0$). Once the complex field distribution or *wavefront* $E(x, y; z = 0, \omega)$ is known the wavefront at other positions along z (or equivalently in time), is easily calculated by Fourier trans-

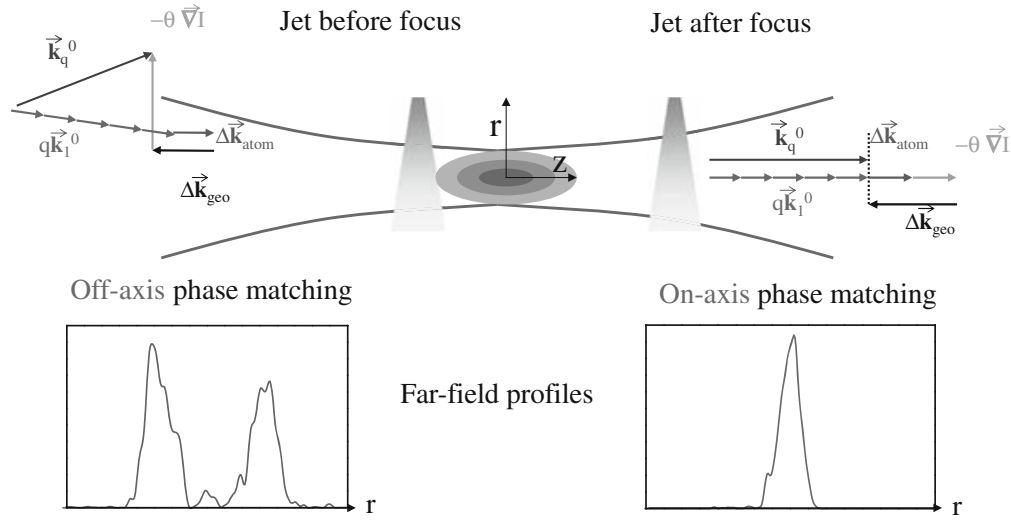


Figure 5.5: Illustration of typical phase-matching behaviour for HHG. Normally the jet is positioned **after** the focus to phase-match short trajectories. (Figure from: [76].)

forming the wavefront from position space to momentum space, and then applying the free-space propagator from Fourier optics [83, 84, 85]:

$$\mathcal{H}_e(k_x, k_y; z, \omega) = \frac{\tilde{E}(k_x, k_y; z, \omega)}{\tilde{E}(k_x, k_y; z=0, \omega)} = \exp\left\{\frac{i(k_x^2 + k_y^2)z}{2k_0}\right\} \exp(-ik_0 z) \quad (5.3.1)$$

where,

$$\tilde{E}(k_x, k_y; z, \omega) = \mathcal{F}_{x,y}\{E(x, y; z, \omega)\} \quad (5.3.2)$$

and $\mathcal{F}_{x,y}$ is the two dimensional spatial Fourier transform. The development of this propagator used the paraxial approximation, which requires the beam to primarily propagate in the positive $\hat{z}$ direction ($k_z^2 \gg k_x^2 + k_y^2$). Note that this propagator is defined for a given optical frequency component ω . To propagate a pulse, the electric field is first Fourier transformed from the time domain to the optical frequency domain $E(x, y; z=0, t) \Rightarrow E(x, y; z=0, \omega)$, and then each optical frequency component is propagated separately.

Figure 5.6 shows an example of propagating a continuous wave beam, with a wavelength of $1.8 \mu\text{m}$, from an initial focus at $z=0$ to a distance of 30 cm. The example shows the far-field spot size for the case of an initial beam waist of $\omega_0 = 10 \mu\text{m}$

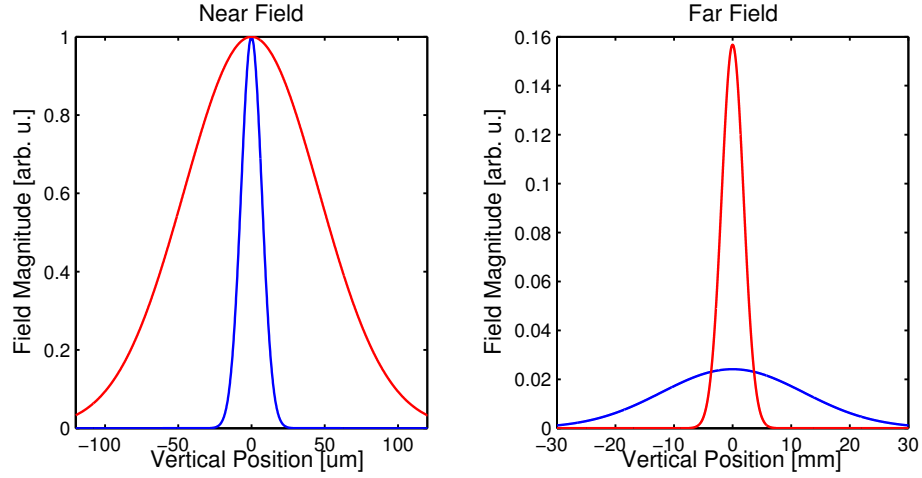


Figure 5.6: Free-space propagation of a $1.8\ \mu\text{m}$ laser beam from a focus at $z = 0$ (left panel), to a position 30 cm after the focus (right panel). The blue curves show the profile of a beam that starts with a $10\ \mu\text{m}$ focal spot, and the red curves correspond to a beam with an initial spot size of $65\ \mu\text{m}$. The smaller spot size at the focus diverges more rapidly.

(blue), as well as $\omega_0 = 65\ \mu\text{m}$ (red). As expected small features at the focus are associated with a more rapidly diverging beam. In the simulations that are described in the next section the free-space propagator is used for both the fundamental and XUV fields. The fundamental starts from an initial focus and is propagated to a position a few millimeters after the focus, corresponding to the ‘jet’ position. The SFA is then evaluated for each spatial grid point in the x-y plane, and the XUV emission at each point is added to the XUV field at that point. The total XUV field is then propagated into the far-field. This simulation assumes propagation in homogeneous media. A spatially varying index of refraction could be introduced into this model by taking small steps along the $\hat{z}$ direction and updating the propagation constant k_0 , which could be made to include both spatial and frequency dependence. This procedure is very similar to the split operator method which is used in Ch. 6 to model spectral broadening of the fundamental laser beam due to self-phase modulation.

5.4 Wavefront Curvature

High harmonic generation in a macroscopic sample was simulated starting with a $1.8\,\mu\text{m}$, 15 fs laser pulse that is initially at a focus ($z=0$) with a spot size of $w_0 = 65\,\mu\text{m}$. Each frequency domain Fourier component of the field was propagated forward using Eq. 5.3.1 in order to calculate the field distribution 5 mm after the focus. This is the ‘jet’ position for optimal phase matching in the simulation. The laser intensity and phase at $z = 5\text{ mm}$ is shown in Fig. 5.7 (a), where the quadratic phase front indicates that the beam is diverging. If the intensity-dependent dipole phase was constant the harmonic phase would be q times that of the fundamental, resulting in a harmonic wavefront that is also diverging with the same confocal parameter as the fundamental. The intensity-dependent dipole phase reduces the curvature of the XUV wavefront, and significantly increases the collimation of the XUV beam [80]. In this simulation the field is modeled in one transverse dimension ($\hat{y}$) and propagated in the $\hat{z}$ dimension. Each of the $N_y = 2^{12}$ spatial grid points along the $\hat{y}$ direction contains $N_t = 2^{13}$ time/frequency points. The numerical model is capable of propagating a two-dimensional field, but for time and memory considerations the present example is limited to simulating harmonic generation from a one-dimension string of emitters. This model also assumes that the index of refraction is the same for the fundamental and XUV fields and that it is not evolving with time. To model harmonic generation, the SFA was evaluated at each spatial grid point along the $\hat{y}$ direction and used to determine the amplitude and phase of the HHG emission at the jet position. The amplitude and phase of the HHG field of the 163rd harmonic of $1.8\,\mu\text{m}$ (112.5 eV), is shown in Fig. 5.7(b). The wavefront is curving outward with a large radius of curvature near the axis indicating a well-collimated beam. The harmonic field at the jet position was propagated into the far-field ($z = 300\text{ mm}$), using the free-space propagator (Eq. 5.3.1). The resulting spectrum, shown in Fig. 5.8 (top), consists of a series of well-collimated harmonics in the plateau region. So far the discussion in this chapter has focused on phase matching for high harmonics produced with ideal Gaussian laser beams. The disastrous effect that a small amount of intensity noise has on harmonic generation is discussed in the next section.

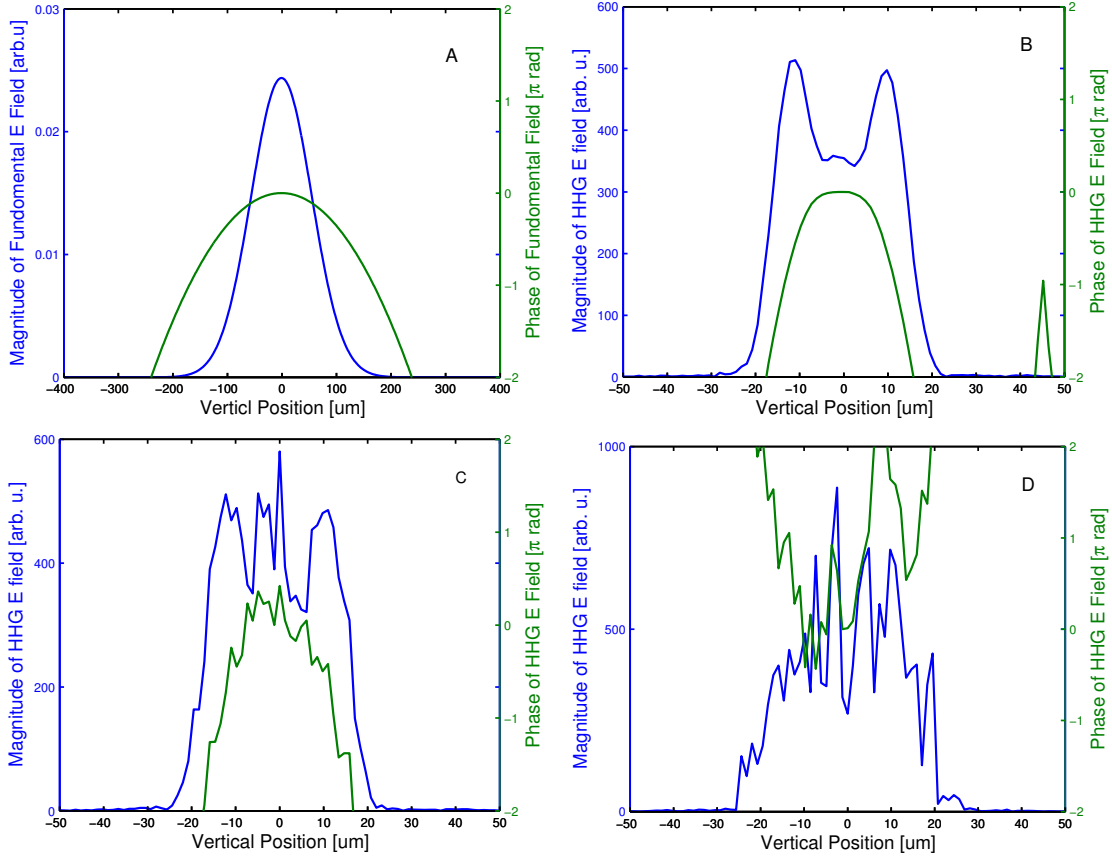


Figure 5.7: Near-field plots: **A)** Fundamental laser field amplitude (blue), and phase (green), 5 mm after the laser focus. **(B-D)** Amplitude (blue), and phase (green), of the wavefront of the 163rd harmonic of $1.8 \mu\text{m}$ (112.5 eV), generated with the medium located 5 mm after the laser focus. Panels B through D show the effect adding 0%, 1%, and 5% amplitude noise to the laser electric field.

5.5 HHG Sensitivity to Mode Quality

In high-harmonic generation the phase of the XUV emission depends linearly on the laser intensity. If the mode quality in the laser focus is poor, the erratic variation in intensity across the focus maps onto the spatial phase of the XUV wavefront. This can lead to destructive interference when harmonic emission from atoms across the focus combines in the far-field. Harmonic generation with long wavelength sources is more sensitive to this as the continuum electron travels further from the parent ion before recombining. Figure 5.7(b) shows the amplitude and phase of the XUV wavefront that was calculated by placing a string of emitters 5 mm after the focus of

an ideal $1.8\ \mu\text{m}$, 15 fs beam with a spot size of $w_0 = 65\ \mu\text{m}$. The phase of the harmonic emission follows a smooth parabolic contour leading to well collimated harmonics in the far-field Fig. 5.8 (top). The effect of poor mode quality is modeled by scaling the fundamental field in the $\hat{y}$ direction, with normally distributed amplitude noise. The noise has a standard deviation (1σ), equal to one percent of the noise-free field amplitude at that grid point. Note that 1% spatial noise on the electric field is equivalent to 0.01% intensity noise. The near-field profile Fig. 5.7(c) shows that the noise on the laser electric field had some effect on the harmonic field distribution, but most importantly caused phase fluctuations which are $> \pi/4$ from point to point across the near-field XUV wavefront. In the far-field these variations scatter energy off-axis reducing harmonic yield, particularly on the high energy side of the spectrum. The result is even more dramatic when 5% field (0.25% intensity), noise is added to the fundamental beam. The near and far-field results are shown in Fig. 5.7(d) and Fig. 5.8 (bottom), respectively. Five percent field noise destroyed phase matching on the high energy side of the spectrum. Figure 5.9 shows the harmonic yield on-axis (far-field), for varying levels of spatial noise on the fundamental laser beam. Clearly, mode quality plays a critical role in phase matching for high energy harmonics.

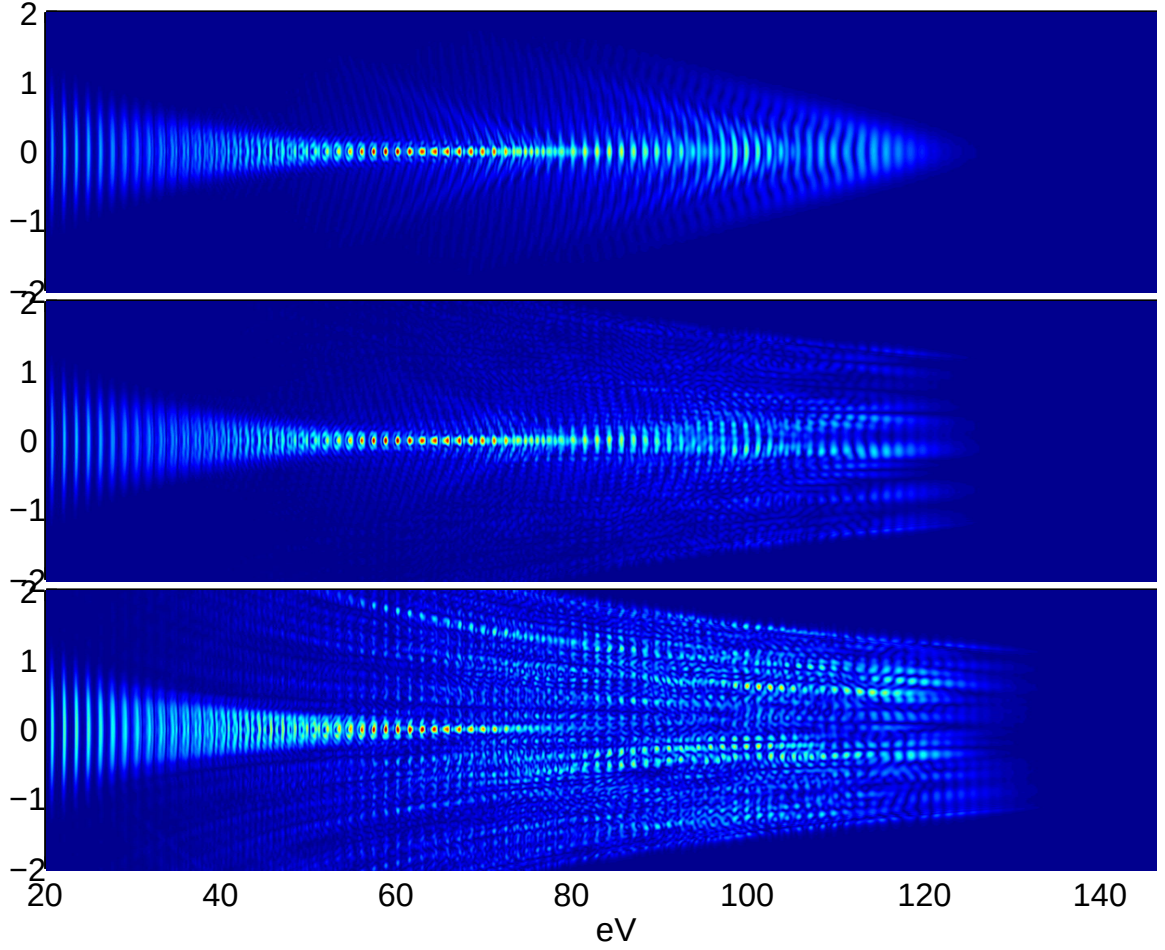


Figure 5.8: Far-field ($z=3\text{-D0 cm}$) HHG spectrum for harmonics produced from a plane of emitters located 5 mm after the laser focus. (top) For a high quality spatial mode the far-field spectrum consists of well-collimated harmonics. (middle) Far-field spectrum from harmonics produced from a laser mode that was modulated with 1% spatial noise at the jet position. (bottom) Far-field spectrum with 5% noise modulating the laser field at the jet position.

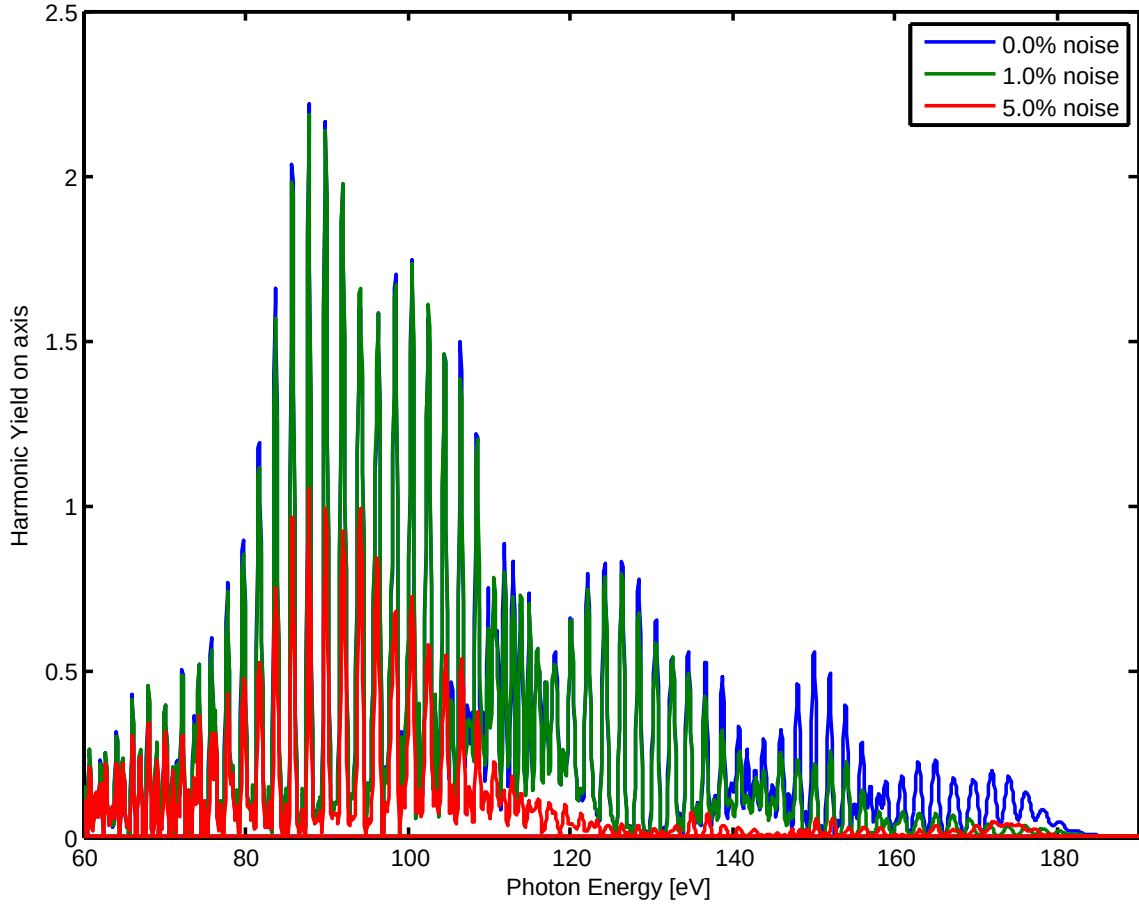


Figure 5.9: Harmonic yield on-axis in the far-field (30 cm) after the laser focus. Plot is for the same data shown in Fig. 5.8. The legend indicates the noise that was added to the laser electric field at the focus, expressed as a percentage of the peak field amplitude. Five percent field noise (0.25% intensity noise) destroys phase matching on the high energy side of the spectrum which is consistent with the experimental result shown in Fig. 5 of the paper that starts on page 71

5.6 What does that iris really do?

In the paper that follows we describe re-imaging the output from a hollow core fiber into the HHG target chamber. The beam profile after the hollow core fiber follows a Bessel intensity distribution [86], which we truncated at the first zero of the Bessel function immediately before the beam was focused into the target chamber. It is well known in the high harmonic community that harmonic generation often improves when an iris is installed at the entrance of an HHG target chamber. This has led to a number of theoretical investigations [78, 87]. Nisoli et al. [87] showed that a tightly focused Bessel beam can produce a periodic intensity modulation along the propagation direction as the fundamental beam moves through a focus, leading to a form of quasi phase matching. C. Jin and C.D. Lin [78] recently explored the experimental conditions that we used for our xenon paper [5] (Chapter 10), and found that for our focusing conditions Bessel and Gaussian modes produced equivalent results.

Most theoretical work starts with an idealized fundamental laser beam with either a Gaussian or Bessel mode. The previous section suggests that one of the most important effects of using an iris to clip a laser beam prior to focusing it into an HHG target chamber is that it reduces high spatial frequency noise in the focus. Section 5.5 demonstrated that 0.25% intensity noise was sufficient to completely destroy phase matching on the high energy side of the spectrum. This was modeled by starting with a diffraction-limited beam and propagating it to a ‘jet position’ located 5 mm after the focus. Noise was added to the field amplitude at each grid point along $\hat{y}$. From diffraction theory we know that the smallest features that can be realized at a focus is limited by the numerical aperture of the beam coming into that focus. An example of this was shown in Fig. 5.6 where a small focal spot is associated with a large spot size in the far-field. The only way to have modulations inside the diffraction limited focal spot of a laser beam is to have light coming into the focus at larger angles compared to the main laser mode. This won’t be news to people who are used to working with optics, but what might be surprising is how little it takes to seriously impact HHG. In addition to poor mode quality originating inside an OPA, it is common for the edges of a beam to slightly overfill the optics used to bring the beam to the target chamber, resulting in diffraction. The diffracted light spreads more rapidly than the fundamental mode, and may reach the spectrometer focusing optic significantly

shifted from the fundamental. If this light crosses the focus it can adversely affect harmonic yield despite having relatively low intensity. When an iris is put in the beam and arranged to slightly clip the fundamental laser mode before focusing, it limits the highest spatial frequency at the focus to that of the fundamental mode, insuring a regular intensity distribution at the focus.

High harmonic generation with a spatially filtered optical parametric amplifier

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Abstract

Numerous applications of high harmonic generation (HHG), such as attosecond pulse synthesis, depend on the ability to increase the electron recollision energy, which is a quadratic function of the driver wavelength. High-energy infrared pulses obtained from an optical parametric amplifier (OPA) are thus attractive for driving the HHG process, thereby offering the opportunity to yield shorter attosecond pulses. However, the increase in driver wavelength is often outweighed by the poor spatial quality of the OPA source. In this paper, we demonstrate that HHG using OPA signal pulses is significantly improved by spatial filtering in a hollow-core fibre prior to focusing in the gas target in comparison with the unfiltered case. Ion yield measurements in combination with beam profile monitoring in the far field enabled control over the interaction volume. For similar interaction volumes, we observe that with less than half the energy per pulse, the HHG yield can increase by one order of magnitude with spatial filtering. The comparison between the harmonic yields in argon and krypton, and their respective dependence on the peak laser intensity, provide experimental evidence that strongly suggests that the enhancement is due to improved phase matching.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

High harmonic generation (HHG) from atoms driven by intense femtosecond lasers is recognized as the key approach for the production of coherent XUV pulses of attosecond duration [1, 2]. During the last decade, there have been considerable advances in the technology to generate and to characterize attosecond laser pulses [3–6]. Currently, the shortest duration for an isolated attosecond pulse is 80 as [7]. Since the minimum pulse duration is limited by the spectral bandwidth, higher-order harmonics are necessary to further decrease the duration of attosecond pulses [8].

According to the three step model for HHG [9], the cutoff energy of the harmonic spectrum is given by $I_p + 3.17U_p$, where I_p is the ionization potential of the atom and $U_p \propto I\lambda^2$ is the laser ponderomotive energy, which is proportional to the laser intensity I and the square of its wavelength λ . Since the

laser intensity needs to be kept low to avoid depletion of neutral atoms [2], one therefore needs to increase the wavelength of the driving laser field into the infrared spectral region in order to extend the cutoff energy, as well as to produce harmonics in noble gas that have preferably large ionization potentials, such as neon and helium.

High-energy infrared pulses, such as those obtained from an optical parametric amplifier (OPA) seeded by a Ti:sapphire laser, are thus attractive for driving the HHG process [10–13]. The extended cutoff that can be obtained using such an infrared driver laser will thereby offer the opportunity to generate shorter attosecond pulses and at the same time extend their applicability as short-wavelength XUV sources, provided that its degree of coherence is high. On the other hand, the increase of the driver wavelength inherently leads to a drop in the conversion efficiency of the HHG process—expected to scale as $\propto \lambda^{-5}$ for the single-atom response [14]—

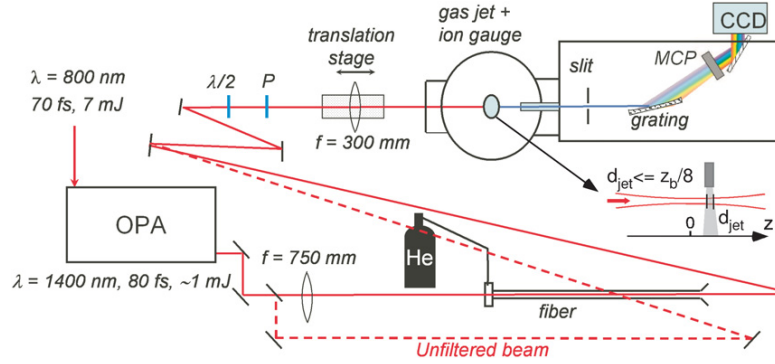


Figure 1. Experimental layout showing the optical parametric amplifier, the hollow-core fibre and the high harmonic source, and spectrometer chambers. d_{jet} and z_b are the diameter of the gas jet and confocal parameter, respectively.

which reduces the brightness of the harmonic emission. Moreover, the poor spatial quality of most available OPA sources, especially those which are seeded by parametric superfluorescence [15], can further worsen the conversion efficiency of the process to a point that outweighs the original benefit of the extended cutoff. This is in addition to the fact that the XUV beam will inherit the quality of the pump beam, which in many cases may ruin its potential as a coherent XUV source [16, 17]. Therefore, methods aimed at improving the quality of the OPA beams are necessary to take full advantage of the increase of the driver wavelength.

In this paper, we demonstrate that it is essential to spatially filter the OPA beam to maximize the efficiency of the high-order harmonic generation process. We compare the HHG produced in krypton and argon using the signal output of an OPA that is filtered through a hollow-core fibre, with those for the unfiltered case. Near- and far-field images of the two beams show that the inhomogeneities in the OPA intensity distribution are smoothed with filtering. In parallel to recording the spectra, we also monitor the number of ions produced in the gas using a fast ionization gauge, which gives an indication of the number of emitters participating in the detected harmonic field. For the same laser intensity and beam diameter at focus, we observe similar ionization probabilities but the HHG yield can be an order of magnitude larger for the spatially filtered beam. The different behaviours between the two considered gases with increasing laser fluences are also investigated and provide strong evidence that the observed enhancement in the harmonic yield is the result of better phase matching.

2. Experimental details

The experiment was conducted at the advanced laser light source (ALLS) using the 100 Hz, 100 mJ, 25 fs, Ti:sapphire laser system. A schematic of the experiment is shown in figure 1. Laser pulses of 7 mJ at 800 nm were directed to an optical parametric amplifier (TOPAS-HE from light conversion) and frequency shifted to 1400 nm signal pulses. The seed pulse of this OPA system is produced via parametric superfluorescence and suffers from a poor spatial mode quality. The pulse energy and temporal duration were measured to be about 1.3 mJ and 80 fs, respectively. Spatial filtering was

performed using a hollow-core fibre (400 μm in diameter, 100 cm long) attached at one end to a closed gas cell and supported on an aluminium V-groove [18].

Since the intent was to use the fibre to spatially filter the beam but not to change the pulse duration, the fibre was filled with helium gas to avoid self-phase modulation. Helium gas flowed through the fibre from the cell at 1.2 atm of pressure and leaked into air at the exit. We verified that the input and output spectra remained identical. The OPA signal beam was focused with an $f = 750$ mm plano-convex lens into the fibre in order to favour the coupling into the fundamental EH_{11} spatial mode, which has the highest transmissivity and whose spatial profile is a zeroth-order Bessel function of the first kind [19–21]. At the output, the laser beam was collimated with an $f = 300$ mm concave silver mirror. At 800 nm, typical transmission through a 100 cm length hollow-core fibre ranges in between 50 and 60% [22, 23]. However, due to the low quality of the mode at 1400 nm, the transmission efficiency was nearer 30%.

The filtered and unfiltered beams were focused by an $f = 300$ mm broadband antireflection-coated lens mounted on a motorized translation stage into a thin pulsed gas jet with a 500 μm aperture backed with a pressure of 3 atm of either argon or krypton. The jet is mounted on a three-axis manipulator for adjustment of its position. The laser energy could be continuously varied using a half-waveplate followed by a Glan-calcite polarizer. We have verified that reabsorption by the gas was not a limiting factor in this experiment as the harmonic yield showed a quadratic dependence as a function of gas backing pressure for pressures below 3.5 atm [27]. The position of the laser focus relative to the centre of the jet was determined using the signal of an ion detector, consisting of a wire biased at -20 V, a high-frequency decoupling circuit and an amplifier connected to a digital oscilloscope, which was held a few inches below the gas nozzle. The ion yield was monitored in parallel with the harmonic radiation throughout the experiment. The harmonic emission, collimated in the direction of the laser beam, entered a flat-field XUV spectrometer chamber through a 3 mm pinhole. The light incident on the 500 μm wide slit of the spectrometer was dispersed by a variable groove density concave grating (Hitachi) and imaged onto a chevron micro-channel plate (MCP) sensitive to wavelengths below 100 nm.

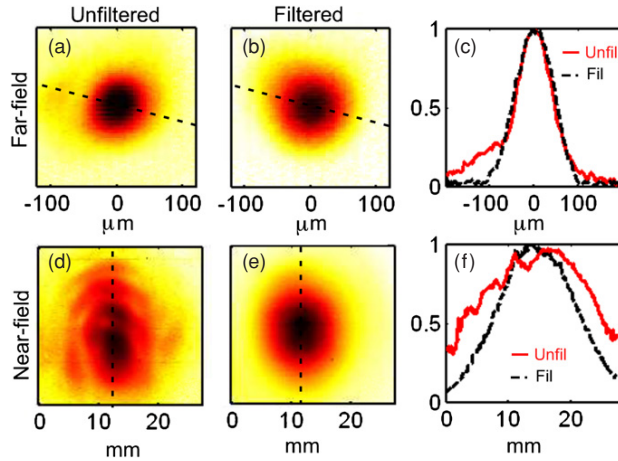


Figure 2. Far- and near-field intensity distributions of the spatially filtered ((b) and (e)) and unfiltered beams ((a) and (d)) that were recorded with an infrared vidicon camera. Axes are equal; (c) Normalized lineouts along the dashed lines in (a) and (b). (f) Lineouts along the dashed lines in (d) and (e).

A phosphor readout screen coupled to the MCP converted the amplified signal to visible light that was then imaged onto a 14-bit CCD camera placed outside the chamber. The gas jet and CCD camera were synchronized to the laser at a repetition rate of 7 Hz, and the ion detector at a rate of 100 Hz.

Near- and far-field intensity distributions of the spatially filtered and unfiltered beams were recorded with an infrared vidicon camera (Hamamatsu model C2741-03 ER) and are shown in figure 2. To record far-field beam profiles, the beam was focused by the 300 mm lens into a 20 $\times$ microscope objective mounted on the camera. The maximum beam intensity did not exceed the saturation level of the sensor when both near- and far-field beam distributions were captured with the camera. The near-field intensity distributions were recorded by sending the collimated beams directly into the camera after 5 m of free propagation in air. One can see from figures 2(d) and (e) that the energy in the unfiltered beam is non-uniformly distributed as compared to the smooth Bessel profile of the filtered beam. It is reasonable to expect that the unfiltered beam will present equally important phase variations. In addition, we observe that the spatial power distribution in the near-field pattern of the unfiltered beam varies considerably with propagation distance contrarily to the beam from the output of the fibre, indicating wavefront distortion. Wavefront inhomogeneities in amplitude and phase are known to translate into phase mismatch between adjacent portions of the interaction volume across the diameter of the beam [16].

The diameter of the filtered beam at the focusing lens was set by the position of the collimating concave silver mirror in order to match the FWHM of the central portion of the unfiltered beam's focal spot, as shown in figure 2(c). The far-field patterns are very similar except that a small fraction of the total energy is spread in a halo about the central focal spot in the unfiltered case. The power contained within the FWHM radius is $\sim 40\%$ and $\sim 58\%$ of the total power for the unfiltered

and filtered beams, respectively. The beams, however, had different Rayleigh lengths due to their different spatial energy distributions; the distance by which the spot size increased by a factor of $\sqrt{2}$ was 4 mm and 9 mm for the filtered and unfiltered beams, respectively. The corresponding confocal parameters are thus 8 mm and 18 mm. In spite of this, we estimate that the interaction volumes at a given average laser intensity should be equivalent for both beams since their confocal parameters z_b are much longer than the gas medium length, i.e. $d_{\text{jet}} \leq z_b/8$. The interaction volume is therefore essentially determined by the lateral beam spot size and by the ionization potential of the gas. Since the jet has a cylindrical symmetry, we estimate the diameter of the jet at the beam focus to be less than 1 mm by monitoring the change in the harmonic emission and ion yield as the jet is moved in the direction perpendicular to the laser propagation axis.

3. Results

Before generating harmonics, we first directly measured the laser intensity for each beam in a different apparatus, a velocity map imaging (VMI) electron spectrometer, which generates a 2D image of the velocity distribution of the electrons produced at the laser focus [24]. The same focusing optics and input window were used as with the harmonic generation chamber. From the measurement of the $2U_p$ electrons produced by circularly polarized light, one can accurately determine the laser intensity at focus [25]. For the filtered beam, the focal spot intensity per unit energy was measured to be $7.6 \times 10^{11} \text{ W cm}^{-2} \mu\text{J}^{-1}$, twice that of the unfiltered beam, which was $3.6 \times 10^{11} \text{ W cm}^{-2} \mu\text{J}^{-1}$. The energy of the unfiltered beam was adjusted so that the peak laser intensity was the same as that of the unfiltered beam. The maximum energy was 500 mJ for a maximum peak intensity for both beams of $1.8 \times 10^{14} \text{ W cm}^{-2}$, which is close to the saturation intensity in krypton.

The time-integrated ion signal, measured in the HHG chamber at various positions of the laser focus relative to the centre of the gas jet and for different laser intensities, is shown in figure 3. Each data point is the computed average over 100 laser shots. The position that corresponds to the peak of the ion count corresponds to the laser focal spot overlapping with the thin jet ($z = 0$). Negative values of z correspond to the focus being placed in front of the jet. The ion signal decreases more rapidly with distance when the beam is filtered as confirmed by the different confocal parameters for the two beams. The ion yield gives us an indication of the number of emitters and therefore shows us how the interaction volumes grow with the laser intensity in each gas. The number of ions produced by the unfiltered beam is equal to (in argon) or larger (in krypton) than that produced by the filtered beam, suggesting slightly more emitters in krypton. However, as we will see next, the harmonic yield obtained with the filtered beam is much larger in all cases.

The total harmonic yield in argon and krypton, averaged over 100 laser shots, is shown in figure 4 as a function of the focus position and peak laser intensity. The total yield was obtained by integrating each spectrum between 10 eV and

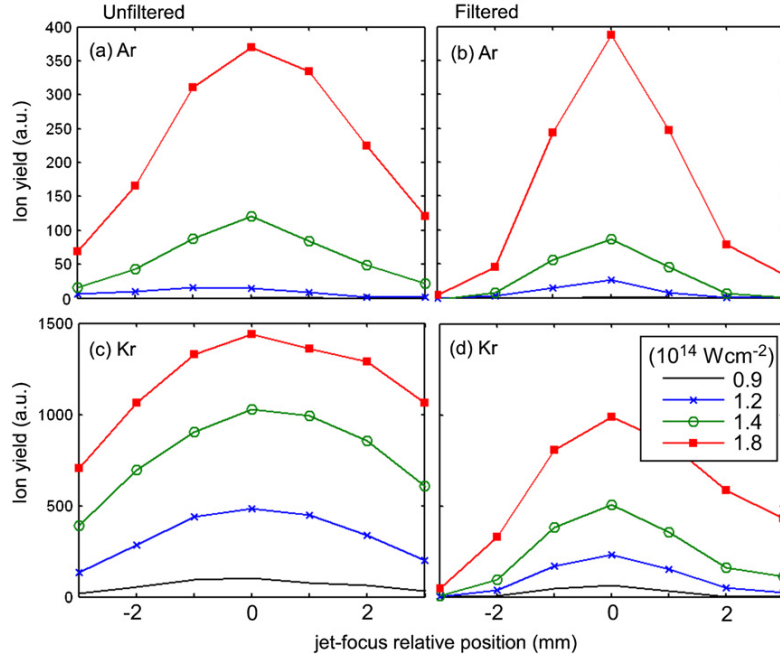


Figure 3. Comparison of the total ionization yield between the fibre-filtered and unfiltered pump beams in argon ((a) and (b)) and in krypton ((c) and (d)). The ion signal is shown as a function of laser intensity at focus and relative distance between the focus and the center of the gas jet. The legend applies to all graphs.

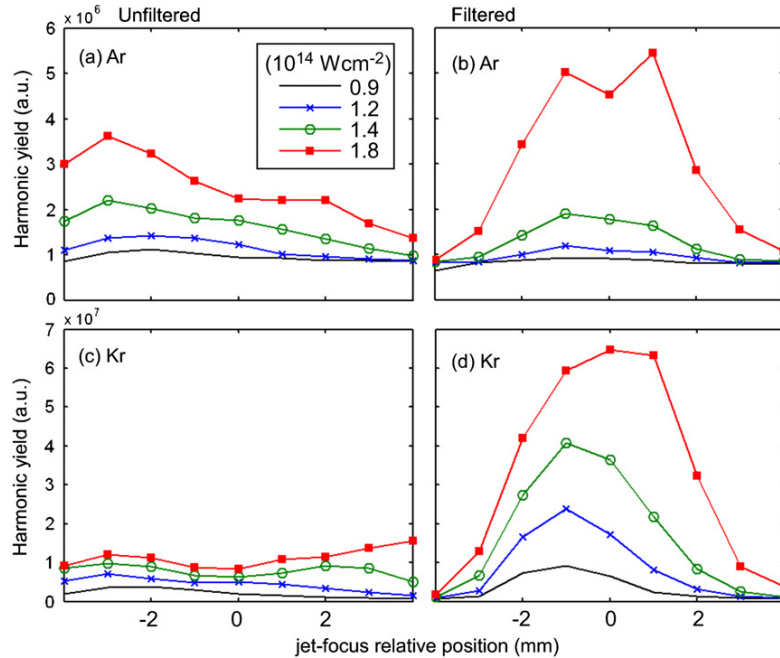


Figure 4. Comparison of the total harmonic yield (integrated over the 10–65 eV range) between the fibre-filtered and unfiltered pump beams in argon ((a) and (b)) and in krypton ((c) and (d)). The signal is shown as a function of the laser intensity at focus and relative distance between the focus and the centre of the gas jet. The legend applies to all graphs.

65 eV. For the filtered beam, an increase in the HHG yield of up to 7.7 for Kr and of 2.5 for Ar is observed compared to the unfiltered case. If we narrow the integration range to the emission of photons above 30 eV, i.e. the critical range for attosecond pulse synthesis, this ratio reaches 9.5 in krypton and 3.0 in argon. In agreement with previous studies using

Bessel beams [20, 21], the HHG yield is maximum when the filtered beam is focused near the centre of the gas jet. In figure 5, we show the average harmonic spectra in krypton produced with the filtered and unfiltered beams at three z positions of the focus relative to the jet and at a laser intensity at focus of $1.8 \times 10^{14} \text{ W cm}^{-2}$. In all three cases, filtering

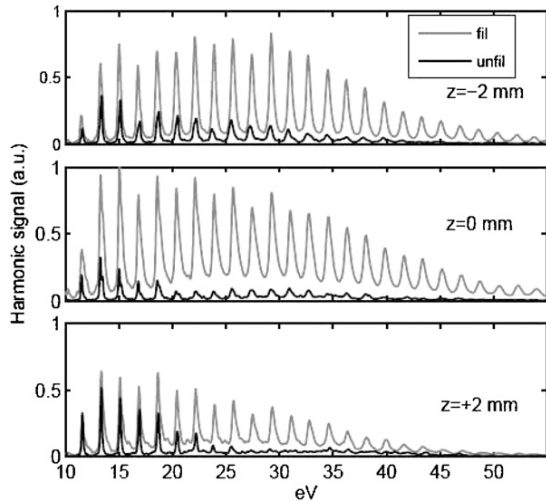


Figure 5. Average harmonic spectra in krypton produced with the fibre-filtered (grey) and unfiltered (black) 1400 nm beams at three z positions of the focus relative to the jet and at a laser intensity at focus of $1.8 \times 10^{14} \text{ W cm}^{-2}$. The signal scale is normalized to the strongest signal.

of the beam not only produces brighter plateau harmonics but also extends the cutoff energy by at least 10–15 eV.

4. Discussion

Harmonic generation is optimized when phase matching is achieved, i.e. when the difference between the phase front of the harmonic field and that of the driving laser field is minimized. One of the main causes of variation in the nonlinear polarization induced by the laser field is due to rapid variations of the dipole phase with intensity I , especially for mid-infrared lasers since it scales as $\lambda^3 I$ [33]. It induces a spatially and temporally dependent phase term in the medium which influences the generation of the macroscopic field. This phase modulation is all the more important as the peak intensity is high. Another contribution is that imparted by the phase variations in the IR laser wavefront itself which imprints on the generated field and impairs the ability for all emitting source points to interfere in the far field. As clearly explained in [32], all the processes which cause different time histories of the rays emerging from two points in the medium will affect phase matching. For example, additional phase mismatch may arise from time and space variations of the ionization which creates an electron density that varies from place to place. Nevertheless, phase-matching calculations that take into account both axial and transverse mismatch are difficult to perform without complete knowledge of the experimental laser wavefront. Indeed, a comparative study between a Gaussian and a Bessel beam under the same experimental conditions for the generation of harmonics has revealed fundamental differences in the phase-matching conditions that were directly linked to the beam mode [21]. Also, current models of the macroscopic response of the medium assume a uniform transverse wavefront and most neglect phase variations across the diameter of the beam in order to concentrate on axial

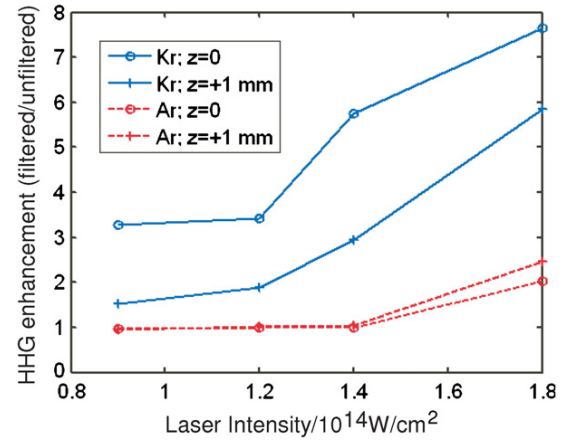


Figure 6. Ratio between the total harmonic yield obtained with the spatially filtered beam and the unfiltered beam as a function of laser intensity in both argon (red dashed lines) and krypton (blue solid lines) at a focus position of $z = 0$ (open circles) and $z = +1 \text{ mm}$ (plus signs).

phase-matching conditions [33]. However, a few groups have investigated the effect of time-dependent phase variations between two non-symmetrical points across the diameter of a Ti:Sa beam on the phase-matching conditions and the subsequent spatial coherence between the two point sources [16, 31, 32]. As briefly discussed in [16], this is similar to the case where the fluctuations arise from inhomogeneities in the IR wavefront. In conditions similar to ours, they find that the coherence is highly sensitive to irregularities in the beam profile. Very recently, measurements of the wavefronts of an 800 nm beam and its 13th harmonic were performed by Gautier *et al* [29] and have shown that the quality of the HHG beam is directly correlated with that of the IR pump.

The harmonic yield ratios between the filtered and unfiltered conditions are plotted in figure 6 as a function of peak laser intensity at the position $z = 0$ for the two gases. The improvement in the harmonic yield with spatial filtering increases with the laser intensity in both gases. In light of the above discussion, we believe that this can be explained by the difference in the transverse extension of the interaction volume in connection to the spatial uniformity of the beam over this area, in a similar fashion as in [32]. At low laser fluence, only a portion limited to the centre of the focal spot will produce sufficient ionization leading to detectable harmonic emission. In this case, the transverse uniformity of the pump beam only needs to be satisfactory over the small area contributing to the emission for efficient buildup. As such, we observe no improvement with filtering at these low peak intensities, i.e. the gain is close to 1. Spatial uniformity of the intensity distribution becomes more important as the peak intensity grows since (1) a larger area can contribute to the emission of the harmonics (source points further apart will be more sensitive to phase variations) and (2) because the phase mismatch due to the atomic dipole phase also increases with I . Smoothing of the laser wavefront is thus likely to have a more significant effect on the harmonic yield gain for the higher peak laser intensities.

In a similar way, the difference in the transverse extension of their respective interaction volumes can also explain the observed differences in the harmonic yield ratios between argon and krypton. The amount of ionization during the time window in the pulse that a particular harmonic is generated is calculated using single-atom ADK (Ammosov–Delone–Krainov) tunnelling ionization rates [28]. Using the model presented in [26], we find that at the maximum laser intensity used during the experiment the ionization probability at the peak of the pulse is 7% in argon compared to 50% in krypton. The emitting volume will thus be proportionally smaller in argon compared to krypton. Instead of producing an estimate of the total ionization over the intensity profiles presented in figure 2, one way to verify this assumption is to compare our results at intensities where both gases exhibit similar ADK ionization curves. The intensity at which the ADK ionization rate in krypton is the same as the rate in argon at the maximum laser intensity is close to $1.2 \times 10^{14} \text{ W cm}^{-2}$. From figure 6, we find that the harmonic yield gain in krypton at $1.2 \times 10^{14} \text{ W cm}^{-2}$ is 3.4 at $z = 0$, in close agreement to the value of 2 that is measured in argon for an intensity of $1.8 \times 10^{14} \text{ W cm}^{-2}$. At $z = 1 \text{ mm}$, the respective gains are both close to 2. Moreover, as one can see from figure 3, the corresponding ion yields are also similar. In the same way, we believe that if we could attain a peak intensity of $2.8 \times 10^{14} \text{ W cm}^{-2}$ with the filtered beam, the increase factor in argon would then be similar to the factor measured in krypton at $1.8 \times 10^{14} \text{ W cm}^{-2}$ since their ionization rates match at these intensities.

In this experiment we made sure that the interaction volumes were equivalent for the filtered and unfiltered conditions by (1) using a gas jet that is much thinner than the confocal parameter of either beam and (2) by adjusting the divergence of the filtered beam such that their focal spot sizes were identical. In addition, we observe that, at constant laser intensity, the ion yield for the filtered beam is either slightly lower than or equal to the yield of the unfiltered beam, which rules out the possibility that the observed improvement in the harmonic generation efficiency is due to an increase in the number of emitters for the filtered condition. The ion yield measurements, performed simultaneously with the recording of the HHG spectra, confirm that the interaction volumes are highly similar for both laser conditions. Moreover, the comparison between the results obtained in argon and krypton at various laser intensities strongly suggests that spatial filtering improves the phase matching between contributing source points across the emitting volume, which results in a more efficient buildup of the harmonic signal. As mentioned earlier, due to its smaller ionization potential, a larger fraction of the atoms will be ionized in krypton compared to argon at a given peak laser intensity; hence, a larger area of the IR beam will contribute to HHG. In agreement with our results, we find that the larger the area contributing to the signal, the better is the improvement in the harmonic yield after smoothing of the laser intensity profile. In support of improved phase matching, we observe an extension of the spectrum towards higher energies with the filtered beam (cf figure 5) and a better spatial quality of the produced harmonics which are

characterized by a low divergence (less than 5 mrad for all orders) that is maintained over a wider range of focus positions.

5. Conclusion

We report significant improvement of the high-order harmonic generation using a 1400 nm OPA beam that is spatially filtered by a hollow-core fibre. The harmonic signal integrated above 30 eV increases by almost a factor of 10 relative to the unfiltered case using only half the energy per pulse. In the future, the next step will be to produce few-cycle IR laser pulses with the hollow-core fibre setup in order to obtain higher intensities per pulse and generate shorter attosecond pulses from the ionization of high I_p atoms. At the moment, our laser system is not carrier-envelope phase (CEP) stabilized and cannot be used to produce isolated attosecond laser pulses. However, our work clearly demonstrates an additional advantage to perform pulse compression of OPA laser pulses with the hollow-core fibre approach.

Acknowledgments

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References

- [1] L’Huillier A and Balcou P 1993 *Phys. Rev. Lett.* **70** 774
- [2] Brabec T and Krausz F 2000 *Rev. Mod. Phys.* **72** 545
- [3] Hentschel M *et al* 2001 *Nature* **414** 509
- [4] Baltuska M *et al* 2003 *Nature* **421** 611
- [5] Sansone G *et al* 2006 *Science* **314** 443
- [6] Mashiko H, Gilbertson S, Li C, Khan S D, Shakya M M, Moon E and Chang Z 2008 *Phys. Rev. Lett.* **100** 103906
- [7] Goulielmakis E *et al* 2008 *Science* **320** 1614
- [8] Corkum P B and Krausz F 2007 *Nature Phys.* **3** 381
- [9] Corkum P B 1993 *Phys. Rev. Lett.* **71** 1994
- [10] Bellini M 2000 *Appl. Phys. B* **70** 773
- [11] Shan B and Zenghu C 2002 *Phys. Rev. A* **65** 011804
- [12] Shan B, Cavalieri A and Chang Z 2002 *Appl. Phys. B* **74** S23
- [13] Takahashi E J, Kanai T, Nabekawa Y and Midorikawa K 2008 *Appl. Phys. Lett.* **93** 041111
- [14] Tate J, Auguste T, Muller H G, Salières P, Agostini P and DiMauro L F 2007 *Phys. Rev. Lett.* **98** 013901
- [15] Cerullo G and De Silvestri S 2003 *Rev. Sci. Instrum.* **74** 1
- [16] Salières P, L’Huillier A, Antoine Ph and Lewenstein M 1999 *Adv. At. Mol. Opt. Phys.* **41** 83
- [17] Hergott J T *et al* 2003 *J. Opt. Soc. Am. B* **20** 171
- [18] Légaré F, Litvinyuk I V, Dooley P W, Quéré F, Bandrauk A D, Villeneuve D M and Corkum P B 2003 *Phys. Rev. Lett.* **91** 093002
- [19] Marcetili E A J and Schmeltzer R A 1964 *Bell Syst. Tech. J.* **43** 1783
- [20] Nisoli M *et al* 2002 *Phys. Rev. Lett.* **88** 033902
- [21] Altucci C *et al* 2003 *Phys. Rev. A* **68** 033806
- [22] Mashiko H, Nakamura C M, Li C, Moon E, Wang H, Tackett J and Chang Z 2007 *Appl. Phys. Lett.* **90** 161114

- [23] Cavalieri A L *et al* 2007 *New J. Phys.* **9** 242
- [24] Eppink A T J B and Parker D H 1997 *Rev. Sci. Instrum.* **68** 3477
- [25] Alnaser A S, Tong X M, Osipov T, Voss S, Maharjan C M, Shan B, Chang Z and Cocke C L 2004 *Phys. Rev. A* **70** 023413
- [26] Spielmann C, Burnett N H, Sartania S, Koppitsch R, Schnrer M, Kan C, Lenzner M, Wobrauschek P and Krausz F 1998 *IEEE J. Sel. Top. Quantum Electron.* **4** 249
- [27] Brichta J-P, Wong M C H, Bertrand J B, Bandulet H-C, Rayner D M and Bhardwaj V R 2008 *Phys. Rev. A* submitted
- [28] Ammosov M, Delone N and Krainov V 1986 *Sov. Phys.—JETP* **64** 1191
- [29] Gautier J *et al* 2008 *Eur. Phys. J. D* **48** 459
- [30] Ditmire T, Gumbrell E T, Smith R A, Tisch J W G, Meyerhofer D D and Hutchinson M H R 1996 *Appl. Phys.* **65** 313
- [31] Ditmire T, Tisch J W G, Gumbrell E T, Smith R A, Meyerhofer D D and Hutchinson M H R 1997 *Appl. Phys. B* **65** 313
- [32] Le Déroff L, Salières P, Carré B, Joyeux D and Phalippou D 2000 *Phys. Rev. A* **61** 043802
- [33] Gaarde M, Tate J L and Schafer K J 2008 *J. Phys. B: At. Mol. Opt. Phys.* **41** 132001

Chapter 6

Optical Parametric Amplification and Pulse Compression

This chapter is based on the following papers:

1. M. Giguère, B. E. Schmidt, **A. D. Shiner**, M.-A. Houle, H. C. Bandulet, G. Tempea, D. M. Villeneuve, J.-C. Kieffer, and F. Légaré, “Pulse compression of submillijoule few-optical-cycle infrared laser pulses using chirped mirrors,” *Opt. Lett.*, vol. 34, no. 12, pp. 1894–1896, 2009
2. B. E. Schmidt, P. Béjot, M. Giguère, **A. D. Shiner**, C. Trallero-Herrero, É. Bisson, J. Kasparian, J. Wolf, D. M. Villeneuve, J. Kieffer, P. B. Corkum, and F. Légaré, “Compression of 1.8 μm laser pulses to sub two optical cycles with bulk material,” *Applied Physics Letters*, vol. 96, p. 121109, Mar. 2010

Section 2.4 showed the example of HHG in hydrogen, where 210 eV photons are produced through an extreme nonlinear interaction involving several hundred 0.69 eV photons. While the end result of HHG experiments is clearly in the realm of extreme nonlinear optics, the technologies used to produce short femtosecond laser pulses rely on low order non-linear processes to shift and broaden the laser spectrum. In all cases the mechanism for producing new optical frequencies, or moving energy between fields at different wavelengths, is rooted in the acceleration of charges. The response of a material to an electric field is described by its polarization $P(t)$, and the second time derivative of the polarization leads to radiation. In linear optics the polarization is proportional to the magnitude of the field $P(t) = \chi^{(1)}E(t)$, and its frequency dependence is responsible for the wavelength-dependent index of refraction, which causes chromatic dispersion. As the field strength increases the electrons in the material are

no longer able to keep up with the changing field, and the polarization develops a nonlinear response. The different orders of nonlinearity are seen by expanding the polarization in powers of the electric field [81]:

$$P(t) = \chi^{(1)}E(t) + \chi^{(2)}E(t)^2 + \chi^{(3)}E(t)^3 + \dots \quad (6.0.1)$$

The propagation of electromagnetic waves is governed by the wave equation [88]:

$$\nabla^2 \mathbf{E} - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}^L}{\partial t^2} + \mu_0 \frac{\partial^2 \mathbf{P}^{NL}}{\partial t^2} \quad (6.0.2)$$

where $\mathbf{P}^L$ is the linear polarizability (first term in Eq. 6.0.1) and $\mathbf{P}^{NL}$ describes the nonlinear response, which acts as a nonlinear source term in the wave equation. It is often convenient to express the linearly polarized electric field $\mathbf{E}(z, t)$ as ([88] (2.3.21)):

$$\mathbf{E}(z, t) = \frac{1}{2} \hat{x} \{ A(z, t) \exp[i(k_0 z - \omega_0 t)] + c.c. \} \quad (6.0.3)$$

where the field is assumed to only propagate in the forward $\hat{z}$ direction, $A(z, t)$ is the pulse envelope and k_0 is its wave number. To solve for the evolution of a pulse, the ansatz (Eq. 6.0.3) is substituted into the wave equation. After following through the derivatives we find that carrier frequency term $\exp[i(\omega_0 t)]$ is common to every term, on both sides of the equation, and cancels [88]. Even for few-cycle pulses, it is sufficient to propagate the complex envelope $\mathbf{A}(z, t)$ forward in space and time and multiply the carrier back in at the end [89], provided that any backward propagating component of the beam is negligible.

6.1 Linear Dispersion and Spectral Phase

To calculate the effect of linear dispersion, the nonlinear source term $\partial_t^2 \mathbf{P}^{NL}$ in Eq. 6.0.2 is set to zero and the equation is Fourier transformed into the frequency domain. Note that a tilde over a variable or operator is used to indicate that it has been Fourier transformed into the optical frequency domain. The propagation of the envelope $\tilde{\mathbf{A}}(z, \omega)$ is described by:

$$\frac{\partial \tilde{A}(z, \omega)}{\partial z} = \tilde{\mathbb{D}} \tilde{A}(z, \omega) \quad (6.1.1)$$

with dispersion operator $\tilde{\mathbb{D}} = i k(\omega)$ and wave number $k(\omega) \approx n(\omega)\omega/c$. If the index of refraction $n(\omega)$ was constant, the relative phases between frequency components in a pulse would remain locked, and its temporal profile would not change as it propagates through material. In practice $n(\omega)$ is not constant and material dispersion plays an important role for generating and maintaining short pulses. To understand how spectral phase affects a pulse, it is helpful to separately consider the role of the different terms in a series expansion of the wavenumber $k(\omega)$ around the carrier frequency ω_0 .

$$k(\omega) = k_0 + (\omega - \omega_0)k_1 + \frac{1}{2}(\omega - \omega_0)^2k_2 + \frac{1}{6}(\omega - \omega_0)^3k_3 + \dots \quad (6.1.2)$$

where $k_0 \equiv k(\omega_0)$ and:

$$k_m = \left(\frac{d^m k}{d\omega^m} \right)_{\omega=\omega_0} \quad (m = 0, 1, 2, \dots). \quad (6.1.3)$$

Figure 6.1 shows the affect of different terms in the series expansion of $k(\omega)$ on a 3-cycle pulse. In all cases the spectrum remains the same; only the relative phase between spectral components will change. The frequency-independent dispersion term k_0 shifts the spectral phase by the same amount for all wavelengths as a pulse propagates. Adding this constant offset to the spectral phase in the frequency domain causes a shift in the relative phase between the carrier and the envelope in time, as shown in Fig. 6.1 (a). Methods for measuring and controlling the carrier envelope phase (CEP) are described in Ch. 7. The linear term $(\omega - \omega_0)k_1$ causes the pulse envelope to move forward or backward in time. The quadratic term $\frac{1}{2}(\omega - \omega_0)^2k_2$ symmetrically broadens the pulse, whereas the cubic term $\frac{1}{6}(\omega - \omega_0)^3k_3$ contributes a spectral phase shift that is asymmetric in frequency, leading to broadening that is asymmetric in time (Fig. 6.1 h). Analytic expressions for k_1 and k_2 are given below [88]:

$$k_1 = \frac{1}{v_g} = \frac{n_g}{c} = \frac{1}{c} \left(n + \omega \frac{dn}{d\omega} \right) \quad (6.1.4)$$

$$k_2 = \frac{1}{c} \left(2 \frac{dn}{d\omega} + \omega \frac{d^2n}{d\omega^2} \right) \quad (6.1.5)$$

where v_g is the group velocity of the pulse and k_2 is responsible for group velocity dispersion.

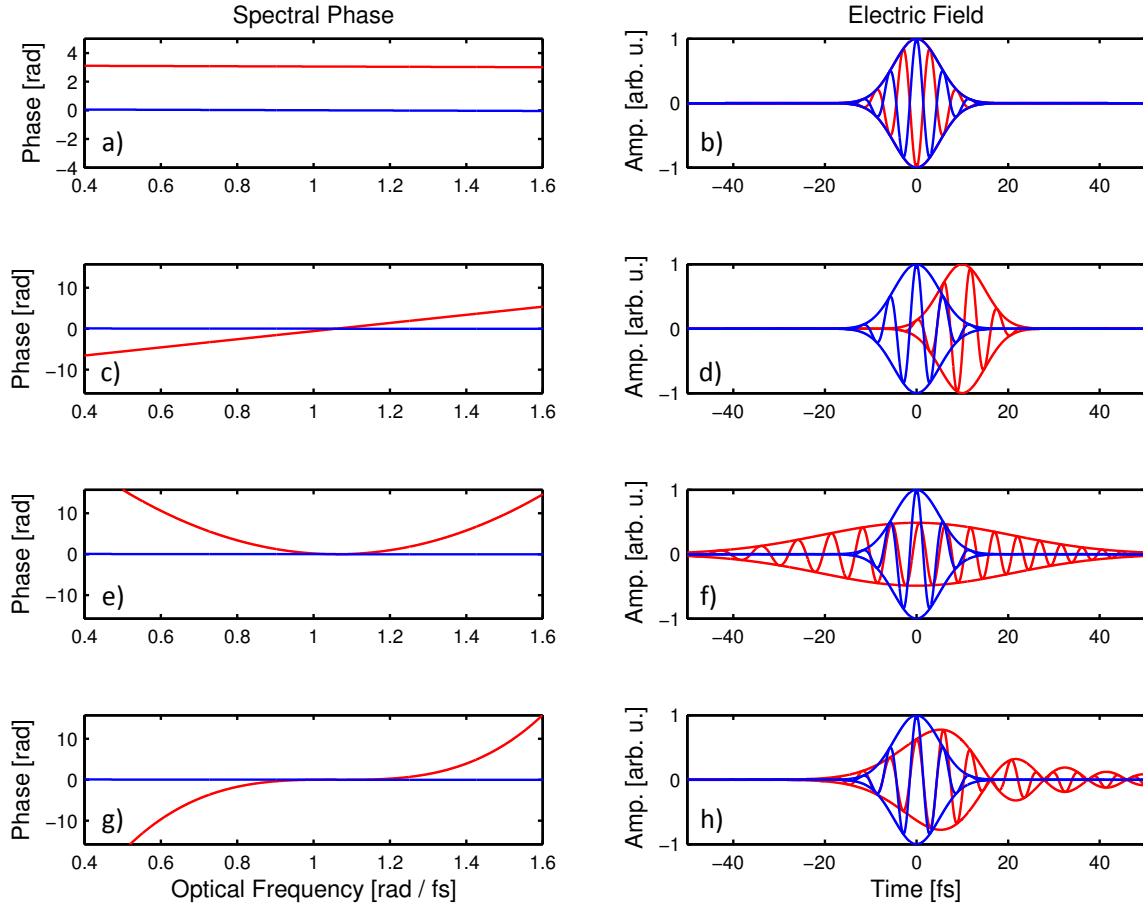


Figure 6.1: Effect of different terms in the series expansion of $k(\omega)$ on a 3-cycle transform limited pulse showing the change in spectral phase (left column), and the corresponding change in the temporal profile of the pulse (right column). Results are shown for the frequency-independent term (a-b), linear spectral phase (c-d), quadratic phase (e-f), as well as cubic phase (g-h).

6.2 Nonlinear Material Response

It is easiest to solve for the nonlinear material response in the time domain where, in most cases only the second or third term in the series expansion of $P(t)$ is included. In the Fourier domain, the second order response is given by the convolution of two electric fields:

$$\tilde{P}^{(2)}(\omega) = \chi^{(2)} \tilde{E}(\omega) \otimes \tilde{E}(\omega) \quad (6.2.1)$$

thus, all pair-wise combinations of frequency components in the electric field(s) contribute to the second order polarization. If we consider the interaction between fields with discrete frequencies labeled ($m = 1, 2, 3$):

$$\mathbf{E}(z, t)_m = A_m \exp[i(k_m z - \omega_m t)] + c.c. \quad (6.2.2)$$

and treat the case where two of the frequency components interact with a nonlinear medium, the polarization is given by:

$$P^{(2)}(t) = \chi^{(2)} [E_1(t) + E_2(t)]^2 \quad (6.2.3)$$

$$P^{(2)}(t) = \chi^{(2)} [A_1 e^{i(k_1 z - \omega_1 t)} + A_2 e^{i(k_2 z - \omega_2 t)} + c.c.]^2 \quad (6.2.4)$$

$$\begin{aligned} P^{(2)}(t) = \chi^{(2)} [& A_1^2 e^{i 2(k_1 z - \omega_1 t)} + A_2^2 e^{i 2(k_2 z - \omega_2 t)} + \dots \\ & 2 A_1 A_2 e^{i[(k_1 + k_2) z - (\omega_1 + \omega_2) t]} + 2 A_1 A_2^* e^{i[(k_1 - k_2) z - (\omega_1 - \omega_2) t]} + c.c.] + \dots \quad (6.2.5) \\ & 2 \chi^{(2)} [A_1 A_1^* + A_2 A_2^*] \end{aligned}$$

The terms on the first line of Eq. 6.2.5 correspond to second harmonic generation and lead to the production of frequency components at $2\omega_1$ and $2\omega_2$. The terms on the second line correspond to sum frequency generation ($\omega_1 + \omega_2$), and difference frequency generation ($\omega_1 - \omega_2$) and the last line gives zero frequency terms. In an optical parametric amplifier, sum and difference-frequency generation is used to exchange energy between a strong pump beam and the longer wavelength signal and idler beams. This process is discussed in the next section.

6.3 Optical Parametric Amplification

The titanium sapphire laser used for all experiments presented in this thesis produces ~ 45 fs pulses at a centre wavelength of 800 nm. To generate longer wavelengths we

used an Optical Parametric Amplifier (OPA), which essentially takes one photon from the pump laser ω_p and splits it into two photons labeled as the signal ω_s and the idler ω_i , where $(\omega_p > \omega_s > \omega_i)$. This splitting must satisfy energy conservation such that $\omega_p = \omega_s + \omega_i$, as well as momentum conservation $\mathbf{k}_p = \mathbf{k}_s + \mathbf{k}_i$. The division of energy between the signal and idler photons is adjustable, so an OPA provides a tunable source of femtosecond pulses spanning the range of $1.2\ \mu\text{m}$ to $1.6\ \mu\text{m}$ (signal), and $1.6\ \mu\text{m}$ to $2.4\ \mu\text{m}$ (idler). The basic arrangement for an OPA is shown in Fig. 6.2, where in this example the pump radiation is divided, part of it is used for seeding and the rest is used for the first and second amplification stages. An OPA always starts by producing a seed at the signal wavelength, usually using either superfluorescence from focusing the pump laser into a nonlinear crystal, or white light seeding from self-phase modulation in a sapphire window. In either case the objective is to produce a small amount of radiation at the desired signal wavelength. The seed and one of the pump beams are combined in a nonlinear crystal, where energy is transferred from the pump to the seed and the excess energy leads to the production of photons at the idler wavelength. Exiting the crystal, the seed (now referred to as the signal beam), is directed into one or more amplification stages where it is combined with additional pump radiation, and the process repeats.

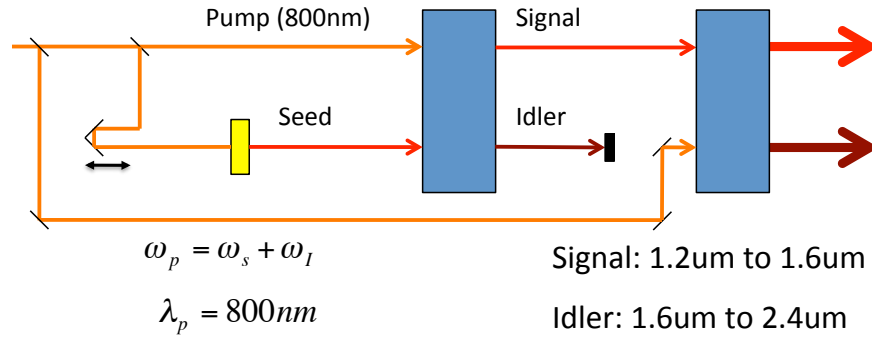


Figure 6.2: Overview of Optical Parametric Amplification.

The underlying mechanism of an OPA relies on the second-order material response $(\chi^{(2)}E(t)^2)$, to couple energy between the pump, signal and idler fields. The nonlinear wave equation can be written as three coupled wave equations[90]:

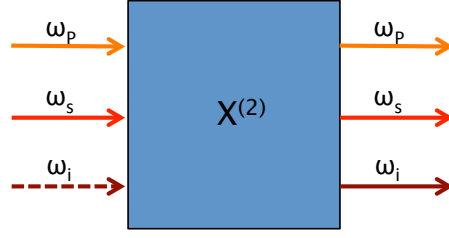


Figure 6.3: Illustration of three-wave mixing.

$$\frac{\partial A_s}{\partial z} + \frac{1}{v_{gs}} \frac{\partial A_s}{\partial t} = -i \frac{\omega_s d_{\text{eff}}}{n_s c} A_i^* A_p \exp(-i \Delta k z) \quad (6.3.1)$$

$$\frac{\partial A_i}{\partial z} + \frac{1}{v_{gi}} \frac{\partial A_i}{\partial t} = -i \frac{\omega_i d_{\text{eff}}}{n_i c} A_s^* A_p \exp(-i \Delta k z) \quad (6.3.2)$$

$$\frac{\partial A_p}{\partial z} + \frac{1}{v_{gp}} \frac{\partial A_p}{\partial t} = -i \frac{\omega_p d_{\text{eff}}}{n_p c} A_s A_i \exp(+i \Delta k z) \quad (6.3.3)$$

where d_{eff} is the nonlinear optical coefficient, which relates to $\chi^{(2)}$ and depends on the propagation direction and polarization of the beams in the nonlinear material. A weak seed entering a nonlinear crystal will exchange energy with the pump field and grow. Each time a seed photon is created from the annihilation of a pump photon, an idler photon is also created with the excess energy between the pump and signal photons. Efficient energy transfer requires the beams to travel together through the crystal, which is satisfied when the wave vector mismatch $\Delta k = k_p - k_s - k_i$ is close to zero. Phase matching allows us to select the desired wavelength, and to maximize their amplification. In practice, the OPA wavelengths are chosen by controlling which seed wavelength overlaps with the pump at the input to the nonlinear medium, as well as by setting the orientation of the nonlinear crystal.

6.4 Pulse Compression: Overview

As described in the introduction to this thesis, the minimum duration for a pulse is limited by its spectral bandwidth. The OPA at ALLS produced nearly transform-limited pulses with durations of around 45 fs. In order to produce shorter pulses, we need to broaden the spectrum and then compress the pulse by flattening its spectral phase. For spectral broadening we focused the laser beam into a one meter long,

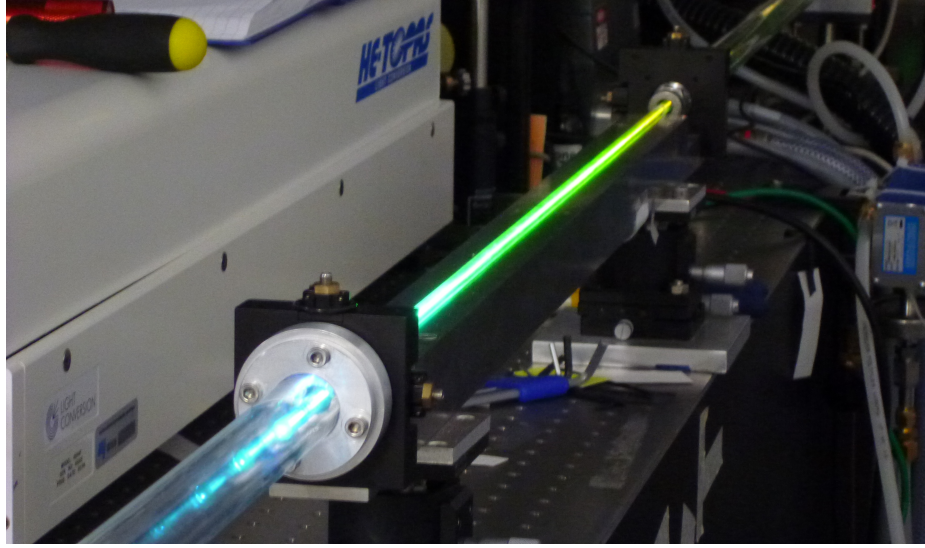


Figure 6.4: Hollow core fiber used for spectral broadening. The $1.8\,\mu\text{m}$ laser pulse propagates from the background to the foreground in the photo. The green colour that is visible to the eye is third harmonic generation in argon.

$400\,\mu\text{m}$ diameter, hollow core fiber (HCF), filled with argon gas. The fiber tightly confines the laser so that a high intensity is maintained throughout the length of the fiber. Self-Phase Modulation (SPM), in argon broadens the spectrum, while the fiber waveguide acts as a spatial filter, reducing nonlinear coupling into higher order modes during SPM. A photograph of the argon filled HCF used at ALLS is shown in Fig. 6.4.

6.5 Self-Phase Modulation

Equation 6.2.5 lists the terms arising from the second-order material response to an applied electric field. Each term contained the product of two electric fields or one electric field with itself. In contrast, self-phase modulation is a third-order process involving the product of three fields. In isotropic media the nonlinear polarization is given by [81]:

$$\tilde{P}^{(2)}(t) = 6\epsilon_0\chi_{1122}^{(3)}(\mathbf{E}(t)\mathbf{E}(t)^*)\mathbf{E}(t) + 3\epsilon_0\chi_{1221}^{(3)}(\mathbf{E}(t)\cdot\mathbf{E}(t))\mathbf{E}(t)^* \quad (6.5.1)$$

The first term is proportional to the intensity ($\mathbf{E}^* \mathbf{E} = |\mathbf{E}|^2 \propto I$), and is responsible for the nonlinear refractive index. The vectorial nature of the second term couples energy between polarization states and is responsible for cross phase modulation [88]. For spectral broadening, we use linearly polarized light and the lowest order spacial mode so the $(\mathbf{E} \cdot \mathbf{E})\mathbf{E}^*$ term is zero. With SPM, the index of refraction can be written as $n(t) = n_0 + n_2 I(t)$, where n_0 is the linear index of refraction and $n_2 I(t)$ is the nonlinear contribution that is proportional to the instantaneous intensity $I(t)$. Here we assume an instantaneous material response, and for now assume that the interaction length is short enough so that there is no temporal re-shaping of the pulse during SPM. The action of SPM is to cause a time (intensity), dependent phase shift as the pulse propagates through the medium:

$$\Phi_{nl}(t) = -n_2 I(t) \frac{\omega_0 L}{c} \quad (6.5.2)$$

where L is the propagation distance and c is the speed of light. A time-dependent phase shift is associated with the creation of new frequencies [84]. The instantaneous frequency is $\omega(t) = \omega_0 + \delta\omega(t)$, where

$$\delta\omega(t) = \frac{d}{dt} \Phi_{nl}(t). \quad (6.5.3)$$

Figure 6.5 shows the result of a split-operator calculation of the spectrum of an 80 fs, $1.8 \mu\text{m}$ pulse as it propagates down a 1 m long HCF filled with argon. This calculation used the same material parameters as the simulation by B  jot [30] but only included the effects of SPM and self-steepening, as well as the full linear material dispersion from the Sellmeier equations. The propagation method is similar to that described in Agrawal [88] as well as Dudley [91] but uses a more sophisticated, fourth order split-step algorithm [92]. As the pulse propagates down the fiber, its spectral bandwidth increases from 61 nm at the entrance of the fiber to roughly 482 nm at the exit. If the simulation only considers the effect of SPM the pulse will exit the fiber with a positive chirp and the same duration that it had going in. For pulses with very large bandwidth, such as the case in Fig. 6.5, there is an intensity-dependent change in the group velocity. As the pulse propagates, the index of refraction is larger near the peak of the pulse, (compared to the leading edge), and the pulse slows down near the peak causing the energy in the falling edge to *pile up*. This intensity-dependent change in group velocity is known as *self-steepening*. Figure 6.6 shows the calculated temporal

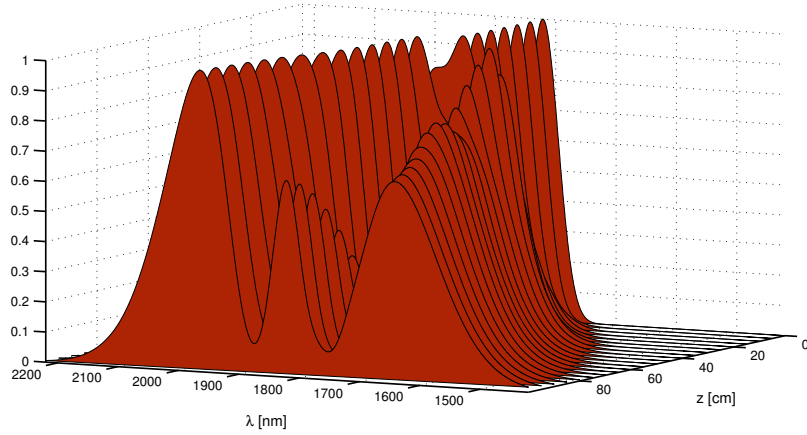


Figure 6.5: Calculated spectrum (λ), for different propagation distances (z), along an argon-filled hollow core fiber including the effect of dispersion, self-phase modulation, and self-steepening. The spectral bandwidth increases from 61 nm to ~ 482 nm. (1 mJ, $1.83 \mu\text{m}$, 80 fs, $\omega_0 = 128.7 \mu\text{m}$)

profile of the pulse at the output of the fiber. When self-steepening is included, the pulse becomes asymmetric and is shifted toward later times. Self-steepening affects both the temporal envelope as well as the spectral phase of the pulse exiting the fiber. Compressing the pulse requires us to flatten the spectral phase as much as possible, so it is necessary to include the action of self-steepening when modeling propagation. Following B  jot [30], the full model for the propagation of the pulse through the argon filled fiber is:

$$\partial_z \tilde{A}(z, \omega) = \left(\tilde{\mathcal{D}} + \tilde{\mathcal{N}} - \frac{\tilde{\alpha}}{2} \right) \tilde{A}(z, \omega) \quad (6.5.4)$$

where α accounts for absorption during propagation. The linear dispersion operator,

$$\tilde{\mathcal{D}} = i[k(\omega) - k_0 - (\omega - \omega_0)k_1] \quad (6.5.5)$$

is the same as the operator in Sec. 6.1, except that the k_0 and k_1 contributions are subtracted from the wave vector. By doing this we transform to a frame that is moving with the pulse at its group velocity. The nonlinear operator $\mathcal{N}$ is given by:

$$\mathcal{N} = i k_0 (1 + i \tau_{\text{shock}} \partial_t) \Delta n \quad (6.5.6)$$

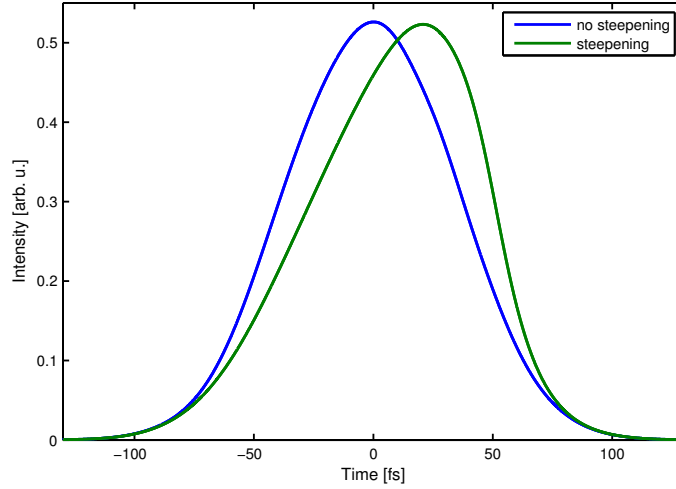


Figure 6.6: Effect of self-steepening for a $1.8\ \mu\text{m}$, 80 fs, 1 mJ pulse propagating in a $400\ \mu\text{m}$ diameter hollow fiber filled with argon. The plot shows the time domain pulse exiting the fiber with and without the self-steepening term.

where the ∂_t term leads to self-steepening and $\Delta n = \sum_{m=1}^5 n_{2m}|A|^{2m}$ is the instantaneous Kerr effect. In the full calculation B  jot and co-worker included up to the tenth power of $|A|$ in the nonlinear source term [30]. The simulation results in this section are limited to the second order $|A|^2$ term. The nearly 500 nm wide spectrum at the output of the fiber would support a pulse duration of around 10 fs if fully compressed. However, we find that the duration at the output of the fiber is comparable to the duration at the input because the pulse is significantly chirped. To achieve a shorter pulse the beam at the output of the fiber needs to be compressed.

6.6 Pulse Compression: Results

The pulse exiting the fiber has a significant positive chirp, which in this case means that it has a predominantly quadratic spectral phase with the red frequency components in the leading edge of the pulse and the blue components in the trailing edge. To compress the pulse, these frequency components must be brought together in time, which corresponds to flattening the spectral phase. For most applications at 800 nm this is accomplished by reflecting the beam off of a series of *chirped mirrors* [93]. A chirped mirror is made by designing a dielectric stack with alternating

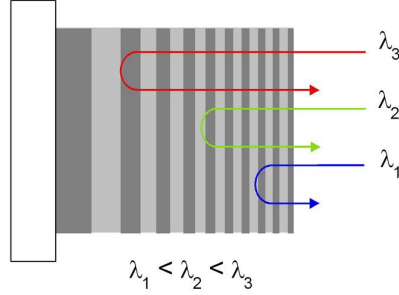


Figure 6.7: Example of a chirped mirror. Red frequency components propagate further into the substrate compared to blue components. This results in negative dispersion for a reflected pulse. (Figure from: [94])

layers of glasses that have different indices of refraction (Fig. 6.7). The separation between the layers makes the stack reflective at some wavelengths and transparent at others. In a chirped mirror this separation varies throughout the stack. The mirror is designed so that the leading (red), tail of the pulse travels deep into the mirror before reflecting back, whereas the trailing (blue), tail of the pulse is reflected close to the surface. After one or more bounces off of chirped mirrors, all of the wavelength components in the pulse emerge at the same time with a flat spectral phase and a pulse duration that is ideally only limited by the spectral bandwidth. The thickness of the individual layers can be carefully tailored to correct for the spectral phase from a particular laser system. The use of chirped mirrors has been very successful for pulse compression at 800 nm, and chirped mirrors are now commercially available from several manufactures.

When this thesis started, chirped mirrors in the infrared were not available. Through a collaboration with Femtolasers a set of mirrors were fabricated for use at $1.4\text{ }\mu\text{m}$, and were successfully used to compress a 1425 nm, 400 μJ pulse to a duration of 13 fs. The pulse compression results were published in Optics Letters [10] in a paper that is presented on p. 94.

We then compressed the $1.8\text{ }\mu\text{m}$ idler beam from the OPA. The original plan was to first demonstrate that we could compress at $1.4\text{ }\mu\text{m}$ and then design a set of chirped mirrors for use at $1.8\text{ }\mu\text{m}$. As it turns out, the anomalous dispersion of normal glasses such as fused silica can be used for compression at $1.8\text{ }\mu\text{m}$. When an 800 nm beam passes through fused silica it experiences normal dispersion. Its red frequency

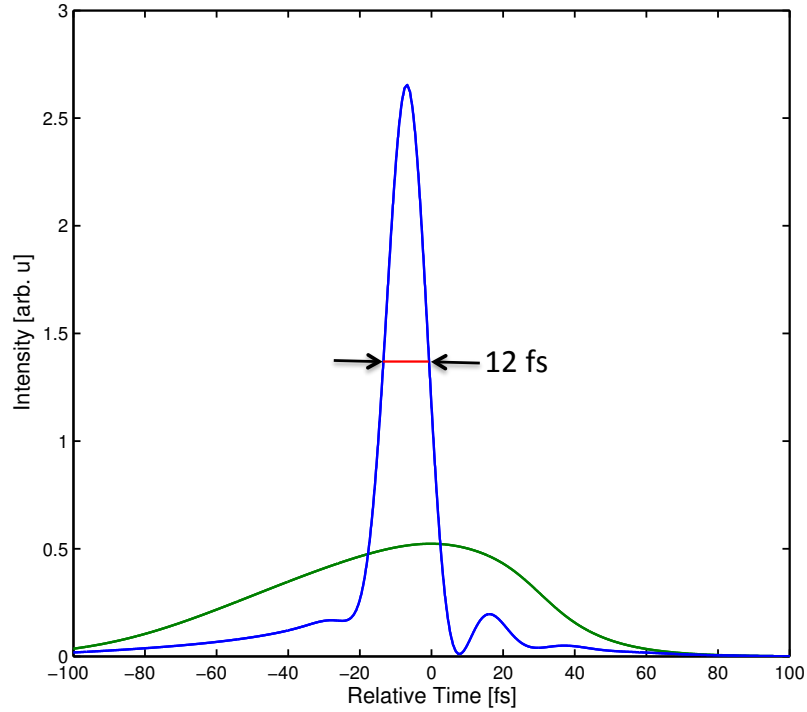


Figure 6.8: Pulse before and after linear propagation through 3.2 mm of fused silica. The duration goes from 83.3 fs to 12.0 fs. This model includes self-phase modulation, self-steepening and linear dispersion. (1 mJ, $1.83 \mu\text{m}$, $T_{\text{in}} = 80 \text{ fs}$, (E-field 1/e radius) = $128.7 \mu\text{m}$)

components travel faster than the blue ones and the pulse emerges positively chirped. At $1.8 \mu\text{m}$, fused silica exhibits anomalous dispersion and the situation is reversed with the blue end of the pulse emerging first. This has the very convenient property that it cancels the positive chirp introduced through SPM. Broadening the $1.8 \mu\text{m}$ pulse and then compressing it with linear propagation in 3 mm of fused silica resulted in a pulse duration of 11.5 fs (less than two optical cycles). This is demonstrated numerically by taking the simulated pulse at the exit of the fiber (Fig. 6.5), and numerically propagating it through 3 mm of fused silica. The temporal profile before and after the fused silica is shown in Fig. 6.8.

6.7 Pulse Compression: Applications

By broadening the infrared pulse from the OPA and then compressing it with chirped mirrors at $1.4\,\mu\text{m}$ and fused silica at $1.8\,\mu\text{m}$ we have developed a nearly ideal laser source for HHG. This has enabled us to significantly increase the HHG cutoff while maintaining a reasonable harmonic yield. Reducing the number of cycles in a laser pulse means that there are fewer opportunities to ionize the sample, thereby allowing for higher intensities to be reached before the sample is fully ionized. Figure 6.9 shows the calculated dependence of the sub-cycle ionization rate (red), on the laser electric field magnitude (blue), for xenon atoms exposed to 50 fs as well as 11 fs pulses. For the long pulse, ionization is concentrated around the most intense half cycles at low intensities (a), but moves to earlier times when the field strength increases (b), with the result that the sample is fully ionized prior to the peak of the pulse. By reducing the number of half-cycles higher peak intensities can be reached before the sample is fully ionized (c). This $1.8\,\mu\text{m}$ 11 fs pulse (< 2 optical cycles), pulse has been used to extend the cutoff from low IP systems such as xenon (Ch. 10). Figure 5 of the paper that starts on Page 119 shows HHG spectra in xenon for different laser wavelengths and pulse durations. The cutoff increases dramatically with increasing wavelength and decreasing pulse duration. By amplifying the OPA we have produced pulses with $> 1\,\text{mJ}$ at $1.8\,\mu\text{m}$ and 2-3 optical cycles. These were sufficient to generate high harmonics in neon with photon energies extending out to $> 400\,\text{eV}$, well in excess of the carbon K-edge at $283\,\text{eV}$ (Fig. 6.10).

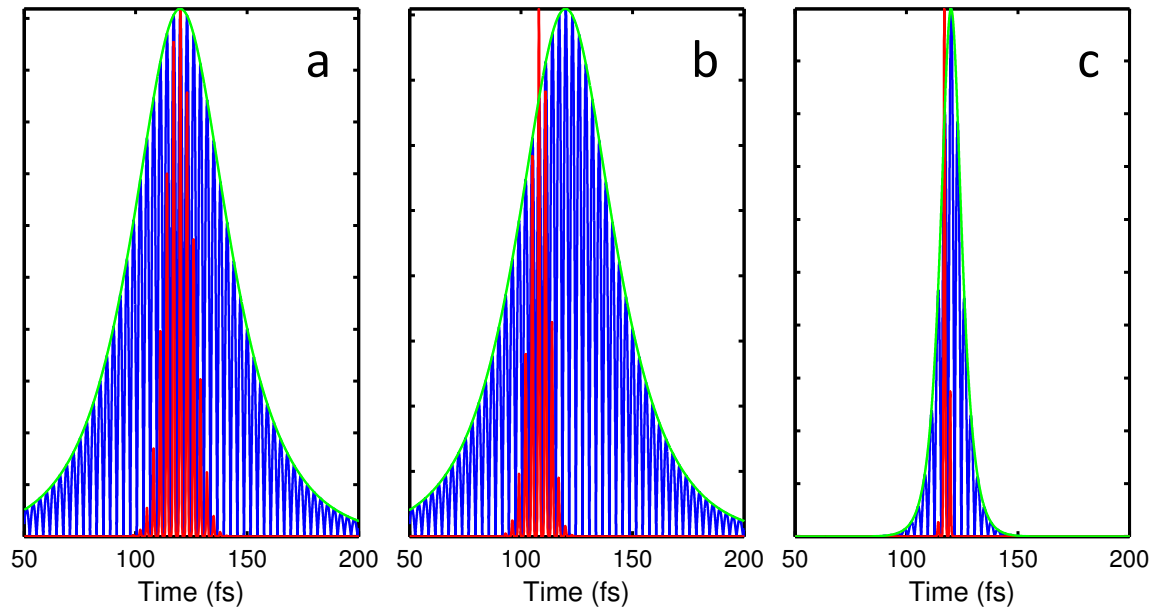


Figure 6.9: Xenon ionization rate (red), in a $1.8\mu\text{m}$ laser field shown with its absolute value in (blue), for different pulse durations and peak intensities. (a) Below the saturation intensity, ionization peaks near the maximum of the most intense half-cycles ($9.1 \cdot 10^{13}\text{W}/\text{cm}^2$; 50fs). (b) At higher intensity ($2.5 \cdot 10^{14}\text{W}/\text{cm}^2$; 50fs) ionization starts earlier and no neutral population survives to the peak of the pulse. (c) By using a shorter 11 fs pulse, there are fewer opportunities to ionize and a higher intensity can be used, with population surviving to the peak of the pulse ($3.5 \cdot 10^{14}\text{W}/\text{cm}^2$).

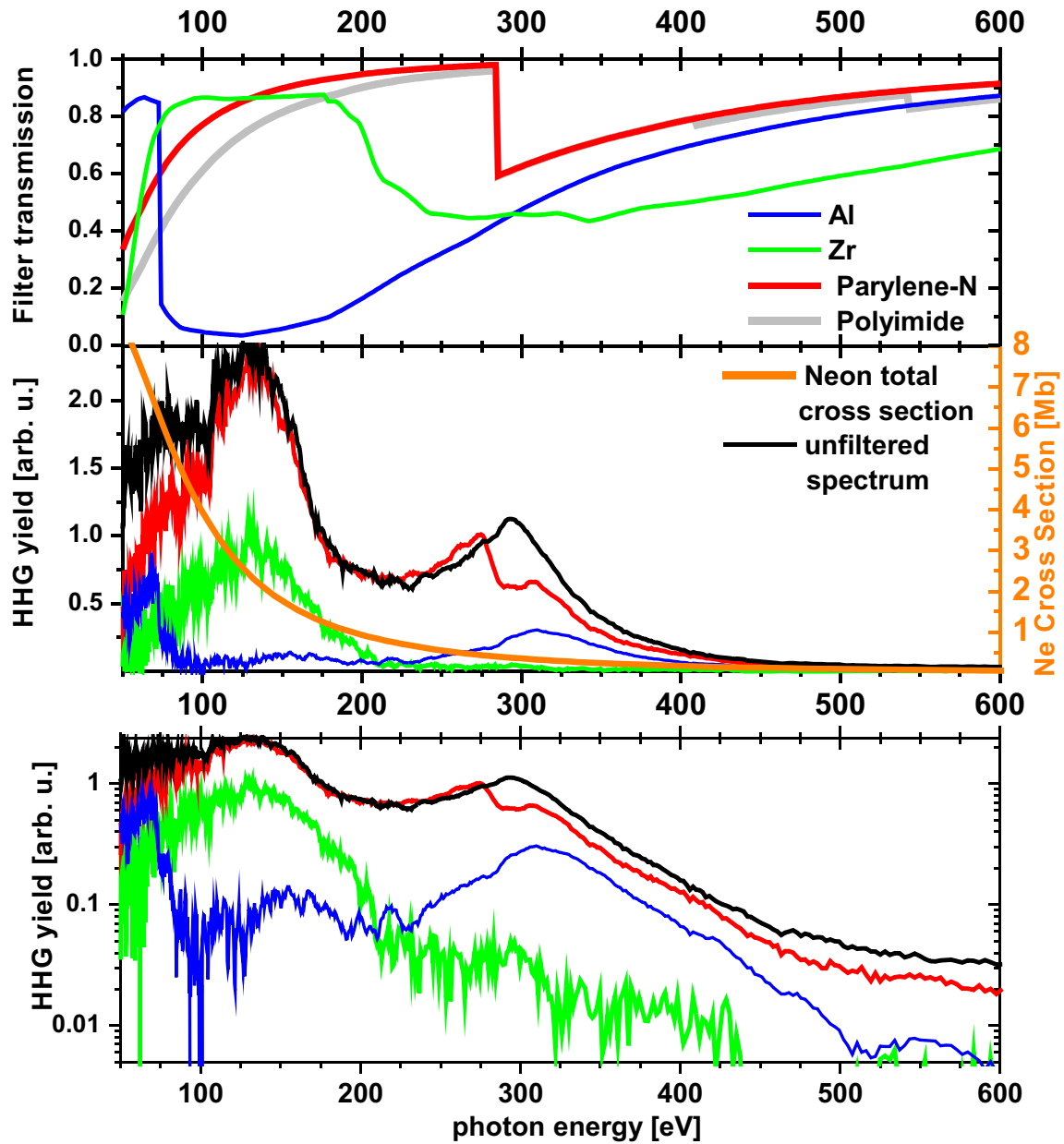


Figure 6.10: HHG spectra produced from neon driven with a $1.8\ \mu\text{m}$ ~ 2 cycle laser pulse. Various plastic and metal filters were inserted into the XUV beam to calibrate the HHG spectrum. The carbon edge at 283 eV is clearly visible and the spectrum extends out to 400 eV.

Pulse compression of submillijoule few-optical-cycle infrared laser pulses using chirped mirrors

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We report generation of 400 μJ , 13.1 fs, 1425 nm optical parametric amplifier laser pulses. Spectral broadening of a 100 Hz optical parametric amplifier laser source is achieved by self-phase modulation in an argon-filled hollow-core fiber, and dispersion compensation is performed using chirped mirrors. This laser source will be useful for ultrafast time-resolved molecular orbital tomography. © 2009 Optical Society of America
OCIS codes: 320.5520, 320.7090.

Generation of intense few-optical-cycle laser pulses at 800 nm has been the key technological breakthrough for attosecond science—high-harmonic generation (HHG) [1–4] and pump–probe molecular imaging [5,6]. At the moment, using carrier-envelope-phase (CEP)-stabilized 1.5 cycle Ti:Sa laser pulses and an interaction media with a large ionization potential (neon, $I_p=21$ eV), pulse duration of 80 attoseconds has been generated [7]. Since the cut-off of the harmonic spectra scales with $I\lambda^2$ (I is the laser intensity and λ the laser wavelength) [8], few-optical-cycle IR laser pulses are required to further decrease the duration of attosecond laser pulses [9]. Such development will also be highly beneficial for molecular orbital tomography, since shorter harmonic wavelength implies better spatial resolution [10].

For HHG, submillijoule of energy per laser pulse is required. At 800 nm, submillijoule few-optical-cycle laser pulses are commonly generated using the combination of self-phase modulation (SPM) through propagation of submillijoule multicycle pulses in a hollow-core fiber filled with a noble gas followed by dispersion compensation using chirped mirrors [11,12]. This is the standard experimental approach for dispersion compensation of 800 nm few-optical-cycle laser pulses and used in many laboratories worldwide. In the IR, chirped mirrors have been used for dispersion compensation in Cr:forsterite oscillators [13] but never to compress broadband laser pulses resulting from nonlinear propagation in a hollow-core fiber setup. Recently, we demonstrated generation of stable broadband IR spectra using this approach, in which we used an acousto-optic programmable dispersive filter for dispersion compensation obtaining 20 fs pulse duration at 1300 nm with 10 μJ of energy per pulse [14]. In this Letter, we demonstrate the generation and characterization of 400 μJ , 13.1 fs, 1425 nm laser pulses by using IR chirped mirrors for dispersion compensation. This approach is a robust method to generate intense IR few-optical-cycle laser pulses and can be easily inte-

grated with submillijoule femtosecond optical parametric amplifier (OPA) laser systems that are becoming widely available, and recently extended to sub-10 mJ level [15].

In the IR spectral range, few-optical-cycle laser pulses have been generated using other approaches that are generally more complex than the one proposed in this Letter: (1) 740 μJ , 15.6 fs at 2.1 μm using an optical parametric chirped-pulse amplifier [16]; (2) 270 μJ , 17 fs at 2.1 μm using pulse compression by filamentation [17]; and (3) 1.2 mJ, 17 fs at 1.5 μm using difference-frequency generation of CEP-stabilized few-optical-cycle 800 nm laser pulses followed by type II parametric amplification [18].

The experiment was conducted at the Advanced Laser Light Source using the 100 Hz, 100 mJ, 35 fs Ti:Sa laser system (Thales Laser). Using 7 mJ of Ti:Sa, 1.2 mJ, 50 fs, 1425 nm signal laser pulses are produced using an OPA based on a seed generated by parametric superfluorescence (HE-TOPAS, Light Conversion) with pulse to pulse stability of 2%–3% rms. At the output of the OPA, the pulse energy is attenuated using the combination of an achromatic half-wave plate with Glan-Calcite polarizer. The 1425 nm laser beam is coupled into the hollow-core fiber compressor using an $f=1$ m plano-convex lens. The hollow-core fiber is supported on an aluminum V-groove and attached on both ends to closed gas cells. The hollow-core fiber is evacuated and filled with argon by maintaining 1.7 Atm of pressure in both cells. At the output, the laser beam was collimated using a 1 m concave silver mirror. Typically, the optical transmission was near 40% owing to the poor spatial quality of the OPA laser beam.

Figure 1 shows typical spectra after the fiber for low (30 μJ , dashed-dotted curve) and high (400 μJ , solid curve) output energies (measured with NIR256 spectrometer from Ocean Optics, Inc.). At low energy, SPM is not observed, and the OPA signal spectra are unchanged upon propagation in the hollow-core fiber. For high energy, we observed spectral broadening owing to nonlinear propagation predominantly causing

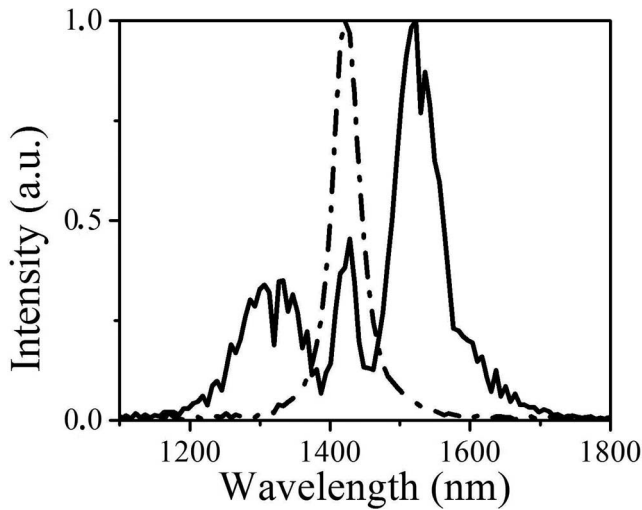


Fig. 1. OPA spectra after propagation into the argon-filled hollow-core fiber as a function of output energy. Dashed-dotted curve, 30 μJ ; solid curve, broadened spectrum with 400 μJ corresponding to the 13.1 fs pulse of Fig. 3(b).

SPM. Assuming Fourier transform-limited pulse duration for the broadened spectra, pulse duration at the FWHM of the intensity profile is 12.2 fs. At 1425 nm, this is equivalent, in terms of the number of cycles, to 7 fs pulse duration at 800 nm.

To first approximation, the phase accumulated by SPM is linear and positive in the time domain corresponding to a positive group-delay dispersion (GDD) in the spectral domain [11,12]. Prism-based negative delay lines introduce significant higher-order dispersion [mainly third-order dispersion (TOD)] being thus unsuitable for few-optical-cycle pulse compression. Consequently, dispersive mirrors were designed to introduce negative GDD and small, negative TOD to compensate for the substantially quadratic chirp carried by the pulses as well as for the positive TOD of glasses eventually used in the setup. We designed and manufactured a pair of dispersive mirrors exhibiting high reflectance ($R > 99.5\%$) and controlled dispersion (GDD = -150 fs^2 and TOD = -15 fs^3 at 1425 nm) in the wavelength range of 1000 nm to 1700 nm. Each mirror consists of 73 layers. SiO_2 and Nb_2O_5 were used as coating materials. The large absolute value of the GDD is a consequence of the increased multilayer thickness (as compared with similar mirrors for the 800 nm range). Decreasing the number of layers and therewith the overall thickness would result in smaller absolute GDD values at the expense of reduced bandwidth and/or lower reflectance. These design constraints can be overcome by employing materials with a higher index ratio, but the deposition of these materials does not fulfill the necessary thickness accuracy standards yet (to our knowledge). Large GDD (absolute) values per bounce are, however, not a drawback, since they result in a very compact, high-throughput compressor. Fine tuning of the GDD, if necessary, can be achieved with thin positive dispersion glass (e.g., SF10) wedges.

Temporal characterization of the laser pulses was performed using a home-build second-harmonic-

generation frequency-resolved optical gating (SHG-FROG) [19]. Since phase matching is extremely critical for few-optical-cycle laser pulse measurements, SHG was generated by recombining both FROG optical arms focused using a 50 cm focal length convex silver mirror inside a 10 μm thickness Type I β -barium borate (BBO) crystal ($\theta \approx 21^\circ$).

Figure 2 shows, on a linear scale, an experimental (a) and a retrieved (b) SHG-FROG spectrogram for the shortest laser pulse duration we obtained. Dispersion compensation has been achieved by adding one round trip on the chirped mirrors with $\sim 1.5 \text{ mm}$ of SF10. In Fig. 3(a), we present the intensity autocorrelation associated with the experimental SHG-FROG traces. The black curve corresponds to the shortest pulse duration, whose FWHM of the autocorrelation is 18.4 fs, corresponding to pulse duration of 13.0 fs, assuming Gaussian intensity profile. In Fig. 3(b), we are showing the retrieved temporal profile of the laser pulses obtained using the SHG-FROG reconstruction. The fact that the obtained FWHM duration of 13.1 fs is only 1.07 times the Fourier limit (12.2 fs assuming flat spectral phase) proves the good compressibility of the SPM broadened spectra at 1425 nm. The intensity profile is asymmetric with a flat temporal phase covering the main pulse, shown as dashed-dotted curve in Fig. 3(b). Pedestals might origin from uncompensated high-order phase. At 1425 nm, 13.1 fs is less than three optical cycles, equivalent to about 7.5 fs pulse duration at 800 nm. Moreover, we have calculated that 70% of the energy is concentrated in the central part of the laser pulse temporal intensity profile. We have determined the direction of the time axis by overcompensating the dispersion with one round trip on the chirped mirrors and added positive dispersion with SF10 windows.

For comparison with the shortest pulse, a cleaner temporal pulse shape can be obtained by reducing

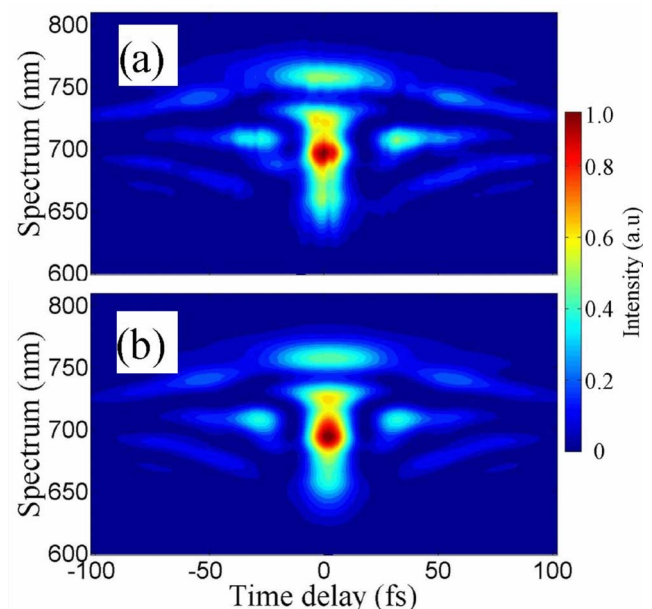


Fig. 2. (Color online) (a) Experimentally measured and (b) retrieved SHG-FROG traces for the compressed 13.1 fs laser pulse of Fig. 3(b).

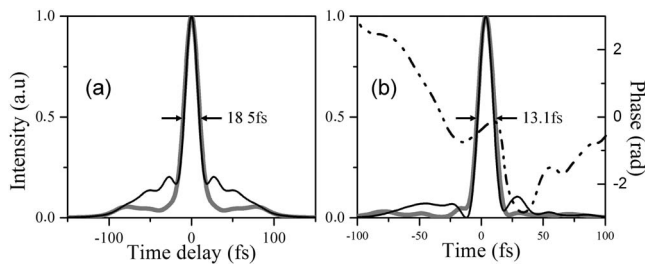


Fig. 3. (a) SHG intensity autocorrelation and (b) retrieved intensity profile and temporal phase obtained from the SHG-FROG reconstruction. The gray curves correspond to the temporally clean 14.5 fs pulse, whereas the black curves show results for the shortest FWHM duration of 13.1 fs.

propagation nonlinearity at reduced argon pressure of 1.4 atm (gray curves in Fig. 3). The retrieved FWHM of intensity is 14.5 fs, whereby the main pulse carries more than 86% of the total energy. This clean pulse was used to experimentally prove the chirped mirror design by measuring an SHG-FROG trace with removed chirped mirrors in the setup. In this case the retrieved spectral phase for the temporally broadened pulse exhibits mainly quadratic behavior (data not shown). From this the second-order dispersion could be determined to be about -330 fs^2 , which is in very good agreement with calculated value of -300 fs^2 for one round trip.

In this Letter, we have shown that hollow-core fiber pulse compression techniques used at 800 nm can be transferred in the IR. We have generated and characterized 400 μJ , 13.1 fs laser pulses at 1425 nm. This is equivalent in terms of number of cycles to 7.5 fs pulse duration at 800 nm. By using a more powerful OPA input [15] and a differentially pumped fiber [12], it should be possible to scale up the output energy. The present pulse duration is limited by the spectral broadening in the fiber owing to the relatively long pulse duration of the OPA, not by the bandwidth of the chirped mirrors. However, the compression of ultrabroadband spectra requires precise dispersion control of TOD by accordingly designed chirped mirrors. At the moment, our few-optical-cycle IR laser pulses are not CEP stabilized. It will require using a CEP-stabilized Ti:Sa amplifier and using an OPA based on signal seed generated by white light continuum. As clearly explained by Corkum and Krausz [9], generation of CEP-stabilized few-optical-cycle laser pulses in the IR will be highly beneficial for the generation of shorter attosecond laser pulses, to perform high temporal resolution time-resolved molecular orbital tomography combined with high spatial resolution, and for studying recollision physics using electrons with higher energy than what can be provided using Ti:Sa laser technology.

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Foundation for Innovation, the Canadian Institute for Photonic Innovations, the Natural Sciences and Engineering Research Council of Canada, and the Fonds Québécois de la Recherche sur la Nature et les Technologies.

References

1. M. Hentschel, R. Kienberger, Ch. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. B. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, *Nature* **414**, 509 (2001).
2. R. Kienberger, E. Goulielmakis, M. Uiberacker, A. Baltuska, V. Yakovlev, F. Bammer, A. Scrinzi, Th. Westerwalbesloh, U. Kleineberg, U. Heinzmann, M. Drescher, and F. Krausz, *Nature* **427**, 817 (2004).
3. I. J. Sola, E. Mével, L. Elouga, E. Constant, V. Strelkov, L. Poletto, P. Villoresi, E. Benedetti, J.-P. Caumes, S. Stagira, C. Vozzi, G. Sansone, and M. Nisoli, *Nat. Phys.* **2**, 319 (2006).
4. G. Sansone, E. Benedetti, F. Calegari, C. Vozzi, L. Avaldi, R. Flammini, L. Poletto, P. Villoresi, C. Altucci, R. Vellota, S. Stagira, S. De Silvestri, and M. Nisoli, *Science* **314**, 443 (2006).
5. F. Légaré, K. F. Lee, I. V. Litvinyuk, P. W. Dooley, A. D. Bandrauk, D. M. Villeneuve, and P. B. Corkum, *Phys. Rev. A* **72**, 052717 (2005).
6. A. Hishikawa, A. Matsuda, M. Fushitani, and E. J. Takahashi, *Phys. Rev. Lett.* **99**, 258302 (2007).
7. E. Goulielmakis, M. Schultze, M. Hofstetter, V. S. Yakovlev, J. Gagnon, M. Uiberacker, A. L. Aquila, E. M. Gullikson, D. T. Attwood, R. Kienberger, F. Krausz, and U. Kleineberg, *Science* **320**, 1614 (2008).
8. P. B. Corkum, *Phys. Rev. Lett.* **71**, 1994 (1993).
9. P. B. Corkum and F. Krausz, *Nat. Phys.* **3**, 381 (2007).
10. J. Itatani, J. Lévesque, D. Zeidler, H. Niikura, H. Pépin, J. C. Kieffer, P. B. Corkum, and D. M. Villeneuve, *Nature* **432**, 867 (2004).
11. S. Sartania, Z. Cheng, M. Lenzner, G. Tempea, Ch. Spielmann, F. Krausz, and K. Ferencz, *Opt. Lett.* **22**, 1562 (1997).
12. A. Suda, M. Hatayama, K. Nagasaka, and K. Midorikawa, *Appl. Phys. Lett.* **86**, 111116 (2005).
13. C. Chudoba, J. G. Fujimoto, E. P. Ippen, H. A. Haus, U. Morgner, F. X. Kärtner, V. Scheuer, G. Angelow, and T. Tschudi, *Opt. Lett.* **26**, 292 (2001).
14. F. Légaré, M. Naji, P. Lassonde, D. Comtois, V. Crozatier, T. Oksenhendler, H. Anis, and J. Kieffer, *Opt. Lett.* **33**, 2824 (2008).
15. E. J. Takahashi, T. Kanai, Y. Nabekawa, and K. Midorikawa, *Appl. Phys. Lett.* **93**, 041111 (2008).
16. X. Gu, G. Marcus, Y. Deng, T. Metzger, C. Teisset, N. Ishii, T. Fuji, A. Baltuska, R. Butkus, V. Pervak, H. Ishizuki, T. Taira, T. Kobayashi, R. Kienberger, and F. Krausz, *Opt. Express* **17**, 62 (2009).
17. C. P. Hauri, R. B. Lopez-Martens, C. I. Blaga, K. D. Schultz, J. Cryan, R. Chirila, P. Colosimo, G. Doumy, A. M. March, C. Roedig, E. Sistrunk, J. Tate, J. Wheeler, L. F. DiMauro, and E. P. Power, *Opt. Lett.* **32**, 868 (2007).
18. C. Vozzi, F. Calegari, E. Benedetti, S. Gasilov, G. Sansone, G. Cerullo, M. Nisoli, S. De Silvestri, and S. Stagira, *Opt. Lett.* **32**, 2957 (2007).
19. R. Trebino, K. W. DeLong, D. N. Fittinghoff, J. N. Sweetser, M. A. Krumbügel, B. A. Richman, and D. J. Kane, *Rev. Sci. Instrum.* **68**, 3277 (1997).

Compression of 1.8 μm laser pulses to sub two optical cycles with bulk material

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We demonstrate a simple scheme to generate 0.4 mJ 11.5 fs laser pulses at 1.8 μm . Optical parametrically amplified pulses are spectrally broadened by nonlinear propagation in an argon-filled hollow-core fiber and subsequently compressed to 1.9 optical cycles by linear propagation through bulk material in the anomalous dispersion regime. This pulse compression scheme is confirmed through numerical simulations. © 2010 American Institute of Physics. [doi:10.1063/1.3359458]

In the past decade, attosecond technology based on high harmonic generation has been developed permitting ultrafast measurements with ~ 100 as temporal resolution.^{1–3} Further reduction in the duration of isolated attosecond pulses and higher photon energies requires intense longer-wavelength carrier envelope phase (CEP) stable few-cycle pulses. Those requirements can be fulfilled using the Idler of a white light seeded optical parametric amplifier (OPA)⁴ with an appropriate pulse compression scheme. In this letter, we demonstrate a robust approach for pulse compression at 1.8 μm to below two optical cycle duration.

Different approaches to generate intense IR few-cycle laser pulses have been demonstrated as follows: (1) 0.74 mJ 15.6 fs at 2.1 μm using an optical parametric chirped-pulse amplifier (OPCPA),⁵ (2) pulse self-compression by filamentation; 0.27 mJ 17.9 fs at 2.1 μm ⁶ and 1.5 mJ 19.8 fs at 1.5 μm ,⁷ (3) 1.2 mJ 17 fs at 1.5 μm utilizing difference frequency generation of few-optical-cycle 800 nm laser pulses followed by type II parametric amplification,⁸ and (4) 0.4 mJ 13.1 fs at 1.4 μm using spectral broadening in a hollow-core fiber (HCF) and dispersion compensation with chirped mirrors.⁹

In this letter, we demonstrate the generation of 0.4 mJ 11.5 fs laser pulses at 1.8 μm . Similar to our recent work using the OPA Signal wavelength,⁹ we spectrally broaden the Idler via nonlinear propagation in a HCF. Instead of chirped mirrors or adaptive devices for dispersion compensation, we show that laser pulses can be efficiently compressed utilizing solely the properties of fused silica (FS) in the anomalous dispersion regime below the third order dispersion (TOD) limit of bulk material. Although compression with glass in the anomalous dispersion regime was applied to mid-IR wavelengths (~ 6 μm),¹⁰ the authors discussed the limitation of bulk material compression to be applicable to multi-cycle pulses only.

The present approach distinguishes itself from previous setups in the reduced complexity, as shown in the sketch of the experimental layout in Fig. 1(a). The IR laser pulses are generated with a superfluorescence-seeded three stage OPA (HE-TOPAS, Light conversion) pumped by 7 mJ, 45 fs Ti:Sa pulses. A conversion efficiency of 35% (Signal+Idler) with 3% intensity fluctuation is typically achieved at the OPA output. At 1.83 μm , the spectral full width at half maximum (FWHM) is 67 nm corresponding to a TL of 73 fs. This OPA spectrum, shown as shaded gray in Fig. 1(b), is measured with an Ocean Optics NIR 256 spectrometer corrected for spectral sensitivity. The IR laser beam is coupled into a 1 m long HCF (400 μm in diameter, argon pressure of 140 ± 10 kPa, 1 mm CaF₂ windows) using a $f=1$ m plano-convex CaF₂ lens. At the output, the laser beam is collimated

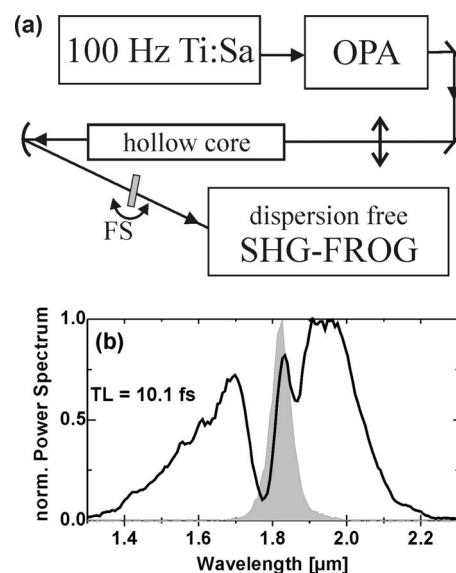


FIG. 1. (a) Experimental layout comprising a CPA pumped high energy OPA whose spectrum is broadened via propagation in a HCF. (b) Experimental spectra of the OPA Idler before (shaded gray) and after broadening (solid black). An asymmetric broadening towards the blue spectral side is visible.

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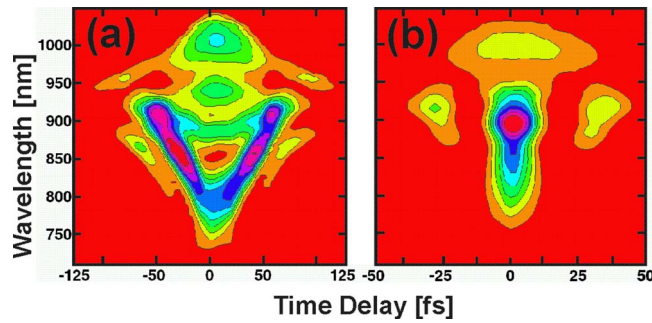


FIG. 2. (Color online) Experimental SHG-FROG traces for the uncompressed pulse after propagation in the HCF (a) corresponds to a pulse duration of 68 fs. Propagation through 3 mm of FS leads to a compressed 11.5 fs pulse depicted in (b) on a different time scale.

with an $R=2$ m concave silver mirror. As shown by the solid line of Fig. 1(b), with 0.89 mJ at the input of the HCF, significant spectral broadening is observed corresponding to a TL of 10.1 fs. This spectrum clearly shows an asymmetry with wider extension towards shorter wavelengths.

Complete pulse characterization is carried out with a home built SHG-FROG (second harmonic generation frequency resolved optical grating).¹¹ Beam splitting is attained by geometrical beam separation to achieve ultrabroadband and dispersion free operation.¹² A high degree of phase matching and negligible geometrical temporal smear¹³ is obtained by achromatically focusing both optical arms with a 500 mm focal length convex silver mirror into a Type I BBO crystal ($\theta=21^\circ$) of 10 μm thickness. The SHG-FROG spectrograms are measured with a USB2000 spectrometer from Ocean Optics and corrected for spectral sensitivity. In addition, spectral distortion of the ultrabroadband SHG is accounted for by applying a cubic correction function to the measured SHG-FROG spectrogram.¹³

After spectral broadening in the HCF, the pulse must be temporally compressed. At 800 nm, this is commonly carried out with chirped mirrors which are not available at 1.8 μm . Fortunately, the unique characteristics of the 1.8 μm source mean that the negative group delay dispersion (GDD) of FS is able to compensate for the spectral phase introduced by self-phase modulation (SPM) in argon. Because the zero dispersion of FS lies at ~ 1.3 μm , it exhibits negative GDD throughout the entire spectral range of interest. Once the appropriate FS thickness is found, a small angular tilt of the glass is sufficient to obtain the shortest pulse duration. In this manner a compression factor of 6.5 is achieved compared to the OPA output pulses.

The pulse duration immediately after the evacuated fiber was measured to be 75 fs [Fig. 3(c), gray line] in agreement with the TL (73 fs) of the OPA spectra presented in Fig. 1(b) as shaded gray. Inserting argon leads to significant broadening and the corresponding SHG-FROG traces of the uncompressed and fully compressed pulses are shown in Fig. 2. Surprisingly, linear propagation through a simple piece of FS is sufficient to achieve excellent compression. Figure 3 presents the reconstructed electric field in the spectral (a, b) and the temporal domains (c). The direction of the time axis was determined by comparing the spectral phase with and without the 3 mm FS. The asymmetric shape of the reconstructed power spectrum [solid black line in Fig. 3(a)] is in reasonable agreement with the measured one given by the black squares. The spectral phase for the compressed pulse [black

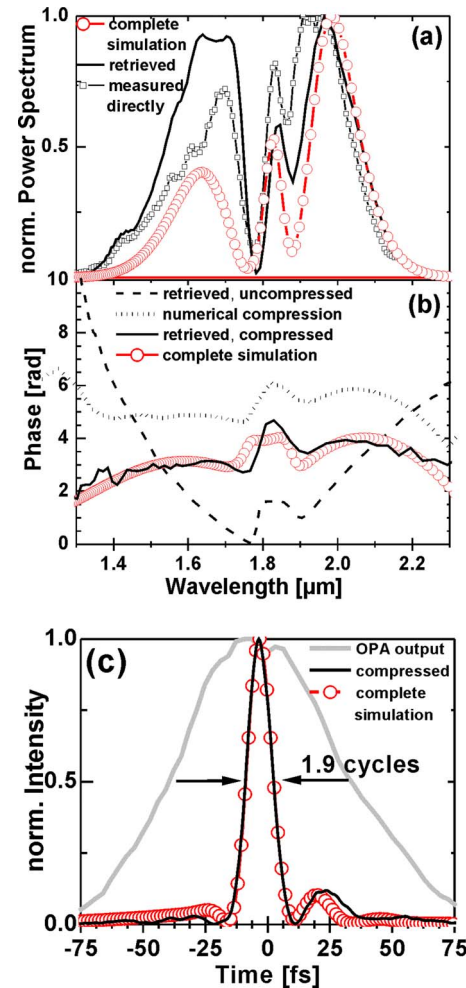


FIG. 3. (Color online) (a) Power spectral densities after nonlinear propagation in the HCF. (b) SHG-FROG retrieved spectral phases for uncompressed (black dashed line) and compressed pulses (black solid line) are compared with full numerical simulations (circles). The spectral phase named numerical compression (black dotted line) has been obtained by numerically propagating the retrieval of the uncompressed pulse (dashed line) through 3 mm of FS. (c) Temporal intensities corresponding to the spectra given above whereby compression down to 11.5 fs is achieved with 3 mm FS after the HCF.

solid line in Fig. 3(b)] is significantly flattened with respect to the uncompressed phase (black dashed line). In the case of pure SPM, one would expect symmetric broadening of the power spectrum around the fundamental accompanied by a symmetric phase function. However, the uncompensated phase [black dashed line] is slightly steeper on the red side compared with the blue side. This spectral asymmetry of the uncompensated pulse corresponds to a temporal asymmetry whereby the pulse exhibits a much steeper trailing edge than the leading one. Linear propagating this pulse through 3 mm of FS generates a very short 11.5 fs pulse [solid black line in Fig. 3(c)]. The fairly clean compressed pulse contains 78% of total energy in the main part and the FWHM duration is only 1.14 times the TL duration of 10.1 fs. Residual higher order phases which are not compensated by the bulk compressor are likely to cause the small pedestals on both sides of the main pulse. We mention that in addition to the rather clean temporal shape on the femtosecond time scale, the present approach is free of superfluorescence background as was reported in case of OPCPA.⁵ When the seed is blocked in the OPA, no light is observed at the output of the fiber.

It is pointed out that this almost TL and relatively clean temporal shape is remarkable because it denotes dispersion compensation not only of the GDD but also of the TOD to a large extent. Most of the commonly used glasses, like FS, exhibit normal dispersion ($GDD > 0$) below $1\ \mu\text{m}$ and anomalous dispersion ($GDD < 0$) beyond the zero dispersion wavelength centered between $1.2\text{--}2\ \mu\text{m}$. Thus bulk material is potentially suitable for pulse compression of IR laser pulses. However, all glasses exhibit positive TOD throughout their entire transmission range. Uncompensated TOD causes any femtosecond pulse to deviate from the TL pulse shape even if the GDD is zero. For instance, assuming a Gaussian shape at center wavelength of $1.83\ \mu\text{m}$, a TOD of only $1000\ \text{fs}^3$ (3 mm FS plus 1 mm CaF_2) causes a TL pulse to broaden from 10.1 to 14.7 fs. To confirm that linear propagation through bulk material (intensity $\approx 5 \times 10^9\ \text{W}/\text{cm}^2$) compresses the pulse after the HCF, we calculated the spectral phase introduced by 3 mm of FS according to the Sellmeier equation. This phase was then added to the retrieved phase of the uncompressed pulse [black dashed curve in Fig. 3(b)]. The result [black dotted line in Fig. 3(b)] matches the retrieved phase of the compressed pulse. On the one hand, this numerical cross check proves the compression mechanism due to linear propagation in FS. On the other hand, it also demonstrates the reliability of the SHG-FROG retrieval though the retrieved power spectrum shows a lower peak on the blue side when compared to the directly measured spectrum. However, by comparing the retrieved spectral phase of compressed and uncompressed pulse we obtain the exact refractive index of FS in the range from 1.4 to $2.2\ \mu\text{m}$. Now the question about the origin of the negative TOD component arises. To address this question, two possible explanations are discussed.

On the one hand, our experimental results might be viewed in the context of pulse self-compression. This explanation can be ruled out since we have to add 3 mm of FS subsequent to the HCF setup to obtain the shortest pulse. At $1.8\ \mu\text{m}$, FS provides significant negative GDD ($-68\ \text{fs}^2/\text{mm}$). Furthermore, in contrast to experimentally observed self-compression,^{6,7,14,15} the laser power of 10 GW in our experiment is roughly two times below the estimated critical power for self-focusing.

On the other hand, the pronounced spectral asymmetry suggests that self-steepening^{15–17} of the pulse takes place in addition to SPM. This temporal reshaping of the trailing edge then leads to an asymmetric spectral phase opposed to the positive TOD of bulk material. To prove this qualitative explanation, we performed numerical simulations. The nonlinear Schrödinger equation¹⁸ was solved in one dimension where the action of self-steepening could be turned on and off numerically. Numerical results based on exact experimental conditions like input pulse duration, pulse energy, and pressure are given by the circles in Fig. 3 and a detailed description is in preparation for a longer article.¹⁹ Briefly, if self-steepening is neglected, it was not possible to simulate an asymmetric power spectrum as shown in Fig. 3(a). Contrary, the symmetry of the spectrum is broken because of self-steepening which is known to promote the higher frequency region of the spectrum.¹⁵ It furthermore enables calculating an asymmetric phase like the dashed curve of Fig. 3(b). Its linear propagation through 3 mm of FS leads to the plot shown by the circles in Fig. 3(b) which is in excellent

agreement with the experimentally retrieved phase [solid black line]. The corresponding temporal intensity [circles in Fig. 3(c)] closely resembles the experimentally retrieved pulse. Neglecting self-steepening leads to a pulse duration of 14.4 fs with an increased pedestal.

In conclusion, we have demonstrated a very simple and robust approach for the generation of submillijoule two-cycle $1.8\ \mu\text{m}$ laser pulses via nonlinear propagation in a HCF followed by dispersion compensation utilizing the anomalous dispersion of bulk material. This straight forward approach was confirmed by numerical simulations that demonstrate the action of self-steepening is to generate a spectral phase opposite to that of FS. At the moment, our sub two-cycle laser pulses are not CEP stabilized, but this is feasible using the Idler of a white light seeded OPA.⁴

We are very grateful for the help and the time spent on the laser system by François Poitras and Antoine Laramée. The authors acknowledge the support of the Canada Foundation for Innovation, the Canadian Institute for Photonic Innovations, the Natural Sciences and Engineering Research Council of Canada, and the Fonds Québécois de la Recherche sur la Nature et les Technologies.

- ¹M. Hentschel, R. Kienberger, Ch. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. B. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, *Nature (London)* **414**, 509 (2001).
- ²G. Sansone, E. Benedetti, F. Calegari, C. Vozzi, L. Avaldi, R. Flammini, L. Poletto, P. Villoresi, C. Altucci, R. Vellota, S. Stagira, S. De Silvestri, and M. Nisoli, *Science* **314**, 443 (2006).
- ³E. Goulielmakis, M. Schultze, M. Hofstetter, V. S. Yakovlev, J. Gagnon, M. Uiberacker, A. L. Aquila, E. M. Gullikson, D. T. Attwood, R. Kienberger, F. Krausz, and U. Kleineberg, *Science* **320**, 1614 (2008).
- ⁴A. Baltuška, T. Fuji, and T. Kobayashi, *Phys. Rev. Lett.* **88**, 133901 (2002).
- ⁵X. Gu, G. Marcus, Y. Deng, T. Metzger, C. Teisset, N. Ishii, T. Fuji, A. Baltuška, R. Butkus, V. Pervak, H. Ishizuki, T. Taira, T. Kobayashi, R. Kienberger, and F. Krausz, *Opt. Express* **17**, 62 (2009).
- ⁶C. P. Hauri, R. B. Lopez-Martens, C. I. Baga, K. D. Schultz, J. Cryan, R. Chirila, P. Colosimo, G. Doumy, A. M. March, C. Roedig, E. Sistrunk, J. Tate, J. Wheeler, L. F. Di Mauro, and E. P. Power, *Opt. Lett.* **32**, 868 (2007).
- ⁷O. D. Mücke, S. Ališauskas, A. J. Verhoef, A. Pugžlys, A. Baltuška, V. Smilgevičius, J. Pocius, L. Giniūnas, R. Danielius, and N. Forget, *Opt. Lett.* **34**, 2498 (2009).
- ⁸C. Vozzi, F. Calegari, E. Benedetti, S. Gasilov, G. Sansone, G. Cerullo, M. Nisoli, S. De Silvestri, and S. Stagira, *Opt. Lett.* **32**, 2957 (2007).
- ⁹M. Giguère, B. E. Schmidt, A. D. Shiner, M.-A. Houle, H.-C. Bandulet, G. Tempea, D. M. Villeneuve, J.-C. Kieffer, and F. Légaré, *Opt. Lett.* **34**, 1894 (2009).
- ¹⁰N. Demirdöven, M. Khalil, O. Golonzka, and A. Tokmakoff, *Opt. Lett.* **27**, 433 (2002).
- ¹¹R. Trebino, K. W. DeLong, D. N. Fittinghoff, J. N. Sweetser, M. A. Krumhölz, B. A. Richman, and D. J. Kane, *Rev. Sci. Instrum.* **68**, 3277 (1997).
- ¹²I. Z. Kozma, P. Baum, U. Schmidhammer, S. Lochbrunner, and E. Riedle, *Rev. Sci. Instrum.* **75**, 2323 (2004).
- ¹³A. Baltuška, M. S. Pshenichnikov, and D. A. Wiersma, *Opt. Lett.* **23**, 1474 (1998).
- ¹⁴N. L. Wagner, E. A. Gibson, T. Popmintchev, I. P. Christov, M. M. Murnane, and H. C. Kapteyn, *Phys. Rev. Lett.* **93**, 173902 (2004).
- ¹⁵G. Yang and Y. R. Shen, *Opt. Lett.* **9**, 510 (1984); G. Stibenz, N. Zhavoronkov, and G. Steinmeyer, *Opt. Lett.* **31**, 274 (2006).
- ¹⁶N. Aközbek, M. Scalora, C. M. Bowden, and S. L. Chin, *Opt. Commun.* **191**, 353 (2001).
- ¹⁷A. L. Gaeta, *Phys. Rev. Lett.* **84**, 3582 (2000).
- ¹⁸G. P. Agrawal, *Nonlinear Fiber Optics*, 3rd ed. (Academic, San Diego (2001)).
- ¹⁹P. Béjot, B. E. Schmidt, J. Kasparian, J.-P. Wolf, P. B. Corkum, and F. Légaré, "IR pulse compression with bulk material: Experiments versus simulations," (unpublished).

Chapter 7

Carrier-Envelope Phase Control

This chapter is based on the following papers:

1. B. E. Schmidt, **A. D. Shiner**, P. Lassonde, J.-C. Kieffer, P. B. Corkum, D. M. Villeneuve, and F. Légaré, “CEP stable 1.6 cycle laser pulses at $1.8\ \mu\text{m}$,” *Optics Express*, vol. 19, no. 7, pp. 6858–6864, 2011
2. B. E. Schmidt, **A. D. Shiner**, M. Giguère, P. Lassonde, C. Trallero-Herrero, J.-C. Kieffer, P. B. Corkum, D. M. Villeneuve, and F. Légaré, “High harmonic generation with long wavelength few-cycle laser pulses,” *Journal of Physics B: Atomic, Molecular and Optical Physics*, vol. 45, no. 7, p. 074008, 2012

7.1 Introduction to CEP Control

Figure 6.1 demonstrated that changing the frequency-independent term in the spectral phase of a pulse shifts the carrier of the pulse in time relative to the pulse envelope. For very short (2-cycle) pulses the Carrier-Envelope Phase (CEP) can be used to direct the most intense half cycle of the laser electric field in either the positive or negative direction, or the CEP can be continuously varied between these extremes, with some CEP values associated with two symmetric half cycles. Carrier-envelope control is important for a number of processes, such as attosecond pulse generation [95, 96, 97], control over electron re-scattering, as well as for steering electron motion and localization during chemical processes such as D_2^+ dissociation [98]. Huge effort was directed toward the development of CEP stable femtosecond oscillators during the late 20th century, the successful development of which was cited in the 2005 Nobel Prize in Physics that was shared by John Hall and Theodor Hänsch. Stabilized femtosecond

oscillators are now recognized as the standard tool for relating optical frequencies in the world's national metrology laboratories [99]. Techniques were then developed to amplify the phase-stable output from an oscillator in order to produce mJ level phase-stable pulses at 800 nm [100].

At ALLS we have developed the white light (WL), seeded OPA as a source for high power, CEP stable, sub-2-cycle pulses at $1.8\mu\text{m}$. It was known for some time that WL seeded OPAs can exhibit passive CEP stability [101, 102]. However, using a CEP stable pulse to control a physical process, such as ionization, requires a pulse that is short enough for there to be significant asymmetry between the positive and negative field directions. The development of pulse compression technology at OPA wavelengths, and particularly at $1.8\mu\text{m}$, has triggered renewed interest in developing OPAs as CEP stable sources.

7.2 Origin of Passive CEP Stability

In a typical 800 nm Ti:Sapphire laser, the CEP is actively stabilized by electronic circuits, which take measurements of the laser repetition rate and carrier-envelope phase offset, and apply corrections to actuators inside the laser, in order to drive these parameters to desired set point values. Maintaining stable pulses during amplification, particularly when the pulses are spread out across the gratings in the amplifier's stretcher/compressor, places stringent demands on the mechanical stability of the system.

In a WL seeded OPA the idler pulses are intrinsically CEP stable, regardless of the stability of the pump laser. Following [102], this can be seen by first letting φ_j represent the CEP of a pulse where $j = \{s, p, i, w4\}$ for the signal, pump, idler, and white light generation seed pulses respectively. The seed is generated through self-phase modulation in a sapphire plate, which is a coherent process that maintains the same (possibly random), CEP as the pump laser, to within an additive constant [103].

$$\varphi_{w4} = \varphi_p + \frac{\pi}{2} \quad (7.2.1)$$

The signal beam is derived from the seed and will therefore have the same CEP to within an additive constant c_1 . The CEP of the idler is the difference between the

CEP of the pump and signal beams.

$$\varphi_s = \varphi_{w4} + c_1 \quad (7.2.2)$$

$$\varphi_i = \frac{-\pi}{2} + \varphi_p - \varphi_s \quad (7.2.3)$$

Because the seed is derived from the pump and has the same (random) CEP, the phases cancel resulting in an idler pulse with stable CEP.

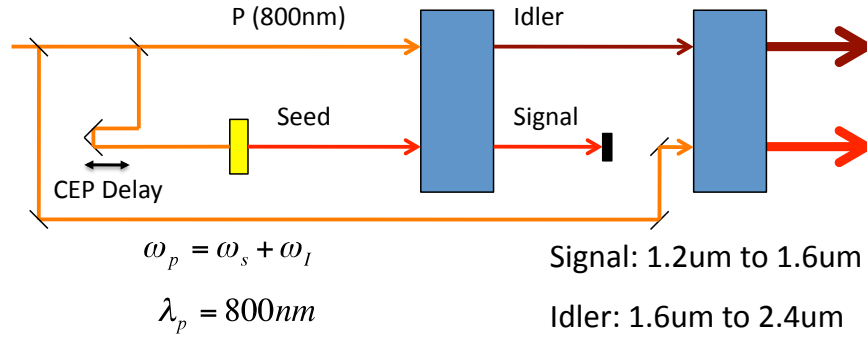


Figure 7.1: Amplification scheme used in modified HE TOPAS-C from Light Conversion. By modifying the path length between the Pump and the WL Seed going into the first gain stage, we can control the absolute CEP of the idler. The idler maintains it stable CE Phase as it is amplified in additional gain stages. Any path length difference between the pump and the idler causes a change in the CE Phase of the signal beam, leaving the idler unaffected. With this approach, interferometric stability is only required between the pump and seed prior to the first gain stage.

7.3 f-2f CEP Measurement

The problem of measuring the (relative) CEP of a many cycle femtosecond laser pulse was solved in 2000 by Jones et al. [104] who showed that it could be measured by broadening the spectrum of the pulse to greater than one octave, and then frequency doubling the red side of the spectrum so that it overlaps with the undoubled blue side. The CEP in the frequency-doubled spectrum is twice that of the undoubled spectrum, so the CEP appears as an interference between the doubled and undoubled spectra. Following Baltuška et al. [101] this can be shown mathematically by expressing the

laser field as:

$$E_L(t) = \frac{1}{2\pi} \int \sqrt{I_L(\omega)} \exp[i(\phi_L(\omega) - \omega t + \varphi)] d\omega + c.c. \quad (7.3.1)$$

where $\phi_L(\omega)$ is the the frequency-dependent spectral phase and φ is the CEP. To gain access to the CEP term, the spectrum is broadened to greater than one octave through SPM, which preserves the CEP. The broadened spectrum is then frequency doubled through second harmonic generation (SHG), which is the convolution of all pairwise combinations of frequency components with the resulting field [101]:

$$E_{shg}(\omega) \propto \exp(i2\varphi) \int \chi^{(2)}(\omega : \omega', \omega - \omega') \sqrt{I_{WL}(\omega') I_{WL}(\omega - \omega')} \\ \times \exp[i(\phi_{WL}(\omega') + \phi_{WL}(\omega - \omega'))] d\omega' \quad (7.3.2)$$

which increases CEP phase by a factor of two. To measure the CEP, the doubled spectrum is time delayed by τ_0 and then combined with the undoubled spectrum as shown in Fig. 7.2. The resulting interferogram is measured with a spectrometer:

$$S(\omega) = (1 - a) I_{WL}(\omega) + a I_{shg}(\omega) + 2 \sqrt{a(1 - a) I_{WL}(\omega) I_{shg}(\omega)} \\ \times \cos(\phi_{shg}(\omega) - \phi_{WL}(\omega) + \omega \tau_0 + \varphi) \quad (7.3.3)$$

For correctly chosen τ_0 , fringes are observed in the spectrum where $I_{WL}(\omega)$ and $I_{shg}(\omega)$ overlap. Changes in the CEP φ cause the position of the fringes to shift. The exact fringe position depends on the details of the nonlinear propagation in the SHG crystal, which means that in practice it is very difficult to know exactly what $\phi_{shg}(\omega)$ is. However, relative changes in the CEP only affect the φ term, and can be observed as a relative shift in the position of the fringes in the interferogram. Figure 7.3(a) shows a typical f-2f interferogram for the WL seeded OPA at ALLS. The laser spectrum, which is centred at $1.8 \mu\text{m}$, is spectrally dispersed with a prism to enable the region between $\sim 600 \text{ nm}$ and $1.2 \mu\text{m}$ to be measured. Note that the spectrometer response is a rapidly decreasing function of wavelength for wavelengths longer than $\sim 1 \mu\text{m}$. Spectral domain interference fringes are visible in the region where the fundamental and second harmonic spectra overlap. The fringes are isolated by multiplying the spectrum with a super-Gaussian filter (shown in red), before the spectrum is Fourier-transformed into the time domain Fig. 7.3(c). The time domain waveform consists

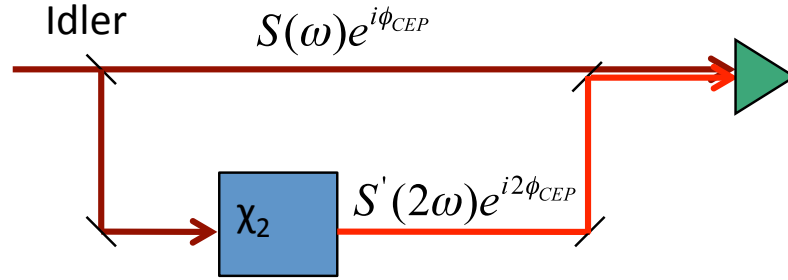


Figure 7.2: In an f-2f interferometer, the laser spectrum is broadened to span > 1 octave and then divided. Part of the spectrum is doubled a nonlinear crystal ($\chi^{(2)}$), before the two beams recombine at the entrance of a spectrometer. The CEP is detected in the spectral interference between the doubled and undoubled beams.

of a large peak at what is effectively zero time, with a side band on either side. The separation between the main peak and the side bands is characteristic of the time delay between the fundamental and second harmonic pulses. To extract the phase of the fringes in Fig. 7.3(a), a super-Gaussian filter is applied around one of the side bands in Fig. 7.3(c). The filtered waveform is then transformed back to the frequency-domain where we plot the amplitude in Fig. 7.3(b) and phase in Fig. 7.3(d). The green curve in Fig. 7.3(d) shows a linear fit to the filtered phase in the region where the magnitude (Fig. 7.3(b)), is non-zero. Changes in the CEP cause the filtered phase response curve to translate vertically. The intercept of the linear fit with the vertical axis is taken as our measure of the *relative* CEP.

7.4 CEP Measurement and Control

In a white light seeded OPA, the seed is phase-coherent with the pump and has the same CEP to within an additive constant. When combined in a DFG crystal these phases cancel, resulting in an idler pulse with a constant CEP regardless of the phase of the pump pulse. This process requires interferometric stability between the pump and WL seed prior to the first DFG crystal. The CEP can be controlled by adjusting the path length between the pump and seed arms. A schematic diagram of the HE-TOPAS -C from Light Conversion is shown in Fig. 7.4. The pump laser is divided by beam splitter BS0, and the transmitted beam is used to generate the seed, and also pumps the first two gain stages. Beam splitter BS2 is used to separate the fraction of

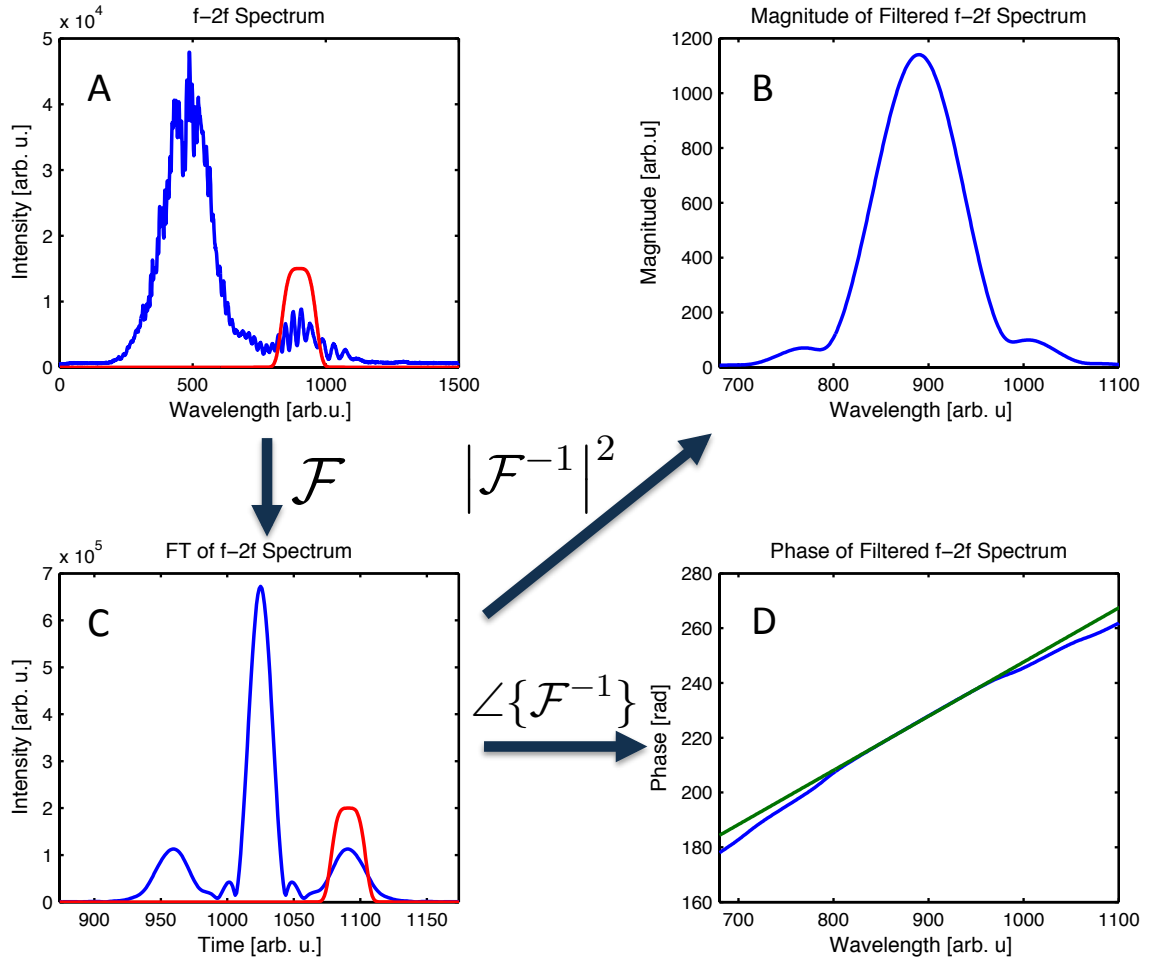


Figure 7.3: Use of spectral interferometry to extract the CEP from an f-2f interferogram.

the pump beam which is used for white light generation (transmitted), and the part which is used for the first gain stage (reflected). The transmitted beam is delayed by mirrors M1 and M2 before focusing into a sapphire window for white light generation WLG. The broadened spectrum combines with the pump beam on dichroic mirror DM1 before passing into the first nonlinear crystal NC1, where the signal and idler beams are produced. In order to control the absolute CEP, we replaced the M1 mirror with a mirror that is mounted on a piezoelectric (PZT), actuator. A photograph of the redesigned M1 mirror assembly is shown in Fig. 7.5. The PZT actuator provides an

HE-TOPAS-C (Ser. # 08504) layout

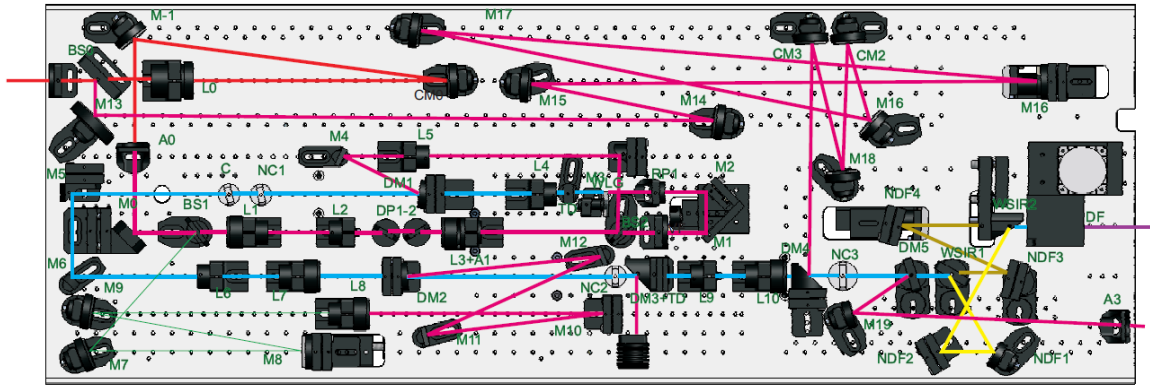


Figure 7.4: Schematic of the optical layout used in the HE TOPAS-C that is installed at the NRC. The ALLS version of this OPA was modified so that the idler beam is amplified following the first gain stage. The M1 mirror mount was also modified to include a PZT actuator which allows for control of the absolute CE Phase of the idler.

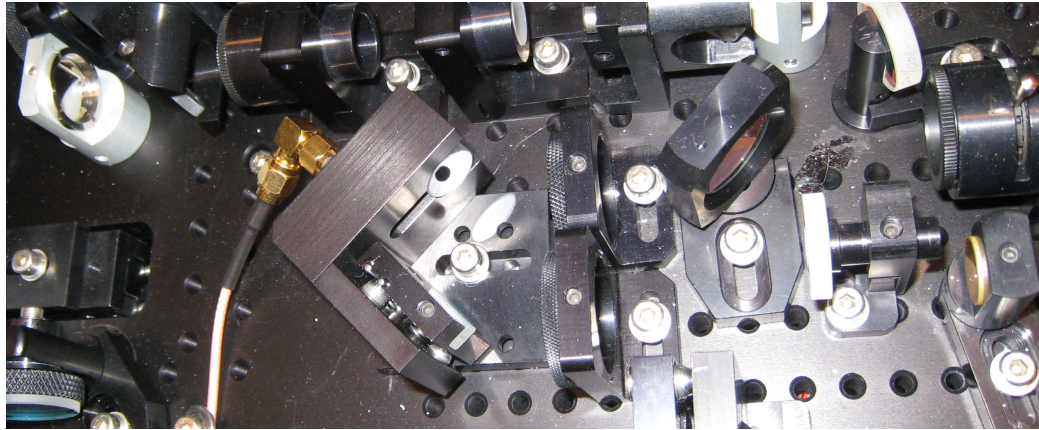


Figure 7.5: Photo of the modified M1 mirror assembly on HE TOPAS-C at ALLS. The mirror mount has been replaced with one that includes a PZT actuator that allows for CE Phase control.

electronically controllable means of translating the M1 mirror, varying the path length difference between the seed and pump arms and with it the absolute CEP. Figure 4 of the publication which is presented starting on Page 119 shows an example of the PZT actuator shifting the position of f-2f fringes.

7.5 Active CEP Control

In the publication on page 119, we show that a WL seeded OPA exhibits a reasonable degree of CEP stability (410 mrad RMS; 10-shot average). The stability of the CEP is limited by mechanical vibrations in the first gain stage, as well as by slow changes in the path length difference between the pump and seed arms, which will cause slow drifts in the CEP. The more important limitation to the long-term (and short-term), stability that is achievable with an OPA comes from nonlinear white light generation process. As shown in Section 6.5, the nonlinear source term in SPM is the product of three fields in the time domain (see Eq. 6.5.1). This term is essential for the generation of new frequencies, but it will also couple energy between different spatial modes. Any variation of mode quality, or the temporal profile of the pulse used for white light generation, will shift the CEP of the idler beam. To help counteract this, we have developed a slow feedback loop which takes a measurement of the CEP from an f-2f interferometer, and applies a correction to the M1 PZT actuator to maintain the CEP at a desired *setpoint* value. For cases where the laser system is operating well, this feedback loop is useful for compensating slow drifts in the CEP as well as for setting the OPA to a desired CEP value. In cases where there is instability in the pump laser intensity, or spatial mode, the feedback loop works to counteract this noise source. Figure 7.6 shows the CEP control loop that is used with the OPA. A spectrometer measures f-2f spectra at 10 to 15 Hz, and a value for the CEP is determined from each measurement using the procedure outlined in Sec. 7.4. These measurements are filtered with an N point First in First Out FIFO moving average filter. The difference between the output from the filter and the desired setpoint value is the *error* signal $e(t)$. The feedback loop uses a proportional integral (PI), controller which calculates the *control* signal to apply to the PZT actuator with the following equation [105]:

$$V(t)_{\text{PZT}} = K_c \left[e(t) + \frac{1}{T_i} \int_0^t e(t') dt' \right] \quad (7.5.1)$$

The output from the control loop goes to a digital-to-analog converter and a high voltage amplifier before being applied to the PZT actuator. The software for the CEP control loop as well as the hardware for the CEP controller were developed by the author. The system was tested at the NRC using an OPA and f-2f interferometer which were setup by Giulio Vampa and Bruno Schmidt. At the time, the laser

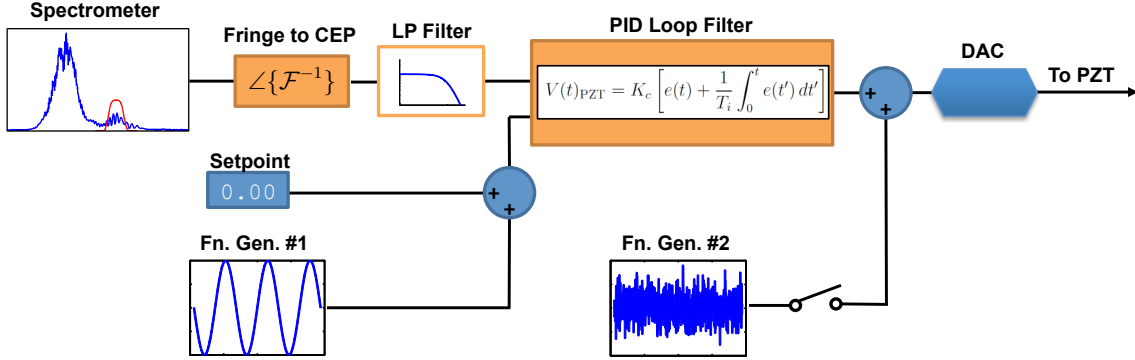


Figure 7.6: Schematic of the CEP control loop. The f-2f spectrum is measured, converted to a CEP value, and low-pass (LP) filtered. The difference between the measured CEP and the setpoint is the error signal $e(t)$ for the PID loop. The setpoint can be a constant value or an arbitrary waveform generated with **Function Generator #1**. The PID loop issues a control signal to the PZT actuator and works to minimize the error signal. The control signal from the PID loop combines with the output from **Function Generator #2**. A digital-to-analog converter (DAC) converts the signal to a voltage, which is then amplified and applied to the PZT actuator inside the OPA.

system was not operating as well as the system at ALLS, and so provided a good opportunity to test the CEP control loop. Fig. 7.7(a) shows the measured CEP from the OPA with the servo loop disabled. The laser could not be used for CEP experiments in these conditions. Fig. 7.7(b) shows the result when the control loop is on. The blue curve is the measured CEP (in-loop measurement), and the green curve shows the voltage applied to the PZT actuator. The control loop has significantly reduced the measured CEP noise, and yields an RMS stability of 322 mRad RMS. The system remained locked for more than 2200 seconds, until the required PZT voltage exceeded the 150 V limit of the actuator. Figure 7.7(c) shows the result of applying a sinusoidal modulation to the CEP setpoint. The setpoint value is plotted in red and the measured CEP (blue), tracks the oscillating setpoint value. Note that these are preliminary results that are intended to show that the feedback loop can be used to control the CEP and to remove noise and slow drifts in phase. The ability of a control loop to counteract noise is always limited by the quality of the measurement. The CEP measurements presented in this section were all acquired with an f-2f interferometer that was located *inside* the control loop. Proper evaluation of the CEP noise requires a measurement with a second f-2f interferometer that is located outside of the control loop. Efforts are currently underway to construct a second f-2f interferometer for

out-of-loop measurements.

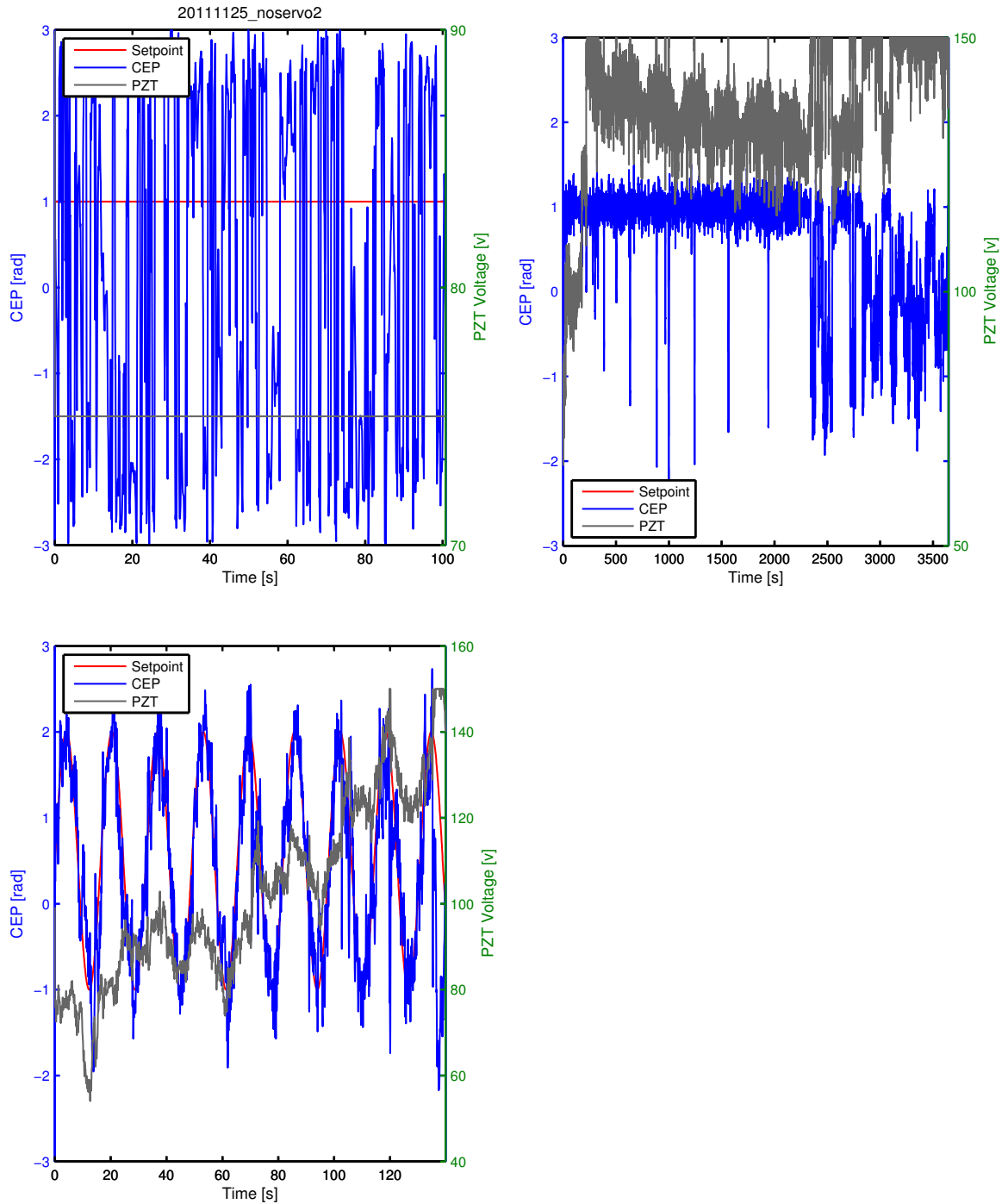


Figure 7.7: CEP control results. Top Left: Free-running OPA without control loop. Top Right: Control loop enabled with a constant setpoint value. For the first 2000 seconds, the loop maintained the CEP lock with a mean value of 0.947 rad and a standard deviation of 322 mrad (in-loop measurement). Lower Left: CEP phase following a sinusoidally-varying setpoint value.

CEP stable 1.6 cycle laser pulses at 1.8 μm

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Abstract: By using the novel approach to pulse compression that combines spectral broadening in hollow-core fiber (HCF) with linear propagation in fused silica (FS), we generate 1.6 cycle 0.24 mJ laser pulses at 1.8 μm with a repetition rate of 1 kHz. These pulses obtained with a white light seeded optical parametric amplifier (OPA) are shown to be passively carrier envelope phase (CEP) stable.

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OCIS codes: (320.5520) Pulse compression; (190.4970) Parametric oscillators and amplifiers.

References and links

1. E. Goulielmakis, V. S. Yakovlev, A. L. Cavalieri, M. Uiberacker, V. Pervak, A. Apolonski, R. Kienberger, U. Kleineberg, F. Krausz, "Attosecond Control and Measurement: Lightwave Electronics," *Science* **317**, 769-775 (2007).
2. M. L. Cowan, B. D. Bruner, N. Huse, J. R. Dwyer, B. Chugh, E. T. J. Nibbering, T. Elsaesser, R. J. D. Miller, "Ultrafast memory loss and energy redistribution in the hydrogen bond network of liquid H_2O ," *Nature* **434**, 199-203, (2005).
3. P. B. Corkum and F. Krausz, "Attosecond science," *Nature Phys.* **3**, 381-387 (2007).
4. A. L. Cavalieri, N. Müller, Th. Uphues, V. S. Yakovlev, A. Baltuška, B. Horvath, B. Schmidt, L. Blümel, R. Holzwarth, S. Hendel, M. Drescher, U. Kleineberg, P. M. Echenique, R. Kienberger, F. Krausz, U. Heinzmann, "Attosecond spectroscopy in condensed matter," *Nature* **449**, 1029-1032 (2007).
5. M. Schultze, M. Fieß, N. Karpowicz, J. Gagnon, M. Korbman, M. Hofstetter, S. Neppl, A. L. Cavalieri, Y. Komninos, Th. Mercouris, C. A. Nicolaides, R. Pazourek, S. Nagele, J. Feist, J. Burgdörfer, A. M. Azzeer, R. Ernstorfer, R. Kienberger, U. Kleineberg, E. Goulielmakis, F. Krausz, V. S. Yakovlev, "Delay in Photoemission," *Science* **328**, 1658-1662 (2010).
6. N. A. Papadogiannis, B. Witzel, C. Kalpouzos, D. Charalambidis, "Observation of Attosecond Light Localization in Higher Order Harmonic Generation," *Phys. Rev. Lett.* **83**, 4289-4292 (1999).
7. M. Drescher, M. Hentschel, R. Kienberger, G. Tempea, C. Spielmann, G. A. Reider, P. B. Corkum, F. Krausz, "X-ray Pulses Approaching the Attosecond Frontier," *Science* **291**, 1923-1927 (2001).
8. Y. Mairesse, A. de Bohan, L. J. Frasinski, H. Merdji, L. C. Dinu, P. Monchicourt, P. Breger, M. Kovačev, R. Taïeb, B. Carré, H. G. Muller, P. Agostini, P. Salières, "Attosecond Synchronization of High-Harmonic Soft X-rays," *Science* **302**, 1540-1543 (2003).
9. G. Sansone, E. Benedetti, F. Calegari, C. Vozzi, L. Avaldi, R. Flammini, L. Poletto, P. Villoresi, C. Altucci, R. Velotta, S. Stagira, S. De Silvestri, M. Nisoli, "Isolated Single-Cycle Attosecond Pulses," *Science* **314**, 443-446 (2006).
10. E. Goulielmakis, M. Schultze, M. Hofstetter, V. S. Yakovlev, J. Gagnon, M. Uiberacker, A. L. Aquila, E. M. Gullikson, D. T. Attwood, R. Kienberger, F. Krausz, U. Kleineberg, "Single-Cycle Nonlinear Optics," *Science* **320**, 1614-1617 (2008).
11. P. B. Corkum, "Plasma perspective on strong field multiphoton ionization," *Phys. Rev. Lett.* **71**, 1994-1997 (1993).
12. G. Tempea, M. Geissler, M. Schnürer, T. Brabec, "Self-Phase-Matched High Harmonic Generation," *Phys. Rev. Lett.* **84**, 4329-4332 (2000).
13. V. S. Yakovlev, M. Ivanov, F. Krausz, "Enhanced phase-matching for generation of soft X-ray harmonics and attosecond pulses in atomic gases," *Opt. Exp.* **15**, 15351-15364 (2007).

14. M.-C. Chen, P. Arpin, T. Popmintchev, M. Gerrity, B. Zhang, M. Seaberg, D. Popmintchev, M. M. Murnane, H. C. Kapteyn, "Bright, Coherent, Ultrafast Soft X-Ray Harmonics Spanning the Water Window from a Tabletop Light Source," *Phys. Rev. Lett.* **105**, 173901 (2010).
 15. I. J. Sola, E. Mével, L. Elouga, E. Constant, V. Strelkov, L. Poletto, P. Villoresi, E. Benedetti, J.-P. Caumes, S. Stagira, C. Vozzi, G. Sansone, M. Nisoli, "Controlling attosecond electron dynamics by phase-stabilized polarization gating," *Nat. Phys.* **2**, 319 – 322, (2006).
 16. Z. Chang, P. Corkum, "Attosecond photon sources: the first decade and beyond [Invited]," *J. Opt. Soc. Am. B* **27**, B9-B17 (2010).
 17. X. Gu, G. Marcus, Y. Deng, T. Metzger, C. Teisset, N. Ishii, T. Fuji, A. Baltuška, R. Butkus, V. Pervak, H. Ishizuki, T. Taira, T. Kobayashi, R. Kienberger, F. Krausz, "Generation of carrier-envelope-phase-stable 2-cycle 740-μJ pulses at 2.1-μm carrier wavelength," *Opt. Exp.* **17**, 62-69 (2007).
 18. C. P. Hauri, R. B. Lopez-Martens, C. I. Blaga, K. D. Schultz, J. Cryan, R. Chirila, P. Colosimo, G. Doumy, A. M. March, C. Roedig, E. Sistrunk, J. Tate, J. Wheeler, L. F. DiMauro, E. P. Power, "Intense self-compressed, self-phase-stabilized few-cycle pulses at 2 μm from an optical filament," *Opt. Lett.* **32**, 868-870 (2007).
 19. O. D. Mücke, S. Ališauskas, A. J. Verhoef, A. Pugžlys, A. Baltuška, V. Smilgevičius, J. Pocius, L. Giniūnas, R. Danielius, N. Forget, "Self-compression of millijoule 1.5 μm pulses," *Opt. Lett.* **34**, 2498-2500 (2009).
 20. C. Vozzi, F. Calegari, E. Benedetti, S. Gasilov, G. Sansone, G. Cerullo, M. Nisoli, S. De Silvestri, S. Stagira, "Millijoule-level phase-stabilized few-optical-cycle infrared parametric source," *Opt. Lett.* **32**, 2957-2959 (2007).
 21. B. E. Schmidt, P. Béjot, M. Giguère, A. D. Shiner, C. Trallero-Herrero, É. Bisson, J. Kasparian, J.-P. Wolf, D. M. Villeneuve, J.-C. Kieffer, P. B. Corkum, F. Légaré, "Compression of 1.8 micron laser pulses to sub two optical cycles with bulk material," *Appl. Phys. Lett.* **96**, 071111 (2010).
 22. P. Béjot, B. E. Schmidt, J. Kasparian, J.-P. Wolf, F. Légaré, "Mechanism of hollow-core-fiber infrared-supercontinuum compression with bulk material," *Phys. Rev. A* **81**, 063828 (2010).
 23. E. Bourkoff, W. Zhao, R. I. Joseph, D. N. Christodoulides, "Evolution of femtosecond pulses in single-mode fibers having higher-order nonlinearity and dispersion," *Opt. Lett.* **12**, 272-275, (1984).
 24. Kubota, M. Nakazawa, "Compensation of nonlinear chirp generated by self-steepening using third order dispersion of a grating pair," *Opt. Comm* **66**, 79-82 (1988)
 25. N. Kuse, Y. Nomura, A. Ozawa, M. Kuwata-Gonokami, S. Watanabe, Y. Kobayashi, "Self-compensation of third-order dispersion for ultrashort pulse generation demonstrated in an Yb fiber oscillator," *Opt. Lett.* **35**, 3868-3870 (2010).
 26. R. Trebino, K. W. DeLong, D. N. Fittinghoff, J. N. Sweetser, M. A. Krumbügel, B. A. Richman, and D. J. Kane, *Rev. Sci. Instrum.* **68**, 3277-3295 (1997).
 27. A. Baltuška, M. S. Pshenichnikov, D. A. Wiersma, "Second-Harmonic Generation Frequency-Resolved Optical Gating in the Single-Cycle Regime," *IEEE Journal of Quantum Electronics*, **35**, 459-478, (1999).
 28. M. Kakehata, H. Takada, Y. Kobayashi, K. Torizuka, Y. Fujihira, T. Homma, and H. Takahashi, "Single-shot measurement of carrier-envelope phase changes by spectral interferometry," *Opt. Lett.* **26**, 1436-1438 (2001).
 29. A. Baltuška, T. Fuji, and T. Kobayashi, "Controlling the Carrier-Envelope Phase of Ultrashort Light Pulses with Optical Parametric Amplifiers," *Phys. Rev. Lett.* **88**, 133901 (2002).
 30. A. Baltuška, T. Fuji, T. Kobayashi, "Visible pulse compression to 4 fs by optical parametric amplification and programmable dispersion control" *Opt. Lett.*, **27**, 306-308 (2002)
 31. D. Herrmann, C. Homann, R. Tautz, M. Scharer, P. St.J. Russell, F. Krausz, L. Veisz, E. Riedle, "Approaching the full octave: noncollinear optical parametric chirped pulse amplification with two-color pumping," *Opt. Exp.* **18**, 18752-18762 (2010).
 32. A modified version of HE TOPAS is available upon request from Light Conversion (private communication with Light Conversion).
 33. H.-C. Bandulet, D. Comtois, A. D. Shiner, C. Trallero-Herrero, N. Kajumba, T. Ozaki, P. B. Corkum, D. M. Villeneuve, J.-C. Kieffer, F. Légaré, "High harmonic generation with a spatially filtered optical parametric amplifier," *J. Phys. B: At. Mol. Opt. Phys.* **41**, 245602 (2008).
 34. R. Trebino, "Frequency-Resolved Optical Gating: The Measurement of Ultrashort Laser Pulses," Kluwer Academic Publishers, (2000).
 35. H. Wang, M. Chini, E. Moon, H. Mashiko, C. Li, and Z. Chang, "Coupling between energy and phase in hollow-core fiber based f-to-2f interferometers," *Opt. Exp.*, **17**, 12082-12089 (2009).
 36. T. Fuji, N. Ishii, C. Y. Teisset, X. Gu, Th. Metzger, A. Baltuska, N. Forget, D. Kaplan, A. Galvanauskas, F. Krausz, "Parametric amplification of few-cycle carrier-envelope phase-stable pulses at 2.1 μm," *Opt. Lett.* **31**, 1103-1105 (2006).
 37. T. Kanai, A. Suda, S. Bohman, M. Kaku, S. Yamaguchi, K. Midorikawa, "Pointing stabilization of a high-repetition-rate high-power femtosecond laser for intense few-cycle pulse generation," *App. Phys. Lett.* **92**, 061106 (2008).
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1. Introduction

Engineering light forms is one of the major challenges in the ultrafast optics community [1]. Expanding those capabilities from established Titanium Sapphire (Ti-Sa) wavelengths towards the infrared spectral range will be particularly useful to study ultrafast energy redistribution in the condensed phase like in the case of correlations in liquid water where the hydrogen-bonded network dephases on a sub-50 fs time scale [2]. Aiming to investigate electron correlations [3-5], the time scale of interaction has to be decreased to the sub fs time scale which is accessible via the process of high harmonic generation (HHG) [6-8]. Furthermore, CEP stable few-cycle laser sources have been the key breakthrough for generating isolated attosecond (asec) pulses [1,9,10]. Those are generated upon recombination of laser field-driven tunneled and accelerated electrons within two thirds of an optical cycle of the electric field oscillation [11].

Currently, CEP stabilized few-cycle laser pulses are mostly generated using ~800 nm Ti-Sa lasers, providing the basis for the synthesis of isolated asec pulses by HHG in gases [3]. CEP stabilization allows one to control the waveform of the electric field under the intensity envelope of with asec precision. Using this asec control, one can restrict the generation of cut-off XUV photons within only one half-cycle leading to isolated pulses with durations near 100 asec [9,10]. Furthermore, in view of increasing the cut-off energy, it has been numerically simulated that few-cycle driving pulses enable self-phase matching in HHG where the group velocity mismatch of XUV and IR driving field is balanced by the harmonics emission phase [12]. This emission phase is determined by the phase of the recolliding electron wave packet and thus depends on varying parameters of the driving field throughout the gaseous target [13]. Alternatively, the approach of quasi-phase matching where HHG takes place in a short hollow fiber can be utilized as well [14]. Complementary to few-cycle driving laser pulses for generation of isolated asec fields, a variety of optical gating techniques has been successfully demonstrated to overcome difficulties in pulse compression [15, 16]. However, one advantage of few-cycle driving fields lies in their reduced amount of ionization, i.e. reduced depletion of the neutral gas target, before the main peak of the electric field can generate the highest energy XUV photons.

Presently, asec laser pulses are generated using XUV photons in the spectral range of 50-110 eV [9,10]. Further reduction of duration requires shorter XUV wavelengths. With HHG having a cut-off at $I_p + 3.17U_p$ (I_p is the ionization potential of the atom and $U_p \sim I \times \lambda^2$, I is the laser intensity and λ the wavelength of the driving laser field) [11], it is evident that further reduction of the duration of the asec pulses requires CEP stabilized few-cycle laser pulses at wavelengths longer than 800 nm [3]. In this letter, we report a simple approach for generation of 1 kHz sub-millijoule CEP stabilized 1.6 cycle laser pulses at 1.8 μm wavelength. The basic idea is to utilize narrowband high power parametric amplification in type-II beta barium borate (BBO) nonlinear crystal followed by subsequent spectral broadening in a HCF. Compression is achieved by linear propagation through appropriate bulk material like FS.

2. Experimental section

To generate intense CEP stabilized IR few-cycle laser pulses, different approaches have been investigated. They are; (1) the optical parametric chirped-pulse amplifier (OPCPA) providing sub-mJ 15.1 fs laser pulses at 2.1 μm [17], pulse self-compression by filamentation at ~2 μm [18,19], and the generation of CEP stabilized 1.5 μm few-cycle laser pulses by DFG (difference frequency generation) of few-cycle 800 nm laser pulses followed by broadband type II parametric amplification [20]. None of these approaches has been able to generate sub-millijoule sub-two cycle CEP stabilized laser pulses.

Recently, we have demonstrated a novel scheme to compress infrared laser pulses to sub-two optical cycle duration [21]. Briefly, non CEP stabilized 1.8 μm laser pulses with duration of 73 fs from a 100 Hz superfluorescence seeded OPA were spectrally broadened via nonlinear propagation in argon filled HCF. Subsequent dispersion compensation was

performed by linear propagation through a 3 mm thickness fused silica (FS) window. It exhibits anomalous dispersion (zero dispersion around 1.3 μm) causing the group velocity dispersion (GVD) to be negative over the entire spectral range of the broadened spectrum. Unexpectedly, third order dispersion (TOD) being positive for all materials has not limited pulse compression with the FS window. Numerical modeling revealed that in addition to self-phase modulation (SPM) as the main driver for spectral broadening in the HCF, self-steepening also occurs causing the trailing pulse edge to become steeper than the leading one, enforcing an asymmetric temporal envelope shape. Again, this temporal asymmetry leads not only to an asymmetric power spectrum but, more important for subsequent compression with bulk material, also to an asymmetry of the spectral phase. This phase asymmetry largely balances the TOD of FS in the range from 1.4 – 2.2 μm [22]. The positive second order phase due to SPM is cancelled by the negative GVD of the anomalous dispersive FS. Similar approaches to balance the effect of self-steepening during nonlinear propagation in single mode fibers by grating pairs have been discussed already in mid 80's [23, 24] and were recently implemented to compensate TOD in an Yb fiber laser oscillator [25]. However, above mentioned schemes were demonstrated with multi-cycle pulses. Pulse characterization was performed using a home-built all reflective SHG-FROG (second harmonic generation – frequency resolved optical gating [26]) which was also used for the experiments described in this paper. The design took into account all considerations for characterization of few-cycle pulses, like for instance phase matching and temporal smear, as described by Baltuška *et al.* [27]. A detailed description of the SHG-FROG apparatus can be found in Ref. [21]. Confirmation of the CEP stability was carried out with a collinear f-2f interferometer [28]. A 0.25 mm thick type-I BBO ($\theta = 23^\circ$) was used as doubling crystal in combination with transmission polarizer (Thorlabs, LP-NIR) to project the blue spectral part of the supercontinuum generated in the HCF and the frequency doubled fundamental onto the same polarization axis.

Besides the increase of repetition rate to 1 kHz, the hereinafter described setup fulfills two important requirements towards isolated asec pulse generation, (i) we employed CEP self stabilization by means of white light seeded OPA [29] and (ii) demonstrated the capability of generating quasi single-cycle pulses of the driving laser, i.e. 1.6 cycle intensity FWHM duration which corresponds to 9.5 fs. CEP control of few-cycle pulses is necessary to generate isolated attosecond pulses [9, 10].

Figure 1 shows the schematic layout of the experimental setup comprising high energy OPA (upper part) and pulse compression stage (lower part). OPA pumping is carried out by a non CEP stabilized kHz Ti-Sa amplifier delivering 3.8 mJ, 35 fs pulses at 800 nm center wavelength (KMLabs, Boulder, USA) which is split into four paths, represented as dashed lines with corresponding numbering. The splitting ratio for the various paths is indicated in figure 1 as percentage rate for the reflected part of the beam. All nonlinear BBO crystals in the OPA are cut for type-II phase matching at $\theta = 28^\circ$. Passive CEP stabilization is achieved via difference frequency generation (DFG) of the seeding white light continuum in path 1 (gray line in figure 1) carrying the same CEP fluctuations as the split fraction of the pump beam in path 2. As a result of DFG via three wave interaction in the first BBO 1, three beams, pump, Signal and Idler herein after identified by the subscripts p, s and i, emerge from the crystal. Conservation laws for energy ($\omega_p = \omega_s + \omega_i$) and momentum ($\vec{k}_p = \vec{k}_s + \vec{k}_i$) have to

be fulfilled with ω denoting the center frequency and $\vec{k}$ the wave vector. The latter requirement is also referred as the phase matching condition

$$\Delta\vec{k} = \vec{k}_s + \vec{k}_i - \vec{k}_p = 0 \quad (1)$$

which becomes increasingly difficult to satisfy as the bandwidth increases to support shorter pulses. Violating the phase matching condition causes temporal and spatial chirp, but can be minimized using non-collinear interaction with tilted pulse fronts. This approach has been used by several research groups to generate ultra broadband laser pulses [30, 31].

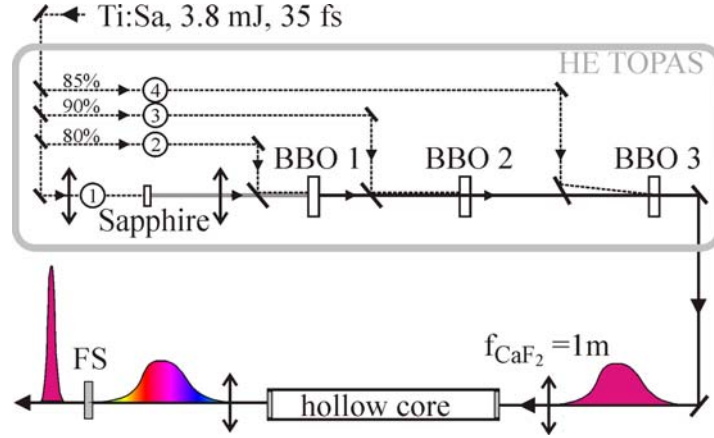


Fig. 1. Schematic of the pulse compression experimental setup. Ti-Sa laser pulses with 35 fs duration and 3.8 mJ of energy are frequency shifted at 1.8 μm using a white light seeded Optical Parametric Amplifier consisting of three parametric amplification stages. Output energy of 650 μJ and 35 fs pulse duration at 1.8 μm is coupled to a hollow-core fiber and subsequently compressed solely by a FS window to 1.6 optical cycles.

Because of the challenges with broadband parametric amplification, we have develop an approach based on narrowband OPA systems delivering high peak power infrared laser pulses (HE-TOPAS from Light Conversion) followed by spectral broadening in a HCF. Generally, OPA techniques enable passive CEP stabilization because of conservation of the offset phase during parametric interaction [29]:

$$\phi_p = \phi_s + \phi_i + \pi/2 \quad (2)$$

From Eqn. 2, it is evident that for equally fluctuating CEP of ϕ_p and ϕ_s , ϕ_i is intrinsically CEP stable [29]. The broadband seeding Signal generated through SPM in the Sapphire window of path 1 has the same, random, CE Phase as the pump beam. For the Idler after BBO 1 this phase cancels resulting in an Idler with constant CEP as long as the path length between seed and pump is maintained. To facilitate this we have modified the commercial OPA such that the Idler rather than the Signal is amplified after BBO 1 [32]. The effect of this is that interferometric stability is only required for the first gain stage between arms one and two in the Fig. 1. The CEP of the idler is fully defined after BBO 1 and any changes in path length after this shifts the, still random, phase of the Signal while leaving the Idler unaffected. In the commercial OPA, DFG in BBO 1 is carried out in a non-collinear interaction. Considering Eqn. 1, it can be seen that this causes the pump, Signal and Idler pulses to be spatially spread. Moreover, Idler pulses exhibit a spatial chirp if the intersecting pump and Signal beams contain more than a single frequency. Thus, we rearranged this stage to be collinear and selected the Idler by means of dichroic optics. This Idler beam was then amplified further in BBO 2 and 3 whereby pumping in BBO 3 is carried out under a small angle ($\sim 0.5^\circ$) to separate Signal and Idler spatially in front of the HCF. A CaF_2 plano-convex lens with 1 m focal length is used to couple the light into the HCF being supported on an aluminum V-groove. Both fiber ends are attached to closed gas cells with 1 mm thickness, anti-reflection coated CaF_2 windows to minimize material dispersion. At the output, the laser beam was collimated using a $f=1$ m concave silver mirror. The HCF is evacuated and filled with argon at a constant pressure of 1.6 Atm.

3. Results and discussion

At the output of the OPA, after BBO 3, 650 μJ of pulse energy at 1.77 μm was available at a repetition rate of 1 kHz. Pulse intensity duration of 35 fs at full width half maximum has been characterized using SHG-FROG. The collimated beam exhibits an $1/e^2$ intensity width of about 10 mm. In comparison with our previous experiments using superfluorescence seeded OPA [19], we now start with already shorter input pulses (35 fs vs 73 fs) for spectral broadening in the HCF. During propagation of the laser pulses in the HCF, the duration at the output remains almost unchanged. The broadened spectra extend from 1.15 μm to 2.15 μm and the measured spectral density is shown in figure 2(c) (gray line). An asymmetric shape with a wider extension towards the blue spectral side is observed due to self-steepening during nonlinear propagation. The spectral sensitivity of the IR spectrometer (NIR 256 Ocean optics) was precisely determined with a broadband calibration lamp.

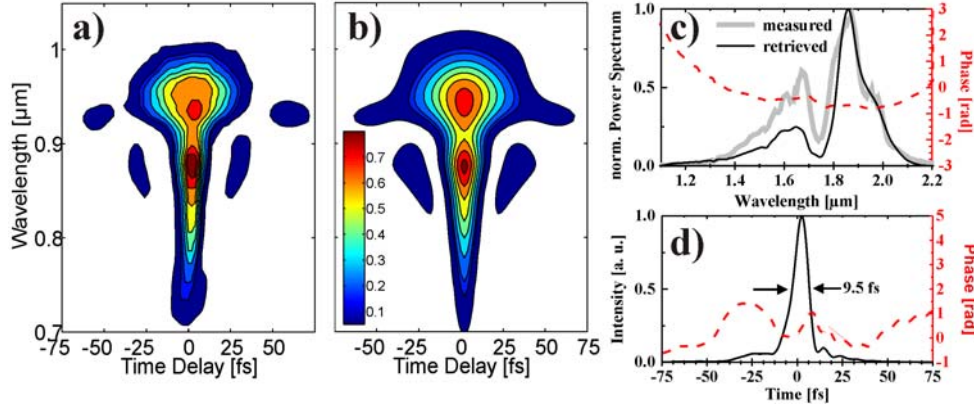


Fig. 2. Results of pulse compression. Measured (a) and retrieved (b) SHG-FROG traces of the 1.6 cycle pulse after compression with 1.5 mm FS. Lowest contour line at 4% intensity value is followed by successively increasing steps of 10%. (c): measured (gray) and retrieved spectral intensity (black) and phase (dashed red) with corresponding time domain representation in (d).

Flattening the spectral phase with 1.5 mm FS led to 1.6 cycle laser pulses with 240 μJ pulse energy without detecting any superfluorescence or remaining pump beam background after the HCF. Good agreement down to the 4% intensity level between measured and retrieved spectrograms can be verified in figure 2(a) and 2(b), respectively. Excellent spatial properties were observed at the output due to the spatial cleaning provided by HCF [33]. In addition, it should be noted that less FS was used to compress the pulses than in our previous experiment with 73 fs input laser pulses (1.5 mm vs 3 mm). The amount of dispersion to compensate depends on the ratio between input and output pulse duration. In figure 2(c), based on the SHG-FROG reconstruction, the corresponding retrieved power spectral density (black) and the spectral phase (dashed red) after passing through the FS window are presented. Considering that our laser pulses have almost an octave spanning spectra, the agreement between measured and retrieved spectral density is very good. Prior to SHG-FROG reconstruction, the measured spectrogram is filtered according to reference [34] in order to take into account the spectral distortions of the SHG process as well as the spectral response of the VIS-NIR spectrometer (Model USB2000, Ocean Optics) in the FROG apparatus.

Temporal intensity and phase of the 9.5 fs pulse are presented in figure 2(d) as solid black and red dotted curves. Residual uncompensated higher order spectral phase in figure 2(c) leads to the non-transform limited temporal shape. Still, the main peak carries 83% of the total pulse power and will be an excellent tool to study strong field processes like HHG, nonsequential double ionization, and above threshold ionization. Since full control over the electric field is desirable in attosecond science and in general for strong field phenomena, we

confirmed passive CEP stabilization which is a necessary prerequisite for controlling the position of the electric field underneath of the intensity envelope. Though not visible on the linear scale of the measured spectrum of figure 2(c) (gray curve), a small fraction of the spectrum extends into the NIR where it can interfere with the SHG of the strong IR part of the spectrum. This allowed us to measure the f-2f interferogram without additional need for a white light generation stage. From the f-2f interferogram presented in figure 3 a RMS jitter of 350 mrad for the CEP has been characterized.

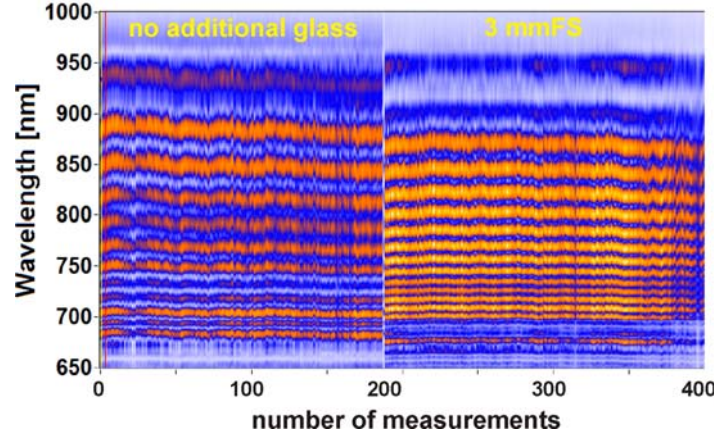


Figure 3. Passive carrier envelope phase stability after the hollow-core fiber. Each measurement is averaged over 10 laser shots. The reduced fringe spacing on the right side (3 mmFS) compared to the left one (no additional glass) corresponds the group delay of 3 mm FS introduced before frequency doubling.

We believe that 350 mrad represents an upper bound for the true CEP fluctuations from our OPA. Wang *et al.* recently demonstrated strong intensity to CEP coupling in a HCF. They show that a 1% change in intensity results in a 128 mrad shift in the measured CEP at 800nm despite no change in the CEP of the actual waveform [35]. Due to the nonlinear nature of the OPA process, energy fluctuations are unavoidable. We measured 6% energy fluctuation after the HCF (3.5% after the OPA). This energy fluctuation in the HCF translates into spectral phase fluctuations in particular in the wings of the broadband spectra, causing the f-2f interferogram to over report CEP errors. This artifact has been also discussed by Fuji *et al.* for few-cycle IR laser pulses generated using OPCPA [36]. Pointing instability into the HCF also contribute to the measured variation in CEP. In our case, it should be possible to reduce energy fluctuation in the HCF and the associated CEP jitter by implementing beam pointing stabilization as described in [37]. However, the measurement of figure 3 clearly shows that the CEP stability is preserved even if temporal reshaping of the pulse intensity envelope during nonlinear propagation in terms of self-steepening occurs.

4. Conclusion

In conclusion, we have reported a simple and robust experimental approach to generate CEP stabilized sub-millijoule 1.6 cycle laser pulses at 1.8 μm wavelength. Future improvements to this source will include beam pointing stabilization as well as the integration of a white light seeded high energy OPA to the 100 Hz 100 mJ Ti-Sa laser system of the Advanced Laser Light Source (INRS-EMT) to have sub-5 mJ Idler laser pulses to couple to the HCF [32]. This will be the ideal IR laser source for HHG and to extend attosecond pulse synthesis in the sub-eV spectral range.

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High harmonic generation with long-wavelength few-cycle laser pulses

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Abstract

We report the extension of hollow-core fibre pulse compression to longer wavelengths. High-energy multi-cycle infrared pulses are generated via optical parametric amplification and subsequently broadened in the fibre. 2.5-cycle pulses at the Signal wavelength (1.4 μm) and 1.6-cycle pulses at the Idler wavelength (1.8 μm) in the sub-millijoule regime have been generated. New compression schemes can be applied at 1.8 μm and beyond. In this manner, 1.6-cycle carrier envelope phase stable pulses were generated by linear propagation in the anomalous dispersion regime of bulk glass which surprisingly enables compression below its third-order dispersion limit. Furthermore, a dispersion-free way of controlling the carrier envelope phase is demonstrated. Moreover, we experimentally confirm the increase in high-harmonic cut-off energy with driving laser wavelength and demonstrate the beneficial effect of few-cycle pulses which enable higher saturation intensities on target compared to multi-cycle pulses. It will be an ideal tool for future synthesis of isolated attosecond pulses in the sub-keV regime. With this laser source, we revealed for the first time multi-electron effects in high harmonic generation in xenon.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

In 1987, Fork *et al* achieved a long-lasting world record of pulse compression down to 6 fs at ~ 620 nm wavelength based on spectral broadening through nonlinear propagation of 50 fs laser pulses in a single-mode fibre (SMF) followed by phase compensation up to cubic order with a combination of prism and grating compressors [1]. This unique laser source rapidly paved the way for a variety of time-resolved spectroscopy experiments requiring high temporal resolution [2–5]. Limited to a few nanojoules of energy per pulse due to the SMF, those pulses were not intense enough to study strong field processes such as high harmonic generation (HHG).

In 1996, Nisoli *et al* extended the approach of spectrally broadening multi-cycle pulses to the millijoule energy level of

high-power Ti:Sa chirped pulse amplifiers. They replaced the SMF by a hollow-core fibre (HCF) filled with gas as the nonlinear medium [6]. Dispersion compensation with a prism pair subsequent to the HCF enabled generation of 10 fs pulses at 0.8 μm centre wavelength. By the end of the 1990s, generation of intense 5 fs pulses at Ti–Sa wavelength became possible due to invention of chirped mirrors (CM). Meanwhile, strategies to stabilize the carrier envelope phase (CEP) of laser pulses were developed [7, 8] leading to the first demonstration of isolated attosecond pulses generated by the process of HHG in gas medium in 2001 [9]. Nowadays, attosecond metrology is used to probe the fastest electronic processes in atoms [10–12], molecules [13] and condensed matters [14]. Recent advancement in CM technology enabled dispersion compensation over an octave of bandwidth providing

single-cycle Ti-Sa laser pulses [15] which were used to generate an 80 attosecond pulse duration in the sub-100 eV spectral range [16].

Obtaining even shorter x-ray pulses via HHG requires extending attosecond metrology to higher photon energies and larger spectral bandwidth [17]. Higher photon energies have been demonstrated with laser sources delivering longer wavelengths compared to Ti-Sa [18–21]. This can be understood by the three-step model as described by Corkum in 1993 [22], which predicts the cut-off photon energy (E_C) of the HHG spectra to be given by E_C (eV) = $I_p + 3.17U_p$ (cut-off law), with I_p being the ionization potential of the atom and $U_p \sim I\lambda^2$. Increasing laser intensity (I) on target, however, must not exceed the threshold for depletion of population in the neutral state which strongly depends on I_p . One approach to overcome this limitation of generating higher cut-off energy is to increase the driving laser wavelength. While several research groups have shown cut-off extension with longer laser wavelength [18–21], isolated attosecond pulses have not yet been demonstrated at photon energies higher than 110 eV. With the aim of transferring established strategies for the generation of isolated attosecond pulses from 0.8 μm to the IR spectral range, a few research groups have been working to develop laser sources delivering CEP stabilized few-cycle pulses at longer wavelength.

Different approaches have been investigated to generate mJ level CEP stabilized few-cycle laser pulses. They are (1) the optical parametric chirped-pulse amplifier providing sub-mJ laser pulses at 2.1 μm [23, 24], pulse self-compression by filamentation at ~ 2 μm [25, 26] and the generation of CEP stabilized 1.5 μm few-cycle laser pulses by difference frequency generation (DFG) of ultra-broadband 0.8 μm laser pulses followed by broadband parametric amplification [27]. In this paper, we show that spectral broadening in an argon-filled HCF followed by a proper dispersion compensation scheme (CM at 1.4 μm and a fused silica window at 1.8 μm [28]) is a robust and simple approach to obtain few-cycle IR pulses. It can be extended to even longer wavelengths, as long as glasses are not absorbing [29]. In addition, starting with a white light seeded optical parametric amplifier provides passive CEP stabilization [30].

Non-CEP stabilized pulses have been applied to perform HHG in xenon described in [31]. Here we expand on these results, showing the spectral evolution of HHG from xenon as a function of wavelength and laser pulse duration. We verify the extension of the XUV spectrum with longer laser wavelength according to the cut-off law. Furthermore, we demonstrate how few-cycle pulses allow higher intensities on target before depletion of ionization of the neutral ground state compared to longer pulse durations. This new laser technology enabled our research team to probe the giant resonance of xenon in the proximity of 100 eV and to extend the cut-off up to 160 eV.

2. Experimental method

The following experiments have been performed on two different laser systems at the advanced laser light source in Varennes, Canada. They share the common principle

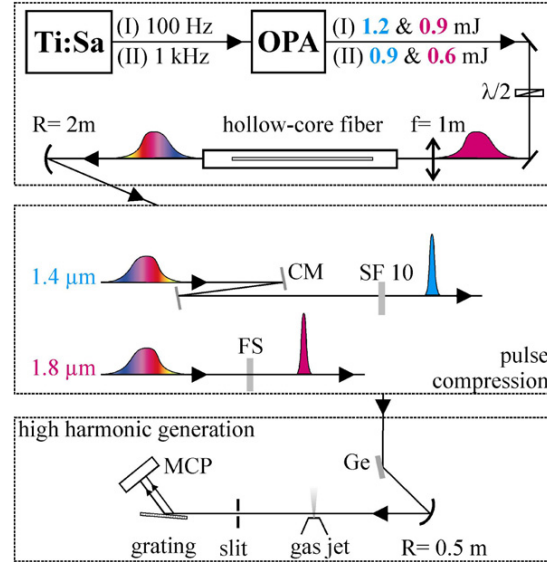


Figure 1. General sketch of the experimental setup representing two different laser systems (I) and (II). The upper panel shows IR pulse generation via commercial OPA systems pumped by a Ti-Sa amplifier. A plano-convex lens couples the light into a standard hollow-core fibre for spectral broadening in an argon atmosphere. Signal pulses are compressed with chirped mirrors (CM), whereas Idler pulses are compressed only with an FS window to sub-two cycle duration. Blue is chosen for Signal wavelengths, whereas red represents Idler pulses in the middle panel. Those pulses are focused in a thin gas jet of xenon and the resulting XUV light from HHG is recorded with an imaging spectrometer and a micro channel plate (MCP, lower panel).

of starting from narrowband, high-energy IR laser pulses generated via type II amplification with a commercially available OPA. After that, the OPA output is spectrally broadened in an HCF to achieve the large bandwidth required for few-cycle pulse generation, as shown in the upper panel of figure 1. The fact that we performed pulse compression experiments with two different Ti:Sa laser systems which are not CEP stabilized underlines the universality of our approach. Each Ti:Sa amplifier is pumping a different type of high-energy OPA. Firstly, we worked with a 100 Hz, 12 mJ, 45 fs laser system (Thales) from which 6 mJ was split to pump a superfluorescence seeded high-energy OPA (Light Conversion, HE-TOPAS, labelled (I) in figure 1). Secondly, a 1 kHz, 3.8 mJ, 35 fs Ti-Sa laser (KMLabs) was used in combination with a white light seeded OPA likewise from Light Conversion (labelled (II) in figure 1). The advantage of using a white light rather than fluorescence seeded OPA lies in the passive CEP stabilization of Idler pulses in the first case [32].

In the last stage of both OPAs, pumping is performed at a slight angle ($<1^\circ$) with respect to the seed in order to separate Signal and Idler beams spatially after some distance. In all cases, coupling into the HCF is carried out with a 1 m plano-convex lens made of CaF_2 , a material also used for all other windows in the setup due to its small amount of dispersion in the 1–2 μm spectral range. The HCF length is

1 m with an inner diameter of 400 μm which is filled with argon for spectral broadening. Pressure of argon was adjusted to control the spectral width at the HCF output and to minimize instabilities.

For both laser systems (100 Hz and 1 kHz), Signal and Idler wavelengths were compressed in different ways as will be explained in the following. Signal pulses around 1.4 μm were compressed with one round trip on CMs which were designed in collaboration with Femtolasers [33]. They introduce negative group delay dispersion (GDD) to compensate for the primarily quadratic chirp stemming from self-phase modulation (SPM) in the HCF, as well as small, negative third-order dispersion (TOD) to balance positive TOD of glasses eventually used in the setup [33].

Different to this established pulse compression method with CM which is sketched in the middle panel of figure 1, we established a new scheme for compression of Idler few-cycle pulses around a central wavelength of 1.8 μm . It simply utilizes linear propagation through a single window of FS instead of CM to balance the spectral phase introduced in the HCF. At a first glance, this seems very straightforward. Since many glasses exhibit anomalous dispersion in the IR range they provide negative GDD to cancel the major part of the spectral phase arising from SPM. However, all dielectric media provide positive TOD throughout their transparent spectral range. To our surprise, the experiment shows that linear propagation in bulk material after the HCF can lead to pulse compression below its TOD limit (TOD limit means full compensation of GDD only). This can be explained by the fact that in addition to SPM in the HCF also self-steepening of the pulse trailing edge occurs during nonlinear propagation. Assuming a symmetric temporal shape of the input pulse, pure SPM causes an intensity-induced symmetric variation of the pulse temporal phase without changes to the pulse envelope. This, in turn, causes a symmetric broadening of the power spectrum with respect to the OPA's fundamental wavelength as well as a symmetric spectral phase which can be approximated by a parabola centred around the fundamental. On the other hand, self-steepening imprints a temporal reshaping of both the pulse envelope and temporal phase which become asymmetric. In the spectral domain, this behaviour translates to an asymmetric power spectrum and what is essential for the new compression scheme, also to an asymmetry of the spectral phase. This additional phase asymmetry on top of the symmetric SPM is nicely opposed by the TOD of bulk material. This intuitive physical explanation was verified by numerical modelling of nonlinear propagation in the HCF and linear propagation in FS afterwards [29]. It is worth mentioning that self-steepening results from a nonlinear polarization of the argon gas without the need for plasma generation. Furthermore, this compression scheme was numerically proven to support clean single cycle pulses and experimentally quasi-single-cycle pulses of about 1.5 cycles have been demonstrated [30, 34].

Pulse characterization was carried out with a home-built second harmonic generation–frequency resolved optical gating

(SHG-FROG) setup⁴ and more details can be found in the literature [28, 33].

Subsequent to dispersion compensation, the few-cycle IR pulses were applied to study HHG at different wavelengths and with different pulse durations. A schematic diagram of the HHG spectrometer and source chamber is shown in the lower panel of figure 1. Before entering the vacuum chamber, an arrangement for variable attenuation over the Signal and Idler spectral range from 1.1 to 2.3 μm has been installed. It enables energy control without changing the optical path after the HCF. This is realized by placing an achromatic half wave plate prior to the HCF whose spectral range is sufficient to cover the range from 1.4 to 1.8 μm . Depending on its rotation of the laser polarization, a corresponding fraction of light is reflected off the Germanium window (Ge in the lower panel of figure 1) which is mounted at Brewster's angle. Nonlinear propagation in the HCF is not affected by rotation of polarization.

The beam is then focused inside the vacuum chamber into a thin pulsed jet of xenon with an $R = 0.5$ m spherical silver mirror. The jet has a 500 μm aperture; thus, it is much thinner than the Rayleigh range of the optical beam. It is typically operated at a backing pressure of 2–3 atm leading to an estimated number density of gas atoms in the jet of around $2 \times 10^{17} \text{ cm}^{-3}$. The intensity was determined by varying the input pulse energy and measuring the amount of ionization in the gas jet. Ions were detected with a negatively biased wire mesh (not shown in figure 1) a few centimetres away from the focus. By fitting the measured ion signal versus input energy to the calculated ion signal based on the Yudin–Ivanov ionization model [35], the laser intensity can be determined [36]. This procedure is described in more detail in [28, 35].

The XUV light passes from the source to the detector chamber via a differential pumping tube after which the XUV beam enters the spectrometer through an $\sim 140 \mu\text{m}$ slit. The XUV spectrum is dispersed by a 1200 l mm^{-1} spherical holographic grating (Shimadzu) which was chosen because of its significantly lower second-order diffraction compared to equivalent ruled gratings [37, 38]. The dispersed spectrum is imaged onto a microchannel plate (MCP) intensifier backed by a phosphor screen (Burle) and the spectrum is digitized with a high dynamic range digital camera (PCO).

3. Experimental results and discussions

As mentioned above, pulse compression has been investigated at 1.4 and 1.8 μm . Firstly, we used a parametric superfluorescence seeded OPA pumped by a 100 Hz Ti–Sa laser system and those results are presented in section 3.1. The same scheme was realized with a different Ti–Sa laser (1 kHz), pumping a white light seeded OPA for which the results are presented in section 3.2. In both cases, the OPAs were tuned to generate a set of conjugated Signal and Idler pulses roughly at the same wavelengths to provide comparable conditions. This set of wavelengths was chosen because both OPAs show the highest output power and best stability. Section 3.3 describes

⁴ We employed geometrical beam splitting with a half mirror and used a thin type I BBO crystal of 10 μm thickness. Vacuum windows, etc were accounted for in the FROG measurement.

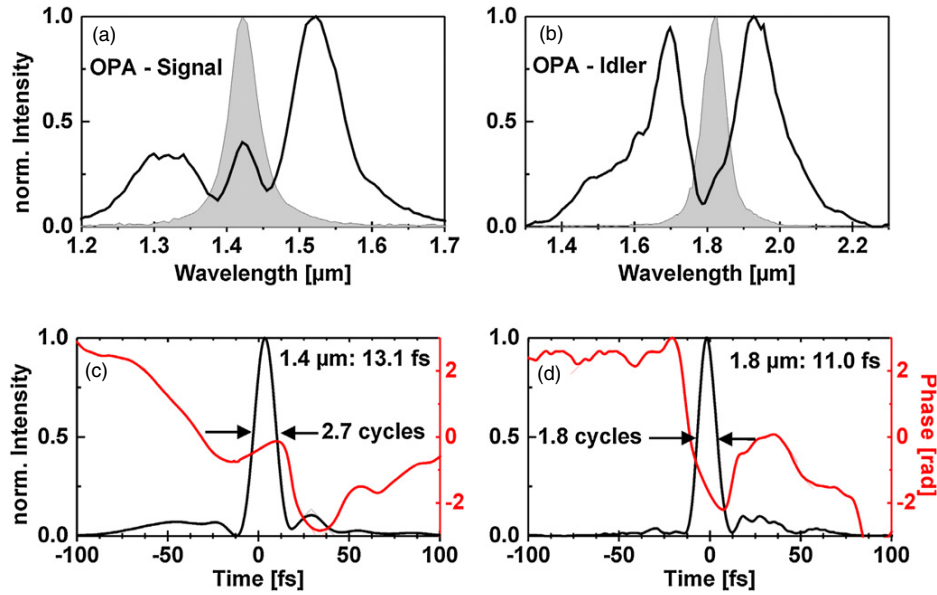


Figure 2. Results with superfluorescence seeded OPA. OPA output pulses (shaded grey) and after broadening in the hollow-core fibre (black) for (a) the Signal at $1.425 \mu\text{m}$ wavelength and (b) the Idler at $1.82 \mu\text{m}$ wavelength. Retrieved temporal intensity (black) and phase (red) for (c) Signal pulse compression with chirped mirrors and (d) Idler pulses after linear propagation through FS only.

a novel scheme for controlling the CEP in a white light seeded OPA.

Finally, the 100 Hz setup of section 3.1 was used for HHG in xenon and XUV spectra for different wavelengths and pulse durations of the driving laser are presented in section 3.4. Thanks to the long-wavelength few-cycle pulses, the XUV spectrum for low I_p species like xenon extends far enough enabling us to observe for the first time multi-electron correlation effects in xenon, in particular the giant resonance around 100 eV photon energy.

3.1. Superfluorescence seeded OPA (case I)

The 100 Hz setup, case (I) in figure 1, employing a superfluorescence seeded OPA delivers Signal and Idler pulses whose energies are 1.2 mJ and 0.9 mJ, respectively. Corresponding OPA spectra are shown in figures 2(a) and (b) as shaded grey curves. Those spectra are associated with pulse durations in the range of 50–60 fs. Pulse durations remained unchanged after the HCF. Effects of pulse self-shortening were not observed since the input power in each case was well below the critical power for self-focusing. Fibre output spectra are shown as black curves in figures 2(a) and (b), each normalized by their maximum value.

A transmission of 40% was achieved for the Signal pulses leading to 0.4 mJ output energy after the HCF. After passing the CM, SHG-FROG traces were recorded for different pressures of argon and with different SF10 windows to fine-tune dispersion. In this way, the shortest duration of 13.1 fs was achieved with 1.7 bar of argon. The SHG-FROG retrieved temporal intensity is plotted in figure 2(c) in black as well as the temporal phase in red.

Now, we present results achieved with the OPA Idler on the 100 Hz laser system. Coupling into the HCF resulted in a

transmission of about 50%, slightly higher than in the Signal case with the same HCF. We attribute this to improvement mode quality of the Idler beam. Up to 0.45 mJ pulse energy was obtained after the fibre with a duration of about 50 fs and the broadened spectrum for 1.2 bar of argon is given in figure 2(b). An equivalent of about 4 mm FS was used to generate 11 fs pulses at $1.8 \mu\text{m}$ wavelength. This denotes about 36 GW of pulse power within 1.8 optical cycles FWHM, an ideal source to study strong field processes such as HHG. A rather clean temporal shape is visible in the plot of figure 2(d) which is carrying about 77% of total energy in the main pulse.

3.2. White light seeded OPA (case II)

Now we switch to the second Ti–Sa laser system. Besides increasing the repetition rate to 1 kHz, it is capable of delivering shorter pulses at $0.8 \mu\text{m}$ as compared to the 100 Hz system. The white light seeded OPA generates Signal and Idler pulses with energies of 0.9 and 0.6 mJ, respectively. Although the last amplification crystal in both high-energy TOPAS, superfluorescence and white light seeded, has the same cut and thickness, the spectra for the latter are slightly broader, as can be seen when comparing the shaded grey curves of figures 3(a) and (b) and 2(a) and (b). In accordance with the broader spectra, the OPA pulses were slightly shorter in the range of 30–40 fs.

Spectral broadening of Signal pulses in 2.3 bar of argon leads to the continuum shown in figure 3(a) as a black curve. Since the energy directly in front of the HCF was 0.8 mJ and we obtained a transmission of about 42%, we measured 0.35 mJ pulse energy after the HCF. The same CM setup as described in section 3.1 leads to pulse compression down to 12.1 fs, shown in figure 3(c) as a black curve. The shorter duration of 12.1 fs

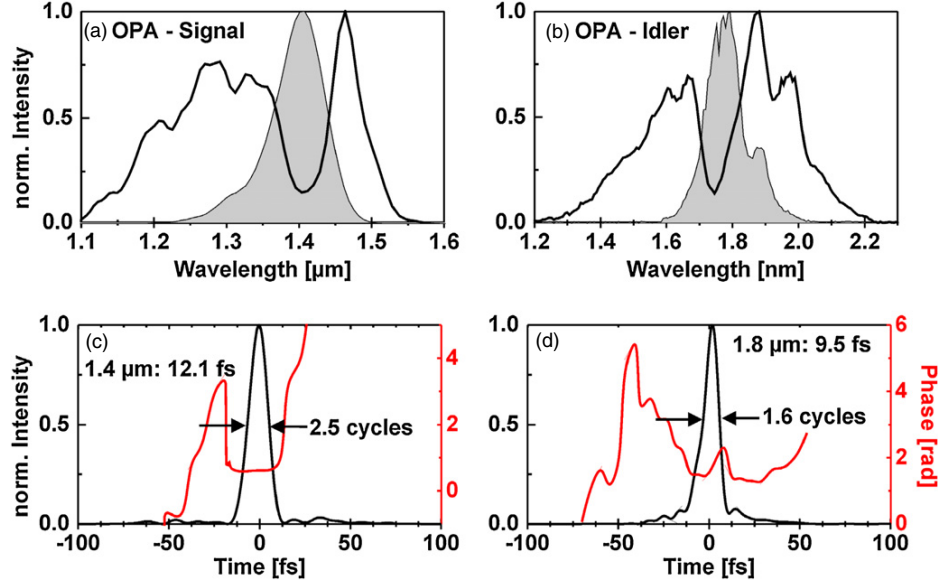


Figure 3. White light seeded OPA results. Output spectra of OPA (shaded grey) and after the hollow-core fibre (black) for (a) the Signal at $1.4 \mu\text{m}$ wavelength and (b) the Idler at $1.78 \mu\text{m}$ wavelength. Retrieved temporal intensity (black) and phase (red) for (c) Signal pulse compression with chirped mirrors, and (d) Idler pulses after linear propagation through FS only.

compared to 13.1 fs in section 3.1 is linked to the shorter input pulse duration coming from the white light seeded OPA which was 42 fs compared to 60 fs from the superfluorescence seeded one.

From a physical point of view, it is obvious that shorter input pulses require less SPM to achieve the same spectral broadening as longer input pulses. The broad spectrum in figure 3(a) exhibits only one minimum in the supercontinuum whereas the one of figure 2(a) shows two minima and a third peak arises at the central wavelength. From simulations, it can be seen that stronger SPM leading to a three peaked continuum is also accompanied by a more complex spectral phase function which shows an increasing number of oscillations around the central wavelength limiting the potential for clean pulse compression.

The same arguments hold for the Idler bulk pulse compression scheme for which the input pulse duration of 35 fs was shorter on the 1 kHz system compared to the situation in section 3.1. The input spectrum before the HCF is shown in figure 3(b) as a shaded grey curve and spectral broadening in 2.3 bar argon is given by a black curve. After the fibre, linear propagation through 1.5 mm of FS suitably compresses the spectral phase to 9.5 fs FWHM duration of intensity. This corresponds to 1.6 optical cycles at $1.8 \mu\text{m}$ wavelength.

The transform-limited (TL) duration for the spectrum in figure 3(b) is 9.2 fs and the residual uncompensated higher order spectral phase is responsible for observed deviation from the theoretical value. Although pedestals arise when aiming for the shortest FWHM duration, the main peak still carries 83% of the total pulse power. To emphasize the high fidelity of phase compensation with bulk material we calculated the effect of uncompensated TOD on few-cycle pulse compression. Assuming perfect cancellation of the GDD component, solely

the travel through 1.5 mm FS would broaden a TL 9.5 fs pulse to 11.9 fs.

Another important attribute of the 1.6-cycle pulse of figure 3(d) is its passive CEP stability which was confirmed with an $f-2f$ interferometer. To build this, no additional broadening stage had to be implemented to generate the octave spanning spectra since the blue spectral wing in figure 3(b) extends out to $0.7 \mu\text{m}$ which cannot be seen in the linear plot. Thus, only a type I BBO for SHG of IR and a polarizer to project both the broadband spectrum and its doubled fundamental is required to perform $f-2f$ measurements. An *rms* stability of 350 mrad was characterized in this manner [30] on the 1 kHz laser system.

3.3. CEP control

Very recently, the 100 Hz laser system was upgraded with a white light seeded OPA yielding pulse durations of 1.9 optical cycles which are passively stabilized [32]. The passive CEP stabilization stems from DFG between a white light continuum ($\Phi_{\text{white light}}$) and a remaining portion of the 800 nm pump beam (Φ_{pump}), both sharing the same randomly fluctuating CEP. Since the DFG process conserves not only the quantities of energy and momentum but also the phases, the CEP of the Idler (Φ_{Idler}) is constant:

$$\Phi_{\text{Idler}} = \Phi_{\text{pump}} - \Phi_{\text{white light}} - \Phi/2 = \text{const.} \quad (1)$$

From equation (1) it can be seen that an additional constant phase offset added to Φ_{pump} or $\Phi_{\text{white light}}$ transfers to a phase shift of the Idler pulse. We implemented this phase shifter in the white light arm of the OPA by moving a steering mirror with a piezoelectric actuator [39]. We carefully checked that the mirror movement on the order of the wavelength does neither affect the few-cycle pulse compression nor the CEP

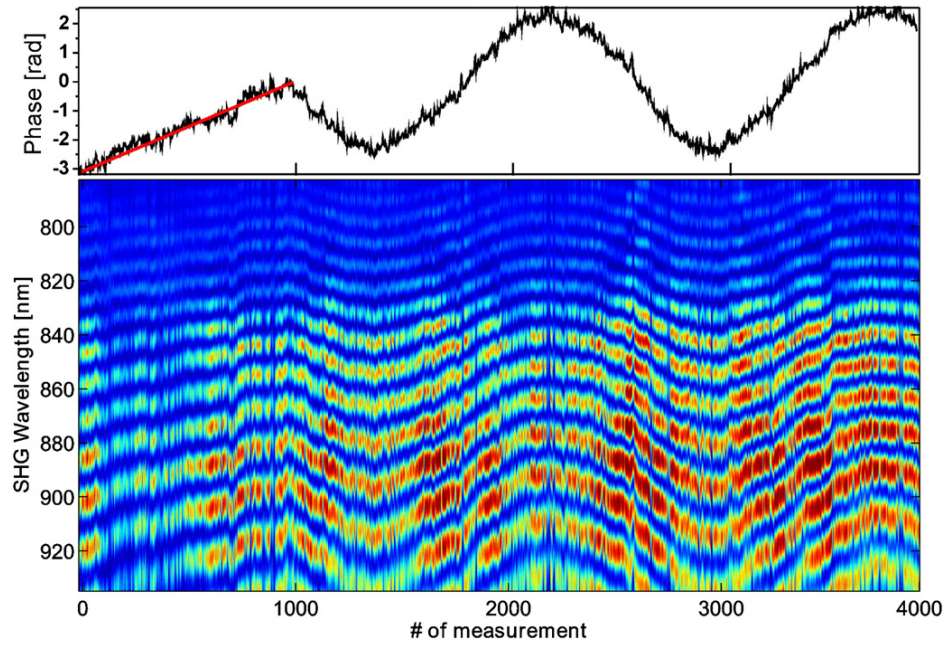


Figure 4. Passive CEP stabilization and control without a feedback loop over an acquisition time of 6.7 min. A linear phase ramp is applied for the first thousand samples and a sinusoidal modulation for the lasting part.

stability. This approach for CEP control is fast, dispersion free and fully wavelength and bandwidth independent since it circumvents changing material dispersion as done in many experiments [7].

Figure 4 demonstrates the passive CEP stabilization and CEP control without a feedback loop for a set of 4000 consecutive $f-2f$ interferograms in the lower panel. Each interferogram is averaged over ten laser shots denoting a total acquisition time of 6.7 min. The value for the relative CEP is shown in the upper graph of figure 4. For the first thousand measurements, the CEP was continuously ramped from $-\pi$ to 0 and a sinusoidal modulation was applied for the rest of the CEP scan. In the region of the CEP linear slope (red line in the upper panel), an *rms* stability of 410 mrad was evaluated based on 10-shot averaging. To reduce this value we will investigate the influence of beam pointing fluctuations when coupling into the fibre as well as from unwanted ionization stemming from residual air inside the HCF vacuum assembly.

3.4. HHG in xenon using few-cycle IR laser pulses

In this section, the application of few-cycle IR pulses for HHG is presented. In particular, we compare HHG spectra obtained at different wavelengths as well as different pulse durations. We utilized the 100 Hz, non-CEP stabilized, superfluorescence seeded OPA for HHG and the results are shown in figure 5. Spectra attained with Signal and Idler pulses, respectively, are linked to bluish and reddish colors. The thin black curve shows HHG with a long pulse of 63 fs at 800 nm wavelength at an intensity of $8.8 \times 10^{13} \text{ W cm}^{-2}$. All intensity values are calculated by fitting ionization data according to the Yudin–Ivanov ionization model [35] as described in section 2. This has the advantage of determining the intensity in the

actual interaction region regardless of focusing conditions and positioning of the focus relative to the gas jet.

An important aspect is to define conditions under which it is possible to compare HHG spectra taken at very different wavelengths (0.8–1.8 μm) and different pulse durations (23–1.8 cycles). Therefore, we present HHG spectra recorded close to the corresponding saturation intensity for each individual condition. The saturation intensity is defined when 43% of the neutral ground-state population of the gas is ionized by the most intense part of the laser pulse on axis.

Furthermore, all plots of figure 5 are normalized to their corresponding maximum at about 20 eV. The aim of this comparison, however, is not to evaluate the absolute efficiency of HHG under different conditions but to demonstrate the cut-off extension when (i) moving to longer wavelengths and (ii) using few-cycle pulses. All spectra are averaged over hundreds of laser shots to achieve a good signal-to-noise ratio in the cut-off region. The spectrum plotted as a blue curve in figure 5 is measured with a 12-cycle pulse (55 fs) at 1.4 μm . It shows a comb-shaped XUV spectrum up to the cut-off at about 65 eV, typical for HHG with multi-cycle pulses due to spectral interferences occurring from many half-cycle emissions. Reducing the Signal pulse duration to 3.2 cycles (15 fs at 1.425 μm) yields the cyan curve of figure 5 which is touching the 90 eV margin. The spectrum becomes smooth beyond 60 eV. It agrees with the picture of a reduced modulation contrast in the HHG spectrum since XUV emission is restricted to less and less half optical cycles within the pulse envelope. Moreover, the decreased modulation contrast is not due to reduced spectrometer resolution as can be seen when compared with the HHG spectrum obtained with the multi-cycle pulses at 1.82 μm , depicted as the red curve. For an equal number of 12 cycles (73 fs at 1.8 μm) for the Idler

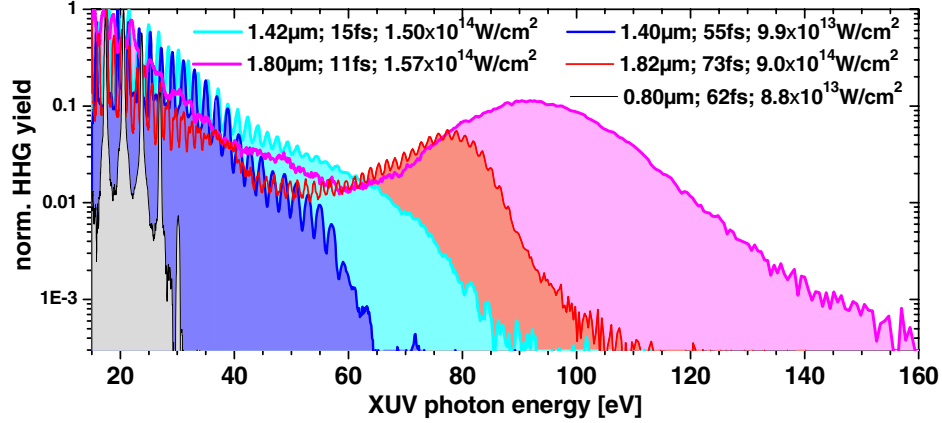


Figure 5. Semi-log plot of high harmonic yield in xenon at different driving laser wavelengths (grey: 0.8 μm ; bluish: 1.4 μm ; reddish: 1.8 μm). Each measurement was performed at intensity close to the corresponding saturation intensity and shows the benefit of increased driving wavelength and decreased pulse duration, respectively. Pulse durations and intensities are given in the legend. The striking enhancement around 95 eV originates from the giant resonance in xenon, an effect due to multi-electron correlation upon the recombination step in HHG.

Table 1. Comparison of conditions (duration, intensity) under which HHG was driven at three different wavelengths.

Wavelength (μm)	Duration (number of cycles)	Saturation intensity ($10^{14} \text{ W cm}^{-2}$)	Experimental intensity ($10^{14} \text{ W cm}^{-2}$)	Calculated cut-off (eV)	Measured cut-off (eV)
0.8	23 (long)	0.84	0.88	28.5	30
1.4	12 (long)	0.99	0.99	70	65
	3.2 (short)	1.37	1.5	102	90
1.8	12 (long)	0.98	0.90	100	105
	1.8 (short)	1.52	1.57	166	160

pulse, a clearly modulated spectrum extends out to 105 eV (red shaded curve). Table 1 summarizes the experimental finding of figure 7. Measured and calculated cut-off photon energies are in good agreement from 30 to 160 eV for all laser wavelengths and pulse durations.

Another conspicuous feature of the red curve is the fact that harmonic yield increases at the cut-off region. It rises from 55 to 80 eV before it starts to roll off in contrast to the blue curves for the OPA Signal pulses which only decrease monotonically. This increase continues even further when HHG is driven by the 1.8-cycle pulse (11 fs) at 1.8 μm . The striking enhancement at 95 eV in figure 5 is attributed to the giant resonance in xenon, which was first observed in photoionization measurements at synchrotron sources [40]. The enhancement originates from the 4d sub-shell ($\text{Xe} = [\text{Kr}] 4d^{10} 5s^2 5p^6$) and was explained theoretically by the combined effects of a shape resonance as well as collective interactions between the ten electrons that fill the 4d shell [41]. Electron exchange between the 4d and 5p shells during photoionization caused the observed increase in the photoionization cross section of the higher lying 5p state. Coming back to our measurements, it is remarkable that multi-electron correlations also take place during the time-reversed photo-recombination step in HHG. Our observation was confirmed by strong field approximation calculations in which a Coulomb interaction of the returning continuum electron (tunnel ionized from the highest 5p orbital) with the lower lying 4d shell enables energy exchange during the recombination step [31]. Table 1

summarizes the experimental finding of figure 5. Measured and calculated cut-off photon energies are in good agreement from 30 to 160 eV for all laser wavelengths and pulse durations.

It is important to consider limiting factors when aiming for a cut-off at high photon energy. We restrict the following discussion to the single atom response, i.e., neglecting all propagation phenomena and re-absorption of the medium. Obviously, the ultimate limit for an intensity increase is reached when the sample is completely ionized. From table 1, it can be seen that to further increase the saturation intensity one has to reduce the number of cycles during which ionization can deplete the neutral ground state before the maximum of the electric field. At very high intensities, however, different mechanisms like ionization gating can be beneficial for the generation of isolated attosecond pulses [42]. As demonstrated in figure 5, the cut-off extends proportional to the square of the driving laser wavelength; however, one has to pay the price of strong reduction of efficiency [36, 43]. Finally, the cut-off law is halted when the photoionization cross-section of the atom used for HHG vanishes. Because of this, HHG in xenon driven by the sub-two-cycle pulse does not follow the cut-off law anymore even if intensity is further increased (data not shown). As can be seen from the smooth spectral curve obtained with the sub-two-cycle pulse (magenta in figure 5), the entire spectrum is smooth for acquisition conditions comparable to the case of the long pulse (red curve). The spectrum is smooth not only in the cut-off region above 100 eV but also below—in an area where

the XUV spectrometer is capable of resolving individual harmonics when driven by multi-cycle pulses. This indicates that propagation effects like non-adiabatic sub-cycle dynamics might take place as discussed in [44]. The spectral FWHM of the magenta curve in figure 5 is about 30 eV centred 95 eV and corresponds to a TL pulse duration of 23 attoseconds which is close to the temporal atomic unit (24.2 attoseconds). This corresponds to the classical time it takes for the electron of a hydrogen atom to travel along one atomic unit of distance (1 atomic unit = 0.529 Å).

4. Conclusion

Multi-cycle OPA pulses are spectrally broadened in a hollow-core fibre and can be compressed with CM around 1.4 μm and solely with bulk material around 1.8 μm or longer wavelengths. This approach was successfully demonstrated on two completely different laser and OPA systems, leading to 1.6- and 1.8-cycle Idler pulses, respectively. It confirms the robustness of our approach which is based on available commercial technologies. Passive CEP stabilization was demonstrated with white light seeded OPAs. Wavelength and bandwidth-independent CEP control was carried out by shifting the phase between white light and pump beam before the DFG in the OPA. By applying tunable IR pulses for HHG, we verified experimentally that the cut-off increases proportional to the square of the driving laser wavelength. Furthermore, we show that saturation intensities at a given wavelength can be increased when driving HHG with few-cycle pulses. Finally, 1.8-cycle pulses at 1.8 μm wavelength enabled cut-off extension in xenon up to 160 eV, enabling for the first time observation of multi-electron correlation effects in HHG around 100 eV, in particular the giant resonance in xenon.

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References

- [1] Fork R L, Brito Cruz C H, Becker P C and Shank C V 1987 *Opt. Lett.* **12** 483
- [2] Mathies R A, Brito Cruz C H, Pollard W T and Shank C V 1988 *Science* **240** 777
- [3] Becker P C, Fragnito H L, Brito Cruz C H, Fork R L, Cunningham J E, Henry J E and Shank C V 1988 *Phys. Rev. Lett.* **61** 1647
- [4] Becker P C, Fork R L, Brito Cruz C H, Gordon J P and Shank C V 1988 *Phys. Rev. Lett.* **60** 2462
- [5] Fragnito H L, Bigot J-Y, Becker P C and Shank C V 1989 *Chem. Phys. Lett.* **160** 101
- [6] Nisoli M, De Silvestri S and Svelto O 1996 *Appl. Phys. Lett.* **68** 2793
- [7] Apolonski A, Poppe A, Tempea G, Spielmann C, Udem Th, Holzwarth R, Hänsch Th W and Krausz F 2000 Controlling the phase evolution of few-cycle light pulses *Phys. Rev. Lett.* **85** 740
- [8] Jones D J, Diddams S A, Ranka J K, Stentz A, Windeler R S, Hall J L and Cundiff S T 2000 Carrier-envelope phase control of femtosecond mode-locked lasers and direct optical frequency synthesis *Science* **288** 635
- [9] Hentschel M, Kienberger R, Spielmann C, Reider G A, Milosevic N, Brabec T, Corkum P, Heinzmann U, Drescher M and Krausz F 2001 Attosecond metrology *Nature* **414** 509
- [10] Schultze M *et al* 2010 Delay in photoemission *Science* **328** 1658
- [11] Eckle P, Pfeiffer A, Cirelli C, Staudte A, Dörner R, Müller H G, Büttiker M and Keller U 2008 Attosecond ionization and tunneling delay time measurements *Science* **322** 1525
- [12] Goulielmakis E *et al* 2010 Real-time observation of valence electron motion *Nature* **466** 739
- [13] Sansone G *et al* 2010 Electron localization following attosecond molecular photoionization *Nature* **465** 763
- [14] Cavalieri A L *et al* 2007 Attosecond spectroscopy in condensed matter *Nature* **449** 1029
- [15] Cavalieri A *et al* 2007 Intense 1.5-cycle near infrared laser waveforms and their use for the generation of ultra-broadband soft-x-ray harmonic continua *New J. Phys.* **9** 242
- [16] Goulielmakis E *et al* 2008 Single-cycle nonlinear optics *Science* **320** 1614
- [17] Corkum P B and Krausz F 2007 Attosecond science *Nature Phys.* **3** 381
- [18] Takahashi E J, Kanai T, Ishikawa K L, Nabakawa Y and Midorikawa K 2008 Coherent water window x-ray by phase-matched high-order harmonics *Phys. Rev. Lett.* **101** 253901
- [19] Shan B and Chang Z 2001 Dramatic extension of the high-order harmonic cut-off by using a long-wavelength driving field *Phys. Rev. A* **65** 011804
- [20] Popmintchev T, Chen M C, Bahabad A, Gerrity M, Sidorenko P, Cohen P, Christov P, Murnane M M and Kapteyn H C 2009 Phase matching of high harmonic generation in the soft and hard x-ray regions of the spectrum *Proc. Natl Acad. Sci.* **106** 10516
- [21] Chen M-C, Arpin P, Popmintchev T, Gerrity M, Zhang B, Seaberg M, Popmintchev D, Murnane M M and Kapteyn H C 2010 Bright, coherent, ultrafast soft x-ray harmonics spanning the water window from a tabletop light source *Phys. Rev. Lett.* **105** 173901
- [22] Corkum P B 1993 Plasma perspective on strong field multiphoton ionization *Phys. Rev. Lett.* **71** 1994
- [23] Gu X *et al* 2007 Generation of carrier-envelope-phase-stable 2-cycle 740- μJ pulses at 2.1- μm carrier wavelength *Opt. Exp.* **17** 62
- [24] Hong K-H, Huang S-W, Moses J, Fu X, Lai C-J, Cirmi G, Sell A, Granados E, Keathley P and Kärtner F X 2011 High-energy, phase-stable, ultrabroadband kHz OPCPA at 2.1 μm pumped by a picosecond cryogenic Yb:YAG laser *Opt. Exp.* **19** 15538
- [25] Hauri C P *et al* 2007 Intense self-compressed, self-phase-stabilized few-cycle pulses at 2 μm from an optical filament *Opt. Lett.* **32** 868
- [26] Mücke O D, Ališauskas S, Verhoeef A J, Pugžlys A, Baltuška A, Smilgevičius V, Pocius J, Giniūnas L, Danielius R and Forget L 2009 Self-compression of millijoule 1.5 μm pulses *Opt. Lett.* **34** 2498
- [27] Vozzi C, Calegari F, Benedetti E, Gasilov S, Sansone G, Cerullo G, Nisoli M, De Silvestri C and Stagira C 2007 Millijoule-level phase-stabilized few-optical-cycle infrared parametric source *Opt. Lett.* **32** 2957

- [28] Schmidt B E *et al* 2010 Compression of 1.8 micron laser pulses to sub two optical cycles with bulk material *Appl. Phys. Lett.* **96** 121109
- [29] Béjot P, Schmidt B E, Kasparian J, Wolf J-P and Légaré F 2010 Mechanism of hollow-core-fiber infrared supercontinuum compression with bulk material *Phys. Rev. A* **81** 063828
- [30] Schmidt B E, Shiner A D, Lassonde P, Kieffer J-C, Corkum P B, Villeneuve D M and Légaré F 2011 CEP stable 1.6 cycle laser pulses at 1.8 μm *Opt. Exp.* **19** 6858
- [31] Shiner A D, Schmidt B E, Trallero-Herrero C, Wörner H J, Patchkovskii S, Corkum P B, Kieffer J C, Légaré F and Villeneuve D M 2011 *Nature Phys.* **7** 464
- [32] Baltuška A, Fuji T and Kobayashi A 2002 Controlling the carrier-envelope phase of ultrashort light pulses with optical parametric amplifiers *Phys. Rev. Lett.* **88** 133901
- [33] Giguère M, Schmidt B E, Shiner A D, Houle M-A, Bandulet H C, Tempea G, Villeneuve D M, Kieffer J-C and Légaré F 2009 Pulse compression of submillijoule few-optical-cycle infrared laser pulses using chirped mirrors *Opt. Lett.* **34** 1894
- [34] Li C, Wang D, Song L, Liu J, Liu P, Xu C, Leng Y, Li R and Xu Z 2011 Generation of carrier-envelope phase stabilized intense 1.5 cycle pulses at 1.75 μm *Opt. Exp.* **19** 6783
- [35] Yudin G L and Ivanov M Y 2001 *Phys. Rev. A* **64** 013409
- [36] Shiner A D *et al* 2009 Wavelength scaling of high harmonic generation efficiency *Phys. Rev. Lett.* **103** 073902
- [37] Chowdhuri M B *et al* 2007 Spectroscopic comparison between 1200 grooves/mm ruled and holographic gratings of a flat-field spectrometer and its absolute sensitivity calibration using bremsstrahlung continuum *Rev. Sci. Instrum.* **78** 023501
- [38] Yamazaki T *et al* 1999 Comparison of mechanically ruled versus holographically varied line-spacing gratings for a soft-x-ray flat-field spectrograph *Appl. Opt.* **38** 4001
- [39] Thai A, Hemmer M, Bates P K, Chalus O and Biegert J 2011 Sub-250-mrad, passively carrier-envelope-phase-stable mid-infrared OPCPA source at high repetition rate *Opt. Lett.* **36** 3918
- [40] Becker U *et al* 1989 Subshell photoionization of Xe between 40 and 1000 eV *Phys. Rev. A* **39** 3902
- [41] Kutzner M, Radojević V and Kelly H P 1989 Extended photoionization calculations for xenon *Phys. Rev. A* **40** 5052
- [42] Ferrari F, Calegari F, Lucchini M, Vozzi C, Stagira S, Sansone G and Nisoli M 2010 High-energy isolated attosecond pulses generated by above-saturation few-cycle fields *Nature Photonics* **4** 875
- [43] Tate J, Auguste T, Muller H G, Salières P, Agostini P and DiMauro L F 2007 Scaling of wave-packet dynamics in an intense midinfrared field *Phys. Rev. Lett.* **98** 013901
- [44] Jin C, Le A-T, Trallero-Herrero C A and Lin C D 2011 Generation of isolated attosecond pulses in the far field by spatial filtering with an intense few-cycle mid-infrared laser *Phys. Rev. A* **84** 043411

Chapter 8

Photo-Recombination Cross-Sections and the Cooper Minimum

This chapter is based on the following paper:

1. **A. D. Shiner**, B. E. Schmidt, C. Trallero-Herrero, P. B. Corkum, J-C. Kieffer, F. Légaré, and D. M. Villeneuve, “Observation of cooper minimum in krypton using high harmonic spectroscopy,” *Journal of Physics B: Atomic, Molecular and Optical Physics*, vol. 45, no. 7, p. 074010, 2012

8.1 Measuring Cross-Sections

In the third step of HHG, photo-recombination, the continuum electron re-encounters the parent ion where it recombines leading to the emission of an XUV photon. This is reminiscent of photo-recombination experiments where a tunable source of electrons is directed onto ions, and researchers measure the probability for an electron of a given energy to combine with an ion leading to the emission of a photon [106].

The photo-recombination transition dipole for an incoming electron described by the scattering wave state Ψ_k^+ to the ground state of an atom Ψ_i is (length form) [70]:

$$d_{k,n}(\omega) = \langle \Psi_i | \mathbf{r} \cdot \hat{\mathbf{n}} | \Psi_k^+ \rangle \quad (8.1.1)$$

where the continuum electron initially has momentum $\mathbf{k}$, and recombination leads to

the emission of a photon polarized along $\hat{n}$. Researchers normally detect the power spectrum of the emitted photons, which can be calculated as Fourier components of the *induced dipole* $\mathbf{d}(t)$ acceleration:

$$P(\omega) \propto |a(\omega)|^2 = \left| \int \frac{d^2 d(t)}{dt^2} e^{i\omega t} dt \right|^2 \approx \omega^4 |d(\omega)|^2 \quad (8.1.2)$$

In HHG, the distribution of electrons that are driven back to the parent ion, and probe the Photo-Recombination Cross-Section (PRCS), is called the returning electron wavepacket. This wave-packet can be calculated to a reasonable approximation using the Strong Field Approximation which was developed in Sec. 2.4.

8.2 Transition matrix elements

The eigen-functions for a single particle in a spherically symmetrical potential $V(r)$ are separable in spherical coordinates. Solutions take the form $r^{-1}P_{nl}(r)Y_{lm}(\Omega)$ for bound states and $r^{-1}P_{\epsilon l}(r)Y_{lm}(\Omega)$ for continuum states, where r is the radial distance, ϵ is the energy of the state, $P_{\epsilon l}$ is the radial wavefunction, and l is the orbital angular momentum. With these Ansatz, the one-electron radial wavefunctions are found as solutions to the following differential equation [107]:

$$\frac{d^2 P_{\epsilon l}}{dr^2} + 2 \left[\epsilon - V(r) - \frac{l(l+1)}{2r^2} \right] P_{\epsilon l}(r) = 0. \quad (8.2.1)$$

Many of the features of the Photoionization Cross-Section (PICS), for atoms like argon and xenon can be understood by considering a single active electron in a suitable effective potential:

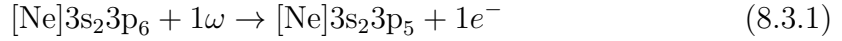
$$V_{eff}(r) \equiv V(r) + \frac{l(l+1)}{2r^2}, \quad (8.2.2)$$

which is substituted into Eq 8.2.1. For a pure Coulomb potential with nuclear charge Z and $V(r) = -Z/r$, we find that Eq. 8.2.2 is single-welled starting with a strong repulsive centrifugal barrier at small r and becoming an attractive Coulomb field at large r . The effective potentials used to model atoms with multiple electrons have more complicated behaviour, with the effective potential at positions inside the outermost orbitals deviating strongly from pure Coulombic behaviour [107]. For some cases with large angular momentum $l \geq 2$ the effective potential is double-welled

which leads to broad maxima in the PICS termed “Shape Resonances” [108, 107, 47]. Similarly, nodes in the radial wavefunction (solutions to Eq. 8.2.1), are responsible for “Cooper Minima” [109]. Both of these features have been observed in our HHG spectra and their origins are explained in the following sections.

8.3 Cooper Minima

Cooper minima originate from a node in the bound state wavefunction, which causes one of the dipole transition matrix elements to go to zero at a particular energy. In HHG, Cooper minima have been observed in argon [110, 111] but had not been observed in krypton or xenon due to the limited bandwidth for high harmonics produced with 800 nm lasers. We have now generated an HHG spectrum in krypton with our 1.8 μm , 11 fs source, which shows a clear Cooper minimum at 85 eV. The resulting publication is presented at the end of this chapter. As an example of a Cooper minimum consider the photoionization for neutral argon, which proceeds as:



where the radial matrix element connects the ground state: $|n=3, l=1\rangle$ to the continuum $|\epsilon, l=0, 2\rangle$. The transition dipole is given by:

$$D \propto c_0 a_0 \langle n=3, l=1 | \mathbf{r} | \epsilon, l=0 \rangle + c_2 a_2 \langle n=3, l=1 | \mathbf{r} | \epsilon, l=2 \rangle \quad (8.3.2)$$

where $|\epsilon, l=0\rangle$ is an s-wave scattering state, and $|\epsilon, l=2\rangle$ is a d-wave state. The coefficients $a_l = \langle Y_g^{(m=0)} | \cos(\theta) | Y_l^{(m=0)} \rangle$ come from the angular integral (which is not energy dependent), Y_l^m are the spherical harmonics, and c_l is the weighting of each partial wave. For atoms that exhibit a Cooper minimum, the ground state wavefunction has a node which causes the integrand for one of the terms in Eq. 8.3.2 to change sign. The radial wavefunctions for d-wave scattering in argon and neon, are plotted in Fig. 8.1 [108]. For neon the PICS is a monotonically decreasing function of photon energy because both the ground state $|n=2, l=1\rangle$ and the corresponding d-wave matrix element $\langle n=2, l=1 | \mathbf{r} | \epsilon, l=2 \rangle$ are always positive. The situation is different for argon, where the ground state wavefunction contains a node and has a positive as well as a negative overlap with the continuum wavefunction. This leads to continuum states where the d-wave matrix element passes through zero at an energy

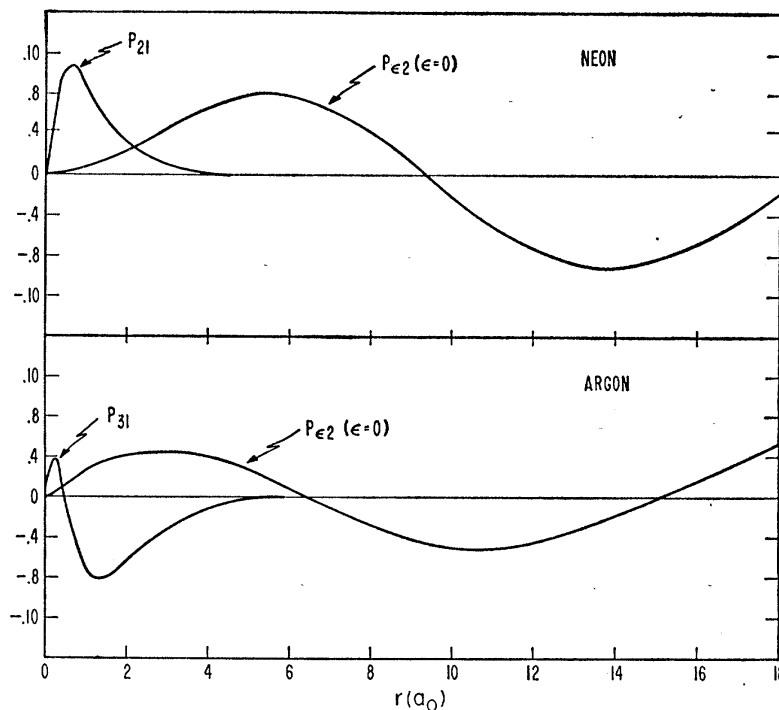


Figure 8.1: Radial wavefunction for bound states ($P_{n,l}$) as well as continuum states ($P_{\epsilon,l}$) in neon ($n=2$) and argon ($n=3$). (Figure from: [108])

of around 50 eV, resulting in a minimum in the 3p cross-section that is clearly visible in Fig. 1.8 (page 17). Figure 2 of the publication at the end of this chapter shows similar behaviour for the d-wave integral for the krypton PICS. Krypton also exhibits a Cooper minimum which in this case occurs at around 85 eV.

8.4 Shape Resonances

As shown in the preceding section, zeros in the radial matrix element cause minima in the photoionization cross-section. In contrast, potential barrier effects can cause a significant enhancement in the cross-section. An example of this is the $4d \rightarrow \epsilon f$ transition in xenon. The cross-section for this transition starts out close to zero at the threshold for ionization, and climbs to a broad maximum near 100 eV. This distinctly non-hydrogenic behaviour is caused by a shape resonance. Chapter 10 presents xenon results where this type of enhancement is observed for the first time in gas-phase HHG.

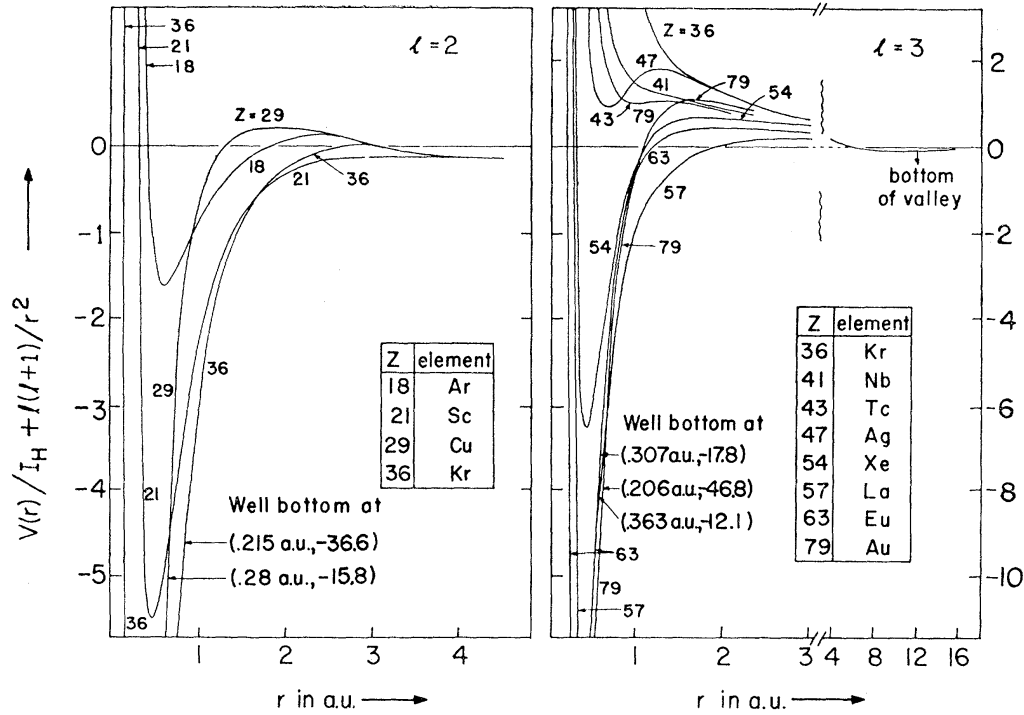


Figure 8.2: Effective potential for xenon (right panel curve 54) (Figure from: [108])

This section shows how the origin of the enhancement can be understood qualitatively with a one electron effective potential model. The effective potential for the xenon 4d state is plotted in Fig. 8.2 (right panel curve 54) [108]. This curve is double wellled, with a barrier between roughly 1 a.u. and 4 a.u. The consequence of this is that the final-state radial wavefunctions have very low amplitude at small r for photoelectron energies that are comparable to the barrier height [107]. For higher-energy continuum states the barrier concentrates the wavefunction amplitude inside the barrier region, where there is strong overlap with the bound 4d wavefunction. As explained in [107], the 4d state is an eigen-function of the inner well and the 4f and $\epsilon = 0.0$ a.u. states are eigen-functions of the outer well. The $\epsilon = 0.3$ a.u state is an eigen-function of both wells. The PICS for the Xe 4d state is plotted in Fig. 8.4 using the Herman Skillman potential [112]. The calculated curve 8.4 (a) has the expected behaviour of a maximum that is delayed from threshold and strongly peaked. The measured cross-section Fig. 8.4 (b) is broader and peaks at higher energy. This behaviour is correctly accounted for by including electron correlations [113, 114].

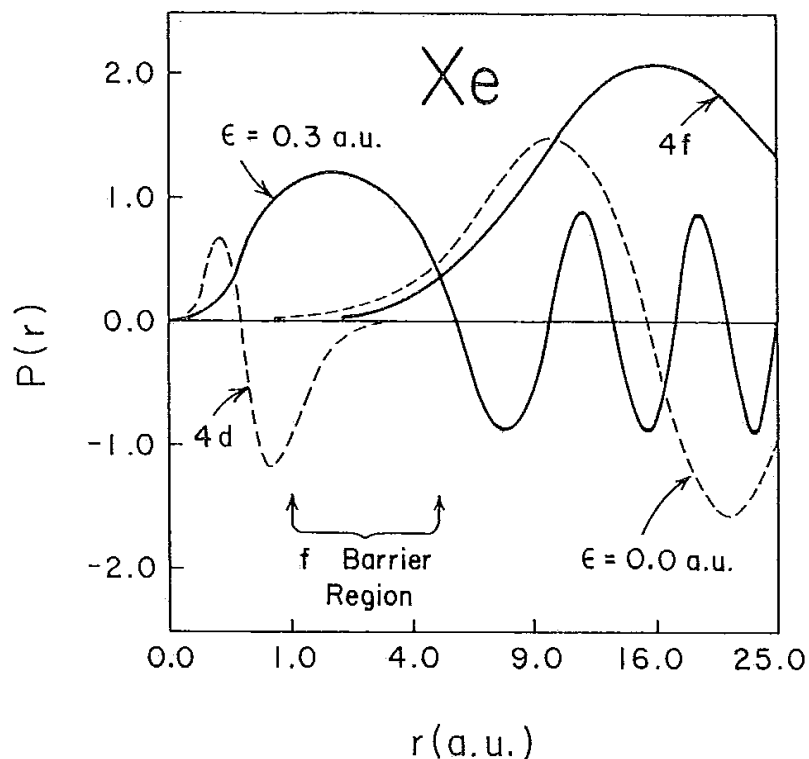


Figure 8.3: Radial wavefunctions for the xenon 4d state, the 4f excited state, and the $\epsilon = 0.0$ a.u. and $\epsilon = 0.3$ a.u. continuum states (all normalized to unity). The barrier from the effective potential prevents the 4f wavefunction from overlapping with the 4d state. The continuum wavefunctions also have minimal overlap with the 4d state for energies less than 0.3 a.u. Energies above 0.3 a.u. are comparable to the height of the barrier, causing the the continuum wavefunction to accumulate inside the barrier region, leading to a large enhancement in the transition probability. (Figure from: [107])

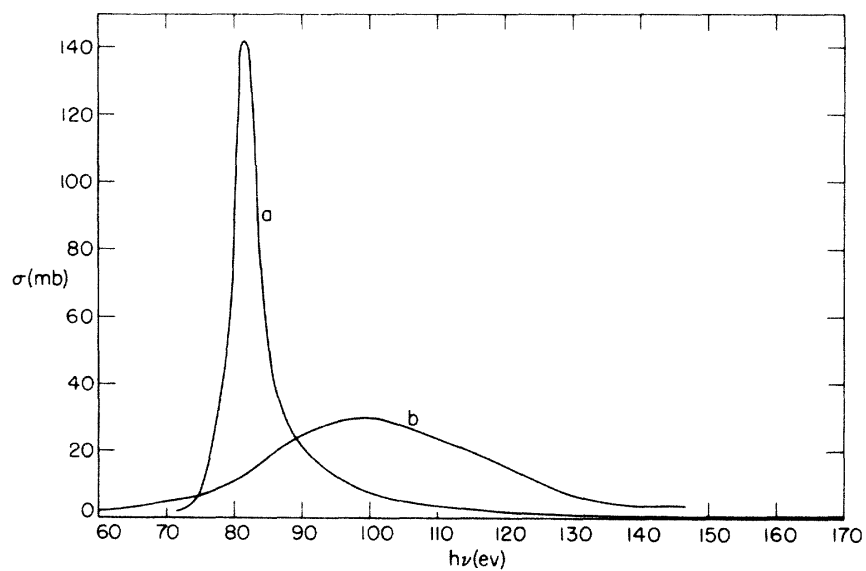


Figure 8.4: (a) Photoionization cross-section calculation for the $4d^{10}$ state of xenon, using a single active electron in a Herman-Skillman effective potential. The experimentally measured cross-section is plotted in (b). (Figure from: [112])

Observation of Cooper minimum in krypton using high harmonic spectroscopy

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Abstract

High harmonic spectroscopy utilizes the methods of attosecond science to study electronic properties of atoms and molecules. We use a 1.8 μm 11 fs laser source to generate high harmonic spectra beyond 150 eV. The Cooper minimum in krypton is clearly visible in these spectra, and would otherwise be difficult to observe with 800 nm laser sources. We relate the shape of the spectrum to the photoionization cross section of krypton.

(Some figures in this article are in colour only in the electronic version)

High harmonic generation (HHG) can be described semi-classically as a three-step process [1, 2] in which an intense laser removes an electron from an atom or molecule, accelerates the electron, and then recombines the electron with the parent ion. The resulting emission extends to the extreme ultraviolet (XUV) and is composed of odd harmonics of the driving laser frequency. HHG provides a source of attosecond pulses [3–6]. The recombination of the recolliding electron also provides a measure of the electronic structure of molecules [7, 8]. Chemical reaction dynamics have been followed by using HHG as a probe [9, 10].

The cutoff frequency in HHG is given by [11] as $\Omega_{\text{max}} = 3.17U_p + I_p$, where $U_p = F^2/(4\omega^2)$ is the ponderomotive energy, F is the peak laser electric field, ω is the laser angular frequency, and I_p is the ionization potential (atomic units are used). U_p scales as $I\lambda^2$, where I is the peak laser intensity, and λ is the laser wavelength. To extend the cutoff frequency, it is advantageous to use a longer laser wavelength [12, 13]. On the negative side, the efficiency of the HHG process falls rapidly with increasing laser wavelength [14, 15].

High harmonic spectroscopy is a field of spectroscopy in which photorecombination cross sections (PRCSs) are measured by recording the HHG spectrum [16–18]. PRCSs are related to photoionization cross sections (PICSs) through

a frequency-dependent prefactor [16, 19]. It has been shown that the PICS can be accurately retrieved to 150 eV using high harmonic spectroscopy [8]. We show here that we can record a Cooper minimum in the photorecombination of krypton.

A Cooper minimum occurs in photoionization when the transition dipole matrix element falls to nearly zero [20], usually as a result of one photoionization partial wave transition moment changing sign while the other partial wave moments are small. The Cooper minimum has been clearly seen in argon [21–24] at 50 eV. The Cooper minimum in krypton is more difficult to detect because it is at a higher energy of 85 eV, and it is not as deep. In addition, due to krypton's low ionization potential, it is difficult to get the high harmonic spectrum to extend past 80 eV using 800 nm laser sources. Using a novel laser source, we resolve the krypton Cooper minimum in HHG.

Experiments were performed at the Advanced Laser Light Source (ALLS) at INRS-EMT. The laser source for the HHG experiment is based on frequency conversion from the Ti:Sa wavelength of 800 nm to 1.8 μm with commercial optical parametric amplifier (OPA) technology followed by subsequent pulse compression. A Ti:Sa amplifier delivers 45 fs pulses with 6 mJ pulse energy at a repetition rate of 100 Hz. Frequency conversion to the IR spectral range is

achieved by utilizing the idler out of a fluorescence seeded high energy OPA (HE TOPAS, Light Conversion). It provides 52 fs pulses with pulse energy of 950 μJ which are focused into an argon-filled hollow-core fibre (HCF) by a $f = 1$ m lens made of CaF_2 . Pulse shortening is achieved with nonlinear propagation in the HCF and linear propagation through bulk material in the anomalous dispersion regime [25]. Self-phase modulation (SPM) in the 1 m long fibre is the main driving source for spectral broadening of the initial OPA spectrum. After the fibre, the pulse duration is roughly that of the input pulse but its spectrum is significantly broadened. Collimation of the continuum is achieved with a mirror of 1 m focal length.

Self-steepening causes the spectral phase to become asymmetric, distinct from pure SPM which leads to the symmetric spectrum and phase. The symmetric part of the spectral phase is compensated afterwards by the negative group delay dispersion (GDD) introduced by the glass in the anomalous dispersion regime. Compared to other approaches [26–29], the glass compression scheme achieves the cancellation of the spectral phase-asymmetry arising from self-steepening during nonlinear propagation. It is balanced by the positive third order dispersion (TOD) of the glass. Thus, compression is not limited by the TOD of the bulk material which is always positive. A more detailed discussion and full numerical simulations of the propagation effects can be found in [30].

Several materials contribute to the pulse propagation after the fibre. CaF_2 windows on the HCF (1 mm thickness) and at the entrance of the high-harmonic chamber (3 mm thickness) have a minor effect because of very low dispersion. Compression mainly arises from the transmissive polarizer (nanoparticle polarizer, Thorlabs). Fine tuning is achieved by small tilts of the fused silica plate (2.0 mm thickness). Intensity control is achieved by rotating the half-wave plate in front of the HCF in combination with the polarizer located after the fibre. Pulse characterization is carried out with a home-built second harmonic generation frequency resolved optical gating (SHG-FROG) [31] setup designed for dispersion-free few-cycle pulse measurement [25]. This setup produces 11 fs pulses, corresponding to 1.8 optical cycles at 1.8 μm wavelength, with ~ 300 μJ pulse energy delivered to the HHG target chamber.

The gas jet in the HHG chamber (Parker pulsed valve) was operated with a constant backing pressure of ~ 3 bar of krypton. The gas density in the focus was estimated to be 10^{17} cm^{-3} . The thickness of the gas sample was 0.5 mm, in all cases much shorter than the Rayleigh range of the laser focus. The laser was focused ~ 2 mm before the gas jet to maximize phase matching for the short trajectories.

Harmonic radiation produced in the gas jet passed through a vertical slit with a 140 μm width into an XUV spectrometer where the beam was spectrally dispersed by a spherical grating (Shimadzu 30-002) and then imaged onto a microchannel plate (MCP) backed by a phosphor screen (Burle APD 3115 32/25/8 I EDR MgF2 P20). The resulting spectrum was recorded with a high dynamic range digital camera. In all cases, we averaged the observed spectra over many laser shots. The resulting harmonics propagated as a low divergence beam with

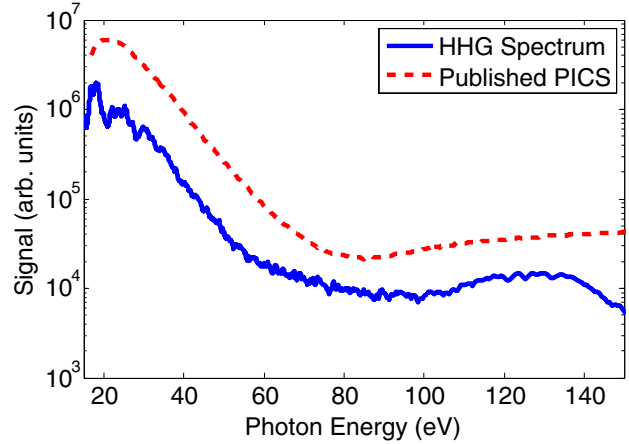


Figure 1. The blue curve shows the measured HHG spectrum from a Kr target. The raw spectrum $S(\lambda)$ has been converted from wavelength to frequency by multiplying by the Jacobian, i.e. $S(\Omega) = S(\lambda) d\lambda/d\Omega$. A minimum is evident around 90 eV. The dotted red curve shows the PICS for Kr [32]. The two curves are shifted vertically by an arbitrary amount in order to show them separately.

no visible annular structure, consistent with short trajectory only.

The HHG spectrum of Kr is shown in figure 1 for a laser intensity of $1.8 \times 10^{14} \text{ W cm}^{-2}$. The spectrum extends beyond 150 eV. The raw spectrum from the XUV spectrometer has been corrected in the conversion from wavelength to frequency by multiplying by the Jacobian, i.e. $S(\Omega) = S(\lambda) d\lambda/d\Omega$. Also shown in the figure is the experimentally measured PICS [32] for Kr, showing a minimum around 85 eV.

The PRCS is related to the more common PICS by the relation [16, 19]

$$\frac{d^2\sigma^r}{\omega^2 d\Omega_n d\Omega_k} = \frac{d^2\sigma^i}{c^2 k^2 d\Omega_n d\Omega_k}. \quad (1)$$

Here, Ω_k is the solid angle of the emitted photoelectron, Ω_n is the solid angle of the photon polarization direction, and k is the final momentum of the photoelectron. In photoionization experiments, the outgoing electron direction is measured relative to the photon polarization direction as angle θ , and its anisotropy is characterized by the asymmetry parameter β , where $-1 < \beta < 2$. In HHG from atoms, the XUV is polarized parallel to the recolliding electron direction, so $\hat{n} \parallel \hat{k}$ and $\theta = 0$. The angle-resolved PICS is related to the angle-integrated cross section and the asymmetry parameter as follows [33]:

$$\frac{d\sigma^i}{d\Omega} = \frac{\sigma^i}{4\pi} [1 + \beta^i P_2(\cos(\theta))], \quad (2)$$

where $P_2(\cos(\theta)) \equiv \frac{3}{2} \cos^2(\theta) - \frac{1}{2}$. To compare our measured HHG spectra with previously published PICSs $\sigma^i(E)$ and asymmetry parameters $\beta^i(E)$, we use equation (2) to determine the $\theta = 0$ contribution.

We proceed to demonstrate that the observed minimum is a Cooper minimum [20]. A Cooper minimum occurs when the PICS goes through a minimum. The infrared laser will remove an electron from the 4p orbital; therefore,

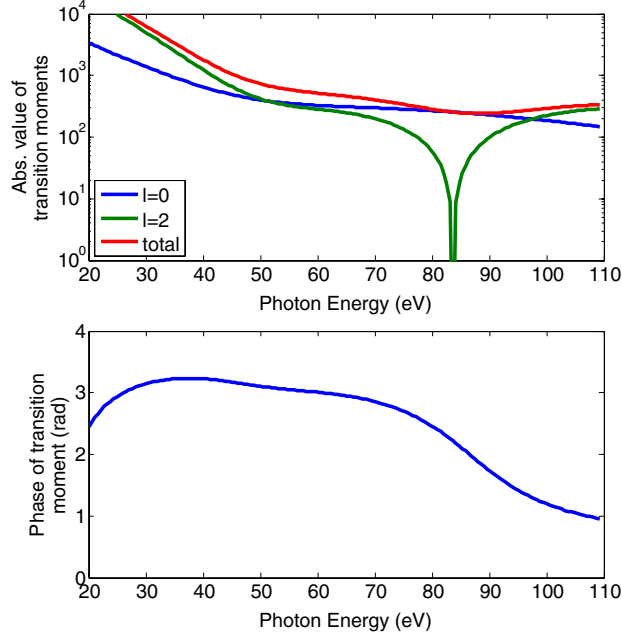


Figure 2. Top: the calculated absolute values of the transition dipoles for krypton for continuum wavefunction partial wave components $\ell = 0$ and $\ell = 2$. The total transition moment shows a minimum around 85 eV, which is caused by the $\ell = 2$ component going through a minimum at 83 eV. Bottom: the phase of the total transition moment as a function of photon energy. There is a gradual phase shift between 80 and 100 eV, showing the expected behaviour around a Cooper minimum.

the recombination will occur back to the 4p state. The Cooper minimum in argon around 50 eV is very prominent and has been extensively studied [21–24]. For both Ar and Kr, the continuum wavefunction associated with ionization from the p orbital will be composed of both s- and d-wave character. We first calculate the bound state 4p wavefunction using the procedure described by [21], which we summarize below.

The continuum wavefunction is calculated in three dimensions using an effective one-electron potential from the literature. Since the potential of the atomic ion is spherically symmetric, the electronic wavefunction is represented as a product of radial and angular components:

$$\psi_c(r, \theta, \phi) = \sum_{\ell m} \frac{1}{r} R_{\ell}(r) Y_{\ell}^m(\theta, \phi), \quad (3)$$

where R_{ℓ} is the radial wavefunction and Y_{ℓ}^m are spherical harmonics. The radial coordinate is treated numerically, whereas the two angular coordinates are treated analytically using angular momentum algebra. We use the independent particle model potential from [34] and include the centrifugal term for the reduced wave equation:

$$V_{\ell}(r) = \frac{\ell(\ell+1)}{2r^2} - \frac{1}{r} - \frac{Z-1}{H(e^{r/d}-1)+1} \quad (4)$$

with parameters $Z = 36$, $d = 0.617$, $H = 2.532$. The continuum functions are generated by numerically integrating the radial wave equation for a positive kinetic energy of the electron using the Numerov algorithm. To normalize the

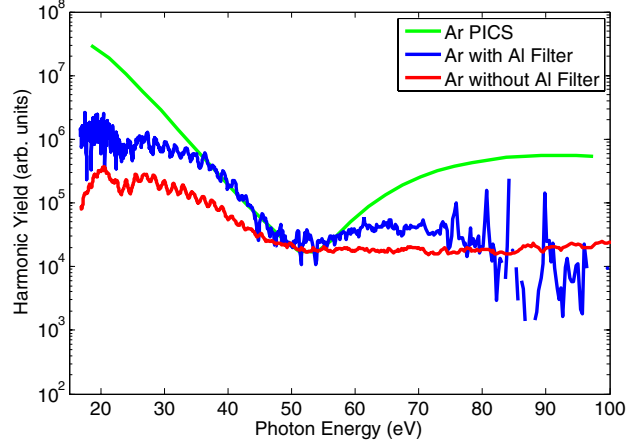


Figure 3. High harmonic spectra generated in argon. The red curve shows the spectrum recorded with the XUV spectrometer in the usual way. The Cooper minimum that is located around 50 eV is indistinct and has less contrast than previously seen. The lower contrast is due to higher frequency emission appearing at lower frequencies due to second order diffraction from the diffraction grating. When an aluminium filter is placed in the XUV beam (blue curve) with an L-shell absorption edge at 75 eV, the higher emission is cut, thereby increasing the contrast of the Cooper minimum and shifting it to 52 eV. The green curve shows the calculated PICS, from [21]. Note that the HHG spectrum shown in figure 1 was recorded with a different diffraction grating with reduced second order diffraction efficiency.

continuum wavefunctions, we match them asymptotically at large r using the formalism of Starace [33] for a potential composed of a short range potential plus a Coulomb tail.

The bound state wavefunction ψ_{4p} is calculated by GAMESS [35] using a correlation-corrected polarized triple zeta basis set (cc-pVTZ) for krypton. The photoionization dipole moment is given by

$$d = \langle \psi_{4p} | r | \psi_c \rangle \quad (5)$$

as a function of photon energy. Note that this model accounts for loss of electron kinetic energy from the Coulomb potential, by relating the photon energy Ω to the asymptotic electron kinetic energy E by $\Omega = E + I_p$, and therefore the ‘dispersion relation’ is self-consistent. Figure 2 shows the results of this calculation. The contribution to the total dipole moment from both angular momentum components is shown. The $\ell = 2$ partial wave component goes through zero near 83 eV, resulting in a gradual phase shift between 80 and 100 eV. The total dipole moment has a small minimum around 85 eV.

We now turn to the high harmonic spectrum of argon, shown in figure 3. The Cooper minimum in argon has been extensively studied in the context of high harmonic spectroscopy [21–24], so we do not need to revisit it in detail. The purpose of this section is to highlight a possible shortcoming of using a broad high harmonic spectrum. In particular, we show that the spectral content at higher energy can appear at lower energy due to second order diffraction of the diffraction grating used to disperse the spectrum. In most previous experiments, the second order diffraction is not an issue, because the HHG spectrum with 800 nm sources falls off

with increasing XUV photon energy. With near-infrared laser sources, particularly few-cycle sources, the HHG spectrum can be flat or even show a peak at a higher photon energy, which appears in second order in the low energy region of the spectrum.

Figure 3 shows the spectrum of argon recorded under similar conditions to krypton. The raw spectrum and one recorded with a 200 nm aluminium filter are shown. The aluminium filter has an L-edge at 75 eV that blocks emission above that energy. It can be seen that the spectrum with the aluminium filter (blue curve) clearly shows the presence of the Cooper minimum, whereas without the filter (red curve) the minimum has been filled in by energy from the 100 eV region of the spectrum by second order diffraction. We experienced a similar problem when recording the spectrum of xenon, due to the very strong emission around 100 eV which appeared as a second peak at 50 eV. This was improved when we replaced the Hitachi grating with a Shimadzu grating. The Shimadzu grating has a lower second order diffraction amplitude. All the spectra shown in this paper were recorded with the Shimadzu grating.

In conclusion, we have demonstrated that a 1.8 μm , 11 fs laser source can record a broad high harmonic spectrum in noble gases. These spectra contain features that reflect the PICS of the target, and in particular reveal the presence of Cooper minima. This work reinforces the current interpretation that HHG is a new form of spectroscopy that is sensitive to electronic properties of the valence shell of the target.

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References

- [1] Corkum P B 1993 *Phys. Rev. Lett.* **71** 1994
- [2] Schafer K J, Yang B, DiMauro L F and Kulander K C 1993 *Phys. Rev. Lett.* **70** 1599
- [3] Scrinzi A, Ivanov M Y, Kienberger R and Villeneuve D M 2006 *J. Phys. B: At. Mol. Opt. Phys.* **39** R1
- [4] Corkum P B and Krausz F 2007 *Nat. Phys.* **3** 381
- [5] Sansone G *et al* 2006 *Science* **314** 443
- [6] Chang Z and Corkum P B 2010 *J. Opt. Soc. Am. B* **27** B9
- [7] Itatani J, Levesque J, Zeidler D, Niikura H, Pépin H, Kieffer J C, Corkum P B and Villeneuve D M 2004 *Nature* **432** 867
- [8] Shiner A D, Schmidt B E, Trallero-Herrero C, Wörner H J, Patchkovskii S, Corkum P B, Kieffer J-C, Légaré F and Villeneuve D M 2011 *Nat. Phys.* **7** 464
- [9] Wörner H J, Bertrand J B, Kartashov D V, Corkum P B and Villeneuve D M 2010 *Nature* **466** 604
- [10] Wörner *et al* H J 2011 *Science* **334** 208
- [11] Lewenstein M, Balcou P, Ivanov M Y, L'Huillier A and Corkum P B 1994 *Phys. Rev. A* **49** 2117
- [12] Sheehy B, Martin J D D, DiMauro L F, Agostini P, Schafer K J, Gaarde M B and Kulander K C 1999 *Phys. Rev. Lett.* **83** 5270
- [13] Shan B and Chang Z 2001 *Phys. Rev. A* **65** 011804
- [14] Tate J, Auguste T, Muller H G, Salières P, Agostini P and DiMauro L F 2007 *Phys. Rev. Lett.* **98** 013901
- [15] Shiner A D, Trallero-Herrero C, Kajumba N, Bandulet H-C, Comtois D, Légaré F, Giguere M, Kieffer J-C, Corkum P B and Villeneuve D M 2009 *Phys. Rev. Lett.* **103** 073902
- [16] Le A-T, Lucchese R R, Tonzani S, Morishita T and Lin C D 2009 *Phys. Rev.* **80** 013401
- [17] Le A-T, Morishita T and Lin C D 2008 *Phys. Rev. A* **78** 023814
- [18] Frolov M V, Manakov N L, Sarantseva T S, Emelin M Y, Ryabikin M Y and Starace A F 2009 *Phys. Rev. Lett.* **102** 243901
- [19] Landau L D and Lifshitz E M 1977 *Quantum Mechanics: Non-Relativistic Theory (Course of Theoretical Physics vol 3)* 3rd edn (New York: Pergamon)
- [20] Cooper J W 1962 *Phys. Rev.* **128** 681
- [21] Wörner H J, Niikura H, Bertrand J B, Corkum P B and Villeneuve D M 2009 *Phys. Rev. Lett.* **102** 103901
- [22] Farrell J P, Spector L S, McFarland B K, Bucksbaum P H, Gühr M, Gaarde M B and Schafer K J 2011 *Phys. Rev. A* **83** 023420
- [23] Colosimo P *et al* 2008 *Nat. Phys.* **4** 386
- [24] Takahashi E J, Kanai T, Nabekawa Y and Midorikawa K 2008 *Appl. Phys. Lett.* **93** 041111
- [25] Schmidt B E *et al* 2010 *Appl. Phys. Lett.* **96** 121109
- [26] Hauri C P *et al* 2007 *Opt. Lett.* **32** 868
- [27] Vozzi C, Calegari F, Benedetti E, Gasilov S, Sansone G, Cerullo G, Nisoli M, Silvestri S D and Stagira S 2007 *Opt. Lett.* **32** 2957
- [28] Gu X *et al* 2009 *Opt. Express* **17** 62
- [29] Mücke O D, Ališauskas S, Verhoeef A J, Pugžlys A, Baltuška A, Smilgevičius V, Pocius J, Giniūnas L, Danielius R and Forget N. 2009 *Opt. Lett.* **34** 2498
- [30] Béjot P, Schmidt B E, Kasparian J, Wolf J-P and Légaré F 2010 *Phys. Rev. A* **81** 063828
- [31] Trebino R, DeLong K W, Fittinghoff D N, Sweetser J N, Krumbügel M A, Richman B A and Kane D J 1997 *Rev. Sci. Instrum.* **68** 3277
- [32] Huang K N, Johnson W R and Cheng K T 1981 *At. Data Nucl. Data Tables* **26** 33
- [33] Starace A F 1982 Theory of atomic photoionization *Handbuch der Physik*, vol 31 (Berlin: Springer) pp 1–121
- [34] Ganas P S and Green A E S 1971 *Phys. Rev. A* **4** 182
- [35] Schmidt M W *et al* 1993 *J. Comput. Chem.* **14** 1347

Chapter 9

High Harmonic Generation in C_2H_4

This chapter is based on the following paper:

1. C. Trallero-Herrero, B. Schmidt, **A. D. Shiner**, P. Lassonde, É. Bisson, J.-C. Kieffer, P. Corkum, D. Villeneuve, and F. Légaré, “High harmonic generation in ethylene with infrared pulses,” *Chemical Physics*, vol. 366, no. 1-3, pp. 33 – 36, 2009

To demonstrate the benefit of producing harmonics with a long wavelength source, particularly for systems with low ionization potentials, we measured HHG spectra from C_2H_4 using the $1.8\mu\text{m}$ Idler beam from the OPA, and compared this to the results with both 15 fs and 35 fs laser pulses at 800 nm. The improvement with the long wavelength source was dramatic, with the cutoff harmonic increasing from 30 eV for measurements with the 800 nm 35 fs source, to $> 70\text{ eV}$ with the $1.8\mu\text{m}$ 73 fs source. The HHG spectrum generated with the $1.8\mu\text{m}$ source largely covers the range of photon energies where the C_2H_4 photoionization cross section is non-zero.



High harmonic generation in ethylene with infrared pulses

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ABSTRACT

We produce harmonics in the molecule ethylene (C_2H_4) with two different wavelengths and three pulse durations. Due to the low ionization potential ($I_p = 10.5$ eV) of C_2H_4 , longer wavelengths are needed to extend the maximum photon energy produced. Our results show that regardless of the strong dependence of the efficiency of the harmonic generation process with wavelength, at 1820 nm the cutoff observed is larger than that obtained with 800 nm, 15 fs pulses. We obtain harmonics with energies exceeding 70 eV at 9.0×10^{13} W/cm² with a 73 fs 1820 nm pulse. Under these circumstances, the limiting factor for the further extension of the cutoff seems to be the photoionization cross section of the molecule.

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1. Introduction

Through the process of high harmonic generation (HHG), new possibilities in chemical physics have arisen. In many aspects, this process can be seen as a new type of atomic and molecular spectroscopy which has lead to the full reconstruction of a molecular orbital [1]. Tomography of molecular orbitals has yet to overcome many hurdles and for this reason alternative approaches has been proposed [2]. However, one aspect which can be improved relatively easy is to increase the spatial resolution by increasing the driving field's wavelength [3]. In the strong field approximation (SFA), the generation of harmonics can be explained in a simple, quasi-classical three step model; Ionization, acceleration under the influence of the electric field, and recombination [4]. Once the electron recombines it emits a photon with energy up to,

$$E_p = I_p + 3.17U_p, \quad (1)$$

with I_p being the ionization potential, and U_p the ponderomotive energy. Thus, the final step in HHG can be seen as the inverse process to photoionization. Therefore, the amplitude of the harmonics obtained in a single shot measurement can contain information about the inverse process; a bound electron absorbing a photon of energy ε and going to the continuum [5,1,6]. This relationship can be exploited to retrieve, in a table top experiment, information about the photoionization cross section of molecules and atoms in the XUV regime. As mentioned above, the main limitation to this approach is the fact that the maximum XUV photon energy is limited by the ponderomotive energy. Since $U_p \propto \lambda^2$, with λ the center wavelength of the driving field, it is expected that by using longer wavelengths the cutoff of the HHG would extend. Furthermore, at high intensities, molecules with low I_p ionize more easily at shorter wavelengths compared to longer wavelengths. The latter has implications for the generation of harmonics due to the fact that, after total ionization, the HHG process in the neutral molecule will cease since there is no interference between the continuum and the ground state and the amplitude of the third step (recombination) is zero. Unfortunately, it was predicted theoretically [7–9] and recently shown experimentally [10] that the efficiency of this process decreases exponentially with wavelength $HHG \propto \lambda^x$ with $x \sim -5$ to -6 . This result means that, in principle, one of the more “trivial” ways of extending the cutoff is not a viable one. We should emphasize that this unfavourable dependence of HHG with wavelength is only for a thin jet geometry where phase matching does not play a crucial role. Recent results show that in a waveguide geometry the decrease in XUV brightness with wavelength can be overcome by enhancing phase matching conditions [11].

In the case of molecular imaging, the importance of extending this cutoff lies in that, by expanding the spectrum of the XUV photons, to all possible non-zero values of the dipole moment of the ground state, we can gather enough information to image a molecular orbital [1]. Spatially this means that the photons emitted in the recombination step must have a wavelength small compared to the size of the molecular orbital features. By no means this is the only reason why a higher cutoff is desired and recently some groups have reported on the generation of harmonics with long wavelengths geared towards other applications [11–13]. In this paper, we report of a HHG spectra obtained in the molecule ethylene

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(C₂H₄) that expands to 75 eV, covering an energy range in which the photoionization cross section of C₂H₄ decreases by a factor of 60 [14].

2. Experimental setup

The experiment was conducted at the Advanced Laser Light Source (ALLS) using two laser sources. A schematic of the experiment is shown in Fig. 1. For 800 nm experiments we used a 5 kHz 500 μ J, 35 fs Ti:sapphire laser system. At this wavelength we used pulses of durations 35 fs and 15 fs. Pulse shortening was achieved via spectral broadening in Ar at atmospheric pressure in a 250 μ m diameter hollow-core fiber. Subsequent to the capillary, compensation of material dispersion and pulse compression down to 15 fs was achieved by five round trips on chirped mirrors. The same fiber can be used under vacuum, to avoid self-phase modulation and produce spatially filtered 35 fs pulses after compensating for the second order dispersion induced by the chirped mirrors (CM in Fig. 1) with the amplifier grating compressor. Pulse durations at this wavelength were measured with an auto-correlator. For the 1820 nm wavelength experiment, we utilized a 100 Hz, 100 mJ, 45 fs, Ti:sapphire laser system. Laser pulses of 7 mJ at 800 nm were directed to an optical parametric amplifier (HE-TOPAS from Light Conversion) and frequency shifted to 1820 nm idler pulses. The pulse energy and temporal duration were measured to be about 800 μ J and 73 fs. Pulse duration was measured using a second harmonic generation frequency resolved optical gating apparatus. Spatial filtering was performed using a hollow-core fiber (400 μ m diameter, 1 m long). As in the case of 800 nm, the fiber was under vacuum to avoid self-phase modulation. We verified that the input and output spectra remained identical.

For both wavelengths we focused into the fiber with a $f=1000$ mm plano-convex lens. At the output, the laser beam was collimated with an $R=2000$ mm concave silver mirror. Spatially filtering the beams was crucial in comparing different lasers since it has been observed that it can greatly increase the harmonic efficiency [15–17]. The filtered beam was focused by an $R=500$ mm concave silver mirror into a thin pulsed gas jet with a 500 μ m aperture backed with a pressure of roughly 3 atm. The jet was mounted on a three-axis manipulator for adjustment of its position. The laser energy could be continuously varied using a half-waveplate in front of the fiber, and a Germanium mirror at Brewster's angle acting as a polarizer (P in Fig. 1). The position of the laser focus relative to the center of the jet was determined using the signal of an ion detector, consisting of a mesh biased at

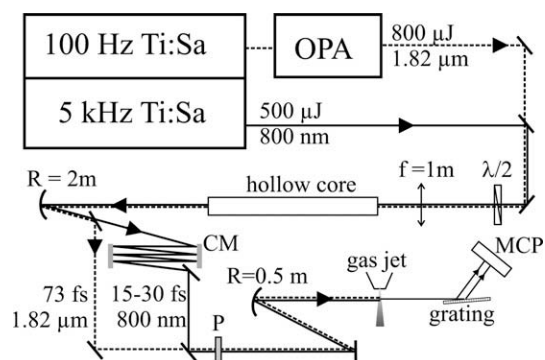


Fig. 1. Optical setup used. Solid line is the 800 nm beam path. The dashed lines show the path for the 1820 nm beam. The diameter of the hollow core fiber was 250 μ m for 800 nm and 400 μ m for the 1820 nm wavelength. For the longer 800 nm pulse (35 fs), the beam follows the same path (solid line) shown here and the dispersion introduced by the chirped mirrors (CM) was compensated by means of the compressor grating. For curved mirrors we quote the radius of curvature $R = 2 \times f$.

–500 V, a high frequency decoupling circuit and an amplifier connected to a digital oscilloscope. The ion detector assembly was held a few inches below the gas nozzle. The harmonic emission, collimated in the direction of the laser beam was dispersed by a variable groove density concave grating (Hitachi 001-0266) and imaged onto a chevron microchannel plate (MCP) sensitive to wavelengths below 100 nm.

3. Results

Since the focusing conditions vary drastically from 800 nm to 1820 nm, we could not rely on monitoring the energy and pulse durations as a measure of laser intensity. For intensity calibration, we used the space averaged ionization yield with respect to pulse energy in atomic Xe, measured in a separate experiment under the same conditions as C₂H₄. We then fit to this curve an ionization model which gives the intensity for a given pulse energy and duration. This procedure is outlined in [10] and [18]. We used the Yudin–Ivanov nonadiabatic ionization model [19] since it is capable of calculating ionization rates at all regimes, from multiphoton to pure tunnelling. By using an atomic specie for the intensity calibration we avoid many of the complications present in modeling molecular ionization. The intensity calibration for the 800 nm, 15 fs data was done in a different way. We start with the intensity calibration at the same wavelength, 35 fs pulses, using the ionization yield as described above. We then find a pulse energy where the HHG spectra from the 800 nm, 15 fs pulse has the same cutoff as the 15 fs, 800 nm pulse. Since the intensity for the 35 fs case is known and the cutoff does not depend on pulse duration, but only on peak intensity (for the same wavelength and I_p), we then know that the 15 fs pulse has the same peak intensity. This allows to obtain the proportionality factor between energy and intensity which we extrapolate for all energies. The accuracy of this process is then corroborated using Eq. (1) by observing the position of the cutoff at different peak intensities. All the spectra shown in this paper have been corrected by the transformation Jacobian from pixels in the MCP/camera to photon energy $J = \frac{d_{\text{pixel}}}{d\varepsilon}$, with ε the photon energy. Since the grating dispersion is almost linear with wavelength, the Jacobian $J(\varepsilon) \approx \frac{1}{c\varepsilon}$. This correction becomes important for high energies where the resolution of the MCP/eV is lower and different energies pile up in a single pixel, thus giving an unrealistic harmonic yield at higher energies. The XUV photon energy is calculated using the grating equation [20]. As the cutoff we used the highest harmonic that is clearly visible above the noise floor.

We produced harmonics in an isotropic sample of C₂H₄. This molecule has $I_p = 10.51$ eV which is a typical value for organic compounds. Harmonics were produced with three different laser pulses. Firstly 800 nm 35 fs, representing a typical pulse out of a commercial Ti:Sa system. Secondly, we used an 800 nm, 15 fs pulse, obtained as explained in the previous section, and finally, we used 1820 nm light from a HE-TOPAS. Fig. 2 shows harmonics produced with 800 nm, 35 fs pulses and intensity $I \approx 9.8 \times 10^{13}$ W/cm² focused in C₂H₄. As expected, the cutoff obtained is around 30 eV, confirming that at the focus there is good phase matching. This is achieved by using a 250 μ m fiber to spatially filter the otherwise Gaussian beam; a technique that has proven to increase the HHG yield [15–17]. In order to extend the cutoff, another common technique is to use ultrashort short pulses obtained after broadening the spectrum via self-phase modulation in a capillary. The cutoff extension in this case is two fold. Firstly because of the mentioned mode filtering, and secondly because short pulses allow for atoms or molecules to experience higher intensities before ionization saturation occurs. In Fig. 3, we show harmonics obtained with 800 nm, 15 fs pulses in ethylene at the following two intensities $I \approx 9.8 \times 10^{13}$ W/cm² (black curve) and

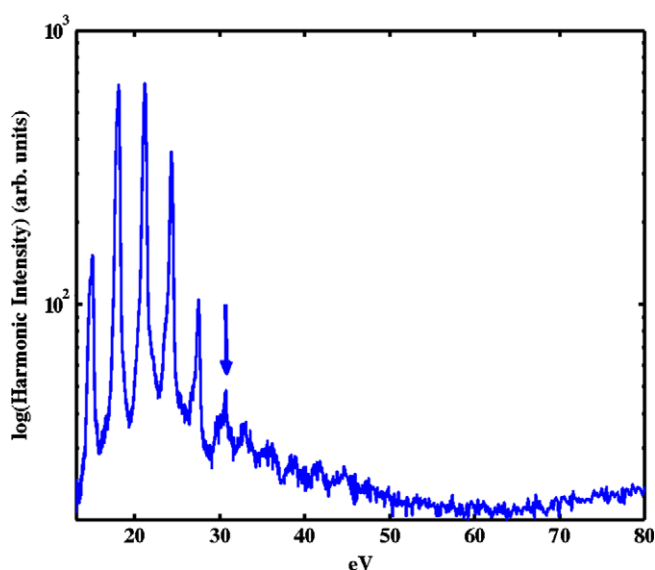


Fig. 2. Harmonics generated with 800 nm 35 fs pulses in C_2H_4 with intensity $I \approx 9.8 \times 10^{13} \text{ W/cm}^2$. The arrow points the spectrum's cutoff determined as the highest harmonic above the noise floor.

$I \approx 1.45 \times 10^{14} \text{ W/cm}^2$ (red curve). Intensities obtained by means of the semiclassical cutoff law are $I_c = 1.0 \times 10^{14} \text{ W/cm}^2$ for a cutoff of 30.4 eV and $I \approx 1.5 \times 10^{14} \text{ W/cm}^2$ for a cutoff of 40 eV. The close agreement between the calculated semiclassical and observed cutoff tells us that for these intensities HHG in ethylene is still following the cutoff law. This is a strong indication that the ionization step is still not saturated. By comparing this result to Fig. 2 we observe that the cutoff is the same when both pulses have similar intensities, as it should be according to Eq. (1) but now we can extend the maximum energy of the XUV photons by increasing the intensity of the driving field. Even though 15 fs is a very modest pulse duration compared to shorter durations obtained nowadays, it does represent a fairly easy attainable one. The spectrum at the highest intensity agrees with that obtained in [21] for similar values of the intensity.

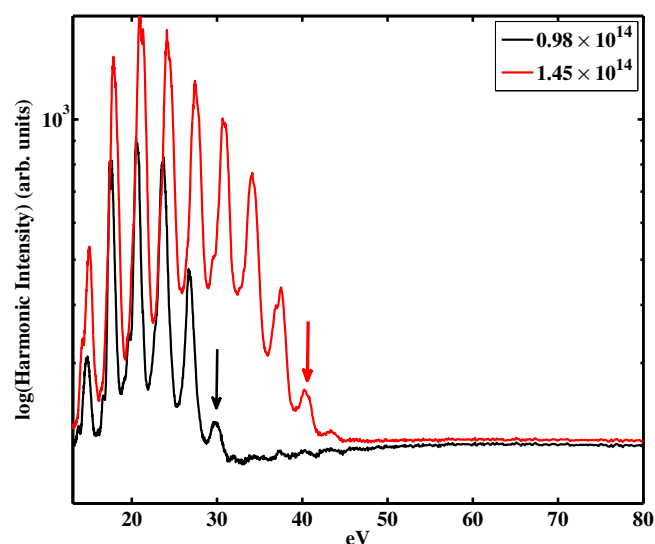


Fig. 3. Harmonics generated with 800 nm 15 fs pulses in C_2H_4 with intensities $I \approx 9.8 \times 10^{13} \text{ W/cm}^2$ (black) and $I \approx 1.45 \times 10^{14} \text{ W/cm}^2$ (red). The arrows points the spectrum's cutoff determined as the highest harmonic above the noise floor. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Finally, we proceed to use the idler beam out of the HE-TOPAS at 1820 nm. Although we obtained 800 μJ out of the HE-TOPAS, at the chamber only 250 μJ remained. These losses came from absorption in air, and poor reflectivity of the mirrors, but mainly at the fiber, where the transmission efficiency is around 50%. However, it was shown that despite great losses in this filtering process, HHG can benefit greatly from it. Ref. [15] quotes enhancements of a factor of 10 at 1400 nm, using the signal from the same HE-TOPAS. For the idler, we expect this enhancement factor to be even larger due a poorer mode quality. Fig. 4 shows harmonics obtained with 1820 nm, 73 fs and $I \approx 9.0 \times 10^{13} \text{ W/cm}^2$ pulses in ethylene. As previously mentioned, the strong power law dependence of harmonic efficiency predicts that at this wavelength the obtained spectra is roughly $(1820/800)^6 \sim 141$ times less bright than at 800 nm. To compensate for this we increased the MCP gain while insuring that the detector is still in the linear regime for amplification. Despite the very unfavourable HHG efficiency at this wavelength we obtained photon energies up to 75 eV. However, the cutoff law Eq. (1) predicts a maximum energy of 94 eV. One might argue that a reason for discrepancy is a difference in phase matching. We argue this is not the case as we spatially filtered all beams and the 800 nm produced harmonics follow Eq. (1) very closely. Another cause could be that since we are using a very long pulse the sample is totally ionized and therefore, the HHG process is saturated. Still, results shown in [21] and in this article for 800 nm predict that fairly high intensities are needed to saturate the first ionization in this molecule. Also, a large error in the intensity calibration is a possible reason for the discrepancy. At this point we have no quantitative measurement on the error for the intensity calibration method used, however, it does work remarkably well at other wavelengths where the cutoff is not as large. Furthermore, if the intensity was 20–25% smaller, the ionization rate and the harmonic process, would drop drastically due to the exponential dependence that tunnel ionization has with field strength. If this is indeed the case, the large discrepancy between the predicted and observed cutoff at this wavelength might be due to effects other than just ionization saturation or a large error in the estimation of the intensity.

According to the SFA model, there are three steps in the generation of harmonics; ionization, acceleration in the field, and recombination. The last step involves the emission of a XUV photon and can be described by the recombination dipole moment,

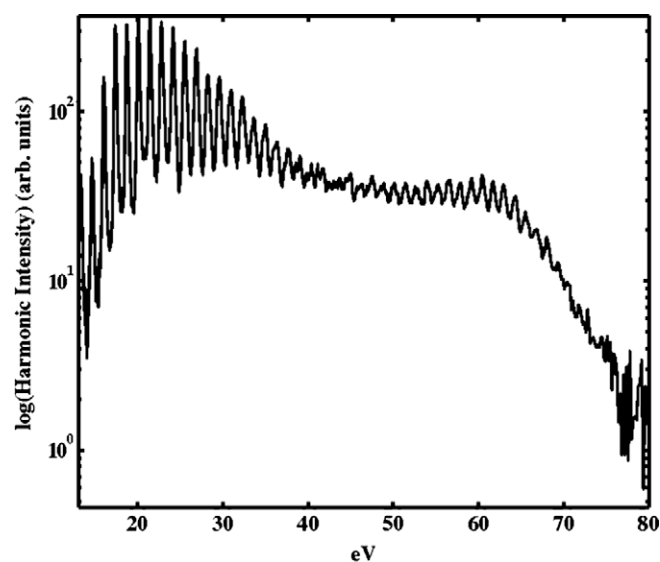


Fig. 4. Harmonics generated with 1820 nm 80 fs pulses in C_2H_4 with intensity $I \approx 9.0 \times 10^{13} \text{ W/cm}^2$.

$\langle \Phi(v) | x | \Psi_g \rangle$, where $\Phi(v)$ is the continuum wave function with electron's velocity v , and Ψ_g is the electron's ground state. This dipole moment can also describe the inverse process, that is, ionization through a photon with energy $\varepsilon = I_p + K(v)$, producing an electron with kinetic energy $K(v)$. HHG requires a non-zero photoionization cross section (dipole moment) to produce a photon of energy ε . In C_2H_4 , this matrix element approaches zero, decreasing by a factor of roughly 60 from 20 eV to 100 eV [14,22]. This will reduce the yield at high photon energies when compared to noble gases.

4. Concluding remarks

We have generated harmonics in ethylene with the following three different pulses; 800 nm with durations of 35 fs and 15 fs, and 1820 nm with a duration of 73 fs. At such a long wavelength, harmonics can be measured by spatially filtering the idler beam out of a HE-TOPAS. We find that due to the dependence of the cutoff with λ^2 , the cutoff extends reaching energies up to 75 eV. This value differs largely from the 94 eV one predicted by theory. Since we use the same spatial filter for both the 800 nm and 1820 nm experiments, and the former follows the theoretical prediction Eq. (1) quite closely, we argue that phase matching should not be the case for the large discrepancy observed in the latter. Furthermore, we observe continually increasing harmonics with intensity up to 1.45×10^{14} W/cm², at 800 nm which is an intensity much larger than the maximum intensity used at 1820 nm, thus eliminating the possibility of ionization saturation. For this case (1820 nm), the limiting factor for the maximum photon energy obtainable is likely the photoionization cross section which is related to HHG through the ionization/recombination dipole moment. At first, this could be seen as a limitation, however, it might open the possibility to study photoionization of molecules in the XUV regime, using table top optics. Perhaps of more relevance to the atomic and molecular physics community is the conclusion that we can expand the spectrum of the XUV photons, to all possible non-zero values of the dipole moment of the ground state, providing enough information to, in principle, image molecular orbitals [1]. Further experiments should still be done to test our findings. We plan to use higher peak intensities to observe a better contrast between the observed and calculated cutoff. This can be achieved by using higher pulse energies or pulse compression techniques, both of which we are currently implementing based on recent developments [23,24].

Acknowledgements

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References

- [1] J. Itatani, J. Levesque, D. Zeidler, H. Niikura, H. Pepin, J.C. Kieffer, P.B. Corkum, D.M. Villeneuve, Tomographic imaging of molecular orbitals, *Nature* 432 (2004) 867.

- [2] W. Boutu, S. Haessler, H. Merdji, P. Breger, G. Waters, M. Stankiewicz, L.J. Frasinski, R. Taieb, J. Caillat, A. Maquet, P. Monchicourt, B. Carre, P. Salieres, Coherent control of attosecond emission from aligned molecules, *Nat. Phys.* 4 (7) (2008) 545.
- [3] V.-H. Le, A.-T. Le, R.-H. Xie, C.D. Lin, Theoretical analysis of dynamic chemical imaging with lasers using high-order harmonic generation, *Phys. Rev. A* 76 (1) (2007) 013414.
- [4] M. Lewenstein, P. Balcou, M.Y. Ivanov, A.L. L'Huillier, P.B. Corkum, Theory of high-harmonic generation by low frequency laser fields, *Phys. Rev. A* 49 (1994) 2117.
- [5] T. Brabec, M.Y. Ivanov, P.B. Corkum, Coulomb focusing in intense field atomic processes, *Phys. Rev. A* 54 (4) (1996) R2551.
- [6] M.V. Frolov, N.L. Manakov, T.S. Sarantseva, M.Y. Emelin, M.Y. Ryabikin, A.F. Starace, Analytic description of the high-energy plateau in harmonic generation by atoms: can the harmonic power increase with increasing laser wavelengths?, *Phys. Rev. Lett.* 102 (24) (2009) 243901.
- [7] J. Tate, T. Augustine, H.G. Muller, P. Salieres, P. Agostini, L.F. DiMauro, Scaling of wave-packet dynamics in an intense midinfrared field, *Phys. Rev. Lett.* 98 (1) (2007) 013901.
- [8] K. Schiessl, K.L. Ishikawa, E. Persson, J. Burgdörfer, Quantum path interference in the wavelength dependence of high-harmonic generation, *Phys. Rev. Lett.* 99 (25) (2007) 253903.
- [9] M.V. Frolov, N.L. Manakov, A.F. Starace, Wavelength scaling of high-harmonic yield: threshold phenomena and bound state symmetry dependence, *Phys. Rev. Lett.* 100 (17) (2008) 173001.
- [10] A.D. Shiner, C. Trallero-Herrero, N. Kajumba, H.-C. Bandulet, D. Comtois, F. Légaré, M. Giguère, J.-C. Kieffer, P.B. Corkum, D.M. Villeneuve, Wavelength scaling of high harmonic generation efficiency, *Phys. Rev. Lett.* 103 (7) (2009) 073902.
- [11] T. Popmintchev, M.-C. Chen, A. Bahabad, M. Gerrity, P. Sidorenko, O. Cohen, I.P. Christov, M.M. Murnane, H.C. Kapteyn, Phase matching of high harmonic generation in the soft and hard X-ray regions of the spectrum, *P.N.A.S.* 106 (26) (2009) 10516.
- [12] P. Colosimo, G. Doumy, C.I. Blaga, J. Wheeler, C. Hauri, F. Catoire, J. Tate, R. Chirila, A.M. March, G.G. Paulus, H.G. Muller, P. Agostini, L.F. DiMauro, Scaling strong-field interactions towards the classical limit, *Nat. Phys.* 4 (5) (2008) 386.
- [13] G. Doumy, J. Wheeler, C. Roedig, R. Chirila, P. Agostini, L.F. DiMauro, Attosecond synchronization of high-order harmonics from midinfrared drivers, *Phys. Rev. Lett.* 102 (9) (2009) 093002.
- [14] J. Berkowitz, *Atomic and Molecular Photoabsorption: Absolute Total Cross Sections*, Academic Press, 1979.
- [15] H.-C. Bandulet, D. Comtois, A.D. Shiner, C. Trallero-Herrero, N. Kajumba, T. Ozaki, P.B. Corkum, D.M. Villeneuve, J.-C. Kieffer, F. Legare, High harmonic generation with a spatially filtered optical parametric amplifier, *J. Phys. B* 41 (24) (2008) 245602 (7pp.).
- [16] M. Nisoli, E. Priori, G. Sansone, S. Stagira, G. Cerullo, S. De Silvestri, C. Altucci, R. Bruzzese, C. de Lisio, P. Villoresi, L. Poletto, M. Pascolini, G. Tondello, High-brightness high-order harmonic generation by truncated Bessel beams in the sub-10-fs regime, *Phys. Rev. Lett.* 88 (3) (2002) 033902.
- [17] C. Altucci, R. Bruzzese, C. de Lisio, M. Nisoli, E. Priori, S. Stagira, M. Pascolini, L. Poletto, P. Villoresi, V. Tosa, K. Midorikawa, Phase-matching analysis of high-order harmonics generated by truncated Bessel beams in the sub-10-fs regime, *Phys. Rev. A* 68 (3) (2003) 033806.
- [18] S. Hankin, D. Villeneuve, P. Corkum, D. Rayner, Intense-field laser ionization rates in atoms and molecules, *Phys. Rev. A* 64 (2001) 013405.
- [19] G.L. Yudin, M.Y. Ivanov, Nonadiabatic tunnel ionization: looking inside a laser cycle, *Phys. Rev. A* 64 (1) (2001) 013409.
- [20] T. Kita, T. Harada, N. Nakano, H. Kuroda, Mechanically ruled aberration-corrected concave gratings for a flat-field grazing-incidence spectrograph, *Appl. Opt.* 22 (4) (1983) 512.
- [21] N. Kajumba, R. Torres, J.G. Underwood, J.S. Robinson, S. Baker, J.W.G. Tisch, R. de Nalda, W.A. Bryan, R. Velotta, C. Altucci, I. Procino, I.C.E. Turcu, J.P. Marangos, Measurement of electronic structure from high harmonic generation in non-adiabatically aligned polyatomic molecules, *New J. Phys.* 10 (2) (2008) 025008 (17pp.).
- [22] G. Cooper, T.N. Olney, C. Brion, Absolute UV and soft X-ray photoabsorption of ethylene by high resolution dipole (e,e) spectroscopy, *Chem. Phys.* 194 (1) (1995) 175.
- [23] E.J. Takahashi, T. Kanai, Y. Nabekawa, K. Midorikawa, 10 mJ class femtosecond optical parametric amplifier for generating soft X-ray harmonics, *Appl. Phys. Lett.* 93 (4) (2008) 041111.
- [24] M. Giguère, B.E. Schmidt, A.D. Shiner, M.-A. Houle, H.C. Bandulet, G. Tempea, D.M. Villeneuve, J.-C. Kieffer, F. Légaré, Pulse compression of submillijoule few-optical-cycle infrared laser pulses using chirped mirrors, *Opt. Lett.* 34 (12) (2009) 1894.

Chapter 10

Electron Correlation Effects in Xenon

This chapter is based on the following paper:

1. **A. D. Shiner**, B. E. Schmidt, C. Trallero-Herrero, H. J. Wörner, S. Patchkovskii, P. B. Corkum, J.-C. Kieffer, F. Légaré, and D. M. Villeneuve, “Probing collective multi-electron dynamics in xenon with high-harmonic spectroscopy,” *Nature Physics*, vol. 7, p. 464467, Mar. 2011

Chapter 8 discussed the origin of the Cooper minimum in the krypton PICS and showed that this feature is clearly visible in HHG spectra. This chapter also explained that a centrifugal barrier in the effective potential seen by a bound electron can cause the PICS to have a small value close to the threshold for ionization, and a peak at a higher energy. This peak is referred to as a ‘shape resonance,’ and a prototypical example for this is the 4d state of xenon. The energy levels for xenon are shown in Fig. 10.1, and the state resolved PICS is shown in Fig. 10.2. In the following paper we show that the effect of a shape resonance can be seen in the HHG spectra from xenon. This is at first surprising, because the shape resonance affects the 4d state ($I_p = 12.1$ eV) whereas HHG originates from the highest occupied 5p orbital ($I_p = 68$ eV). In the traditional single active electron view of HHG, an electron tunnels out from a state, accelerates in the continuum, and then returns to the state from which it came. It is essential that coherence is maintained between the continuum electron and the hole left behind in the parent ion in order for the XUV

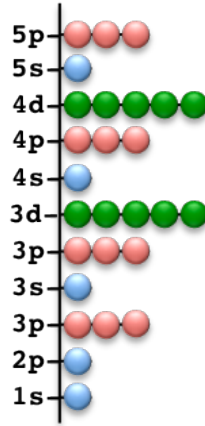


Figure 10.1: Xenon energy levels.

photon emitted during re-combination to phase-match. HHG is fundamentally different than photo-recombination in a plasma, because the HHG process for all atoms in the ensemble is driven by the same laser field, which imposes an overall phase relation between the emitters. In a plasma, the cloud of ions and electrons recombine at random times and there is no phase relation between photons emitted from different atoms. Incoherent plasma emission radiates into 4π steradians, in contrast to coherent HHG emission which can phase match and propagate as a collimated beam of radiation.

In order to explain the appearance of the shape resonance in our HHG spectra we have extended the SFA model of Ch. 2 to include an inelastic re-scattering step. When the continuum electron returns to the parent ion it inelastically scatters, giving some of its energy to the ion and promoting a 4d electron up to fill the 5p hole. Some time later the continuum electron returns to fill the 4d hole and a ~ 100 eV photon is emitted. The paper that follows shows the first observation of electron correlation and exchange effects during HHG in xenon.

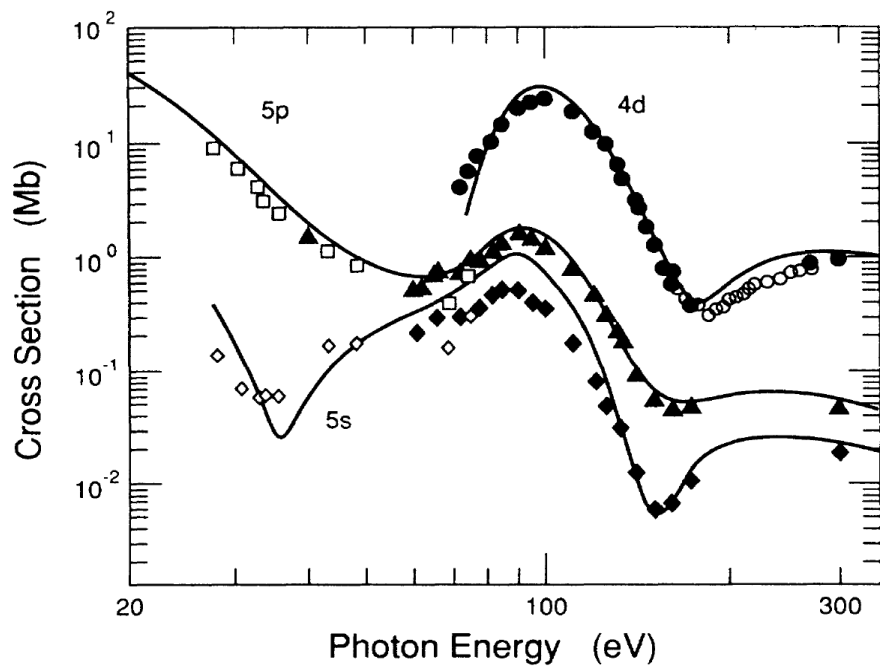


Figure 10.2: Xenon photoionization cross section. The symbols are measurements from synchrotron sources and the solid curves are from theoretical calculations. (Figure from: [114])

Probing collective multi-electron dynamics in xenon with high-harmonic spectroscopy

A. D. Shiner^{1†}, B. E. Schmidt^{1,2‡}, C. Trallero-Herrero^{1,3}, H. J. Wörner^{1,4}, S. Patchkovskii¹, P. B. Corkum¹, J.-C. Kieffer², F. Légaré² and D. M. Villeneuve^{1★}

High-harmonic spectroscopy provides a unique insight into the electronic structure of atoms and molecules^{1–5}. Although attosecond science holds the promise of accessing the timescale of electron–electron interactions, until now, their signature has not been seen in high-harmonic spectroscopy. We have recorded high-harmonic spectra of atoms to beyond 160 eV, using a new, almost ideal laser source with a wavelength of 1.8 μm and a pulse duration of less than two optical cycles. We show that we can relate these spectra to differential photoionization cross-sections measured with synchrotron sources. In addition, we show that the high-harmonic spectra contain features due to collective multi-electron effects involving inner-shell electrons, in particular the giant resonance in xenon. We develop a new theoretical model based on the strong-field approximation and show that it is in agreement with the experimental observations.

Measuring and understanding the electronic structure and correlated dynamics of matter on its natural timescale represents the main thrust of ultrafast laser science. Electron correlations affect essential properties of complex systems ranging from configuration interactions in molecules to cooperative phenomena in solids, such as superconductivity. Our knowledge of the electronic structure of matter originates from several decades of research on photoionization and photoelectron spectroscopy^{6–8}, mainly driven by the development of synchrotron-based sources. Recent advances in strong-field physics have opened an alternative approach to probing both the electronic structure^{1,9} and the dynamics^{10–12} of molecules using table-top laser sources. These new methods rely on the recollision of an electron, removed from the molecule by a strong laser field, with its parent ion¹³, as illustrated in Fig. 1a. The electronic structure of the molecule is encoded in the emitted high-harmonic spectrum through the amplitude and phase of the photorecombination matrix elements^{4,11,14,15}.

We use high-harmonic spectroscopy to investigate a new class of collective electronic dynamics—induced and probed by the recombining electron. The kinetic energy of the returning electron is usually much larger than the difference between electronic energy levels of the parent ion. Consequently, inelastic scattering followed by recombination is energetically possible, as illustrated in Fig. 1b. Using the xenon atom as an example, we demonstrate that such processes indeed occur and that they can locally enhance the efficiency of high-harmonic generation (HHG) by more than one order of magnitude. We show that such a seemingly complex pathway contributes significantly to the phase-matched process. This observation uncovers a new

unexpected facet of high-harmonic spectroscopy—it provides access to electron correlations that are otherwise very difficult to observe. Our results indicate that electron–electron excitations may be ubiquitous in high-harmonic spectroscopy experiments. High-harmonic spectroscopy gives access to multi-electron dynamics through their spectral signature, much as in photoionization studies, but it offers the additional potential of attosecond temporal resolution. Until recently, HHG was interpreted within the single-active-electron approximation. HHG experiments in preformed, transition-metal plasmas¹⁶ demonstrated significant enhancements of a single harmonic order. These enhancements were recently interpreted¹⁷ to be caused by Fano autoionizing resonances and $3d\text{--}3p$ transitions, underlining the importance of multi-electron effects in HHG.

We used a new laser source that is almost ideal for spectroscopic studies¹⁸, having a wavelength of 1.8 μm and a duration of less than 2 optical cycles. The source is described in the Methods section and in the Supplementary Information. With its long wavelength, it creates a recollision electron whose energy can exceed 100 eV, even for low-ionization-potential systems such as small organic molecules.

HHG spectra were recorded for the noble gases, argon, krypton and xenon, using a thin phase-matched gas jet described in the Methods section. Results are shown in Figs 2 and 3 and demonstrate the smooth spectra that are possible with this laser source. The limit of 160 eV is imposed by the spectrometer. We now show how the photoionization cross-section (PICS) can be measured with these spectra. It has been shown that the three-step model for HHG (ref. 13) can be approximately factorized into three terms^{1,2,15,19} corresponding to ionization (I), electron propagation (W), and recombination (σ^r). Although the range of validity for this expression has not been fully explored, it is sufficiently accurate to allow us to compare the HHG spectra of Xe and Kr, both of which involve ionization from p orbitals.

$$S(E_\Omega) = I(F, \omega)W(E)\sigma^r(E) = \mathbb{W}(E)\sigma^r(E)$$

Here $S(E_\Omega)$ is the HHG power spectrum, $E_\Omega = E + I_p$ is the emitted photon energy, E is the electron kinetic energy, ω is the laser frequency, F is the laser field and I_p is the ionization potential. The photorecombination cross-section σ^r is related to the photoionization cross-section σ^i through the principle of detailed balance^{2,20}.

To demonstrate that our laser source produces an almost flat recolliding electron wave packet, we plot the differential PICS for krypton²¹ together with our measured HHG spectrum in Fig. 2. The good agreement tells us that $\mathbb{W}(E)$ is virtually flat. The development

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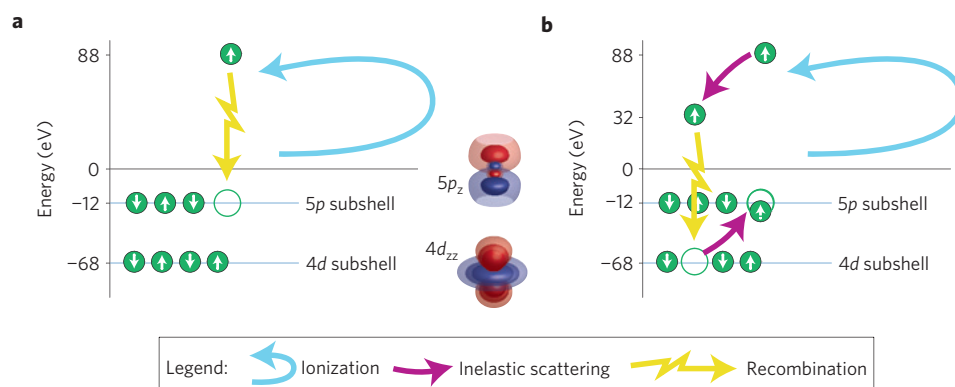


Figure 1 | Steps for harmonic generation. **a**, In the usual three-step model, an electron is tunnel ionized from the valence shell, accelerates in the continuum and then recombines to the state from which it came. **b**, With inelastic scattering, the returning electron can promote a lower-lying electron into the valence band and then recombine to the vacancy in the lower-lying state. In both cases, a 100 eV photon is emitted by recombination to a 5p vacancy (**a**) or a 4d vacancy (**b**).

of this new source allows us to see many features through HHG that were previously not accessible with traditional laser sources. For example in the krypton spectrum (Fig. 2), we observe a Cooper minimum at 80 eV. Cooper minima have been reported in HHG experiments for argon^{4,22}, but not for krypton. With our new 1.8 μm source, we are able to report the first observation of the Cooper minimum in krypton in HHG.

We now use the measured Kr HHG spectrum and the PICS from the literature to accurately determine the recolliding electron wave-packet spectrum $\mathbb{W}(E)$ by setting $\mathbb{W}(E) = S_{\text{Kr}}(E_{\Omega}) / \sigma_{\text{Kr}}^i(E_{\Omega})$, using a similar approach to that used in ref. 1. We then divide the measured HHG spectrum for xenon with this term, to extract the photoionization cross-section for xenon: $\sigma_{\text{Xe}}^i = S_{\text{Xe}}(E_{\Omega}) / \mathbb{W}(E)$. As this procedure divides one spectrum by another, experimental details such as grating reflectivity and detector response cancel out. In addition, the proportionality factor^{2,20} relating σ^i and σ^r approximately cancels out when the ionization potentials are similar. The experimentally derived σ_{Xe}^i is plotted as the blue line in Fig. 3, together with the PICS from synchrotron experiments. The excellent agreement shows that the HHG spectrum contains detailed information about the electronic structure of atoms, imprinted through the photorecombination cross-section. It is remarkable that the intense laser field can be neglected.

We now study the xenon results, shown in Fig. 3. The most striking feature is the pronounced peak around 100 eV. This peak has been extensively studied in the context of photoionization, and led to the development of a theory that included electron–electron correlations during photoionization²³. The 100 eV peak is interpreted as being due to the influence of 4d electrons, which have a large photoionization cross-section in this region owing to a shape resonance. Energy-resolved measurements²⁴ have shown that the photoionization cross-section of the 5p shell is strongly enhanced around 100 eV through electron–electron interactions with the 4d subshell.

The first step in HHG is tunnel ionization from the most weakly bound electrons, the 5p electrons in the case of xenon. The probability of tunnel ionizing a 4d electron whose binding energy is 68 eV is vanishingly small (10^{-51}). So how is it that the photorecombination process in HHG can manifest the influence of the 4d shell? Figure 1b illustrates the Coulomb interaction between the returning continuum electron and a bound 4d electron. An exchange of energy takes place, resulting in the promotion of a 4d electron to fill the 5p hole. This causes the continuum electron to lose 56 eV (the difference between binding energies of the two subshells), and hence to have the correct kinetic energy to experience the quasibound continuum state that enhances the 4d cross-section. The decelerated electron then recombines to the 4d

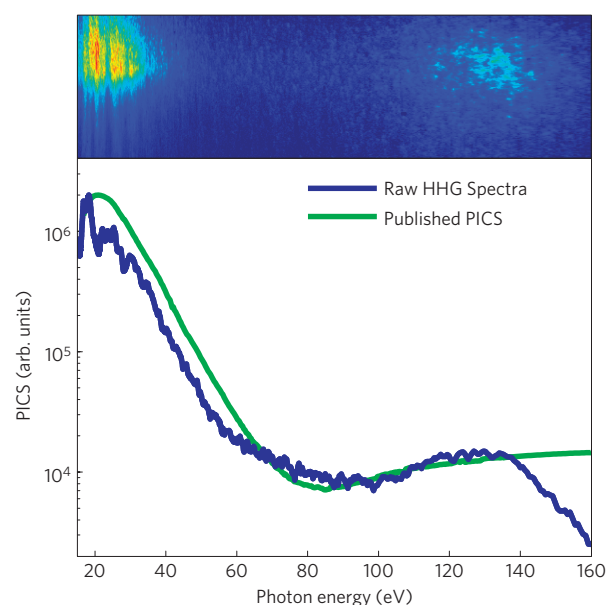


Figure 2 | Results for krypton. Top, raw HHG spectrum from krypton at $1.8 \times 10^{14} \text{ W cm}^{-2}$ in an image taken from a CCD (charge-coupled device) camera. The horizontal scale has been stretched to be linear in frequency rather than wavelength. Bottom, experimental spectrum (blue) from integrating the CCD image vertically, including the Jacobian of the transformation from wavelength to frequency; published photoionization cross-section²¹ (PICS; green).

hole and emits a photon whose energy is the same as that of the direct channel, owing to energy conservation.

The smooth green curve in Fig. 3 is derived from a complete multi-electron calculation²⁵ that includes contributions from all subshells. We now describe a simplified model in which electron correlations are included in the recombination step of HHG. In the Supplementary Information, we show how this is included in a strong-field approximation model. Briefly, we include the Coulomb interaction between the returning electron that was ionized from the 5p state and the other electrons of the ion. For clarity, we include only the 4d state, although by summation other states can be easily included. Following ref. 23, the recombination dipole moment can be written as $D(\Omega) = d_{5p} + d_{4d}A_c$, where the single electron dipole moments are $d_{5p} = \langle \phi_{5p} | r | \chi_{5p} \rangle$ and $d_{4d} = \langle \phi_{4d} | r | \chi_{4d} \rangle$. The Coulomb interaction term is $A_c = \langle \phi_{5p} \chi_{4d} | (|r_1 - r_2|^{-1}) | \phi_{4d} \chi_{5p} \rangle / \Delta I_p$. Here the bound states (labelled by ϕ) are wavefunctions in a Hartree–Fock

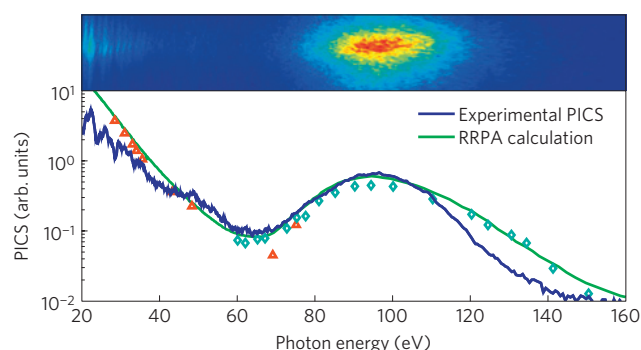


Figure 3 | Results for xenon. Top, the raw HHG spectrum from xenon at an intensity of $1.9 \times 10^{14} \text{ W cm}^{-2}$. The horizontal scale has been stretched to be linear in frequency rather than wavelength. Bottom, experimental HHG spectrum divided by the krypton wave packet (blue) and the relativistic random-phase approximation (RRPA) calculation of the xenon photoionization cross-section (PICS) from ref. 25 (green). The red and green symbols are PICS measurements from refs 31 and 24 respectively, each weighted using the anisotropy parameter calculated in ref. 25.

model, and electron exchange is included through antisymmetrized products. The continuum states correlated with the hole states are labelled as χ_{5p} and χ_{4d} . Each continuum state contains a full range of angular momentum components, only two of which contribute to the recombination.

Figure 4a shows the individual dipole moments for the $5p$ and $4d$ channels, as well as the Coulomb interaction term A_c . It can be seen that the direct $5p$ channel alone cannot explain the observed peak at 100 eV. The total calculated cross-section including the electron–electron correlations is shown in Fig. 4b, along with the experimentally recorded normalized HHG spectrum from Fig. 3. Our simplified model considers recombination with the $4d$ ($m=0$) level, ($A_c=0$ for the other m levels) and neglects further interactions between the other $4d$ electrons. The model in ref. 25, green curve in Fig. 3, includes second-order interactions between the other $4d$ electrons. We also note that the same calculation done for krypton $4p/3d$ or for xenon $5p/5s$ does not predict a giant resonance peak, in agreement with the experiment. It is the coincidence of two factors that gives rise to the 100 eV peak in xenon—the strong electron–electron coupling just above the threshold of the $4d$ level, and the strong transition dipole of the $4d$ state at the same energy.

Although we have successfully predicted the observed HHG spectrum in xenon, it is not obvious that the $4d$ channel will be phase matched, a necessity for HHG. Does the electron–electron interaction cause a loss of the coherence that is necessary for HHG phase matching? The generalized HHG model described in the Supplementary Information shows that coherence is indeed maintained. In the strong-field approximation, the ion evolves with a phase given by its total energy; the continuum electron gains a phase given by the classical action. After the collisional excitation, both the ion and the continuum electron exchange energy, but the total energy remains the same, meaning that the excitation time does not affect the emission phase, and coherence is maintained.

The observation of the 100 eV peak in xenon in the HHG spectrum is striking in several ways. It reinforces the observation that the HHG spectrum is largely determined by the photoionization cross-section^{2,4,15}. In fact it has been predicted¹⁵ that the xenon giant resonance will appear in HHG spectra. The agreement between our measured spectrum and the previously measured photoionization cross-section is remarkable because it represents a clear observation of electron–electron correlations and excitation of the ion in HHG. Indeed, it is quite likely that collisional excitation followed by recombination to an inner-shell orbital is a general phenomenon in HHG whose experimental evidence has been directly observed

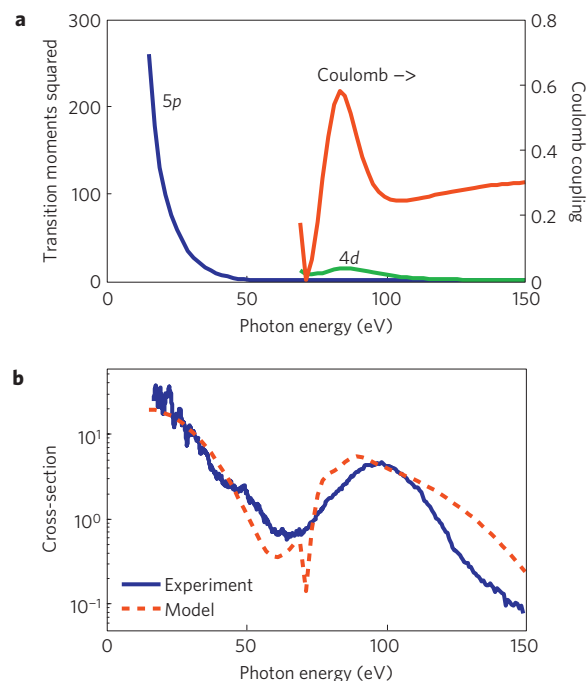


Figure 4 | Prediction of our theoretical model. **a**, The single-electron transition dipole moments (squared) for recombination to $5p$ holes and $4d$ holes in xenon. The electron–electron Coulomb interaction $|A_c|^2$ responsible for excitation of a $4d$ electron to the $5p$ hole is shown as a red curve (plotted against the right axis). **b**, The red dashed curve shows the predicted photorecombination cross-section, including both the direct $5p$ channel and the indirect electron–electron interaction through the $4d$ channel. The blue solid curve shows the experimentally obtained HHG cross-section of xenon from Fig. 3.

for the first time as a result of the newly developed sub-two-cycle infrared laser source.

Our results have three important implications. First, the high selectivity of tunnel ionization shows how photorecombination dynamics of the valence-shell orbital of molecules can be isolated and measured. In the photoionization of atoms, individual orbitals are easily identified through the kinetic energy of the photoelectrons. In molecules, however, photoelectron spectra of different orbitals overlap, inhibiting the selective observation of the PICSs over large energy intervals. Second, they show that the single-active-electron approximation can break down for HHG. Electron correlations beyond exchange interactions have been neglected until recently. We show that Coulomb correlations need to be included even at the qualitative level. Such interactions are ubiquitous in complex atoms and molecules. The giant resonance that we observe in high-harmonic spectroscopy for the first time is indeed known from photoionization experiments to persist from atoms into molecules and into the solid state. Third, owing to the factorization of HHG to explicitly include the field-free photorecombination cross-section, they demonstrate how the large body of spectroscopic data accumulated over the past decades will be useful in choosing optimal media for attosecond pulse generation in the quest to extend the photon energy^{26,27}.

Our results also suggest new opportunities for future experiments. We have shown that HHG provides access to electronic structure information of inner shells through an unexpected mechanism. This opens the possibility to study electronic structure and correlations in inner shells of molecular systems where very little is known. Giant resonances as we have observed in xenon also offer interesting systems for attosecond time-resolved studies. Converting the 30 eV measured width of the resonance into a lifetime, we

obtain 23 attoseconds. Using an attosecond pulse centred at 100 eV to photoionize xenon from the 4d shell into the region of the giant resonance and subsequently streaking the photoelectron, should reveal the signature of the multi-electron dynamics. Such a measurement carried out in neon has recently revealed an unexpected temporal shift between the emission from different subshells²⁸. Alternatively, the timescale of the electron correlations might be directly observable in high-harmonic experiments, if the electron is accelerated by the laser field after interaction, resulting in a phase shift of the emission; such an effect could be probed by varying the laser intensity or wavelength.

Methods

Source. Harmonic generation was driven by a 1.8 μm , 11 fs laser pulse. Details of this source have been published elsewhere¹⁸ and are also given in the Supplementary Information. In short, the 800 nm radiation from a Ti:sapphire laser (Thales, 100 Hz, 30 fs) was downshifted by an optical parametric amplifier (Light Conversion TOPAS-HE) producing approximately 1 mJ of radiation at 1.8 μm with a nominal pulse duration of 52 fs. This pulse was directed into a hollow-core fibre filled with 1.2 bar of argon gas, where its spectrum was broadened through self-phase modulation and self-steepening, and then compressed through anomalous dispersion in 2.0 mm of fused silica and a transmissive polarizer. This resulted in a nearly transform-limited 11 fs pulse, which is only 1.9 optical cycles in duration. The spatial quality of the beam is excellent, owing to the spatial filtering properties of the fibre. The laser pulse is collimated after the hollow-core fibre, then focused into the vacuum chamber with a mirror with a focal length of 250 mm. The focus was positioned before the gas jet, and optimized to favour phase-matched short trajectories and to suppress long trajectory harmonics.

Phase matching. Our intention is to apply high-harmonic spectroscopy to various rare-gas atoms. Unlike gas-source geometries that are designed to optimize production of extreme-ultraviolet emission, we employ a thin gas jet target in which collective effects are minimized. The gas sample is emitted from a pulsed valve with a 500 μm orifice and a backing pressure of 3 bar. We have previously verified that the extreme-ultraviolet emission scales quadratically with gas pressure²⁹, demonstrating that the target is operating in the phase-matched regime. The resulting high-harmonic spectrum is therefore a reflection of the single-atom response.

Spectrometer. Harmonic radiation produced in the gas jet passed into an extreme-ultraviolet spectrometer where the beam was spectrally dispersed by a 1,200 line mm^{-1} spherical holographic grating (Shimadzu 30-002). This type of grating has significantly lower second-order dispersion when compared with typical mechanically ruled gratings. The dispersed spectrum was imaged with a microchannel plate backed by a phosphor screen. The resulting spectrum was recorded with a high-dynamic-range digital camera.

Spectrometer calibration. The spectrometer was calibrated by putting oxygen into the pulsed jet and using several millijoules of circularly polarized 800 nm radiation while increasing the microchannel plate gain. We then recorded the plasma emission lines, using a method similar to that in ref. 30. By assigning the observed recombination lines to oxygen transitions known from the literature, we achieve a calibration of the spectrometer. Details of this procedure are given in the Supplementary Information.

Intensity calibration. The absolute laser intensity in the jet was calibrated by measuring the ion yield as a function of laser pulse energy. The intensity dependence of the mode-integrated ion yield for the target gas was then calculated using the Yudin–Ivanov non-adiabatic ionization model parameterized by the measured laser wavelength and pulse duration. The proportionality factor relating the pulse energy measured outside the chamber to the peak intensity in the interaction volume was found by fitting the measured ion yield to the intensity-dependent yield calculated from the model. We have published this procedure as part of our previous work²⁹ and show details in the Supplementary Information.

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References

- Itatani, J. *et al.* Tomographic imaging of molecular orbitals. *Nature* **432**, 867–871 (2004).
- Le, A.-T., Lucchese, R. R., Tonzani, S., Morishita, T. & Lin, C. D. Quantitative rescattering theory for high-order harmonic generation from molecules. *Phys. Rev. A* **80**, 013401 (2009).
- Mairesse, Y., Levesque, J., Dudovich, N., Corkum, P. B. & Villeneuve, D. M. High harmonic generation from aligned molecules—amplitude and polarization. *J. Mod. Optics* **55**, 2591–2602 (2008).
- Wörner, H. J., Niihara, H., Bertrand, J. B., Corkum, P. B. & Villeneuve, D. M. Observation of electronic structure minima in high-harmonic generation. *Phys. Rev. Lett.* **102**, 103901 (2009).
- Haessler, S. *et al.* Attosecond imaging of molecular electronic wavepackets. *Nature Phys.* **6**, 200–206 (2010).
- Berkowitz, J. *Photoabsorption, Photoionization and Photoelectron Spectroscopy* (Academic, 1979).
- Kimura, K., Katsumata, S., Achiba, Y., Yamazaki, T. & Iwata, S. *Handbook of Hel Photoelectron Spectra* (Japan Scientific Societies Press, 1981).
- Becker, U. & Shirley, D. A. (eds) *VUV and Soft X-Ray Photoionization* (Plenum Press, 1996).
- Torres, R. *et al.* Probing orbital structure of polyatomic molecules by high-order harmonic generation. *Phys. Rev. Lett.* **98**, 203007 (2007).
- Baker, S. *et al.* Probing proton dynamics in molecules on an attosecond timescale. *Science* **312**, 424–427 (2006).
- Smirnova, O. *et al.* High harmonic interferometry of multi-electron dynamics in molecules. *Nature* **460**, 972–977 (2009).
- Wörner, H. J., Bertrand, J. B., Kartashov, D. V., Corkum, P. B. & Villeneuve, D. M. Following a chemical reaction using high-harmonic spectroscopy. *Nature* **466**, 604–607 (2010).
- Corkum, P. B. Plasma perspective on strong field multiphoton ionization. *Phys. Rev. Lett.* **71**, 1994–1997 (1993).
- Le, A.-T., Lucchese, R. R., Lee, M. T. & Lin, C. D. Probing molecular frame photoionization via laser generated high-order harmonics from aligned molecules. *Phys. Rev. Lett.* **102**, 203001 (2009).
- Frolov, M. V. *et al.* Analytic description of the high-energy plateau in harmonic generation by atoms: Can the harmonic power increase with increasing laser wavelengths? *Phys. Rev. Lett.* **102**, 243901 (2009).
- Ganeev, R. A. High-order harmonic generation in a laser plasma: A review of recent achievements. *J. Phys. B* **40**, R213–R253 (2007).
- Frolov, M. V., Manakov, N. L. & Starace, A. F. Potential barrier effects in high-order harmonic generation by transition-metal ions. *Phys. Rev. A* **82**, 023424 (2010).
- Schmidt, B. E. *et al.* Compression of 1.8 μm laser pulses to sub two optical cycles with bulk material. *Appl. Phys. Lett.* **96**, 121109 (2010).
- Frolov, M. V., Manakov, N. L., Sarantseva, T. S. & Starace, A. F. Analytic formulae for high harmonic generation. *J. Phys. B* **42**, 035601 (2009).
- Landau, L. D. & Lifshitz, E. M. *Quantum Mechanics Non-Relativistic Theory* 3 edn (Course of Theoretical Physics, vol. 3, Pergamon Press, 1977).
- Huang, K. N., Johnson, W. R. & Cheng, K. T. Theoretical photoionization parameters for the noble gases argon, krypton, and xenon. *At. Nucl. Data Tables* **26**, 33–45 (1981).
- Minemoto, S. *et al.* Retrieving photorecombination cross sections of atoms from high-order harmonic spectra. *Phys. Rev. A* **78**, 061402(R) (2008).
- Amusia, M. Y. & Connerade, J.-P. The theory of collective motion probed by light. *Rep. Prog. Phys.* **63**, 41–70 (2000).
- Becker, U. *et al.* Subshell photoionization of Xe between 40 and 1,000 eV. *Phys. Rev. A* **39**, 3902–3911 (1989).
- Kutzner, M., Radojević, V. & Kelly, H. P. Extended photoionization calculations for xenon. *Phys. Rev. A* **40**, 5052–5057 (1989).
- Popmintchev, T. *et al.* Phase matching of high harmonic generation in the soft and hard X-ray regions of the spectrum. *Proc. Natl Acad. Sci. USA* **106**, 10516–10521 (2009).
- Vozzi, C. *et al.* High-order harmonics generated by 1.5 μm parametric source. *J. Mod. Opt.* **57**, 1008–1013 (2010).
- Schultze, M. *et al.* Delay in photoemission. *Science* **328**, 1658–1662 (2010).
- Shiner, A. D. *et al.* Wavelength scaling of high harmonic generation efficiency. *Phys. Rev. Lett.* **103**, 073902 (2009).
- Farrell, J. P., McFarland, B. K., Bucksbaum, P. H. & Gühr, M. Calibration of a high harmonic spectrometer by laser induced plasma emission. *Opt. Express* **17**, 15134–15144 (2009).
- Fahlman, A., Krause, M. O., Carlson, T. A. & Svensson, A. Xe 5s, 5p correlation satellites in the region of strong interchannel interactions, 28–75 eV. *Phys. Rev. A* **30**, 812–819 (1984).

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Author contributions

A.D.S. and B.E.S. contributed equally to this work. B.E.S., F.L. and J.-C.K. developed the laser source. A.D.S., B.E.S. and C.T.H. carried out the experiments. H.J.W. interpreted the xenon results. A.D.S. analysed the data. S.P. and D.M.V. provided the theoretical parts. A.D.S., H.J.W., P.B.C. and D.M.V. wrote the manuscript.

Additional information

The authors declare no competing financial interests. Supplementary information accompanies this paper on www.nature.com/naturephysics. Reprints and permissions information is available online at <http://npg.nature.com/reprintsandpermissions>. Correspondence and requests for materials should be addressed to D.M.V.

Probing collective multi-electron dynamics in xenon with high harmonic spectroscopy (Supplementary Online Material)

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1 Calculating HHG Spectra

The Photo-Recombination Cross Section (PRCS) is related to the more common Photoionization Cross Section (PICS) by the relation ^{1,2}

$$\frac{d^2\sigma^r}{\omega^2 d\Omega_n d\Omega_k} = \frac{d^2\sigma^i}{c^2 k^2 d\Omega_n d\Omega_k} \quad (1)$$

Here, Ω_k is the solid angle of the photoelectron direction, Ω_n is the solid angle of the photon polarization direction, and k is the final momentum of the photoelectron. In photoionization experiments, the outgoing electron direction is measured relative to the photon polarization direction as angle θ , and its anisotropy is characterized by the asymmetry parameter β , where $-1 < \beta < 2$. In HHG from atoms, the xuv is polarized parallel to the recolliding electron direction, so $\hat{n} \parallel \hat{k}$ and $\theta = 0$. The angle resolved photoionization cross section is related to the angle integrated cross section and the asymmetry parameter as follows ³:

$$\frac{d\sigma^i}{d\Omega} = \frac{\sigma^i}{4\pi} [1 + \beta^i P_2(\cos(\theta))] \quad (2)$$

Where $P_2(\cos(\theta)) \equiv \frac{3}{2} \cos^2(\theta) - \frac{1}{2}$. To compare our measured HHG spectra with previously published photoionization cross sections $\sigma^i(E)$ and asymmetry parameters $\beta^i(E)$, we use eq. (2) to determine the $\theta = 0$ contribution, then use eq. (1) to convert to a PRCS.

Kr Published Photoionization Cross Section Data: State resolved photoionization cross sections for Kr were calculated by Huang et al.⁴ who used the Relativistic Random Phase Approximation (RRPA). After ionization of the Kr ground state the ion is left in a superposition of the 2P_j states with $j = \{\frac{1}{2}, \frac{3}{2}\}$. The PI cross section for each state along the direction $\theta = 0$ was calculated using Eq. (2) and the contributions from the two states were combined with the statistical weighting $(2j+1)$. The resulting PI cross section is shown in Fig. 1. Huang et al. did not provide data for photon energies above ~ 140 eV. In this region the PICS was extrapolated by fitting a second order polynomial to the published PICS from 100 eV to 140 eV. The results are shown as the red curve in Fig. 1.

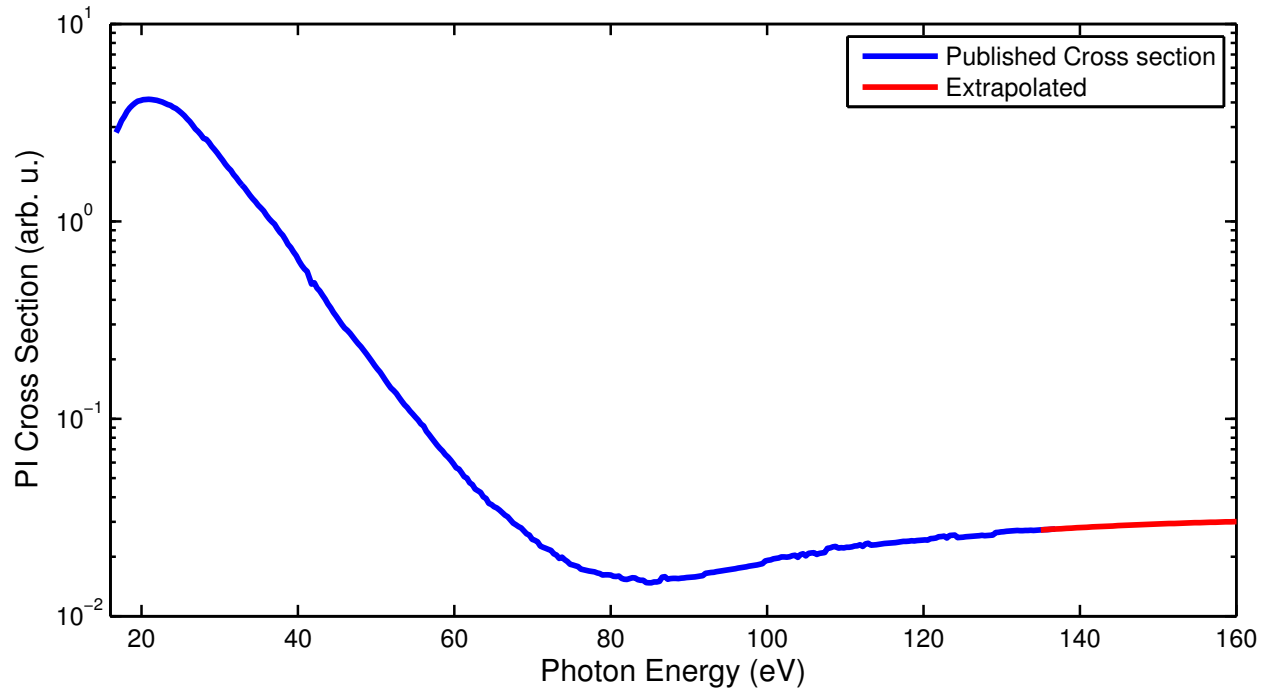


Figure 1: Photoionization cross section for Kr along the ($\theta = 0$) direction combining the contributions from channels that leave the ion in the $^2P_{1/2}$ and $^2P_{3/2}$ states. Data was taken from⁴ and extrapolated to 160 eV.

2 Strong Field Approximation

The Strong Field Approximation (SFA) is commonly used to calculate HHG emission⁵. The usual expression for the time-dependent dipole $d(t)$ in a one-electron system is

$$d_1(t) = -i \int d\mathbf{k} \int_0^t dt_1 \langle \psi_0 | \mathbf{r} | \mathbf{k} + \mathbf{A}(t) \rangle e^{-iS} \mathbf{E}(t_1) \cdot \langle \mathbf{k} + \mathbf{A}(t_1) | \mathbf{r} | \psi_0 \rangle \quad (3)$$

Here $\mathbf{k}$ is the canonical momentum, $\mathbf{A}$ is the vector potential of the laser field, and $S = \int_{t_1}^t dt'' (\mathbf{k} + \mathbf{A}(t''))^2/2 + I_p(t - t_1)$ is the classical action, with I_p being the ionization poten-

tial.

Following Patchkovskii et al.⁶, we rewrite this expression in the multi-electron form to explicitly include the ion and the various phase terms.

$$\begin{aligned} d_2(t) = & -i \int d\mathbf{k} \int_0^t dt_1 \left\langle \psi_N \left| e^{iE_N t} \mathbf{r} e^{-iE^+(t-t_1)} e^{-iS_v(t,t_1)} \right| \mathbf{k} + \mathbf{A}(t), \psi^+ \right\rangle \\ & \times \left\langle \mathbf{k} + \mathbf{A}(t_1), \psi^+ \left| \hat{V}_L(t_1) e^{-iE_N t_1} \right| \psi_N \right\rangle \end{aligned} \quad (4)$$

Here, $\psi_N(1\dots n)$ is the n -electron ground state of the neutral atom, E_N is the total energy of the neutral, $\psi^+(1\dots n-1)$ is the $n-1$ -electron wave function of the ion following ionization, E^+ is the total energy of the ion, $V_L = xE_L(t_1)$ is the interaction Hamiltonian due to the laser field, and $S_v(t, t_1) = \int_{t_1}^t (\mathbf{k} + \mathbf{A}(t''))^2 dt''/2$ is the Volkov phase of the electron in the continuum. We explicitly show the energy of the neutral system, E_N , rather than the more usual ionization potential I_p , because we will soon introduce an excited state of the ion.

Now we include the Coulomb interaction between the returning electron and the $n-1$ electrons in the ion. In the S-matrix description, this is an additional interaction at time t_2 somewhere between ionization time t_1 and emission time t . The time-ordering is $(0 \leq t_1 \leq t_2 \leq t)$. In the case of xenon, the ionic states correspond to the ion with a 5p hole (ψ_1^+) and a 4d hole (ψ_2^+). The expression can easily be extended to additional ionic states. The continuum electron wave functions associated with each ionic state are written as χ_1 and χ_2 to simplify the expression. Electron n is ionized (without showing the explicit antisymmetrization).

$$\begin{aligned} d_3(t) = & \int d\mathbf{k} \int_0^t dt_1 \int_{t_1}^t dt_2 \\ & \times \left\langle \psi_N \left| e^{iE_N t} \mathbf{r} e^{-iS'_v(t,t_2)} \right| \chi_2(t_2), \psi_2^+ \right\rangle \\ & \times \left\langle \chi_2(t_2), \psi_2^+ \left| \sum_{i=1}^{n-1} |\mathbf{r}_i - \mathbf{r}_n|^{-1} \right| \chi_1(t_2), \psi_1^+ \right\rangle \\ & \times e^{-iS_v(t_2,t_1)} \left\langle \chi_1(t_1), \psi_1^+ \left| \hat{V}_L(t_1) e^{-iE_N t_1} \right| \psi_N \right\rangle (E_2^+ - E_1^+)^{-1} \end{aligned} \quad (5)$$

Here the classical action is split into two parts, before and after t_2 :

$$S_v = \frac{1}{2} \int_{t_1}^{t_2} d\tau (\mathbf{k} + \mathbf{A}(\tau))^2 + E_1^+(t_2 - t_1) \quad (6)$$

$$S'_v = \frac{1}{2} \int_{t_c}^t d\tau K_2 + E_2^+(t - t_2), \quad (7)$$

where K_1 is the kinetic energy of continuum electron 1 before collision and $K_2 = K_1 - (E_2^+ - E_1^+)$ is the kinetic energy of continuum electron 2 after collision, which we assume is not affected by

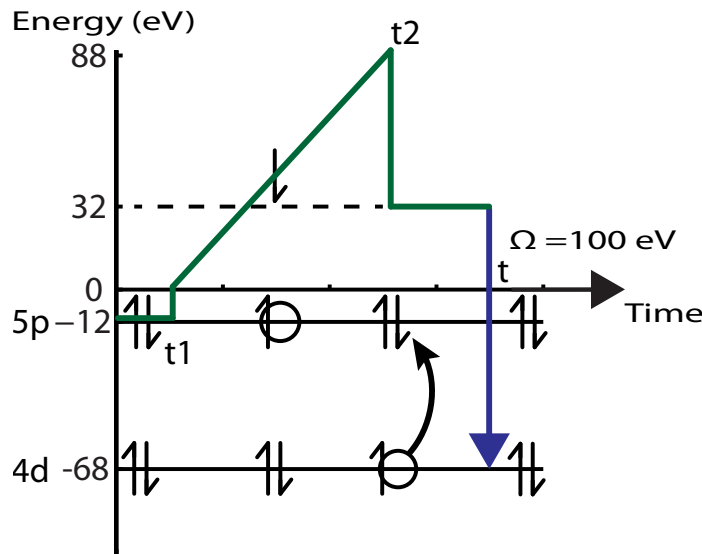


Figure 2: Time evolution of the continuum electron in HHG. From time zero to t_1 the electron waits to be ionized by the laser field. At t_1 the electron is field ionized and accelerates in the continuum. At t_2 the continuum electron re-encounters the parent ion causing the exchange of the 5p hole with a 4d electron losing 56 eV in the process. At time t the continuum electron fills the 4d hole and a 100 eV photon is emitted.

the laser field during its brief existence. A schematic diagram of this 4-step process is shown in Fig. 2.

We calculate the effect of the extra 5p-4d coupling by considering only the recombination part of the three-step model (see main text, eq. 1). The recombination moment $D(\Omega)$ is the sum of the direct 5p channel, D_{5p} and the indirect 4d channel, D_{4d} .

$$D_{5p}(\Omega) = \langle \phi_{5p} | \mathbf{r} | \chi_{5p} \rangle \quad (8)$$

$$D_{4d}(\Omega) = \langle \phi_{4d} | \mathbf{r} | \chi_{4d} \rangle \left\langle \chi_{4d}, \phi_{5p} \left| |\mathbf{r}_1 - \mathbf{r}_2|^{-1} \hat{A} \right| \chi_{5p}, \phi_{4d} \right\rangle (E_2^+ - E_1^+)^{-1} \quad (9)$$

Here, $\hat{A}$ is the antisymmetrization operator. The continuum wave functions are taken as scattering wave solutions of the ionic potential which are asymptotically matched to hydrogenic Coulomb functions of asymptotic kinetic energy K such that $\Omega = K + I_p$, where I_p is the ionization potential of the corresponding orbital vacancy (5p or 4d). This approach is described in more detail in Wörner et al. ⁷. The single-electron orbital wave functions ϕ_{5p} and ϕ_{4d} are taken from a quantum chemistry calculation using GAMESS ⁸ with small-core relativistic plus polarization correlation consistent triple zeta basis set (cc-pVTZ-PP and Stoll-Dolg effective core potentials) ⁹. The 5p and 4d orbitals are taken in the frozen-core approximation, in which the other orbitals are assumed to be unchanged upon ionization. Such orbitals are very close to the corresponding Dyson orbitals, which include rearrangement of the other electrons. For noble gas atoms, this rearrangement is

very small.

In converting from transition dipole moment in the length form to photoionization cross section, as shown in Fig. 4 in the main paper, we use the following expression³:

$$\frac{d\sigma}{d\Omega_{sr}} = \frac{4\pi^2}{c} \Omega \left(\frac{e^2}{m\hbar^2} \right) \left| (2(\Omega - I_{5p}))^{1/2} \mathbf{D}_{5p} + (2(\Omega - I_{4d}))^{1/2} \mathbf{D}_{4d} \right|^2 \quad (10)$$

where I_{5p} and I_{4d} are the ionization potentials for removal of a 5p and a 4d electron, and Ω is the emitted photon energy.

All of the integrals above are in general complex, meaning that the emission from each channel is not necessarily in phase. Nevertheless, the calculation predicts that the interference between channels is generally constructive, in agreement with the experiment.

In HHG experiments, phase matching is essential to producing coherent collimated xuv emission. This condition is usually met because the attosecond emission is synchronized to the optical cycle. This condition requires that the atom coherently returns to the initial ground state, otherwise the emission is incoherent. It is less obvious that the channel which includes coupling between 5p and 4d electrons is phase-matched, since the electron-electron interaction could potentially randomize the phase. However the analysis above shows that, as long as the total energy of the system (continuum electron plus ion core) is conserved, the emission phase is deterministic. If the laser field accelerates the continuum electron after the electron-electron interaction and before recombination, an additional phase term will appear. Since the interaction occurs very close to the ion core, and just before recombination, we neglect this effect. This additional phase might be an experimental observable that will give access to the time of the electron-electron interaction on an attosecond time scale.

3 Source

The experimental layout comprising the optical setup for IR few-cycle pulse generation and the high-harmonic chamber is depicted in Fig. 3. The source is based on the frequency conversion from Ti:Sa wavelength of 800 nm to 1.8 μm with commercial OPA technology followed by subsequent pulse compression. The starting point is a Ti:Sa amplifier delivering 40 fs pulses with 6 mJ pulse energy at a repetition rate of 100 Hz. Frequency conversion to the IR spectral range is achieved by utilizing the idler out of a fluorescence seeded high energy OPA (HE TOPAS, Light Conversion). It provides 52 fs pulses with pulse energy of 950 μJ which are focused into the hollow-core fiber (HCF) by a $f = 1$ m lens made of CaF_2 . Pulse shortening is realized due to the combined action of nonlinear propagation in the HCF and linear propagation through bulk material in the anomalous dispersion regime¹⁰. Self-phase modulation (SPM) in the 1 m long fiber is the main driving source for spectral broadening of the initial OPA spectrum shown as shaded gray in Fig. 4a. After the fiber, the pulse duration roughly equals the input duration but its spectrum signif-

icantly broadened (black line in Fig. 4a). Collimation of the continuum is carried out with a mirror of 1 m focal length. The broadened power spectrum given by the black line in Fig. 4a is slightly asymmetric towards the blue spectral side as a result of self-steepening¹¹. More important for the subsequent compression, however, is that self-steepening also causes the spectral phase to become asymmetric, distinct from pure SPM which bears both, symmetric spectrum and phase. As expected, this symmetric part of the spectral phase is compensated afterwards by the negative group delay dispersion (GDD) introduced by the glass in the anomalous dispersion regime. The distinctiveness of the glass compression scheme, compared to other approaches^{12–15}, lies in the cancelation of the spectral phase-asymmetry arising from self-steepening during nonlinear propagation. It is balanced by the positive third order dispersion (TOD) of the glass during linear propagation of the expanded, low intensity, beam. Thus, compression is not limited by the TOD of bulk material which is always positive. The flat spectral phase after compression is shown as red line in Fig. 4a with the corresponding temporal profile given in Fig. 4b. A more detailed discussion and full numerical simulations of the mutually compensating propagation effects can be found in Ref.¹⁶.

Several materials contribute to the pulse compression subsequent to the fiber. CaF₂ windows (not shown in Fig. 3) on the HCF (1 mm thickness) and at the entrance of the high-harmonic chamber (3 mm thickness) have a minor effect because of very low dispersion. Compression mainly arises from the transmissive polarizer **P** (nanoparticle polarizer, Thorlabs) and fine tuning is achieved by small tilts of the Fused Silica **FS** plate (2.0 mm thickness). Intensity control is achieved by rotating the half-wave plate $\lambda/2$ in front of the HCF in combination with the polarizer located after the fiber. Pulse characterization is carried out with a home built second harmonic generation - frequency resolved optical gating (SHG-FROG)¹⁷ setup designed for dispersion free few-cycle pulse measurement¹⁰.

In this straight-forward manner 11 fs pulses, corresponding to 1.8 optical cycles at 1.8 μm wavelength (Fig. 4b), with $\sim 300 \mu\text{J}$ pulse energy have been delivered to the target chamber.

4 XUV Spectrometer

A schematic diagram of the HHG spectrometer and source chamber is shown in Fig. 3. The laser enters the source chamber through a 3 mm thick CaF₂ window and is focused below a thin pulsed gas jet by an $f = 25 \text{ cm}$ spherical mirror. The jet has a 500 μm aperture and backing pressure of 2–3 atm. It is mounted on a three-axis manipulator for adjustment relative to the laser focus. We estimate the number density under the jet to be $2 \cdot 10^{17} \text{ cm}^{-3}$. The focusing optics are chosen such that the jet is much thinner than the Rayleigh range. An ion detector consisting of an electrically biased mesh was located directly below the jet. The resulting harmonics pass from the source to the detector chambers through a differential pumping tube. The harmonic beam enters the spectrometer through a $\sim 140 \mu\text{m}$ slit and is dispersed by a 1200 l/mm spherical holographic grating (Shimadzu 30-002). This type of grating has been shown to have significantly lower second order dispersion compared to equivalent ruled gratings^{18,19}. The dispersed spectrum is imaged

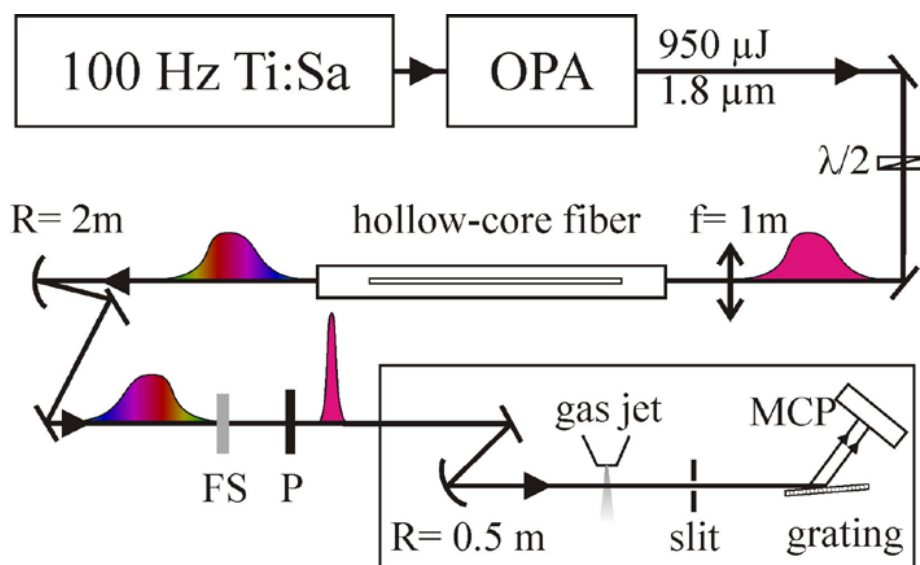


Figure 3: Experimental setup The IR spectral range is accessed by utilizing the idler of a high energy OPA, pumped at 800 nm. Confined propagation in a hollow-core fiber filled with Ar at atmospheric pressure broadens the spectrum which is subsequently compressed by bulk material (CaF_2 vacuum windows, transmissive polarizer (P), 2 mm FS for fine tuning). A half-wave plate ($\lambda/2$) in front of the fiber is used to attenuate the beam in combination with the polarizer. High harmonics are generated by focusing this beam ($f = 25$ cm) into a thin gas jet and their spectral characterization is carried out by imaging the entrance slit onto a multichannel plate (MCP) with a concave grating.

onto a microchannel plate intensifier backed by a phosphor screen (Burle APD 3115 32/25/8 I EDR MgF2 P20). The resulting spectrum was recorded with a high dynamic range digital camera.

Calibration In HHG spectroscopy it is common to calibrate the spectrometer by fitting the positions where harmonics are observed to their expected position based on odd multiples of the laser photon energy and the grating equation. This procedure works well for low harmonic orders but can introduce significant errors when a large number of harmonics are produced. The accuracy of calibrations using the observed HHG spectra is very sensitive to our knowledge of the laser wavelength and can be seriously degraded by the ‘blue shift’²⁰ which causes the spectrum to shift in energy at high harmonic orders. The wavelength scale of the spectrometer was calibrated by recording oxygen recombination lines and comparing their positions to the wavelengths in the NIST database. To do this we put oxygen into the pulsed jet and focus several millijoules of circularly polarized 800 nm radiation to the same position where we normally focus the 1.8 μm laser when producing harmonics. We then observed the spectrum resulting from photo-recombination of free electrons with oxygen ions in the laser produced plasma. This procedure is similar to the one described by Farrell et al.²¹ as well as²². The resulting spectrum is shown in Fig. 5 along with the positions of oxygen lines that we expect based on NIST data.

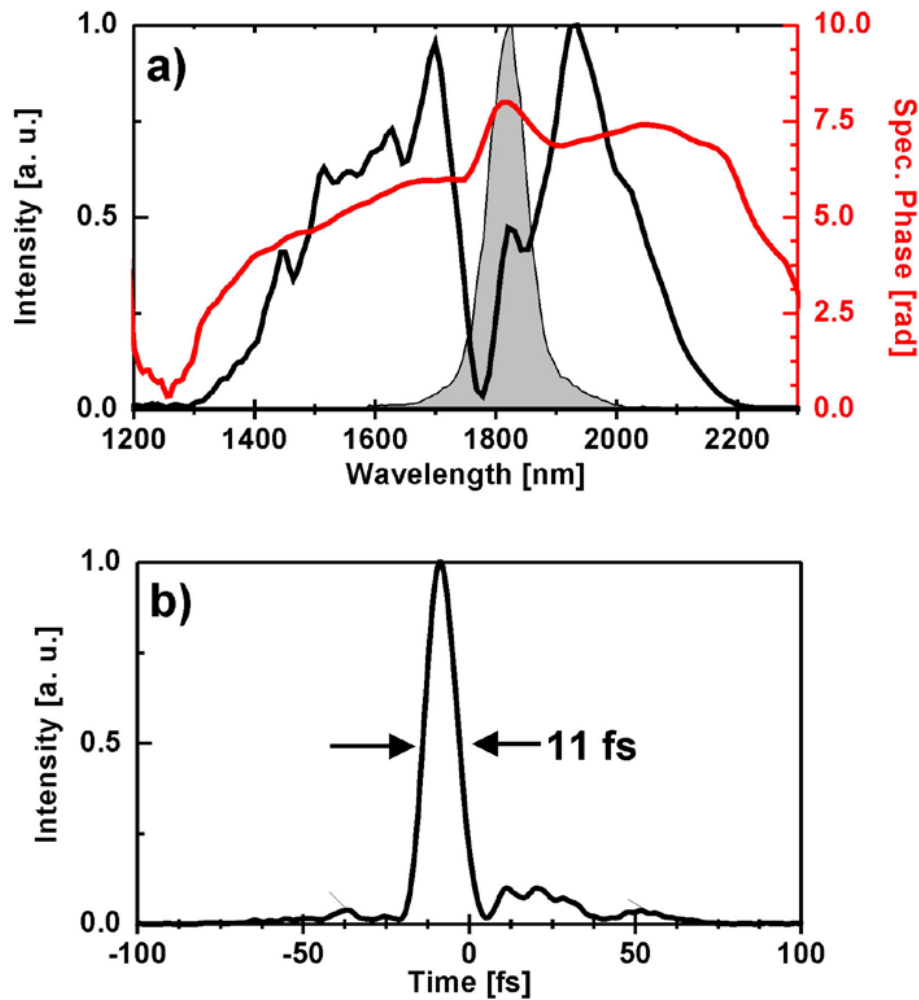


Figure 4: **Pulse characterization** (a) spectral broadening (black) after the hollow-core fiber is compared to OPA input bandwidth (shaded gray) which corresponds to a 52 fs pulse. The retrieved flat spectral phase compressed by the bulk material used in the setup is shown as a red curve. (B) Temporal intensity of the compressed 1.8 cycle pulse.

Note that NIST gives the position and relative intensity for oxygen transitions, many of which are very close together. To generate the spectrum shown we assumed that each transition had a secant squared line shape with an amplitude given by the relative amplitudes from NIST. For a given wavelength we then sum the contribution from each transition. This procedure gives a reasonable approximation for the centre wavelength expected for each line in the observed spectrum.

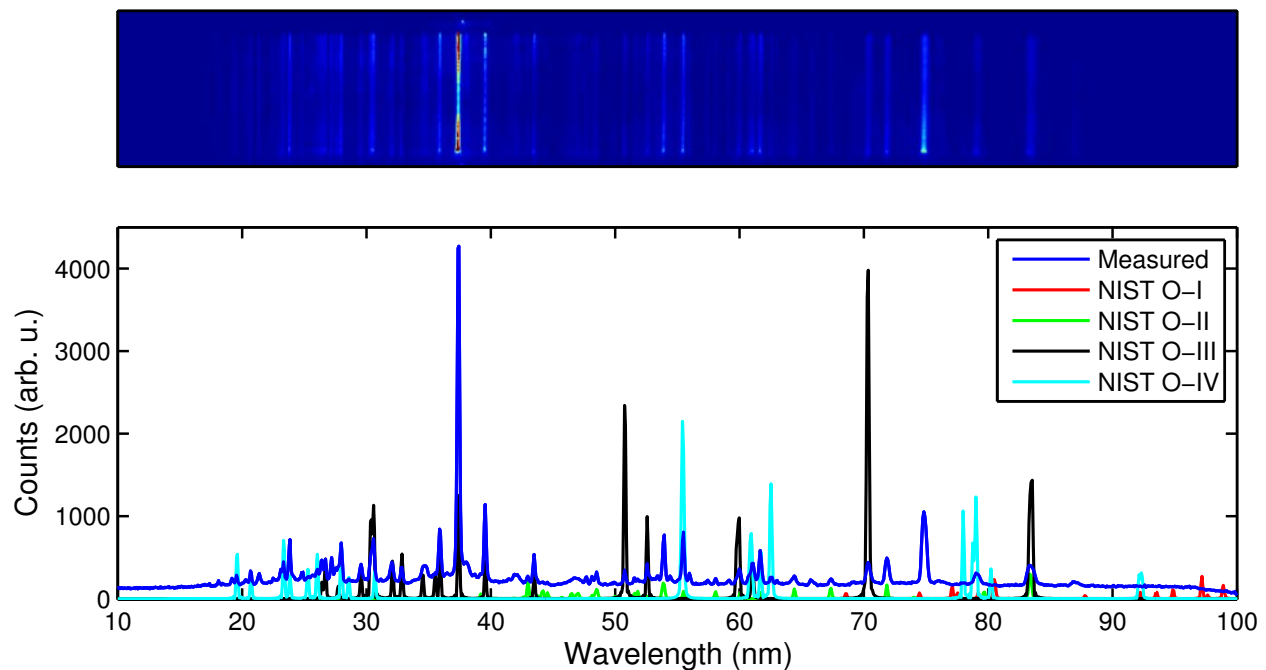


Figure 5: Image of measured spectrum of oxygen recombination lines (top). The lower panel shows the measured spectrum (blue) along with the absorption spectra of Oxygen-I through IV as tabulated by NIST. Note that the NIST data is largely based on photo-absorption measurements. In our experiment, the spectrum is produced by photo-recombination of electrons in the laser produced plasma. While the centre wavelengths for lines corresponding to the two processes will be nearly identical, their relative strengths are expected to differ.

5 Intensity Calibration

We use an ion detector consisting of a wire mesh, biased at -500 V, located below the pulse jet to measure the number of ions produced during harmonic generation. In²³ we showed that the ion signal is proportional to the number of ions produced in the interaction volume. To calibrate the laser intensity we measure the ion yield N_i as a function of pulse energy and fit this data to the intensity dependent ionization rate from theory. The measured ion yield is plotted as symbols in Fig. 6. The solid curves were calculated using the Yudin-Ivanov nonadiabatic ionization model²⁴ integrated over a Gaussian spatial intensity distribution. This model includes both quasi-static tunneling as well as the wavelength-dependent multiphoton contribution to the ionization probability. We minimize the square error between theory and experiment by scaling the horizontal (intensity) axis of the measured data and the vertical (ionization yield) axis of the theoretical curve. This fitting procedure determines the absolute intensity of the laser in the interaction volume for a given pulse energy²⁵.

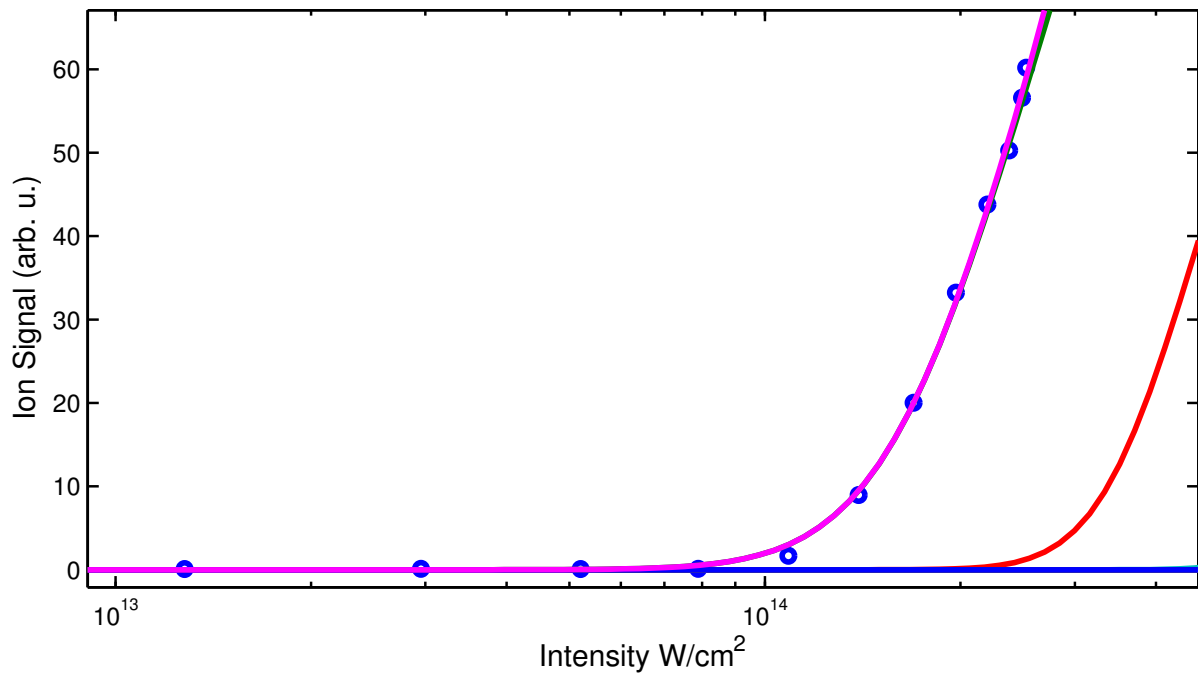


Figure 6: Points are the experimentally measured ion signal from Xenon when ionized by our 11fs $1.8\mu m$ pulse for a range of pulse energies. The solid lines show the Yudin-Ivanov ionization rate integrated over the focal volume.

6 Phase Matching

We have tacitly assumed that the experimentally observed HHG spectrum reflects the single-atom response of xenon. HHG is a collective process in which the emission from each atom is added coherently. Phase mismatch occurs if the driving laser field has a different phase velocity than the xuv emission. If phase mismatch is present, the observed HHG spectrum contains both the single-atom response and a contribution from the collective response. Phase mismatch has been extensively studied²⁶.

Phase mismatch can become more significant with longer-wavelength driving lasers for two reasons. First, the lower frequency laser is more affected by the medium, particularly by the free electrons following ionization of the atoms. Second, the xuv frequencies are much higher, commensurate with the higher cutoff energy, and require less of a temporal shift of the driving field in order to become out of phase. Nevertheless, it has been shown that xuv emission from capillary gas targets can extend beyond 200 eV in a phase-matched process²⁶. In the present experiment, we use an extremely thin gas jet target to minimize the possibility of phase mismatch. Additionally, the very short laser pulse reduces the effect of phase mismatch due to plasma formation.

To test the supposition that the HHG spectrum corresponds to the single atom response, we performed several tests. We note that the HHG spectra of Kr and Xe are consistent with the PICS of the single atoms; Xe would be expected to be more sensitive to plasma since its ionization potential is lower than that of Kr, yet both spectra match the PICS.

In Fig. 7 we show how the spectrum of xenon changes as the laser intensity is changed. As the intensity increases, the cutoff frequency also increases and goes through the giant resonance at 100 eV. Beyond this point, the spectrum only increases in total power, but does not change shape. If phase mismatch from plasma dispersion were present, we would expect that the higher intensity spectrum would change due to the presence of more plasma.

We also tested the scaling of the HHG emission as a function of the gas pressure in the jet. If the process is perfectly phase matched, the HHG emission should scale quadratically with gas pressure. We previously demonstrated this behavior in the same gas jet²³. We show the scaling of the HHG emission in the present experiment in Fig. 8. The scaling power is seen to be somewhat greater than quadratic; if phase mismatch were an issue, the scaling power would be less than 2. There was some indication that, at the lowest gas pressure, the giant resonance peak in xenon was at 120 eV rather than 100 eV; however the signal was barely above the noise level at this pressure. As the pressure was increased, the peak was clearly centered at 100 eV.

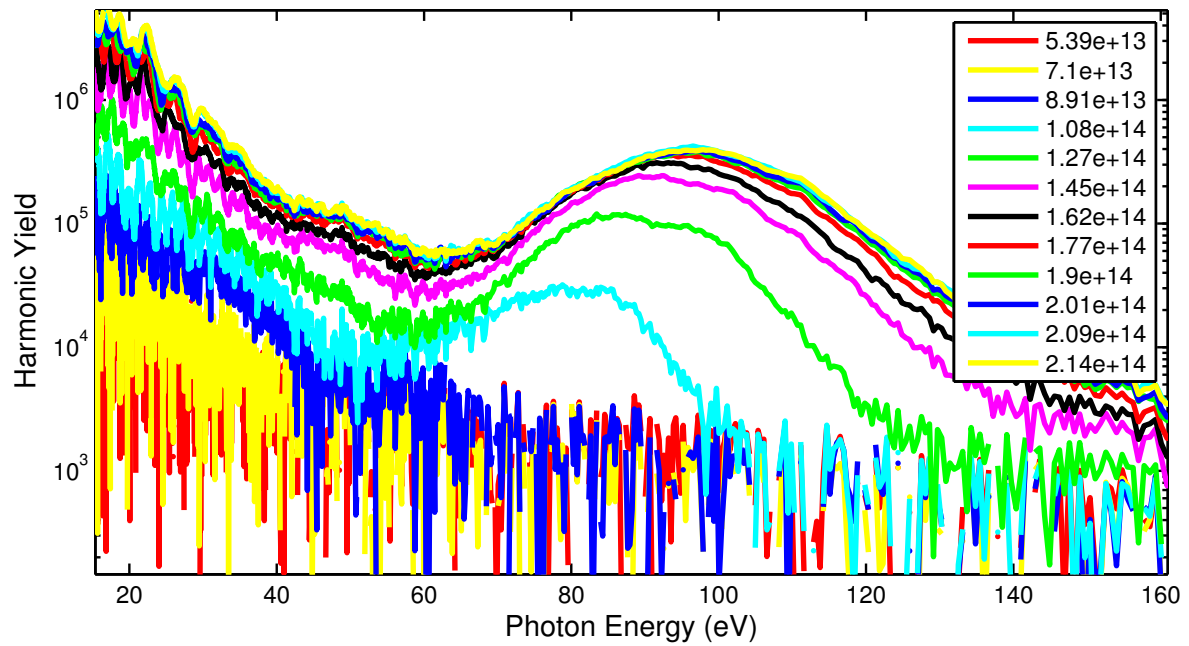


Figure 7: HHG yield in xenon for a range of laser intensities (see legend in W/cm^2). The pulsed jet backing pressure was ~ 3 bar.

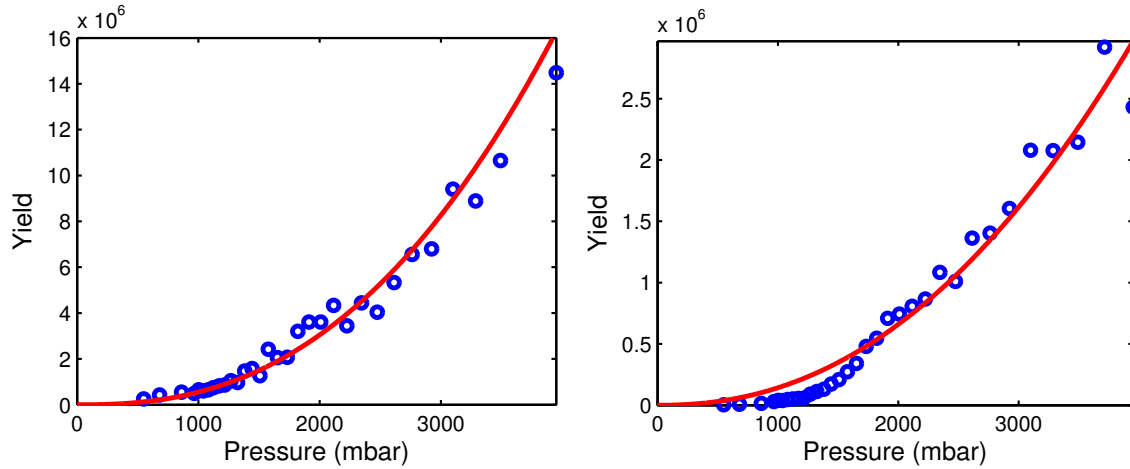


Figure 8: Integrated harmonic yield with respect to pressure. For each pressure the harmonics were integrated over a specified range of photon energies and the result was fit to the function $a_0 P^b$ where P is pressure. The results are (left) 20eV-100eV, $b = 2.46$, (right) 90eV-110eV, $b = 2.21$.

7 Argon PICS

As described in the main text of this paper we analyze the xenon HHG spectra by normalizing it to the returning electron wavepacket $\mathbb{W}(E)$ measured for a krypton reference atom. The xenon PICS is then determined by dividing its measured HHG spectrum by the $\mathbb{W}(E)$ from krypton. We used a similar procedure in an effort to extract the argon PICS from HHG measurements.

Comparing our results, Fig. 9, to the published cross section, we find that the expected deep Cooper minimum in argon is barely present in our unfiltered measurements. The reason for this is that a fraction of the harmonics from the high energy side of the spectrum, at around 100 eV, are diffracted in to the second order by the spectrometer grating and overlap with the 50 eV harmonics which are observed in first order. As described in Sec. 4, the Shimadzu holographic grating was chosen for these experiments because it has significantly lower second order dispersion compared to similar ruled gratings, but interference from second order diffraction is still present in our measured spectrum.

To show that this is the case, we placed a 200 nm thick aluminum filter in the xuv beam before the grating and recorded HHG spectra in both argon and krypton. Aluminum transmits xuv photons in the range of 17 eV to 73 eV and strongly attenuates higher energy harmonics.

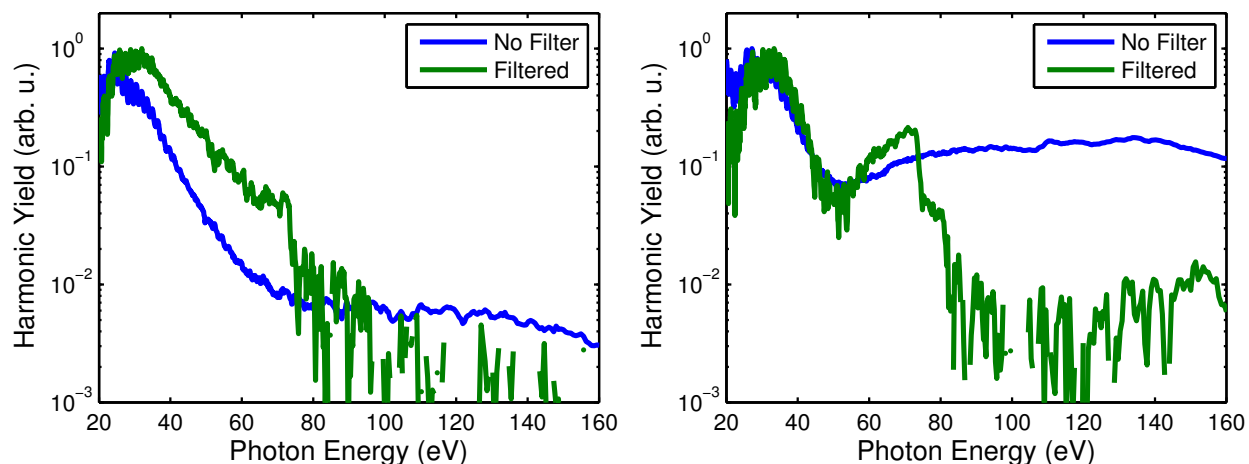


Figure 9: HHG spectra for krypton (left) and argon (right) measured with and without an aluminum filter, green and blue curves respectively. The filtered and unfiltered spectra were scaled vertically in these plots.

As expected, the filter blocks harmonics with energies $> 74\text{ eV}$. The shape of the krypton spectrum in the transmission band of the filter is largely unaffected by the filter because the real krypton spectrum is much brighter at 50 eV compared to 100 eV, so any second order contribution to the measured spectrum is negligible. The situation is different for argon, where bright $\sim 100\text{ eV}$

harmonics fill in the Cooper minimum at 54 eV. Placing the filter in the beam blocks the ~ 100 eV harmonics and a much deeper Cooper minimum is observed as shown in Fig. 9 (green).

1. Landau, L. D. & Lifshitz, E. M. *Quantum Mechanics Non-Relativistic Theory*, vol. 3 of *Course of Theoretical Physics* (Pergamon Press Ltd., New York, 1977), 3 edn.
2. Le, A.-T., Lucchese, R. R., Tonzani, S., Morishita, T. & Lin, C. D. Quantitative rescattering theory for high-order harmonic generation from molecules. *Phys. Rev. A* **80**, 013401 (2009).
3. Starace, A. F. Theory of atomic photoionization. In Mehlhorn, W. (ed.) *Handbuch Der Physik Vol. XXXI*, 1–121 (Springer, Berlin, 1982).
4. Huang, K. N., Johnson, W. R. & Cheng, K. T. Theoretical photoionization parameters for the noble gases argon, krypton, and xenon. *Atomic and Nuclear Data Tables* **26**, 33 – 45 (1981).
5. Lewenstein, M., Balcou, P., Ivanov, M. Y., L’huillier, A. & Corkum, P. B. Theory of high-harmonic generation by low-frequency laser fields. *Phys. Rev. A* **49**, 2117–2132 (1994).
6. Patchkovskii, S., Zhao, Z., Brabec, T. & Villeneuve, D. M. High harmonic generation and molecular orbital tomography in multielectron systems. *J. Chem. Phys.* **126**, 114306 (2007).
7. Wörner, H. J., Niihara, H., Bertrand, J. B., Corkum, P. B. & Villeneuve, D. M. Observation of electronic structure minima in high-harmonic generation. *Physical Review Letters* **102**, 103901 (2009).
8. Schmidt, M. W. *et al.* General atomic and molecular electronic structure system. *J. Comput. Chem.* **14**, 1347 (1993).
9. Peterson, K., Figgen, D., Goll, E., Stoll, H. & Dolg, M. Systematically convergent basis sets with relativistic pseudopotentials. ii. small-core pseudopotentials and correlation consistent basis sets for the post-d group 16 - 18 elements. *J. Chem. Phys.* **119**, 11113 (2003).
10. Schmidt, B. E. *et al.* Compression of 1.8 μm laser pulses to sub two optical cycles with bulk material. *Appl. Phys. Lett.* **96**, 121109 (2010).
11. Yang, G. & Shen, Y. R. Spectral broadening of ultrashort pulses in a nonlinear medium. *Opt. Lett.* **9**, 510–512 (1984).
12. Hauri, C. P. *et al.* Intense self-compressed, self-phase-stabilized few-cycle pulses at 2 μm from an optical filament. *Opt. Lett.* **32**, 868–870 (2007).
13. Vozzi, C. *et al.* Millijoule-level phase-stabilized few-optical-cycle infrared parametric source. *Opt. Lett.* **32**, 2957–2959 (2007).
14. Gu, X. *et al.* Generation of carrier-envelope-phase-stable 2-cycle 740- μj pulses at 2.1- μm carrier wavelength. *Opt. Express* **17**, 62–69 (2009).

15. Mücke, O. D. *et al.* Self-compression of millijoule 1.5 μm pulses. *Opt. Lett.* **34**, 2498–2500 (2009).
16. Béjot, P., Schmidt, B. E., Kasparian, J., Wolf, J.-P. & Legaré, F. Mechanism of hollow-core-fiber infrared-supercontinuum compression with bulk material. *Phys. Rev. A* **81**, 063828 (2010).
17. Trebino, R. *et al.* Measuring ultrashort laser pulses in the time-frequency domain using frequency-resolved optical gating. *Review of Scientific Instruments* **68**, 3277–3295 (1997).
18. Chowdhuri, M. B. *et al.* Spectroscopic comparison between 1200 grooves/mm ruled and holographic gratings of a flat-field spectrometer and its absolute sensitivity calibration using bremsstrahlung continuum. *Review of Scientific Instruments* **78**, 023501 (2007).
19. Yamazaki, T. *et al.* Comparison of mechanically ruled versus holographically varied line-spacing gratings for a soft-x-ray flat-field spectrograph. *Appl. Opt.* **38**, 4001–4003 (1999).
20. Wahlström, C.-G. *et al.* High-order harmonic generation in rare gases with an intense short-pulse laser. *Phys. Rev. A* **48**, 4709–4720 (1993).
21. Farrell, J. P., McFarland, B. K., Bucksbaum, P. H. & Gühr, M. Calibration of a high harmonics spectrometer by laser induced plasma emission. *Opt. Express* **17**, 15134–15144 (2009).
22. Fill, E. *et al.* xuv spectra of optical-field-ionized plasmas. *Phys. Rev. E* **51**, 6016–6027 (1995).
23. Shiner, A. D. *et al.* Wavelength scaling of high harmonic generation efficiency. *Phys. Rev. Lett.* **103**, 073902 (2009).
24. Yudin, G. L. & Ivanov, M. Y. Nonadiabatic tunnel ionization: Looking inside a laser cycle. *Phys. Rev. A* **64**, 013409 (2001).
25. Hankin, S. M., Villeneuve, D. M., Corkum, P. B. & Rayner, D. M. Intense-field laser ionization rates in atoms and molecules. *Phys. Rev. A* **64**, 013405 (2001).
26. Popmintchev, T. *et al.* Phase matching of high harmonic generation in the soft and hard x-ray regions of the spectrum. *Proc. Nat. Acad. Sci. U. S. A.* **106**, 10516 (2009).

Chapter 11

Conclusions and Outlook

The first half of this thesis focused on developing techniques to extend the HHG cutoff by generating harmonics with long-wavelength OPA based laser sources. The $I\lambda^2$ scaling of the harmonic cutoff with laser wavelength suggested that there should be a significant benefit to going to longer wavelengths. Unfortunately, while the cutoff does increase with the square of the laser wavelength, this is accompanied by a corresponding decrease in harmonic yield. The first major campaign of this thesis was to experimentally measure the scaling of harmonic yield with wavelength. A systematic study was conducted where, in the words of one referee: “almost all of the possible experimental errors have been avoided or corrected for.” The conclusion of this study is that the harmonic yield scales as $\lambda^{-6.3\pm1.1}$ in Xe and $\lambda^{-6.5\pm1.1}$ in Kr at constant intensity [9]. These results suggest that it will be difficult to produce high harmonics in low density gas jets with long wavelength sources. One solution to this has been the development of new gas sources based on capillary waveguide, where pressures of several atmospheres can be employed to compensate for the decrease in yield from the individual emitters [115].

The wavelength scaling experiments required careful control over the harmonic generation process, which have lead to a number of improvements in the HHG experiment. First, we developed an ion detector (Ch. 4), for monitoring ionization during HHG. This detector was originally used as a diagnostic when aligning the timing of the pulsed jet to the laser pulse. The realization that the ion signal was proportional to the number of ions produced in the focus made the detector a much more useful tool, which is now routinely used for intensity calibration of the laser pulse, measurement of the temporal and spatial profile of the gas pulse, and for measuring laser

beam parameters. In addition to the HHG chamber at ALLS, ion detectors have now been installed in all HHG target chambers at the NRC as well as in the chambers of a number of our collaborators.

The other major improvement from the wavelength scaling experiments came from spatially filtering the OPA beam and the renewed appreciation for the role that mode quality plays during HHG. We were initially very reluctant to install a spatial filter, as we were concerned that there would not be sufficient laser intensity for HHG after losing energy in the filter. It turned out that harmonic yield significantly improved. The introduction to Chapter 5 shows that the strong coupling between laser intensity at the focus and the phase of the XUV wavefront implies that maintaining excellent mode quality is essential to HHG experiments, particularly in the IR.

The result of the wavelength scaling study suggests that it would be difficult to produce very high energy $\sim$ keV high harmonics in a gas jet simply by going to longer and longer laser wavelengths. The other approach for increasing the HHG cutoff is to reduce the number of optical cycles in the laser pulse, enabling the use of higher laser intensity. We developed pulse compression at $1.4\text{ }\mu\text{m}$ by broadening the OPA spectrum and then compressing the pulse with multiple bounces off of specially-designed chirped mirrors. This resulted in a 13.1 fs 400 μJ pulse (Ch. 6). We then compressed the OPA idler beam at $1.8\text{ }\mu\text{m}$ using anomalous dispersion in fused silica glass instead of chirped mirrors. This resulted in a sub-two-cycle pulse, which has now been amplified to $>1\text{ mJ}$ and used to produce harmonics in neon that extended out past the carbon K edge to $>400\text{ eV}$.

In addition to developing the OPA as a short pulse, long wavelength, high power source, we have also taken advantage of the intrinsic carrier envelope phase stability that is possible with white light seeded OPAs (Ch. 7). It has been known for some time that OPAs can produce CEP stable pulses [101], but taking advantage of CEP control to drive physical processes such as ionization [116, 57], dissociation [98], and attosecond pulse generation [95, 96, 97] requires a pulse that is at most a few optical cycles long. By combining pulse compression with CEP control at $1.8\text{ }\mu\text{m}$, we have developed a <2 cycle pulse which is ideally suited to studying these processes.

The development of pulse compression technology at $1.8\text{ }\mu\text{m}$ enabled us to produce large HHG spectra in low I_p systems. The second half of this thesis focused on analyzing HHG spectra in an effort to learn about the electronic structure of the generating medium. The first step was to compare harmonics produced in C_2H_4 using

both 800 nm and un-compressed 1.8 μm laser pulses. With the long wavelength source the HHG cutoff increased from 40 eV to > 70 eV with the XUV spectrum covering most of the region where the C_2H_4 photoionization cross section is non-zero. We then compressed the 1.8 μm source and used it to produce the largest HHG spectra ever reported in krypton and xenon. The electronic structure of these atoms was clearly apparent in their HHG spectra. The krypton spectrum (Ch. 8), showed a clear Cooper minimum at 85 eV, which results from the $l=2$ radial dipole matrix element passing through zero.

In xenon we produced harmonics with photon energies in excess of 140 eV (Ch. 10). This spectrum shows clear evidence of core excitation during the recombination step of HHG through electron-electron correlations, which manifest as a broad enhancement in harmonic yield near 100 eV [5].

Moving forward there are several areas where new experiments can be developed that build on the results of this thesis:

First: The short-pulse long-wavelength source allowed us to increase the HHG cutoff in xenon by a factor of 5 compared to results with an 800 nm source. The > 140 eV spectrum enabled us to make the first observation of electron correlation and exchange effects in xenon during HHG. The electron correlation and exchange step takes place when the continuum electron re-encounters the parent ion. So far we have assumed that this interaction is instantaneous, and that the laser field does not vary during the interaction. Because the electron correlations are moderated by the Coulomb potential, the interaction starts to take place when the continuum electron is relatively far from the parent ion. It may be possible to influence this process by varying the laser wavelength or intensity, which would open a path to control over electron correlation effects.

There has been a lot of research where HHG was used to study molecular targets with the goal of observing dynamics. In most cases molecules have low ionizing potentials so the HHG spectrum generation with 800 nm sources is limited to a few tens of electron volts. The 1.8 μm few-cycle pulse will greatly increase the cutoff for harmonics produced in molecules, enabling better HHG spectroscopy and higher resolution orbital tomography [67]. This source will also allow broad HHG spectra to be produced with lower laser intensities, which will be helpful when attempting to

interpret HHG spectra from molecules with closely spaced energy levels [117].

Second: We have demonstrated phase-matched high harmonics in neon that extended past 400 eV. This radiation can be focused into a secondary target chamber, where it can be used for microscopy as well as spectroscopy in the water window. If fully compressed, this bandwidth would support pulse durations of < 10 attoseconds.

Third: One of the most exciting directions in the near term will be the use of weak perturbation beams, at the same wavelength as the fundamental, to control the spatial and temporal properties of HHG. Chapter 5 showed that 0.25% intensity noise across the laser focal spot was sufficient to completely destroy phase matching on the high energy side of the spectrum. This resulted from the strong coupling between the laser intensity and the phase of the induced dipole. The implication of this is that it is essential to very carefully control mode quality when producing high energy harmonics. But the news is not all bad; this result also suggests that a weak perturbing field can be used to shape the phase front of the XUV beam in a controllable way. During the last experiment campaign of this PhD, we tried an experiment that was lead by Kyung Taec Kim where a weak 0.1% perturbation beam was made to cross the laser focus at a small angle, as shown in Fig. 11.1. This perturbation caused a periodic phase shift and intensity modulation across the laser focus which distorted the XUV wavefront, causing the beam in the far field to deflect vertically. By varying the time delay between the pump and perturbation fields, the fringes in the focus shift and the XUV beam in the far field oscillates up and down. For a given delay, the position of the HHG spectrum was integrated along the energy axis in the neighbourhood of 70 eV in order to determine the vertical profile of the beam. This profile is plotted as a function of the time delay between the pump and perturbation beams in Fig. 11.2. At large delays the perturbation pulse does not affect HHG, as it does not overlap with the pump pulse in time. In the absence of a perturbing field, HHG produces a well collimated XUV beam that is observed as a small spot on-axis at the detector. As the perturbation pulse advances in time, and starts to overlap with the pump pulse, it cause the XUV beam to deflect vertically. Several oscillations of the XUV beam are observed corresponding to the cross correlation between the pump and perturbation pulses which where each ~ 3 cycles long. This result shows that the weak perturbation field can deflect the XUV

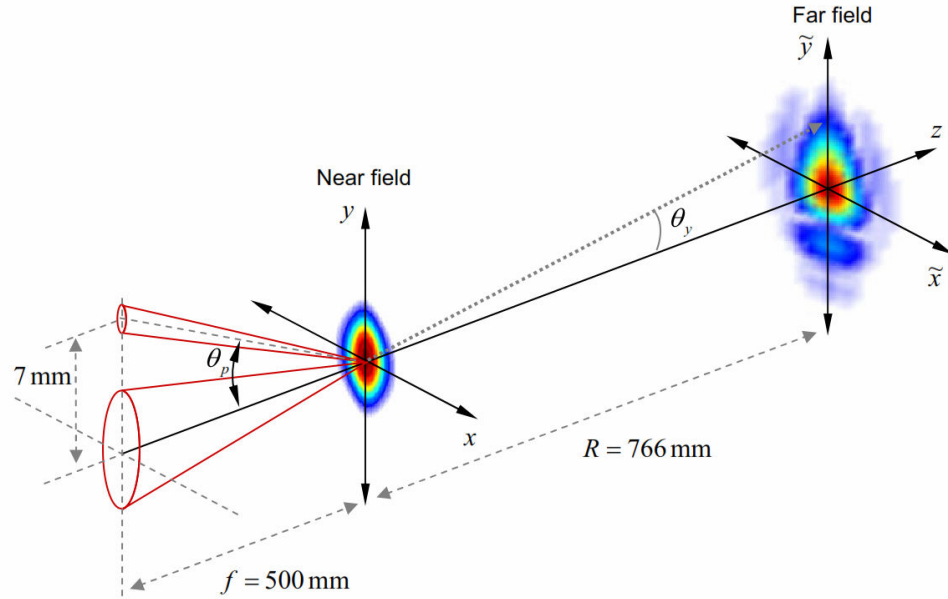


Figure 11.1: Geometry for perturbing the HHG wavefront. A weak perturbation field crosses the laser focus at a small angle. Interference between the pump and perturbation beams causes an intensity modulation across the laser focus. This modulation perturbs the HHG process, causing the XUV beam to deflect vertically in the far-field. (Figure from: [1])

beam by nearly one beam diameter, and that this can be done in a controllable way. It is important to bear in mind that while harmonics are being produced with the several-hundred-microjoule $1.8\,\mu\text{m}$ pump beam, the control field is only 0.1% of this. While an absorptive filter placed in the pump beam might have a very limited life expectancy, it would be straightforward to put complex masks into the probe beam. Figure 11.3 shows a proposed experimental setup for generating an XUV beam with a programmable wavefront. A beam splitter separates a small fraction of the pump laser beam which is directed onto a micro-mirror array. Micro-mirror arrays are used in most video projectors and consist of around 1 million individually programmable mirrors which in a projector either deflect the beam towards the screen or toward a beam dump. Development kits such as the ‘DLP LightCommander’ from Texas Instruments are available, which provide a (1024 x 768) mirror array that can be controlled with a computer and updated at 500 Hz. For the setup in Fig. 11.3, the array would be set to imprint a programmable intensity distribution onto the

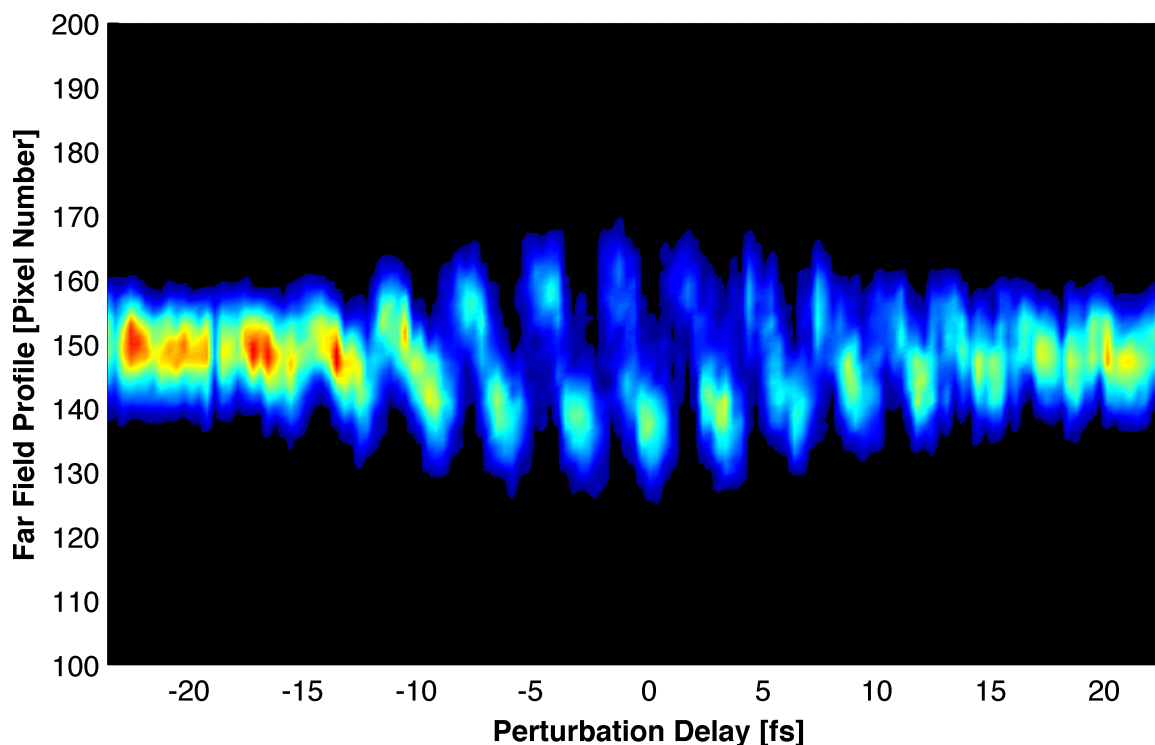


Figure 11.2: The far-field HHG intensity distribution along the vertical direction is plotted in false colors for argon high harmonics at ~ 70 eV (vertical axis). The relative time delay between the pump and perturbation pulses is plotted on the horizontal axis. At large delays the perturbation field does not overlap the pump pulse in time, and a collimated beam is observed on-axis. When the pulses do overlap (~ 0 fs delay), the intensity distribution at the laser focus causes the XUV beam to deflect vertically as seen in the far field profile.

probe beam before it recombines with the pump and is focused into the HHG target chamber. This arrangement could be programmed to produce the same intensity distribution that was used to deflect the XUV beam in Figure 11.2. It could be used to deflect the XUV beam out of the path of the fundamental, or it could be used to imprint a quadratic phase front into the XUV beam, causing the beam to focus. It may also be possible to produce a more complex wavefront that results in two XUV foci that intersect at one point in space, and in time, opening the door to all-optical XUV-pump/XUV-probe experiments. It is enough to make a student wonder if he really wants to graduate!

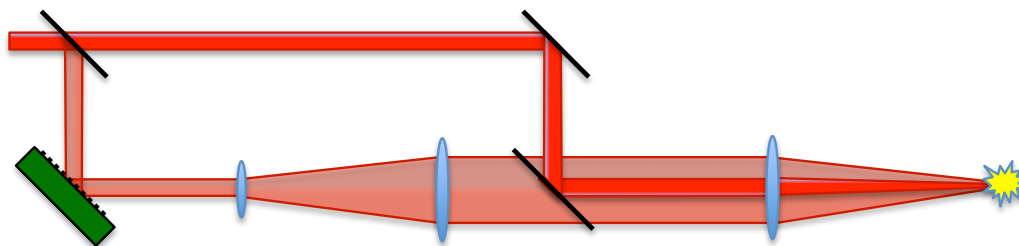


Figure 11.3: Proposed setup for HHG wavefront control. A spatial intensity modulation is imprinted onto a weak perturbation beam by a micro-mirror array (shown in green). A telescope increases the size of the perturbation beam which recombines with the strong pump and the two beams focus together into the gas target in the HHG target chamber. The spatial intensity distribution on the probe beam modifies the phase of the XUV wavefront.



Figure 11.4: Ditch beside the parking lot at ALLS.

Appendix A

List of Variables

Symbol	Description
α	Ion detector sensitivity
$a_{\text{ion}}(t)$	Probability amplitude for ionization
$a_{\text{pr}}(t)$	Probability amplitude for propagation
$a_{\text{rec}}(t)$	Probability amplitude for recombination
$\mathcal{A}$	Vector potential
A	Laser field envelope
χ	Electric susceptibility
c (Ch. 4)	Neutral atom concentration
c	Speed of light
$\mathbf{d}(\omega)$	Dipole transition matrix element
$\mathbf{D}(\omega)$	Induced transition matrix element
$\mathbb{D}$	Dispersion operator
e	Electron charge
$e(t)$ (Ch. 7)	Error signal
F	Electric field strength
E (Ch. 5,6)	Electric field strength
E (Ch. 2)	Energy
$\mathcal{E}$	Peak electric field strength
E_c	Cutoff energy
$\mathcal{F}_{x,y}$	Two dimensional spatial Fourier transform operator
ϕ	Carrier envelope phase

Symbol	Description
Γ	Instantaneous ionization rate
γ	Keldysh parameter
$\hbar$	Planck constant divided by 2π
$\hat{H}$	Hamiltonian
$\mathcal{H}_e(k_x, k_y; z, \omega)$	Free space propagator
$I(t)$	Intensity
I_p	Ionization potential
I_r (Ch. 4)	Radial dependence of intensity $I(r)$
I_{sat}	Saturation intensity for ionization
k (Ch. 5, 6)	Wavenumber
k (Ch. 2)	Canonical momentum
K	Proportional gain
l (Ch. 4)	Thickness of gas sample
l	Orbital angular momentum
λ	Laser wavelength
m_e	Electron mass (a.u.)
$\Delta\nu$	Frequency 1/e bandwidth
$\mathbf{n}$	Refractive index
$n(t)$	Ground state population (Eq 2.4.17)
$N(t)$	Ground state population (Eq 2.2.1)
$\mathbb{N}$	Nonlinear operator
Ω	Solid angle
$P(\omega)$	HHG power spectrum
$\mathbb{P}$	Polarization
$P(r)_{\text{ion}}$ (Ch. 4)	Probability for ionization at position r by end of pulse
$P_{\epsilon,l}(r)$ (Ch. 8)	Radial wavefunction
$\Phi(z, r)$ (Ch. 5)	Intensity-dependent dipole phase
$\phi(\omega)$	Spectral phase
ψ	Wavefunction
q	Order of nonlinear process / high harmonic
S	Classical action
σ_r	Photo-recombination cross-section
σ_I	Photoionization cross-section

Symbol	Description
t	Time
T_i	Integration Time
$\Delta\tau$	Pulse duration (1/e)
τ	Tunneling time
t_b	Birth time
t_e	Emission time
$\theta(t)$	Instantaneous electric field phase
$\mathbf{v}$	Velocity
V	Potential energy
w	Laser 1/e field radius
w_0	Laser 1/e field radius at the focus
ω_L	Laser optical frequency
x	Electron Position

Bibliography

- [1] K. T. Kim, C. Zhang, A. D. Shiner, S. E. Kirkwood, E. Frumker, G. Gariepy, A. Naumov, D. M. Villeneuve, and P. B. Corkum, “Manipulating quantum paths for space-time characterization of attosecond pulses,” *Nature*, 2012. (Manuscript ID: 2012-08-09966, Submitted August, 2012).
- [2] A. D. Shiner, C. Trallero-Herrero, N. Kajumba, B. E. Schmidt, Kyung Taec Kim, H.-C. Bandulet, D. Comtois, F. Légaré, J.-C. Kieffer, D. M. Rayner, P. B. Corkum, and D. M. Villeneuve, “Characterization of ionization and intensity during high harmonic generation,” 2012. (In Preparation).
- [3] A. D. Shiner, B. E. Schmidt, C. Trallero-Herrero, P. B. Corkum, J.-C. Kieffer, F. Légaré, and D. M. Villeneuve, “Observation of cooper minimum in krypton using high harmonic spectroscopy,” *Journal of Physics B: Atomic, Molecular and Optical Physics*, vol. 45, no. 7, p. 074010, 2012.
- [4] C. Trallero-Herrero, C. Jin, B. E. Schmidt, A. D. Shiner, J.-C. Kieffer, P. B. Corkum, D. M. Villeneuve, C. D. Lin, F. Légaré, and A. T. Le, “Generation of broad xuv continuous high harmonic spectra and isolated attosecond pulses with intense mid-infrared lasers,” *Journal of Physics B: Atomic, Molecular and Optical Physics*, vol. 45, no. 1, p. 011001, 2012.
- [5] A. D. Shiner, B. E. Schmidt, C. Trallero-Herrero, H. J. Wörner, S. Patchkovskii, P. B. Corkum, J.-C. Kieffer, F. Légaré, and D. M. Villeneuve, “Probing collective multi-electron dynamics in xenon with high-harmonic spectroscopy,” *Nature Physics*, vol. 7, p. 464467, Mar. 2011.
- [6] B. E. Schmidt, A. D. Shiner, P. Lassonde, J.-C. Kieffer, P. B. Corkum, D. M. Villeneuve, and F. Légaré, “CEP stable 1.6 cycle laser pulses at 1.8 μm ,” *Optics Express*, vol. 19, no. 7, pp. 6858–6864, 2011.
- [7] B. E. Schmidt, P. Béjot, M. Giguère, A. D. Shiner., C. Trallero-Herrero, É. Bissou, J. Kasparian, J. Wolf, D. M. Villeneuve, J. Kieffer, P. B. Corkum, and F. Légaré, “Compression of 1.8 μm laser pulses to sub two optical cycles with bulk material,” *Applied Physics Letters*, vol. 96, p. 121109, Mar. 2010.

- [8] C. Trallero-Herrero, B. Schmidt, A. D. Shiner, P. Lassonde, É. Bisson, J.-C. Kieffer, P. Corkum, D. Villeneuve, and F. Légaré, “High harmonic generation in ethylene with infrared pulses,” *Chemical Physics*, vol. 366, no. 1-3, pp. 33 – 36, 2009.
- [9] A. D. Shiner, C. Trallero-Herrero, N. Kajumba, H.-C. Bandulet, D. Comtois, F. Légaré, M. Giguère, J.-C. Kieffer, P. B. Corkum, and D. M. Villeneuve, “Wavelength scaling of high harmonic generation efficiency,” *Physical Review Letters*, vol. 103, no. 7, p. 073902, 2009.
- [10] M. Giguère, B. E. Schmidt, A. D. Shiner, M.-A. Houle, H. C. Bandulet, G. Tempea, D. M. Villeneuve, J.-C. Kieffer, and F. Légaré, “Pulse compression of sub-millijoule few-optical-cycle infrared laser pulses using chirped mirrors,” *Opt. Lett.*, vol. 34, no. 12, pp. 1894–1896, 2009.
- [11] H.-C. Bandulet, D. Comtois, A. D. Shiner, C. Trallero-Herrero, N. Kajumba, T. Ozaki, P. B. Corkum, D. M. Villeneuve, J.-C. Kieffer, and F. Légaré, “High harmonic generation with a spatially filtered optical parametric amplifier,” *Journal of Physics B: Atomic, Molecular and Optical Physics*, vol. 41, no. 24, p. 245602 (7pp), 2008.
- [12] B. E. Schmidt, A. D. Shiner, M. Giguère, P. Lassonde, C. Trallero-Herrero, J.-C. Kieffer, P. B. Corkum, D. M. Villeneuve, and F. Légaré, “High harmonic generation with long wavelength few-cycle laser pulses,” *Journal of Physics B: Atomic, Molecular and Optical Physics*, vol. 45, no. 7, p. 074008, 2012.
- [13] H. Akagi, T. Otobe, A. Staudte, A. D. Shiner, F. Turner, R. Dörner, D. M. Villeneuve, and P. B. Corkum, “Laser Tunnel Ionization from Multiple Orbitals in HCl,” *Science*, vol. 325, no. 5946, pp. 1364–1367, 2009.
- [14] P. Dubé, A. A. Madej, J. E. Bernard, L. Marmet, and A. D. Shiner, “A narrow linewidth and frequency-stable probe laser source for the $^{88}\text{Sr}^+$ single ion optical frequency standard,” *Applied Physics B: Lasers and Optics*, pp. 40–+, Feb. 2009.
- [15] A. D. Shiner, A. A. Madej, P. Dubé, and J. E. Bernard, “Absolute optical frequency measurement of saturated absorption lines in Rb near 422 nm,” *Applied Physics B: Lasers and Optics*, vol. 89, pp. 595–601, Dec. 2007.
- [16] A. D. Shiner, B. E. Schmidt, C. Trallero-Herrero, H. J. Wörner, S. Patchkovskii, J.-C. Kieffer, P. B. Corkum, F. Légaré, and D. M. Villeneuve, “Probing collective multi-electron dynamics with high harmonic spectroscopy - the giant resonance in xenon,” in *The 3rd International Conference on Attosecond Physics*, (Hokkaido University, Sapporo, Japan), 2011. (Oral Presentation).

- [17] A. D. Shiner, B. E. Schmidt, C. Trallero-Herrero, H. J. Wörner, S. Patchkovskii, P. B. Corkum, J.-C. Kieffer, F. Légaré, and D. M. Villeneuve, "Probing collective multi-electron dynamics with high harmonic spectroscopy - the giant resonance in xenon," in *Quantum Electronics and Laser Science Conference*, (Baltimore, Maryland, USA), p. QMC1, Optical Society of America, 2011. (Oral Presentation).
- [18] F. Legare, B. E. Schmidt, P. Béejot, M. Giguère, A. D. Shiner, C. Trallero-Herrero, É. Bisson, J. Kasparian, J.-P. Wolf, D. M. Villeneuve, J.-C. Kieffer, and P. B. Corkum, "Intense sub-2 optical cycle laser pulses at 1.8 micron for high harmonic generation," *APS Meeting Abstracts*, p. L6007, Mar. 2010.
- [19] C. Trallero-Herrero, B. Schmidt, A. D. Shiner, J.-C. Kieffer, P. Corkum, F. Legare, and D. Villeneuve, "Time Reversing High Harmonic Generation: Towards table top XUV spectroscopy," *APS Meeting Abstracts*, p. 3006, Mar. 2010.
- [20] A. D. Shiner, C. Trallero-Herrero, N. Kajumba, H.-C. Bandulet, D. Comtois, F. Légaré, M. Giguère, J.-C. Kieffer, P. B. Corkum, and D. M. Villeneuve, "Scaling of high harmonic generation efficiency with wavelength," in *Attosecond Physics 2009*, (Manhattan Kansas), 2009. (Poster Presentation).
- [21] F. Légaré, B. E. Schmidt, A. D. Shiner, C. Trallero-Herrero, H. J. Wörner, M. Giguère, P. B. Corkum, J.-C. Kieffer, and D. M. Villeneuve, "High harmonic generation using 1425 nm sub-3 cycles laser pulses," in *Attosecond Physics 2009*, (Manhattan Kansas), 2009. (Oral Presentation).
- [22] A. D. Shiner, C. Trallero-Herrero, N. Kajumba, H.-C. Bandulet, D. Comtois, F. Légaré, J.-C. Kieffer, P. B. Corkum, and D. M. Villeneuve, "Scaling of high harmonic generation efficiency with wavelength," in *Ultra-Fast Dynamic Imaging of Molecules II*, (Ischia Island (Naples), Italy), 2009. (Poster Presentation).
- [23] A. D. Shiner, C. Trallero-Herrero, N. Kajumba, P. B. Corkum, D. Villeneuve, B. Heidi-C, C. Daniel, F. Légaré, and J.-C. Kieffer, "Scaling of high harmonic generation efficiency with wavelength," in *2008 Gordon Research Conference on Multiphoton Processes*, 2008. (Poster Presentation).
- [24] A. D. Shiner, A. A. Madej, P. Dubé, and J. E. Bernard, "Absolute measurement of Rb transition frequencies at 422-nm using a femtosecond optical frequency comb," in *Deutsche Physikalische Gesellschaft Summer School on Attosecond Physics, Bad Honnef, Germany.*, 2007. (Poster Presentation).
- [25] H.-C. Bandulet, D. Comtois, T. Ozaki, J.-C. Kieffer, and F. Légaré, "High harmonic generation with a spatially-filtered optical parametric amplifier," in *2008 Gordon Research Conference on Multiphoton Processes*, 2008. (Poster Presentation).

- [26] A. Staudte, H. Akagi, D. Pavicic, A. D. Shiner, F. Turner, R. Doerner, D. M. Villeneuve, M. Y. Ivanov, and P. B. Corkum, "Alignment-free molecular frame tunneling probabilities," in *39th Annual Meeting of the APS Division of Atomic, Molecular, and Optical Physics*, 2008.
- [27] C. Trallero-Herrero, A. D. Shiner, N. Kajumba, C. Bandulet, Heidi, D. Comtois, F. Légaré, J.-C. Kieffer, P. Corkum, and D. Villeneuve, "Scaling of high harmonic generation efficiency with wavelength," in *39th Annual Meeting of the APS Division of Atomic, Molecular, and Optical Physics*, 2008.
- [28] H. Akagi, A. Staudte, A. D. Shiner, F. Turner, D. M. Villeneuve, M. Y. Ivanov, and P. B. Corkum, "Molecular frame angular distribution of tunnel ionization probability of HCL molecule determined by correlation measurement between electron and fragment ion," in *Second Annual Meeting of the Japan Society of Molecular Science*, 2008.
- [29] A. D. Shiner, C. D. Schaaff, R. C. Madter, R. S. Mathew, M. F. Habicher, and Q. A. Luong, "Over-voltage protection circuit." US Patent Number 20030076642, Filed April 24, 2003, Accepted April 25, 2006.
- [30] P. Béjot, B. E. Schmidt, J. Kasparian, J.-P. Wolf, and F. Legaré, "Mechanism of hollow-core-fiber infrared-supercontinuum compression with bulk material," *Phys. Rev. A*, vol. 81, p. 063828, Jun 2010.
- [31] A. H. Zewail, "Femtochemistry: Atomic-scale dynamics of the chemical bond using ultrafast lasers (nobel lecture)," *Angewandte Chemie International Edition*, vol. 39, no. 15, pp. 2586–2631, 2000.
- [32] G. Sansone, T. Pfeifer, K. Simeonidis, and A. I. Kuleff, "Electron correlation in real time," *ChemPhysChem*, vol. 13, no. 3, pp. 661–680, 2012.
- [33] M. W. Schmidt and M. S. Gordon, "The construction and interpretation of mcsf wavefunctions," *Annual Review of Physical Chemistry*, vol. 49, no. 1, pp. 233–266, 1998.
- [34] E. Dagotto, "Correlated electrons in high-temperature superconductors," *Rev. Mod. Phys.*, vol. 66, pp. 763–840, Jul 1994.
- [35] L. Cederbaum and J. Zobeley, "Ultrafast charge migration by electron correlation," *Chemical Physics Letters*, vol. 307, no. 34, pp. 205 – 210, 1999.
- [36] D. E. Spence, P. N. Kean, and W. Sibbett, "60-fsec pulse generation from a self-mode-locked ti:sapphire laser," *Opt. Lett.*, vol. 16, pp. 42–44, Jan 1991.
- [37] J.-C. Diels, *Ultrashort Laser Pulse Phenomena*. San Diego: Academic Press, 1996.

- [38] P. B. Corkum and F. Krausz, “Attosecond science,” *Nature Physics*, vol. 3, pp. 381–387, June 2007.
- [39] P. M. Paul, E. S. Toma, P. Breger, G. Mullot, F. Aug, P. Balcou, H. G. Muller, and P. Agostini, “Observation of a train of attosecond pulses from high harmonic generation,” *Science*, vol. 292, no. 5522, pp. 1689–1692, 2001.
- [40] M. Hentschel, R. Kienberger, C. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, “Attosecond metrology,” *Nature*, vol. 414, pp. 509–513, Nov. 2001.
- [41] “Exploring matter with the light of the SOLEIL synchrotron,” SOLEIL Communication Department. L’Orme des Merisiers, Saint-Aubin, BP48 91192 Gif-sur-Yvette Cedex, Dec. 2006. www.synchrotron-soleil.fr.
- [42] J.-F. Adam, J.-P. Moy, and J. Susini, “Table-top water window transmission x-ray microscopy: Review of the key issues, and conceptual design of an instrument for biology,” *Review of Scientific Instruments*, vol. 76, no. 9, p. 091301, 2005.
- [43] J. Kirz and C. Jacobsen, “The history and future of x-ray microscopy,” *Journal of Physics: Conference Series*, vol. 186, no. 1, p. 012001, 2009.
- [44] M. D. Seaberg, D. E. Adams, E. L. Townsend, D. A. Raymondson, W. F. Schlotter, Y. Liu, C. S. Menoni, L. Rong, C.-C. Chen, J. Miao, H. C. Kapteyn, and M. M. Murnane, “Ultrahigh 22 nm resolution coherent diffractive imaging using a desktop 13 nm high harmonic source,” *Opt. Express*, vol. 19, pp. 22470–22479, Nov 2011.
- [45] A. Ravasio, D. Gauthier, F. R. N. C. Maia, M. Billon, J.-P. Caumes, D. Garzella, M. Géléoc, O. Gobert, J.-F. Hergott, A.-M. Pena, H. Perez, B. Carré, E. Bourhis, J. Gierak, A. Madouri, D. Mailly, B. Schiedt, M. Fajardo, J. Gauthier, P. Zeitoun, P. H. Bucksbaum, J. Hajdu, and H. Merdji, “Single-shot diffractive imaging with a table-top femtosecond soft x-ray laser-harmonics source,” *Phys. Rev. Lett.*, vol. 103, p. 028104, Jul 2009.
- [46] I. McKinnie and H. Kapteyn, “High-harmonic generation: Ultrafast lasers yield X-rays,” *Nature Photonics*, vol. 4, pp. 149–151, Mar. 2010.
- [47] U. Becker and D. Shirley, *VUV and Soft X-Ray Photoionization*. Physics of Atoms and Molecules, Plenum Press, 1996.
- [48] R. Weinkauf, P. Schanen, A. Metsala, E. W. Schlag, M. Brgle, and H. Kessler, “Highly efficient charge transfer in peptide cations in the gas phase: threshold effects and mechanism,” *The Journal of Physical Chemistry*, vol. 100, no. 47, pp. 18567–18585, 1996.

- [49] R. Weinkauf, E. W. Schlag, T. J. Martinez, and R. D. Levine, "Nonstationary electronic states and site-selective reactivity," *The Journal of Physical Chemistry A*, vol. 101, no. 42, pp. 7702–7710, 1997.
- [50] H. Hennig, "Charge migration due to electron correlation in molecules: Development of a non-dyson-method," in *Special Topics in Nanoelectronics Seminar Series*, (Technische Universität Dresden), February 28th, 2005.
- [51] A. I. Kuleff, J. Breidbach, and L. S. Cederbaum, "Multielectron wave-packet propagation: General theory and application," *The Journal of Chemical Physics*, vol. 123, no. 4, p. 044111, 2005.
- [52] M. Protopapas, C. H. Keitel, and P. L. Knight, "Atomic physics with super-high intensity lasers," *Reports on Progress in Physics*, vol. 60, no. 4, p. 389, 1997.
- [53] B. M. Smirnov, *Physics of Atoms and Ions*. Graduate Texts in Contemporary Physics, Berlin: Springer-Verlag, 2003.
- [54] P. Eckle, A. N. Pfeiffer, C. Cirelli, A. Staudte, R. Drner, H. G. Muller, M. Bttiker, and U. Keller, "Attosecond ionization and tunneling delay time measurements in helium," *Science*, vol. 322, no. 5907, pp. 1525–1529, 2008.
- [55] C. P. J. Martiny, M. Abu-samha, and L. B. Madsen, "Counterintuitive angular shifts in the photoelectron momentum distribution for atoms in strong few-cycle circularly polarized laser pulses," *Journal of Physics B: Atomic, Molecular and Optical Physics*, vol. 42, no. 16, p. 161001, 2009.
- [56] M. Lein, "Streaking analysis of strong-field ionisation," *Journal of Modern Optics*, vol. 58, no. 13, pp. 1188–1194, 2011.
- [57] G. L. Yudin and M. Y. Ivanov, "Nonadiabatic tunnel ionization: Looking inside a laser cycle," *Phys. Rev. A*, vol. 64, pp. 013409–+, July 2001.
- [58] M. V. Ammosov, N. B. Delone, and V. P. Krainov *Sov. Phys. JETP*, vol. 64, p. 1191, 1986.
- [59] N. B. Delone and V. P. Krainov, "Tunneling and barrier-suppression ionization of atoms and ions in a laser radiation field," *Uspekhi Fizicheskikh Nauk*, vol. 41, pp. 469–485, May 1998.
- [60] C. Cohen-Tannoudji, B. Diu, and F. Laloë, *Quantum Mechanics*. Paris, France: Hermann and John Wiley & Sons, Inc., 1977.
- [61] M. Lewenstein, P. Balcou, M. Y. Ivanov, A. L’huillier, and P. B. Corkum, "Theory of high-harmonic generation by low-frequency laser fields," *Phys. Rev. A*, vol. 49, pp. 2117–2132, Mar. 1994.

- [62] M. Lewenstein, P. Salières, and A. L’Huillier, “Phase of the atomic polarization in high-order harmonic generation,” *Phys. Rev. A*, vol. 52, pp. 4747–4754, Dec 1995.
- [63] E. Hasović, M. Busuladžić, A. Gazibegovic-Busuladžić, D. Milosevic, and W. Becker, “Simulation of above-threshold ionization experiments using the strong-field approximation,” *Laser Physics*, vol. 17, pp. 376–389, 2007.
- [64] M. Y. Ivanov, “Strong field approximation: Basic formalism and high harmonic generation,” in *Lecture Notes on Strong Field Physics*, ch. 5, 2002.
- [65] M. Y. Ivanov, “Dealing with complicated integrals: saddle point-type methods and some applications,” in *Lecture Notes on Strong Field Physics*, ch. 1, 2002.
- [66] V. S. Yakovlev, M. Ivanov, and F. Krausz, “Enhanced phase-matching for generation of soft x-ray harmonics and attosecond pulses in atomic gases,” *Opt. Express*, vol. 15, no. 23, pp. 15351–15364, 2007.
- [67] J. Itatani, J. Levesque, D. Zeidler, H. Niikura, H. Pépin, J. C. Kieffer, P. B. Corkum, and D. M. Villeneuve, “Tomographic imaging of molecular orbitals,” *Nature*, vol. 432, pp. 867–871, Dec. 2004.
- [68] M. V. Frolov, N. L. Manakov, T. S. Sarantseva, M. Y. Emelin, M. Y. Ryabikin, and A. F. Starace, “Analytic description of the high-energy plateau in harmonic generation by atoms: Can the harmonic power increase with increasing laser wavelengths?,” *Phys. Rev. Lett.*, vol. 102, p. 243901, Jun 2009.
- [69] M. V. Frolov, N. L. Manakov, T. S. Sarantseva, and A. F. Starace, “Analytic formulae for high harmonic generation,” *Journal of Physics B: Atomic, Molecular and Optical Physics*, vol. 42, no. 3, p. 035601, 2009.
- [70] A.-T. Le, R. R. Lucchese, S. Tonzani, T. Morishita, and C. D. Lin, “Quantitative rescattering theory for high-order harmonic generation from molecules,” *Physical Review A (Atomic, Molecular, and Optical Physics)*, vol. 80, no. 1, p. 013401, 2009.
- [71] T. Morishita, A.-T. Le, Z. Chen, and C. D. Lin, “Accurate retrieval of structural information from laser-induced photoelectron and high-order harmonic spectra by few-cycle laser pulses,” *Phys. Rev. Lett.*, vol. 100, p. 013903, Jan 2008.
- [72] J. Tate, T. Augustine, H. G. Muller, P. Salières, P. Agostini, and L. F. Dimauro, “Scaling of Wave-Packet Dynamics in an Intense Midinfrared Field,” *Physical Review Letters*, vol. 98, p. 013901, Jan. 2007.
- [73] K. Schiessl, K. L. Ishikawa, E. Persson, and J. Burgdörfer, “Quantum Path Interference in the Wavelength Dependence of High-Harmonic Generation,” *Physical Review Letters*, vol. 99, p. 253903, Dec. 2007.

- [74] M. V. Frolov, N. L. Manakov, and A. F. Starace, “Wavelength scaling of high-harmonic yield: Threshold phenomena and bound state symmetry dependence,” *Physical Review Letters*, vol. 100, no. 17, p. 173001, 2008.
- [75] S. M. Hankin, D. M. Villeneuve, P. B. Corkum, and D. M. Rayner, “Intense-field laser ionization rates in atoms and molecules,” *Phys. Rev. A*, vol. 64, p. 013405, Jun 2001.
- [76] P. Salières and I. Christov, *Macroscopic Effects in High-Order Harmonic Generation*, vol. 134 of *Strong Field Laser Physics, Springer Series in Optical Sciences*, pp. 261–280. Berlin: Springer-Verlag, 2009.
- [77] T. Brabec and F. Krausz, “Intense few-cycle laser fields: Frontiers of nonlinear optics,” *Reviews of Modern Physics*, vol. 72, pp. 545–591, Apr. 2000.
- [78] C. Jin and C. D. Lin, “Comparison of high-order harmonic generation of ar using truncated bessel and gaussian beams,” *Phys. Rev. A*, vol. 85, p. 033423, Mar 2012.
- [79] R. W. Boyd, *Nonlinear Optics*. San Diego, California: Academic Press, 2nd ed., 2003.
- [80] P. Saures, A. L’huillier, P. Antoine, and M. Lewenstein, “Study of the spatial and temporal coherence of high-order harmonics,” vol. 41 of *Advances In Atomic, Molecular, and Optical Physics*, pp. 83 – 142, Academic Press, 1999. (Also available on arXiv: arXiv:quant-ph/9710060v1).
- [81] R. W. Boyd, *Nonlinear Optics*. San Diego, California: Academic Press, 3rd ed., 2008.
- [82] P. Balcou, P. Salières, A. L’Huillier, and M. Lewenstein, “Generalized phase-matching conditions for high harmonics: The role of field-gradient forces,” *Phys. Rev. A*, vol. 55, pp. 3204–3210, Apr 1997.
- [83] T.-C. Poon and T. Kim, *Engineering optics with MATLAB*. Engineering optics with MATLAB, by T.-C. Poon and T. Kim. Singapore: World Scientific Publishing, 2006, 2006.
- [84] J. F. James, *A student’s guide to Fourier transforms*. Cambridge UK: Cambridge University Press, 1995.
- [85] E. O’Neill, *Introduction to Statistical Optics*. New York: Dover Publications Inc., 1992.
- [86] E. A. J. Marcatili and R. A. Schmeltzer, “Hollow Metallic and Dielectric Waveguides for Long Distance Optical Transmission and Lasers,” *The Bell System Technical Journal*, pp. 1783–1809, July 1964.

- [87] M. Nisoli, E. Priori, G. Sansone, S. Stagira, G. Cerullo, S. De Silvestri, C. Altucci, R. Bruzzese, C. de Lisio, P. Villoresi, L. Poletto, M. Pascolini, and G. Tondello, "High-brightness high-order harmonic generation by truncated bessel beams in the sub-10-fs regime," *Phys. Rev. Lett.*, vol. 88, p. 033902, Jan 2002.
- [88] G. Agrawal, *Nonlinear Fiber Optics*. New York: Elsevier, 4 ed., 2007.
- [89] G. Genty, P. Kinsler, B. Kibler, and J. M. Dudley, "Nonlinear envelope equation modeling of sub-cycle dynamics and harmonic generation in nonlinear waveguides," *Opt. Express*, vol. 15, pp. 5382–5387, Apr 2007.
- [90] G. Cerullo and S. D. Silvestri, "Ultrafast optical parametric amplifiers," *Rev. Sci. Inst.*, vol. 74, no. 1, pp. 1–18, 2003.
- [91] J. M. Dudley and J. R. Taylor, *Supercontinuum Generation in Optical Fibers*. Cambridge,: Cambridge University Press, 2010.
- [92] G. Muslu and H. Erbay, "Higher-order split-step fourier schemes for the generalized nonlinear schrödinger equation," *Mathematics and Computers in Simulation*, vol. 67, no. 6, pp. 581 – 595, 2005.
- [93] G. Tempea, F. Krausz, C. Spielmann, and K. Ferencz, "Dispersion control over 150 thz with chirped dielectric mirrors," *Selected Topics in Quantum Electronics, IEEE Journal of*, vol. 4, pp. 193 –196, mar/apr 1998.
- [94] P. Dombi, V. S. Yakovlev, K. O'Keeffe, T. Fuji, M. Lezius, and G. Tempea, "Pulse compression with time-domain optimized chirped mirrors," *Opt. Express*, vol. 13, pp. 10888–10894, Dec 2005.
- [95] E. Goulielmakis, V. S. Yakovlev, A. L. Cavalieri, M. Uiberacker, V. Pervak, A. Apolonski, R. Kienberger, U. Kleineberg, and F. Krausz, "Attosecond control and measurement: Lightwave electronics," *Science*, vol. 317, no. 5839, pp. 769–775, 2007.
- [96] G. Sansone, E. Benedetti, F. Calegari, C. Vozzi, L. Avaldi, R. Flammini, L. Poletto, P. Villoresi, C. Altucci, R. Velotta, S. Stagira, S. De Silvestri, and M. Nisoli, "Isolated single-cycle attosecond pulses," *Science*, vol. 314, no. 5798, pp. 443–446, 2006.
- [97] E. Goulielmakis, M. Schultze, M. Hofstetter, V. S. Yakovlev, J. Gagnon, M. Uiberacker, A. L. Aquila, E. M. Gullikson, D. T. Attwood, R. Kienberger, F. Krausz, and U. Kleineberg, "Single-cycle nonlinear optics," *Science*, vol. 320, no. 5883, pp. 1614–1617, 2008.

- [98] M. F. Kling, C. Siedschlag, A. J. Verhoef, J. I. Khan, M. Schultze, T. Uphues, Y. Ni, M. Uiberacker, M. Drescher, F. Krausz, and M. J. J. Vrakking, "Control of electron localization in molecular dissociation," *Science*, vol. 312, no. 5771, pp. 246–248, 2006.
- [99] S. T. Cundiff, J. Ye, and J. L. Hall, "Optical frequency synthesis based on mode-locked lasers," *Review of Scientific Instruments*, vol. 72, pp. 3749–3771, Oct. 2001.
- [100] A. Baltuška, T. Udem, M. Uiberacker, M. Hentschel, E. Goulielmakis, C. Gohle, R. Holzwarth, V. S. Yakovlev, A. Scrinzi, T. W. Hänsch, and F. Krausz, "Attosecond control of electronic processes by intense light fields," *Nature*, vol. 421, pp. 611–615, Feb. 2003.
- [101] A. Baltuška, M. Uiberacker, E. Goulielmakis, R. Kienberger, V. Yakovlev, T. Udem, T. Hansch, and F. Krausz, "Phase-controlled amplification of few-cycle laser pulses," *Selected Topics in Quantum Electronics, IEEE Journal of*, vol. 9, pp. 972 – 989, july-aug. 2003.
- [102] A. Baltuška, T. Fuji, and T. Kobayashi, "Controlling the carrier-envelope phase of ultrashort light pulses with optical parametric amplifiers," *Phys. Rev. Lett.*, vol. 88, p. 133901, Mar 2002.
- [103] G. Cerullo, A. Baltuška, O. Mücke, and C. Vozzi, "Few-optical-cycle light pulses with passive carrier-envelope phase stabilization," *Laser & Photonics Reviews*, vol. 5, no. 3, pp. 323–351, 2011.
- [104] D. J. Jones, S. A. Diddams, J. K. Ranka, A. Stentz, R. S. Windeler, J. L. Hall, and S. T. Cundiff, "Carrier-Envelope Phase Control of Femtosecond Mode-Locked Lasers and Direct Optical Frequency Synthesis," *Science*, vol. 288, pp. 635–640, Apr. 2000.
- [105] S. Haykin and B. Van Veen, *Signals and Systems*. New York: John Wiley & Sons, 1999.
- [106] M. A. Levine, R. E. Marrs, J. R. Henderson, D. A. Knapp, and M. B. Schneider, "The electron beam ion trap: A new instrument for atomic physics measurements," *Physica Scripta*, vol. 1988, no. T22, p. 157, 1988.
- [107] A. F. Starace, "Theory of atomic photoionization," in *Handbuch Der Physik Vol. XXXI* (W. Mehlhorn, ed.), pp. 1–121, Berlin: Springer, 1982.
- [108] U. Fano and J. W. Cooper, "Spectral distribution of atomic oscillator strengths," *Rev. Mod. Phys.*, vol. 40, pp. 441–507, Jul 1968.

- [109] J. W. Cooper, "Photoionization from outer atomic subshells. a model study," *Phys. Rev.*, vol. 128, pp. 681–693, Oct 1962.
- [110] S. Minemoto, T. Umegaki, Y. Oguchi, T. Morishita, A.-T. Le, S. Watanabe, and H. Sakai, "Retrieving photorecombination cross sections of atoms from high-order harmonic spectra," *Physical Review A (Atomic, Molecular, and Optical Physics)*, vol. 78, no. 6, p. 061402, 2008.
- [111] H. J. Wörner, H. Niikura, J. B. Bertrand, P. B. Corkum, and D. M. Villeneuve, "Observation of electronic structure minima in high-harmonic generation," *Physical Review Letters*, vol. 102, no. 10, p. 103901, 2009.
- [112] J. W. Cooper, "Interaction of maxima in the absorption of soft x rays," *Phys. Rev. Lett.*, vol. 13, pp. 762–764, Dec 1964.
- [113] M. Y. Amusia and J.-P. Connerade, "The theory of collective motion probed by light," *Reports on Progress in Physics*, vol. 63, no. 1, p. 41, 2000.
- [114] M. Kutzner, V. Radojević, and H. P. Kelly, "Extended photoionization calculations for xenon," *Phys. Rev. A*, vol. 40, pp. 5052–5057, Nov 1989.
- [115] T. Popmintchev, M.-C. Chen, D. Popmintchev, P. Arpin, S. Brown, S. Ališauskas, G. Andriukaitis, T. Balčiunas, O. D. Mücke, A. Pugzlys, A. Baltuška, B. Shim, S. E. Schrauth, A. Gaeta, C. Hernandez-Garcia, L. Plaja, A. Becker, A. Jaron-Becker, M. M. Murnane, and H. C. Kapteyn, "Bright coherent ultrahigh harmonics in the kev x-ray regime from mid-infrared femtosecond lasers," *Science*, vol. 336, no. 6086, pp. 1287–1291, 2012.
- [116] P. Dietrich, F. Krausz, and P. B. Corkum, "Determining the absolute carrier phase of a few-cycle laser pulse," *Opt. Lett.*, vol. 25, pp. 16–18, Jan 2000.
- [117] C. Jin, J. B. Bertrand, R. R. Lucchese, H. J. Wörner, P. B. Corkum, D. M. Villeneuve, A.-T. Le, and C. D. Lin, "Intensity dependence of multiple orbital contributions and shape resonance in high-order harmonic generation of aligned n_2 molecules," *Phys. Rev. A*, vol. 85, p. 013405, Jan 2012.