Characterization of second harmonic generation in chiral metamaterials

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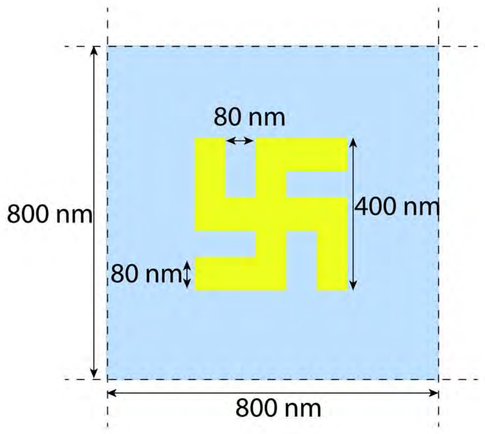
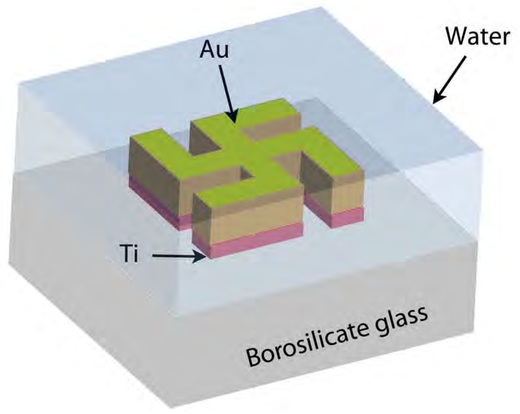
Introduction

Circular dichroism (CD), which is a differential absorption of left and right circularly polarized light, is a highly useful tool for detecting and characterizing chiral organic molecules. However, traditional methods of measuring the CD response of a sample characteristically produce prohibitively weak signals for small sample volumes due to the usually large difference in scale between the wavelength of the probing light and the molecules under study.

Recently, it has been shown that chiral metallic (plasmonic) metamaterials in the form of arrays of chiral-shaped gold nanostructures fabricated on a glass substrate can improve the sensitivity of CD measurements of chiral organic molecules adhered to their surface [1]. Moreover, it is well-known that second harmonic generation (SHG)-CD, defined as a disparity between the second harmonic signals produced by a material excited by either left or right circularly polarized light, is in general more highly pronounced than ordinary CD [2].The present study attempts to characterize the microscopic SHG-CD response of chiral metallic metamaterials with the aim of achieving a better understanding of the mechanisms that govern the enhancement of the SHG-CD response.

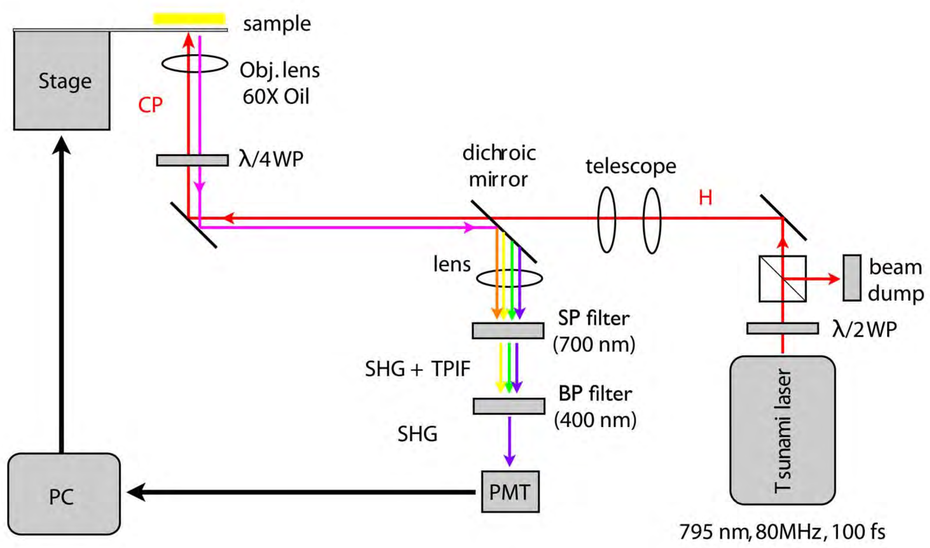
Methodology





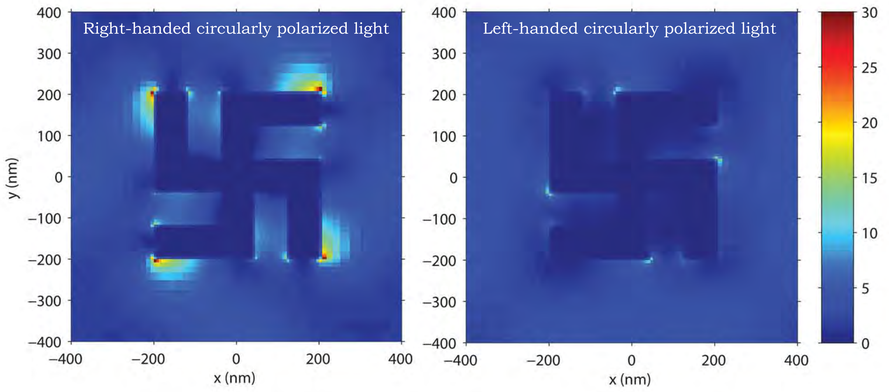
The chiral plasmonic metamaterial studied consisted of an array of right-handed gold gammadia deposited on a borosilicate glass substrate with the assistance of a 5 nm titanium adhesion layer. The gammadia array is a square lattice with an 800nm period, and consists of 400nm square gammadia with 80nm arm length and a thickness of 100nm (see above image).

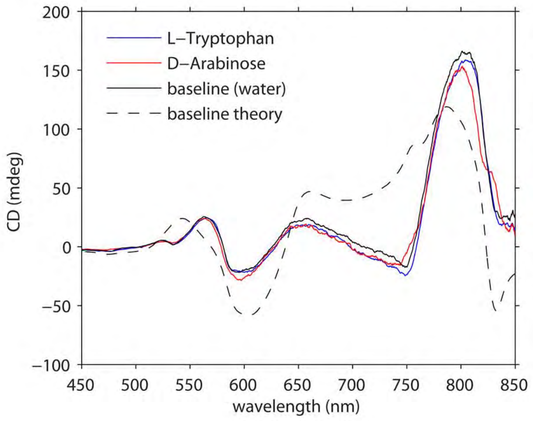
First, the metamaterial's response to circularly polarized light was simulated through the use of a commercial finite difference time domain (FDTD) package, Lumerical (version 8). UV-Visible spectroscopy was then performed using a commercial CD spectropolarimeter (the Jasco J810) in order to obtain the CD spectra of the metamaterial in the presence of water or tryptophan. Finally, SHG microscopy was attempted using the setup illustrated below.



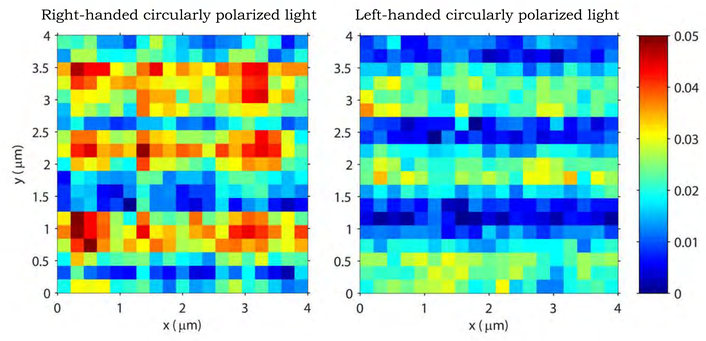
Results

Pictured on the right are FDTD simulated images revealing the electric field density near a right-handed gold gammadion illuminated with either right-handed circularly polarized light (left) or left-handed circularly polarized light (right). It is evident from these images that the electric field intensity near the gammadion is significantly greater when illuminating with right-handed circularly polarized light as opposed to left-handed circularly polarized light.





The CD spectra of right-handed gammadia were measured using the Jasco-J810 spectropolarimeter first in water, and then in solutions of L-Tryptophan and D-Arabinose. The results of that measurement are presented in the graph on the left. Included in the graph is the FDTD simulated CD spectrum of RH gammadia in water. Note the peaks at approximately 550 nm, 650nm and 800nm, each corresponding to a different plasmon resonance of the metamaterial. It can be seen that the positions of these peaks (especially the 800nm peak) shift when the water is replaced by either L-Tryptophan or D-Arabinose. Finally, images of the gammadion array were taken using the microscope setup. By scanning the sample through the beam, a two dimensional image of the SHG response of the gammadion array was obtained (see right). When illuminated with right-handed circularly polarized the SHG response of each gammadion was found to be very large (left image) whereas illumination of the array with left-handed circularly polarized light produced a much weaker result (right image).



Conclusion

The CD and SHG-CD response of chiral metallic metamaterials was characterized through the use of simulation, UV-Visible spectropolarimetry and SHG-CD microscopy. The SHG-CD observed in the microscopy experiment is attributed to the presence of more highly pronounced electric field "hot spots" in the metamaterial when illuminating with right-handed circularly polarized light as opposed to left-handed. Further research will involve studying the SHG response of the metamaterials in the presence of chiral liquid molecules adsorbed at the gold surface.

References / Acknowledgement

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[1] E. Hendry et al., Nat. Nanotechnol. 5, 783 (2010).
[2] L. Persechini and J. F. McGilp, "Chiral second-harmonic generation from small organic molecules at surfaces", Phys. Status Solidi B 249, No. 6, 1155–1159 (2012).