10th International Congress on Advanced Electromagnetic Materials in Microwaves and Optics – Metamaterials 2016
Crete, Greece, 17-22 September 2016

Giant Nonlinear Optical Activity from Planar Metasurfaces

Guixin Li[1], Shumei Chen[2, 5], Franziska Zeuner[1], Martin Weismann[3], Bernhard Reineke[1], Ventsislav Kolev Valev[4], Kok Wai Cheah[5], Nicolae Coriolan Panoiu[3], Shuang Zhang[2], and Thomas Zentgraf[1]

[1]Department of Physics, University of Paderborn, Warburger Straße 100 D-33098 Paderborn, Germany
[2]School of Physics & Astronomy, University of Birmingham, Birmingham B15 2TT, UK
[3]Department of Electronic and Electrical Engineering, University College London, Torrington Place, London, WC1 E7JE, UK
[4]Department of Physics, University of Bath, Claverton Down, Bath, BA2 7AY, UK
[5]Department of Physics, Hong Kong Baptist University, Kowloon Tong, Hong Kong

Abstract **- Second harmonic generation circular dichroism (CD) is more sensitive to the handedness of chiral materials than its linear optical counterpart. In this work, we show that 3D chiral structures are not necessary for introducing strong CD for harmonic generations. Specifically, we demonstrate giant CD for both second harmonic generation and third harmonic generation on suitably designed ultrathin plasmonic metasurfaces. It is experimentally and theoretically verified that the overwhelming contribution to this nonlinear CD is of achiral origin. The results shed new light on the origin of the nonlinear CD effect in achiral planar surfaces.**

I. INTRODUCTION

In plasmonic nanomaterials, light can excite coherent oscillations of the surface electrons, which lead to strongly enlarged electromagnetic near-fields. Such enlargements are useful to enhance the light-matter interaction with chiral molecules, leading to vast improvements in sensitivity to the chiral purity of pharmaceuticals, agrochemicals, biomolecules, etc. Chiral plasmonic nanomaterials in particular have been investigated for their selective interaction with circularly polarized light, which can enable, for instance, highly localized control of circularly polarized light emission. On the other hand, there has been a growing interest in the investigation of the nonlinear optical properties of plasmonic nanostructures and optical metamaterials due to the associated strong enhancements of electromagnetic fields and the capability to engineer the structural symmetry of their unit cell [1-5]. In particular, nonlinear optical CD of metamaterials and plasmonic nanostructures have been demonstrated. However, the observed nonlinear optical CD in most experiments arises either from the chirality in 3D nanostructures, or from the extrinsic contribution in the case of 2D structures when the fundamental beam is incident at oblique angles. It should be pointed out that even seemingly 2D chiral materials, such as monolayers of chiral thin-film nanostructures are intrinsically 3D chiral as recently demonstrated in the linear and in the nonlinear optical regimes. In such nanostructures, chirality arises from the presence of a substrate on one side of the thin-films and not on the other.

Here, we demonstrate giant CD for both second and third harmonic generation on ultrathin plasmonic metasurfaces with broken in-plane mirror symmetry. Despite the symmetry breaking along the surface normal direction due to the presence of substrate, this CD is primarily of achiral origin.

II. EXPERIMENT

The Trisceli- and Gammadion-type plasmonic nanostructures are fabricated on ITO coated BK7 glass substrates by using electron-beam lithography and a lift-off process. As shown in Fig. 1a and 1b, these two types of gold nanostructures with thickness of 30 nm are arranged in triangular and square lattices with periods of 400

nm and 500 nm, respectively. Fig. 1c and 1d show the measured transmission spectra of the plasmonic nanostructures for horizontally polarized incident and transmitted light by using Fourier transform infrared spectrometer. The dips in the transmission spectra correspond to the excitation of localized surface plasmon polariton modes of the Trisceli- and Gammadion-type plasmonic nanostructures at wavelength of 1165 nm and 1230 nm, respectively, which are confirmed by the numerical simulations.

We use a femtosecond laser beam under normal incidence to the sample surface to interact with the nanostructures. Moreover, we used a low numerical aperture objective (NA=0.1 to minimize the projection of a polarization component along the propagation direction. The signals of harmonic generation are then collected in the transmission direction. To characterize the frequency dependent nonlinear optical activity, the wavelength of illuminating laser is tuned between λ_o =1100 nm and 1400 nm and the circularly polarized SHG and THG signals are recorded with a spectrometer. We found that SHG and THG signals from the plasmonic nanostructures have opposite polarization states as compared to that of the illuminating laser. However, we also measured extremely low SHG and THG with the same polarization state as that of the illuminating laser although they are theoretically forbidden in the dipole approximation but can be observed due to the imperfections of nanofabrication. [3-5]

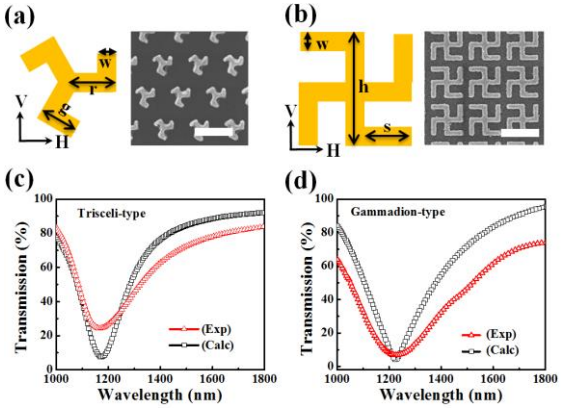

Fig. 1. Geometry parameters and transmission properties of the plasmonic nanostructures. (a) and (b) Schematic view of single Trisceli- and Gammadion- type plasmonic nanostructures and scanning electron microscopy images of the fabricated periodic pattern (scale bar: 500 nm). The 30 nm-thick gold structures are arranged in triangular and square lattices with periods of 400 nm and 500 nm, respectively. The geometry parameters of the gold nanostructures are w = 60 nm, r = 110 nm, g = 100 nm, h = 410 nm and s = 175 nm. (c)and (d) Measured ('Exp') linear transmission spectra of the Trisceli- and PFO coated Gammadion- type nanostructures for linearly (horizontally) polarized light, exhibiting localized plasmon resonance at wavelength of 1165 nm and 1230 nm, respectively, which agree well with the calculated ('Calc') transmission spectra.

Based on the measured spectral results for both SHG and THG, we obtained the nonlinear CDs by (I_{LCP}-I_{RCP})/(I_{LCP}+I_{RCP}) (Fig. 2a and 2c). The SHG-CD varies from negative to positive values, crossing zero around 1150 nm and reaching near-unity value of 0.98 at 1280 nm. On the other hand, the THG-CD reaches its maximum value of 0.79 at 1280 nm. We note that this is the first observation of THG-CD on planar chiral plasmonic structures. Furthermore, the maximum value of the measured nonlinear SHG-CD is much higher than that of the linear CD effect observed previously from the Gammadion-type plasmonic nanostructures. [6] We also found very similar trends of the nonlinear CDs from the simulated SHG and THG signals (Fig. 2b and 2d), where the simulated maximum values of the SHD-CD and the THG-CD are 0.637 at 1271 nm and 0.435 at 1301 nm, respectively. These values are somewhat lower than their respective measured values.

III. Conclusion

In summary, we demonstrate near unity nonlinear circular dichroism for both second and third harmonic generations with suitably designed ultrathin Trisceli- and Gammadion-type plasmonic nanostructures with three- and four-fold rotational symmetry. These two kinds of nanostructures allow a symmetry controlled generation of circularly polarized SHG and THG, respectively. Both giant SHG-CD and THG-CD are experimentally observed for the planar plasmonic nanostructures with negligible linear optical activity. Importantly, the observed SHG-

CD is identified to have an achiral origin. The simple fabrication of two dimensional plasmonic structures and wider applicability of SHG and THG techniques also enable more freedoms in designing chip-type nonlinear optoelectronic devices.

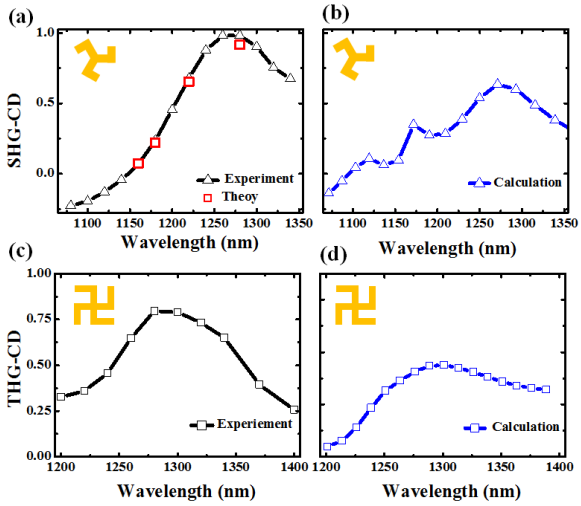

Fig. 2. SHG-CD and THG-CD from the Trisceli- and Gammadion- type plasmonic nanostructures. (a) and (c) Measured (Exp) wavelength dependent SHG-CD and THG-CD. The values for the squares in (a) are obtained from the calculation (Calc) of SHG-CD based on the experimentally determined values of the effective nonlinear susceptibilities. Both SHG-CD and THG-CD have a broadband response for fundamental wavelength between 1200 nm and 1350 nm. The SHG-CD experiences a sign change at wavelength of ~ 1150 nm, which is close to the theoretical prediction of 1100 nm. (b) and (d) Corresponding numerically simulated (Simulation) nonlinear CD spectra for the structures. Both of the calculated SHG and THG spectra show similar trend as the measured ones. While the peak of SHG-CD is close to the measured value, the calculated THG has a much higher deviation from our experimental data, which can be attributed to the imperfections of nanofabrication.

ACKNOWLEDGEMENT

The authors would like to acknowledge support from Cedrik Meier by providing his electron beam lithography system. This work was financially supported by EPSRC (Grant No EP/J018473/1), Leverhulme (grant no. RPG-2012-674) and the Deutsche Forschungsgemeinschaft (Grant Nos. TRR142/A05 and ZE953/7-1).

REFERENCES

[1] M. Kauranen, A. V. Zayats, Nature Photon. 6, 737, 2012.

[2] M. Lapine, I. V. Shadrivov, Y. S. Kivshar, Rev. Mod. Phys. 86, 1093, 2014.

[3] K. Konishi, T. Higuchi, J. Li, J. Larsson, S. Ishii, M. Kuwata-Gonokami, Phys. Rev. Lett. 112, 135502, 2014.

[4] S. M. Chen, G. X. Li, F. Zeuner, W. H. Wong, E.Y. B. Pun, T. Zentgraf, K. W. Cheah, S. Zhang, Phys. Rev. Lett. 113, 033901, 2014.

[5] G. X. Li, S. M. Chen, N. Pholchai, W. H. Wong, E.Y. B. Pun, K. W. Cheah, T. Zentgraf, S. Zhang, Nature Mater. 14, 607, 2015.

[6] M. Kuwata-Gonokami, N. Saito, Y. Ino, M. Kauranen, K. Jefimovs, T. Vallius, J. Turunen, Y. Svirko, Phys. Rev. Lett. 95, 227401, 2005.