From Nonlinear Optics to Nonlinear Plasmonics: Giant Nonlinear Polarization Effects in Metamaterials

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Abstract: We report that engineering of chiral and nonlinear optical properties in plasmonic metamaterial allows the observation of Nonlinear Optical Activity that is millions of times stronger than in natural materials offering a potential for practical applications.
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Through observation of giant intensity-dependent polarization effects we provide a powerful illustration that nanoscale nonlinear plasmonics of metamaterials offers extremely strong effects unfolding in nanoscale volumes of nonlinear medium that could lead to applications in modulation of light intensity and polarization in nanophotonic devices.

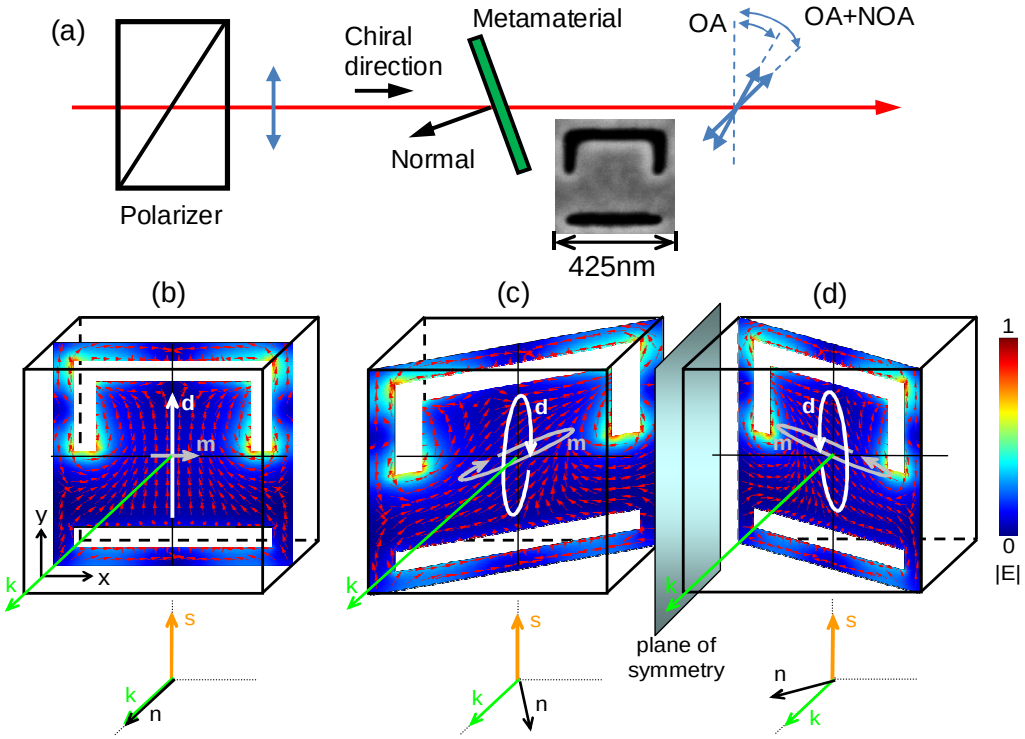

Fig. 1. (a) Observation of intensity-dependent polarization rotation (Nonlinear Optical Activity) in a gold plasmonic metamaterial along a chiral direction where nonlinearity is resonantly enhanced by nanoscale confinement of light. The inset shows an SEM image of a metamaterial unit cell. (b) Non-chiral and (c,d) chiral experimental arrangements of opposite handedness. The electric field magnitude (color map) and instantaneous direction (arrows) are shown alongside the meta-molecule's effective electric *d* and magnetic *m* dipoles, which are responsible for chiral scattering at oblique incidence. The sign of optical activity changes with the handedness of the mutual arrangement of incident wave vector *k*, metamaterial orientation *s* and surface normal *n*.

Giant intensity-dependent polarization rotation (Nonlinear Optical Activity) was observed at a plasmonic resonance of a gold nanostructure which had been periodically nanostructured with an array of asymmetric split ring apertures, see Fig. 1(a). The structure's optical properties were studied using a femtosecond tunable laser with 115 fs pulse duration and 80 MHz repetition rate. The easy-to-fabricate planar metamaterial exhibits exceptionally large linear and Nonlinear Optical Activity at oblique incidence, when it forms a chiral arrangement with the direction of

incidence. Two experimental arrangements of opposite handedness, leading to Nonlinear Optical Activity of opposite sign, are shown by Fig. 1(c,d). For example at 20 deg incidence, the 50 nm thick film rotates the polarization state light by 29 deg in the low intensity limit, see Fig. 2(a). At an average power of only 1.3 mW (peak intensity 1 GW/cm^2) we observe a nonlinear decrease of polarization rotation by about 1 deg, see Fig. 2(b). This corresponds to a specific nonlinear polarization rotation (deg/cm) per unit of intensity (W/cm^2) of 6×10^{-4} deg cm/W, compared to 10^{-11} deg cm/W for natural lithium iodate crystal where the effect was first discovered. Apart from polarization rotation, the structure also shows circular dichroism and relatively weak linear birefringence and linear dichroism, which all show similarly enormous nonlinear changes.

The 7 orders of magnitude improvement results from the fact that the structure's optical activity is linked to a plasmonic resonance at 956 nm, which also leads to a several orders of magnitude increase of nonlinear two-photon absorption via the local field enhancement mechanism. Pump-probe measurements of the underlying nonlinear absorption indicate that the metamaterial's nonlinear response if faster than 100 femtoseconds [1], allowing the modulation of intensity and polarization of light with light at THz rates.

Observation of these giant polarization effects provides a powerful illustration that nanoscale nonlinear plasmonics of metamaterials offers extremely strong effects unfolding in nanoscale volumes of nonlinear medium that could lead to applications in ultrafast modulation of light intensity and polarization in nanophotonic devices.

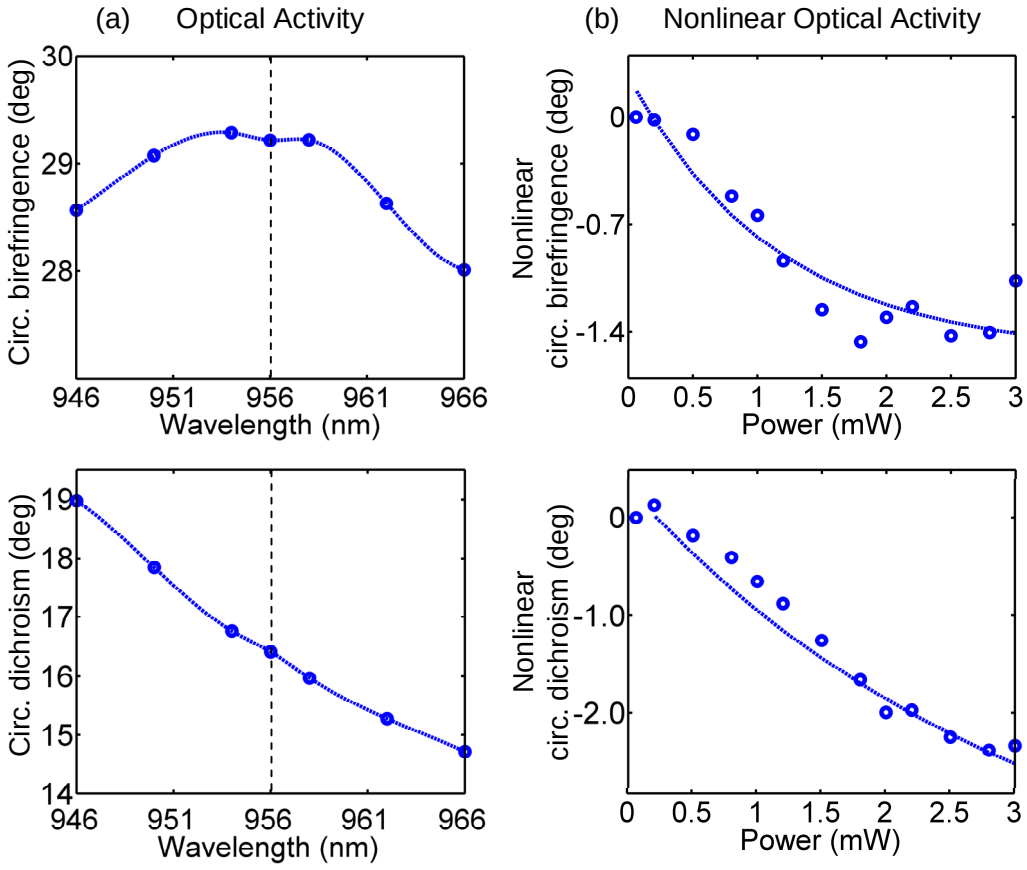

Fig. 2. (a) Linear and (b) Nonlinear Optical Activity in terms of circular birefringence (polarization rotation) and circular dichroism and their dependence on intensity at 956 nm. The angle of incidence is 20 deg in both cases.

[1] M. Ren, B. Jia, J. Ou, E. Plum, J. Zhang, K. F. MacDonald, A. E. Nikolaenko, J. Xu, M. Gu and N. I. Zheludev, "Nanostructured plasmonic medium for terahertz bandwidth all-optical switching," Adv. Mater. doi: 10.1002/adma.201103162 (2011).