Third Harmonic Generation in Fishnet Metamaterials

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Abstract: Optical third harmonic generation in gold-based fishnet metamaterials is studied. Pumping the magnetic resonance of the metamaterial showed an additional contribution to third-order optical nonlinearities rising from antisymmetric structure of the currents in metamaterial layers.

Plasmonic metamaterials represent nanostructured films of noble metals, which possess optical properties not observable for bulk metallic medium. It was shown recently that depending on their geometry metamaterials possess unique optical properties that can used for optical cloaking [1], superlens [2] and negative refraction [3]. In contrast to linear optical properties of metamaterials with negative refraction, which are well understood, their nonlinear optical properties began to attract the attention of researchers only recently. Experiments are largely limited to the spectral dependence of optical harmonic generation in such metamaterials [4]. In this paper we study the angular dependence of third harmonic generation in fishnet metamaterials [5]. It was shown that such structures possess an effective negative refractive index in the near infrared region of the electromagnetic spectrum that is associated with the excitation of the effective magnetic resonance, due to excitation of surface plasmons in the metamaterial. Also, subpicosecond-resolution third harmonic generation chronograms revealed pronounced photoinduced change in third-order optical nonlinearities of metamaterial.

The proposed structure was defined by e-beam-lithography and lift-off technique on a SiO_2 substrate. The structure has a period of 500 nm in both lateral directions. The thicknesses of the Au films and the intermediate dielectric MgO film are 20 nm and 35 nm, respectively. Spectroscopy of the linear absorption for different angles of incidence with the angle varying from 0^o to 50^o was carried out. The normal incidence IR absorption spectrum demonstrates an absorption peak at 1.54 μm corresponding to the excitation of the magnetic plasmon mode, which position is blue-shifted as the angle of incidence is increased. For the nonlinear measurements a setup based on an optical parametric amplifier (OPA) was used operating at wavelengths of 1.49, 1.54, 1.56, and 1.60 μm and having an average output power of 3 mW focused to a 300 μm spot from the air side of the sample. The OPA was pumped by a Nd:YAG laser with a pulse duration of 5 ps and a repetition rate of 5 kHz. The resulting fluence took values up to 700 $\mu J/cm^2$ in the plane of the sample. The sample was placed on a six-axis positioning stage such that during the angular spectroscopy the beam was always focused into the same spot. The forward propagating third harmonic generation (THG) signal pulses were detected by a photomultiplier tube and gate-integrated by an oscilloscope. We used the p-p polarization configuration—illuminating with p-polarized light and selecting only the p-polarized part of forward propagating light before the detector. For all measurements spectral filtering before the detector was used for picking up the desired wavelength. With these filters the third harmonic response was orders of magnitude larger than signals at other wavelength, i.e., at the pump wavelength. The averaged THG signal from the pure SiO_2 substrate measured outside the metamaterial area was at least one order of magnitude lower than that from the metamaterial area. Contributions from the substrate were therefore safely neglected.

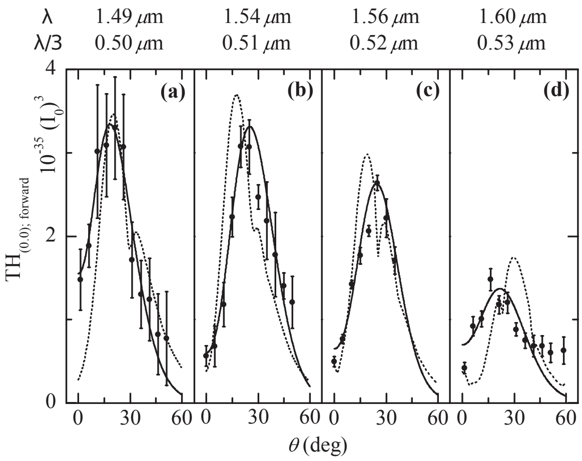

Fig.1. Panels (a)–(d) show the third harmonic signal as a function of the angle of incidence for different wavelengths in the spectral vicinity to the magnetic resonance. The black dots represent the experimental data and the dotted lines represent the simulation results. The solid lines are analytically derived curves.

The third harmonic intensity was measured and simulated in the forward zeroth diffraction order with the fundamental wavelength exciting the magnetic resonance. The angular spectra of THG are provided in Figs. 1(a)–1(d) for the fundamental wavelengths of 1.49, 1.54, 1.56, and 1.60 μm, respectively. The magnetic resonance position for normal incidence is 1.54 μm. The maximum of the THG signal is seen at angles of incidence around 20°. The appearance of this maximum is detailed in the discussion section and is believed to be caused by the interference of THG from the individual layers forming the fishnet metamaterial. The simulation shows an agreement with the experimental values.

Plasmon-enhanced THG at the magnetic resonance of fishnet metamaterials was reported previously [4]. It was shown that the THG spectra obey the principles of the local-field enhanced nonlinear response. It was proposed that the wavelength dispersion of the THG efficiency is defined by the spectral line of the magnetic resonance cubed. The maximum of THG at angles of about 20° can neither be explained by means of dispersion of the local field factor at the fundamental frequency nor with the linear transmission characteristics at the third harmonic wavelength. Finally, the position of the maximum does not coincide with the angular position of the propagating diffraction order appearance. An analytical model was elaborated showing that, first, this feature is caused by retardation effects, and second, it is specific to the antisymmetric electric current structure of the magnetic resonance [6].

Finally, investigation of the temporal dynamics of THG using the pump-probe technique was performed. The experimental data indicate the existence of a delay in the nonlinear response of the metamaterial. The delay is attributed to the ultrafast relaxation processes in gold characterized by time constant of 1 ps that is comparable to the electron-phonon relaxation time.

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