Giant femtosecond optical nonlinearity of gold metamaterial nanostructures

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Abstract

We demonstrate that the magnitude and sign of the third-order nonlinear susceptibility of plasmonic metals can be controlled by nanostructuring. By patterning a gold film on the nanoscale to support a high quality plasmonic mode, enhancement of its two-photon absorption coefficient by two orders of magnitude is realized. With a response time of <115 fs, nonlinear gold metamaterials have potential applications in ultrafast optical limiting and all-optical data processing with terahertz bandwidth.

1. Introduction

Nonlinear optical processes such as Raman scattering, frequency mixing and multi-photon absorption are usually weak phenomena which become important only at very high intensities. This fundamental challenge has been addressed very successfully in surface enhanced Raman spectroscopy, where the Raman scattering cross-section is amplified up to 15 orders of magnitude by large local electric fields in the vicinity of resonant plasmonic nanostructures. More recently, second and third harmonic generation were observed directly from plasmonic nanostructures [1, 2] and plasmon-enhanced ultrafast nonlinear bleaching has been reported for metamaterial resonators hybridized with carbon nanotubes [3].

Here we demonstrate that the magnitude and sign of the third-order nonlinear absorption of gold films can be enhanced and controlled by nanopatterning. In our experiments, patterning of a gold film with plasmonic nanostructures leads to two order of magnitude enhancement of the metal's two-photon absorption coefficient, which reaches $\beta_{\mathrm{MM}} = 7.7 \times 10^{-6}$ m/W at the plasmonic resonance. The nonlinear response is shown to be ultrafast (<115 fs), making nonlinear gold nanostructures a promising component for ultrafast optical limiting and all-optical data processing.

2. Controlling optical nonlinearity with plasmonic resonances

The metamaterial nanostructure consists of a 50 nm thick gold film supported by a quartz substrate and is periodically patterned with an array of aperture-type asymmetrically split plasmonic ring meta-molecules, see inset of Fig. 1(a). The unit cell size is 425 nm. The structure supports a narrow plasmonic

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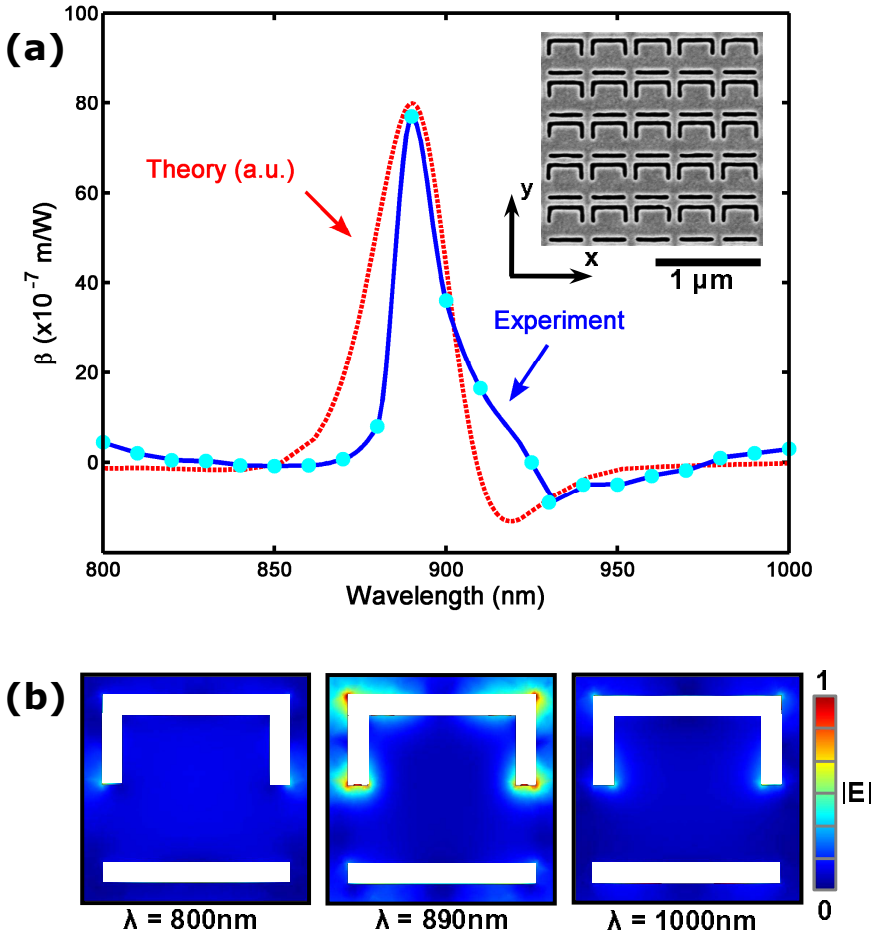

Fig. 1: Plasmonic resonant enhancement of the metal's ultrafast nonlinearity by a metamaterial nanostructure: (a) The measured ($\beta_{\mathrm{MM}}^{\mathrm{exp}}$, data points) and theoretically predicted ($\beta_{\mathrm{MM}}^{\mathrm{theo}}$ in arbitrary units, dashed line) effective two-photon absorption coefficients of the nanostructured gold film. The inset shows a SEM image of the plasmonic metamaterial. (b) Simulations of the resonant (890 nm) and off-resonant (800 nm and 1000 nm) local electric fields 10 nm below the metamaterial's gold surface.

resonance at about 890 nm, which can be excited by incident y-polarized light and is associated with large electric field enhancement within the gold film, see Fig. 1(b).

We studied the metamaterial's nonlinear absorption properties around its plasmonic resonance wavelength using a microscopic opened aperture Z-scan method [4]: The sample was scanned through the focus of a femtosecond laser beam and the intensity-dependence of the transmitted signal was recorded using lock-in detection. Using the fitting procedure described by Ref. [4], the two-photon absorption coefficient β_{MM} was retrieved, see blue curve in Fig. 1(a), which shows a sharp peak at the plasmonic resonance (890 nm) reaching 7.7×10^{-6} m/W. The corresponding third-order nonlinear susceptibility is about $\mathrm{Im}(\chi_{MM}^{(3)}) = 3.6 \times 10^{-6}$ esu. We investigated the dynamic behavior of this giant nonlinear response using pump-probe spectroscopy: The pump-induced changes of the metamaterial's transmission were measured as a function of the time delay between y-polarized high energy pump and low energy probe pulses. The presence of the pump pulse leads to an additional two-photon absorption process involving a pump and a probe photon and therefore leads to a reduction of the transmitted probe light. The resultant two-photon absorption was found to be symmetric with respect to the pump-probe pulse delay and proportional to the optical autocorrelation function of the laser pulses, indicating that the nonlinear process must be faster than the optical pulse duration ($\sim$115 fs).

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As explained below, their ultrafast and exceptionally large third-order nonlinearity makes nanostructured gold films very promising components for optical limiters, modulators and saturable absorbers. For example, at the plasmonic resonance, two-photon absorption is so large that the transmitted power is essentially constant for input intensities between 3.3-5 GW/cm^2, and thus the structure acts as an ideal optical limiter with a response time of less than 115 fs. Furthermore, two-photon absorption was observed to lead to a modulation of the metamaterial's transparency of up to 57%, allowing ultrafast all-optical modulation of light signals at THz rates. In contrast to unstructured gold, we found that nanostructuring can even reverse the sign of β_{MM}, see Fig. 1(a) at 930 nm. Here the metamaterial becomes more transparent under intense laser exposure, which paves a way towards novel saturable absorbers for mode-locking of lasers. Compared to nonlinear materials used in typical semiconductor saturable absorber mirrors (SESAM), which are usually used in such applications, nanostructured gold films offer higher nonlinearity which can be controlled by design.

Following Ref. [5], enhancement of the metal's two-photon absorption coefficient due to amplification of the local electric field $\mathbf{E_{Loc}}$ near the metamaterial's plasmonic resonance frequency can be predicted as

$$\beta_{\mathrm{MM}}^{\mathrm{theo}} = \mathrm{Re}(f)\frac{n_{\mathrm{Au}}^2}{n_{\mathrm{MM}}^2}\beta_{\mathrm{Au}} \tag{1}$$

with the nonlinearity enhancement factor

$$f = \frac{\int_{\mathrm{Au}} \mathbf{E_{Loc}}^2 |\mathbf{E_{Loc}}|^2 \mathrm{d}V}{\mathbf{E_0}^2 |\mathbf{E_0}|^2 V_{\mathrm{Au}}}. \tag{2}$$

n_{MM} and n_{Au} are the effective refractive indices of metamaterial and unstructured gold, respectively. β_{Au} is the two-photon absorption coefficient of bare gold, while $\mathbf{E_0}$ is the incident electric field and V_{Au} is the gold volume. As illustrated by Fig. 1(a), the experimentally-observed dramatic resonant enhancement of the two-photon absorption coefficient of gold is accurately reproduced by this theoretical model.

3. Conclusion

In conclusion, we found that the third-order nonlinearity of plasmonic metal films can be controlled and greatly enhanced by nanostructuring. By structuring a gold film with a plasmonic metamaterial pattern we enhanced its two-photon absorption coefficient by two orders of magnitude. The giant nonlinear response is a consequence of resonant local field enhancement near the metamaterial's plasmonic resonance. With an ultrafast response time shorter than 115 fs and an exceptionally high two-photon absorption coefficient, nanostructured gold films are an excellent candidate for practical applications as ultrafast optical limiters, terahertz bandwidth optical modulators and mode-locking devices for femtosecond pulsed lasers.

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