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Third-harmonic spectroscopy of electric and magnetic resonances in all-dielectric oligomers

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Abstract

We perform third harmonic generation (THG) spectroscopy of silicon nanodisk oligomers to reveal the contributions of magnetic and electric dipolar resonances to the third-order nonlinearity. Due to the local field enhancement of the pump beam upon excitation of the oligomer resonances, the THG is strongly enhanced with respect to the bulk crystalline silicon. By changing the oligomer geometry we bring both resonances to a partial overlap, which causes a strong THG enhancement for wavelengths of the strongest overlap. This effect is explained in the framework of constructive interference of the third harmonic signal generated by the electric and magnetic dipolar resonances.

1. Introduction

Already since the development of the Mie scattering theory it was realized that high-index dielectric particles can exhibit artificial magnetic resonances due to the excitation of circular displacement currents [1]. Recently, this fundamental phenomenon was observed experimentally throughout the entire visible and infrared spectral ranges for silicon nanospheres [2, 3], opening completely new functionalities for nanophotonics devices. In particular, the interference between the electric and magnetic optically induced dipoles results in azimuthally symmetric unidirectional scattering [5]. Such unique scattering phenomena have resulted is a vast number of works on high-index dielectric nanostructures with magnetic resonances at optical wavelengths, including silicon disks and oligomers. Importantly, it was predicted that in multi-particle silicon oligomers, the electric and magnetic dipolar moments can interfere, leading to a novel type of Fano resonances [4], which are highly sensitive to the changes of the environment. These results have prompted towards the investigation of nonlinear effects in all-dielectric resonant structures.

However, the nonlinear scattering of magento-electric all-dielectric nanostructures have never been studied to date. In addition, silicon is a material with a high third-order nonlinearity, therefore a number of interesting phenomena can be expected in the nonlinear scattering of high-index dielectric nanoparticles.

In this work, for the first time to our knowledge, we study the nonlinear scattering from silicon disk trimers in the vicinity of the electric and magnetic resonances. We show that by engineering of the modes of such trimers we can control the nonlinear interference of the magnetic and electric dipoles, including strong enhancement of THG and the presence of nonlinear Fano-type resonances.

2. Experimental approach

Silicon nanodisk trimers (Fig. 1(a)) were fabricated via electron-beam lithography (EBL) on backside polished silicon-on-insulator (SOI) wafers. The EBL process is followed by a directive reactive-ion etching process using the obtained electron-beam resist pattern as an etch mask [6]. A scanning electron micrograph of a fabricated trimer is shown in Fig. 1(b). In this way, different samples of trimers were fabricated, where the size (d) of the individual disks was varied. In addition, two sets of sample with different distances (D) between the three disks of the trimer were fabricated, as seen in the top and bottom panels of Fig. 2. This allows for precision engineering of the mutual position of the electric and magnetic resonances of the trimers.

The extinction spectrum of an array of trimers is also calculated utilizing finite element method. Fig. 1(c) shows the scattering cross section spectrum for trimers with $d = 380\,\text{nm}$, $D = 455\,\text{nm}$, $h = 260\,\text{nm}$ and an array period $p = 2\,\mu\text{m}$. The spectrum reveals two distinct peaks of scattering, which correspond to the electric and magnetic resonances of the trimers. The long wavelength resonance is the fundamental magnetic resonance, while the resonance at $0.91\,\mu\text{m}$ is the electric resonance of the trimer; the identification of the resonances is derived through multipole decomposition (not shown). We note that these resonances appear similar to the electric and magnetic resonances of individual silicon spheres or nanodisks, however the two modes of the trimer are not orthogonal and can interfere, forming the Fano-type resonances. The corresponding electric field profiles of the two modes are shown in Figs. 1(d,e), respec-

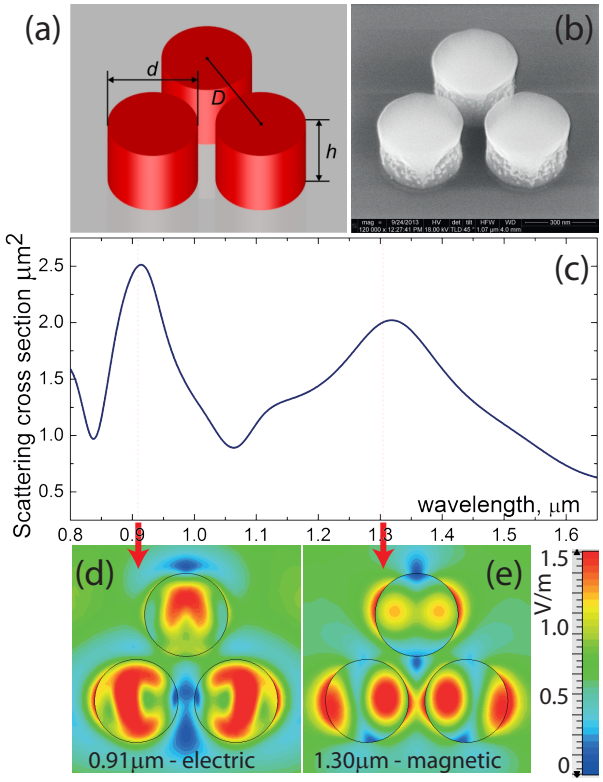

Figure 1: (a) Schematic of a nanodisk trimer characterized by the disk diameter d, center-to-center distance D, and height h. (b) SEM image of a SOI-based Si nanodisk trimer. (c) Simulated transmission spectrum of a rectangular array of trimers with $d = 380\,\text{nm}$, $D = 455\,\text{nm}$, $h = 260\,\text{nm}$, and array period $p = 2\,\mu\text{m}$. The resonant wavelength values correspond to the electric resonance at $\lambda_{\text{ED}} = 0.91\,\mu\text{m}$ and magnetic resonances at $\lambda_{\text{MD}} = 1.30\,\mu\text{m}$. (d,e) Corresponding local electric field ($|E|$) distributions for horizontal incident polarisation.

tively. The strong localization of the optical fields inside the silicon disks is clearly visible. Therefore, nonlinear effects such as THG are expected to be strongly enhanced at the resonance frequencies. However it is an open question what would be the spectral dependence of the THG due to the possible interference of both resonances.

To understand the process of nonlinear interference, we perform nonlinear THG spectroscopy on different silicon trimer arrays. We use a titanium-sapphire oscillator pumping an optical parametric oscillator, which produces a train of 200 fs pulses with a carrier wavelength adjustable from 1 μm to 1.6 μm. The beam is modulated by a chopper at 2.7 kHz and focused by an aspheric lens to a waist of 20 μm in diameter, leading to a maximal peak intensity value of $I = 2\,\text{GW/cm}^2$. The pump beam is focused in a way that it passes the silicon handle wafer first, so that the transmitted TH beam is not absorbed in the bulk silicon of the SOI wafer. The generated TH radiation is collected and filtered out from the pump by blue glass filters. The collimated TH is directed to the cathode of a photomultiplier tube (PMT). The voltage output of the PMT is analyzed with a lock-in amplifier coupled to the optical chopper. The signal is proved to be of TH origin by checking its cubic dependence on the pump power and measuring its spectrum directly. The TH is found to be always polarized along the polarization plane of the pump beam. A white light source and an IR spectrometer are also included into the setup to obtain linear transmission spectra of the trimers without moving the sample.

Bulk silicon possesses high intrinsic third-order nonlinear susceptibility of up to $\chi^{(3)} = 2.5 \cdot 10^{-10}\,\text{esu}$ in the spectral range of interest [7]. To unambiguously disclose wavelength-dependent contributions coming specifically from the nanodisk trimers and their dipolar resonances, for each pump wavelength THG was measured consecutively from the trimers arrays and from the adjacent area where the top silicon layer was etched away. Since the source of both TH signals is bulk silicon, the ratio of these signal values cancels $\chi^{(3)}$ dispersion out. This ratio is referred to as THG enhancement (THGE); it represents the modifications the nanodisks bring to the THG from the unstructured area of the sample.

3. Results and discussions

The linear extinction spectra of the silicon trimers are shown in Fig. 2 with gray curves. Similar to the numerical simulations, each curve is seen to contain two peaks. Note, however, that in the numerical simulations the substrate was not taken into account and hence the theoretical spectra are blue shifted. The first peak of extinction represents the electric dipolar resonance of the trimer and is situated at 1060 nm for all the samples under study. The second one corresponds to the magnetic dipolar resonance, which spectral position varies from sample to sample.

The spectra of THGE in nanodisk trimer arrays are shown in Fig. 2 with blue/red circles. It is seen that, even though the relative area (filling factor) of the nanodisks is only $(3\pi r^2)/p^2 \approx 0.08$, the disks demonstrate enhancement of THG by a factor of up to 2.2 if compared with the bulk 500-μm thick Si substrate. For all the samples under study, THGE is greater than unity in the spectral range of the resonances. Also, each spectrum contains a number of oscillatory-type features.

In each spectrum obtained for the larger disks shown in Fig. 2(b,d), it is possible to highlight two separate areas which correspond to either electric or magnetic dipolar resonances of the trimer. The peaks of the THGE function coincide with the corresponding peaks in the extinction spectrum within sub-20 nm accuracy. This falls within the well-known framework of local field-enhanced nonlinearities: the n-th-order nonlinear polarization of a medium is proportional to the local field correction factor brought to the power of n: $p^{(n)}(\mathbf{r}) \propto L^n(\omega, \mathbf{r})$, where $L(\omega, \mathbf{r})$ is a coefficient connecting the external electric field at the ω frequency and the local electric field observed at the point $\mathbf{r}$ within the nonlinear medium. For a resonant medium, the space-averaged local field factor can be approximated with

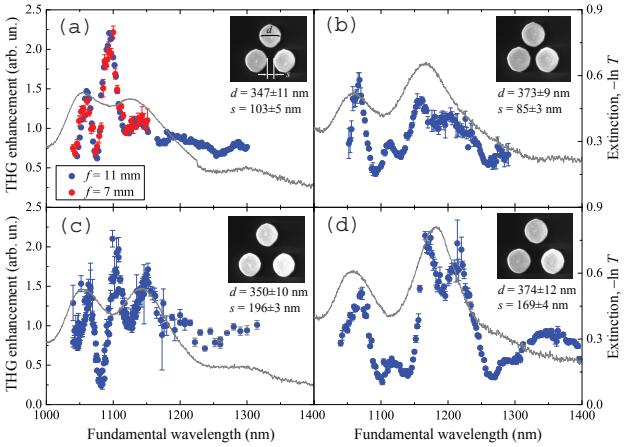

Figure 2: Experimental transmitted THG spectra (blue filled circles) and extinction spectra (grey curves) for the silicon nanodisk trimer samples with the dimensions shown in the corresponding insets. The interparticle distance is calculated as $s = D - d$. Red filled circles denote the spectrum obtained using a lens with $f = 7$ mm as opposed to all the other data obtained with a lens with $f = 11$ mm.

the expression [8]:

$$L(\omega) = A + \frac{B}{\omega_{\text{res}} - \omega + \imath\gamma}, \qquad (1)$$

where γ is the damping factor of the resonance, ω_{res} is the central frequency of the resonance, and A is close to unity for materials that are transparent for the pump radiation. Eq. (1) indicates that the position of the THGE maximum should coincide with the maximum of extinction.

In contrast, if the resonances are brought closer in the frequency domain, as accomplished in Fig. 2(a,c) with smaller disks, the maximal THGE is observed in between the two extinction peaks. Moreover, a three-peak structure is revealed for the THGE. This comes into a contradiction with Eq. (1), since a linear sum of two resonances produces a double peak structure in the spectrum of L^3 with two maxima situated exactly in the sites of the extinction resonances. We have verified experimentally that the observed oscillations are not a result of Fabry-Pérot fringes in the buried SiO_2 layer, which are weak in our experiment and have approximately 1.5 times larger frequency in the spectral domain. Moreover, changing the numerical aperture of the focusing system from 0.045 to 0.07 yielded the same results as depicted in Fig. 2(a) with red circles. This fact indicates minor influence of the effect on the angular dispersion and distinguish it from Fabri-Pérot fringes. We conjecture the two side peaks of the three-peak structure to be affiliated with corresponding extinction resonances, while the one in middle is a result from a collective action of both resonances. While further insight is needed to deeper understand the interference phenomena of the nonlinear scattering problem, it is clear from our experimental studies that by adjusting the resonances position one can achieve a flexible platform of tailoring nonlinear optical response of all-dielectric optical oligomers and possibly all-dielectric metamaterials.

4. Conclusions

In conclusion, third harmonic generation spectroscopy is applied to silicon-based nanodisk oligomers. THG is found to be increased in the spectral ranges that correspond to excitation of the electric and magnetic dipolar resonances of the high-index silicon trimers. Simultaneous excitation of the partially-overlapping electric and magnetic resonances produces strongly enhanced nonlinear signal in the area of spectral overlap , which is connected with constructive nonlinear action of the resonances.

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