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

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Abstract: We characterize experimentally the nonlinear optical response of silicon nanodisk oligomers using third-harmonic generation spectroscopy and reveal the contributions of magnetic and electric dipolar resonances, local field enhancement, and nonlinear interference.

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1. Introduction

Already since the development of Mie scattering theory, it was realized that high-index dielectric particles can exhibit artificial magnetic resonances due to the excitation of circular displacement currents. This fundamental phenomenon was recently explored for silicon nanospheres in the entire visible spectral range [1, 2]. The resonances of three and more dielectric particles have also been recently studied [3], opening new opportunities for all-dielectric nanophotonics and metamaterials. Importantly, multi-particle oligomer structures also exhibit electric and magnetic eigenmodes, which are however not orthogonal and can interfere in the near-field region. Therefore, open questions are how to characterize the nonlinear response enhanced by two resonances and how such near-field interference can be examined. While the linear scattering of the oligomers is not dependent on the particular near-field distribution [3], there is a strong dependence of the nonlinear effects, such as harmonic generation, on the near-field of the particles and their clusters.

However, the optical nonlinearities originated from the magnetoelectric response of all-dielectric nanostructures have never been studied. Furthermore, silicon is a material with a high third-order nonlinearity, therefore strong nonlinear phenomena can be expected in the nonlinear optical response of high-index dielectric nanoparticles. Here we analyze, for the first time to our knowledge, the nonlinear response from silicon disk oligomers (in the form of trimers) in the vicinity of the electric and magnetic resonances. We demonstrate that by engineering the resonant modes of such trimers we can control the nonlinear interference of the magnetic and electric dipoles, resulting in the observation of a strong enhancement of third-harmonic generation (THG) and nonlinear Fano-type resonances.

2. Experimental approach

Silicon nanodisk trimers are fabricated via electron-beam lithography on backside polished silicon-on-insulator (SOI) wafers. Different samples of trimers are fabricated with the size (d) of the individual disks varied. In addition, two sets of samples with different distance (s) between the three disks of the trimer are fabricated. This allows for precise engineering of the mutual position of the electric and magnetic resonances of the trimers.

In our experiments, we use a tunable ($\lambda = 1 - 1.6\,\mu$m) optical parametric oscillator (200 fs) to perform nonlinear THG spectroscopy on different silicon trimer arrays. The laser beam is focused by an aspheric lens to a waist of 20 μm in diameter, resulting in a maximum peak intensity of $I = 2\,\mathrm{GW/cm^2}$. The generated third-harmonic (TH) beam is collimated and directed to a photomultiplier tube. The voltage output of the latter is proved to be of TH origin by checking its cubic dependence on the pump power and measuring THG spectrum directly. The TH radiation 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.

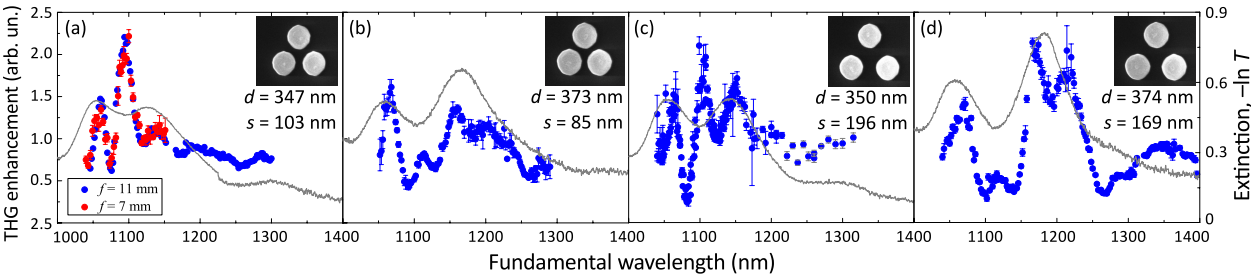

Fig. 1. Experimental transmitted THG enhancement spectra (blue filled circles) and extinction spectra (grey curves) for the silicon nanodisk trimer samples with the dimensions shown in the corresponding insets. Red filled circles denote the THG enhancement spectrum obtained using a lens with $f = 7$ mm as opposed to all the other data obtained with a lens with $f = 11$ mm.

The bulk silicon possesses high intrinsic third-order nonlinear susceptibility of up to $\chi^{(3)} \approx 2.5 \cdot 10^{-10}$ esu in the spectral range of interest [4]. Therefore, a normalization procedure was carried out 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. The ratio of these signal values is referred to as a THG enhancement; it represents the modifications the nanodisks bring to the THG from the unstructured area of the sample. The THG enhancement spectra are measured for all nanodisk trimers in transmission and reflection geometry.

3. Results and discussions

The linear extinction spectra of the silicon trimers are shown in Fig. 1 with gray curves; each curve is seen to contain two peaks. 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 THG enhancement spectra in nanodisk trimer arrays are shown in Fig. 1 with blue/red circles. It is seen that, even though the relative area (filling factor) of the nanodisks is only ≈ 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, THG enhancement is greater than unity in the spectral range of the resonances.

In each spectrum obtained for the larger disks shown in Figs. 1(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 THG enhancement function coincide with the corresponding peaks in the extinction spectrum, indicating strong local field enhancement at the resonances. However, if the resonances are brought closer in frequency, as accomplished in Figs. 1(a,c) with smaller disks, the maximal THG enhancement is observed in between the two extinction peaks. Moreover, a three-peak structure is revealed for the THG enhancement. We conjecture that the two side peaks of the spectral structure are affiliated with the corresponding extinction resonances, while the middle peak is a result from a collective action of both resonances. Our experimental studies evidence that adjusting the resonances position we can achieve a flexible platform of tailoring nonlinear optical response of all-dielectric oligomers and possibly all-dielectric metamaterials.

4. Conclusions

We have applied the THG spectroscopy to characterize the nonlinear response of silicon-based nanodisk oligomers. We have found that the THG response increases in the spectral ranges corresponding to the excitation of the electric and magnetic dipolar resonances of the high-index silicon nanodisks. We have observed that the simultaneous excitation of the overlapping electric and magnetic resonances produces strong enhancement of THG in the area of spectral overlap connected with the constructive nonlinear optical response of the resonances.

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