Third-Harmonic Generation from Silicon Oligomers and Metasurfaces

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Abstract: Third-harmonic generation spectroscopy of silicon oligomers and metasurfaces reveals the nonlinear spectra reshaping with electric and magnetic dipolar Mie-type resonances and up-conversion increase by two orders of magnitude as compared to the bulk silicon.

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1. Introduction

It is known that structuring bulk media on the nanoscale brings new functionalities into the optical [1] and nonlinear-optical [2] response. The boost in the nonlinear-optical processes is defined by the local-field enhancement stemming from geometrical resonances, e.g., surface plasmon resonances in metallic nanostructures [3]. However, using metal nanostructures for high-optical-power applications is not desirable due to relatively low photoinduced thermal damage threshold. In this regard, the recently emerged all-dielectric nanostructures and metasurfaces [4,5] are of special interest since no ohmic losses are inherent to these structures. Their nonlinear-optical properties, however, have only started to be tackled [6] and are yet to be determined. In this work, we demonstrate a set of silicon-based nanostructures—nanodisk oligomers and metasurfaces—that improves the third-harmonic generation (THG) process with Mie-type electric and magnetic dipolar resonances by over two orders of magnitude with respect to the bulk unstructured silicon used as a control. We also observe a considerable reshaping of the THG spectrum by fine tuning the magnetic resonance spectral position resulting in a pronounced interference pattern and additional THG enhancement.

2. Results and Discussion

The etch mask for the metasurface was defined by electron-beam lithography on a silicon-on-insulator wafer with the top Si layer being 220-nm- or 260-nm-thick. The process was followed by reactive-ion etching using the obtained electron-beam resist pattern as a mask. Several nanodisk-based oligomer (trimer) arrays and metasurfaces with different disk diameter values were fabricated. A scanning electron micrograph of one of the metasurface samples with a nanodisk diameter of 500 nm is shown in the inset of Fig.1(a). The transmittance (T) spectroscopy reveals a feature at a wavelength of 1.24 μm shown with $-\ln T$ spectrum in Fig.1(a). This feature is a result of the interference of the two main modes of the nanodisk—the electric and magnetic dipolar resonances [6]. THG spectroscopy was carried out for the silicon metasurface samples. In order to take the dispersion of the silicon nonlinear susceptibility into account, the THG signal from the sample area was normalized by that from the area of the wafer where the top silicon layer was etched away. The resultant normalized THG is plotted as a function of the pump wavelength in Fig.1(a) with purple dots. The maximum enhancement of about 100 is seen in the vicinity of the transmittance spectrum feature associated with the resonant polarization and magnetization. At the pump wavelength of 1.26 μm the THG radiation could be seen with naked eye under standard laboratory illumination conditions.

Packing the nanodisks into the trimer arrangement provides more versatility in tuning the nonlinear optical response, as shown in Fig.1(c-e). Careful selection of the disk diameter and interdisk spacing makes it possible to finely tune the position of the low-energy (magnetic dipolar) resonance while keeping the high-energy (electric dipolar) resonance at a fixed position. Doing so brings considerable changes in the THG spectra about. Specifically, when the resonances

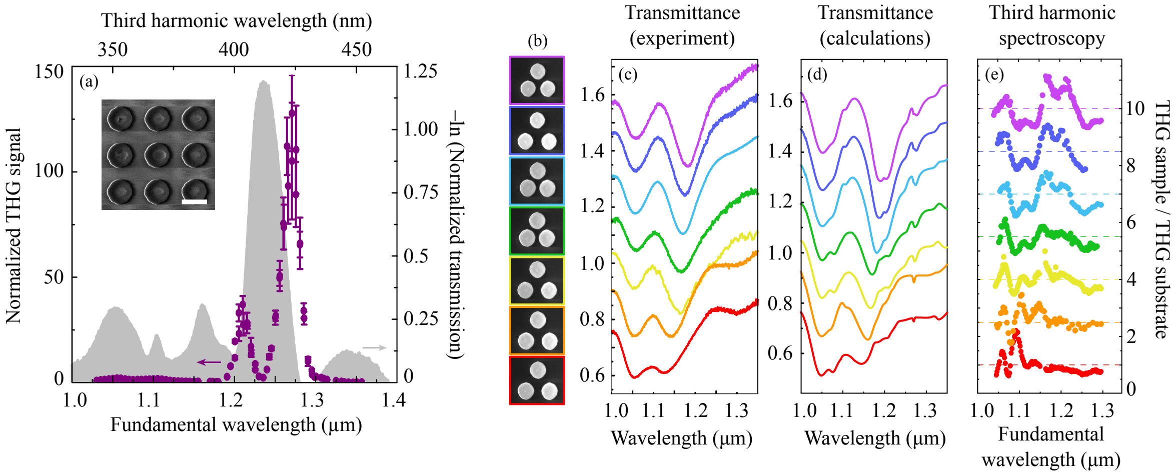

Fig. 1. (a) Third-harmonic generation spectrum of the metasurface (purple dots) revealing 100-fold enhancement of the signal as compared to the bulk silicon slab. The experimental transmittance spectrum is given with the gray area. The inset shows a scanning electron microscope (SEM) image of the sample; the scale bar is 500 nm. (b) SEM micrographs of the silicon nanodisk trimers under study. The diameter of the upmost disks is 350 nm. (c) Experimental and (d) calculated transmittance spectra of the corresponding trimers showing the electric dipolar resonance fixed at 1.05 μm and the magnetic dipolar resonance shifting from 1.13 μm to 1.18 μm. The bottommost spectrum is to scale, others are shifted for better readability. (e) Third-harmonic generation spectra of the corresponding trimers. The bottommost spectrum is to scale, others shifted for better readability.

are brought to a partial overlap, additional enhancement of the THG is observed in between the resonances, as shown with red dots in Fig.1(e).

3. Conclusions

In conclusion, we demonstrate a flexible platform for enhanced third-order nonlinearities based on silicon nanodisk oligomers and metasurfaces. The third-harmonic generation data reveals the increase of the nonlinear signal by two orders of magnitude as compared to the bulk silicon and reshaping of the nonlinear spectra by exciting electric and magnetic resonances. Supported by our studies, we can positively conclude that silicon nanodisk-based structures with Mie-type resonances are prospective new objects for observing enhanced nonlinear-optical response from nanosystems, which may find applications in silicon photonics and novel optical telecom devices.

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