Cascaded Second Order Optical Nonlinearities in a Dielectric Metasurface

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Abstract: In this work, we analyze the second and third harmonic signal from a dielectric metasurface in conjunction with polarization selection rules to unambiguously demonstrate the occurrence of cascaded second-order nonlinearities.

1. Introduction

In recent years, the rapid development in artificially structured materials such as nanoscale resonators arrayed as metasurfaces has led to achieve optical frequency mixing at increasingly smaller scales. [1] An outstanding question though has been the microscopic nature of the higher order nonlinear processes observed from these nonlinear metasurfaces. While traditional thick, transparent crystals require a gradual, phase-matched accumulation of the converted light over macroscopic distances (compared to the wavelength of light), which unavoidably limit harmonic generation to one process at a time, light conversion in metasurfaces occurs within lengths comparable to the wavelength of the light causing all nonlinear processes to occur simultaneously. [2,3] This could promote harmonic generation via a cascade of several frequency mixing steps. Given the high second order nonlinearity of III-V semiconductors and the lack of phase matching in nonlinear metasurfaces, several of the frequency mixing products observed experimentally could thus be explained by cascaded second order nonlinearities. For example, harmonic generation at three times the pump frequency, ω, could be achieved by traditional third-harmonic generation ($\chi^{(3)}: THG$) from a χ^3 direct process or via a cascaded process of second harmonic generation (SHG) and sum frequency generation (SFG), that we will refer as $\chi^{(2)}:\chi^{(2)}(3\omega)$. [4,5] Identifying the origin of light conversion in these systems is essential as cascaded harmonic generation could compete for efficiencies with conventional direct frequency mixing processes or even surpass them.

2. Results and Discussion

In this work, we analyze the harmonic signals at 2ω and 3ω measured from a highly nonlinear metasurface in conjunction with polarization selection rules, symmetry considerations and self-consistent modeling to unambiguously demonstrate the occurrence of cascaded second-order nonlinearities. The metasurface is made of arrays of subwavelength GaAs nanocylinders, isolated from a GaAs/AlGaAs substrate by a AlGaO spacer layer as shown in figure 1a. We choose this design as the metasurface is known to exhibit intense harmonic signals at multiples of the pump frequency enhanced by the excitation of Mie-like optical resonant modes at the fundamental (i.e., pump) frequency (see figure 1b). The linear reflectance spectrum shows a broad resonance with two distinct peaks characteristic of two Mie-like optical modes, namely an electric dipole mode centered around 1200nm and a magnetic dipole mode centered around 1400nm.

To demonstrate the presence of $\chi^{(2)}:\chi^{(2)}(3\omega)$, we need to find ways of distinguishing third harmonic photons between $\chi^{(2)}:\chi^{(2)}(3\omega)$ and $\chi^{(3)}$:THG. Figure 1c shows measured (in reflection) and simulated normalized intensity of the harmonic signal at 2ω and 3ω as a function of incident polarization when optically pumping the magnetic dipole mode at 1400nm. Since GaAs crystal belongs to the point-group symmetry $\bar{4}3$m, traditional $\chi^{(3)}$:THG exhibit a fourfold symmetry with respect to crystal orientation as commonly observed for cubic crystals. [6,7] This result is confirmed by our nonlinear optical modeling with a commercial finite-element solver as shown in figure 1d, middle panel. However, in our experiment (fig. 1d, top panel), we find that our third wave mixing signal exhibit a twofold symmetry which cannot be explained by $\chi^{(3)}$:THG alone. To explain this, we include $\chi^{(2)}:\chi^{(2)}(3\omega)$ in our model and consider its interference with χ^3:THG (fig 1d, bottom panel). We can retrieve the twofold symmetry observed in our experiment demonstrating the occurrence of cascaded second order nonlinearities in our metasurface.

To correctly model $\chi^{(2)}:\chi^{(2)}(3\omega)$, we need to consider the polarization symmetry rule of SHG since $\chi^{(2)}:\chi^{(2)}(3\omega)$ crucially depends on it. In our experiment, we also find that measured SHG has a twofold symmetry with respect to incident polarization (Fig 1c, top panel). However, simulated SHG considering second order optical nonlinearities of bulk GaAs only predicts a four-fold symmetry (Fig 1c, middle panel). This discrepancy can be solved by including surface nonlinearities of GaAs (fig1c, bottom panel), as it possesses a different crystal symmetry (mm2) to that of the bulk. Our SHG result reaffirms the importance of surface-induced nonlinearities in these types of nanophotonic systems. Using this self-consistent modeling that is representative of the physics of our metasurface, we deduct the strength of each harmonic process and find that $\chi^{(2)}:\chi^{(2)}\,(3\omega)$ can be of comparable strength to $\chi^{(3)}:THG$. Moreover, it seems to be primarily generated via surfaces second order nonlinearities.

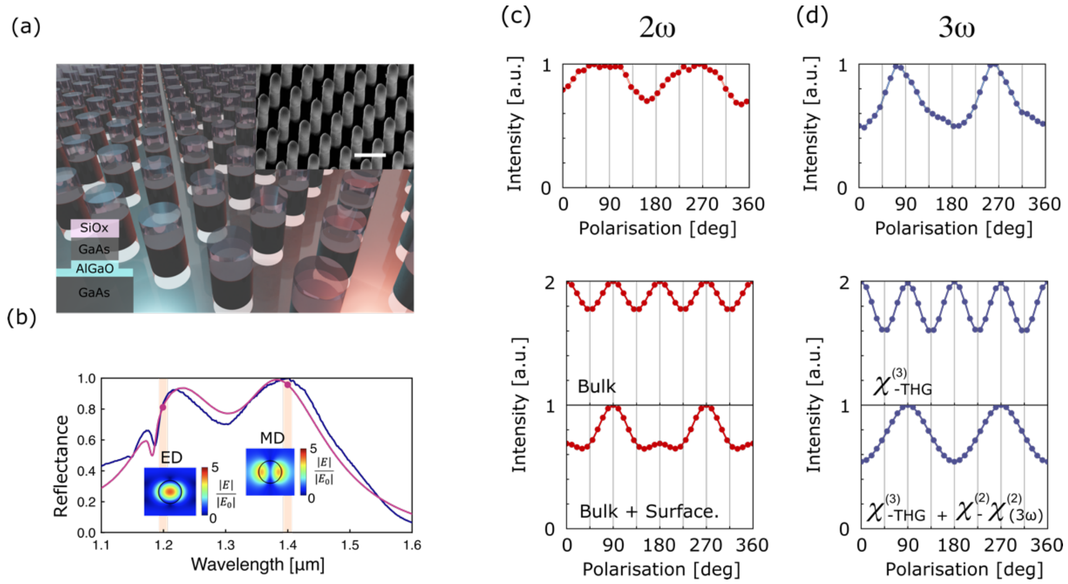

Fig 1. (a) Artistic representation of the dielectric metasurface considered in this work Top inset is a tilted view of a scanning electron micrograph (SEM) of the metasurface. White scale bar represents 1µm. Bottom inset shows the different materials that constitutes each resonator. (b) Simulated (solid purple) and measured (solid blue) linear reflection spectra of our metasurface. Top views of normalized electric field distribution of the resonators are displayed for the electric dipole mode at 1200nm and the magnetic dipole mode at 1400nm. (c, d) Measured (top panel) and simulated (middle and bottom panels) nonlinear harmonic intensities at (c) 2ω and (d), 3ω and for the magnetic dipole mode excitation ($\lambda = 1400\ nm$) while varying the polarization of the incident beam from 0 to 360 degrees. Fourfold symmetry is characteristic of SHG from bulk nonlinearities, and of third wave mixing via a direct third order process: $\chi^{(3)}$: THG. Twofold symmetry indicates the presence of SHG from surface nonlinearities at 2ω, and of third wave mixing at 3 ω via a cascaded second order process: $\chi^{(2)}:\chi^{(2)}(3\omega)$.

The demonstration of cascaded optical nonlinearities in our dielectric metasurface suggests that alternative pathway to third wave mixing, and more broadly high harmonic generation, is achievable at the nanoscale. This is facilitated by the relaxation of phase-matching. Such understanding and analysis could thus be used to significant enhance efficiencies for frequency-mixing processes as well as their quantum equivalent such as spontaneous parametric down-conversion.

3. References

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