Ge$_2$Sb$_2$Te$_5$-based nanocavity metasurface for enhancement of third harmonic generation

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Abstract

The third-order nonlinear processes in nanophotonic devices may have great potentials for developing ultra-compact nonlinear optical sources, ultrafast optical switches and modulators, etc. It is known that the performance of the nonlinear nanophotonic devices strongly relies on the optical resonances and the selection of appropriate nonlinear materials. Here, we demonstrate that the third harmonic generations (THG) can be greatly enhanced at subwavelength scale by incorporating $\alpha$-Ge$_2$Sb$_2$Te$_5$ ($\alpha$-GST) into the nanocavity metasurface. Under pumping of a near-infrared femtosecond laser, the THG from the nanocavity metasurface is $\sim$50 times stronger than that from the bare GST planar film. In addition, the nanocavity metasurface also provides a powerful platform for characterizing the third-order nonlinear susceptibility of the active medium in the cavity. We expect that the GST-based nanocavity metasurface could open new routes for achieving high efficiency nonlinear nanophotonic devices.

1. Introduction

Harmonic generation in nonlinear optical materials, such as second and third harmonic generations (SHG and THG), represent an important route for extending the spectral range of a laser source [1]. While the bulky optical crystals have been widely used in the commercial laser system, subwavelength thick optical films usually exhibit week nonlinear optical responses which limit their practical applications [2, 3]. Given the fact nonlinear optical sources at subwavelength scale may have important applications in on-chip optical switching, optical computing and so on, there is strong requirement for developing high efficiency nonlinear nanophotonic devices. Among various strategies, incorporating nonlinear materials into the artificial structures such as optical superlattice [4], photonic crystals [5, 6], and metamaterials [7, 8] have been proven to be a useful way to achieve high efficiency nonlinear nanophotonic devices. For example, optical superlattice has been used to improve the efficiency of SHG [4], THG [9], and entangled photons [10]. By engineering the photonic band structures, enhancement of THG from photonic crystals was demonstrated [11–13]. In addition, metamaterials which consist of subwavelength meta-atoms in three dimensions were proposed to enhance the SHG [14], and four-wave mixing processes [15, 16]. Note that, most of the previous artificial structures, including the photonic crystals and metamaterials, are bulky and thus may affect their practical applications in integrated photonics. This constraint could be partially addressed by the newly developed photonic metasurfaces, which consist of two dimensional spatially variant plasmonic or dielectric meta-atoms [17]. In the last decade, there have been many achievements in
manipulating light–matter interaction with metasurfaces [17–21]. By locally engineering the properties of meta-atoms, efficient control of the phase, polarization and amplitude of light have been successfully realized in the linear optical regime. For example, the metasurface-based multifunctional devices such as vortex waveplates [17], high efficiency holograms [22], ultrathin metalenses [23], and metasurfaces for quantum information technology [24–27] have been developed.

The concept of linear optical metamaterial has been successfully applied to the nonlinear optical regime [28, 29]. Unlike in the bulky materials [1–3], such as the conventional optical crystals, photonic crystals and metamaterials, the phase matching requirement for nonlinear optical processes on the metasurfaces is relaxed due to the short light–matter interaction length. By designing the plasmonic meta-atoms with inversion symmetry breaking properties, enhanced SHG from split ring resonators [30–32], multi-resonant meta-atoms [33], metal–quantum well hybrid system [34], semiconductor meta-atoms [35] and plasmonic meta-atoms [36, 37] were experimentally verified. In addition, by applying the concept of geometric phase, also called Pancharatnam–Berry phase [38, 39], to nonlinear metasurfaces, we can locally control the polarization, phase of nonlinear waves with unprecedented abilities [37]. For example, nonlinear metasurface holograms [40], nonlinear metasurfaces [41] based on SHG process were successfully demonstrated. In the meantime, the manipulation of THG waves has been demonstrated on metal–dielectric hybrid metasurfaces [42, 43], silicon metasurfaces [44–46] and so on. Apart from the consideration of symmetry and material constituents of the meta-atoms, various resonant mechanisms such as magnetic resonance [44], Fano resonance [45] and bound state in the continuum [46] were utilized to improve the nonlinear optical efficiency of dielectric metasurfaces. Despite the rapid progress of nonlinear metasurfaces in recent years [29], novel designs and high performance nonlinear materials still need to be proposed to meet the requirements for practical applications [47].

In this work, we propose a new scheme to improve the THG efficiency of a metasurface by taking into account of both the selection of active materials and the resonant mechanisms. To this end, we incorporate the α–GST material, which has high third-order nonlinear susceptibility [48], into a metal–dielectric–metal nanocavity. The nanocavity metasurface consists of the gold nanodisk/SiO2/α–GST/SiO2/gold reflector multilayer. The two SiO2 layers act as barrier layers to prevent interlayer diffusion and interfacial reaction of the materials of α–GST and gold. In the Fabry–Perot nanocavity, the gold nanodisk and the gold reflector act as the two mirrors, and the SiO2/α–GST/SiO2 tri-layer represents the spacer. The localized plasmonic resonance of the gold nanodisk together with the Fabry–Perot nanocavity formed by the two metallic layer are expected to strongly confine the energy of fundamental wave (FW) inside the α–GST and gold layers. As shown in figure 1(a), for a normally incident linearly polarized FW with frequency ω, the THG wave at frequency of 3ω from the nanocavity metasurface can be detected in the back-reflection direction. Figure 1(b) shows the schematic diagram of a unit cell of the nanocavity metasurface. The gold circular nanodisks are arranged in a square lattice with the period of P.

2. Design, fabrication and characterization of the nanocavity metasurface

We firstly characterize the complex refractive index and the thickness of the α–GST film which was deposited on a cleaned crystalline silicon substrate by using RF sputtering method. As shown in figure 2(a),
we measured the wavelength dependent refractive index from the visible to near infrared regime by using the spectroscopic ellipsometer. To match the spectral range of our nonlinear optical measurement system, we optimized the geometrical parameters of the nanocavity metasurface to obtain a cavity resonance mode located in the near infrared regime. The period of the square lattice is $P = 300$ nm along both $x$- and $y$-axes, the radius of the gold nanodisk is $R = 84$ nm, and the thickness of the gold nano-disk is 30 nm. The thicknesses of the SiO$_2$, $\alpha$-GST and gold reflector are 3 nm, 24 nm and 100 nm, respectively.

In figure 2(b), the reflection spectra of the nanocavity metasurface and the planar multilayer (SiO$_2/\alpha$-GST/SiO$_2$/gold reflector) were calculated by using the commercial software COMSOL Multiphysics. For a linearly polarized wave, the plasmonic nanocavity metasurface exhibits an intense resonance at the wavelength of $\lambda \sim 1380$ nm, which corresponds to the Fabry–Perot cavity mode. In comparison, the planar multilayer is highly reflective for $\lambda > 1100$ nm. To verify the simulated results, we fabricated the planar multilayer by using e-beam and RF sputtering deposition techniques. The gold nanodisk array was subsequently fabricated on the planar multilayer by using the electron beam lithography and lift-off process. Figure 2(c) presents the scanning electron microscope image of the fabricated gold plasmonic metasurface. The measured reflection spectra of the nanocavity metasurface and the planar multilayer are shown in figure 2(d). As was observed, the calculated reflection spectra agree well with the measured ones, while the discrepancy may come from the geometrical deviations between the fabricated and designed devices.

### 3. Nonlinear optical measurement

The THG responses of the nanocavity metasurface were experimentally characterized by using a femtosecond laser based optical setup (figure S1). Under the pumping of linearly polarized FWs with the wavelength ranging from 1280 nm to 1560 nm, the linearly polarized THG signals emitted from the metasurface were measured. In the measurements, the energy loss of the incident FW and the generated THG waves when passing through or reflected by the optical elements were taken into account. As shown in figure 3(a), for a horizontally (H, along $x$-axis) polarized FW, the wavelength dependent THG responses...
Figure 3. Characterization of the THG responses on the α-GST based nanocavity metasurface. (a) The measured wavelength dependent THG responses of the nanocavity metasurface. ‘HFW – HTHG’ and ‘HFW – VTHG’ represent the polarization states of the incident FW and the measured THG waves (H: horizontal polarization, V: vertical polarization). (b) The measured H- and V-polarized THG spectra for the H-polarized FW at λ = 1402 nm. (c) The circle symbols represent the measured power of the THG signals under different pumping power of the FW at λ = 1402 nm. The fitted slope value (red line) is 2.903. (d) The measured wavelength dependent THG response of the planar multilayer.

with H and vertical (V, along y-axis) polarizations were obtained. It is also found that the intensity of H-polarized THG signals are much stronger than the V-polarized ones. The strongest H-polarized THG responses is observed at the fundamental wavelength of ~1392 nm, which is close to the resonant wavelength at 1378 nm in figure 2(d). At the stable wavelength of 1402 nm of the femtosecond laser, the typical THG spectra with H and V polarizations were measured. As shown in figure 3(b), the polarization of the THG signal is mainly along horizontal direction. In figure 3(c), we investigate the effect of power of the H-polarized FW on the efficiency of the H-polarized THG waves. The power dependent curve has a slope value of 2.903, which is close to the theoretical value of 3.0, indicating the third-order nonlinear optical processes. In figure 3(d), we also measured the wavelength dependent THG responses from the planar multilayer, which has a configuration of SiO2/α-GST/SiO2/gold reflector. It can be found that the THG signal generated from the multilayer is at almost the noise level. Thus, by using a design of nanocavity metasurface, the THG signal is ~50 times stronger than that from the planar multilayer.

To compare the THG contributions from each constituent materials that form the nanocavity metasurface, a control experiment was then conducted. As shown in figure S2, we designed and fabricated a SiO2 based nanocavity metasurface, which is composed of gold nanodisks/SiO2/gold reflector multilayer. Similarly, the SiO2 based nanocavity metasurface shows a measured cavity resonance at the wavelength of ~1415 nm. From the results in figures S2(b) and (d), it is found that the measured reflection spectra agree well with the simulated ones. We also characterized the THG responses of the SiO2 based nanocavity metasurface by using the same pumping conditions as that for the α-GST based nanocavity metasurface (figure S3). At fundamental wavelength of 1402 nm, the THG signal from the α-GST based metasurface is more than 10 times stronger than that of the SiO2 based metasurface (figures 3(a) and S3(a)).

4. Nonlinear optical calculation

To understand the THG enhancement mechanism of the nanocavity metasurface, a theoretical model based on the COMSOL Multiphysics and the self-developed codes was built. The nonlinear optical responses of the metasurface is obtained through the three-step calculations. Firstly, the spatially variant nonlinear
polarization is described by \( \mathbf{P}_3 = \varepsilon_0 \chi^{(3)} \mathbf{E}^3 \), where \( \mathbf{E} \) is the electric field of the FW, \( \chi^{(3)} \) is the third-order nonlinear susceptibility of the materials. Secondly, the nonlinear polarization in the near field is used as the nonlinear source in the COMSOL Multiphysics model to radiate THG waves. Lastly, the wavelength dependent far field THG waves is detected by the monitor in the COMSOL Multiphysics model. From the weak nonlinear responses in the SiO2 based nanocavity metasurface, the third-order susceptibility \( \chi^{(3)} \) of SiO2 is neglected in all the calculations. By comparing the measured THG responses with the calculated ones, we obtain the wavelength dependent third-order susceptibility \( \chi^{(3)} \) of gold, which is comparable with the experimentally reported values [49, 50].

Finally, we study the THG responses from \( \alpha \)-GST based nanocavity metasurface. In the simulation, a horizontally polarized (H, along x-axis) FW is normally incident onto the nanocavity metasurface. At the cavity resonant wavelength of 1382.6 nm, the total electric field distributions in the x–z plane are plotted in figure 4(a). Figures 4(b) and (c) show the electric field distributions (Ex and Ey) at the interface (x–y plane) between the gold nanodisk and the SiO2 passivation layer. It can be found that the electric field is strongly localized at the edge of the gold nanodisk, and the Ex component is much stronger than Ey. This explains why the measured H-polarized THG signal in figure 3(b) is dominant. Based on the measured \( \chi^{(3)} \) of gold in figure S4, the wavelength dependent third-order susceptibility of GST film can be extracted from the measured THG responses in the \( \alpha \)-GST based nanocavity. The calculated \( \chi^{(3)} \) of \( \alpha \)-GST is shown in figure 4(d), which is close to the values in reference [51]. It should be noted that although third-order susceptibility of \( \alpha \)-GST is lower than that of gold, the THG contribution from GST is much stronger (as shown in figure S5), this is mainly because the effective mode volume in gold is much smaller [52].

5. Conclusion

In summary, we have proposed a novel way to boost the efficiency of THG at subwavelength scale by utilizing the \( \alpha \)-GST based nanocavity metasurface. Compared to the planar multilayer, the THG efficiency from the \( \alpha \)-GST based nanocavity metasurface enhanced by up to 50 times. By combining the theoretical

Figure 4. Simulated distribution of the total electric field for H-polarized FW at the wavelength of 1382.6 nm. (a) The total electric field distribution in x–z plane (y = 0). (b) and (c) The Ex and Ey distributions at the interface (x–y plane) of gold nanodisk and SiO2 film. (d) The third-order susceptibility of \( \alpha \)-GST film extracted from the measured THG responses.
model with the nonlinear optical measurements, we are able to extract the wavelength dependent third-order susceptibilities of gold and GST layers. The methodology proposed in this work can be also utilized to enhance other nonlinear optical processes. We expect that the nanocavity metasurface represents a promising platform for developing ultra-compact and highly efficient nonlinear optical sources at the nanoscale.

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Data availability statement

The data generated and/or analysed during the current study are not publicly available for legal/ethical reasons but are available from the corresponding author on reasonable request.

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