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Optothermally controllable multiple high-order harmonics generation by $Ge_2Sb_2Te_5$-mediated Fano clusters

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ABSTRACT

Substantial enhancement of nonlinear high-order harmonics generation based on Fano-resonant nanostructures has received growing interest due to their promising potential for developing integrated and advanced next-generation nanophotonic devices. In this study, going beyond conventional subwavelength structures, we propose an optothermally functional hetero-metallodielectric asymmetric eight-member octamer cluster composed of a central silicon nanodisk and peripheral disks with a phase-change material ($Ge_2Sb_2Te_5$). Using full electromagnetic calculations, we show that in the amorphous phase of the surrounding nanoparticles, the oligomer acts as an all-dielectric cluster, while in the crystalline regime, the octamer turns into a hybrid metallodielectric assembly. Exploiting the exquisite ability of supporting distinct Fano lineshapes at different wavelengths depending on the phase of $Ge_2Sb_2Te_5$, we showed the generation of both second and third harmonics at amorphous and crystalline phases of GST nanodisks, respectively with the produced harmonic wavelengths of 425 nm and 317 nm, respectively. Our calculations for the corresponding conversion efficiencies revealed significant enhancements as $\eta_{SHG} = 0.0081\%$ and $\eta_{THG} = 0.012\%$ for SHG and THG, respectively. Such an exquisite feature of multiresonant optothermally tunable cluster allows generation of several harmonics with substantial intensities using a single system for future photonics applications.

1. Introduction

High-harmonic generation (HHG) is a conventional phenomenon, has been observed initially in gas atoms (i.e. Ar, Kr, and Xe) due to multiple multiphoton ionization, resulting from the absorption of large number of incoming photons *via* nonlinear processes [1–3]. By producing wide variety of wavelengths from a single optical source, nonlinear HHG provides broad range of applications in modern nanophotonics technology and attosecond optics [3–10]. So far, several strategies have been developed to convert the fundamental incident beam frequency into the intense higher-order harmonics by subwavelength bulk solids with large field-induced of susceptibilities ($\chi^{(2)}$ or $\chi^{(3)}$) including but not limited to plasmonic antennas and waveguides [11–15], all-dielectric structures [16–21], photonic crystals [22,23], ring resonators [24–26], two-dimensional (2D) and chiral materials [27–31], optoelectronic platforms [32,33], etc. On the other hand, very recently, intense and efficient ($P_{2\omega,3\omega}/P_{\omega}^2$) nonlinear spectral behavior of higher-harmonics have been reported in both Fano-resonant fully metallic, all-dielectric, and metallodielectric nanoparticle clusters with either simple or complex geometries, taking the advantage of dark side of plasmons [15,17,19]. The loss-less nature of dark-side of plasmons helps to produce substantially strong and efficient fundamental resonances and relatively enhanced high-order harmonics and other pronounced spectral features in either molecular and atomic scales [15,19,34–36]. It is well-accepted that efficient scattering of generated harmonics signal into the far-field radiation can be realized by breaking the centrosymmetry of nanoparticle assemblies, resulting in strong electromagnetic field localization and formation of hotspots in nanosystems [37,38]. Although all of the elucidated mechanisms provide significant intensities and high efficiencies for generation of both odd and even harmonics, they suffer from limited tunability and high nonradiative and scattering losses [39,40]. It is shown that introducing low-loss and electrically controllable atomically-thin graphene layer to the subwavelength systems result in formation of higher-harmonics with enhanced tunability *via* tuning the doping concentration of graphene sheet [41–44]. However, manipulation and integration of atomically-thin layers with bulk systems require complex nanofabrication techniques, costly processes and also suffer from limited mechanical flexibilities [38]. Moreover, in these systems, due to the weak field enhancement at the fundamental frequency, the near-field intensity in the

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higher-harmonics cannot be significantly boosted [45]. Consequently, finding an approach to control the nonlinear spectral response with high-harmonic intensity and functionality allows for developing advanced, tunable, and integrated nanophotonic devices. This can be realized by using optically controllable materials in the geometry of the artificially engineered structure like thermally controllable phase-change materials (PCMs) [46,47], and semiconductors (e.g. InSb) [48,49]. The latter option needs for external heating/cooling system to tune the corresponding intrinsic carrier density, which is limiting its efficacy in ultracompact photonic systems. On the other hand, as new and exotic members of optical materials, optothermally controllable PCMs based on chalcogenide compounds offer novel and promising methods to address the inherent lack of tunability in conventional bulk solids [43]. PCMs have a wide range of applications in space related technologies due to having inherent radiation-hard feature. Such exquisite feature led to developing of several astronomical devices such as 4-Mbit tool etc. [50] On the other hand, recent progresses in advanced nanophotonics technology have witnessed broad utilization of PCMs in designing optical devices such as antennas [51,52], rewritable data storages [53], modulators [47,54–56], and beam steering metamaterials [57,58]. Possessing significantly different dielectric functions at two different phases (amorphous and crystalline states) of PCMs (e.g. VO_2 [46,59,60], AgInSbTe [61], $Ge_xSb_yTe_z$ [47,55–57]) at room-temperature, enabled emerging of novel optical devices with exotic properties. This exquisite feature of PCMs becomes more interesting, when we analyze the switching timescale between opposite phases and their rapid reversibility, which is around few tenth of nanoseconds [62]. This interplay between phases can be realized by applying heat, optical or electrical pulses as external thermal stimuli [46].

In this study, we show generation of multiple higher-harmonics using an eight-member asymmetric Fano-resonant hetero-metallodielectric octamer assembly composed of central dielectric (silicon) and peripheral PCM ($Ge_2Sb_2Te_5$ or GST) nanodisks. Using the opposite behavior of GST nanodisks at below and above its critical temperature (~477 °C) [63], we showed that, due to having inherent asymmetric geometry in octamer [64], the proposed nanoassembly can be effectively tailored to support distinguished Fano dips at different wavelengths depending on the phase of the GST particles. By adjusting the fundamental wavelength of Fano dips in two different states of GST, we efficiently generated both second- (SHG) and third harmonics (THG) at different temperatures with high intensity. Our full electromagnetic analyses showed that in the amorphous regime of GST (a-GST), the structure acts as a full dielectric octamer cluster and supports a Fano minimum around $\lambda \sim 875$ nm. In contrast, when the GST state switches to the crystalline phase (c-GST), the satellite nanoparticles act similar to the metallic components. This results in the formation of a hybrid metallodielectric cluster, enabled to sustain Fano mode at $\lambda \sim 1050$ nm. This functionality is exploited for developing a platform to generate higher-harmonics according to the phase of the PCM and the fundamental wavelength of Fano lineshapes.

1.1. Excitation of Fano resonances

Fig. 1a demonstrates the scattering spectra for the proposed hetero-metallodielectric octamer cluster under intense plane wave radiation for both phases of GST nanoparticles and also contains a schematic for the asymmetric compositional cluster as an inset. The peripheral nanoparticles are GST compound and the central one is silicon, deposited on a glass (SiO_2) substrate with the relative permittivity of $\varepsilon \sim 2.1$. The corresponding dimensions of the optimized octamer are set as follows: the diameter of central and surrounding disks are 190 nm and 128 nm, respectively, with the homogenous height of 60 nm, and gap distance of 20 nm between neighboring nanoparticles. Here, we used experimental permittivity values reported by Palik [65], and Shportko et al. [66] for silicon and GST, respectively. Specifically, for the effective permittivity of GST at crystallization level, we employed the effective-medium

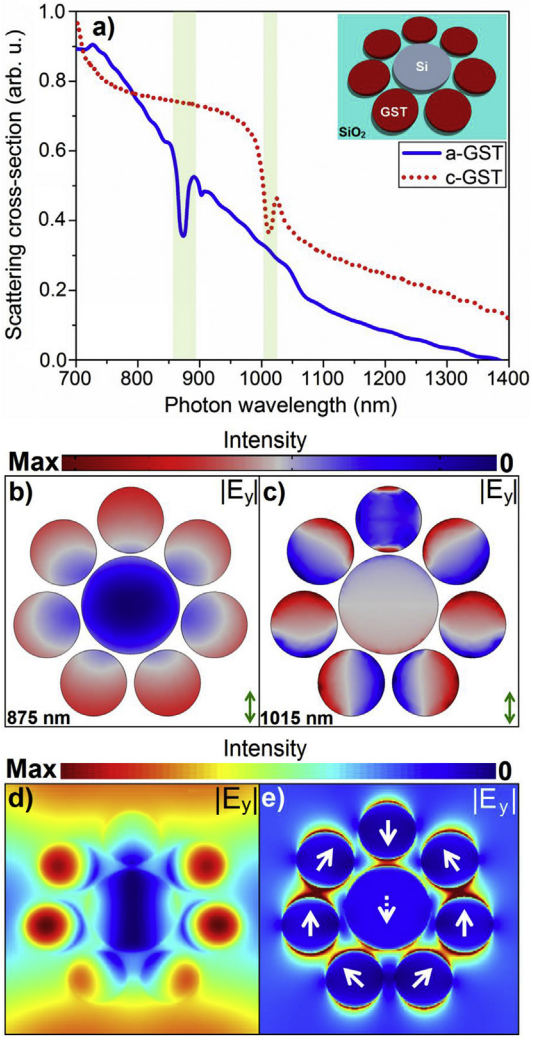

Fig. 1. (a) Scattering cross-section spectrum of the hetero-metallodielectric octamer cluster in both a-GST and c-GST phases of the surrounding nanoparticles. The inset is the schematic representation of the octamer assembly. **(b), (c)** The $|E_y|$-field maps for resonant modes excitation at the fundamental wavelength of the Fano modes for a-GST and c-GST states of cluster, respectively, obtained by FEM analysis. **(d), (e)** The $|E_y|$-field maps for resonant modes excitation and localization at the fundamental wavelength of the Fano modes for a-GST and c-GST states of cluster, respectively, obtained by FDTD analysis.

expression based on Lorentz-Lorenz theory [55,56,67,68].

Focusing on the spectral response of the octamer, we first analyze the optical properties of the cluster consists of satellite a-GST nanodisks. In this regime, the surrounding nanodisks have dielectric properties and the entire cluster can be considered as an all-dielectric nanoassembly. Employing discrete dipole approximation (DDA) [69], previous studies have shown that both symmetric and asymmetric all-dielectric nanoparticle oligomers can be tailored to support pronounced Fano resonances across the visible to the near-infrared region (Vis-NIR), originating from the destructive interference of the resonant and non-resonant modes excited in central and peripheral nanoparticles, respectively [70–72]. In the current structure, in the all-dielectric limit and under the linear polarization beam illumination, the Fano dip is induced at $\lambda \sim 875$ nm, which is recognized as a fundamental resonance frequency for the all-dielectric oligomer. Fig. 1b illustrates the *xy*-plane electric-field (E_y) plot for the electric dipole mode excitation and totally opposite resonant behavior and resonance mismatch between the central and satellite nanodisks, consistent with the DDA mechanism.