Active Control over the Interplay between the Dark and Hidden Sides of Plasmonics Using Metallodielectric Au–Ge$_2$Sb$_2$Te$_5$ Unit Cells

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ABSTRACT: We present a systematic study for a metallodielectric nanoplasmonic unit cell to support both Fano and toroidal resonances in the near-infrared region. Using full electromagnetic computations, we show that a combination of phase-changing and metallic materials allows for designing a unit cell with different spectral responses at different phases of the phase-changing material (here Ge$_2$Sb$_2$Te$_5$ or simply GST). This phenomenon of switching between toroidal response and Fano resonant mode is confirmed using both finite-element and finite-difference time-domain methods. Having active control over the interplay between narrow and tunable Fano and toroidal modes at different wavelengths with fast transition would allow for developing efficient and practical photonic devices for communication and imaging applications.

INTRODUCTION

The interaction of electromagnetic (EM) radiation with matter in the subwavelength regime has led to significant progress in technological development of advanced integrated nanophotonic devices in the past two decades. Such an interaction conventionally results in the excitation of dominant electric ($\vec{p}$) and magnetic ($\vec{m}$) dipolar and multipolar momenta with strong far-field fingerprints in a wide range of spectra. The type of interference of the optically excited modes determines the nature of the induced resonances in a given nanosystem. For both destructive and constructive interactions, geometry of nanoantenna plays a fundamental role in the type and nature of the excited modes. In the retarded limit, the excitation of a sharp asymmetric Fano resonance is the unique and direct result of an interference between spectrally narrow dark and broad bright modes via a destructive interaction. Taking advantage of pronounced Fano dips with high-quality factor (high-Q) in both all-dielectric and metallic nanostructures, several all-optical and optoelectronic devices have been developed including but not limited to biochemical sensors, modulators, photodetectors, transistors, molecular spectroscopic devices, nonlinear optics, colorimetric sensors, photothermal heat generation, and flexible filters. At the Fano minimum position, the dipolar bright mode is suppressed, and the absorption cross-section enhances substantially; therefore, the radiating beam weakly couples to the free space. This results in significant damping in both radiative and nonradiative losses with the dramatic reduction of absorption losses.

In the recent years, a third family of nonradiative resonant momenta known as toroidal moments ($\vec{T}$) has been increasingly studied. Toroidal moments have dramatically weak far-field optical signatures, which are masked by classical EM multipolar far-field radiation. In terms of condensed matters and nuclear physics the toroidization concept operates based on both time-reversal ($t \rightarrow -t$) and space inversion ($r \rightarrow -r$) symmetry. In the toroidal multipole family, the dipolar toroidal mode ($\vec{T} = \vec{p} \times \vec{m} = \sum q_j \vec{r}_j \times (\vec{r}_j \times d\vec{r}_j/dt)$) has received growing interest to employ for nano- and microscale devices (i.e., metamaterials, switches, and sensors). Toroidal dipolar response can be visualized as a vortex of closed-loop circular magnetic current flowing on the surface of a torus. Such a hybrid behavior of toroidal momentum allows for substantial and direct influence of electric polarization by the magnetic field and vice versa. Recent studies have reported the formation and observation of pronounced toroidal modes in the microwave to the terahertz domain in both planar and 3D artificial unit cells due to the weak far-field effect of...
Conventional multipoles. Besides, dipolar and multipolar toroidal resonances have successfully been excited in plasmonic hexamer nanoholes and metallic V-grooves along the visible to the near-infrared region (NIR), respectively. Recently, Bao et al. have proposed a numerical approach to induce toroidal dipoles under radially polarized beam excitation in symmetric plasmonic hexamer nanodisks.

The radiation patterns produced by both toroidal and Fano lineshapes cannot efficiently couple to free space; hence, we expect strong localization of the EM field squeezed in a single point at the position of these resonances. Although both Fano and toroidal modes can be exploited for development of advanced optical devices, they largely suffer from the lack of spectral tunability. Using coupling of mirrored antisymmetric Fano-resonant classical planar antennas, Gupta et al. have shown that the antialigned magnetic dipolar resonators can be modified to support toroidal dipole moment via morphological variations. Despite the exquisite features of the induced resonances in regular resonators, lack of geometrical flexibility in either planar or 3D unit cells limits possessing active control over the excited modes. Designing a unit cell with the ability of supporting different resonances (i.e., both Fano and toroidal resonances) without physical changes in the design would allow for tailoring a metasurface with exquisite controllability over the spectral response. Such an advantage can be obtained by using heat-, voltage-, and/or light-intensity-dependent materials in the geometry of the proposed design.

Recently, Bao et al. have proposed a numerical approach to induce toroidal dipoles under radially polarized beam excitation in symmetric plasmonic hexamer nanodisks.

Newly, phase-changing materials (PCMs) (i.e., vanadium dioxide (VO$_2$), AglnSbTe, and Ge$_2$Sb$_2$Te$_5$ (GST) and phase-changeable atomic monolayers (i.e., MoS$_2$) have been introduced as chalcogenide alloys for fabricating efficient nonvolatile and rewritable data storage and low-loss nanophotonic devices. Having two different phases including amorphous and crystalline states allows the PCMs to switch between dielectric and semimetallic properties by applying either optical pulses or electrical biases. As one of the most common PCMs, Ge$_2$Sb$_2$Te$_5$ (GST-225 or GST) shows reasonably fast and large refractive index variance between amorphous (a-GST) and crystalline (c-GST) phases in the NIR. For instance at the C-band (telecommunication bandwidth, $\lambda \sim 1550$ nm), the a-GST and c-GST reflections have the refractive indices: $n_{a-GST} \sim 18.89 + i2.175$ (Im($n$)/Re($n$) < 0.1) and $n_{c-GST} \sim 48.61 + i18.68$ (Im($n$)/Re($n$) < 0.3), respectively.

In this work, by going beyond classical plasmonic resonators, we describe a novel metallodielectric plasmonic unit cell consisting of a blend of metallic and PCM parts to support both Fano and toroidal dipolar moment along the NIR at different states of the employed PCM. Taking advantage of the switching between amorphous and crystalline phases of the PCM in the designed plasmonic resonator, we demonstrated the excitation of distinctly different resonant modes in the unit cell without any geometrical variations. Using electromagnetic computations, we show that a careful combination of metallic and GST parts helps to have active control over the interplay between Fano and toroidal dipolar resonances by varying the magnetic resonance circulation direction. We also analyzed the NIR switching mechanism of the proposed structure to develop practical telecommunication devices.

**METHODS**

**Electromagnetic Calculations.** Numerical analyses were performed using the three-dimensional (3D) finite-difference-time-domain (FDTD) method (Lumerical 2017) and finite-element method (FEM) (COMSOL Multiphysics). The dielectric functions of gold and glass host were taken from experimentally defined Johnson–Christy and Palik constants, respectively. The workplace is surrounded with perfectly matched layers (PMLs) as boundaries with the number of 64 and spatial grid size of $\Delta x = \Delta y = \Delta z = 1$ nm for meshing the structure. We used a plane wave pulse to excite plasmons in the unit cell with the duration of 500 fs and average power of $P_p = 8.8$ nW. To define the magnetic surface current plot, a divergence current module was applied to the FDTD model.
Modeling the Phase Switching of GST. The permittivity data for the GST arc layer was obtained from experimentally measured constants by Shportko et al.\(^{59}\) for Ge\(_2\)Sb\(_2\)Te\(_5\). Then, the effective permittivity at crystallization level of the PCM layer was defined based on the Lorentz–Lorenz effective-medium expression\(^{60}\)

\[
(e_{\text{eff}}(\lambda) - 1/e_{\text{eff}}(\lambda) + 2) = f_c(e_c(\lambda) - 1/e_c(\lambda) + 2) + f_f(e_f(\lambda) - 1/e_f(\lambda) + 2)
\]

where \(f_j\) is the volume function of the \(j\)th phase as \(0 \leq f_j = n_j / \sum n_j \leq 1\), in which \(n_j\) is the corresponding density of the \(j\)th phase. For the crystallization of GST sections of the unit cells, we used an incident continuous wave (Gaussian beam) with the irradiation power of \(P_0 = 3.2\) µW, beam fluence of \(\phi = 60\) J m\(^{-2}\), and repetition of \(f_s = 10\) kHz. Using these settings, for the sample with the distance of \(\tau\) from the source, the light fluence is defined by \(F(\tau) = 2P_0 \exp(-2r/w^2)/\pi w^2\), where \(w\) is the waist of the Gaussian beam. Finally, the produced thermal power can be calculated as a function of distance and time delay \((t_0)\) using \(T(\tau,t) = (AQ\phi/1.77\tau^2)\exp(-t-t_0)^2/\tau^2\),\(^{62}\) where \(\tau\) is the time constant of the irradiation beam.

**RESULTS AND DISCUSSION**

The schematic and top-view images for the first analyzed symmetric plasmonic unit cell is shown in Figures 1a and 1b, respectively, with geometrical details inside. The required material data and EM computation techniques are explained in the Methods section. The proposed nanoplasmonic system is composed of two nanorings located apart with a rectangular bar in between. Figure 1c illustrates the normalized transmission amplitude for the plasmonic resonator showing formation of an antisymmetric and pronounced Fano resonant (F) mode around \(~1700\) nm. According to the plasmon hybridization theory,\(^{27,43,63−65}\) two nanorings located close to each other with a rectangle resonator in between as a bright electric dipole supporter directly interact and couple to free space. Besides, as the near-field components, the surrounding nanorings sustain dark magnetic dipoles (m), which couple efficiently to the bright electric mode.\(^{65}\) The inset is the rendering for the magnetic moment rotation and direction of the excited charges. The geometrical dimensions are specified in the figure legend. Here, the magnetic moments in both peripheral rings oscillate and destructively couple to the bright mode from the central bar and lead to formation of a distinct Fano minimum. The normalized absorption and reflection spectra are exhibited in Figure 1d, consistent with the optically driven Fano dip. The local E-field maps for the excited plasmons at the Fano dip and bright dipolar mode positions are plotted as snapshots in Figures 1e and 1f. Figure 1g exhibits the surface current density in a vectorial plot at the Fano dip wavelength, confirming that magnetic moments in surrounding nanorings are oscillating in the same direction.

On the other hand, previous studies have shown that breaking the symmetry of plasmonic nanostructures enhances the dark mode and alters the width and position of the Fano resonance.\(^{9,11,15,27,66}\) Conversely, in the present system, breaking the symmetry leads to significant alterations in the magnetic current direction, resulting in unveiling of significant toroidal momentum. Figures 2a and 2b illustrate the schematic and top-view pictures, respectively, for the new unit cell with the broken rings. By removing a certain part of the nanorings with a given arc length (defined by \(L_{\text{arc}} = 2\pi R_a(\theta/360^\circ)\)), we prevent coherent rotation of magnetic moment in both rings, whereas the capacitive gaps play a fundamental role in the elimination of
Figure 3. yz-plane for the magnetic field (IH-field) excitation and direction across the unit cell for (a) Fano dip and (b) toroidal dipole.

Figure 4. (a) Transmission spectra for the toroidal resonator for samples having different arc lengths ($L_{arc}$). (b) and (c) The surface current of resonator at toroidal dipole position for two different arc lengths, 104 and 174 nm, respectively. (d) and (e) The surface current of resonator at magnetic dipole moment for two different arc lengths, 104 and 174 nm, respectively.

excitation of magnetic resonances at the toroidal mode position using FEM analyses. In order to verify formation of toroidal resonance, we plotted the surface current vectorial plot as shown in Figures 2e and 2f. At 1675 nm, the charges oscillate in opposite directions in the surrounding broken nanorings; hence, the magnetic moments do not couple to the bright dipolar mode arising from the central resonator. The magnetic vectorial charge plot demonstrates oscillation of charges in a head-to-tail configuration (toroidal magnetic moment, $\mathbf{T}$) in each isolated nanoring and leading to formation of a big closed-loop arrangement. This magnetic closed-loop current is formed due to opposite rotation of magnetic dipolar momenta in the nanorings stimulated by the capacitive gap regions. Notice in Figure 2f, at the magnetic dipole moment position, that the excited magnetic fields in both rings oscillate in a head-to-head configuration. To verify this claim, we plotted the yz-plane of magnetic (H-field) for the plasmonic structure with and without arc section at both Fano and toroidal momenta wavelengths as shown in Figures 3a and 3b, respectively. Obviously, for the Fano dip position, the charges oscillate coherently and couple to the bright mode (Figure 3a). On the other hand, for the toroidal mode, the charges oscillate in a head-to-tail arrangement across the unit cell (Figure 3b).

Focusing on the toroidal moment as the unique response of the structure, the length of the arc plays an important role in the behavior and position of both toroidal dipole and magnetic modes. To show the effect of arc length, we studied the effect of this parameter on the excited modes for 87 nm < $L_{arc}$ < 209 nm, as shown in Figure 4a. Increasing the arc length (gap distance) leads to a dramatic blue-shift in the position of magnetic dipole to the shorter spectra and finally disappeared and dampened for the largest examined arc length ($L_{arc} = 209$ nm). On the other hand, the toroidal dipole slightly blue-shifted including an
enhancement in its depth. Such a spectral response can be better understood by plotting the current density flowing on the surface of the surrounding nanorings and central block for toroidal dipole and magnetic modes as depicted in Figures 4b–4e for $L_{\text{arc}} = 104$ and 174 nm. At the toroidal dipole wavelength, the direction and intensity were not affected significantly by the morphological variations (see Figures 4b and 4c). In contrast to the Fano resonance, the negligible dependence of the toroidal resonances on major geometrical variations and antisymmetric properties is verified, consistent with the previous works.44 On the other hand, by increasing the length of the arc, the head-to-head magnetic oscillation decayed due to imperfect rotation of the magnetic dipoles (see Figures 4d and 4e). Comparing Figure 4a and surface current flow for two specific arc lengths, the results for the toroidal and magnetic resonance intensities are consistent. The effect of arc length can be further investigated by quantifying the far-field scattering power or intensity for toroidal ($I_{\text{TF}} = 2\omega^2|T_{\parallel}|^2/3c^5$) and magnetic ($I_{\text{mFF}} = 2\omega^2|m|^2/3c^3$) moments.41,68,70 Figure 5a exhibits the quantified scattering intensity profiles for both projected toroidal and magnetic dipole resonant moments for $L_{\text{arc}} = 87$ nm. The appeared distinct peaks are correlated with the targeted resonant modes with dominant scattering power for projected toroidal and magnetic moments normal to the incident beam direction ($z$-axis). Figure 5b illustrates the scattering power profile as a function of varying arc length, showing the intensity of magnetic moment decayed drastically; however, the intensity of the toroidal dipole remained nearly constant with a slight blue-shift to the higher energies.

The provided numerical calculations for the proposed plasmonic unit cell show the formation of strong and pronounced Fano and toroidal resonant modes along the NIR using a fully metallic unit cell in two different regimes. Although supporting both Fano and toroidal resonance modes on the same structure without geometrical modifications helps to design advanced nanoscale devices, the currently analyzed metasurface still lacks tunability on the plasmonic response similar to the analogous nano and microstructures in previous works.30–42,44,67,70 To address this limitation, we show how we can efficiently control the interplay between Fano and toroidal resonances without morphological variations. To this end, we introduce a PCM (here GST225 or GST) into the gap areas (arcs) with the ability to support different amorphous and crystalline states at different temperatures. This approach yields an active switching between two distinctly different resonances. Figure 6a illustrates an artistic picture for the metallic unit cell

![Figure 5](image5.png)

**Figure 5.** (a) Scattering powers of toroidal and magnetic moments ($I_{\text{arc}}$). (b) The scattering power as a function of arc length variations for toroidal and magnetic moments.

![Figure 6](image6.png)

**Figure 6.** (a) Schematic for the metallodielectric unit cell with GST arcs. (b) and (c) The transmission spectra for the arc length variations in a-GST and c-GST states of arcs, respectively. (d) and (e) The E-field maps for the metallodielectric unit cell in c-GST and a-GST phases of arcs, respectively, at the Fano and toroidal wavelengths. (f) and (g) The surface current density for the c-GST and a-GST states of arcs, respectively.
computed the spectral response in Figures 6b and 6c. For the amorphous phase (a-GST), the arc acts as a dielectric material, and due to dominancy of capacitive coupling, we expect the formation of a toroidal moment as well as a weak magnetic moment at lower energies (Figure 6b). By increasing the arc length from 87 to 174 nm, we detected a minor decay in the toroidal dipole moment, while the magnetic dipole damped significantly and blue-shifted similar to the previous analyses in this context. On the other hand, by applying the continuous beam to change the phase of GST sections to the crystalline state (see Methods), the arc behaves as a semimetallic and conductive material, leading to the formation of a Fano dip in the spectral response (Figure 6c). In addition, an intense magnetic dipole appeared at the left side of the Fano dip for shorter arc lengths, leading to the formation of a broad electromagnetically induced transparency (EIT)-like response around ~1400 nm. However, increasing the arc length causes a blue-shift in the position of the magnetic dip, finally vanishing,
while the Fano minimum approximately remains unchanged. The E-field maps for the plasmonic Fano and toroidal modes are illustrated in Figures 6d and 6e (for Lsec = 87 nm). Accordingly, the c-GST arcs in both rings act as a conductive substance, leading to coherent excitation of charges and formation of an asymmetric Fano dip (Figure 6d). On the other hand, for the a-GST, due to the dielectric behavior of the GST arc, the charges oscillate oppositely, whereas the effect of arc section is distinct in Figure 6e. Figures 6f and 6g compare the surface current density for the metallodielectric unit cells with GST substance in two opposite phases, which are consistent with the earlier studies in this work. The dashed circles at the GST position exhibit how charges intensely couple in a capacitive a-GST arc and easily shuttle in c-GST regimes.

Additionally, we carry out a series of computations to enhance the Q-factor of the induced Fano dip and toroidal moment by concentrating on the geometrical modifications. Due to different natures of Fano and toroidal modes, we expect strong dependency of each resonance to specific geometrical components. Starting from the metallodielectric unit cell with the a-GST arc, which can sustain a pronounced toroidal dipole, we analyzed the spectral behavior as shown in Figure 7. The inner radius (Ri) varies in the range of 25–75 nm (see figure legend) for the satellite nanorings, while the other geometries are kept fixed and did not affect the toroidal dipole considerably. A minor blue-shift in the position of toroidal mode to the shorter wavelengths (Figure 7a) is observed. In addition, it is noteworthy that the magnetic dipole (m) is located at longer spectra beyond 2800 nm (not shown). On the other hand, a dramatic decay in the toroidal dipole is observed by increasing the outer radius (Ro) of the particles. Interestingly, the magnetic dipole remarkably red-shifted to the higher energies including a continuous broadening in the line shape (Figure 7b). Finally, we examined the effect of gap distance (Dg) between the central resonator and the surrounding nanorings on the plasmonic response of the toroidal unit cell (Figure 7c). By changing the gap distance between the central bar and the neighboring nanorings homogeneously in the range of 5–50 nm, we observe that the toroidal moment is not affected by the capacitive gap size due to missing coupling between the dipolar electric mode excited from the central bar and the magnetic moment from the peripheral rings. We noticed in the transmission spectra for gap variations that a subtle red-shift in the position of the toroidal dipole is observed including a decay in the amplitude. One should note that due to negligible influence of the central resonator on the toroidal response we did not consider the effect of this component. On the other hand, for the unit cell with the c-GST arc, the induced Fano resonance strongly depends on the gap spots and the central block. Figures 8a–8d exhibit the geometrical variations for the inner and outer radii, gap distance, and central resonator geometries, respectively. It is obvious that Fano resonance is sensitive to the minor geometrical alterations, compared to the toroidal dipole. Being almost fixed for the nanoring variations (Figure 8a), a huge sensitivity for Fano dip is observed for the variations in the offset gap distance and central resonator. Figure 9b shows the red-shift in the position of Fano dip by increasing the gap size including a decay in the asymmetric dark mode. On the other hand, minimizing the size of the middle bar (both L and W) significantly blue-shifts the Fano dip to the shorter wavelengths (Figures 8c and 8d). One should note that the magnetic moment disappeared or is greatly suppressed due to these geometrical variations.

By using the obtained data and careful selection of the dimensions for the unit cell geometries, we plotted the transmission amplitude for a potential telecommunication switch (see Figure 9). The optimized geometries for the metallodielectric unit cell with the high-Q toroidal dipole and asymmetric Fano dip are (Ri/Ro/Dg/W/L) = (95/65/10/70/325) nm. The corresponding Q-factors for toroidal dipole and Fano dip are quantified as 14.8 and 15.6, respectively. As shown in Figure 9, the GST arc supports a toroidal resonance at 1550 nm (global telecommunication wavelength, Off-state) when it is in the amorphous state. By applying an optical pulse and heating the structure, the GST arc switches to the crystalline phase, leading to formation of a Fano dip away from 1550 nm (On-state). The rapid switching between opposite states of GST helps to develop promising and efficient plasmonic devices for practical applications.

■ CONCLUSIONS

To conclude, using PCM-mediated metallodielectric unit cells, we demonstrated fully reversible active control over the interplay between Fano and toroidal plasmonic resonant modes. For the purely metallic unit cell and complete nanorings, we observed excitation of the asymmetric Fano resonant line shape across the NIR. By removing arc-shaped sections of the nanorings, we interfered in the direction of oscillation of magnetic modes and induced toroidal dipole resonance along the same domain. To provide reversible and fully controllable modulation of the excited plasmons and switching between Fano and toroidal moments, we introduced GST arcs as a thermally functional compound in the unit cell. Our numerical results confirm that switching the phase of GST arc between amorphous and crystalline regimes gives rise to variations in the magnetic moment oscillation in both nanorings. This study represents a useful and practical approach to the active tuning and controlling of the excited plasmonic resonances without morphological variations in the geometry of the unit cell. This study opens new avenues to tailor novel and advanced nanoplasmonic devices.

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Notes
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