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OPEN Toroidal Dipolar Excitation in **Metamaterials Consisting of Metal** nanodisks and a Dielectrc Spacer on **Metal Substrate**

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We have investigated numerically toroidal dipolar excitation at optical frequency in metamaterials whose unit cell consists of three identical Aq nanodisks and a SiO₂ spacer on Aq substrate. The near-field plasmon hybridization between individual Aq nanodisks and substrate forms three magnetic dipolar resonances, at normal incidence of plane electromagnetic waves. The strong coupling among three magnetic dipolar resonances leads to the toroidal dipolar excitation, when space-inversion symmetry is broke along the polarization direction of incident light. The influences of some geometrical parameters on the resonance frequency and the excitation strength of toroidal dipolar mode are studied in detail. The radiated power from toroidal dipole is also compared with that from conventional electric and magnetic multipoles.

In 2010, dominant toroidal dipolar response was firstly experimentally observed at microwave, in metamaterials consisting of a three-dimensional (3D) array of four asymmetric split-ring resonators (SRRs)¹. In 2012, toroidal dipolar response was then pushed theoretically to the optical frequency, by scaling down the size of SRRs². The fabrication of 3D array of four asymmetric SRRs is not easy especially at optical wavelengths. In 2013, a simplified two-dimensional (2D) planar scheme was demonstrated in experiment for toroidal dipolar metamaterials which were also comprised of four asymmetric SRRs³. In the past several years, SRRs-based toroidal metamaterials have been drawing a lot of attentions^{4–16}, thanks to their novel electromagnetic properties and a variety of potential applications such as low-threshold lasing⁶, polarization transformers¹², electromagnetically induced transparency (EIT)¹³, and circular dichroism (CD)¹⁶.

Recently, toroidal dipolar response was also investigated in metamolecules with magnetic resonance¹⁷⁻²⁵, plasmonic cavities²⁶⁻³², and high-refractive-index dielectric nanostructures³³⁻⁴¹. For example, the toroidal dipolar response in the optical regime was demonstrated experimentally in metamolecules that were formed by six pairs of asymmetric double-bars¹⁷. The toroidal dipolar response was also showed theoretically in metamolecules consisting of six gold disks on a gold substrate separated by a SiO2 layer, under the excitation of radially polarized light²². The theoretical and experimental evidence of toroidal dipolar response was presented in a plasmonic cavity comprising seven round holes drilled in a thick silver film²⁶. A pronounced spectral feature in far-field scattering related to toroidal dipolar response was observed experimentally in high-refractive-index silicon nanoparticles, when the resonance frequencies of toroidal and electric dipole modes were tuned to be overlapped³⁹.

In this work, we will theoretically study the excitation of toroidal dipolar mode at optical frequency in metamaterials composed of three Ag nanodisks with equal size and a SiO2 spacer on Ag substrate. It is found that under normal incidence of plane electromagnetic waves, the near-field plasmon hybridization between individual Ag nanodisks and substrate forms three magnetic dipolar resonances. The further strong coupling among three

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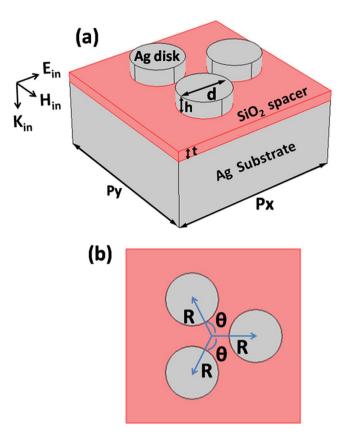


Figure 1. Oblique view (a) and top view (b) of metamaterials supporting a toroidal dipolar mode.

magnetic dipolar resonances will result into the excitation of toroidal dipolar mode, when space-inversion symmetry breaking is introduced in the polarization direction of incident light, through placing the Ag nanodisks in different locations. We have investigated in detail the influences of some geometrical parameters on the resonance frequency and the excitation strength of toroidal dipolar mode. The radiated power from toroidal dipole is also compared with that from conventional electric and magnetic multipoles. We hope that the numerical results presented in this work could be helpful to experimentally observe toroidal dipolar response at optical frequency.

Results

Figure 1 schematically shows the toroidal metamaterials composed of three Ag nanodisks and a SiO₂ spacer on Ag substrate. d and h are the diameter and height of Ag nanodisks, and t is the thickness of SiO₂ spacer. The relative positions of Ag nanodisks are determined by radius R and rotation angle θ . The periods along the x and y axes are p_x and p_y . K_{in} , E_{in} , and H_{in} are the wave vector, electric field, and magnetic field of incident light, respectively.

Figure 2(a) shows the reflection (Ref., red circle) and absorption (Abs., green triangle) spectra of toroidal metamaterials under normal incidence of light, in the frequency range from 360 to 400 THz. The spectra are calculated by the commercial software package "EastFDTD", which is based on finite-difference-time-domain (FDTD) method⁴². In our calculations, the relative permittivity of Ag is from experimental data⁴³, and SiO₂ has a refractive index of 1.45. In Fig. 2(a), there are two resonance modes centered at $f_1 = 379.25$ THz and $f_2 = 384.75$ THz, which correspond to wavelengths of $\lambda_1 = 791$ nm and $\lambda_2 = 780$ nm, respectively. At both f_1 and f_2 resonances, the reflection spectra have a dip, while the absorption spectra have a peak. To find the physical mechanisms of the resonant modes, Fig. 2(b-c) plot the magnetic field distributions at the resonance frequencies of f_1 and f_2 . For resonant mode at f_1 , one can clearly see three field "hotspots" under Ag nanodisks. Moreover, the directions of magnetic fields have a head-to-tail distribution, which implies the excitation of a toroidal dipolar mode¹. However, resonant mode at f_2 does not have such a head-to-tail distribution, though there are also three field "hotspots". It is well known that, the near-field plasmon hybridization between individual metal nanoparticle and a metal substrate can form a magnetic dipolar resonance^{44, 45}, which has been widely explored for perfect absorption^{46–48}. In our case, such plasmon hybridization forms three magnetic dipolar resonances under Ag nanodisks, resulting into the appearance of three field "hotspots". In a similar approach reported in ref. 1, the further interactions among the magnetic dipolar resonances lead to the excitation of the toroidal dipolar mode.

To further demonstrate that resonant mode at f_I is closely related to the excitation of a toroidal dipolar mode, in Fig. 3 we have calculated the radiated power I_p , I_m , I_{EQ} , I_{MQ} , and I_T from electric dipolar moment p, magnetic dipolar moment m, electric quadrupole moment EQ, magnetic quadrupole moment EQ, and toroidal dipolar mement EQ, respectively. In our calculations, the used equations² are expressed as

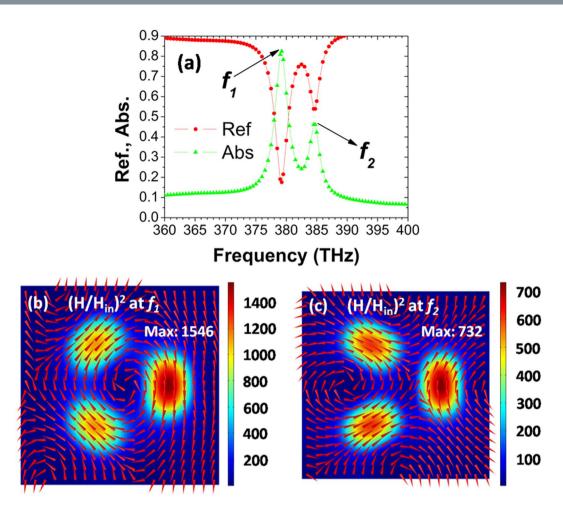


Figure 2. (a) Reflection and absorption spectra of toroidal metamaterials at normal incidence. (b,c) Magnetic field distributions on the xy plane across the center of SiO_2 spacer, at the resonance frequencies of f_1 and f_2 . Red arrows show the directions of magnetic fields, and colors give the intensity of magnetic fields. Geometrical parameters: d = 150 nm, h = 50 nm, t = 30 nm, t = 120 nm, $t = 120 \text{$

$$\mathbf{p} = (1/i\omega) \iiint \mathbf{J} \text{ dv}$$
 (1)

$$m = (1/2c) \iiint (\mathbf{r} \times \mathbf{J}) \, dv$$
 (2)

$$T = (1/10c) \iiint [(r. J)r - 2r^2 J] dv$$
 (3)

$$EQ_{\alpha\beta} = (1/2i\omega) \iiint [(r_{\alpha}J_{\beta} + r_{\beta}J_{\alpha}) - 2(\mathbf{r}. J)\delta_{\alpha\beta}/3] dv$$
(4)

$$MQ_{\alpha\beta} = (1/3c) \iiint [(\mathbf{r} \times \mathbf{J})_{\alpha} r_{\beta} + (\mathbf{r} \times \mathbf{J})_{\beta} r_{\alpha}] dv$$
 (5)

$$I_{p} = (2\omega^{4}/3c^{3})|p|^{2} \tag{6}$$

$$I_{m} = (2\omega^{4}/3c^{3})|\boldsymbol{m}|^{2} \tag{7}$$

$$I_{T} = (2\omega^{6}/3c^{5})|T|^{2} \tag{8}$$

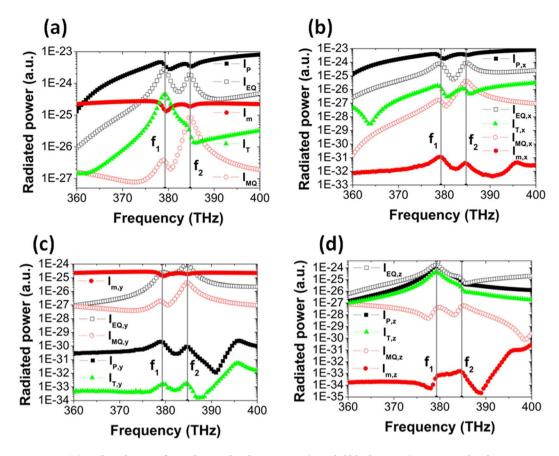


Figure 3. (a) Radiated power from electric dipolar moment (I_p , solid black square), magnetic dipolar moment (I_m , solid red circle), electric quadrupole dipolar moment (I_{EQ} , hollow black square), magnetic quadrupole dipolar moment (I_{MQ} , hollow red circle), and toroidal dipolar mement (I_T , green triangle). (**b,c**) The same as (a) but for x, y and z components of radiated power, respectively. Two vertical gray lines label the resonance frequencies of f_1 and f_2 in Fig. 2(a).

$$I_{\mathbf{EQ}} = (\omega^6 / 5c^5) \sum |\mathbf{EQ}_{\alpha\beta}|^2 \tag{9}$$

$$I_{\mathbf{MQ}} = (\omega^6/40c^5) \sum |\mathbf{MQ}_{\alpha\beta}|^2 \tag{10}$$

where ${\it r}$ is position vector, ${\it J}$ is volume current density, ω is frequency of incident light, c is light speed in vacuum, i is unit imaginary number, $d{\it v}$ indicates the volume integration carried out in a unit cell, \sum represents sigma summation, $\delta_{\alpha\beta}$ is delta function, and α , $\beta = x$, y, z. It is clearly seen in Fig. 3(a) that, the radiated power I_T from toroidal dipolar mement ${\it T}$ has a peak exactly at f_I , which clearly indicates that resonant mode at f_I is closely related with the excitation of a toroidal dipolar mode. Near the frequency of f_I , the radiated power I_T is larger than the radiated power I_M from magnetic dipolar moment ${\it m}$ and the radiated power I_M from magnetic quadrupole moment ${\it MQ}$, but it is still smaller than the radiated power I_p from electric dipolar moment ${\it p}$ and the radiated power I_{EQ} from electric quadrupole moment ${\it EQ}$. By decomposing the x, y and z components of radiated power in Fig. 3(b–d), it is found that the z component $I_{T,z}$ dominates the radiated power I_T , and it can be comparable with the z component $I_{p,z}$ as shown in Fig. 3(d).

Discussion

To study the influence of rotation angle θ on the toroidal dipolar mode, we present in Fig. 4(a) the contour plot of absorption spectra of toroidal metamaterials as a function of light frequency and rotation angle θ . The toroidal dipolar mode will blue-shift until θ increases to about 115°, because of the continuously strengthened interactions of magnetic dipolar resonances between the left two nanodisks and the right one. But, it will have a red-shift when θ is further increased, since the left two nanodisks' interactions are gradually weakened with increasing θ . Figure 4(b-e) show the magnetic field distributions on the xy plane across the center of SiO₂ spacer at a, b, c, and d points, respectively. For these points, the directions of magnetic fields under Ag nanodisks also have a vortex distribution (i.e., a head-to-tail distribution), a character of toroidal dipolar mode. As exhibited in Fig. 4(c), three field "hotspots" are simultaneously the most obvious, suggesting a relatively stronger excitation of toroidal dipolar mode for θ to be about 115°. The right field "hotspot" in Fig. 4(a) and the left two field "hotspots" in Fig. 4(d)

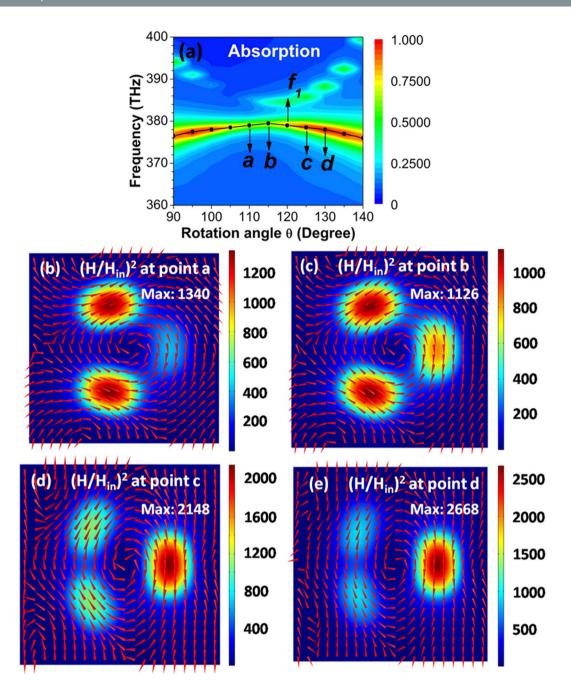


Figure 4. (a) Contour plot of absorption spectra of toroidal metamaterials as a function of light frequency and rotation angle θ at normal incidence. The overlaid black line and solid circles give the resonance position of toroidal dipolar mode. (b-e) Magnetic field distributions on the xy plane across the center of SiO₂ spacer, at a, b, c, and d points. Red arrows show the directions of magnetic fields, and colors give the intensity of magnetic fields.

become much weaker, which indicates a weak excitation of toroidal dipolar mode. When θ is smaller than 110° or larger than 140°, in principle, it is not a toroidal resonance and just is a magnetic dipole resonance.

We have also investigated the influence of radius R on the toroidal dipolar mode. Figure 5(a) shows the contour plot of absorption spectra of toroidal metamaterials as a function of light frequency and radius R. The toroidal dipolar mode is obviously red-shifted as R is varied from 105 to 150 nm, since the interactions of magnetic dipolar resonances among Ag nanodisks become weak with increasing R. Figure 5(b-e) show the magnetic field distributions on the xy plane across the center of SiO_2 spacer at e, f, g, and h points, respectively. At the four points, the directions of the magnetic fields under Ag nanodisks all have a head-to-tail distribution, indicating the excitation of a toroidal dipolar mode. When radius R is increased further, the right field "hotspot" will get stronger, while the left two field "hotspots" will get weaker.

In conclusion, we have theoretically studied the excitation of toroidal dipolar mode at optical frequency in metamaterials composed of three Ag nanodisks and a SiO_2 spacer on Ag substrate. The Ag nanodisks have

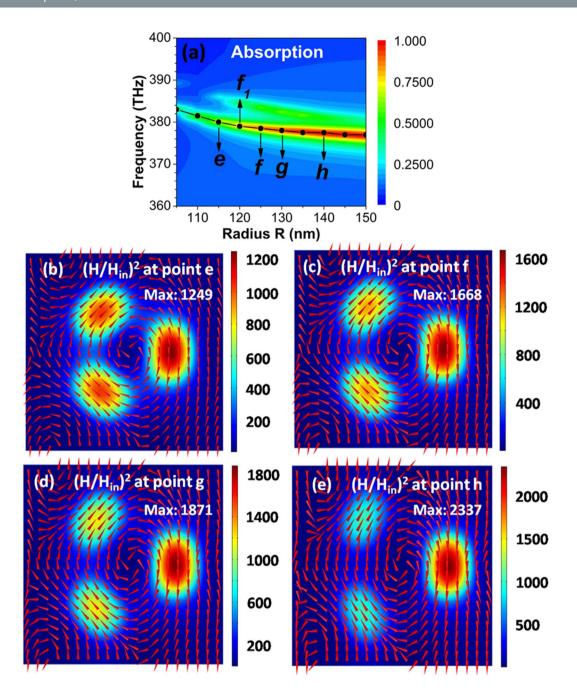


Figure 5. (a) Contour plot of absorption spectra of toroidal metamaterials as a function of light frequency and radius *R* at normal incidence. The overlaid black line and solid circles give the resonance position of toroidal dipolar mode. (**b**-**e**) Magnetic field distributions on the *xy* plane across the center of SiO₂ spacer, at *e*, *f*, *g*, and *h* points. Red arrows show the directions of magnetic fields, and colors give the intensity of magnetic fields.

identical size, but are placed in different locations to break space-inversion symmetry in the polarization direction of incident light. Under normal incidence of linearly polarized light, the near-field plasmon hybridization between individual Ag nanodisks and substrate forms three magnetic dipolar resonances, and their further interactions lead to the excitation of toroidal dipolar mode. We have investigated in detail the influences of some geometrical parameters on the resonance frequency and the excitation strength of toroidal dipolar mode. The radiated power from toroidal dipole is also compared with that from conventional electric and magnetic multipoles. Our designed metamaterials may be helpful to experimentally observe toroidal dipolar response at optical frequency.

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Author Contributions

Chaojun Tang, Qiugu Wang and Jing Chen did the calculations. Chaojun Tang, Qiugu Wang and Jing Chen wrote the manuscript. Chaojun Tang, Jing Chen and Fanxin Liu supervised the project. Chaojun Tang, Bo Yan, Qiugu Wang, Jing Chen, Zhendong Yan, Fanxin Liu, Naibo Chen, and Chenghua Sui discussed the results and reviewed the manuscript.

Additional Information

Competing Interests: The authors declare that they have no competing interests.

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