

All-optical Hall effect by the dynamic toroidal moment in a cavity-based metamaterialZheng-Gao Dong,^{1,2,*} Jie Zhu,² Xiaobo Yin,² Jiaqi Li,¹ Changgui Lu,² and Xiang Zhang²¹*Physics Department and Key Laboratory of MEMS of the Ministry of Education, Southeast University, Nanjing 211189, China*
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Dynamic dipolar toroidal response is demonstrated by an optical plasmonic metamaterial composed of double disks. This response with a hotspot of localized E -field concentration is a well-behaved toroidal cavity mode that exhibits a large Purcell factor due to its deep-subwavelength mode volume. All-optical Hall effect (photovoltaic) is observed numerically attributed to the nonlinear phenomenon of unharmonic plasmon oscillations, which exhibits an interesting analogy with the magnetoelectric effect in multiferroic systems. The result shows a promising avenue to explore various intriguing optical phenomena associated with this dynamic toroidal moment.

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I. INTRODUCTION

A standard multipole expansion method conventionally decomposes the scattering field into contributions of electric and magnetic dipoles, quadrupoles, and so forth.¹ As is known, electric polarization breaks the space-inversion symmetry while magnetization breaks the time-reversal symmetry, and thus they are incomplete when confronting systems in particle physics with simultaneous violations of space-inversion and time-reversal symmetries. In 1957, Dipolar toroidal moment \vec{T} was introduced to interpret the parity violation in weak interactions,² which is now widely acknowledged not only in nucleons,³ atoms,⁴ molecules,⁵ and other elementary particles,⁶ but even in condensed matters such as multiferroic materials.⁷⁻⁹ Historically, \vec{T} was sometimes called an anapole moment, though they are different in a strict sense because the former can radiate while the latter is nonradiating (in the stationary limit they are exactly the same thing). As a classical configuration with \vec{T} , we can consider a conducting solenoid bent into a closed toroid with a poloidal current flowing in.^{2,9-11} In condensed matters, a static \vec{T} characterized by a vortex distribution of head-to-tail magnetic dipoles is associated with various intriguing properties, such as the magnetoelectric effect,^{9,10} dichroism,^{12,13} and nonreciprocal refraction.¹⁴

However, these theoretically predicted properties associated with the elusive \vec{T} are very weak to be observed experimentally from naturally occurring permanent multiferroic materials. Recently, dynamic \vec{T} realized in metamaterials may provide a platform to unveil the intriguing optics associated with it.^{12,15-22} As evidenced by the past decade, various optical phenomena have been demonstrated by metamaterials such as the negative refraction,²³ super-imaging,²⁴ plasmonically induced transparency,²⁵ and cloaking.²⁶ By virtue of metamaterials with \vec{T} , interesting characteristics have been verified at the microwave as well as the infrared regime.^{12,15-22} However, these sophisticated toroidal structures have certain geometrical asymmetries and thus are hard for nanofabrication.

In this paper, before addressing the issue of designing an improved toroidal metamaterial that exhibits unique optical phenomena, it is of benefit to briefly review two elementary metamaterials. One is the cut-wire structure, scattering the field in the way of an electric dipole if the incident light is linearly polarized in the length direction of the wire.²⁷ The other is the split-ring structure, exhibiting a magnetic-dipole-like response

when the light is incident with the H -field polarization parallel to the normal of the split ring.²⁸ Since electric and magnetic dipoles are excited by parallel E - and H -field components, respectively, an optical \vec{T} , characterized by a centrally confined E field perpendicular to and attributed to the H -vortex distribution, could be induced by an incidence polarized parallel to \vec{T} . To mimic the classical toroidal configuration (namely, a current-carrying toroidal solenoid),^{2,9-11} we demonstrate that a metallic metamaterial comprising strongly coupled double disks (DD) is a qualified analogy of natural systems with \vec{T} .

II. SPECTRA AND MULTIPOLE CONTRIBUTIONS

Figure 1(a) schematically shows the excitation configuration of the proposed DD metamaterial (approximately a gap-bearing toroid). The incident light propagates along the x direction ($\vec{k}_{\text{light}} = k_x \hat{x}$) and is polarized perpendicular to the disk ($\vec{E}_{\text{light}} = E_z \hat{z}$) in order to satisfy $\vec{E}_{\text{light}} \parallel \vec{T}$, which can excite a toroidal mode with the induced current density (\vec{J}) shown in Fig. 1(b). To eliminate the influence of unnecessary factors, the disks are set in the vacuum unless it will be specified. For simulations performed by the CST Microwave Studio, silver disks are considered with a dispersive permittivity $\varepsilon(\omega) = \varepsilon_\infty - \omega_p^2 / (\omega^2 + i\omega\gamma)$, where the high-frequency permittivity $\varepsilon(\infty) = 6.0$, the plasma frequency $\omega_p = 1.5 \times 10^{16}$ rad/s, and the collision frequency $\gamma = 7.73 \times 10^{13}$ rad/s.^{29,30}

Total transmission and reflection spectra shown in Fig. 2 are calculated based on a two-dimensional DD array that is single layered in the x direction but with periodicities of 1500 and 490 nm in y and z directions, respectively. According to the H -field map presented in insets of Fig. 2, the first mode at 63.8 THz is a dipolar magnetic resonance, which was extensively studied in magnetic metamaterials comprising the split-ring or double-bar structure.^{31,32} The second mode around 105 THz is a quadrupole magnetic resonance.³³ Moreover, the third mode at 130 THz, characterized by the H vortex as well as the \vec{J} distribution shown in Fig. 1(b), was rarely discussed in literatures. To the interest of this work, it will be demonstrated that this response has a dominant \vec{T} and thus is highlighted for explorations of intriguing optical properties. It should be noted that, to obtain the toroidal response with an obvious H -vortex distribution, the DD gap must be small enough to ensure that paired disks are in strong plasmon coupling. In addition, the

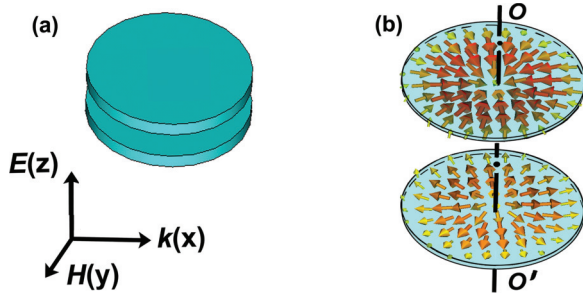


FIG. 1. (Color online) (a) Primary metamaterial element with a response of dynamic toroidal moment, where the radius $r = 600$ nm, thickness $t = 40$ nm, and gap $g = 10$ nm for the silver disk. The coordinate origin is positioned at the gap center. (b) The current distribution for the dipolar toroidal response in the DD (note that the gap between the DD is enlarged intentionally for eye guidance).

scattering/absorption ratio increases with the disk thickness as well as the gap, while the toroidal mode itself suffers a redshift with the reduction of the disk thickness.

In principle, the transmission/reflection spectra can be decomposed into radiating contributions of multipoles, including the electric, magnetic, and toroidal ones. The localized distribution of the volume current density \vec{J} in the silver disks is used to analyze these resonant responses in terms of multipole scattering contributions, according to formulas written as^{15,17,34}

$$\text{dipolar electric moment: } \vec{P} = \frac{1}{i\omega} \int \vec{J} d^3r,$$

$$\text{dipolar magnetic moment: } \vec{M} = \frac{1}{2c} \int (\vec{r} \times \vec{J}) d^3r,$$

$$\text{dipolar toroidal moment: } \vec{T} = \frac{1}{10c} \int [(\vec{r} \cdot \vec{J})\vec{r} - 2r^2\vec{J}] d^3r,$$

$$\text{quadrupole electric moment: } Q_{\alpha\beta} = \frac{1}{i2\omega} \int [r_\alpha J_\beta + r_\beta J_\alpha - \frac{2}{3}(\vec{r} \cdot \vec{J})\delta_{\alpha\beta}] d^3r,$$

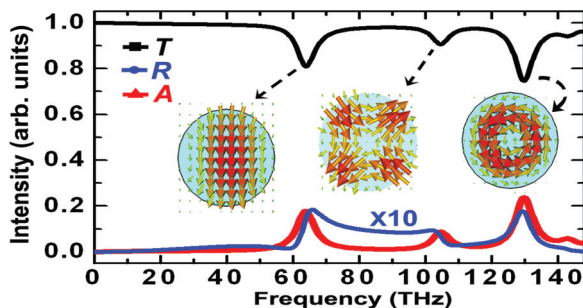


FIG. 2. (Color online) Spectra of transmittance (T), reflectance (R), and absorptance ($A = 1 - T - R$) for the proposed metamaterial configuration. The inset presents maps of the H -field vector on the middle of the gap, corresponding to the three resonant modes in the frequency sequence. The vanishing reflectance implies well-behaved nonscattering characteristics of these modes (note the reflectance curve is multiplied by a factor of 10, so that the peaks and their shapes are clear).

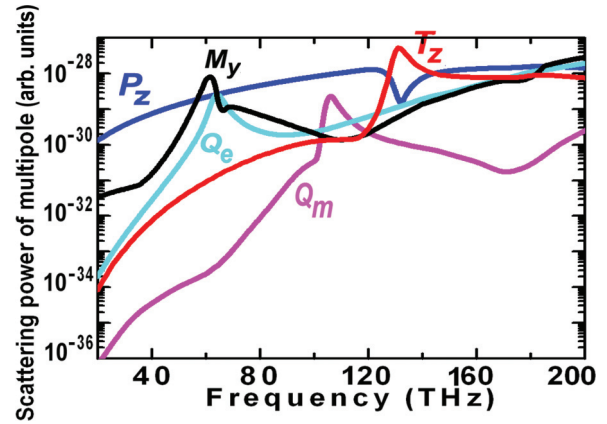


FIG. 3. (Color online) Scattering powers of multipole moment. Note the dominant toroidal moment T_z is responsible for the resonance around 130 THz.

$$\text{quadrupole magnetic moment: } M_{\alpha\beta} = \frac{1}{3c} \int [(\vec{r} \times \vec{J})_\alpha r_\beta + (\vec{r} \times \vec{J})_\beta r_\alpha] d^3r,$$

where c is the speed of light, \vec{r} is the distance vector from the origin (DD gap center) to point (x, y, z) in a Cartesian coordinate system, and $\alpha, \beta = x, y, z$. Accordingly, the decomposed far-field scattering power by these multipole moments can be written as $I_P = 2\omega^4 |\vec{P}|^2 / 3c^3$, $I_M = 2\omega^4 |\vec{M}|^2 / 3c^3$, $I_T = 2\omega^6 |\vec{T}|^2 / 3c^5$, $I_Q^e = \frac{\omega^6}{5c^5} \sum |Q_{\alpha\beta}|^2$, and $I_Q^m = \frac{\omega^6}{40c^5} \sum |M_{\alpha\beta}|^2$. For the resonance at 130 THz as shown in Fig. 3, a dominant scattering power attributed to the toroidal moment T_z is found, which cannot be accounted by common electric and magnetic multipole moments, and thus confirms the unambiguous toroidal mode.

III. CAVITY CHARACTERISTIC

It is interesting to investigate the performance of this toroidal resonance as a cavity mode, since the field confinement in this toroidal cavity (Fig. 4) is different from common cavity modes, such as a whispering-gallery or a Fabry-Perot cavity. This toroidal cavity mode cannot only concentrate the E field into a hotspot at the DD center [Fig. 4(a)], but also confine the H field in a vortex distribution [Fig. 4(b)]. As for the counterintuitive observation of field maps exhibiting a threefold rotational symmetry, it is caused by the imperfectly formed H vortex in the gap when the incident light comes

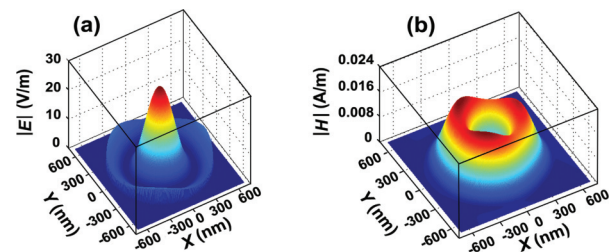


FIG. 4. (Color online) The electromagnetic concentration capability of the toroidal cavity mode in the DD structure. There are simultaneous enhancements for (a) the E field as well as (b) the H field in the DD gap.

along the x direction, since the Ag/air/Ag structure is not in a deep-subwavelength scale. Nevertheless, if an infinitesimal line-current source, oscillating in the z direction, is set in the gap center of the DD, this counterintuitive observation will disappear and vanishing field can be radiated to the outside. According to the simulation, at the hotspot of the DD cavity, the probed maximum of the E -field magnitude is $|\vec{E}_{\text{local}}| = 28.5$ V/m [Fig. 4(a)], indicating an intensity enhancement more than 800 times since the light is incident with an E -field magnitude $|\vec{E}_{\text{light}}| = 1.0$ V/m. Accordingly, the mode volume is calculated to be as small as $0.0011(\frac{\lambda_0}{2})^3$ by using the formula $V_{\text{eff}} = \int \varepsilon(\vec{r})|\vec{E}(\vec{r})|^2 d^3r / \varepsilon(\vec{r}_{\text{max}})\max(|\vec{E}(\vec{r})|^2)$, where $\lambda_0 = c/f_0 = 7.692$ μm is the freespace wavelength corresponding to the resonant frequency $f_0 = 130$ THz, \vec{r}_{max} is the hotspot location, and $\varepsilon(\vec{r}) = 1$ for the vacuum gap. Considering that the quality factor (Q) is about 19.8 (determined by the resonant frequency f_0 divided by the full width at half maximum of the spectrum in Fig. 2), a Purcell factor $F_p = (3/4\pi^2)\lambda_0^3(Q/V_{\text{eff}})$ as large as 1.12×10^4 can be reached. Additionally, such a cavity can be pushed into a deep-subwavelength scale if the gap is squeezed in further. For example, when the gap size is decreased to 4 nm, the frequency of the toroidal response will shift to 89 THz. The results verify that the toroidal cavity mode excited in the DD structure is promising to be used in situations with demand of optical cavity. Also, the giant localized field concentration at the hotspot is attractive for enhanced light emissions and nonlinear effects.

IV. ALL-OPTICAL HALL EFFECT

In multiferroic systems, it is known that the \vec{T} , for which both the spatial and temporal inversion symmetries are broken, can lead to the magnetoelectric effect.⁹⁻¹¹ That is, an applied H field induces the appearance of a built-in E field in the orthogonal direction, and vice versa. As schematically presented in Fig. 5(a), a steady poloidal current homogeneously flowing on the toroid surface will lead to a uniformly distributed H vortex (counter-clockwise), which is described by a \vec{T} perpendicular to the vortex. If an external \vec{B} field is applied along the x direction [Fig. 5(b)], the poloidal current contours with magnetic dipoles unparallel to the applied \vec{B} will be dragged transversally (i.e. shift to the left side of the toroid) in order to orient their magnetic dipoles in the direction of \vec{B} . Such a redistribution of the originally uniform carriers implies an unbalanced accumulation of carriers on the left side of the toroid, resulting in an electric polarization (\vec{P}) in the x direction.⁹⁻¹¹ This phenomenon was observed experimentally in naturally occurring ferrotoroidic materials, so called the magnetoelectric effect ($\vec{P} \propto \vec{T} \times \vec{B}$). For the optical toroidal response demonstrated in this work, its moment is dynamic, instead of the stationary one in ferrotoroidic materials. Nevertheless, a similar magnetoelectric effect can be obtained as well, regardless of its optically dynamic nature. The difference is that a dc polarization, fed from the incident light itself, can be induced without an externally applied \vec{B} field.

For the Ag/air/Ag structure, no obvious magnetoelectric effect can be observed from the centrally positioned H -vortex distribution [Fig. 5(c)]. However, it will emerge if

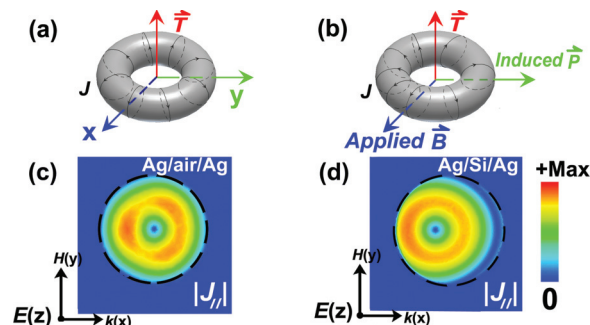


FIG. 5. (Color online) Comparison of the magnetoelectric effect in a classical toroidal system and the DD metamaterial. (a) For a classical toroidal system without an applied magnetic field, poloidal current contours are homogeneously distributed around the toroid. (b) However, they will be nonuniform when a magnetic field is applied, resulting a magnetoelectric effect. For the toroidal DD structure, amplitude distributions of tangential current density ($|J_{\parallel}| = \sqrt{J_x^2 + J_y^2}$) at toroidal resonances are plotted in (c) and (d), corresponding to the Ag/air/Ag (130 THz) and the Ag/Si/Ag (39 THz) structures, respectively (dashed lines indicate the disk profiles). A transversal displacement of the carriers (electrons) toward the left side can be verified for the latter case.

a semiconductor with a high refractive index (e.g. silicon) is filled in the gap, characterized by the left-shifted H vortex in Fig. 5(d). A direct result from the gap-filled Ag/Si/Ag structure is the resonant frequency lowering to 39 THz (approximately equal to $f_0/\sqrt{\varepsilon_{\text{silicon}}}$), which makes the sandwich structure in a deep-subwavelength geometry (the disk diameter is now about 6.5 times smaller than the freespace wavelength of the toroidal resonance). Subsequently, the H -field component of the incident light can be dealt with quasistatic approximation, acting like the applied uniform magnetic field for a stationary magnetoelectric effect in multiferroics.⁹⁻¹¹ Moreover, since \vec{T} is parallel to the incident \vec{E}_{light} , as specified earlier, it is interesting to find that the magnetoelectric relationship $\vec{P} \propto \vec{T} \times \vec{B}$ now means

$$\pm \vec{P} \parallel (\vec{E}_{\text{light}} \times \vec{H}_{\text{light}}) \parallel \vec{k}_{\text{light}},$$

where the sign depends on the positively or negatively charged carrier. Considering that electrons are the carrier in a metal, plenty of unbalanced electrons will be accumulated on the left side of the disks although they are oscillating with \vec{T} . As a result, a dc polarization \vec{P} along the light propagating direction ($\vec{P} \parallel \vec{k}_{\text{light}}$) will be retrieved from the incident light. This intriguing phenomenon is verified by the simulation result in Fig. 5(d), where current contours dragged to the left side of the toroid are similar to that expected in static ferrotoroidic systems [Fig. 5(b)]. Additionally, the magnetoelectric effect for ferrotoroidic materials typically works both ways, which means an external electric field can also induce a magnetization. Therefore, it deserves a further investigation about this reversible effect in such a dynamic toroidal system.

On the other hand, it is apparent that this phenomenon can also be understood in the context of the Hall effect because accumulated electrons on the left side, due to Ampere forces on contours of circulating current, are basically a result of charge carriers flowing in a magnetic field. Although the photoinduced

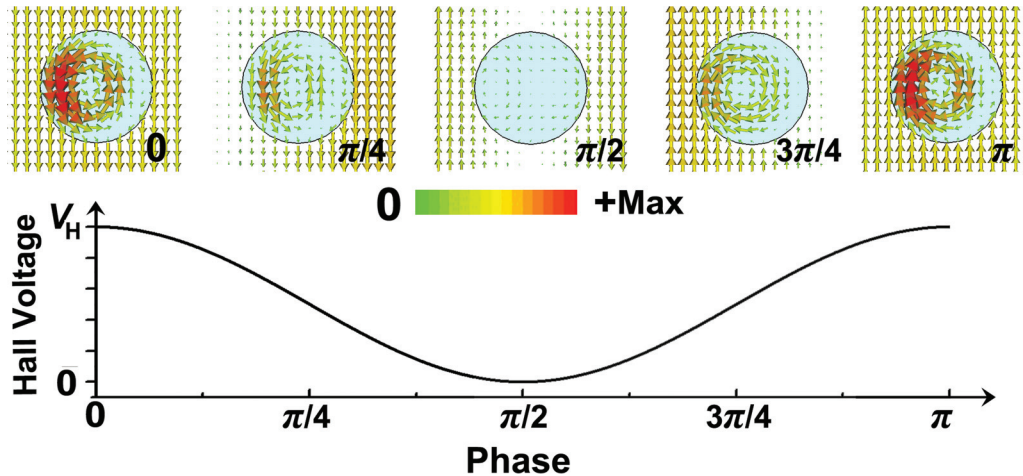


FIG. 6. (Color online) The photovoltaic signal by the all-optical Hall effect. Note the magnetic vortices on the upper panel are plotted only in a half cycle (at 39 THz), while the corresponding rectified Hall voltage is in a full cycle.

V_H is a dc signal, it is pulsating with the phase of the incident wave (Fig. 6), thus acts as an all-optical full-wave rectifier (for optical frequencies, usually only the time-averaged value is considered). It is valuable if amplitude of the rectified V_H can be estimated by calculating the point-to-point voltage difference induced in the DD structure. Unfortunately, we were unable to do such calculations due to postprocessing limitations of the adopted simulation package.

It should also be noted that there is intrinsic difference between this all-optical Hall effect and the magnetoelectric coupling in permanent multiferroic materials since they are originated from different material systems and, moreover, the former is optically dynamic while the latter is static, though an interesting analogy can be drawn between them. To get a closer physical interpretation, the reported effect can be considered as a result of the nonlinear response of unharmonic plasmon oscillations in the metallic DD structure.³⁵ In the literature, this unharmonicity can lead to the second harmonic generation and can be induced through either the structural asymmetry of plasmonic resonators³⁶ or the magnetic component of light acting on oscillating electrons via the Lorentz force.³⁷ As for the considered all-optical Hall effect, the appearance of a rectified V_H is the counterpart phenomenon of the second harmonic generation and also results from the unharmonicity of plasmonic oscillations due to the force from the magnetic component of light. Consequently, the all-optical Hall effect reported here should also not be confused with phenomena termed “optical spin Hall effect”, “Hall effect of light”, and “optical Hall effect”.^{38,39} Usually, an optical Hall effect means

a transversal shift of the light trajectory at an interface of two materials with different refractive indices.

V. SUMMARY

To summarize, we proposed a plasmonic metamaterial with a dominant \vec{T} attributed to the resonantly induced poloidal current, a mode of previously overlooked toroidal charge-current excitation. The resultant toroidal cavity characteristic with a large Purcell factor, due to the E field concentrated at the hotspot, is of great advantage as compared with common cavity modes. Particularly, in analogy to the magnetoelectric effect in ferrotoroidic materials, all-optical Hall effect attributed to the dynamic \vec{T} is demonstrated numerically from the deep-subwavelength Ag/Si/Ag structure. This unprecedented effect due to the dynamic toroidal moment can be interpreted as a counterpart of the nonlinear phenomenon that results from the unharmonicity of plasmonic oscillations induced by the magnetic component of light. In view of the enhancement characteristics of localized plasmon resonance and the orbital nature of the induced current (instead of a spin one), plenty of enhanced optical phenomena connected with the elusive \vec{T} deserve further explorations.

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