Toroidal Moments Probed by Electron Beams

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Abstract. Toroidal moments have recently attracted attentions as an individual class of moments, by expanding the vector potential and decomposing the third component into toroidal moments and magnetic quadrupoles. Toroidal moments sustain peculiar time-space symmetry relations and are known as hybrid modes, which can be only understood by including retardation effects. Here, we outline our recent attempts to realize toroidal moments in void oligomeric metamolecules. These structures are investigated experimentally using electron energy-loss spectroscopy and numerically using finite-difference time-domain method. We discuss about various toroidal moments supported by these structures, the role of symmetry and topology in excitation of these moments, the radiation from electron-induced toroidal moments resolved by cathodoluminescence spectroscopy, and finally hybridization of toroidal moments in coupled oligomers. Our findings outline the necessity of including toroidal moments within the classes of electromagnetic moments, despite the current discrepancy about their significance.

1. Introduction

Toroidal moments (TMs) have been first introduced by Dubovik within the context of electromagnetic theory [1]. By expanding the current density distribution versus Taylor series, and decomposing the terms into solenoidal and rotational terms, Dubovik demonstrated that the rotational (longitudinal) terms in the expansion set are indeed related to the time derivative of the static electric and magnetic multipoles obtained by expanding the charge density distribution or the scalar potential. However, the transverse terms are not related to the moments obtainable by expanding the charge density distribution, and hence support an individual class of moments, within which one finds also the TM. Even at those early times, it became apparent that the TM might not seem to be crucial for expanding the electromagnetic sources versus moments. Nevertheless, due to its specific symmetries, TM supports a peculiar space-time symmetry relation. Hence, it is crucial for understanding charge–current densities of the sort happening in some solid-state systems [2] or metamaterials [3, 4].

Thereafter, various attempts have been performed to realize TMs in both solid-state physics and electromagnetism. Among them, manipulation of spin–current densities to form toroidization domains in materials [2] and realization of dynamic toroidal moments at the microwave [5] and visible ranges [6-8] in meta-molecules have attracted attention. TMs in Cartesian coordinate system and in Gaussian unit can be obtained as

\[ \overline{T} = \frac{1}{6c} \int \vec{r} \times \vec{r} \times \vec{J} d^3r \]  

(1)
where \( \vec{r} \) is the displacement vector and \( \vec{J} \) is the current density distribution. In order to exclude the concomitant excitation of other classes of moments like magnetic quadruples, the best possible configuration for supporting TMs are the poloidal currents flowing along the surface of a sphere or torus. This is in particular better understood by expanding the fields instead of sources, within the spherical coordinate system [9]. In this way, the resulting electric field has three components associated with longitudinal, toroidal and poloidal currents. It is thus directly observed that electromagnetic sources are arranged as electric and magnetic multipoles (obtained from the longitudinal terms) as well as toroidal and poloidal multipoles. The latter terms are often called polar TM and axial TM, respectively [4].

Here, we intensively study the optical excitations in oligomer nano-cavities embedded in a silver thin film experimentally using electron energy-loss spectroscopy (EELS) and numerically using finite-difference time-domain (FDTD) method. This structure has been shown to support toroidal moments, which can be probed using electron beams [8]. We also investigate the radiation of TMs into the far-field using cathodoluminescence (CL) spectroscopy [11] and demonstrate that even in a single oligomer nano-cavity, two classes of TM modes are supported, namely symmetric and antisymmetric configurations. Moreover, whereas the symmetric mode contributes strongly to the radiation, the antisymmetric mode is not observed in the CL spectra. We furthermore discuss the coupling between TMs with the aim to understand hybridization effects and formation of symmetric and antisymmetric TMs [10].

2. Single toroidal moments supported by oligomers

Equation (1) may be realized as a loop of magnetic moments arranged along the circumference of a ring in a head to tail configuration (figure 1a) [1]. Given the duality theory of electromagnetism, such a magnetic loop is dual to the poloidal currents along the surface of a torus. To realize a loop of magnetic moments, we use another electromagnetic theory, the so-called Babinet’s principle. It is well known that metallic nanoparticles can support localized plasmon polaritons, in the form of electric dipole resonances. According to Babinet’s principle, one expect to observe magnetic moments in void nanostructures embedded in metallic thin films, though the magnetic dipole is oriented orthogonal to the electric dipole in the particle at the same polarization for the excited light [12] (figure 1b). A chain of nano-holes in a metallic thin film thus potentially supports a ring of magnetic moments, and hence a toroidal moment will be excited at the direction perpendicular to the thin film (see eq. (1)). This configuration of magnetic moments cannot be excited using linearly polarized light, but radially polarized light or swift electrons should be used instead [8]. This is obviously due to the fact that the polarization of the incident light should be perpendicular to the orientation of the magnetic moments. We fabricated such a structure by milling a heptamer nano-cavity composed of holes with the diameter of 80 nm into a silver film with the thickness of 50 nm. The optical modes of this structure were probed using EELS in the Zeiss SESAM microscope [13], and CL in a VG HB501UX scanning transmission electron microscope (STEM) figure 2a). Two kinds of TMs are observed even in a single oligomer cavity, depending on the electron impact position. The first mode, excited at an energy of \( E = 2.1 \) eV, is composed of a ring of magnetic moments supported by the outer holes. Such a configuration of magnetic moments gives rise to the excitation of a symmetric TM, and is observed in both CL and EELS spectra and confirmed by theoretical simulations. However, when the electron excites the structure at the silver bridge between the holes, another TM is detected, which is composed of an antisymmetric distribution of moments and two magnetic rings circulating in an opposite direction with respect to each other (compare both panels in figure 2b). This latter TM is however only observed in EELS and hidden to CL, and hence does not contribute to the radiation. This is partially due to the small mode volume of this moment, which makes it a quasi-static optical mode, with dissipative losses contributing more significantly than the radiative losses to the overall decay channels.
3. Coupled toroidal moments

Furthermore, we study the long-range coupling between TMs excited in adjacent oligomers. Two heptamers were merged with each other according to the schematic shown in figure 3a. This configuration has been extensively studied in ref. [10], where 3 distinguished TMs have been detected at $E = 1.6 \text{ eV}$, $E = 1.85 \text{ eV}$, and $E = 2.1 \text{ eV}$ (figure 3b). Here, we discuss the higher energy mode at $E = 2.1 \text{ eV}$, which is composed of three anti-parallel TMs as shown by red markers in figure 3c. As we change the distance between the central holes, the induced magnetic moments inside the holes shown by the red circles get frustrated. We notice that this frustration however is in favor of the excitation of TMs with 4 magnetic moments positioned along the circumference of the elliptic ring rather than a circular ring, compared to a single heptamer (compare figure 3c with figure 2b, left panel).

4. Conclusion

In summary, we studied the excitation of polar TM in individual and merged heptamer nano-cavities. We showed that the excitation of TMs in these structures predominates other classes of multipoles at
certain electron impacts. In a single oligomer two symmetric and anti-symmetric TMs are excited, the former contributes significantly to the radiation decay rate, whereas the latter to the dissipative losses. In coupled oligomers depending on the arrangement of nanoholes, frustration of the magnetic moment distribution can be observed. Our findings demonstrate that TMs can be spectroscopically investigated and distinguished from other moments, and may find applications in plasmonics beyond the routinely used dipolar excitations.

Figure 3. (a) A configuration, composed of two merged heptamers inserted in a silver thin film at a thickness of 70 nm, for the investigation of TMs. (b) Simulated EEL spectra for the electron beam scanning along the positions depicted by dashed lines in (a). (c) Spatial distribution of the z-component of the magnetic field at the photon energy of $E = 2.1$ eV, for the electron beam exciting the structure at the impact position depicted at panel a. The magnetic moments inside the holes marked by red color are frustrated. The direction of the induced toroidal moments is shown by black markers.

References
[1] Dubovik V M and Tugushev V V 1990 Phys. Rep. 187 145
[2] Spaldin N A, Fiebig M, and Mostovoy M 2008 J. Phys.: Condens. Matter 20 434203
[3] Papasimakis N, Fedotov V A, Savinov V, Raybould T A, and Zheludev N I 2016 Nature Mater. 15 263
[4] Talebi N, Guo S, and van Aken P A 2018 Nanophotonics 7 93.
[5] Kaelberer T, Fedotov V A, Papasimakis N, Tsai D P, and Zheludev N I 2010 Science 330 1510
[6] Tuz V R, Khardikov V V, and Kivshar Y S 2018 ACS Photon. 5 1871
[7] Miroshnichenko A E, Evlyukhin A B, Yu Y F, Bakker R M, Chipouline A, Kuznetsov A I, et al. 2015 Nature Commun. 6 8069
[8] Ögüt B, Talebi N, Vogelgesang R, Sigle W, and van Aken P A 2012 Nano Lett. 12 5239
[9] Gongora T. A. and Ley-Koo E. 2006 Rev. Mex. Fis. E 52 188
[10] Guo S, Talebi N, and van Aken P A 2018 ACS Photon. 5 1326
[11] Guo S, Talebi N, Campos A, Kociak M, and van Aken P A 2019 ACS Photon. DOI: 10.1021/acsspectrosc.8b01422.
[12] Hentschel M, Weiss T, Bagheri S, and Giessen H 2013 Nano Lett. 13 4428
[13] Talebi N, Sigle W, Vogelgesang R, Koch C T, Fernández-López C, Liz-Marzán L M, Burcu Ö, Rohm M, van Aken P A 2012 Langmuir 28 8867

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