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REVIEW ARTICLE

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Subwavelength optical localization with toroidal excitations in plasmonic and Mie metamaterials

Ruisheng Yang[1] | Jing Xu[1] | Nian-Hai Shen[2] | Fuli Zhang[1] | Quanhong Fu[1] | Junjie Li[3] | Hongqiang Li[4,5] | Yuancheng Fan[1]

[1]Key Laboratory of Light-field Manipulation and Information Acquisition, Ministry of Industry and Information Technology and School of Physical Science and Technology, Northwestern Polytechnical University, Xi'an, China

[2]Ames Laboratory and Department of Physics and Astronomy, Iowa State University, Ames, Iowa, USA

[3]Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing, China

[4]School of Physics Science and Engineering, Tongji University, Shanghai, China

[5]The Institute of Dongguan-Tongji University, Dongguan, Guangdong, China

Correspondence
Yuancheng Fan, Key Laboratory of Light-field Manipulation and Information Acquisition, Ministry of Industry and Information Technology and School of Physical Science and Technology, Northwestern Polytechnical University, Xi'an 710129, China.
Email: phyfan@nwpu.edu.cn

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Abstract
Since the performance of electronic circuits is becoming rather limited in face of intensively increasing of amount of information and related operations, all-optical processing offers a promising strategy for future information system. It would benefit a great deal if the all-optical processing could be implemented within the developed electronic chips of nanoscale structures. In that it is highly desirable to break the diffraction limit of light for achieving effective light manipulations with deep subwavelength structures compatible with the state-of-the-art nanofabrication processes. It is of fundamental importance to get subwavelength optical localization, that is, squeeze light wave into sub-wavelength space for achieving freely manipulating of light fields. This review summarizes the development in realizing subwavelength optical localization by exciting toroidal mode in photonic metamaterials. The toroidal excitations in plasmonic metamaterials and Mie resonant metamaterials, in 3D structures and planar metamaterials, with single or few layers in spectral regime from microwave to optical frequencies are surveyed. Based on the discussion on the configurations of toroidal excitations, the recent development on toroidal-related optical scattering control actively manipulates the toroidal excitations, and promising applications are further investigated and highlighted.

KEYWORDS
dielectric metamaterials, metamaterials, subwavelength optics, toroidal metasurfaces, toroidal moment

1 | INTRODUCTION

Investigation on metamaterials[1-11] and plasmonics[12-19] has stimulated rapid development in subwavelength optics,[20,21] which is emerging as the frontier of modern optics. The key issue in subwavelength optics is to break the diffraction limit of light wave and develop technologies for extreme manipulation of optical field in subwavelength scale, which is compatible with the well-developed nanolithography in modern nanoelectronics. Optically resonant modes are of fundamental importance in realizing enhanced light–matter interactions for their coupling ability with light fields in both the spatial and time domain. For the spatial domain coupling, it is highly desirable to localize light in a possibly subwavelength volume with strong fields for applications in, for example, nonlinear processing.[22,23] Plasmonic excitations in metallic structures have been exploited for realizing light localization at deep subwavelength scale.

Metamaterials[24-43] and its two-dimensional (2D) counterpart metasurfaces[44-68] made from plasmonic resonant[69-73] or Mie resonant building blocks[74-84] are especially promising for the enhancement of light–matter interactions[19,85,86] at a subwavelength scale. Various plasmonic and Mie metamaterials have been proposed for achieving high Q factor response,[87-96] for example, the trapped mode, which is kind of magnetic mode weakly coupled to free space, was suggested to be excited by introducing symmetry breaking in the shape of structural elements for realizing sharp spectral response.[87] Recently, the excitation of toroidal moment in felicitously designed plasmonic and Mie metamaterial is suggested as a new route for achieving strong optical localization and high-Q response.[26,32,97-103] The toroidal current configuration was first considered by Zel'dovich to account parity violation interaction.[104] Later, Dubovik found the possibility to introducing the new class of moment, namely toroidal moment with different time–space symmetry.[105-107] Since then the toroidal moments have already been intensively studied in nuclear, atomic and molecular physics, solid state physics, and electrodynamics.[32,105-113] A static toroidal moment can exist in various materials including metals,[114] glasses,[115] boracites,[116] pyroxens,[117] olivenes,[118] bulky crystals,[119] and biological and chemical macromolecules.[120,121] In addition to static moments, dynamic toroidal moments, also called as toroidal excitations, can be induced by interacting with incident optical fields and make contributions over the entire electromagnetic spectrum.[32,105]

In general, electric multipoles are produced by separating the positive and negative charges over a distance (oscillating charge density), whereas magnetic multipoles are created by the closed circulation of electric current (oscillating current density). Toroidal multipoles are not a part of the standard multipole expansions and originate from the decomposition of the momentum tensors with currents flowing on the surface of the torus along its meridians (oscillating radial components of current density from radiating fields). The electric excitations are strongly coupled to free space with large radiative loss. On the other hand, the generation of electrical and magnetic excitations is always accompanied by prominent induced currents (conduction current in metals or displacement current in dielectric medium), which would inevitably result in large nonradiative loss. However, the toroidal excitations are weakly coupled to free space and the magnetic fields are strongly confined in dielectric surroundings or free space.[122] The weak free-space coupling and unique light localization are crucial for achieving higher Q response and enhanced light–matter interactions. The radiation patterns of these multipoles are shown in the far right column of Figure 1.[123] To take the toroidal resonance apart from the electric and magnetic resonance, the multipolar radiation powers should be calculated first. The structure design should be optimized before experiment to make sure that the toroidal response dominates the far-field scattering power at desired frequencies, where other multipolar radiation powers are significantly suppressed. The radiation power of induced multipoles can be calculated using the following formula[26]:

$$I = \frac{2\omega^4}{3c^3}|P|^2 + \frac{2\omega^4}{3c^3}|M|^2 + \frac{2\omega^6}{3c^5}|T|^2 + \cdots \tag{1}$$

with the electric dipole moment

$$P = \frac{1}{i\omega}\int j d^3 r \tag{2}$$

the magnetic dipole moment

$$M = \frac{1}{2c}\int (r \times j) d^3 r \tag{3}$$

and the toroidal dipole moment

$$T = \frac{1}{10c}\int \left[(r \cdot j)r + 2r^2 j\right] d^3 r \tag{4}$$

where $\boldsymbol{j}$ is the current density, ω is the circular frequency, $\boldsymbol{r}$ is the displacement vector, and c is the speed of light in vacuum.

The toroidal excitations are different from electrical and magnetic excitations in traditional multipole expansions, and it is of higher-Q response (in comparison with

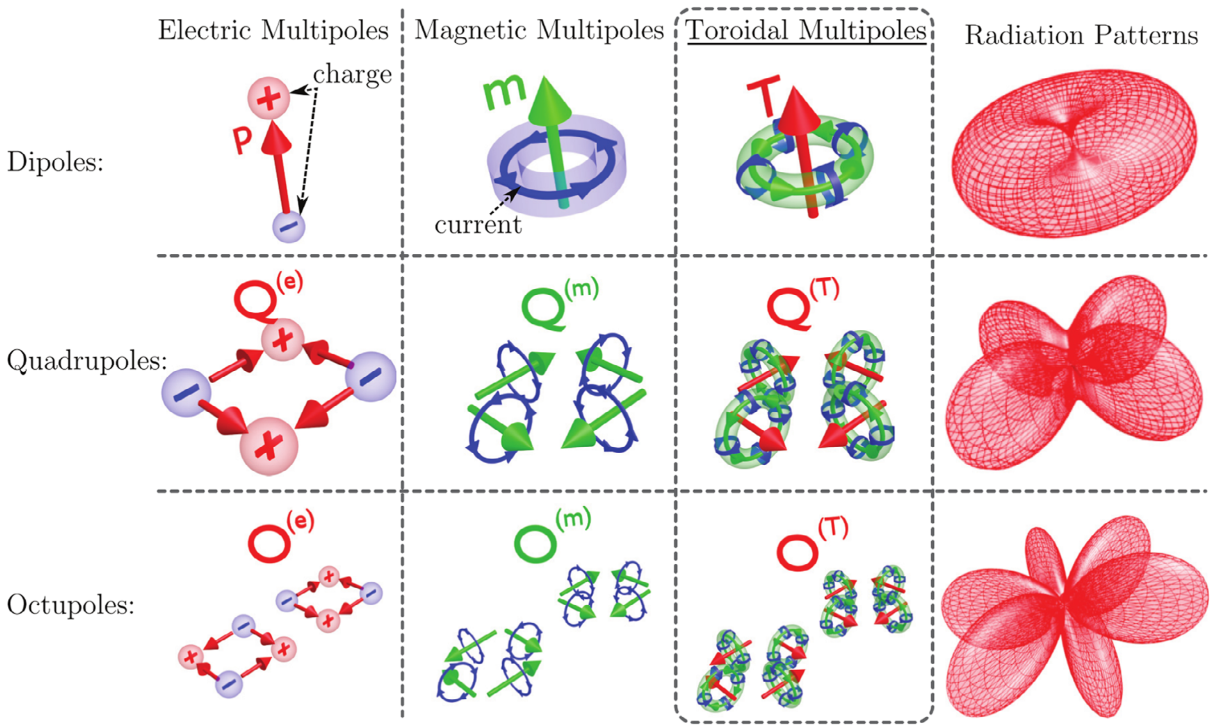

FIGURE 1 Electric multipoles result from the separation of positive and negative charges. Magnetic multipoles originate from the closed circulation of electric current. Toroidal multipoles cannot be easily considered as electric or magnetic multipoles while they are produced by the current that flows along the meridians of torus. Every type of multipole member (dipole, quadrupole, octupole) has its identical far-field power radiation patterns as shown in the far right column. Reproduced with permission. Copyright 2014, American Physical Society[123]

common electric/magnetic dipolar mode), which is due to its weak free-space coupling.[26,32] Toroidal excitations provide opportunity to further increase the light field localization and high-Q response for potential applications in low-power nonlinear processing and strong localized field-based-sensitive photonic applications. In that, toroidal excitations are of fundamental importance for freely controlling the optical signal in deep subwavelength scale. However, it is challenging to excite and detect toroidal excitations in media since toroidal dipoles are mostly weak coupled to free space.

Metamaterial with freely tailorable functions was first introduced to excite toroidal moment in 2007, the negative refraction and backward wave properties were studied in such a toroidal metamaterial.[124] Later in 2009, a toroidal dipole excitation was first reported experimentally in the microwave regime.[125] However, this excitation was hindered by other electric and magnetic multipoles. The first spectrally isolated toroidal dipole dominated resonance was observed in 2010, with metamolecules formed by ring-shaped microwave resonators, where toroidal response was enhanced to a detectable level.[26]

Later, plasmonic metamaterials with toroidal excitations were also observed at terahertz[126] and optical frequencies[98] by scaling down the size of metamolecules. To simplify the fabrication of three-dimensional (3D) plasmonic metamaterials, planar metamaterials or metasurfaces and less challenging patterns were also studied for simplified excitation of toroidal moments.[122] Although the performance of plasmonic metamaterials is restricted by the ohmic damping when reaching the higher frequencies, dielectric metamaterials with low-loss and high refractive index building blocks were also proposed to excite high-Q toroidal resonances for extreme strong subwavelength optical localization by exploiting the Mie resonances.[99] Multipole decomposition of the optical scattering of toroidal structure also illustrates that the interference of multipole modes involving toroidal mode plays an essential role in nanoscale manipulation of light.[127] A kind of nonradiating dark state, namely anapole mode, created by the nontrivial destructive interference between antiphased electric and toroidal dipoles due to their similar far-field scattering patters is also excited in metamaterials.[111,128] The destructive interference between oscillating electric and toroidal dipoles also provides a new approach for the electromagnetically induced transparency with narrow transparency lines.[129,130] These studies again show that toroidal excitations in metamaterials have great potential for the enhancement of optical light localization. In this article, we will review the progress in the development of toroidal excitations in both plasmonic and Mie resonant dielectric metamaterials for subwavelength optical localization. We will discuss various toroidal excitation configurations with subwavelength toroidal modes in 3D and 2D metamaterials or metasurfaces in a wide frequency range. The emerging toroidal excitation that involved scattering of optical wave and actively tunable toroidal metamaterials will also be investigated. Furthermore, a survey of the novel toroidal resonant mode-based applications for example, spaser and high-quality sensing, is conducted. Finally, we will discuss and envision the promising future of toroidal excitations. Hopefully this review can promote the research on subwavelength optical localization associated with toroidal excitations for high-efficient trapping of light, strongly enhanced nonlinear nanophotonics, and all-optical information processing on a chip.

2 | TOROIDAL EXCITATIONS IN PLASMONIC METAMATERIALS

Plasmonics have become one of the most vibrant areas in research with technological innovations impacting fields from telecommunications to medicine. Many fascinating applications of plasmonic nanostructures employ electric dipole, magnetic dipole, and higher-order multipole resonances for the enhancement of light-matter interaction. Besides these multipolar modes that easily radiate into free space, some other types of electromagnetic resonances also exist, such as toroidal modes generated from the decomposition of the momentum tensors, which have been largely overlooked historically. Unlike electric and magnetic multipoles, toroidal multipoles are not a part of standard multipole expansions. Toroidal multipoles with currents flowing on the surface of torus along its meridian have great capability to enhance the light–matter interactions for the unique light field localization, which is originating from the weak or nonradiating feature of the toroidal modes. In particular, it has been shown that the strength of their interaction with electromagnetic fields depends not only on the strength of the fields, but rather on their time derivatives. The rapid development of plasmonic metamaterials provided new ideas and methods for the research of toroidal multipoles. By rationally designing the symmetry of metallic resonators and their space arrangement, we can selectively suppress the fundamental electric and magnetic dipolar modes and increase the toroidal dipole responses to dominate the optical properties of metamaterial.

Herein, a collection of recent progress on toroidal excitations in plasmonic metamaterials is reviewed. Firstly, we intend to discuss the toroidal excitations in 3D plasmonic metamaterial structures.[9,97,98,131,132] In the next section, the planar designs for toroidal excitations are reviewed, which greatly simplifies the fabrication of toroidal metamaterials.[100,102,122,129,133-138] Finally, we expatiate the research proceeding on toroidal excitations in plasmonic cavities.[101,139-142]

2.1 | 3D plasmonic structures for the toroidal excitations

In 2010, the resonant toroidal response was first experimentally observed in metamaterials by Kaelberer et al.[26] The toroidal metamolecule was composed of four rectangular, electrically disconnected metallic wire loops embedded into a low-loss dielectric slab. The loops were located in two mutually orthogonal planes and separated by a distance r (Figure 2A). It is observed two peaks in the metamaterial's reflection spectra and two deeps in transmission spectra corresponding to two modes in which excitations are manifested as resonant features I (Figure 2B) and II (Figure 2C). The radiation powers as a function of the frequency are plotted in Figure 2H, where one can see that the strongest contribution of the metamaterial response at resonance I is provided by the magnetic dipole and that at resonance II is provided by the toroidal dipole. The toroidal dipole scatters more strongly than any other multipoles by almost two orders of magnitude. Compared to the value of quality factors in these two modes, resonance I is located at 16.1 GHz, with a quality factor Q of $\sim$80 and resonance II is located at 15.4 GHz with the Q factor reaching 240 (Figure 2F,G). The higher Q factor of toroidal dipole is due to its strong confinement and weak free-space coupling. In addition to achieving a high-quality factor, toroidal multipoles provide opportunities to further increase the field localization in subwavelength scale due to the weak coupling of the toroidal dipole mode to the free space.

Toroidal metamaterial provides a convincible method to further increase the Q-factor and enhance the field localization at a subwavelength scale. Based on the structure of metallic wire loops, toroidal excitations in plasmonic metamaterials were also studied in optical frequency region.[98] Limited by the dimension and resolution in the method to fabricate metamaterial structural units, the experimental realization of the resonant toroidal response was still full of challenges in higher frequency region. Up to now, a few methods have been developed for the fabrication of vertical split-ring resonators (SRRs) in micro- or nanostructure, such as double exposure e-beam lithographic process,[143] multilayer electroplating,[144] metal stress-driven self-folding method,[145] self-aligned

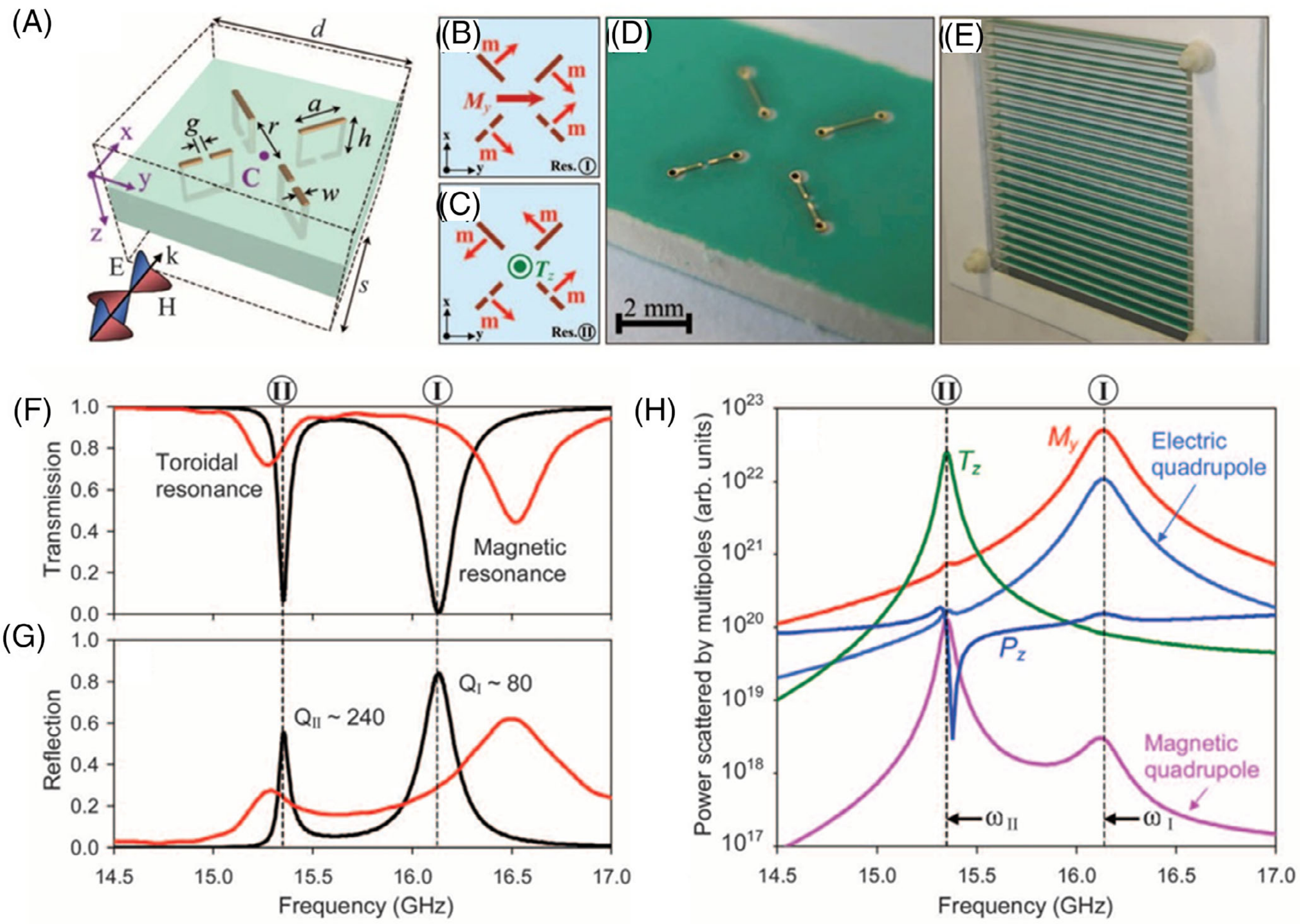

FIGURE 2 Schematic of the electromagnetic metamaterial supporting toroidal dipolar excitation and the resonant toroidal response of the metamaterial. (A) Unit cell of the metamaterial, containing four split wire loops embedded into a dielectric slab. (B,C) Two distinctively different modes of excitation corresponding to magnetic (I) and toroidal (II) dipole resonances, respectively. (D) Close-up photograph of the wire structure of the toroidal metamolecule. (E) Assembled metamaterial slab, 8 mm × 176 mm × 165 mm (green solder resist was removed before the measurements). (F,G) Calculated (black lines) and measured (red lines) transmission and reflection spectra of the metamaterial. (H) Dispersion of scattered power for various multipole moments induced in the metamolecule that contribute to the reflection and transmission spectra of the metamaterial slab. Reproduced with permission. Copyright 2010, AAAS[26]

membrane projection lithography,[146,147] two-photon polymerization process,[148,149] ion beam induced folding.[150,151] A new method for the fabrication of folded 3D metamaterials, which excited the toroidal response in the mid-infrared regime, was proposed by Liu et al. (see Figure 3A,B).[132] In this work, the adoption of metal patterns on dielectric frameworks could greatly expand the fabrication capability, showing great design flexibility and controllability on size, position, and orientation at nanometer level. Compared to the state-of-the-art techniques, not only the weakness of short connection between etch unit and the substrate was overcomed, but also the diversity of 3D structures could be greatly expanded from just metal structures to various combinations of dielectric and metal structures.

Progress in fabrication technology realizes the extension of the toroidal response frequency into optical region while maintaining high-*Q* property, which opens a horizon of optical applications including sensing, lasing spaser, and optical force. Besides the achievement of high quality factor, toroidal response is also used to enhance the field localization. As is well known, the localized spoof surface plasmons (LSSPs)[152-154] is a special surface wave mode, propagating on a periodically subwavelength structured metal surface, which has enhanced energy confinement and dispersion as well as has physical significance to the light–matter interaction in longer wavelength regimes. Realization of the LSSPs with toroidal dipole moments has physical significance to enhancement of the field localization. Toroidal dipole moments excited in LSSPs was detected in a compact planar

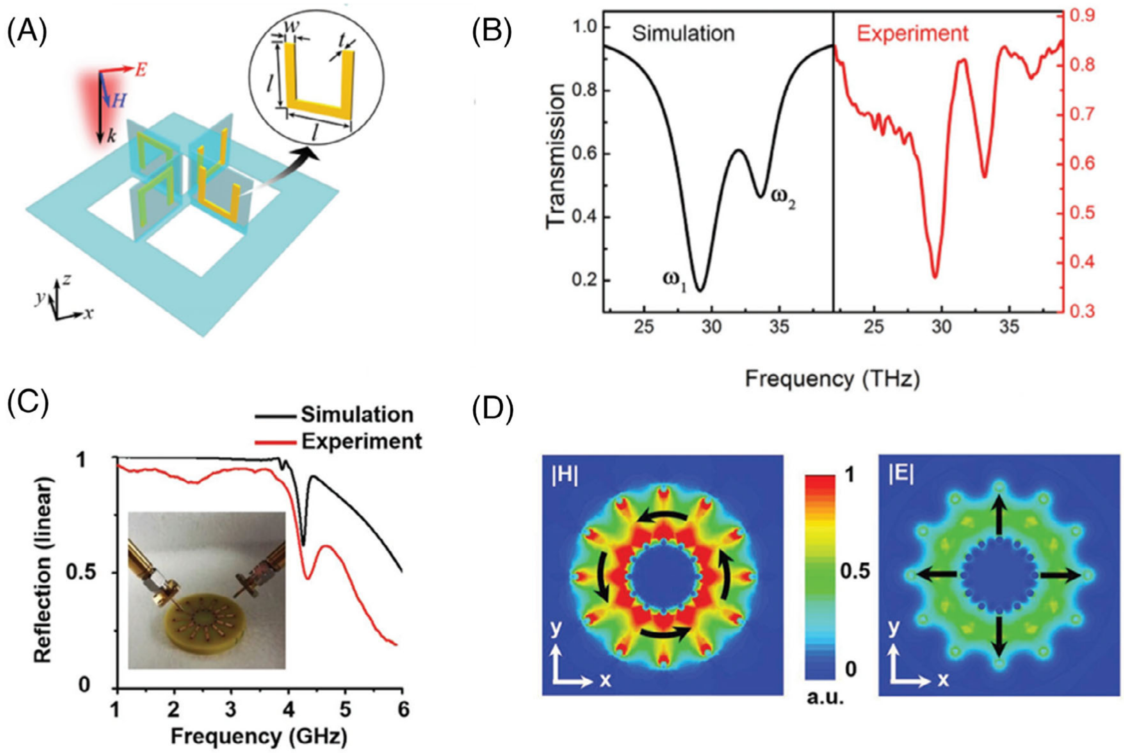

FIGURE 3 3D plasmonic structures for the toroidal excitations. (A) Schematic of a toroidal folded 3D molecule. (B) Transmission spectra of the folded 3D toroidal metamaterials in (A) by simulation and experiment. (C) Simulated (black curve) and experimental (red curve) reflection parameters. The inset is the experimental setup with the top view of the proposed structure consisting of 12 split-ring resonators. (D) Magnetic and electric field distributions of toroidal moments in (C). (A,B) Reproduced with permission. Copyright 2017, Wiley-VCH.[132] (C,D) Reproduced with permission. Copyright 2018, Wiley-VCH[155]

metadisk based on SRRs (see Figure 3C,D).[155] On the one hand, the near-field distributions of the toroidal LSSPs' resonance mode were successfully observed both in simulated and experimental results. On the other hand, the miniaturized device volume of the structure could make a vast difference to the integrated photonic circuits.

2.2 | Planar plasmonic designs to excite toroidal moment

Limited by the dimension and resolution in the method to fabricate metamaterial structural units, the experimental realization of the resonant toroidal response was still full of challenges in higher frequency region. Quasi-planar structures were designed to simplify the fabrication of toroidal metamaterials. The planar-structure-based scheme is not limited to microwave bandwidths but also shows good performance at terahertz bands and even in the optical regime. Compared with 3D metamaterials, the 2D structures offer relatively poor confinement of circulating magnetic field. Nevertheless, we could suppress the undesired multipoles and reveal the toroidal dipole contribution through the careful selection and design of the metamaterial geometry.

A planar structural toroidal metamaterial with the unit cell consists of four asymmetric split-ring resonators (ASRRs) was proposed in microwave region (see Figure 4A).[122] This work has demonstrated that toroidal metamaterials could be constructed through arrangement of planar ASRRs as meta-atoms via manipulating structural symmetry among the meta-atoms. Toroidal geometry together with Fano resonance of the ASRR made an even higher *Q* responsed metamaterial, in which light-matter interaction would be significantly amplified.

A few studies also demonstrated the toroidal dipole responses based on different planar structural metamaterials in higher frequency region. The planar-structure-based scheme simplifies the fabrication of toroidal metamaterials and also shows good performance at terahertz bands and even in the optical range. A metamaterial design with two joint metallic loops equipped two capacitive gaps in each loop was used in solution to excitations of the sharp toroidal dipolar response in the terahertz region (Figure 4C).[135] A resonant mode in which the currents in the two loops of each metamolecule oscillated in opposite directions was excited by controlling the position of the gaps and the polarization of the incident field. This mode further made the realization of suppressed electric and enhanced toroidal dipole response.

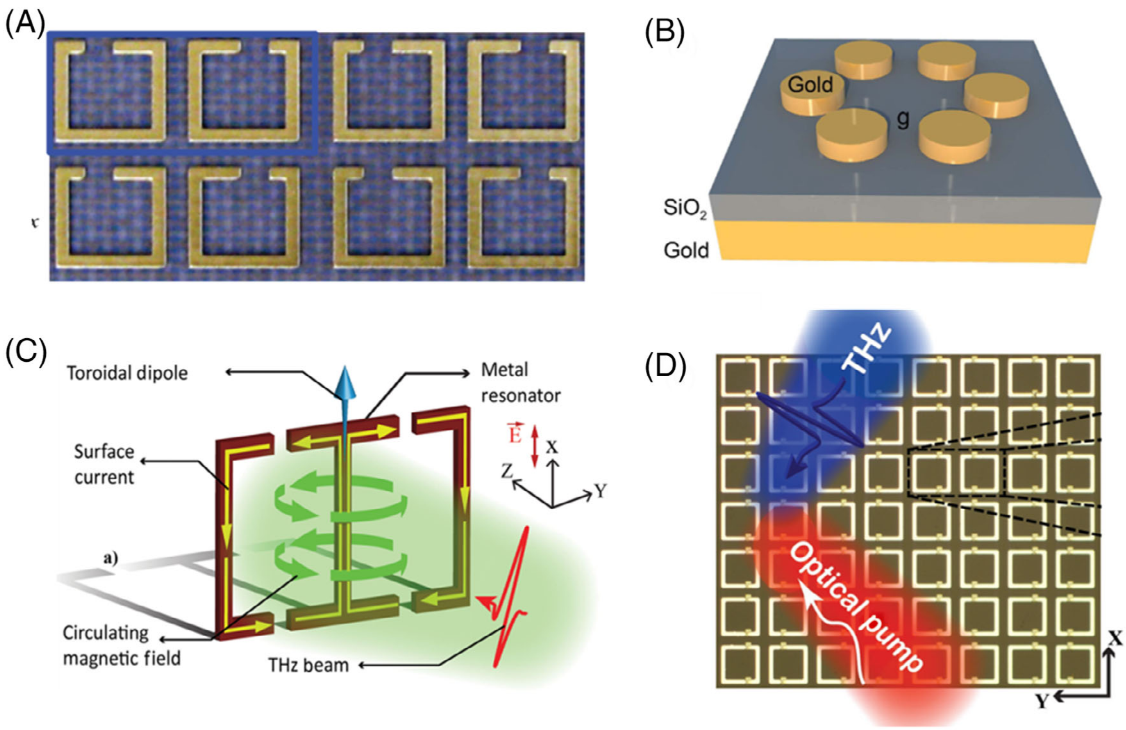

FIGURE 4 Quasi-planar plasmonic structures for the toroidal excitations. (A) Schematic of the unit cell for the ASRR-based planar toroidal metamaterial. (B) A schematic view of the toroidal structure based on metal–dielectric–metal combination. (C) Schematic of toroidal dipole generated due to circulating magnetic field produced by current carrying loops. (D) Microscopic image of fabricated metamaterial sample, along the *XY* plane, illuminated with pump beam in the presence of terahertz pulse. Switching the THz response of unit cell from toroidal dipole to electric or magnetic dipole can be achieved by optical pump. (A) Reproduced with permission. Copyright 2013, American Physical Society.[122] (B) Reproduced with permission. Copyright 2015, Nature Publishing Group.[133] (C) Reproduced with permission. Copyright 2016, Wiley-VCH.[135] (D) Reproduced with permission. Copyright 2018, Wiley-VCH[102]

By tailoring the asymmetry in the structure and the line width, the amplitude and quality factor of toroidal resonance could be tuned. The enhancement of the field localization is also realized by excitation of toroidal response in planar metamaterials. On the basis of this work, the authors demonstrated a toroidal metamaterial switch that could dynamically transform from toroidal dipole moments to electric or magnetic dipole excitations (Figure 4D).[102] The realization of this dynamic switch was attributed to the control of the optical properties of the metamaterial through ultrathin silicon layer as a dynamic material excited in the form of near-infrared femtosecond pulses. By illuminating fabricated sample at different pump powers, photoactive-mediated switching of toroidal resonance was realized. Except the structure of asymmetric split-ring, toroidal response can be excited in other planar structures. For example, in a planar metamaterial that was composed of gold hexamer and bottom gold mirror separated by a layer of silicon dioxide, the toroidal response was realized (Figure 4B).[133] The toroidal dipolar moment could be strongly excited in a metal–dielectric–metal combination in the optical region. The proposed structure could suppress the components of electric and magnetic dipole moment, and the toroidal moment could be formed by a closed loop of the magnetic dipoles that were excited in the top and bottom gold disk under the incident radially polarized light. The toroidal moment gave the dominant contribution in the scattering spectrum.

2.3 | Toroidal excitations in plasmonic cavities

In addition to the 3D and planar designs mentioned earlier, there are some other structures to excite toroidal moment such as plasmonic cavities. The toroidal systems based on plasmonic cavities could also enhance the field localization and may constitute novel approaches to waveguides and resonators. One recent literature, toroidal modes were demonstrated experimentally and theoretically at visible frequencies based on plasmonic cavities. The investigated structures were consisted of seven round holes of 60 nm diameter. The central hole was surrounded by a six-membered ring of holes, and these holes were made by drilling in a free-standing 60-nm-thick silver film (Figure 5A).[140] In this structure, the realization of quadrupolar mode, magnetic dipolar mode, and toroidal mode was induced, respectively, by the radially, azimuthally polarized far-field radiation, and radiation emitted by an electric dipole placed in the central hole at different electric fields. When an electric dipole excitation was placed in the central hole, toroidal mode was induced in the six-membered ring of holes at 2.5 eV and 3.7 eV. As shown in the spectrum of electric and magnetic field distributions, magnetic field distributions encircled the central hole and electric fields flew in radial loops between the central and surrounding holes. The sample volume between the central and surrounding holes acts as a ring, around which toroidal moments could build up. The other work of toroidal moment excitation in optical region was investigated in a structure of circular V-groove array by angle-resolved reflection (Figure 5C).[142] This work showed that a plasmonic toroidal mode around wavelength 700 nm could be excited in nanostructure for incident angles larger than 20°.

Besides these planar-array cavity structures, toroidal moments could also be excited in vertical-array cavity structures, which was made in a thin metal plate and resembled a meridianal cross-section of a toroidal void (Figure 5D).[156] Radiation suppression for metamaterials was achieved in a system based on dumbbell-shaped aperture elements. The scattering contribution from multipolar current mode could be effectively suppressed in an aperture-based structure of higher rotational symmetry. In this article, the proposed toroidal response as an out of the ordinary and nonradiating charge–current excitation realizes the enhancement of field localization and produces very narrow isolated symmetric Lorentzian transparency lines with Q factors reaching 300. In parallel, toroidal dipole moment was also achieved in a metallic metamaterial comprising pair of bars[139] and disks.[141]

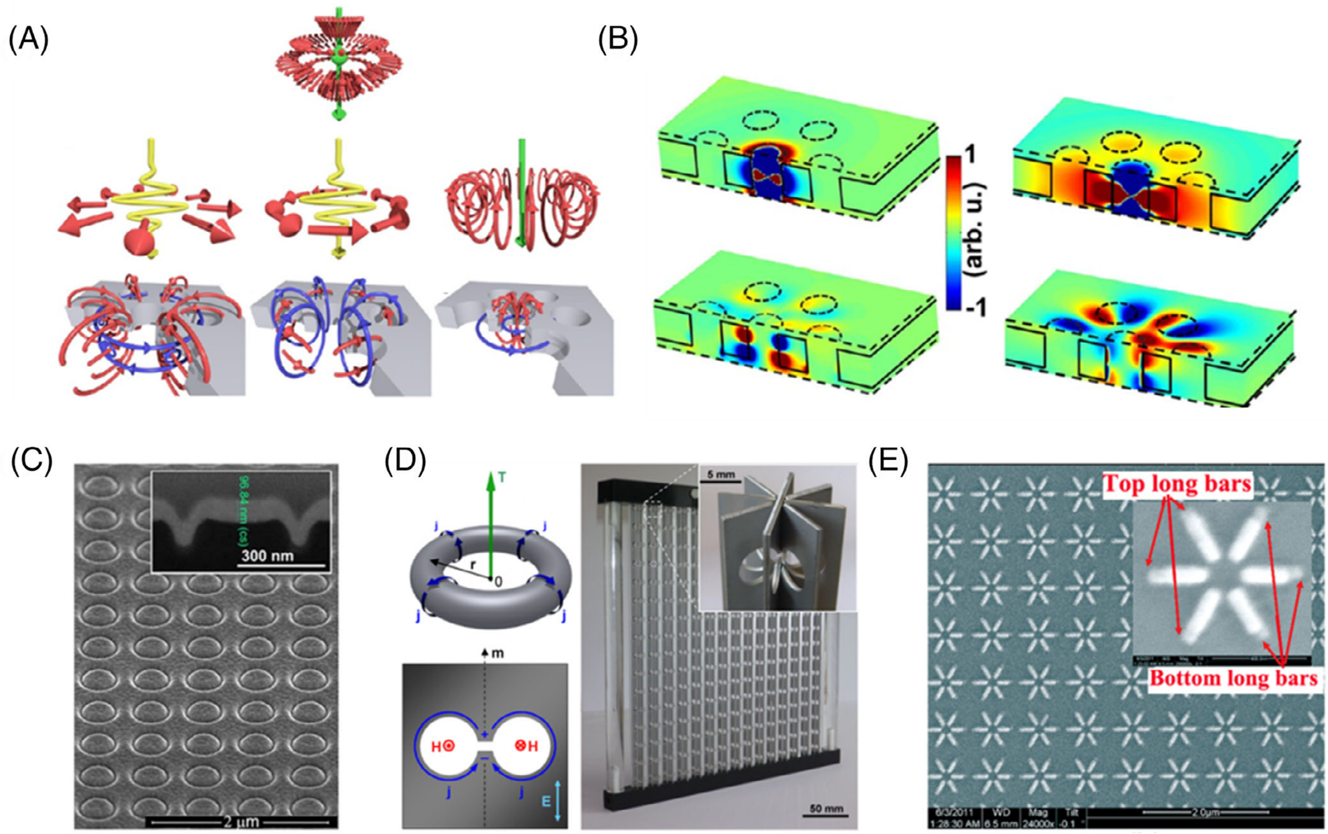

FIGURE 5 Toroidal excitations in plasmonic cavities. (A) Radially, azimuthally polarized far-field radiation, and radiation emitted by an electric dipole placed in the central hole. (B) The electric and magnetic field strength of the fundamental toroidal mode at 2.5 eV (the two pictures in the first column) and 3.7 eV (the two pictures in the second column). (C) Focused-ion-beam image of the sample circular V-groove array. (D) The upper left corner of the picture illustrated the poloidal currents flowing on a surface of a torus along its meridians create toroidal dipole moment T. The bottom left corner of the figure is a schematic of metal screen with a dumbbell-shaped aperture and it is the structural element of toroidal metamaterial. Dashed arrow m represents the axis of its mirror symmetry. The figure at right is the photograph of the assembled metamaterial slab. Inset shows a close-up view of one of the array's column with 8-fold symmetry. (E) The toroidal metamaterial composed of asymmetric double bar resonators and the image of the fabricated sample. (A,B) Reproduced with permission. Copyright 2012, American Chemical Society.[140] (C) Reproduced with permission. Copyright 2014, OSA Publishing.[142] (D) Reproduced with permission. Copyright 2013, Nature Publishing Group.[156] (E) Reproduced with permission. Copyright 2012, AIP Publishing[139]

3 | TOROIDAL EXCITATIONS IN MIE METAMATERIALS

Plasmonic metamaterials with metallic resonators utilizing toroidal designs can effectively couple with external fields to localize light at subwavelength scale. However, when operating in the higher frequencies, the inevitable ohmic loss in plasmonic structures would hinder the toroidal multipole excitations. To eliminate the dissipation loss, all-dielectric metamaterials with high refractive index and low absorption loss resonators are exploited to further localize light.[91,93] The high refractive index enables strong confinement of optical localization at a subwavelength scale and the low material loss is necessary for the low dissipation loss.[30] These resonators support volumetric Mie-type resonance modes associated with strong displacement currents, which are induced by the incident electromagnetic wave for the enhancement of optical localization.[10] By specially engineering all-dielectric metamolecules of toroidal topology, electric and magnetic excitations can be suppressed. Spectrally isolated strong toroidal dipole responses can be excited and enhanced to a detectable value.[103,157-166] A toroidal response can be excited either in a single dielectric particle with larger size or in a cluster (referred as metamolecule) formed by several dielectric particles with a proper arrangement, and the latter excitation often has a stronger toroidal moment. Different from plasmonic metamaterials, only the displacement current rather than conduction current can be induced in Mie metamaterials. The displacement currents can be extracted from the electric near-field distribution inside the dielectric cylinders by utilizing the following formula:

$$j = \frac{\partial P}{\partial t} = \varepsilon_0(\varepsilon - 1)\frac{\partial E}{\partial t} = i\omega\varepsilon_0(\varepsilon - 1)E \tag{5}$$

The multipole moments can be calculated by replacing the displacement currents into the multipole moment formulas. In the next, recent progress on Mie metamaterials for the subwavelength optical localization with strong toroidal excitations will be reviewed.

3.1 | Toroidal responses excited by the all-dielectric metamaterials

The first all-dielectric metamaterial with strong toroidal resonances was considered by Basharin et al. in 2015.[99] They proposed a kind of dielectric toroidal topology (see Figure 6A). The cluster is composed of four symmetric dielectric cylinders close to each other made of ionic crystal $LiTaO_3$ with high permittivity and negligible dissipation loss at terahertz frequency region. In the simulation model, the cylinders are assumed to be infinitely long and the polarization of incident wave is parallel to the axes of the cylinders. Each dielectric cylinder is excited by the incident electric field **E** parallel to the axes of the cylinders through near-field coupling, generating Mie-type resonance. The displacement current rather than conduction current is induced and spatially confined in each cylinder circulating along a closed loop. The magnetic moments **m**, oscillating perpendicular to the axes of the cylinders, are created by the oscillating displacement currents **j** in a narrow range of frequencies. Once the magnetic moments are aligned head to tail, a toroidal dipole **T** of dynamic vortex state with closed loops of the magnetic field would be excited inside the metamolecule. The toroidal dipolar resonance is observed with a full transmission at 1.89 THz (see Figure 6C). The toroidal excitation is confirmed by calculating the local field maps and displacement currents (Figure 6B), where a magnetic vortex field is induced by the displacement currents oscillating along a closed loop in the four cylinders. To further confirm the toroidal dipolar response in the metamaterials, the multipole moments are calculated (see Figure 6D) based on the displacement current distributions inside the metamolecule. As can be seen, the far-field scattering is dominated by the toroidal dipolar excitation around 1.9 THz, where the power scattered by other multipoles are significantly suppressed. The metamaterial with subwavelength clusters of high-index all-dielectric cylinders is operating in the Mie resonant mode, showing the capability for suppressing all other standard multipoles because of the strong toroidal excitations. This work with the special designing strategy paves the avenue to the toroidal resonance in all-dielectric metamaterials with Mie-type resonance.

The toroidal responses excited by the incident wave with electric field parallel to the axes of cylinders in 3D metamaterials were also studied in other works. Inspired by this work, further efforts have been paid to study toroidal excitations in clusters of dielectric cylinders. For example, Tasolamprou et al. presented a thorough investigation of the electromagnetic modes in metamolecules with clusters of dielectric cylinders number from 2 to 8.[165] They found that the metamolecules with an odd number of dielectric cylinders could exhibit enhanced spectral isolation for the toroidal mode. In 2015, a design of a simplified polaritonic $LiTaO_3$ microtube was proposed to excite the dominant toroidal dipolar response in the terahertz regime by Li et al.[162] A dominant toroidal dipolar excitation in a broadband frequency range with high-*Q* response and strong concentrated field at a deep subwavelength scale was found. The design strategy is promising for inspiring some applications, such as high-

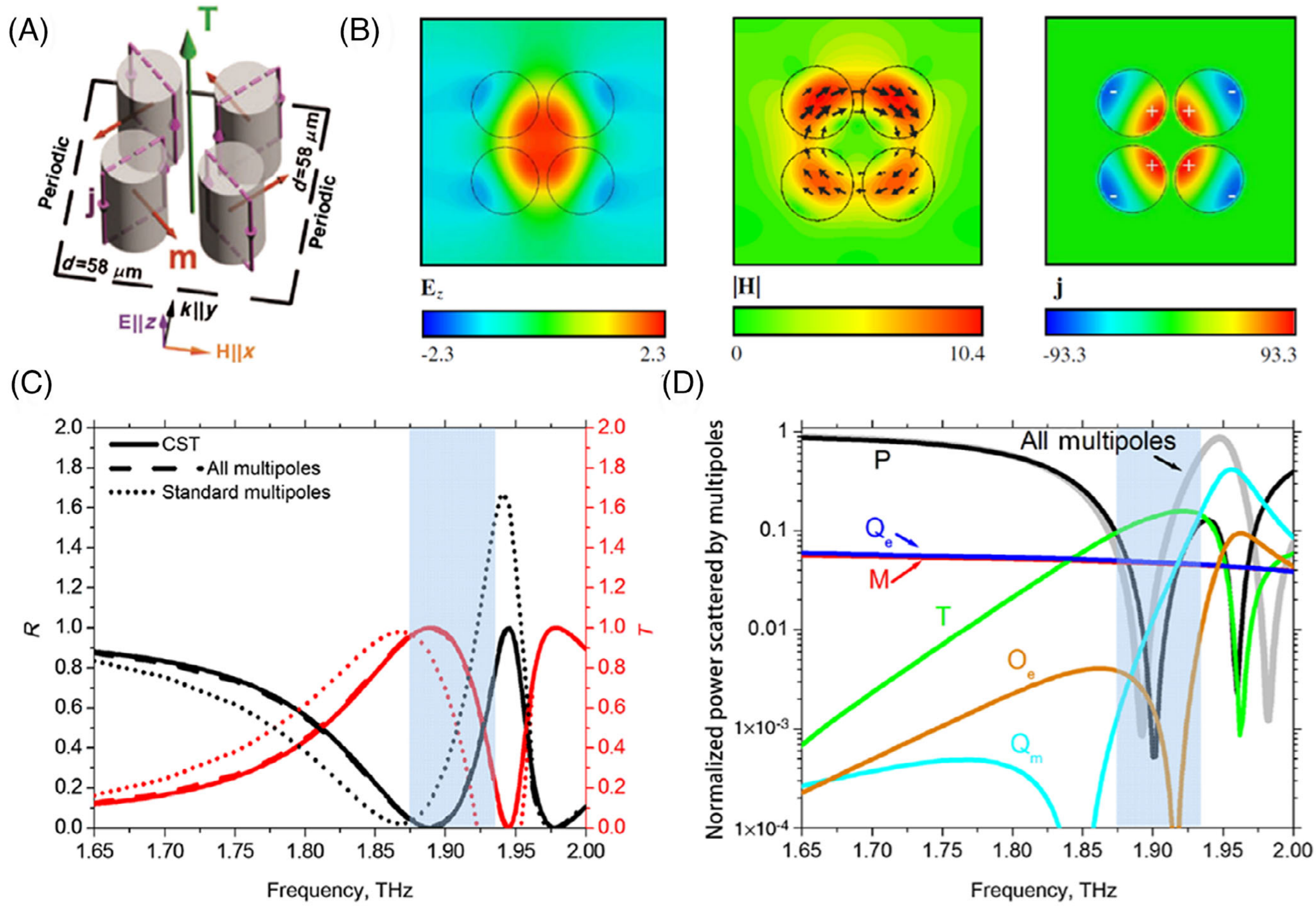

FIGURE 6 Strong toroidal excitations in a Mie-type metamaterial. (A) Schematic of a unit cell of an all-dielectric metamaterial composed of four symmetric $LiTaO_3$ cylinders exhibiting strong toroidal dipolar response in terahertz regime. (B) Calculated distributions of electric field, magnetic field (arrows showing instantaneous directions), and displacement current induced in the metamaterial at the toroidal resonant frequency. (C) Calculated transmission T (red lines) and reflection R (black lines) for the all-dielectric toroidal metamaterial. (D) Calculated normalized scattering power of the six strongest multipolar excitations for the metamaterial. Reproduced with permission. Copyright 2015, American Physical Society[99]

sensitive sensors, nonlinear optics, and particle trapping. In 2017, the toroidal dipolar response in dielectric metamaterials based on clusters of cylindrical particles was firstly measured in experiment in microwave band by Stenishchev et al.[166] These findings of all-dielectric toroidal metamaterials that taking advantage of low-loss Mie-type resonance are significant to further improve the research towards subwavelength optical localization.

3.2 | Toroidal excitations inside the all-dielectric metasurfaces

Although the toroidal excitation in 3D metamaterials is strong enough to a detectable value, the fabrication of the all-dielectric 3D toroidal metamaterials is quiet difficult, especially in higher frequencies. On the other hand, toroidal excitations in 3D dielectric metamaterials often require incident electric field parallel to the axes of dielectric cylinders, making it challenging to measure these responses. To address these issues, recently, planar designs (metasurfaces) are considered to simplify the fabrication and measurement of toroidal metamaterials. In a metasurface, the strong toroidal response can be excited by a normally incident wave under frontal excitation.[158] This requires a special configuration for metasurface. For example, Zografopoulos et al. experimentally demonstrated a single layer all-dielectric metasurface with strong toroidal response at subterahertz frequencies.[159] The metasurface is formed by dodecagonal prismatic elements made of high-resistivity floating-zone silicon with appropriately selected thickness standing on a substrate. The scattering efficiencies of multipolar mode contributions show that a strong dominant toroidal excitation appear at frequency of 93.2 GHz, where other multipole scattering efficiencies are significantly suppressed (see Figure 7A). The electric and magnetic field distributions in one of the metamolecules further confirm the toroidal excitation in the metasurface, where a pair of electric field circulating in a close loop with opposite directions induces magnetic fields oscillating arranged head-to-tail along a loop (Figure 7B). Some other dielectric metasurface designs are also proposed to excite strong toroidal dipolar responses, such as mirrored asymmetric silicon SRRs[167] (Figure 7C) designed by Liu et al. and silicon-based E-shaped metasurface[160] (Figure 7D) designed by Han et al, which support extremely high Q factor due to the weak free-space coupling and strong optical localization at subwavelength scale.

3.3 | Toroidal dipole resonances in all-dielectric oligomer metasurfaces

Since the unique electromagnetic properties of the toroidal excitations that are different from the electric and magnetic multipole modes, toroidal responses in metasurfaces have attracted growing attention in recent years. All-dielectric oligomer metasurfaces are also employed as proper platforms for the excitations of strong toroidal responses and high efficiency trapping of light. Xu et al. proposed a Mie metasurface composed of trimer clusters of high-index dielectric disks exhibiting strong toroidal dipolar responses in the microwave frequency range (see Figure 8A).[158] They directly identified the toroidal modes by the near-field intensity mapping of the electric field in experiment, and two distinct toroidal dipole modes are observed in this metasurface design (Figure 8B). Mie metasurfaces composed of different types of dielectric oligomer disks exhibiting strong toroidal dipolar excitations were further considered by Zhang et al.[157] They systematically studied the metamolecules constructed by low-loss silicon trimer, quadrumer, pentamer, and hexamer disks in the near-infrared band (Figure 8C). All these meta-molecule configurations support strong toroidal dipolar resonance when the polarization of the normally incident plane wave is directed to one of the symmetry axes of the oligomers. In particular, they found that the toroidal dipolar resonances would spectrally disappear when the meta-molecules were protected by even order symmetry, such as C4-symmetry and C6-symmetry. Such works are promising for enriching the diversity of all-dielectric electromagnetic systems with strong toroidal excitations and providing an effective flat-optics platform

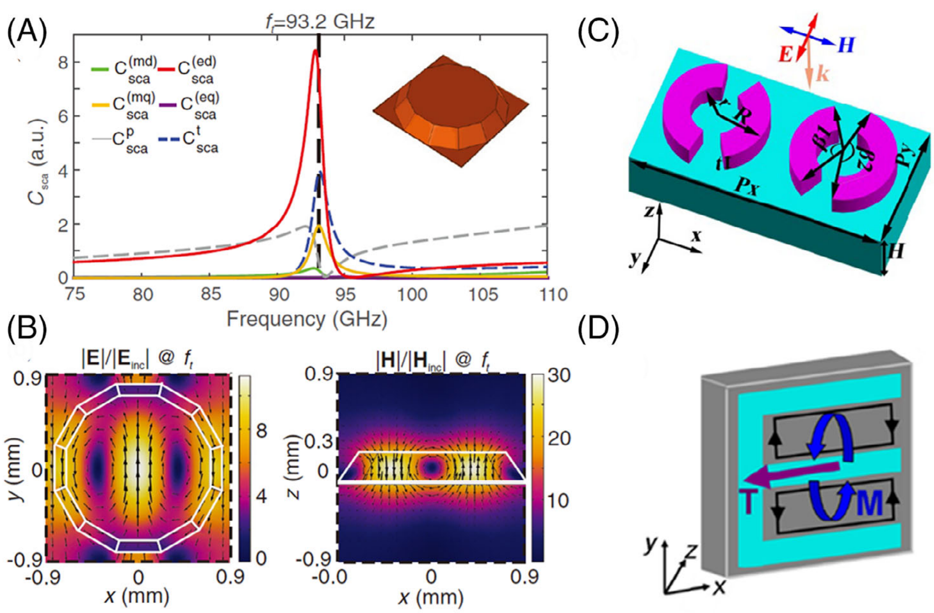

FIGURE 7 Toroidal excitations in all-dielectric metasurfaces. (A) Calculated scattering efficiency of the magnetic dipole (md), magnetic quadrupole (mq), electric dipole (ed), electric quadrupole (eq), magnetic dipole (md), Cartesian electric dipole (p), and toroidal dipole (t) moments, where a strong toroidal dipole resonance appears at 93.2 GHz. The inset shows a unit cell of the all-dielectric toroidal metasurface made of high-resistivity floating-zone silicon. (B) Simulated electric and magnetic field distributions with arrows indicating instantaneous directions, calculated at the toroidal resonant frequency. (C) A unit cell of planar all-dielectric metasurface with two mirrored asymmetric silicon SRRs on a silica substrate supports a strong toroidal dipolar resonance with narrow line width and high quality (Q) factor. (D) A schematic of toroidal dipole excitation in an E-shaped all-dielectric metasurface in the unit cell, with **M** corresponds to the head-to-tail magnetic moment. (A,B) Reproduced with permission. Copyright 2019, Wiley-VCH.[159] (C) Reproduced with permission. Copyright 2017, OSA Publishing.[167] (D) Reproduced with permission. Copyright 2018, OSA Publishing[160]

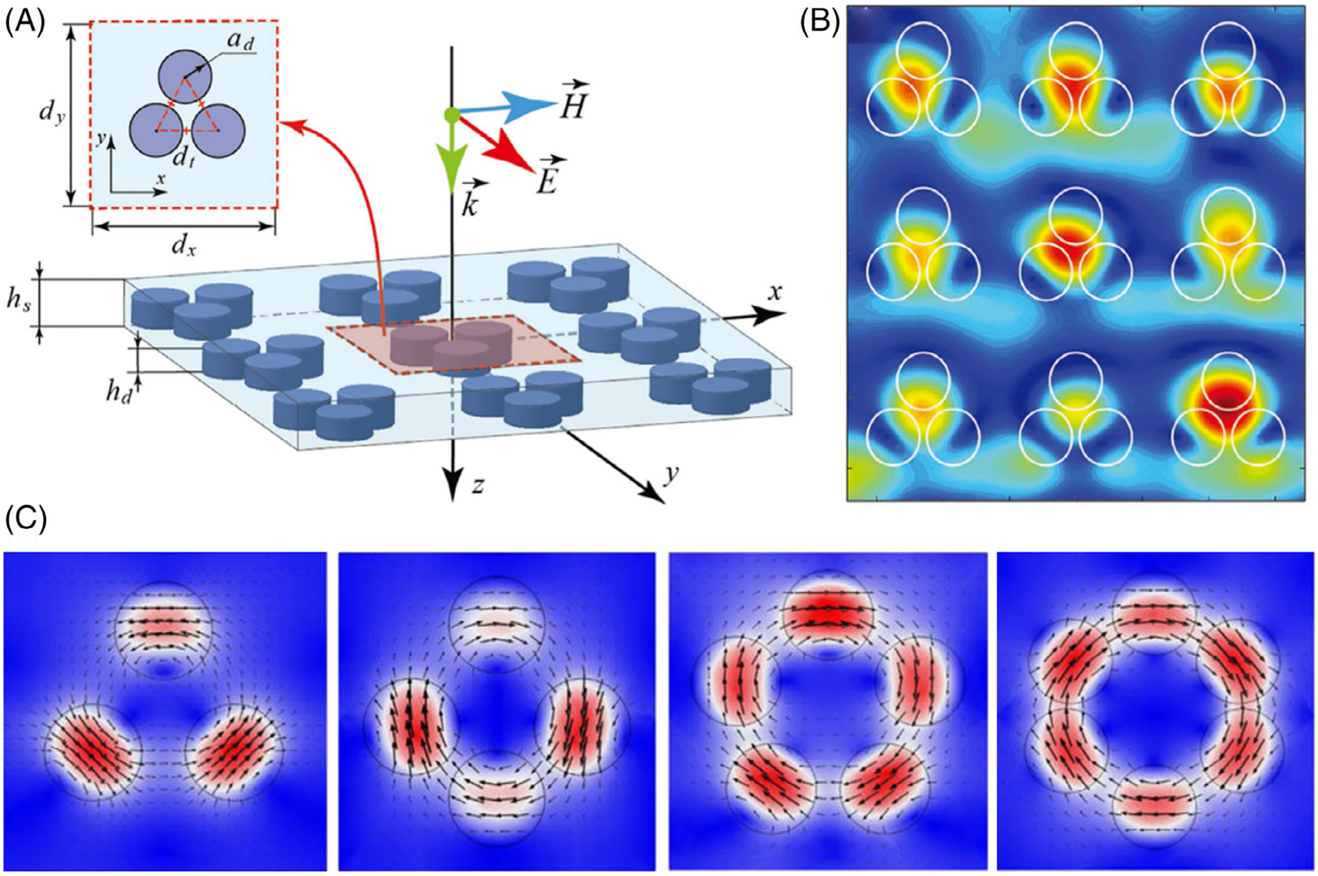

FIGURE 8 Toroidal dipole resonances in all-dielectric oligomer metasurfaces. (A) A schematic view of an all-dielectric metasurface composed of an array of clusters exhibiting strong toroidal dipolar response in microwave regime. Every cluster consists of three high refractive index ceramic disks, forming as a trimer. (B) Measured near-field distribution of the E_z electric field at the toroidal resonant frequency for the all-dielectric metasurface shown in (A). (C) Calculated magnetic field distribution at the toroidal resonant frequency for the dielectric trimer, quadrumer, pentamer, and hexamer cases, respectively. Arrows indicate the directions of magnetic field. (A,B) Reproduced with permission. Copyright 2018, Wiley-VCH.[158] (C) Reproduced with permission. Copyright 2019, Wiley-VCH[157]

for the enhancement of light–matter interactions and confinement of optical light.

4 | RECENT DEVELOPMENT AND APPLICATIONS

4.1 | Anapole excitations in metamaterials

The far-field scattering could be significantly suppressed through the complete destructive interference between antiphased toroidal and electric dipolar moments owing to their similar far-field radiation patterns, which leads to a dark state called anapole.[128] Anapole means "without poles" in Greek and has been employed as a classical model of elementary particles for the description of dark matter in the Universe. The current distribution of an anapole mode is associated with a toroidal dipole moment pointing outward along a torus symmetry axis (see Figure 9A). The oscillating currents are flowing on the surface of a torus along its medians. These poloidal surface currents can induce a set of magnetic dipoles **m** arranged head-to-tail along a loop, resulting in a toroidal dipole **T**. The radiationless properties of anapole can be achieved by exciting a second electric dipole **P** that oscillates out-of-phase with the toroidal dipole **T**, resulting in a complete scattering cancellation of far-field radiation since their scattering patterns are identical to each other. The total far-field scattering contributions can be written as

$$E_{sca} \sim \frac{k^2}{4\pi\varepsilon_0}\{n \times P \times n + ikn \times T \times n\} \tag{6}$$

The far-field radiation will vanish if $E_{sca} = 0$, when the electric and toroidal dipolar moments are out-of-phase with $P = -ikT$. This is the necessary condition for the

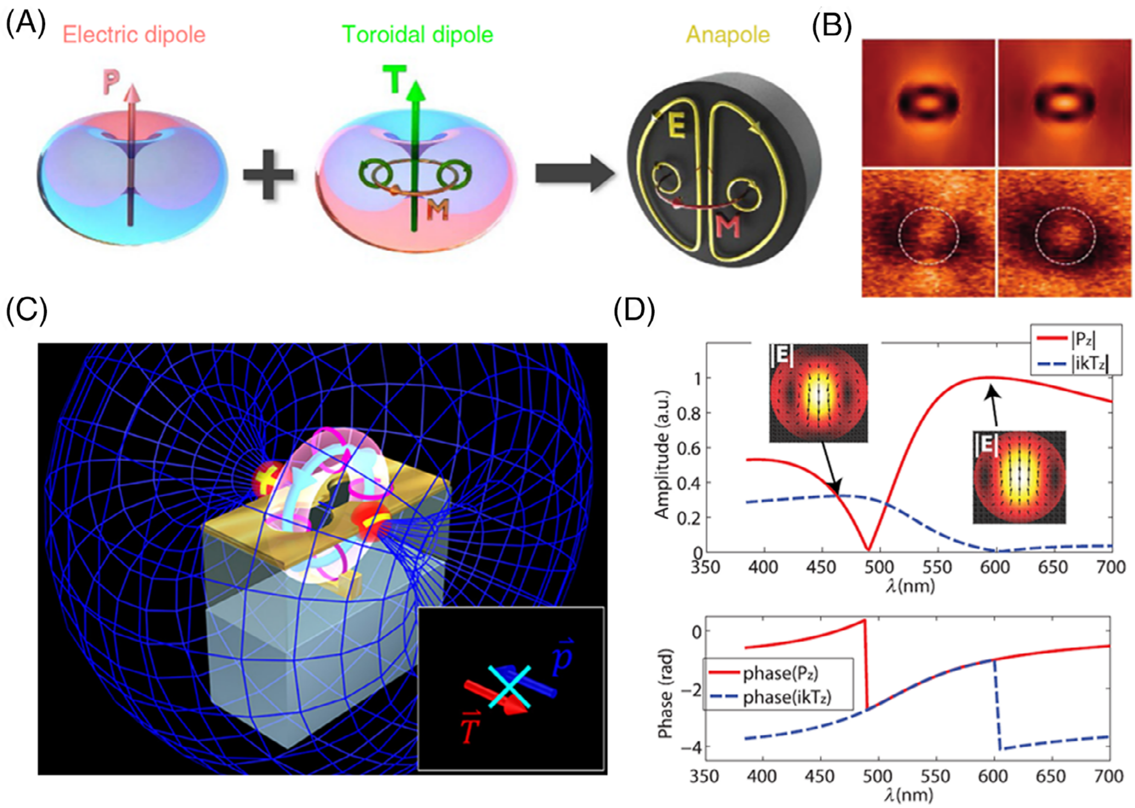

FIGURE 9 (A) Conceptual illustration of an anapole excitation. The toroidal dipole T can be considered as a set of magnetic dipoles m arranged head-to-tail along a loop accompanied by electric poloidal current distribution. The destructive interference between electric dipole P and toroidal dipole T can lead to the completely scattering cancellation in the far field since they have similar radiation patterns. (B) Near-field signature of anapole excitation around a dielectric silicon nanodisk. (C) A schematic shows the nonradiating anapole mode inside a multilayered metamaterial. The structure is composed of a planar array of vertical split-ring resonators suspended in a dielectric medium and covered with a perforated gold film. The radiating field will be significantly suppressed when the induced electric and toroidal dipoles are in antiphase due to the destructive interference. (D) Amplitude and phase of Cartesian dipole moment P_z and toroidal moment ikT_z of the induced displacement current inside a dielectric nanosphere. The left inset figure shows the induced displacement current distribution at a wavelength where the anapole mode is excited. The right inset figure shows the pure electric dipole mode. (A,B) Reproduced with permission. Copyright 2015, Nature Publishing Group.[128] (C) Reproduced with permission. Copyright 2018, American Chemical Society.[171] (D) Reproduced with permission. Copyright 2016, OSA Publishing[168]

excitation of nonradiative anapole mode. A recent work reported by Lei et al. also proved this necessary condition.[168] They calculated the z component of amplitude and phase of Cartesian dipole moment P_z and toroidal moment ikT_z inside a dielectric nanosphere, along with the electric field distributions at anapole and dipole wavelengths (Figure 9D). At the wavelength of anapole excitation, the condition $P = -ikT$ is satisfied in that the electric and toroidal dipolar moments have the same strength but are out of phase, which leads to the total scattering cancellation of the far-field radiation. The study of nonradiating anapole mode may enrich our understanding toward the nonradiating sources and non-scattering objects.

Anapole has a nonzero potential but does not generate field outside and may result in the violation of reciprocity and Aharonov–Bohm like phenomena.[128,169,170] Such a nonradiative excitation originating from the interference of electric and toroidal dipole moments can also provide a new direction for realizing invisibility cloak based the cancellation of radiation scattering.

To experimentally confirm the existence of anapole mode in all-dielectric metasurfaces, Miroshnichenko et al. fabricated silicon nanodisks on a substrate by standard nanofabrication techniques.[128] The dark anapole excitation was observed in the silicon nanodisk through the measurement of scattering spectra and near-field distribution, where a pronounced minimum appeared in the scattering spectra and an associated maximum was found in the near-field energy (Figure 9B). Such a radiationless excitation makes the nanodisk almost invisible in far field and thus provides a new way for the realization of invisibility condition based on the scattering cancellation. Wang et al. also experimentally demonstrated anapole response inside a quasi-planar plasmonic metamaterial.[171] The metamaterial driven by normally incident plane waves could simultaneously excite the antiphased toroidal and electric dipolar moments with nearly same magnitude of amplitude, leading to the anapole mode with strong localized fields but weak far-field radiation (Figure 9C). The fine control strategy for the radiationless anapole modes associated with near-field enhancement would promise important applications in nonlinear optics, sensing and cloak.

4.2 | Tunable toroidal dipole based on metamaterials

With the rapid and impressive progress in toroidal metasurface technology, active and efficient control over induced toroidal resonant modes is increasingly concerned in recent years. The dynamic modulation on toroidal dipole resonance could greatly broaden its application fields. In recent literature studies, many methods were proposed to realize the tunabilities on toroidal dipoles. Recent studies have demonstrated that toroidal moments showed more sensitivity to the incident wave power and variations of the refractive index in contrast to the classical resonant modes. Based on the strong dependence between response of toroidal plasmonic metamodulator and terahertz incident wave power, a toroidal plasmonic metamodulator was proposed by Gerislioglu et al. (see Figure 10A).[172] It was illustrated that the quality of the toroidal resonance would be significantly decreased with the reduced power of incident beam, which could be employed in some applications and devices such as metaswitches and sensors. Besides, effective modulation on toroidal dipole resonances could also be realized by changing the geometric parameters of the metamolecules and the polarization distributions of the incident wave (see Figure 10B).[173] Three different samples with different gap distances between resonators (3/4/5 μm) were fabricated and analyzed in this work. Both experimental and numerical results showed a strong polarization sensitivity and a large modulation depth, which is promising for the development of advanced terahertz applications with polarization-dependent and high-Q properties. Moreover, the tunability of inductive toroidal response was also achieved by changing the electromagnetic parameter of materials. For example, the realization of tunability was obtained by the phase change of silicon.[174] It was found that the currents flew along metallic parts of metamolecules at the dielectric state of Si, but after the transition to the metallic state, currents flowing along the silicon inclusions were dominating. The silicon conductivity variations would lead to the blueshift of toroidal dipolar frequency. In practice applications, the tunability occurs to switch between "invisible" mode and "visible" dipole mode. To switch among these responses, Tian et al. proposed and experimentally demonstrated the mode shifting between a

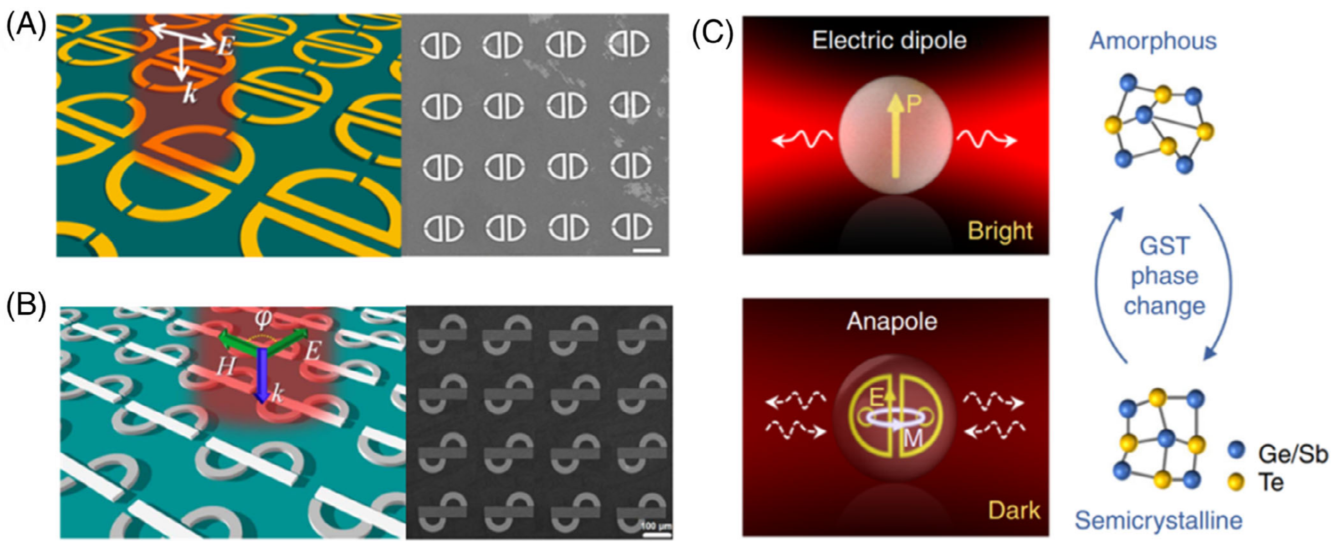

FIGURE 10 (A) Schematics of the toroidal metamolecule arrays and the scanning electron microscopy (SEM) image of the fabricated metamolecules array. (B) Graphical representation of bimetallic plasmonic metamolecules on a silicon host and the SEM image of fabricated plasmonic multipixel structures in arrays. (C) A schematic shows an electric dipolar resonance (bright state) and an anapole excitation (dark state) in a GST sphere. Shifting between these two states can be achieved by employing an intermediate phase transition of the GST material. (A) Reproduced with permission. Copyright 2018, American Physical Society.[172] (B) Reproduced with permission. Copyright 2017, IEEE.[173] (C) Reproduced with permission. Copyright 2019, Nature Publishing Group[175]

bright electric dipole resonance and a dark anapole state by exploiting intermediate phases of high-index and low-loss material GST, as schematically illustrated in Figure 10C.[175] With the phase of GST material changed between amorphous and semicrystalline states, the mode can be switched between a radiative electric dipole resonance and a radiationless anapole state. The arbitrary control of radiation states may promise applications towards tunable meta-devices with scattering on demand.

4.3 | Toroidal-based applications

The formation of toroidal moments with extraordinary properties and tunabilities in electromagnetic configuration opens a horizon of potential applications such as ultrasensitive sensors,[176] metaswitch,[102] molecule detection,[112,177,178] lasing spaser,[179] and toroidal circular dichroism.[131] In the sections that follow, we will review specific examples of applications for toroidal moments in metamaterials.

The toroidal excitations with strong light localization were used to achieve a laser spaser by Huang et al. (see Figure 11A).[179] The paper demonstrated that the toroidal dipole in a near-infrared metamaterial could be capable of lowering the levels of gain threshold for loss compensation, laser emission, and optical magnification. In this way, the authors realized an optical amplifier of coherent radiation induced by toroidal dipoles. Compared with the magnetic dipolar response, toroidal mode guarantees more excellent collective response of the metamaterial such as better coherency and narrower diversion on the beam. Considering the strong high-Q characteristics based on the toroidal response in the planar metamaterials, it was recently verified that a minute quantity of coated analyte on the toroidal dipolar metasurface caused spectral shifts of toroidal resonance, which allows the detection of the dielectric or biochemical environment near the metasurface. High Q toroidal resonances supported strong interaction between the electromagnetic wave and a specific analyte (see Figure 11B). Toroidal responses with high Q property offered a promising platform for sensing devices and

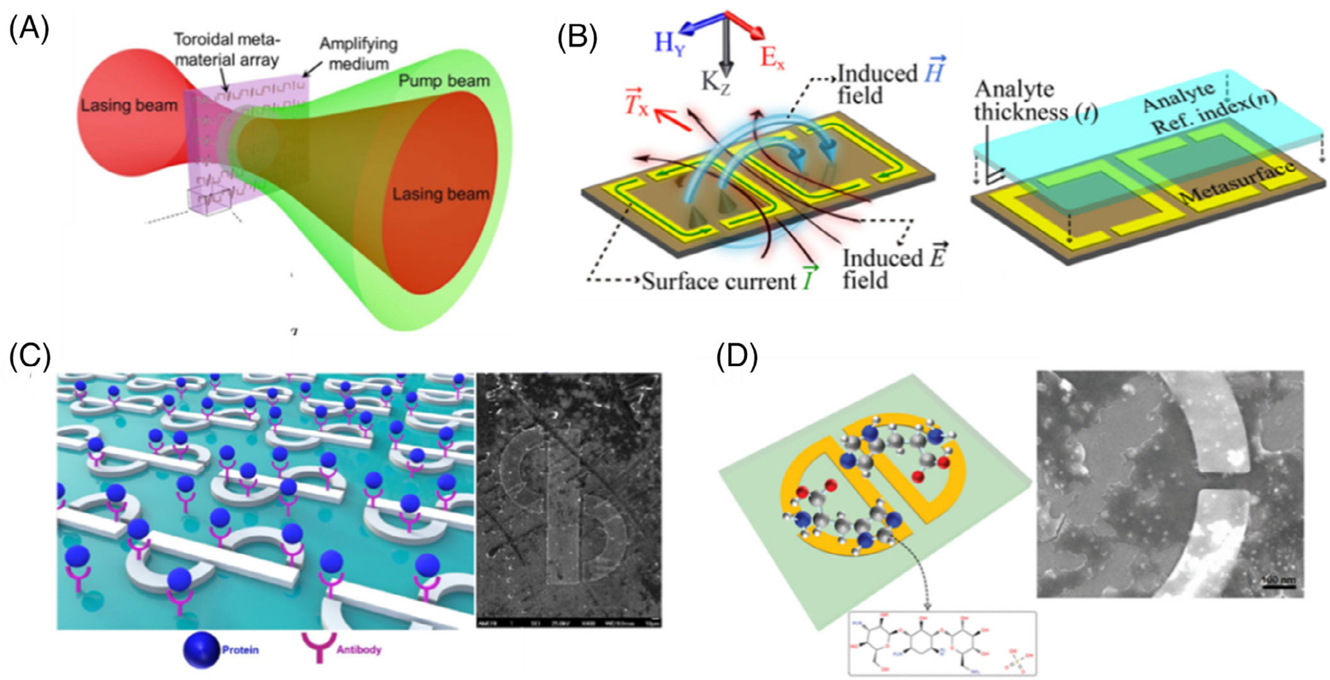

FIGURE 11 Toroidal-based application. (A) Schematic diagram of the plasmonic toroidal lasing spaser. (B–D) Sensing with toroidal metamaterial. (B) Artistic impression of toroidal dipole generated due to the circulating magnetic field produced by surface currents induced in mirrored terahertz asymmetric split-ring resonator configuration and the unit cell of the toroidal metasurface coated with the analyte layer on the top. (C) Schematic demonstration of Zika-virus (ZIKV) envelope protein binding with respective antibody on the toroidal THz plasmonic metasurface, and SEM image of the plasmonic toroidal resonator covered with ZIKV envelope proteins attached to the antibody, respectively. (D) Schematic of the proposed mid-IR plasmonic toroidal meta-atom with the presence of kanamycin sulfate molecules deposited on the surface and the SEM image of an area of metamolecule with the presence and accumulation of kanamycin sulfate molecules at the capacitive opening of the metastructure. (A) Reproduced with permission. Copyright 2013, Nature Publishing Group.[179] (B) Reproduced with permission. Copyright 2017, AIP Publishing.[176] (C) Reproduced with permission. Copyright 2017, American Chemical Society.[177] (D) Reproduced with permission. Copyright 2019, American Physical Society[178]

detecting applications.[176] Analogously, high Q toroidal metasurfaces, acting as excellent photonic devices for high sensitive sensors, have wide applications in the field of dielectric, chemical, liquid, and biological detections.[177] Rapid detection of infectious envelope proteins taking advantages of sharp toroidal moment in a plasmonic metasensor was demonstrated for the high sensitivity, repeatability, reliability, and accuracy (see Figure 11C). In another research of molecular detection,[178] toroidal dipole was experimentally proved to be highly sensitive to molecular concentrations (see Figure 11D).

5 | CONCLUSION AND OUTLOOK

Realizing the localization of light in subwavelength scale is of fundamental importance for free manipulation of light locally and with enhanced light–matter interactions. Its integration with the current developed nanoscale lithography technique is promising for all-light optical information processing on a chip. Toroidal excitations in artificial micro/nano-structured metamaterials provide a novel way for high-quality subwavelength light localization. Progresses in this field include toroidal excitations with strong light localization and high-Q response in plasmonic resonant metamaterials and Mie resonant dielectric structures, toroidal excitation associated optical anapole mode, and actively tunable high-Q toroidal mode are reviewed. It is shown that the light localization platform based on toroidal excitations can be exploited for effective and smart manipulation of light in deep subwavelength scale. We also discussed some new development related to applications of the optical localization based on the toroidal mode, such as toroidal metamaterial spaser and toroidal mode based sensing of environment. It is worth noting that the toroidal excitations based subwavelength optical localization and local optical field manipulation are still in the elementary stage on the whole, although having acquired some novel achievement from theory to some promising applications in, for example, sensing in the past decade. Its applications in optics is still localized, there are many problems that need to be further studied for achieving key technologies in designing freely controlling localization features and radiation of the subwavelength scale light fields and fabrication of high-quality metamaterial-based toroidal configuration that integrate to modern nanophotonic devices and systems.

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CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

ORCID

Ruisheng Yang https://orcid.org/0000-0002-4342-5610
Yuancheng Fan https://orcid.org/0000-0002-7919-4148

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AUTHOR BIOGRAPHIES

Ruisheng Yang received his BS degree in applied physics from the Northwestern Polytechnical University (NPU), in 2016. He is currently a PhD student majored in optical engineering at Advanced Wave-functional Microstructured Materials laboratory in NPU. His current research mainly focuses on near-field optics, nanophotonics, metamaterials, and metagratings.

Jing Xu received her BS degree in optical information science and technology from the Northwestern Polytechnical University (NPU), in 2015. She is currently a PhD student majored in optical engineering at Advanced Wave-functional Micro-structured Materials laboratory in NPU. Her current research interests include microwave metamaterial absorbers and liquid metamaterials.

Yuancheng Fan is currently a Professor at School of Physical Science and Technology, Northwestern Polytechnical University. He received his PhD degree from Tongji University in 2014. He was a joint-training PhD student with Ames Laboratory (Iowa State University) in 2012–2014. He was a Hong Kong Scholar with the Hong Kong University of Science and Technology in 2018–2020. His current research interests are in the field of light-functional micro-structured materials, including toroidal metamaterials, graphene plasmonics, and terahertz metamaterials and dielectric metasurfaces.

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