

Nanoscale chirality enhancement using topology-designed three-dimensional dielectric nanogap antennas

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We have explored, using topology optimization, three-dimensional (3D) chiral-nanogap antennas for circular-to-linear polarization conversion between far and near fields. The topology-designed 3D nanogap structures exhibit a giant chiral dissymmetry ($g = -1.70$) in gap-mode intensity against the handedness of the incident polarization. We have found that the linearly polarized gap field, designed for counterpropagating circularly polarized light, retains optical chirality localized at the subwavelength mode volume. With the nanoscale-chirality enhancement demonstrated, machine-aided designs of 3D chiral nanostructures offer a promising platform for amplifying chiral light-matter interactions with structured light at the nanoscale.

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Introduction. Chirality manifests when an object cannot be superimposed onto its mirror image. Light exhibits chirality, as represented in the forms of circularly polarized light (CPL) and optical vortices. The difference (or dissymmetry) in how left- and right-handed CPLs (l - and r -CPL) interact with a chiral molecule is typically weak due to the extremely small size of molecules relative to the light wavelength. Nevertheless, chiroptical light-matter interactions are pivotal across various fields, including circular-dichroism (CD) spectroscopy [1], chiral synthesis and crystallization in chemistry [2,3], optical trapping [4,5], and spin-state manipulation in quantum information technology [6–8].

Chiral light interactions with nanoscale objects can be enhanced by reducing the mode volume to better match the nanoscale dimensions of the objects. Additionally, a nanoscale mode volume with high chirality enhancement is essential for advancing chiral molecular analysis to the level of single molecules or trace volumes through surface-enhanced spectroscopy [9]. Metallic nanostructures such as gammadions [10,11] and helices [12–14] have been exploited to confine the chiral field at the nanoscale. Yet, designing these structures traditionally relies on heuristic approaches, largely dictated by a finite set of preassigned

design parameters determined by humans. Given the inherent complexity of evanescent near fields interacting with chiral nanostructures, a systematic design strategy is required to explore a broader design space beyond human intuition.

Topology optimization (TO), initially devised for mechanical designs [15], uses gradient-based searches rooted in Maxwell's equations [16,17]. TO enables the handling of large-scale structure designs, comprising hundreds to potentially billions of spatial points, each assigned an appropriate permittivity value, offering a considerable degree of design freedom [18]. This allows for designing structures by traversing topologically discontinuous deformation, a feature unattainable with traditional design strategies that only optimize dimensional design parameters [19] and outer shape. TO has been applied to the design of photonic waveguides [20], nanogap cavities [21], and metallic structures for enhancing Raman scattering [22]. Nonetheless, despite its remarkable flexibility, topology design for chiral photonic structures involving helically structured light has only been partially explored in the context of a periodic diffractive device [23]. The use of full three-dimensional (3D) topology optimization (TO) to create chiral optical nanoantennas with subwavelength mode volumes and to investigate their potentially distinct chiral field properties remains largely unexplored.

This study explores a 3D dielectric chiral-nanogap antenna for nanoscale field confinement and enhancement with increased chiral dissymmetry through 3D TO. Specifically, we present a nanogap antenna that supports a gap mode with a subwavelength mode volume, in which the field becomes linearly polarized upon interaction with

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propagating far-field CPL. Such 3D structures are difficult to conceive by human intuition. The near-field localization and enhancement are analyzed with respect to the handedness of the incoming CPL. We show that the linearly polarized gap field, designed with counterpropagating CPL, retains significantly large optical chirality within the nanogap and that the results are explained by the converging flux of chirality. Finally, far-field CPL emission from the 3D chiral-nanogap antenna, facilitated by the local chirality with a subwavelength mode volume, is proposed.

Topology design of nanogap antenna for CPL. TO began with the configuration illustrated in Fig. 1(a), involving a single titanium dioxide (TiO_2) slab on a silicon dioxide (SiO_2) substrate.

A right-handed circularly polarized Gaussian beam with a 532-nm wavelength was directed onto the substrate via a 0.25-NA (numerical aperture) lens. The beam diameter was 982 nm. We have considered a gap mode that is linearly polarized along the x axis within an 8-nm³ space at the center of the TiO_2 slab. A power flux was defined with its polarization constrained to be linear [24]. TO was then applied to maximize the power flow into the gap position, thereby enhancing the local field energy density.

A typical topology-optimized structure is presented in green in Fig. 1(b) (for perspective views, see Videos S1 and S2 in the Supplemental Material [24]). It exhibited

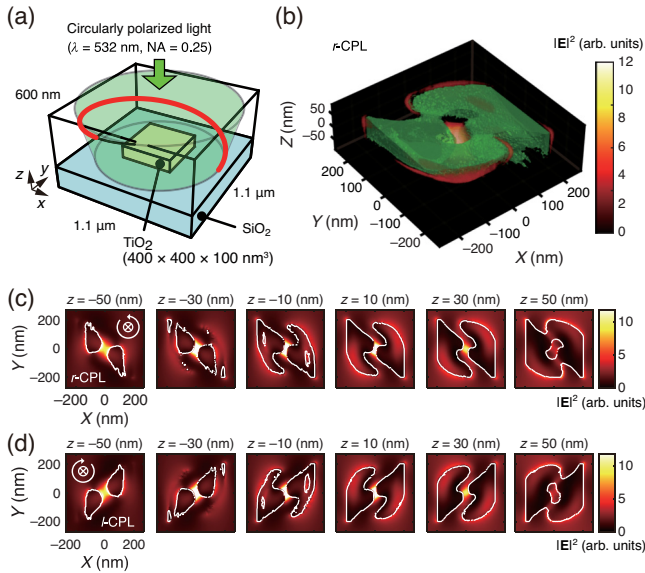


FIG. 1. The topology-designed gap antenna for CPL. (a) A schematic of the TO configuration. (b) A 3D rendering of the antenna structure (green) optimized for incident r -CPL, with the overlaid electric field intensity distribution. (c) The r -CPL-optimized antenna structure (outlined in white) and the associated field intensity distribution at various z positions. (d) Similar to (c), optimized for incident l -CPL.

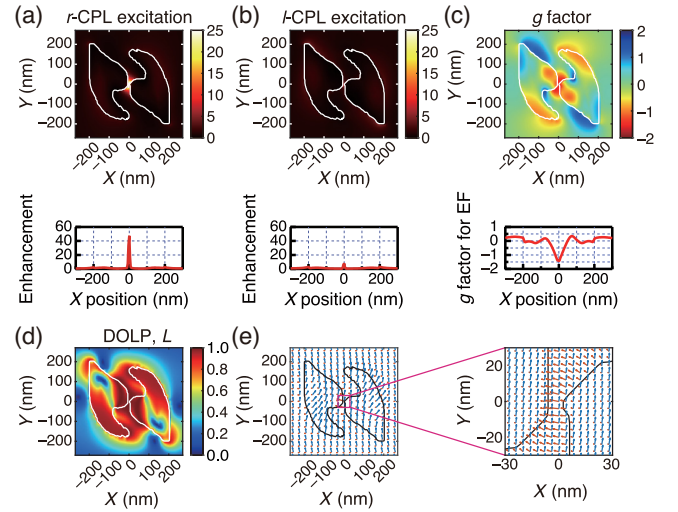


FIG. 2. The near-field properties of the topology-optimized antenna for incident r -CPL. (a),(b) The intensity-enhancement-factor distribution for (a) r -CPL excitation (polarized co-circularly) and (b) l -CPL excitation (polarized cross-circularly), respectively, at the gap plane ($Z = 0$ nm). (c) The dissymmetry factor (g factor) of the field intensity, with a sectional line profile along $Y = 0$ nm. (d) The degree of linear polarization (DOLP), L . (e) A vector representation of the electric (red) and magnetic (blue) fields and their magnified plot.

a vortexlike structure in 3D. Sections of the nanostructure are presented at 20-nm intervals in the z direction in Fig. 1(c) [for the pixel-by-pixel z section, see Fig. S1(a) in the Supplemental Material [24]]. A twisted pattern with two bent arms formed on the topmost surface ($z = 50$ nm), exhibiting twofold symmetry. The observed degree of symmetry was lower than that of gammadions (i.e., four-fold symmetry), which have previously been proposed for plasmonic planar chiral metamaterials [10,25], indicating that intuitive designs may have an excessive degree of symmetry for differentiating the handedness of CPL. An aperture appeared at the center of the top surface. Moving downward in the z direction, a transition occurred; the hole expanded, dividing the structure into two parts at $z = 30$ nm with a nanogap. The gap narrowed to 12 nm at $z = 0$ nm, where the near-field intensity was maximum [Fig. 2(a)]. The formation of the two tips at the nanogap links to the objective function, which prefers the field to be linearly polarized [24]. Two tips can retain a linearly polarized field but cannot retain CPL; at least three tips are required to receive the spin angular momentum (SAM) of light [26]. This explains the absence of two tips on the topmost surface where incoming CPL impinges. The base of the structure forms a diagonal winglike shape at $z = -50$ nm.

In Fig. 1(d), we show the nanostructure obtained using the same configuration but with the opposite handedness of the incident circular polarization, namely, left-handed

CPL (*l*-CPL). The structure was mirror reflected from that obtained for *r*-CPL [Fig. 1(c)]. The mirror asymmetry observed in these examples highlights the sensitivity of TO to the chirality of light.

Near-field properties of the gap mode: Field enhancement. The near-field properties of the nanogap antenna optimized for *r*-CPL [Fig. 1(c)] have been investigated using finite-differential time-domain (FDTD) simulations. In Figs. 2(a) and 2(b), we compare the near-field intensity distributions excited by *r*-CPL (polarized co-circularly) and *l*-CPL (polarized cross-circularly) in the gap plane at $z = 0$ nm. The color bars represent the enhancement factor, the ratio of the field intensity calculated with the antenna to that without the antenna. The highest enhancement occurred at the gap position, with a full width at half maximum (FWHM) of about 7.5 nm, demonstrating the light confinement at the subwavelength scale. TiO_2 , a high-index dielectric at 532-nm wavelength ($n = 2.512 + 0.011i$) [24], causes local field enhancement on the surface. The enhancement factors were 47.4 and 8.0 for co-circularly and cross-circularly polarized excitation, respectively, indicating that the gap antenna optimized for *r*-CPL preferentially couples to the incident *r*-CPL with the same handedness.

To quantify the dissymmetry of the near-field intensity with respect to the handedness of incident CPL, we have adopted the concept of the dissymmetry factor used in CD [1]: $g_{\text{gap}} = 2(I_L - I_R)/(I_L + I_R)$, where I_L and I_R are the gap-mode intensities excited by *l*-CPL and *r*-CPL, respectively. The g factor ranges from -2 and $+2$, similar to the original one. As illustrated in Fig. 2(c), the g factor peaked at -1.42 within the gap, demonstrating high chiral selectivity for the gap-mode intensity depending on the handedness of the incident CPL, despite the entire structure size ($400 \times 400 \times 100 \text{ nm}^3$) being less than a wavelength.

Near-field properties of the gap mode: Polarization states. To describe the near-field polarization, we have calculated a dimensionless linear polarization parameter L using generalized Stokes parameters [24]. This parameter measures the degree of linear polarization (DOLP), where $L = 1$ indicates pure linear polarization and $L = 0$ signifies circular polarization [27]. The calculated L distribution reveals linear polarization within the gap [Fig. 2(d)], with the electric field vector oriented across the nanogap [Fig. 2(e) and Video S9 in the Supplemental Material [24]]. The results show that the topology-designed chiral-nanogap antenna, with its specific geometry, is capable of converting far-field CPL of a specific handedness to the linearly polarized near field with a subwavelength mode volume.

Optical-chirality confinement and amplification at the nanoscale with a topology-designed chiral-gap antenna. The optical chirality, expressed as $\chi = -\omega/$

$(2c^2)\text{Im}(\mathbf{E}^* \cdot \mathbf{H}) = -\omega/(2c^2)|\mathbf{E}||\mathbf{H}|\cos(\delta_{\mathbf{E},\mathbf{H}})$ for a monochromatic wave, is a physical quantity related to the enantioselective excitation of a chiral molecule when exposed to electromagnetic fields [28,29]. Here, $\mathbf{E}$ and $\mathbf{H}$ are the complex time-harmonic electric and magnetic fields, respectively; ω is the angular frequency of the light; c is the speed of light in free space; and $\delta_{\mathbf{E},\mathbf{H}}$ denotes the angle between the $i\mathbf{E}$ and the $\mathbf{H}$ vectors. The amplitude of the chirality is maximized for the fields $i\mathbf{E}$ and $\mathbf{H}$ aligned in parallel or antiparallel with the relative phase shift, satisfying $\cos(\delta_{\mathbf{E},\mathbf{H}}) = \pm 1$ [30]. For propagating plane waves, CPL provides an optimal polarization state, as expressed by $\chi_{r\text{-CPL}} = -\epsilon_0\omega/(2c)E_0^2$, where E_0 is the electric field amplitude [30,31]. Attempts have been made to align local near fields in phase-shifted yet parallel components of linearly polarized $i\mathbf{E}$ and $\mathbf{H}$, via two-dimensional chiral arrangements of achiral metallic nanoslit antennas [31] and achiral dimers of dielectric nanoparticles [32].

Here, we have examined counterpropagating *r*-CPL as the excitation scheme, as shown in Fig. 3(a). This configuration forms a “polarization standing wave” (PSW) [33], where the $\mathbf{E}$ and $\mathbf{H}$ fields are parallel, $\pi/2$ shifted, and linearly polarized, with the azimuthal orientation varying spirally along the propagation axis z . In Ref. [28], Tang *et al.* have analytically examined the optical chirality of PSW for propagating plane waves; this work forms the near-field counterpart using TO.

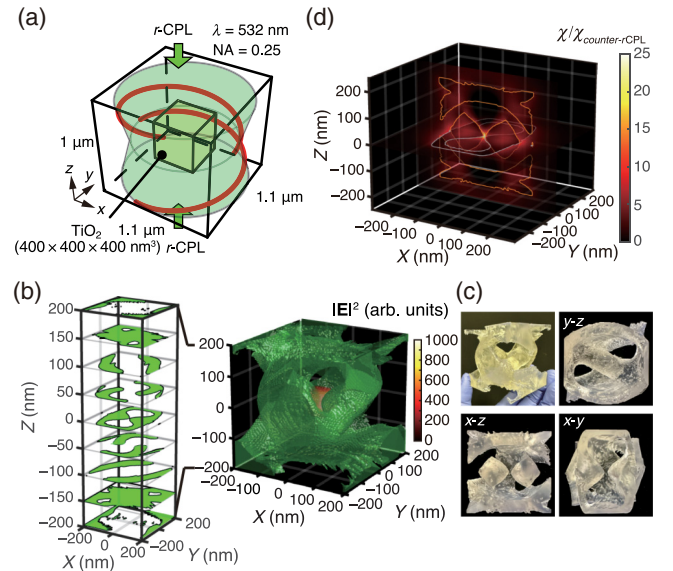


FIG. 3. The topology-designed gap antenna for counterpropagating *r*-CPL. (a) A schematic of the boundary conditions. (b) The 3D model of the antenna with Z-sliced sections, overlaid with the electric field intensity distribution. (c) A 3D-printed mock-up of the chiral-gap antenna at 140 000:1 scale. (d) The optical-chirality density, χ , normalized to structure-free, displayed at the $Z = 0 \text{ nm}$ and $Y = 0 \text{ nm}$ planes under counterpropagating *r*-CPL excitation.

The resultant structure is depicted in Fig. 3(b) as a 3D-rendered model. In Fig. 3(c), we display images of a scaled mock-up at a 140 000:1 ratio, created using a commercial 3D printer, demonstrating its free-standing capability without any checkerboard-pattern artifacts [34]. A complete set of z sections is shown in Fig. S3 in the Supplemental Material [24] and a perspective view of the nanostructure is presented in Videos S10 and S11 in the Supplemental Material [24]. An 8-nm-wide nanogap was created at the $z = 0$ nm plane, with a helical winding structure observed above and below the gap plane, forming a mirror image of its counterpart across the gap plane. The electric field was localized and enhanced at the gap center. The enhancement factors of the electric field intensity calculated for the r -CPL and l -CPL excitations, g_{gap} , and the polarization state near the gap, are summarized in Fig. S4 in the Supplemental Material [24]. The enhancement factor peaked at 1250 for the same counterpropagating r -CPL without the structure. The intensity profile shows an FWHM of 8.2 nm, demonstrating a subwavelength mode volume. Videos S12 and S13 in the Supplemental Material [24] feature optical near fields propagating on the gap antenna, involving r -CPL (co-circular) excitation and cross-circular l -CPL excitation, respectively. The extremum of g_{gap} reached as low as -1.70 within the gap [Fig. S4(c) in the Supplemental Material [24]], indicating an even greater dissymmetry than the structure optimized for a single r -CPL [Fig. 2(c)]. In Figs. S4(e) and Video S14 in the Supplemental Material [24], we show electric and magnetic field vectors aligned in parallel within the gap, a preferable condition for enhancing optical chirality. The distribution of optical chirality for counterpropagating r -CPL excitation is displayed in Fig. 3(d). The enhancement factor reached a maximum of 24.5 in the middle of the gap, with a spatial width of 10.4 nm (FWHM) [Fig. S5(a) in the Supplemental Material [24]], demonstrating the chiral field with a subwavelength mode volume. The optical chirality of a counterpropagating r -CPL in free space has a constant value throughout space and is double that of a single CPL with the same amplitude [28]. Consequently, the structure provides an approximately 50-fold increase in the optical-chirality density within the nanogap compared with that of CPL propagating in free space. As shown in Fig. S5(b) in the Supplemental Material [24], $i\mathbf{E}$ and $\mathbf{H}$ also remained in phase within the gap ($\cos(\delta_{i\mathbf{E},\mathbf{H}}) = 0.98$).

The chirality enhancement shown in Fig. 3(d) exhibited only positive values across the entire structure surface. This behavior contrasts with previously studied plasmonic systems [13,35] and dielectric Mie particles utilizing magnetic multipolar resonances [36], where regions of positive and negative chirality enhancement alternate on the surface, thereby reducing the overall global chirality enhancement. The consistent polarity within the chiral mode volume facilitates its spatial overlap with molecules, making it advantageous for many applications.

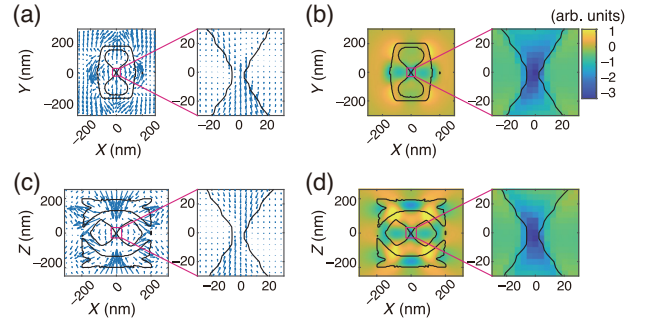


FIG. 4. (a) The SAM density vector $\mathbf{s}$ in the X - Y plane at $Z = 0$ nm and (b) the corresponding SAM divergence $\nabla \cdot \mathbf{s}$. (c), (d) The same as (a), (b) but for the X - Z plane at $Y = 0$ nm.

To gain a deeper insight into the unipolarity in chirality, we have visualized the chirality flux in Fig. 4(a). This flux relates to the optical-chirality density through the continuity equation, expressed as $d\chi/dt + \nabla \cdot \mathbf{F} = 0$, where $\mathbf{F} = (\omega/4)\text{Im}(\epsilon_0 \mathbf{E}^* \times \mathbf{E} + \mu_0 \mathbf{H}^* \times \mathbf{H})$ is the optical-chirality flux. Notably, $\mathbf{F} = (\omega/4)\text{Im}(\epsilon_0 \mathbf{E}^* \times \mathbf{E} + \mu_0 \mathbf{H}^* \times \mathbf{H})$ is proportionally related to the SAM density by a factor of ω^2 [24,37,38]. In the vector plot, the SAM density converges inward due to the gap-antenna structure. With counterpropagating r -CPLs having opposite signs, the SAM flow cancels out at the gap position, creating a distinct negative divergence (sink) of the SAM [Figs. 4(b) and 4(d)]. This sink results in an increase in the local density of optical chirality. These analyses provide an insightful picture in which the unipolarity of the localized chirality arises from the intrinsic chirality of the counterpropagating CPL, fed to the antenna and then “squeezed” into the nanogap, as suggested by the conservation of optical chirality.

Coupled radiation of the point dipole with a 3D chiral-gap antenna. Finally, given that the gap field is linearly polarized [Fig. S4(e) in the Supplemental Material [24]], we have hypothesized that a linear point dipole embedded in the chiral-gap antenna radiates CPL, as a consequence of the reciprocity of the antenna. We have verified this for the gap antenna optimized for counterpropagating r -CPL [Fig. 3(b)]. As depicted in Fig. 5(a), a point dipole was placed at the center of the gap, with the orientation aligned to the gap. In Fig. 5(b), we show the electric field intensities radiating from the dipole with and without the gap antenna. Stronger radiation was seen with the gap antenna, indicating efficient coupling from the near to the far field.

In Fig. 5(c), the left panel presents the phase angles of the E_x and E_y components along the z axis. In the forward ($+z$) direction, E_x leads E_y by a nearly constant phase difference, whereas in the backward ($-z$) direction, E_x lags behind E_y . The right panel shows that the phase difference asymptotically approaches $\pm\pi/2$ in the $\pm z$ direction, indicating that the resultant field vector rotates counterclockwise, or with left-handed rotation. The

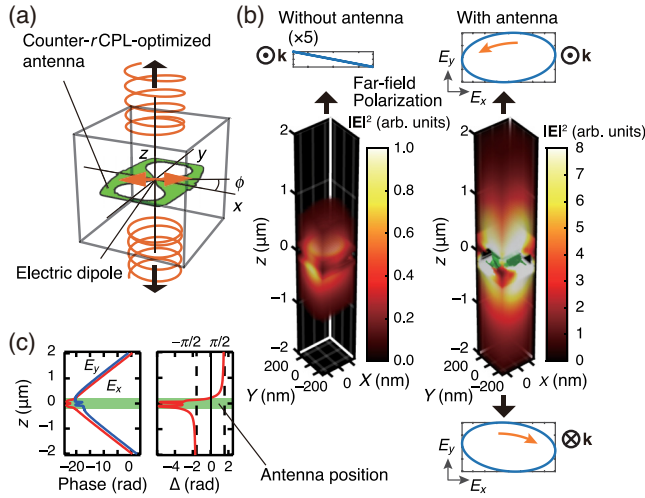


FIG. 5. The coupled radiation of a point dipole with the topology-optimized 3D gap antenna. (a) The simulation model. (b) The electric field intensity of the dipole radiation, with polarization ellipses in the far field normal to the X - Y plane. (c) The phase angle of the E_x (red) and E_y (blue) components along the Z axis (left panel) and the phase difference (right panel).

consistent handedness in both the forward and backward directions is a distinctive characteristic that planar achiral structures, such as gammadions, cannot reproduce; in such structures, the handedness reverses in opposite scattering directions. Consequently, as shown in Fig. 5(b) (right panel), the far-field vector traces an ellipsoidal trajectory.

The polarization state can be decomposed into two orthogonal circular-polarization states, E_L and E_R . The dissymmetry factor of circularly polarized radiation in the far field, $g_{\text{scat}} = 2(I_{\text{scat},L} - I_{\text{scat},R}) / (I_{\text{scat},L} + I_{\text{scat},R})$, where $I_{\text{scat},L}$ and $I_{\text{scat},R}$ are the squared amplitudes of E_L and E_R , respectively, was calculated. The resultant g value was 1.6, demonstrating a significantly high dissymmetry in the coupled radiation of the linear dipole interacting with the chiral cavity, that has a nanoscale mode volume.

Conclusions. We have explored 3D dielectric chiral-nanogap antennas for nanoscale-chirality confinement and enhancement. Our approach has utilized 3D TO to fully explore complex design spaces that are otherwise constrained by human intuition. The resulting 3D chiral-nanogap structure exhibits distinct chiroptical phenomena arising from polarization mode conversion at the nanoscale mode volume. By incorporating PSW with 3D TO, we have demonstrated a 3D chiral-nanogap structure that localizes the chiral field within a nanoscale mode volume with unipolarity, which can inversely radiate a chiral field from a point dipole interacting with the chiral-gap mode. Furthermore, control of chirality flux through a topology-designed 3D structure has been demonstrated, offering a perspective on chiral structural design for enhancement.

The present study has considered only linear polarization for the localized gap mode as the simplest case. Extending this approach to modes with nonzero spin and orbital angular momentum (OAM) represents an intriguing avenue for future research. Higher-order OAM, associated with the larger beam diameter of the Laguerre-Gaussian propagating wave [39], benefits particularly from its nanoscale confinement [26], enabling efficient OAM transfer to nanomaterials. This approach offers a promising pathway to enhance nanoscale interactions mediated by the angular momentum of light.

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