

Article

Local Circular Dichroism Induced by Near-Field Interference in Asymmetric Plasmonic Nanocylinder Trimer

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Abstract

Surface plasmons are collective oscillations of electrons that can confine light within extremely tiny nanoscale spaces, acting to enhance the spectral signals (such as Raman and circular dichroism) of molecules. Borrowed from circular dichroism in chemistry, plasmonic local circular dichroism (CD) refers to the difference in electric field enhancement excited by left-handed (LCP) and right-handed (RCP) circularly polarized light in a nanogap. Nanosphere trimers have been employed to realize local CD. However, spherical nanoparticles exhibit a small gap area and poor structural stability. In this article, we propose asymmetric nanocylinder trimers on SiO₂/Si substrate to realize a strong local CD signal. In contrast to nanospheres, nanocylinder sidewalls increase effective gap areas and can be obtained by nanofabrication. We find that the asymmetric nanocylinder trimer achieves a local CD parameter 0.94 under the excitation of 782 nm. This strong local CD within the gaps mainly originates from the near-field interference between different modes generated by circularly polarized light. Furthermore, the gap distance and symmetry have a significant influence on the local CD. A giant local CD can be obtained for small gaps (1 nm) and right-angle trimers. These results provide a physical basis for the development of plasmonic CD and chirality, and they are of interest for the field of nanosensors, nanoantennas, solar energy conversion, and polarization-dependent photochemistry.

Keywords: surface plasmons; nanocylinder; trimer; near-field enhancement; circular dichroism; chirality

1. Introduction

Localized surface plasmon resonances (LSPs) are collective oscillations of conduction electrons supported by noble metal nanostructures [1]. They can confine light waves to the subwavelength nanoscale [2], laying the physical foundation for the development of miniaturized plasmonic devices, such as surface-enhanced spectroscopy [3–7], nanoantennas [8–13], quantum plasmonics [14–16], nanosensors [17–19], photocatalysis [20,21], and solar cells [22–24]. Plasmonic response (e.g., near-field enhancement and far-field scattering) depends on the size, shape, spatial arrangement and materials of the nanostructures [23,25–35]. The optical properties of metal nanoparticles also rely on the polarizations of incident lights. Plasmonic circular dichroism (CD) generally refers to the difference in reflection and transmission between left-handed (LCP) and right-handed (RCP) circularly polarized light [36–38]. This concept of circular dichroism is adopted from chemistry, which means the differential



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absorption of left-handed and right-handed circularly polarized light by chiral molecules. Plasmonic CD is typically achieved in asymmetric nanostructures, including monomers (e.g., helical structures) and multimers (asymmetric arrangements) [39,40].

The plasmonic CD is usually far-field CD [41], while plasmonic local CD is manifested as the differential electric field enhancement under left-handed and right-handed circularly polarized excitation within the nanogaps [42–44]. The local CD originates from the near-field interference between the different plasmonic modes excited by two linearly polarized components that constitute circularly polarized light [45]. Local CD can emerge in monomer (e.g., helical metal nanoparticles), dimer (e.g., twisted nanorods) or trimer (e.g., asymmetric nanoparticles) [46]. For nanoparticle trimers, local field intensity in right-angle trimers excited by RCP is almost one thousand times larger than the LCP case [43]. However, the inter-gap area of many studies is extremely small (e.g., nanospheres), which results in a limited functional area for local CD. Moreover, nanospheres exhibit poor structural stability due to the small contact area between nanospheres and the substrate.

In this study, we propose an asymmetric circular cylinder [47–56] trimer with an enlarged gap area, which can be fabricated using techniques such as electron beam lithography [49,53,54] and the self-assembly technique [35,52,57]. The electric field enhancement in the nanogap is maximized when the incident polarization is parallel to the dimer axis. We investigate the electric field enhancement of nanocylinder trimers under circularly polarized light excitation. Plasmonic local CD parameters are calculated as the offset angle of nanocylinders [58] varied from 0° (linear) to 120° (equilateral). Local CD depends on structural symmetry. Linear and equilateral triangular trimers exhibit no local CD signals due to their high symmetry, whereas the asymmetric L-shaped (90°) structure yields the local CD parameter ρ of up to 0.94 at 782 nm due to constructive interference of two modes excited by CPL. The electric field enhancement is smaller for larger gaps, and the local CD also shows a dramatic decline as the interparticle gap width increases from 1 nm to 50 nm. The weakening of interparticle plasmonic coupling disrupts the near-field interference conditions required for local CD generation. This study realizes giant local CD via asymmetric nanocylinder trimers and provides theoretical support for the development of plasmonic CD, chirality, and optical activity (OA) [59].

2. Methods

All simulations were performed using the finite-difference time-domain (FDTD) method. The wavelength-dependent complex permittivity of the silver nanocylinders was obtained from the experimental data of Johnson and Christy [60]. The dispersive optical constants of Si and SiO₂ were obtained from the Palik database [61]. These tabulated material data were imported into the FDTD material library and fitted over the investigated wavelength range of 500–1000 nm.

To simulate the light scattering process in open space, the boundary conditions in the x , y , and z directions of the simulation domain are set as perfectly matched layers (PML) to fully absorb outgoing waves and eliminate boundary reflection interference. A non-uniform meshing technique is adopted. To precisely capture the highly localized electric field gradients inside the nanogap, an ultrafine mesh with a resolution smaller than 0.5 nm is applied in the mesh override region around the nanogap.

Light source configuration. The total-field/scattered-field (TFSF) source ($\lambda = 500\text{--}1000$ nm) is employed to divide the simulation domain into total-field and scattered-field regions, which significantly improves the signal-to-noise ratio in scattering characterization. To investigate plasmonic local CD, circularly polarized light (CPL) is generated by superposing two linearly polarized waves with equal amplitude, orthogonal polarization directions, and a phase difference of $\pm\pi/2$. The incident electric field vector E is defined as follows:

Left-handed circularly polarized (LCP) light:

$$E_{\text{LCP}} = E_0 \left(\hat{x} + i\hat{y} \right) e^{ikz} \quad (1)$$

With the phase of the y -component leading that of the x -component by 90° .

Right-handed circularly polarized (RCP) light:

$$E_{\text{RCP}} = E_0 \left(\hat{x} - i\hat{y} \right) e^{ikz} \quad (2)$$

With the phase of the y -component lagging that of the x -component by 90° .

Normalized scattering efficiency Q_{scat} . The scattering cross-section σ_{scat} is the ratio of the total scattered power P_{scat} (power monitor placed in the scattered-field region) to the incident power I_{inc} . To ensure comparability among different multi-body systems, normalized scattering efficiency Q_{scat} is defined as the ratio of the scattering cross-section σ_{scat} to the total geometric projection area σ_{geo} of all nanocylinders:

$$Q_{\text{scat}} = \frac{P_{\text{scat}}}{I_{\text{inc}} \sigma_{\text{geo}}} \quad (3)$$

where $\sigma_{\text{geo}} = \text{the number of cylinders} \times \pi r^2$, and the nanocylinder radius $r = 50$ nm.

Substrate coupling efficiency f_{sub} . To evaluate the potential for photovoltaic applications, f_{sub} is defined as the ratio of the scattered power coupled into the substrate P_{sub} to the total scattered power P_{scat} . Here, P_{sub} is measured using a power monitor placed in the substrate region:

$$f_{\text{sub}} = \frac{P_{\text{sub}}}{P_{\text{scat}}} \quad (4)$$

Electric field enhancement M . To quantitatively characterize the intensity of hotspots at the nanogap, the electric field enhancement M is defined as the ratio of the squared modulus of the local electric field to the squared modulus of the incident electric field at a specific monitoring point/plane:

$$M = \frac{|E_{\text{loc}}|^2}{|E_{\text{inc}}|^2} \quad (5)$$

Local CD parameter ρ . To quantitatively describe the intensity of local CD, a dimensionless factor ρ is defined as the normalized difference of electric field enhancement M (the point is the gap region center between two sidewalls of cylinders) under LCP and RCP excitation:

$$\rho = \frac{M_{\text{RCP}} - M_{\text{LCP}}}{M_{\text{RCP}} + M_{\text{LCP}}} \quad (6)$$

3. Results

3.1. Plasmonic Response of a Silver Nanocylinder

We investigate the optical response of a single silver nanocylinder placed on a SiO_2/Si substrate (shown in Figure 1a). The aspect ratio (AR) of the circular cylinder is defined as the ratio height H to its diameter D . A 20 nm-thick SiO_2 layer is sandwiched between the metallic cylinder and the underlying Si substrate. The total-field/scattered-field (TFSF) light source is used to irradiate the cylinder vertically from the top air side. By placing power monitors at different positions (see Section 2), the scattering cross-section Q_{scat} and substrate coupling efficiency f_{sub} are calculated separately. The Q_{scat} and f_{sub} of nanocylinders ($\text{AR} = 0.5\text{--}6$, $D = 100$ nm) are shown in Figures 1b and 1c, respectively.

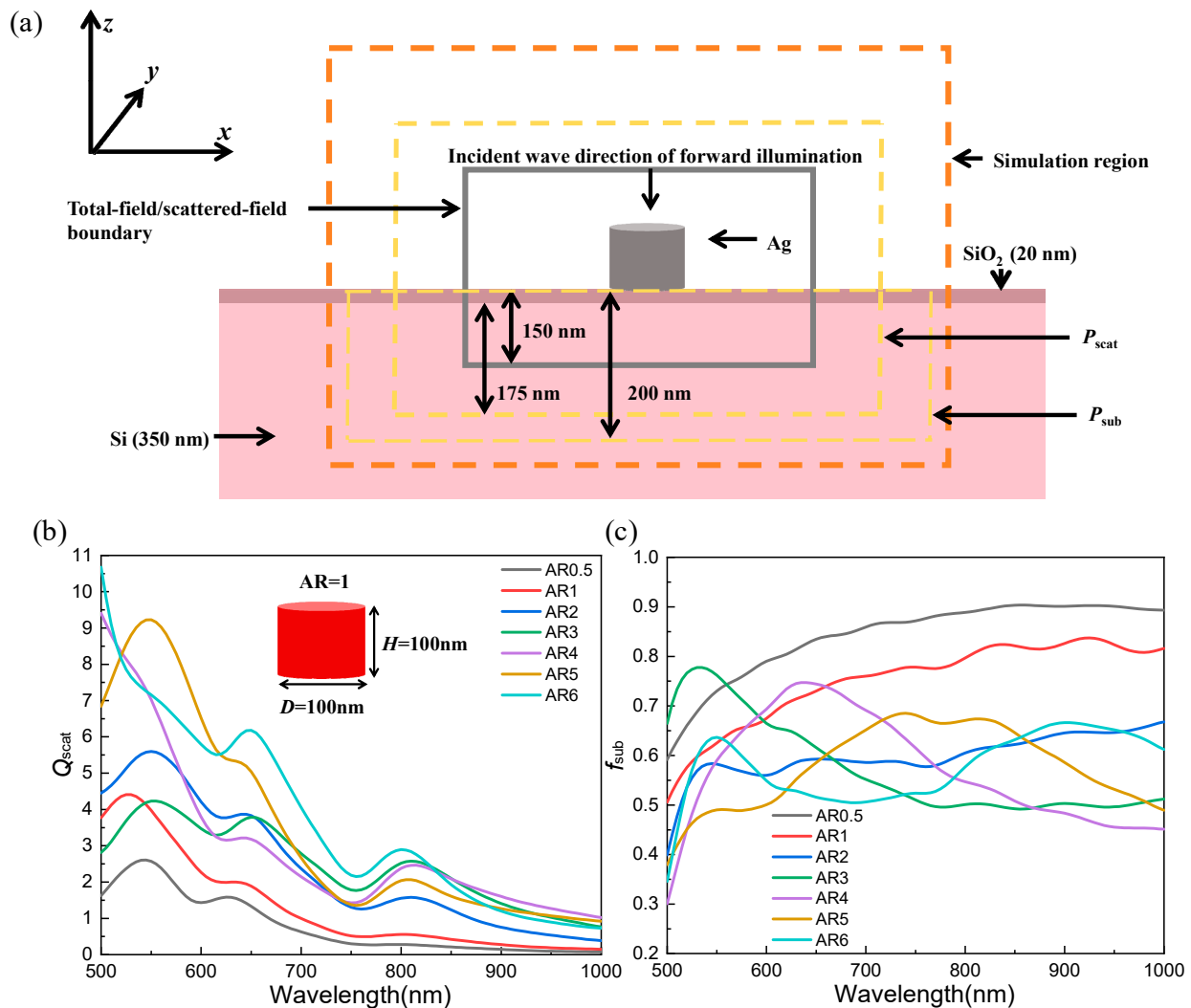


Figure 1. Plasmonic response of a silver nanocylinder. (a) Schematic of the simulation model. (b,c) The scattering cross-section Q_{scat} and substrate coupling efficiency f_{sub} for silver nanocylinders with different aspect ratios (AR = 0.5–6), respectively.

The nanocylinder exhibits a stronger Q_{scat} for larger AR (Figure 1b). However, f_{sub} decreases with the increase of AR (Figure 1c). Although the high AR can enhance the total scattering intensity of the nanocylinder, it restricts the effective coupling of light to the substrate. There is a trade-off between the scattering capacity and coupling efficiency. The nanocylinder of AR = 1 with relatively large Q_{scat} and f_{sub} simultaneously is selected as the basic unit for the subsequent cylinder dimers and trimers.

3.2. Polarization-Dependent Response of Nanocylinder Dimers

We then investigate the scattering cross-section Q_{scat} of the nanocylinder dimer (shown in Figure 2a) for the different gap distance d and incident light polarization angle θ . We select polarization parallel ($\theta = 0^\circ$) and perpendicular ($\theta = 90^\circ$) to the dimer axis (x -axis) for $d = 1$ –200 nm. The calculated results are presented in Figures 2b and 2c, respectively.

For parallel polarization excitation (Figure 2b), the scattering peak position blueshifts from 864 nm ($d = 1$ nm) to 742 nm ($d = 6$ nm). The weak Coulomb attraction between the adjacent surface charges of the dimer enhances the effective restoring force, leading to the increased energy of the longitudinal oscillating mode. When d is larger than ~6 nm, the blueshift will saturate because of weak coupling. For $d = 200$ nm, plasmonic couplings between two nanocylinders nearly disappear, and the scattering is similar to the case of

monomer. The plasmonic response of the silver nanocylinder dimer exhibits anisotropic characteristics. In contrast, the position of the scattering peak remains nearly unchanged, with varying gaps illuminated by perpendicular polarized light, indicating that no obvious interaction occurs between these two nanocylinders. This confirms that the occurrence of strong coupling requires both a parallel polarization direction and an ultra-small gap.

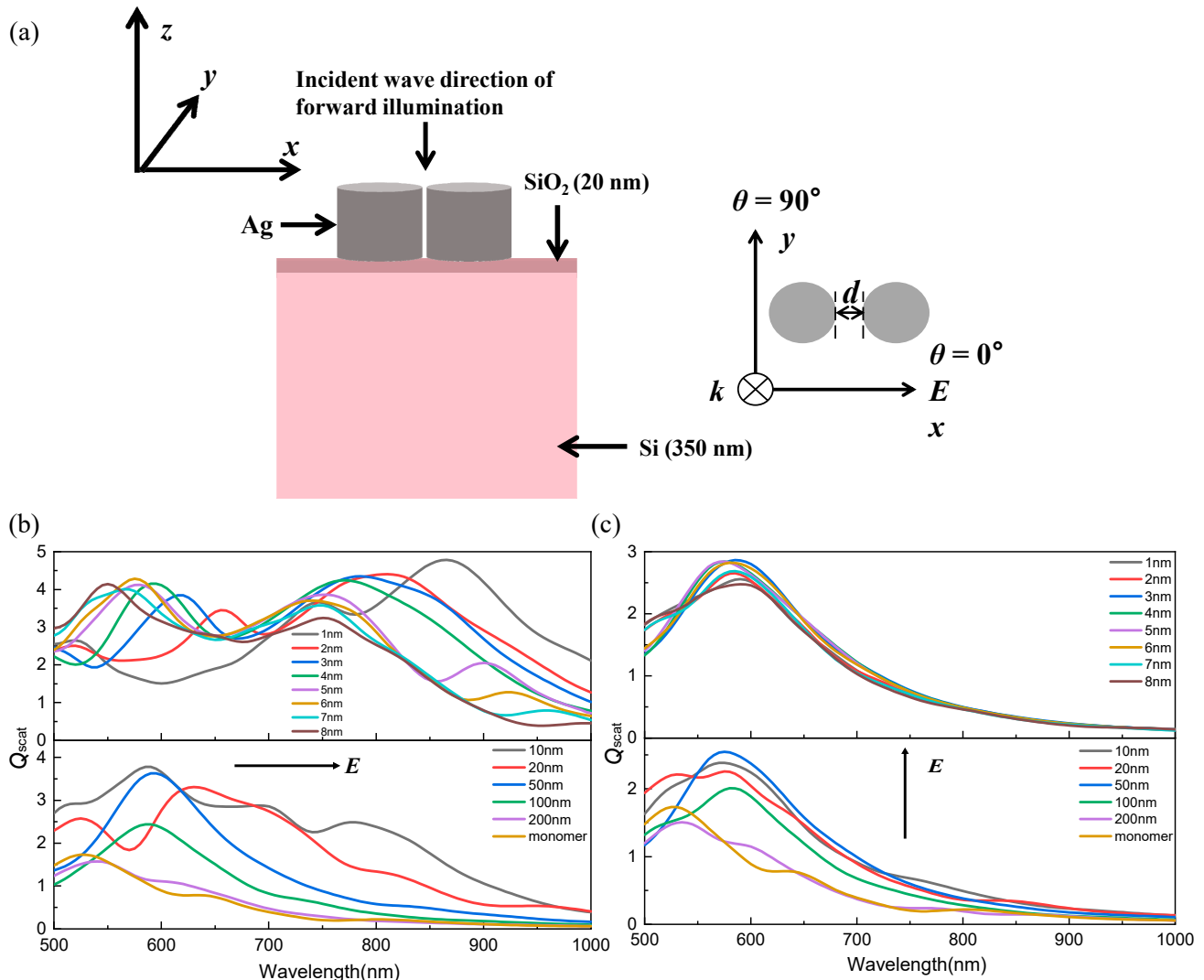


Figure 2. Scatterings of silver nanocylinder dimer under different polarizations and gap distances. (a) Schematic of the nanocylinder dimer. (b,c) Simulated scattering cross-section Q_{scat} for gap distances $d = 1$ –200 nm with the incident light polarization parallel ($\theta = 0^\circ$) or perpendicular ($\theta = 90^\circ$) to the dimer axis (x -axis).

As the polarization deviates from the dimer axis ($d = 1$ nm), the intensity of the resonance peak (~ 864 nm) of Q_{scat} decays gradually (Figure 3a). The electric field enhancement $|E|/|E_0|$ (the ratio of the local electric field to the incident electric field) distributions in x – z at the resonant wavelength of 864 nm for different polarizations (0 – 90°) are shown in Figure 3b–h. We adopt $|E|/|E_0|$ rather than $|E|^2/|E_0|^2$, to avoid excessively rapid growth of electric field value that would obscure the geometric edges of the nanocylinders. The electric field in the gap reaches its maximum (so-called hotspot) for parallel polarization (0°). This is because plasmonic coupling reaches its maximum due to the longitudinal bonding mode, which excites strong capacitive coupling at the gap and tightly confines electromagnetic energy within the gap region.

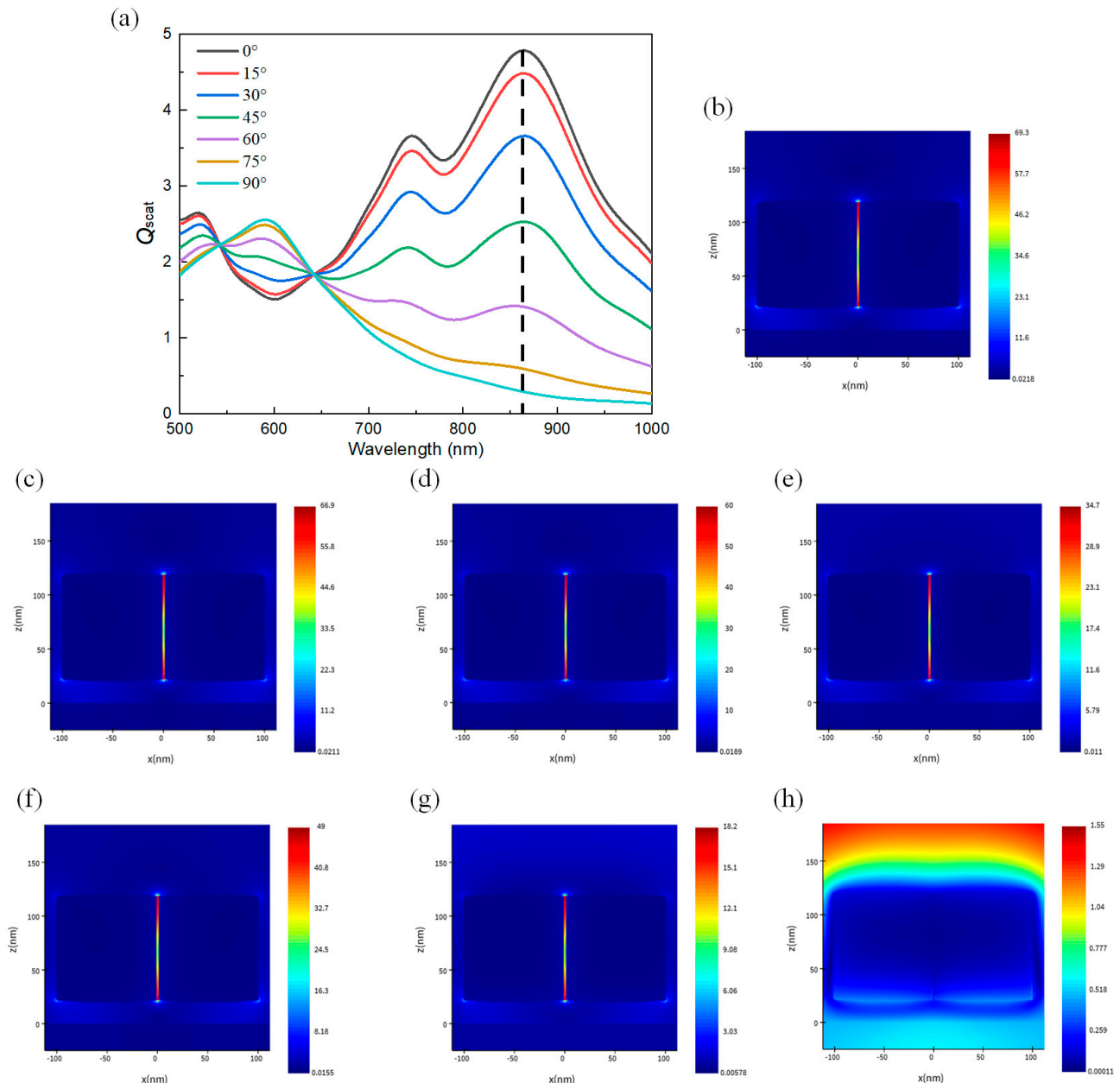


Figure 3. Polarization-dependent response of the nanocylinder dimer with $d = 1$ nm. (a) Far-field Q_{scat} for different incident polarization angle θ (0 – 90°). (b–h) Electric field enhancement ($|E|/|E_0|$) distributions in the x – z plane at 864 nm for different polarization angle θ .

3.3. Local CD of Near-Field Hotspot in Asymmetric Trimers

We select an asymmetric L-shaped trimer (Figure 4a) to study its polarization-dependent electric field enhancement. Here, d_{12} and d_{23} denote the minimum surface-to-surface gap distances between the 1st–2nd and 2nd–3rd nanocylinders, respectively. In Figures 4 and 5, two gap distances are $d_{12} = d_{23} = d = 1$ nm. As shown in Figure 4b, the electric field enhancement $M = |E|^2/|E_0|^2$ is calculated at the gap region center (marked with a red dot) excited by 0° (red curve) or 90° (green curve) polarizations. The 90° polarization yields a pronounced peak at ~ 660 nm with a maximum $M = 1.24 \times 10^4$. The electric field enhancement at the gap region center for 0° polarization is not zero ($M = 0.64 \times 10^4$ at ~ 852 nm); this is because the 3rd nanocylinder affects the near-field coupling of 1st–2nd dimer. These two curves have an intersection at 782 nm, where the electric field enhancement values for these two orthogonal polarizations are equal

($M = 0.43 \times 10^4$). The working wavelength 782 nm is not universal and can be tuned through the geometrical dimensions, gap distances, and dielectric environment of the trimer. The near-field distributions (logarithmic-scale $\log_{10}(M)$) in the x - y plane at 782 nm for two polarizations are shown in Figure 4c.

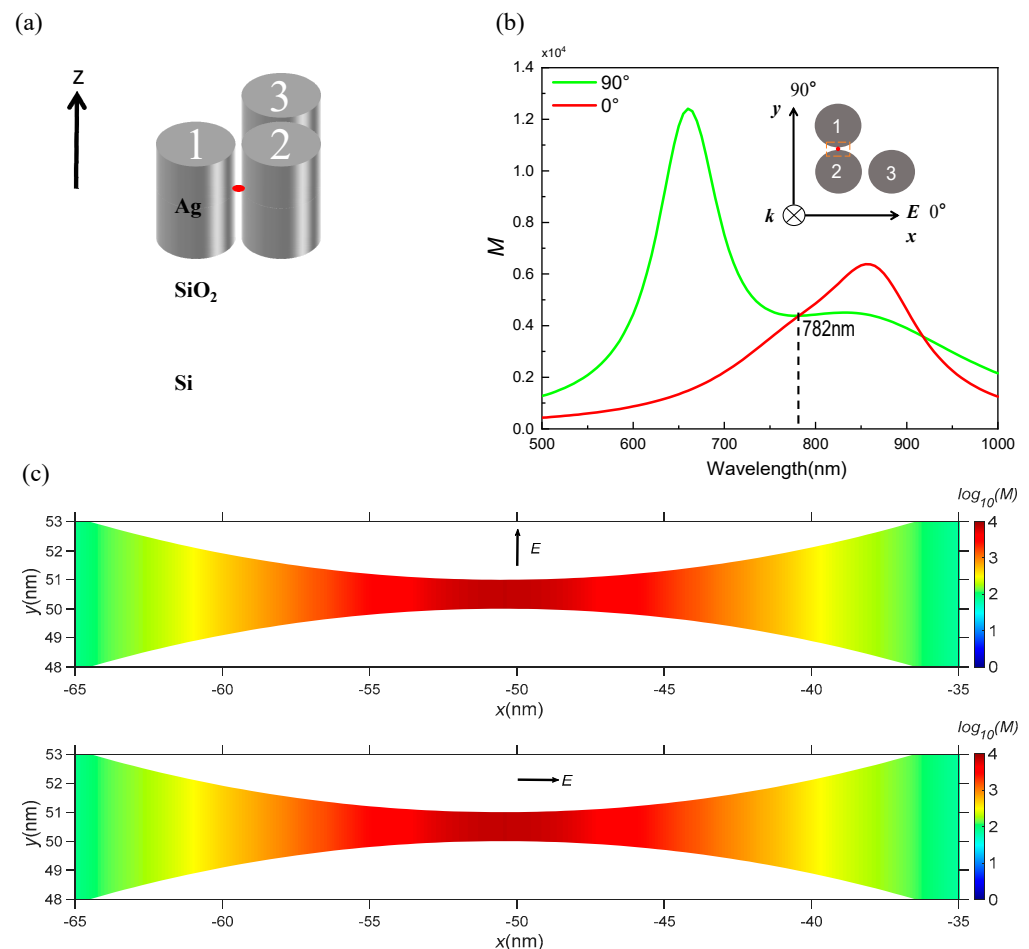


Figure 4. Electric field enhancement response of the L-shaped trimer under linearly polarized excitation. (a) Structure of the L-shaped trimer. (b) Electric field enhancement M at the gap center as a function of wavelength for two orthogonal incident polarizations. The red dots indicate the locations at which M are evaluated. (c) Near-field distributions around 1st–2nd gap in the x - y plane correspond to 0° and 90° polarizations at excitation wavelength 782 nm.

We then excite the trimer with LCP and RCP light to investigate its local CD response. Figure 5a presents electric field enhancement M and local CD parameter ρ (see Section 2) under CPL excitation. A distinct handedness-dependent field enhancement is observed. For LCP excitation (green curve), electric field enhancement stays near zero in most regions with only a weak broad peak ($M = 0.18 \times 10^4$) at ~ 653 nm and drops below 0.04×10^4 at 782 nm. For RCP excitation (red curve), M peaks at ~ 672 nm (1.45×10^4). The local CD parameter ρ (black curve) reaches its maximum value of 0.94 at 782 nm (M remains as high as 1.20×10^4). Figure 5b shows the near-field distributions in the 1st–2nd cylinder gap at 782 nm with LCP (top panel) and RCP (bottom panel) illumination. RCP illumination gives rise to strong constructive interference in the gap with $\log_{10}(M) = 2.5$ – 4.2 , while LCP illumination leads to destructive interference and field attenuation with $\log_{10}(M) = 1.5$ – 2.6 .

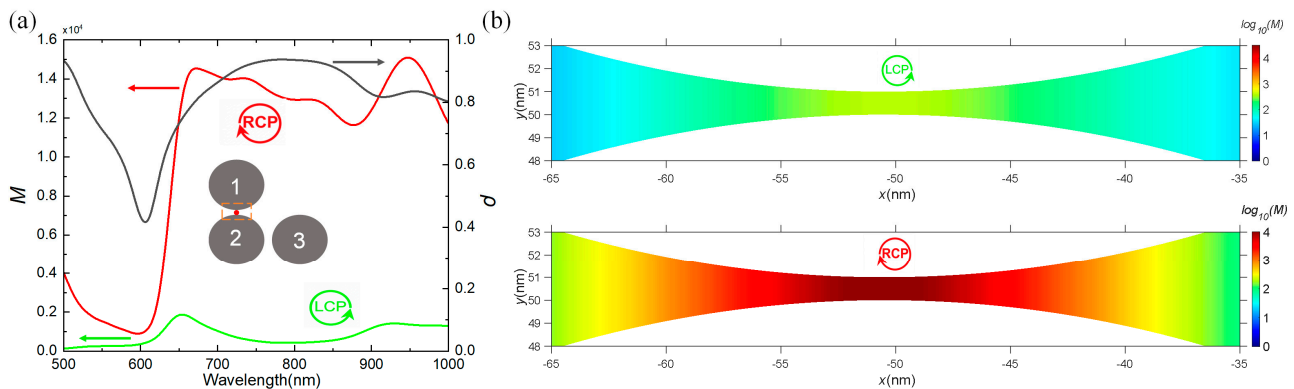


Figure 5. Local CD and electric field enhancement distribution of L-shaped trimer under circularly polarized excitation. (a) Electric field enhancement M under LCP (green curve) and RCP (red curve) excitation, respectively. The local CD parameter ρ (black curve, to the right axis) reaches its maximum value of 0.94 at 782 nm. (b) Electric field enhancement $\log_{10}(M)$ in 1st–2nd cylinder gap under 782 nm illumination with different handedness.

The gap distance influences plasmonic coupling, electric-field enhancement, and the relationship of two modes excited by CPL. Let $g_{\parallel} = |g_{\parallel}| e^{i\Delta_{\parallel}}$ and $g_{\perp} = |g_{\perp}| e^{i\Delta_{\perp}}$ denote the complex field-enhancement coefficients induced by the two orthogonal components of CPL. The corresponding field enhancements are $M_{\parallel} = |g_{\parallel}|^2$ and $M_{\perp} = |g_{\perp}|^2$, while the enhancement under RCP/LCP excitation can be expressed as

$$M_{\text{LCP/RCP}} = \frac{1}{2} \left(|g_{\parallel}|^2 + |g_{\perp}|^2 \right) + |g_{\parallel}g_{\perp}| \cos \left[\Delta_{\parallel} - \left(\Delta_{\perp} \pm \frac{\pi}{2} \right) \right] \quad (7)$$

The giant local CD arises primarily from the intrinsic near-field interference of two modes. At an excitation wavelength of 782 nm, $|g_{\parallel}|^2 \approx |g_{\perp}|^2 = 0.43 \times 10^4$, and $\Delta_{\parallel} - \Delta_{\perp} \approx \pi/2$. The electric-field enhancements at the 1st–2nd gap for LCP and RCP excitation are $M_{\text{LCP}} = 4.23 \times 10^2$ and $M_{\text{RCP}} = 1.31 \times 10^4$, respectively. So, the near fields excited by LCP and RCP are destructive interference and constructive interference, respectively. This provides the condition for giant local CD generation (ρ as high as 0.94) in the L-shaped trimer.

4. Discussion

4.1. Influence of Offset Angle ϕ on Local CD

To investigate the influence of structural symmetry on local CD, we systematically compare trimers with different offset angles ϕ (the 3rd nanocylinder offset the axis of the 1st–2nd dimer). Figure 6a shows CD parameter ρ as a function of ϕ . The ϕ varies continuously from 0° (linear trimer) to 120° (equilateral triangular trimer). When ϕ is 0° or 120° , ρ is nearly zero across the entire spectrum due to the symmetry of these two configurations. For other ϕ , the largest local CD signals appear at 805 nm for $\phi = 30^\circ$, 810 nm for $\phi = 60^\circ$, and 782 nm for $\phi = 90^\circ$. Figure 6b shows ρ as a function of ϕ at 782 nm. ρ increases gradually from 0 at $\phi = 0^\circ$ to a maximum of 0.94 at $\phi = 90^\circ$. It then decreases rapidly when ϕ exceeds 90° and finally near zero at $\phi = 120^\circ$.

The local CD response depends on geometric symmetry. The L-shaped trimer is the optimal geometry for giant local CD. In this structure, the right-angle arrangement of the three nanocylinders leads to the destructive/constructive interference of two modes excited by two orthogonal linear polarizations constituting LCP/RCP, thus generating the strongest local CD.

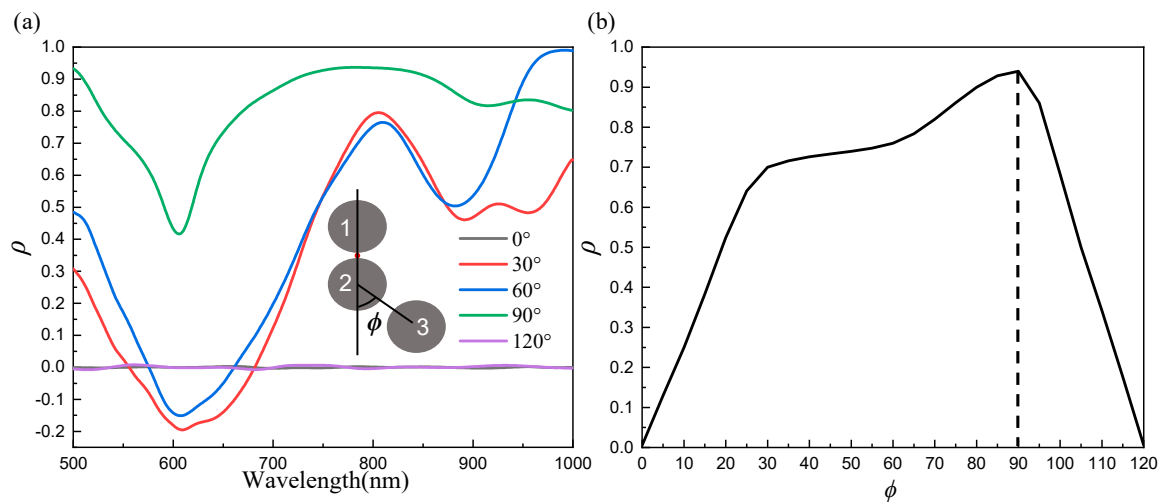


Figure 6. Dependence of local CD parameter ρ on offset angles ϕ . (a) The CD parameter as a function of excitation wavelength for different offset angles ϕ . (b) The CD parameter as a function of ϕ for 782 nm excitation wavelength. The gap distances are 1 nm. The dashed line corresponds to 90° .

4.2. Influence of Gap Distance d on Local CD

Besides the offset angle, the gap distance (1st–2nd, 2nd–3rd) is also a geometric parameter that affects the local CD of the trimer. The local CD parameter ρ as a function of excitation wavelength for different gap distance d in the L-shaped trimer is shown in Figure 7a. The local CD is highly sensitive to gap distance. When d increases from 1 nm to 2 nm or 3 nm, the peak of ρ drops from 0.94 (at 782 nm) to 0.54 (at 731 nm) or 0.47 (at 659 nm). We then calculate ρ as a function of d for the 782 nm excitation wavelength shown in Figure 7b. ρ decreases rapidly as d increases from 1 nm to ~5 nm. For gap distances larger than 10 nm, ρ is less than 0.1 and approaches zero for larger d , and local CD disappears.

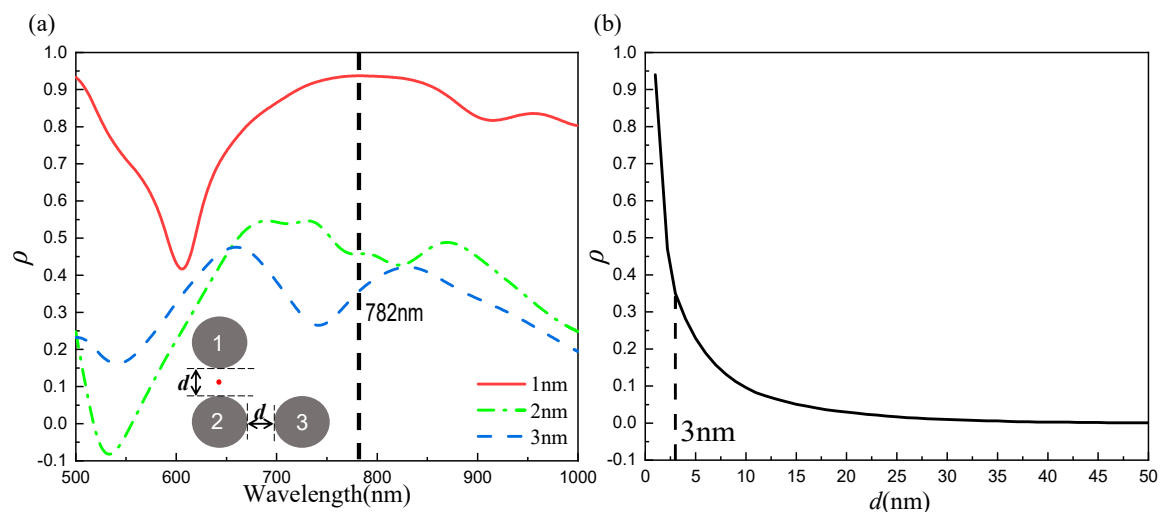


Figure 7. Dependence of local CD parameter ρ on gap distance d . (a) ρ as a function of excitation wavelength for different d . (b) ρ as a function of d for 782 nm excitation wavelength. The offset angle ϕ is 90° .

When d increases from 1 to 3 nm, the local electric-field enhancement decreases rapidly, and the amplitude balance and phase relationship required for the strong constructive and destructive interference of two modes induced by two orthogonal field components constituting CPL is gradually disrupted. The difference between M_{LCP} and M_{RCP} decreases,

leading to a drop in local CD intensity. Therefore, a small gap distance (~ 1 nm) is necessary to generate giant local CD in asymmetric trimer.

When the two gap distances $d_{12} = d_{23}$, the trimer is an achiral structure. If these two gap distances are different, d_{23} might affect local CD between nanocylinders 1st–2nd. To separately investigate the influence of d_{23} , we fix $d_{12} = 1$ nm and only vary d_{23} . The case of $d_{12} = d_{23} = 1$ nm is used as the equal-gap reference, whereas $d_{12} \neq d_{23}$ produces a gap-asymmetric trimer. The local CD parameter ρ as a function of excitation wavelength for different d_{23} is shown in Figure 8a. When d_{23} increases from 1 nm to 2 nm, 3 nm, and 5 nm, the maximum value of ρ decreases from 0.94 to 0.81, 0.63 and 0.43, respectively.

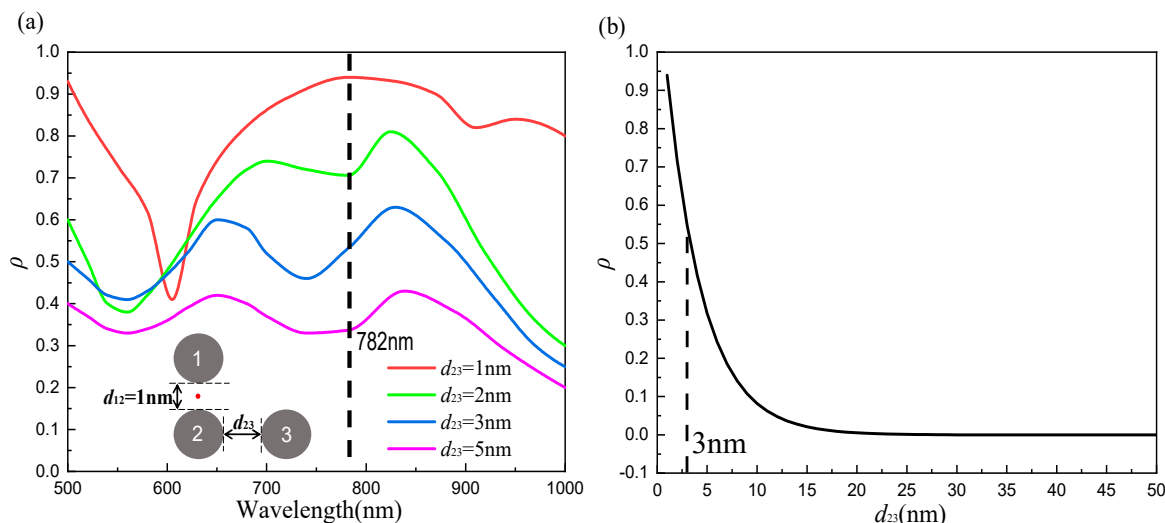


Figure 8. Dependence of the local CD parameter ρ on the gap distance d_{23} , with d_{12} fixed at 1 nm. (a) The local CD parameter ρ as a function of excitation wavelength for different d_{23} . The dashed vertical line indicates an excitation wavelength of 782 nm. (b) The local CD parameter ρ as a function of d_{23} at an excitation wavelength of 782 nm.

We then calculate ρ as a function of d_{23} at the excitation wavelength of 782 nm, as shown in Figure 8b. ρ decreases rapidly as d_{23} increases from 1 nm to approximately 5 nm. When d_{23} increases from 1 nm to 2 nm or 3 nm, and 5 nm, the corresponding ρ values are 0.94, 0.71, 0.53, and 0.34, respectively. For d_{23} larger than approximately 10 nm, ρ becomes less than 0.1 and gradually approaches zero.

The gap distance d_{23} influences the plasmonic coupling between the 3rd nanocylinder and the 1st–2nd nanocylinder dimer, thereby modifying the near-field coupling and local CD within the 1st–2nd nanocylinder gap. Because d_{12} remains fixed at 1 nm, strong near-field enhancement within the 1st–2nd gap is retained. However, increasing d_{23} weakens the interaction of the 3rd nanocylinder with the 1st–2nd dimer and changes the amplitudes and phases of the two plasmonic modes excited by CPL. This gradually disrupts the constructive and destructive interference of the two plasmonic modes and leads to the decrease of local CD in the 1st–2nd gap, which drops with the increase of d_{23} . Compared with Figure 7b, the influence of d_{23} is smaller than that of d_{12} .

4.3. Influence of the SiO₂ Spacer-Layer Thickness t_{SiO_2} on Local CD

To investigate the influence of the Si substrate on local CD, we vary the thickness t_{SiO_2} of the SiO₂ spacer layer between the Si substrate and Ag nanocylinders. Figure 9 shows the local CD parameter ρ evaluated at the center of the 1st–2nd nanocylinder gap for $t_{\text{SiO}_2} = 0, 1, 5, 10, 20$, and 30 nm. The spectra exhibit modest variations in the resonance position and amplitude. The maximum ρ values are approximately 0.89, 0.89, 0.90, 0.92,

0.94, and 0.94 for SiO₂ thicknesses of 0, 1, 5, 10, 20, and 30 nm, respectively. As the SiO₂ thickness increases, the local CD peaks at ~782 nm, rises gradually, and saturates above $t_{\text{SiO}_2} = 20$ nm, where the influence of the Si substrate on local CD weakens. The small differences among the spectra indicate that local CD is mainly governed by the in-plane coupling within the nanocylinder trimer, while the Si substrate provides additional modulation.

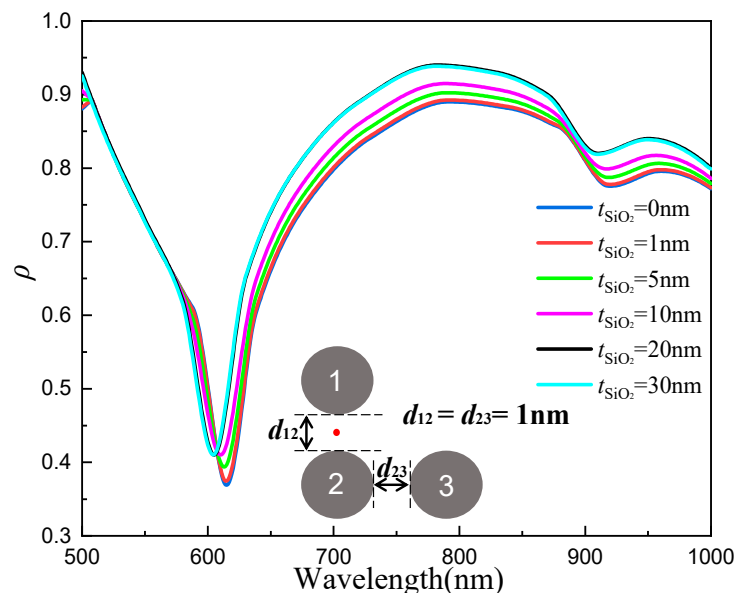


Figure 9. Influence of the SiO₂ spacer-layer thickness t_{SiO_2} on the local CD parameter ρ . The near fields are evaluated at the center of the 1st–2nd nanocylinder gap for $t_{\text{SiO}_2} = 0, 1, 5, 10, 20$, and 30 nm.

4.4. The Local CD of Another Hotspot

The local CD above is concentrated at the hotspot between nanocylinders 1st–2nd, while the hotspot between nanocylinders 2nd–3rd may exhibit a CD response. We evaluate the local CD parameter at the center of the nanocylinders 2nd–3rd for the equal-gap trimer with $d_{12} = d_{23} = 1$ nm. The local CD parameters at the centers of the 1st–2nd and 2nd–3rd nanocylinder gaps are denoted by ρ_{12} and ρ_{23} , respectively. As shown in Figure 10, the two symmetry-related gap-center points exhibit local CD responses with approximately equal magnitudes but opposite signs over the wavelength range investigated.

For this mirror-symmetric configuration, the mirror operation exchanges nanocylinders 1 and 3 and transforms RCP into LCP. Therefore, the calculated local field enhancements at the two symmetry-related monitoring points satisfy $M_{12}^{\text{RCP}} \approx M_{23}^{\text{LCP}}$ and $M_{12}^{\text{LCP}} \approx M_{23}^{\text{RCP}}$, leading to $\rho_{12} \approx -\rho_{23}$. These results demonstrate that the sign of the local CD response depends on the selected observation position. Therefore, the specific hotspot or monitoring position should be clearly identified when discussing the local CD response of the trimer.

An individual hotspot can exhibit pronounced local CD even though the trimer is globally achiral. The equal-gap L-shaped trimer possesses in-plane mirror symmetry and is therefore geometrically achiral. The CD discussed here is the site-resolved local CD evaluated at a selected nanogap rather than the far-field CD of the entire trimer. Its overall far-field CD vanishes under normal incidence.

Although local CD is confined to the hotspot, it can be utilized when molecules are placed within the nanogap. The different local field enhancements under LCP and RCP illumination may produce polarization-dependent SERS signals [62] and surface-enhanced Raman optical activity (SEROA) [59,63]. Polarization-dependent photochemistry [64–66], such as hot-electron generation and plasmonic photocatalysis in the nanogap, may also be realized by local CD.

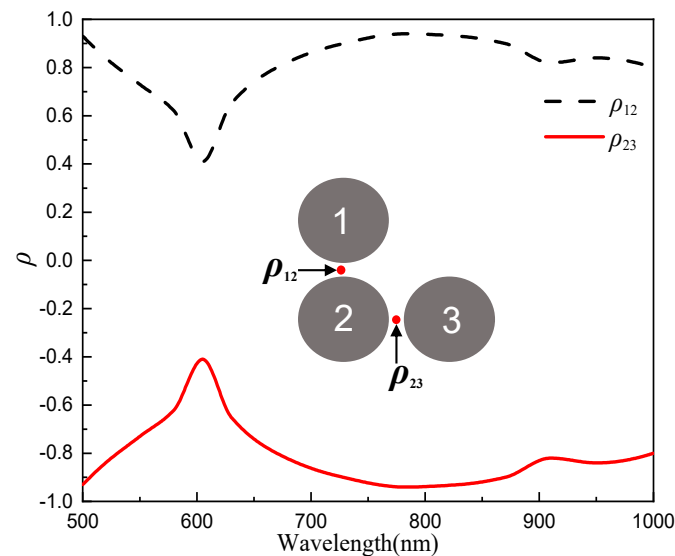


Figure 10. The local CD parameters ρ_{12} and ρ_{23} as functions of excitation wavelength at the centers of the 1st–2nd and 2nd–3rd nanocylinder gaps, respectively, with $d_{12} = d_{23} = 1$ nm. The red dots indicate the locations at which ρ_{12} and ρ_{23} are evaluated. The SiO₂ spacer-layer thickness is $t_{\text{SiO}_2} = 20$ nm.

5. Conclusions

To conclude, we have investigated the electric field enhancement in plasmonic nanocylinders under circularly polarized light excitation. A giant CD parameter $\rho = 0.94$ is obtained in the nanogap of the L-shaped trimer at 782 nm due to the destructive and constructive interference of near field illuminated by CPL light. The local CD is sensitive to geometry and can be tuned by the gap distance d and offset angle ϕ . A small gap ($d = \sim 1$ nm) and L-shaped trimer ($\phi = 90^\circ$) is essential for achieving strong near-field coupling and interference and thus the giant CD. This result exists widely in asymmetric trimers, while nanocylinders are suitable for photovoltaic applications. These findings are of interest in the field of near-field CD, chiral plasmonics and polarization-dependent photochemistry.

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Abbreviations

AR	Aspect ratio
CD	Circular dichroism
CPL	Circularly polarized light
f_{sub}	Substrate coupling efficiency
FDTD	Finite-difference time-domain
LCP	Left-handed circularly polarized light
LSPs	Localized surface plasmon resonances
M	Electric field enhancement
OA	Optical activity

ρ	Local CD parameter
PML	Perfectly matched layers
Q_{scat}	Normalized scattering efficiency
RCP	Right-handed circularly polarized light
SEROA	Surface-enhanced Raman optical activity
Si	Silicon
SiO ₂	Silicon dioxide
TFSF	Total-field/scattered-field

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