

Tailoring enantioselective interaction between chiral molecules and eye-shaped chiral metasurfaces

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Abstract: Chiral metasurfaces have emerged as a useful platform for enhancing the circular dichroism (CD) signals of chiral molecules, and understanding the enantioselective interactions between chiral metasurfaces and chiral molecules is essential. Here, we study the enhanced enantioselective interactions between chiral molecules and eye-shaped chiral metasurfaces by investigating the difference in CD response (ΔCD) and the differential dissymmetric factor of optical chirality (Δg_c) through varying structural parameters of the metasurface. It is shown that strong chiral near-field confinement enhances both ΔCD and Δg_c around the molecule resonance. Meanwhile, the coupling condition of the metasurface with incident light affects the ΔCD response of the metasurface-molecule coupled system but has a negligible effect on Δg_c . These results will advance the metasurface-based chiral molecule sensing for potential applications in pharmaceutical drug development and biomedical diagnostics.

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1. Introduction

Optical metasurfaces are artificially engineered materials composed of subwavelength nanostructures that can manipulate the properties of light. Metasurfaces can be designed to exhibit specific linear and nonlinear optical responses and achieve the desired functionalities. With the enhanced light-matter interaction, optical metasurfaces have found tremendous applications in fields such as light absorbers and emitters [1,2], nanolasers [3], optical imaging [4], optical chiral devices [5], nonlinear frequency conversion [6], optical encryption [7] and biosensing [8,9]. In particular, metasurface-enhanced circular dichroism (CD) spectroscopy is highly attractive for chiral molecule sensing. Chiral molecules, such as amino acids, DNA, and glucose, commonly exist as non-superimposable mirror-image enantiomers with distinct biological properties. Traditional vibrational circular dichroism spectroscopy often exhibits limited detection sensitivity for enantiomer detection and discrimination [10]. Metasurfaces have been widely used for enhancing weak CD signals of chiral molecules by engineering the electromagnetic fields with high optical chirality [11–13]. Furthermore, chiral metasurfaces can generate superchiral electromagnetic near fields, which enable enhanced optical chirality and strong chiroptical coupling with molecules [14,15]. It is important to understand the enantioselective interactions between chiral metasurface and chiral molecules through the study of far-field CD response and superchiral near fields confinement.

In this work, the enhanced enantioselective interactions between chiral molecules and eye-shaped chiral metasurfaces are investigated by varying the structural parameters of metasurfaces. Under circular polarization incidence, the difference in CD response (ΔCD) and the differential dissymmetric factor of optical chirality (Δg_c) of metasurfaces coupled with chiral molecules of opposite handedness are studied. As the minor-axis of the inner disk of metasurface decreases,

the enhanced chiral near-field confinement leads to the increased magnitudes of both ΔCD and Δg_c dip values around the molecule resonance, and the near-field Δg_c follows a linear relationship with the far-field ΔCD . As the spacer layer thickness of metasurface increases, the coupling condition of the metasurface with incident light moves close to the critical-coupling regime, which enhances the far-field ΔCD response of the metasurface-molecule coupled system whereas has little effect on the near-field Δg_c values. The demonstrated results provide insight into the understanding of enantioselective interaction between chiral molecules and chiral metasurfaces, which enables the enhanced selective detection of chiral molecules and advances the metasurface-based molecule sensing for potential applications in pharmaceutical drug development and biomedical diagnostics.

2. Results and discussion

Figure 1(a) shows the schematic of the eye-shaped chiral metasurface unit cell and the simulated near-field electric field distributions under circularly polarized incident light. The designed metasurface is a three-layer structure, with 65 nm-thick top gold layer and 200 nm-thick bottom gold layer and a middle alumina spacer layer of thickness $h = 400$ nm. The eye-shaped resonator on the top layer consists of an outside elliptical slot with major and minor axes of $a_1 = 2934$ nm, and $b_1 = 1334$ nm, and an inside elliptical disk with major and minor axes of $a_2 = 2134$ nm, $b_2 = 200$ nm and a rotation angle of $\theta = -8^\circ$. The periods of the unit cell in x and y directions are $P_x = 2400$ nm and $P_y = 2800$ nm, respectively. The electromagnetic response of the metasurface is simulated with COMSOL under normally incident left- and right-circularly polarized (LCP and RCP) light, with the periodic boundary condition in x and y directions and the perfectly matched layer in the z direction. The permittivities of gold and alumina are taken from literature [8,16]. The simulated electric field distributions at the metasurface resonance are shown in the bottom panel of Fig. 1(a). Strong electric field is concentrated around the tips of the inner elliptical disk under LCP incidence, whereas the electric field is weak under RCP incidence. To study the enantioselective interaction between chiral molecules and eye-shaped chiral metasurfaces, a chiral molecular layer with the thickness of 85 nm is coated on top of the metasurface. Here, a simple case of uniform, isotropic and dense molecular layer is considered. The interaction of electromagnetic wave and chiral molecules is given by the constitutive relations: $\mathbf{D} = \varepsilon_0 \varepsilon \mathbf{E} - i \kappa \mathbf{H}$, $\mathbf{B} = \mu_0 \mu \mathbf{H} + i \kappa \mathbf{E}$, where ε is the permittivity, μ is the permeability, κ is the Pasteur parameter, $\mathbf{E}$ and $\mathbf{H}$ are the complex electric field and magnetic field [17]. The optical properties of chiral molecules are modelled as $\varepsilon = \varepsilon_\infty - \gamma \left(\frac{1}{\hbar\omega - \hbar\omega_0 - i\Gamma} \right)$, $\kappa = \beta \left(\frac{1}{\hbar\omega - \hbar\omega_0 - i\Gamma} \right)$, and $\mu = 1$, where the background permittivity $\varepsilon_\infty = 1.15$, the coefficients $\gamma = 1.2483 \times 10^{-3}$ eV, $\beta = 4.6812 \times 10^{-4}$ eV, the damping constant $\Gamma = 1.56 \times 10^{-3}$ eV, and the resonance frequency $\omega_0 = 3.0235 \times 10^{14}$ rad/s corresponding to the wavelength of 6230 nm, which represent the chiral response of typical amino acids [18,19]. The handedness of chiral molecules is defined by the sign of $+\kappa$ or $-\kappa$. Figure 1(b) shows the absorption spectra of the bare metasurface and the metasurface coupled with chiral molecules. Electromagnetically induced transparency (EIT)-like and electromagnetically induced absorption (EIA)-like spectral features are observed for the metasurface coupled with chiral molecules under LCP and RCP incidence, respectively [9]. However, under the same circular polarization, the absorption responses are different for chiral molecules with $+\kappa$ and $-\kappa$ due to the enantioselective interaction between metasurface and molecules of opposite handedness.

The difference in absorption under LCP and RCP incidence is defined as $CD = A_{LCP} - A_{RCP}$. The CD spectra of metasurface coupled with $+\kappa$ and $-\kappa$ chiral molecules are shown in Fig. 2(a), exhibiting different dip values around the molecule resonance wavelength due to the enantioselective interaction between metasurface and chiral molecules. The difference in CD response for $+\kappa$ and $-\kappa$ chiral molecules is defined as $\Delta CD = CD_{+\kappa} - CD_{-\kappa}$, which represents the far-field characteristics of the coupled system between $+\kappa$ and $-\kappa$ cases. The ΔCD spectrum is

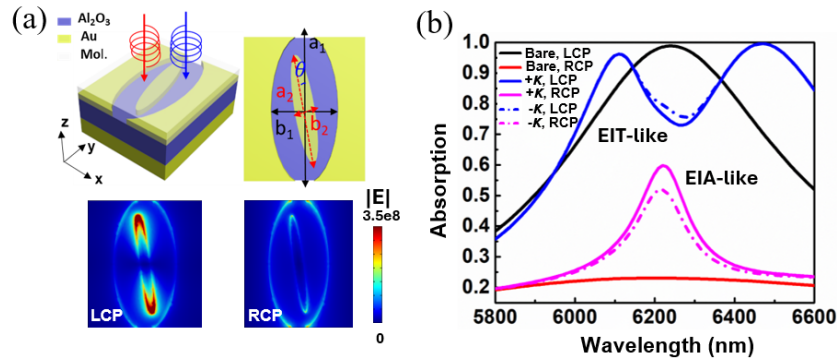


Fig. 1. (a) Schematic of the designed eye-shaped metasurface unit cell and the simulated electric field distributions on resonance under LCP and RCP incidences at the xy plane 5 nm above the top Au layer. (b) Absorption spectra of the bare metasurface and the metasurface coupled $+\kappa$ and $-\kappa$ molecules under LCP and RCP incidences.

plotted in Fig. 2(b) where a dip value of -0.12 around the molecule resonance is observed. Optical chirality C characterizes the chiral properties of electromagnetic fields and can be expressed as $C = \frac{\epsilon_0 \omega}{2} \text{Im}(\mathbf{E}^* \cdot \mathbf{B})$ [20]. Figure 2(c) displays the near-field optical chirality distributions of metasurface coupled with chiral molecules under circular polarizations at the molecule resonance wavelength. The optical chirality C is normalized by C_0 , which is the optical chirality of incident circularly polarized light in free space. Strong optical chirality around the tips of the inner

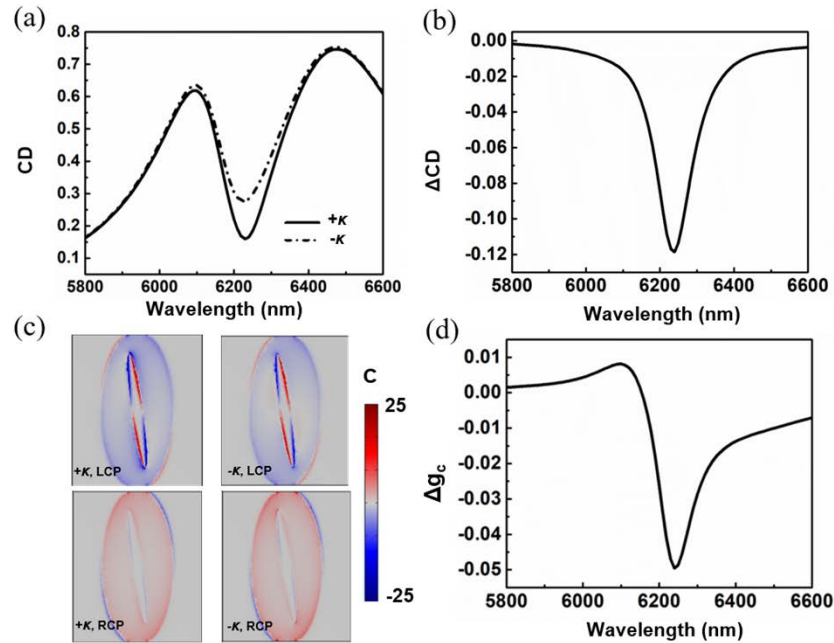


Fig. 2. (a) CD spectra of metasurface coupled with $+\kappa$ and $-\kappa$ chiral molecules. (b) ΔCD spectrum. (c) Optical chirality distributions of metasurface coupled with chiral molecules under circular polarizations at the xy plane 5 nm above the top Au layer at the molecule resonance. (d) Δg_c spectrum.

elliptical disk is obtained for both $+\kappa$ and $-\kappa$ chiral molecules under LCP incidence, however, the optical chirality is weak under RCP incidence. The dissymmetric factor of optical chirality $g_c = \frac{|C_{int,RCP}| - |C_{int,LCP}|}{|C_{int,RCP}| + |C_{int,LCP}|}$ is used to describe the difference in optical chirality distribution under LCP and RCP incidences, where $C_{int,LCP}$ and $C_{int,RCP}$ represent the integrated optical chirality over the entire simulation volume under LCP and RCP incidence, respectively. For metasurface coupled with $+\kappa$ and $-\kappa$ chiral molecules, the differential dissymmetric factor of optical chirality $\Delta g_c = g_{c,+\kappa} - g_{c,-\kappa}$ is considered to describe the near-field characteristics of the coupled system between the $+\kappa$ and $-\kappa$ cases. The Δg_c spectrum is shown in Fig. 2(d) with a dip value of -0.05 around the molecule resonance, manifesting the enantioselective coupling for $+\kappa$ and $-\kappa$ chiral molecules.

Next, the minor-axis length b_2 of the inside elliptical disk of the metasurface is varied to investigate the dependence of enantioselective coupling between metasurface and chiral molecules on the chiral near-field confinement. Figure 3(a) shows the CD spectra of metasurface coupled with $+\kappa$ and $-\kappa$ chiral molecules when b_2 is 600 nm, 400 nm and 200 nm. As the minor-axis length b_2 decreases, the dip of the CD spectrum becomes more pronounced, with the change in CD being greater for the $+\kappa$ case than for the $-\kappa$ case. The ΔCD spectrum plotted in Fig. 3(b) shows the magnitude of ΔCD dip value increases when b_2 is reduced, indicating the enhanced enantioselective coupling between metasurface and chiral molecules. Figure 3(c) shows the optical chirality C distributions of metasurface coupled with $+\kappa$ molecules under circular polarizations when $b_2 = 200$ nm and 600 nm. The stronger confinement of optical chirality around

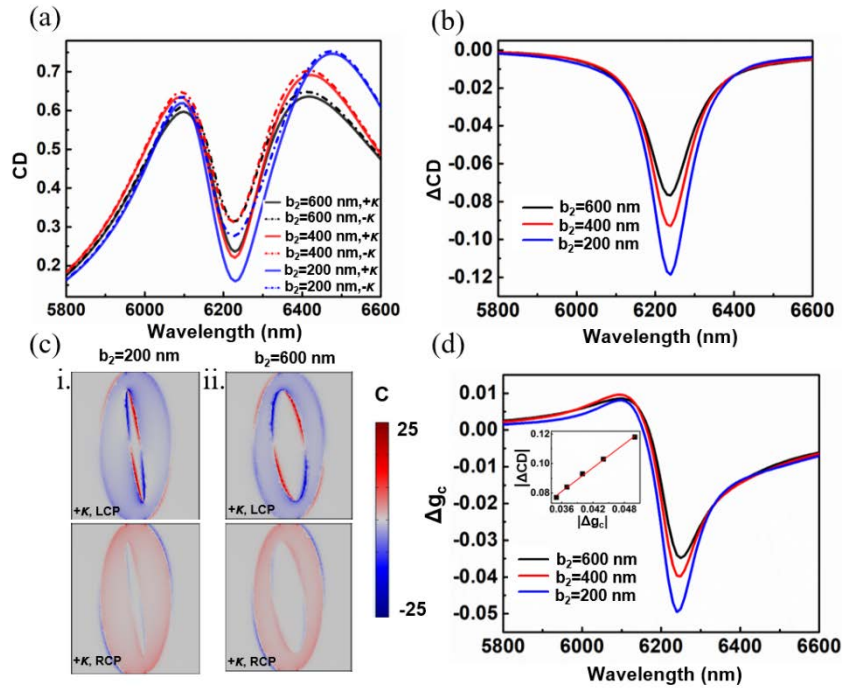


Fig. 3. (a) CD spectra of metasurface coupled with $+\kappa$ and $-\kappa$ chiral molecules with $b_2 = 600$ nm, 400 nm and 200 nm. (b) The corresponding ΔCD spectra for three b_2 cases. (c) Optical chirality distributions of metasurface coupled with $+\kappa$ molecules under LCP and RCP incidences for (i) $b_2 = 200$ nm and (ii) $b_2 = 600$ nm. (d) Δg_c spectra for the three b_2 cases. The inset shows a linear relationship between $|\Delta CD|$ and $|\Delta g_c|$ values at resonance with b_2 increasing from 200 nm to 600 nm in a step of 100 nm.

the tips of the inner elliptical disk is observed for metasurface with the smaller b_2 for both LCP and RCP incidences, which leads to the larger magnitude of the integrated optical chirality. Similarly, the integrated optical chirality is also increased for the metasurface coupled with $-\kappa$ molecules for the smaller b_2 case. Next, Δg_c spectra for the three b_2 cases are examined as shown in Fig. 3(d). The magnitude of Δg_c dip value increases with the decreased b_2 , which is consistent with the observation in the far-field ΔCD spectra in Fig. 3(b). The inset of Fig. 3(d) shows that the Δg_c dip value follows a linear relationship with the ΔCD dip value, as the minor-axis length b_2 is varied to enhance the chiral near-field confinement.

Furthermore, the spacer layer thickness h of the metasurface is varied to investigate the dependence of metasurface-molecule interaction on the coupling condition of the metasurface with incident light. Figure 4(a) shows the CD spectra of metasurface coupled with $+\kappa$ and $-\kappa$ chiral molecules when $h = 150$ nm, 200 nm and 400 nm with b_2 fixed at 400 nm. As the spacer layer thickness h increases, both the CD value of the metasurface and the CD contrast near the molecule resonance wavelength increase, corresponding to the coupling condition of the metasurface with the LCP incident light moving from the under-coupled regime toward the critical-coupling condition while the coupling condition with the RCP incidence remaining in the over-coupled regime. This behavior is related to the changes in the ratio of metasurface radiation loss to absorption loss under different coupling conditions of the metasurface with incident light as the spacer layer thickness varies [21]. In addition, the difference in the CD dip value between $+\kappa$ and $-\kappa$ cases gets larger with the increased spacer layer thickness. Figure 4(b) shows the ΔCD spectra for metasurfaces with different h values, where the magnitude of ΔCD dip value

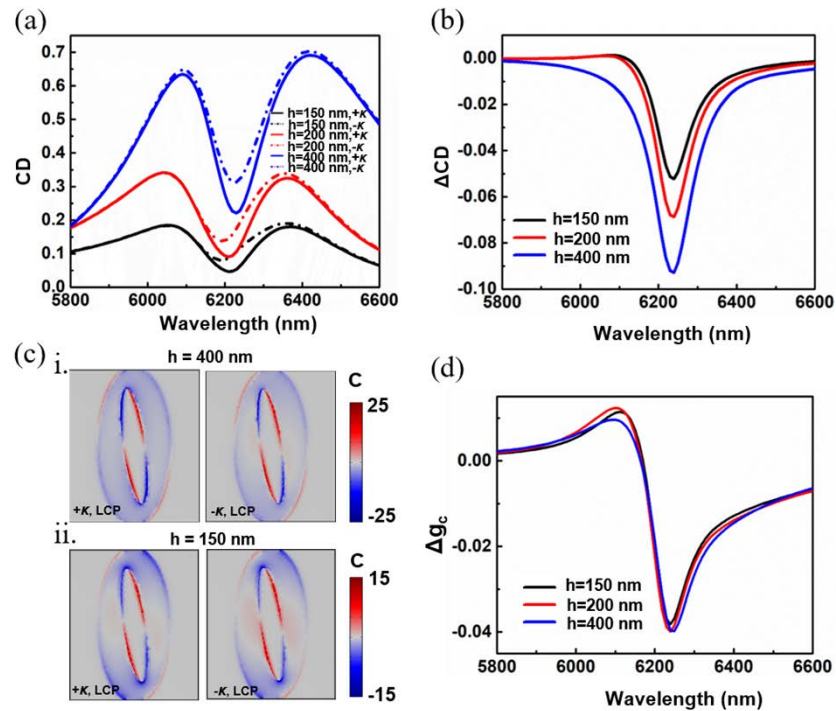


Fig. 4. (a) CD spectra of metasurface coupled with $+\kappa$ and $-\kappa$ chiral molecules with $h = 150$ nm, 200 nm and 400 nm when b_2 is fixed at 400 nm. (b) The corresponding ΔCD spectra. (c) Optical chirality distributions of metasurface coupled with $+\kappa$ and $-\kappa$ molecules under LCP incidence for (i) $h = 400$ nm and (ii) $h = 150$ nm. (d) Δg_c spectra for the three h cases.

increases and the spectrum linewidth broadens when the spacer layer gets thicker, showing the enhanced far-field chiroptical response of the molecule-metasurface system. Figure 4(c) shows the optical chirality C distributions of metasurface coupled with $+\kappa$ and $-\kappa$ molecules under LCP incidence for $h = 150$ nm and 400 nm. The intensity of optical chirality C is enhanced to a similar extent for both the $+\kappa$ and $-\kappa$ cases when h increases from 150 nm to 400 nm. It is noted that under RCP incidence, the intensity of optical chirality C exhibits the same trend. As a result, the Δg_c dip values around the molecule resonance are almost unchanged, as shown in Fig. 4(d). It shows that the coupling condition of the metasurface with incident light has little effect on the near-field Δg_c values, however, it can enhance the far-field ΔCD response of the metasurface-molecule coupled system.

3. Conclusion

We have demonstrated the tailoring of enantioselective interactions between chiral molecules and eye-shaped chiral metasurfaces by investigating the difference in CD response ΔCD and the differential dissymmetric factor of optical chirality Δg_c through varying structural parameters of metasurface. As the chiral near-field confinement becomes strengthened with the decreased minor-axis of the inner disk, the magnitudes of both ΔCD and Δg_c dip values around the molecule resonance increase while the near-field Δg_c follows a linear relationship with the far-field ΔCD . As the spacer layer thickness increases, the coupling condition of the metasurface with incident light transitions close to the critical-coupling regime, which enhances the far-field ΔCD response of the metasurface-molecule coupled system whereas has little effect on the near-field Δg_c values. Our findings are believed to support the understanding of enantioselective interaction between chiral molecules and chiral metasurfaces, enabling enhanced selective detection of chiral molecules with opposite handedness and promoting metasurface-enhanced vibrational circular dichroism for potential applications in pharmaceutical drug development and biomedical diagnostics.

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Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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