Surface Enhanced Circular Dichroism by Electric and Magnetic Dipole Resonance of Cross Shaped Nanoholes

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The near-field interaction of plasmonic nanostructures and chiral molecules induces circular dichroism in achiral nanostructure. The induced circular dichroism (ICD) strength is several orders greater than the molecular inherent CD (MCD), and it generally appears at visual range (VR) where the CD signal is easily detectable. Therefore, the ICD is considered as a potential method to detect single molecular chirality. In this study, cross shaped nanohole (CSN) is proposed for enhancing the MCD signal. We explained the mechanism of MCD and ICD generation and found that the maximum enhancement of the structure reached 450 folds of MCD. Through analysis of the surface plasmon resonance mode, it is found that overlapping of the electric dipole (ED) and magnetic dipole (MD) surface plasmon resonance (SPR) in CSN is essential for the CD enhancement. Furthermore, we investigated the effects of the thickness of structure, length of longer and shorter arms of nanohole, the displacement of shorter arm of the nanohole and x period of CSN on the circular dichroism enhancement, and found the enhancement factor and location of CD enhancement peak to be tunable by the structural parameters.

Keywords: induced circular dichroism, chiral molecules, surface enhanced CD, electric and magnetic dipole resonance

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I. INTRODUCTION

Circular dichroism (CD), absorption difference of right-circularly polarized (RCP) and left-circularly polarized (LCP) light, is widely used in determining chirality of molecules, secondary structure of proteins and stability of biomolecules [1–3]. Especially, CD spectroscopy is routinely used for chirality detection of molecules in drug production, because toxicity or therapeutic properties of drug molecules are related to their chirality [4]. However, molecule inherent CD (MCD) is intrinsically weak. Furthermore, MCD peak in UV region overlaps with absorption peak of molecules and solvent which generates strong noise during the measurement. Since the detection of molecular chirality is difficult, the surface plasmon enhancing technique is, therefore, used to enhance MCD. In the near field interaction of chiral molecules and nanostructure, first, the MCD is enhanced by localized optical chiral field [5, 6], second, the chiral molecule induces CD (ICD) signal in nanostructure. The ICD is several orders higher than the MCD at surface plasmon resonance (SPR) wavelength [7] and it is sensitive to molecular chirality. Therefore, the ICD is considered as a potential method for detecting molecular chirality and realizing single molecular chirality detection.

In past decades, many experimental and theoretical studies have been carried out to enhance the MCD. At the beginning, the chiral plasmonic nanostructures were considered as potential candidates for enhancing the MCD, because chiral nanostructure can enhance localized optical chiral field which contributes to the MCD [7–9]. But lateral studies show that the ICD arisen from interactions of chiral molecule and achiral nanostructure is greater than MCD enhanced by localized optical chiral field [10–19]. Different shapes of achiral nanostructures, such as half sphere [10–12], cross [13], cubic [14, 15] and cylindrical [16, 17] ones have been proposed for enhancing the ICD. The different shape of nanostructures generates different SPR mode, and it is desired to know some specific SPR mode to enhance the ICD to as large extent as possible. Furthermore, different materials such as gold [16], silver [15] and dielectric [17] also considered to enhance the ICD. Although the adjustment of shape and materials indeed enhances the ICD, but no detailed investigation about mechanism of MCD and ICD generation is available, moreover the enhancement factor still does not meet the expectations. Only an enhancement of MCD up to two orders of magnitude in long wave infrared (LW-IR) range [18] has been reported, but LW-IR range is not comparable with molecular vibrational wavelength, therefore the value of ICD is small.
Generally, ICD is larger in the region close to molecular vibrational resonance. Therefore, ICD enhancement in visual (VR) or shortwave infrared (SW-NIR) range is desirable.

In this paper, cross shaped nanohole (CSN) nanostructure is proposed for enhancing the MCD in the VR and SW-NIR range. We explained the mechanism of MCD and ICD generation and found the maximum enhancement of the structure reached to 450 folds of molecular inherent MCD. Through analyzing the surface plasmon resonance mode, it is found that overlapping of the electric (ED) and magnetic dipole (MD) surface plasmon resonance (SPR) is essential for CD enhancement. Furthermore, the effect of structural parameters also investigated. We found that the value and location of CD enhancement peak can be tuned by the structural parameters.

II. STRUCTURE AND COMPUTATIONAL METHOD

Figure 1(a) depicts the proposed cross shaped nanohole structure. The structure is supposed to be prepared on glass substrate. The material of nonohole film is gold, and the refractive index is calculated by Brendel-Bormann model [19]. The thickness of film is set as \( t = 20 \text{ nm} \), periods fixed as \( P_x = 460 \text{ nm} \) and \( P_y = 260 \text{ nm} \), the length of longer arm of nanohole is \( l_1 = 360 \text{ nm} \), the length of shorter arm is \( l_2 = 160 \text{ nm} \), the width of hole is \( w = 20 \text{ nm} \), and the thickness of substrate is \( t_s = 100 \text{ nm} \). CD is defined as

\[
CD = A_{RCP} - A_{LCP},
\]

where \( A_{RCP} \) and \( A_{LCP} \) are the absorption of right-hand circularly polarized (RCP) light and left-hand circularly polarized (LCP) light, respectively. Generally, chiral molecules have vibrational absorption peak and chirality peak in UV region [16], for

![Figure 1](image-url)

FIG. 1. (a) Schematic plot of CSN and chiral medium composed system, (b) The unit cell of CSN.
example, the chiral biomolecule α-helix has larger MCD peak at wavelength 192 nm, and β sheet has a peak and dip around 200 nm. Therefore, the chiral molecules are modeled with a complex refractive index that has absorption peak at 192 nm, as same as molecular chirality parameter.

Finite element method (FEM) was used to perform the simulations, and the electric displacement vector, magnetic field strength and derivative of magnetic field strength was modulated by implementing chiral constitutive relations given by [20]

\[
\begin{align*}
D &= \varepsilon_0 \varepsilon_c E - \frac{ik}{c} H, \\
B &= \varepsilon_0 \varepsilon_c H + \frac{ik}{c} E,
\end{align*}
\]

where \( \kappa \) is the molecular chirality parameter and \( \varepsilon_c \) is the permittivity of the chiral molecule. In all calculations, relative permeability was set as \( \mu = 1 \). The molecular chirality parameter \( \kappa \) and permittivity \( \varepsilon_c \) are given by [21]

\[
\begin{align*}
\varepsilon_c &= \varepsilon_b + Nd\omega_0 \frac{\omega_0^2 - \omega^2 + i\omega\Gamma}{(\omega_0^2 - \omega^2)^2 + \omega^2\Gamma^2}, \\
\kappa &= Nr\omega \frac{\omega_0^2 - \omega^2 + i\omega\Gamma}{(\omega_0^2 - \omega^2)^2 + \omega^2\Gamma^2},
\end{align*}
\]

where \( \varepsilon_b \) is relative permittivity of background solvent, \( N \) is number density of molecules, \( d \) is electric dipole strength, \( r \) is rotatory strength, and \( \omega_0 \) is molecular absorption band frequency. The parameters were set as \( \varepsilon_b = 1.77 \), \( Nd = 0.8 \times 10^{14} s^{-1} \), \( Nr = 0.8 \times 10^{12} s^{-1} \), \( \Gamma = 3.77 \times 10^{14} s^{-1} \), \( \lambda_0 = 192 \text{ nm} \) and \( \omega_0 = 2\pi c/\lambda_0 \). Generally the chirality parameter of a chiral molecule is written in the following tensor form:

\[
\bar{\kappa} = \begin{pmatrix}
\kappa_{xx} & \kappa_{xy} & 0 \\
\kappa_{yx} & \kappa_{yy} & 0 \\
0 & 0 & \kappa_{zz}
\end{pmatrix}
\]

where the diagonal elements are the optical activity contributed by the electric and magnetic dipole of chiral molecules, the off-diagonal elements are optical activity contributed by the dipole-quadruple interactions. For randomly oriented chiral molecules, the off-diagonal elements become zero [22, 23], and the diagonal elements become \( \kappa_{xx} = \kappa_{yy} = \kappa_{zz} = \kappa \). For oriented molecules the five elements of chirality tensor elements are set as \( \kappa_{xx} = \kappa_{yy} = 0.25\kappa \), \( \kappa_{zz} = 2.5\kappa \) and \( \kappa_{xy} = \kappa_{yx} = 0.025\kappa \). Initially, the molecular orientation is defined as parallel
to $z$-axis, and the unitary transformation $\kappa_f = R(\varphi)\kappa_i R^{-1}(\varphi)$ is used for changing of molecular orientation, where, $\kappa_i$ is the initial orientation and $\kappa_f$ is the final orientation, $R(\varphi)$ is unit rotation matrix for angle $\varphi$ which is different for rotations about $x$, $y$ and $z$-axis [24].

III. RESULTS AND DISCUSSION

Figure 2(a) depicts the absorption and CD spectra of the composed system. Two SPR modes marked as I and II appear at 830 nm and 1190 nm in the absorption spectra. CD takes maximum value when molecular orientation is parallel to the $x$ axis. Therefore, the molecular orientation is set as parallel to the $x$ axis in the following. In CD spectra, two peaks appear at 830 and 1160 nm. The CD peak at 1160 nm is slightly deviated from wavelength of mode II. Thus, we investigated the ED and MD resonance mode by analyzing the charge and current distributions at mode I and mode II. The charge and current distributions are shown in Figure 2(b). At mode I, ED is octopolar mode and MD is quadruple mode around the hole. Because the ED and MD resonance overlap, both of the enhanced electric and magnetic field peaks are located at mode I thereby leading to overlapped absorption and CD peaks at this mode. Mode II is contributed from ED resonance at 1100 nm and MD resonance at 1190 nm, the ED resonance mode is quadruple mode while MD resonance mode is dipolar mode. The peak of enhanced electric field is located at wavelength of 1160 nm while the peak of enhanced magnetic field is located at 1190 nm. Thus, the absorption peak and CD peak at mode II observe slight deviation from overlapping.

The CD of composed system originats from the MCD and ICD. The MCD have three sources which are electric part $CD_E$ (Eq. S2), magnetic part $CD_M$ (Eq. S3) and chiral part $CD_C$ (Eq. S4) [25]. Figure 3(a) shows the spectra of $CD_E$, $CD_M$, $CD_C$ and MCD. The $CD_E$ and $CD_M$ are two order larger than the total MCD, but they are opposite to each other and sum of them results in relatively small MCD signal. Without the chiral molecules, intensity of electric and magnetic fields in molecular region are equal for RCP and LCP illuminations, the dipole radiation of chiral molecule generates opposite electric and magnetic field increments for RCP and LCP [26], this generates $CD_E$ and $CD_M$. The $CD_C$ is originated from opposite optical chirality of RCP and LCP.

The ICD originates from symmetry breaking of electric field in CSN by electric field radiated from molecular dipole, and the ICD is proportional to electric field in the nanostructure
FIG. 2. (a) CD and absorption spectrum of CSN, (b) Charge and current distribution at mode I and mode II.

and magnetic field in molecular region (Eq. S16), that is [26]

\[ \text{ICD} \propto \mathbf{E}^{ns^*} \cdot \mathbf{B}^c \]  

(4)

where \( \mathbf{E}^{ns^*} \) is electric field strength in nanostructure, and \( \mathbf{B}^c \) is magnetic flux density in chiral medium. From Eq. (4) and simulation result, it is concluded that the ICD peak is a result of enhanced magnetic field in molecular region and electric field in CSN. From Fig. 3(b) which depicts the MCD, ICD and total CD of whole composed system, it turns out that the ICD dominates the total CD. Therefore, only the ICD is considered in the following.

FIG. 3. (a) spectra of MCD and its components CD_E, CD_M, CD_C. (b) Spectra of MCD, ICD and CD.
In order to further enhance the ICD, the thickness of nanohole film \( t \) was changed from 30 \( nm \) to 10 \( nm \) by step of 5 \( nm \). To clearly represent the enhancement result, the enhancement factor of molecule-CSN composed system was calculated by \( f = \frac{CD}{MCD} \) [25]. Figure 4(a) depicts the enhancement factor spectra for different values of film thickness \( t \). The ICD increased with decrease of thickness and the enhancement factor reached 450 when \( t = 10 \) \( nm \). In order to explain the increasing of ICD, the electric field \( E^{ns} \) and magnetic field \( B^{c} \) were analyzed. Figure 4(b) presents the charge and induced current distributions in CSN. With the decrease of \( t \), the strength of both ED and MD resonance are enhanced at the two modes. Therefore, the electric field strength \( E^{ns} \) is increased (Fig S1) thereby resulting in increase of ICD at both mode I and mode II.

![Figure 4](image)

**FIG. 4.** (a) Enhancement factor spectra for different thickness, (b) Charge and current distribution for different thickness.

The MD resonance at mode II appears around short arm of CSN which means that the MD resonance at mode II will not be affected by any change of longer arm length \( l_1 \) suggesting that the effective region of near field can be controlled by length \( l_1 \). In order to see this effect, we changed the length \( l_1 \) form 360 \( nm \) to 200 \( nm \) by step of 40 \( nm \) and results are shown in Figure 5(a). As expected, the wavelength of mode II does not change and ICD increases with decrease of \( l_1 \). Figure 5(b) presents the charge and induced current distributions for different values of \( l_1 \). At mode II, the MD resonance does not change with \( l_1 \), but the ED resonance is strengthened with decrease of \( l_1 \). Thus, each of electric field strength \( E^{ns} \), magnetic flux density \( B^{c} \) (Fig S2) and ICD increase at mode II. It is worth to note that the increase of enhancement factor is not only originated from the increase of
ICD, the decrease of molecular quantity also contributed to the enhancement. At mode I, the MD resonance appears around long arm of nanohole, therefore, mode I is blue shifts with decrease of $l_1$. Both of ED and MD resonance are weakened at mode I resulting in decrease of ICD.

![Graph showing enhancement factor spectra for different length $l_1$.](image)

**FIG. 5.** (a) Enhancement factor spectra for different length $l_1$, (b) Charge and current distribution for different length $l_1$.

The MD resonance around short arm of CSN at mode II can be modulated by length $l_2$ meaning that the wavelength and value of ICD at mode II can be regulated by $l_2$. Thus, the length $l_2$ was changed from 180 nm to 140 nm by step of 10 nm. Figure 6(a) shows the enhancement factor spectra for different values of $l_2$. At mode I, ICD does not change with $l_2$ whereas at mode II the enhancement factor increased with decrease of $l_2$ and reached 210 when $l_2 = 140$ nm. Figure 6(b) shows the charge and current distributions for different $l_2$. At mode I, ED and MD resonance appear around the long arm. Therefore, ICD is not strongly affected by length of short arm of CSN. With the decrease of $l_2$, the propagation length of induced current increased resulting in red shift of mode II. The strength of MD resonance at mode II remained unchanged but that of ED resonance increased causing an increase in electric field and decrease in magnetic field such that electric field increment is faster as compared to magnetic field decrement. More significantly, distance between ED and MD resonance reduced thereby improving ED and MD contributions to field enhancement which finally resulted in increase of ICD with decrease of $l_2$ at mode II.

After investigating the effect of the length $l_2$ on ICD, we stepped towards investigating the effect of position of short arm on ICD. The short arm is displaced from origin along the
FIG. 6. (a) Enhancement factor spectra for different $l_2$, (b) Charge and current distributions for different $l_2$.

axial directions by step of 10 nm. Figure 7(a) depicts the $f$ spectra for different displacement distance $d$. One can observe that the ICD at mode I increased whereas at mode II it decreased and blue shifted with gradual displacement of short arm. Besides these observations, a new mode can also be seen to emerge at longer wavelengths after mode II. Figure 7(b) shows the charge and current distributions at the mode I and mode II for different values of displacement distance $d$. With gradual increase of $d$, ICD at mode I enhanced as a result of strengthening of quadruple MD resonance. At mode II, however, owing to displacement distance $d$, path length of induced current reduced and MD resonance got weakened while quadrupole ED resonance got stronger with gradually transferring from quadrupole to hexapolar resonance. All these factors pushed the electric field $E^{ns}$ to decrease. As a result, ICD at mode II observed a decrease and blue shift. The new mode is appearing as a result of size match of half of the long arm length and longer part of short arm length of CSN (Fig. S4). At the new mode, the path length of induced current and strength of MD resonanced increase with $d$. Therefore, the new mode is red shifted and the ICD is increased with $d$.

At the last, we investigated the effect of period $P_x$ on ICD by changing $P_x$ from 400 nm to 520 nm by a step of 30 nm and the resulting $f$ spectra are shown in Figure 8(a) where it appears that CD at mode I did not change but at mode II it increased with $P_x$. From the charge and induced current distributions, it can be seen that the change of $P_x$ does not affect the ED and MD at mode I. Therefore, the change of ICD is very small at mode I.
At mode II, the MD resonance increased with $P_x$ while ED resonance decreased. The final result is increase of both $E^{as}$ and $B^c$. Thus the ICD increased at mode II.

IV. CONCLUSION

In this paper cross shaped nanohole is proposed to enhance the circular dichroism of chiral molecules in the visual and shortwave infrared range. We explained the mechanism of MCD and ICD generation and found the maximum enhancement this proposed structure
reached to be 450 folds of chiral molecules inherent circular dichroism signal. Furthermore, we found that the overlapping of electric and magnetic dipole resonance plays a critical role in circular dichroism induction in chiral molecule-nanostructure near field interactions. In addition, we investigated the effect of the thickness of structure, lengths of long and short arms of nanohole, the displacement of short arm of the nanohole, and the period $P_x$ on the circular dichroism enhancement. With decrease of shorter arm of nanohole, ICD increases and blue shifts. Longer periods are found to be beneficial for ICD enhancement. More significantly, not only the enhancement value of ICD but wavelength of enhancement peak can also be tuned by varying these structural parameters.

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Supporting Informations for

Quantitative Study on Circular Dichroism Induction from Achiral Nanostructure-Chiral Matter Near Field Interactions

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S1. ORIGIN OF CD

S1.1 Origin of molecular CD

The CD of chiral molecule contributed three terms which are electric part CD_E, magnetic part CD_M and chiral part CD_C, these are expressed as [1]

$$\text{MCD} = \text{CD}_E + \text{CD}_M \text{CD}_C$$  \hspace{1cm} (S2)

$$\text{CD}_E = \frac{ω}{2} \int_{V_c} \text{Im}(\varepsilon_c)[|E_+|^2 - |E_-|^2]dV_c$$  \hspace{1cm} (S3)

$$\text{CD}_M = \frac{ω}{2} \int_{V_c} \text{Im}(\mu_c) |H_+|^2 - |H_-|^2dV_c$$  \hspace{1cm} (S4)

$$\text{CD}_C = \frac{ω}{2} \int_{V_c} \text{Im}(\kappa)[\text{Re}(E_+ \cdot H_+^*) - \text{Re}(E_- \cdot H_-^*)]dV_c$$  \hspace{1cm} (S5)

S1.2 Origin of induced CD

Absorption of a nanostructure is generally given by[2]

$$A_{ns}^{\pm} = \frac{ω}{2} \int_{V_{ns}} [\text{Im}(\varepsilon_{ns})(E_{n\pm}^0 \cdot E_0^0) + \text{Im}(\mu_{ns})(H_{n\pm}^0 \cdot H_0^0)]dV,$$  \hspace{1cm} (S6)

where, ω is angular frequency of CPL, ε_{ns} and µ_{ns} are relative permittivity and permeability of nanostructure, E_{n\pm}^0 and H_{n\pm}^0 are electric and magnetic field strengths. The magnetic

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susceptibility of plasmonic nanostructure is very small, the magnetic loss can therefore be neglected. In presence of chiral molecule, \( \mathbf{E}_0^d \) will be affected by electric field radiated by chiral molecular dipole. Equation (S6) can then be written as \[3\]

\[
A_{ns}^\pm = \frac{\omega}{2} \text{Im}(\varepsilon_{ns}) \int_{V_{ns}} (\mathbf{E}_0^0 + \mathbf{E}_\pm^d)^* \cdot (\mathbf{E}_0^0 + \mathbf{E}_\pm^d) \, dV, \quad (S7)
\]

where, \( \mathbf{E}_\pm^d \) is dipole radiated electric field. The dipole radiated electric field can be written in terms of Green’s function. For time harmonic electromagnetic fields, the Green functions are dyadic functions, because the source is vector. Thus, the dipole radiated electric field can be written in terms of Green dyadic integral form

\[
\mathbf{E}_d^d(r) = \int_{V'} \begin{pmatrix} \overline{G}_{ee}(r - r') \\ \overline{G}_{em}(r - r') \end{pmatrix} \begin{pmatrix} \mathbf{J} \\ \mathbf{M} \end{pmatrix}, \quad (S8)
\]

where the \( \mathbf{J} \) and \( \mathbf{M} \) electric and magnetic currents. The Green functions satisfy the Maxwell equations. Using chiral constitutive relation to solve the Maxwell equations we derive Green functions for chiral medium[4]

\[
\overline{G}_{ee}(r) = \overline{G}_{ee+}(r) + \overline{G}_{ee-}(r)
\]

\[
\overline{G}_{ee\pm}(r) = -\frac{i\eta}{2} [(k_\pm G_{\pm}(r)) \mathbf{I} \pm \nabla G_{\pm}(r) \times \mathbf{I} + \nabla \nabla \left( \frac{G_{\pm}(r)}{k_{\pm}} \right)], \quad (S9)
\]

\[
\overline{G}_{em}(r) = \overline{G}_{em+}(r) + \overline{G}_{em-}(r)
\]

\[
\overline{G}_{em\pm}(r) = -\frac{1}{2} \left[ \pm (k_\pm G_{\pm}(r)) \mathbf{I} + \nabla G_{\pm}(r) \times \mathbf{I} + \nabla \nabla \left( \frac{G_{\pm}(r)}{k_{\pm}} \right) \right]
\]

where \( G_{\pm}(r) = \frac{e^{-ik_\pm r}}{4\pi r}, \ k_\pm = k(1 \pm \kappa \frac{n_c}{n_e}) \) and \( n_c \) is refractive index of chiral medium. Thus, the dipole radiated electric field is

\[
\mathbf{E}_d^d = i\omega n_{mol} \alpha \overline{G} \cdot \mathbf{E}_c^c + \omega n_{mol} \kappa \overline{G} \cdot \mathbf{B}_c^c. \quad (S10)
\]

The inducted CD in nanostructure is \[5\]

\[
\text{CD}_{ind} = \frac{\omega}{2} \text{Im}(\varepsilon_{ns}) \int_{V_{ns}} (\mathbf{E}_0^0 + \mathbf{E}_+^d)^* \cdot (\mathbf{E}_0^0 + \mathbf{E}_+^d) \, dV
\]

\[
- \frac{\omega}{2} \text{Im}(\varepsilon_{ns}) \int_{V_{ns}} (\mathbf{E}_0^0 + \mathbf{E}_-^d)^* \cdot (\mathbf{E}_0^0 + \mathbf{E}_-^d) \, dV
\]

\[
\frac{\omega}{2} \text{Im}(\varepsilon_{ns}) \int_{V_{ns}} \left( (\mathbf{E}_+^0 + \mathbf{E}_+^d)^* \cdot (\mathbf{E}_+^0 + \mathbf{E}_+^d) - (\mathbf{E}_-^0 + \mathbf{E}_-^d)^* \cdot (\mathbf{E}_-^0 + \mathbf{E}_-^d) \right) \, dV. \quad (S11)
\]

The dipole radiated electric field \( \mathbf{E}_d^d \) is much smaller than original near field in nanostructure. Then neglecting the second order term \( \mathbf{E}_d^d \cdot \mathbf{E}_d^{d*} \) we derive

\[
\text{CD}_{ind} = \frac{\omega}{2} \text{Im}(\varepsilon_{ns}) \int_{V_{ns}} 2\text{Re} \left( \mathbf{E}_+^0 \cdot \mathbf{E}_+^{ds} - \mathbf{E}_-^0 \cdot \mathbf{E}_-^{ds} \right) \, dV. \quad (S12)
\]
According to Eqs. S12 and S15 the ICS is proportional to electric field in nanostructure and field in nanostructure is symmetric to any achiral nanostructure is symmetric to $y$ (or $x$, any achiral structure has symmetric axis) axis, the near field in $xy$ plane has properties $E^x_+(x, y) = E^x_-(x, -y)$, $E^y_+(x, y) = -E^y_-(x, -y)$, $E^z_+(x, y) = E^z_-(x, -y)$, $B^x_+(x, y) = -B^x_-(x, -y)$, $B^y_+(x, y) = B^y_-(x, -y)$, $B^z_+(x, y) = -B^z_-(x, -y)$. Applying coordinate transformation $y \to -y$ (or $x \to -x$, depending on the symmetry axis) to \( \frac{1}{r^2}(x-x')^2E^0_{x'}(x', y')E^x_+(x, y) \), we derive

\[
\frac{1}{r^2}(x-x')^2E^0_{x'}(x', -y')E^x_+(x, -y) = \frac{1}{r^2}(x-x')^2E^0_{x'}(x', y')E^x_-(x, y)
\]  

(Equation S14) means the term \( \frac{1}{r^2}(x-x')^2E^0_{x'}(x', y')E^x_+(x, y) \) is equal for RCP and LCP illumination. Using the same coordinate transformations to other terms, we finally derive

\[
(E^0_+ \cdot E^d_+) - (E^0_- \cdot E^d_+)
\]

\[
= -\frac{3i\omega \eta_{mol} \kappa}{r^2 k} (G_+ + G_-) \frac{1}{r^2} \left( x^2 E^0_{x'} B^{cx}_+ + y^2 E^0_{y'} B^{cy}_+ + z^2 E^0_{z'} B^{cz}_+ \right)
\]

(S15)

According to Eqs. S12 and S15 the ICS is proportional to electric field in nanostructure and magnetic field in molecular region, that is

\[
\text{ICD} \propto E^{ns'} \cdot B^c
\]

(S16)
S2. SUPPORTING PICTURES

The Figure S1(a) show the real and imaginary part of molecular chirality parameter. The Figure S1(b) depicts the absorption of chiral molecules. It can be observed that the chirality peak and absorption peak overlap. Since asymmetric factor of MCD is small and strong absorption generates noise during the CD measurement, the detection of the chirality of molecules in UV region is therefore relatively difficult. But the chirality parameter exponentially decrease with the distance apart from molecular vibrational wavelength. Since, the CD (or ICD) signal is small in infrared (IR) or far IR region, visible or shortwave infrared (SW-IR) region is therefore beneficial for detecting molecular chirality by surface enhancement technique.

FIG. S1. (a) Real and imaginary part of chirality parameter, (b) Absorption of real chiral molecules
FIG. S2. (a) Averaged electric field $E^{ns}$ for different thickness, (b) Averaged magnetic flux density $B^{ns}$ for different thickness.

FIG. S3. (a) Averaged electric field $E^{ns}$ for different length $l_1$, (b) Averaged magnetic flux density $B^{ns}$ for different length $l_1$. 
FIG. S4. (a) Averaged electric field $E^{ns}$ for different length $l_2$, (b) Averaged magnetic flux density $B^{ns}$ for different length $l_2$.

FIG. S5. the new ED and MD resonance modes for displacement $d = 10nm$
FIG. S6. (a) Averaged electric field $E^{ns}$ for different periods $P_x$, (b) Averaged magnetic flux density $B^{ns}$ for different length $P_x$

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