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

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The circular dichroism (CD_{ind}) induced from interaction of chiral molecules with achiral plasmonic metal nanostructures has improved detection limit of molecular chirality. Although experimental results have presented several orders magnitude of CD_{ind} enhancement, but pushing the enhancement limit is restricted by superficial understanding of the CD_{ind} induction mechanism. In this paper, a theory for analyzing the mechanism of CD_{ind} induction, and relationship between CD_{ind} with near field around nanostructure is presented. By introducing dipole approaching of chiral molecule and a near field approximation of molecule-nanostructure interaction, we derived an analytical formula for CD_{ind}. The theory explains the mechanism of CD_{ind} induction which is symmetry breaking of electric field in nanostructure by chiral molecules dipole radiation. Furthermore, the derived formula exhibits CD_{ind} proportional to molecules chirality parameter and number density, and it exponentially increases with the inverse cube of distance between molecule and nanostructure. Most importantly, the formula shows that CD_{ind} is proportional to strength of electric field in nanostructure and magnetic field in molecular region. To verify the analytical results, we studied the CD_{ind} properties of graphene nanohole metasurface (GNM) and chiral molecule composed system. Numerical results were found consistent with analytical results. The investigation results are helpful for understanding mechanism of CD_{ind} induction, pushing the limit of CD_{ind} enhancement and realizing supersensitive detection of molecular chirality.

Keywords: Induced circular dichroism, Surface plasmon, Chiral molecules, Graphene

I. INTRODUCTION

Circular dichroism (CD), absorption difference of right circularly polarized (RCP) and left circularly polarized (LCP) light, is widely used in determining the configurations and conformations of chiral molecules [1–5]. However, CD of chiral molecules (CD_{mol}) is weak and only observable in the UV region. Therefore, it is desired to develop new techniques to increase the limit of sensitivity, so as to achieve the ultimate goal of single-molecule chirality detection. Thus the surface plasmon enhancing technique has been used for enhancing the CD_{mol} [6]. In the interaction between chiral molecules and nanostructure, on one hand, the optical chiral field enhanced by surface plasmon increases the CD_{mol} in the UV region [7], on the other hand, the molecular chirality transfers to nanostructure and generates the induced circular dichroism (CD_{ind}) signal [3, 6]. The CD_{ind} provides a way to detect chiral molecules in the visual or microwave region, which is easily detectable by CD spectroscopy.

In order to further enhance the CD_{ind} signal, effects of different factors such as molecules location [8, 9], density [10, 11] and orientation [13, 19], separation distance of molecule and nanostructure [5], shape of nanostructure [3, 14, 15] on CD_{ind} have been investigated. For nanorod-chiral molecule coupled system, the CD_{ind} is larger when it is placed on ends than placed on sides [8, 16, 17]. Lateral studies have found that the CD_{ind} is proportional to the near field intensity [18], and stronger near field is located at ends of nanorod . The CD_{ind} is sensitive to molecular orientations also [13, 19, 20], and the CD_{ind} is larger when the orientation of largest chirality component is along the strongest near field component [20]. The properties of CD_{ind} are gradually being clear and origin of CD in chiral molecule-nanostructure interaction is archived based on chiral Poynting’s theorem[21]. Even so, further quantitative studies on near field interactions between chiral molecule and nanostructure, mechanism of chirality transfer from chiral molecule to achiral structures are more desirable. Specifically, quantitative studies on interaction of molecular dipole with near field in molecular region and nanostructure, effect of molecule-nanostructure distance, and mechanism of CD_{ind} induction in near field interaction of chiral molecules dipole and achiral nanostructure are helpful for pushing the limit of chirality detection.

In this paper, a theory based on molecular dipole approaching and molecule-nanostructure near field interaction approximation is presented to analyze the CD_{ind} induction in chiral molecule-achiral nanostructure near field interaction, and we use the theory to explain the mechanism of CD_{ind} induction in achiral nanostructure. To prove the theory, a graphene nanohole metasurface (GNM)-chiral molecule composed system is proposed for simulation. The simulated results are consistent with the analytical results. Both simulated and analytical results show that there are linear relations between the CD_{ind} and molecular density, molecular chirality parameter, and the CD_{ind} exponentially increases with the inverse cube of molecule-nanostructure distance. Furthermore, the CD_{ind} is also proportional to electric field strength in nanostructure and magnetic flux density in [20].
molecular region. These results would be helpful for realizing supersensitive detection of molecular chirality.

II. THEORY

Absorption of nanostructure is formally calculated by volume integral \( \frac{\omega}{2} \text{Im}(\varepsilon) \int_{V_{ns}} |E|^2 dV \) [21, 22]. In this case, electric field in nanostructure comes from the contributions of the electric field generated by surface plasmons without chiral molecules and the electric field radiated by molecular dipole. Thus, absorption of the nanostructure is given by [8]

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

where, \( E^0_\pm \) (+ represents RCP light, and - represents LCP light) is the original electric field inside the nanostructure without the chiral molecule, \( E^d_\pm \) is the dipole radiated electric field in nanostructure, \( A_{ns}^{\pm} \) is the absorption of nanostructure, \( \omega \) is the angular frequency of light, \( \varepsilon_{ns} \), \( V_{ns} \) are respectively the permittivity and volume of nanostructure.

The electric fields \( E^d_\pm \) can be written in terms of dyadic Green’s function \( E^d_\pm = i\omega n_{mol}\mathcal{G}^\pm(r)P_{mol}^\pm \), where, \( n_{mol} \) is the molecules number density, \( P_{mol}^\pm \) is dipole strength of molecular chiral medium, and \( \mathcal{G}^\pm(r) \) is the dyadic Green’s function which is the electric field corresponding to molecular dipole at arbitrary location. The Green function \( \mathcal{G}^\pm(r) \) of chiral medium is given by [23]

\[
\mathcal{G}^\pm(r) = \overline{\mathcal{G}}^\pm(r) + \overline{\mathcal{G}}^\mp(r)
\]

where, \( \overline{\mathcal{G}}^\pm(r) = -\frac{i\eta}{2}(k_\perp G^\pm(r))\mathbb{I} + \nabla G^\pm(r) \times \mathbb{I} + \nabla (\frac{G^\pm(r)}{k_\perp}), \)

where, \( \eta = \sqrt{\varepsilon} \) is the electrical impedance of chiral medium, \( \varepsilon, \mu \) are permittivity and magnetic permeability of chiral medium, \( \mathbb{I} \) is unit matrix, and \( r \) is the distance between molecule and location of the observed electric field, \( k_\perp = (k \pm r \hat{z}) \) are wave vectors of RCP and LCP, and the function \( G^\pm(r) \) is equal to \( G^\pm(r) = \frac{1}{4\pi r}e^{-ik_\perp \cdot r}. \) Here, \( k_\perp \) is wave vector of linear polarized light, \( n_c \) is refractive index of chiral molecule. The chiral medium and nanostructure interact through near field, thus implementing the near-field interaction approximation \( k_\perp r \ll 1 \) to calculate the gradients of function \( G^\pm(r) \), we derive

\[
\nabla G^\pm(r) = (ik_\perp + \frac{1}{r})G^\pm(r)\hat{u}_r \approx \frac{1}{r}G^\pm(r)\hat{u}_r,
\]

\[
\nabla \nabla G^\pm(r) = [(ik_\perp + \frac{1}{r})^2 + \frac{1}{r^2}]G^\pm(r)\hat{u}_r \hat{u}_r
\]

\[
- \frac{1}{r^2}G^\pm(r)(\mathbb{I} - \hat{u}_r\hat{u}_r) \approx \frac{3}{r^2}G^\pm(r)\hat{u}_r\hat{u}_r - \frac{1}{r^2}G^\pm(r)\mathbb{I},
\]

where, \( \hat{u}_r \hat{u}_r \) is a diadic and \( \hat{u}_r \) is a unit vector along distance \( r \). Replacing \( \nabla G^\pm(r) \) and \( \nabla \nabla G^\pm(r) \) by their respective relations from Eq. (3) and implementing near field approximation \( k_\perp r \ll 1 \), Eq. (2) can be written as

\[
\overline{\mathcal{G}}^\pm(r) = -\frac{i\eta}{2r^2k_\perp}(3G^\pm(r)\hat{u}_r\hat{u}_r - G^\pm(r)\mathbb{I}).
\]

Dipole moment of chiral molecule is given by [24, 25]

\[
P^\text{mol} = \alpha E^c_\pm - \frac{\kappa}{c}B^c_\pm,
\]

where, \( E^c_\pm \) and \( B^c_\pm \) are respectively electric field strength and magnetic flux density of near field in molecular region, \( \alpha \) and \( \kappa \) are respectively the electric polarizability and chirality parameter of chiral molecules, and \( c \) is the speed of light. Finally, we derive the dipole radiated electric field \( E^d_\pm \)

\[
E^d_\pm = i\omega n_{mol}G^\pm(r)P_{mol}^\pm = \frac{\omega n_{mol}\alpha}{2r^2k_\perp}\mathbb{G}^\pm E^c_\pm + \frac{\omega n_{mol}\kappa}{c}B^c_\pm
\]

\[
= \frac{\omega n_{mol}\alpha}{2r^2k_\perp} [3(G_+ + G_-)\hat{u}_r\hat{u}_r - (G_+ + G_-)\mathbb{I}] \cdot E^c_\pm
\]

\[
+ \frac{\omega n_{mol}\kappa}{2r^2k_\perp} [3(G_+ + G_-)\hat{u}_r\hat{u}_r - (G_+ + G_-)\mathbb{I}] \cdot B^c_\pm.
\]

In order to derive the CD effect, we first calculate field intensity difference in nanostructure

\[
(E^0_+ + E^d_+)^* \cdot (E^0_+ + E^d_+) - (E^0_- + E^d_-)^* \cdot (E^0_- + E^d_-)^* = 2Re \left( (E^0_+ \cdot E^d_+) - (E^0_- \cdot E^d_-) \right).
\]

Replacing \( E^d_\pm \) to calculate the \( (E^0_\pm \cdot E^d_\pm) \), we derive

\[
(E^0_\pm \cdot E^d_\pm) = E^0_\pm \cdot \left\{ \frac{\omega n_{mol}\alpha}{2r^2k_\perp} [3(G_+ + G_-)\hat{u}_r\hat{u}_r - (G_+ + G_-)\mathbb{I}] \cdot E^c_\pm
\]

\[
+ \frac{\omega n_{mol}\kappa}{2r^2k_\perp} [3(G_+ + G_-)\hat{u}_r\hat{u}_r - (G_+ + G_-)\mathbb{I}] \cdot B^c_\pm \right\}.
\]

Suppose the incident field is along \( z \) direction, and the achiral structure is symmetric along \( y \) axis. The near field in any \( xy \) plain has properties \( E^0_\pm(x, y) = E^d_\pm(x, y) \), \( E^0_\pm(x, y) = -E^d_\pm(-x, y) \), \( E^0_\pm(x, y) = E^d_\pm(x, y) \), \( E^0_\pm(x, y) = B^d_\pm(x, y) \), \( E^d_\pm(x, y) = B^d_\pm(-x, y) \). Implementing these to Eq. (8), the field intensity difference in nanostructure becomes

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

\[
\left\{ \frac{-3\omega n_{mol}\alpha}{c r^2} \left[ x^2E^{0\ast}_x B^x + y^2E^{0\ast}_y B^y + z^2E^{0\ast}_z B^z \right]
\]

\[
- \frac{-\omega n_{mol}\kappa}{c r^2} \left[ x^2E^{0\ast}_x B^x + y^2E^{0\ast}_y B^y + z^2E^{0\ast}_z B^z \right] \right\}.
\]
For periodic structure in $x$ and $y$ directions, Eq. (9) can approximate as
\[
\left( E^+_m + E^+_d \right) \cdot \left( E^+_m + E^+_d \right)^* - \left( E^-_m + E^-_d \right) \cdot \left( E^-_m - E^-_d \right)^* = \frac{\eta_{mol}}{2\pi^3} \text{Re}\{i\kappa e^{-ikr}[E^m_{nx} B^m_{nx} + E^m_{ny} B^m_{ny}]\}. \tag{10}
\]

Finally, the CD$_{\text{ind}}$ generated by a molecular dipole strength $P$ is can be written as
\[
\text{CD}_{\text{ind}} = A^+_m - A^-_m = \frac{\eta_{mol} \omega}{4} \text{Im}(\varepsilon_m) \int \frac{1}{r^3} \text{Re}\{-i\kappa e^{-ikr}[E^m_{nx} B^m_{nx} + E^m_{ny} B^m_{ny}]\} dV.
\tag{11}
\]

According to Eq. (11), there are linear relations between the CD$_{\text{ind}}$ and number density of chiral molecules, chirality parameter of chiral molecules, electric field strength in nanostructure, magnetic flux density in chiral molecular region. Moreover, the CD$_{\text{ind}}$ exponentially increases with the inverse cube of distance between chiral molecules and nanostructure.

III. STRUCTURE AND COMPUTATIONAL METHOD

In order to prove Eq. (11), we propose a graphene nanohole metasurface (GNM) and chiral molecule composed system. Generally, the SPR wavelength of graphene is located in microwave region, and the condition $k_\pm r \ll 1$ is completely satisfied. Figure 1(a) depicts the proposed GNM-chiral molecule composed system. The GNM was supposed to be immersed in the bottom of 100 nm thick chiral molecule solution. The periods were fixed as $P_x = P_y = 600 \text{ nm}$, and radius of nanohole was chosen to be 150 nm. Generally, the chiral molecules have absorption and CD$_{\text{ind}}$ peak located in UV region [1], for example the $\alpha$-helix has an absorption and CD$_{\text{mol}}$ peak located at 192 nm. Therefore, the chiral molecules are modeled with complex refractive index and chirality parameter that has absorption and CD$_{\text{ind}}$ peak at 192 nm.

Finite element method (FEM) was used to perform the simulations. To simulate for chiral medium, Maxwell equations were modulated by implementing a chiral constitutive relation. The constitutive relation for the chiral medium is given by [26]
\[
D = \varepsilon_0 \varepsilon_e E - \frac{i\kappa}{c} H,
\]
\[
B = \mu_0 \mu_c H + \frac{i\kappa}{c} E, \tag{12}
\]
where, $\varepsilon_e$, $\mu_c$ are the relative permittivity and magnetic permeability of the chiral molecule, $\kappa$ is the molecular chirality parameter, $\varepsilon_0$ and $\mu_0$ are the vacuum permittivity and magnetic permeability, respectively. The permittivity $\varepsilon_e$ and molecular chirality parameter $\kappa$ are given by [24]
\[
\varepsilon_e = \varepsilon_{\text{sol}} + n_{mol} \omega_0 d \frac{\omega_0^2 - \omega^2 + i\omega\Gamma}{(\omega_0^2 - \omega^2)^2 + \omega^2\Gamma^2},
\]
\[
\kappa = n_{mol} \omega R_{\text{rot}} \frac{\omega_0^2 - \omega^2 + i\omega\Gamma}{(\omega_0^2 - \omega^2)^2 + \omega^2\Gamma^2}, \tag{13}
\]
where, $\varepsilon_{\text{sol}}$ is the relative permittivity of solvent, $d$ represents the electric dipole strength, $R_{\text{rot}}$ is the rotatory strength, $\omega_0$ is molecular absorption band frequency. The parameters were set as $\varepsilon_{\text{sol}} = 1.77$, $d = 1.6 \times 10^{-9} \text{m}^3 \text{s}^{-1}$, $R_{\text{rot}} = 1.6 \times 10^{-11} \text{m}^3 \text{s}^{-1}$, $n_{mol} = 5 \times 10^{22} \text{m}^{-3}$, $\Gamma = 3.77 \times 10^{14} \text{s}^{-1}$, $\omega_0 = 9.8 \times 10^{13} \text{Hz}$. The chirality parameter of a chiral molecule is represented by a matrix. The initial molecular orientation is defined as parallel to $z$-axis, and the unitary transformation $\kappa_i = R(\varphi) \kappa_i R^{-1} (\varphi)$ 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 [20].

The conductivity of graphene was computed within the local random phase approximation, which is a function
of the frequency of incident light [29, 30]:

\[
\sigma(\omega) = \frac{2ikBT}{\pi\hbar^2(\omega^2 - \omega_0^2)} \ln[2\cosh(\frac{e\omega}{2k_BT})] + \frac{\epsilon^2}{4\pi\hbar^2} \left\{ 0.5 + \frac{1}{\pi} \arctan(\frac{-\hbar\omega + 2E_f}{2k_BT}) \right\} - \frac{i}{2\pi} \ln\left[ \frac{(\hbar\omega - 2E_f)^2 + (2k_BT)^2}{(\hbar\omega + 2E_f)^2 + (2k_BT)^2} \right],
\]

(14)

where \( T \) is temperature, \( \tau \) is carrier relaxation time, \( k_B \) is Boltzmann constant, \( v_F \) is Fermi velocity, \( k_BT \) and \( E_f \) represent temperature energy and Fermi energy, respectively. The parameters were set as \( T = 300K \), \( E_f = 0.6eV \), \( v_F = c/300 \) (\( c \) is speed of light), \( \tau = \frac{\rho_{DC}E_f}{ev_F} \) and \( \rho_{DC} = 10^4 \text{ cm/Vs} \).

**IV. RESULTS AND DISCUSSION**

Mechanism of circular dichroism induction: symmetry breaking of electric field in achiral nanostructure. The better aspect of the theory is that it can explain the mechanism of \( CD_{ind} \) induction. We first explain the mechanism of generating different electric fields in nanostructure under RCP and LCP illuminations. The racemic mixture of chiral molecule does not possess chiral properties and it cannot induce \( CD_{ind} \). The radiated electric field by the racemic mixture is

\[
\mathbf{E}^d_{\pm} = \frac{\omega\eta_{mol}^{\alpha}}{2r^2k} \left[ (G_+ + G_-)(3\hat{u}_r\hat{u}_r - I) \right] \cdot \mathbf{E}^d_{\pm}.
\]

(15)

The averaged electric field in achiral nanostructure has symmetry \( \mathbf{E}^{d+}_{\pm} = \mathbf{E}^{d-}_{\pm} \), \( \mathbf{E}^{d+}_{\pm} = \mathbf{E}^{d-}_{\pm} \), and \( \mathbf{E}^{d+}_{\pm} = \mathbf{E}^{d-}_{\pm} \). The symmetry of radiated electric field \( \mathbf{E}^{d+}_{\pm} \) in achiral nanostructure is the same as that of original electric field \( \mathbf{E}^d_{\pm} \), thus the symmetry of electric field does not break. The radiated electric field by pure chiral molecule is given in Eq. (6). Calculating the increments of electric field \( \Delta \mathbf{E}^d_{\pm} = \mathbf{E}^d_{\pm} - \mathbf{E}^{d+}_{\pm} \) by molecular chirality, we derive

\[
\Delta \mathbf{E}^d_{\pm} = \mathbf{E}^d_{\pm} - \mathbf{E}^{d+}_{\pm} = \frac{\omega\eta_{mol}^{\alpha}}{2r^2k} \left[ (G_+ + G_-)(3\hat{u}_r\hat{u}_r - I) \right] \cdot \mathbf{B}^\times_{\pm}.
\]

(16)

The \( x \) component of electric field increments \( \Delta \mathbf{E}^{dx}_{\pm} \) generated by dipole moment along \( x \) direction is

\[
\Delta \mathbf{E}^{dx}_{\pm} = \frac{-i\eta_{mol}k}{4r^3} \mathbf{B}^{\times y}_{\pm} e^{-ikr}.
\]

(17)

In a similar manner, the \( y \) component of electric field increments \( \Delta \mathbf{E}^{dy}_{\pm} \) generated by dipole moment along \( y \) direction is

\[
\Delta \mathbf{E}^{dy}_{\pm} = \frac{-i\eta_{mol}k}{4r^3} \mathbf{B}^{\times x}_{\pm} e^{-ikr}.
\]

(18)

The \( \Delta \mathbf{E}^{dx}_{\pm} \) has symmetry \( \Delta \mathbf{E}^{dx}_{\pm} = -\Delta \mathbf{E}^{dx}_{\pm} \) while \( \mathbf{E}^{dx}_{\pm} = \mathbf{E}^{dx}_{\pm} \), and \( \Delta \mathbf{E}^{dy}_{\pm} \) has \( \Delta \mathbf{E}^{dy}_{\pm} = -\Delta \mathbf{E}^{dy}_{\pm} \) while \( \mathbf{E}^{dy}_{\pm} = \mathbf{E}^{dy}_{\pm} \). In this way, symmetry of electric field in nanostructure is broken by electric field radiated by chiral molecule, and the electric field in nanostructure is different for RCP and LCP illumination. If the electric field increases for one handedness of light, it decreases for opposite handedness of light in same quantity (Fig. S4). This is the mechanism of \( CD_{ind} \) induction in chiral molecule-achiral nanostructure near field interaction.

Comparison of the analytical results to the simulation results. To prove the theory, we proposed the GMN-chiral molecule composed system, and investigated the \( CD_{ind} \) properties. Figure 2(a) depicts the absorption spectra of GNM. Three SPR modes (marked as I, II, and III) in the absorption spectrum are located at wavelengths 13.6 \( \mu \text{m} \), 10.8 \( \mu \text{m} \) and 9.2 \( \mu \text{m} \), respectively. Figure 2(b) depicts the surface charge distribution on GNM. The red color corresponds to positive charge, and the blue color corresponds to negative charge. Mode I is a dipolar mode because the positive and negative charges are clearly separated in two regions. Mode II is a hexapolar mode, the positive and negative charges are located in
FIG. 3. (a) The CD$_{ind}$ spectra for different molecule densities. (b) Linear fitting (sold line) of CD$_{ind}$ and molecule densities.

FIG. 4. (a) The CD$_{ind}$ spectra for different chirality parameter. (b) Linear fitting (sold line) of CD$_{ind}$ (Square) and chirality parameters at the three modes.

the six apexes of hexagon. Mode III is an octupolar mode composed of a hexapolar and dipolar mode. Figure. 2(c) shows the CD$_{ind}$ spectra of GNM. Three CD$_{ind}$ dispersions appear at three modes. Absorption and CD$_{ind}$ are largest at mode I, and smallest at mode III. The CD$_{ind}$ induced by oriented molecules is larger than that in the case of the isotropic molecules and it obtains the maximum when molecular orientation is parallel to x-axis[20]. Therefore, molecular orientation is set as parallel to x-axis in the following.

In order to prove the linear relation between molecular density and CD$_{ind}$, the CD$_{ind}$ for different molecular densities are calculated. For the sake of simplicity, the density of molecule is normalized by $\hat{n} = \frac{n_{mol}}{6.25 \times 10^{21} m^{-3}}$. Figure 3(a) presents the CD$_{ind}$ spectra for different molecule number densities $\hat{n}$. As expected, the CD$_{ind}$ peak value increases with the increase of $\hat{n}$. In order to compare with the analytical result, the linear fitting was made at wavelengths of three modes. Figure 3(b) shows linear fitting of $\hat{n} - CD_{ind}$ relation at mode I (black line), mode II (red line) and mode III (blue line). It can be seen that there are indeed linear relations between $\hat{n}$ and CD$_{ind}$, and the goodness of fitting is 0.99. This is consistent with analytical result of Eq. (11).

Similarly, to prove the linear relation between molecular parameter $\kappa$ and CD$_{ind}$, the CD$_{ind}$ for different molecular chirality parameters are calculated. For the sake of simplicity, the chirality parameter is normalized by $\hat{\kappa} = \frac{\kappa_{mol}}{\kappa_{mol}^0}$. Here, $\kappa_{mol}^0$ is the initial molecular chirality parameter which corresponds to $R_{mol} = 1.6 \times 10^{-11} m^3 s^{-1}$ and $n_{mol} = 5 \times 10^{22} m^{-3}$. The CD$_{ind}$ spectra with the change of $\hat{\kappa}$ is given by Fig. 4(a). The CD$_{ind}$ increases with increasing of $\hat{\kappa}$ at all three modes. Figure 4(b) shows the linear fitting at mode I (yellow line), mode II (olive line) and mode III (orange line). The fitting shows linear relationships between $\hat{\kappa}$ and CD$_{ind}$. Same as fitting of $\hat{n} - CD_{ind}$, goodness of $\hat{\kappa} - CD_{ind}$ is 0.99. These results further prove proposed theory.

In an effort to determine the relation between CD$_{ind}$ and molecule-nanostructure distance $r$, the distance $h_r$
from the GNM to bottom of molecular region is changed by a step of 10 nm. As the GNM is moved from the bottom to the center of the chiral medium, the CD$_{ind}$ at mode I increases and approaches the maximum when GNM reaches the center of chiral medium. The CD$_{ind}$ at mode II, III decreased with increase of $h_r$. According to Eq. (11), the relationship between CD$_{ind}$ and $e^{-ikr}r^3$ is linear, but electric fields $E^{ns}$ and magnetic fields $B^c$ change with $h_r$ thereby affecting the CD$_{ind}$. Through comparing the $E^{ns} \cdot B^c$ and $E^{ns}/r^3B^c$ spectra (Fig 5S), we concluded that the influence of distance $r$ is obvious, and the $E^{ns}/r^3B^c$ spectra are more close to CD$_{ind}$ spectra. To eliminate the effect of $E^{ns}$ and $B^c$ changes and inconvenience for fitting due to different peak positions for different $h_r$, three off-resonance wavelengths 18 $\mu$m, 19 $\mu$m and 20 $\mu$m are chosen to get $e^{-ikr} - CD_{ind}$ relation, because effects of mode shape changes on $E^{ns}$ and $B^c$ for different $h_r$ are negligible at the three wavelengths.

To get $\frac{e^{-ikr}}{r^3} - CD_{ind}$ relation, the averaged molecule-nanostructure distance is calculated by $r = \frac{1}{i} \sum (z_i - z_0)$, here, $z_0$ is the z coordinate of graphene and the $z_i$ is the z coordinate of chiral molecule layers. Figure 5(b) shows the linear fitting at the three wavelengths. The fitting shows linear relations between $\frac{e^{-ikr}}{r^3}$ and CD$_{ind}$, and the $\frac{e^{-ikr}}{r^3} - CD_{ind}$ relations are well consistent with the analytical results.

For the purpose of investigating the effects of $E^{ns}$ and $B^c$ on the CD$_{ind}$, we slightly change the structural parameters to regulate $E^{ns}$ and $B^c$ (Fig. S6). A small additional hole is set at the center of every four adjacent original holes. The radius of additional hole is $R_\alpha$ and is changed from 20 nm to 100 nm by a step of 20 nm. Figure 6(a) shows the CD$_{ind}$ spectra for different $R_\alpha$. After adding the hole, the CD$_{ind}$ decreases at mode I, increases at mode II and mode III. From Figure 6 it can be observed that the shapes of the $E^{ns}$, $B^c$ and CD$_{ind}$ are similar, the reason is that CD$_{ind}$ spectra is regulated by the electric field $E^{ns}$ (Fig.6(b)) and magnetic field $B^c$ (Fig.6(c)). At mode I, CD$_{ind}$ is increased by $E^{ns}$ and $B^c$, whereas at mode II and III the CD gets decreased by $E^{ns}$ and $B^c$ which agrees with Eq. (11).

V. CONCLUSION

Based on chiral molecule dipole approaching and chiral molecule-achiral nanostructure near field approximation, we present a theory for quantitative analysis of induced circular dichroism (CD$_{ind}$) from chiral molecule-achiral
structure near field interaction. The theory clarifies the mechanism of CD\textsubscript{ind} induction which is symmetry breaking of near field by dipole radiated electric field. Furthermore, the CD\textsubscript{ind} properties of graphene nanohole meta-surface and chiral molecule coupled system are simulated to prove the relation. The simulated results are consistent with the theoretical ones. There are linear relations between the CD\textsubscript{ind} and molecular density, molecular chirality parameter, and the CD\textsubscript{ind} exponentially increases with the inverse cube of molecule-nanostructure distance. Finally, a smaller hole is added at the center of four original holes to regulate the near field, and investigate the effect of electric field E\textsuperscript{ns} in nanostructure and magnetic field B\textsuperscript{c} in chiral molecule region on the CD\textsubscript{ind}. As expected, the CD\textsubscript{ind} is proportional to electric field E\textsuperscript{ns} and magnetic field B\textsuperscript{c}. Both, the analytic and simulation results show two ways of enhancing the CD\textsubscript{ind}. First, decreasing the molecule-nanostructure distance, and second, enhancing magnetic field in molecular region or enhancing electric field inside nanostructure. These results are useful for analyzing experimental or calculated data, and beneficial for realizing supersensitive detection of molecular chirality by plasmon enhancing technique.

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

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I. CIRCULAR DICHROISM INDUCTION BY DIPOLE RADIATED ELECTRIC FIELD

Circular dichroism (CD) is absorption difference of right circularly polarized (RCP) and left circularly polarized (LCP) light, and it is a characteristic property of chiral media. The essential difference between chiral and achiral media lies in the constitutive relations. The constitutive relation for a chiral medium is given by [1]

\[
\begin{align*}
D &= \varepsilon_0 \varepsilon_c E - \frac{i \kappa_c}{c} H \\
B &= \mu_0 \mu_c H + \frac{i}{c} E
\end{align*}
\] (S1)

where, \(\varepsilon_c, \mu_c\) are respectively the relative permittivity and magnetic permeability of the chiral molecule, \(\kappa\) is the molecular chirality parameter, \(\varepsilon_0\) and \(\mu_0\) are the vacuum permittivity and magnetic permeability, respectively.

Generally, the absorption of a nanostructure is given by [2]

\[
A_{ns}^{\pm} = \frac{\omega}{2} \int_{V_{ns}} [\text{Im}(\varepsilon_{ns})(E_{ns}^0 \cdot E_{ns}^{0*}) + \text{Im}(\mu_{ns})(H_0^0 \cdot H_0^{0*})] dV,
\] (S2)

where, \(\omega\) is angular frequency of CPL, \(\varepsilon_{ns}\) and \(\mu_{ns}\) are relative permittivity and permeability of nanostructure, \(E_{ns}^0\) and \(H^0_{ns}\) are electric and magnetic field strengths. The magnetic susceptibility of plasmonic nanostructure is very small, therefore magnetic loss can be neglected. In presence of chiral molecule, \(E_{ns}^0\) will be affected by the electric field radiated by dipole moment of chiral molecule. Then Eq. (S2) can be written [3]

\[
A_{ns}^{\pm} = \frac{\omega}{2} \text{Im}(\varepsilon_{ns}) \int_{V_{ns}} (E_{ns}^0 + E_{ns}^d)^* \cdot (E_{ns}^0 + E_{ns}^d) dV,
\] (S3)

where, \(E_{ns}^d\) is dipole radiated electric field which 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

\[
E^d(r) = \int_{V'} \left( \overline{G}_{ee}(r-r') \quad \overline{G}_{em}(r-r') \right) \left( \begin{array}{c} J \\ M \end{array} \right)
\] (S4)

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

\[
\begin{align*}
\overline{G}_{ee}(r) &= \overline{G}_{ee+}(r) + \overline{G}_{ee-}(r) \\
\overline{G}_{ee\pm}(r) &= -\frac{in}{2} [(k_{\pm} G_{\pm}(r)) \mathbb{1} \pm \nabla G_{\pm}(r)] + \mathbb{1} \pm \nabla \left( \frac{G_{\pm}(r)}{k_{\pm}} \right), \\
\overline{G}_{em}(r) &= \overline{G}_{em+}(r) + \overline{G}_{em-}(r) \\
\overline{G}_{em\pm}(r) &= -\frac{1}{2} (\pm(k_{\pm} G_{\pm}(r)) \mathbb{1} \pm \nabla G_{\pm}(r) \times \mathbb{1} \pm \nabla \left( \frac{G_{\pm}(r)}{k_{\pm}} \right))
\end{align*}
\] (S5)
where $G_{\pm}(r) = \frac{e^{-ik_{\pm}r}}{4\pi r}$, $k_{\pm} = k_{0}(1 \pm \frac{\kappa}{n_c})$ and $n_c$ is refractive index of chiral medium. Using the approximation $k_{\pm}r \ll 1$ for near field calculation, and calculating the gradient terms in Eq. (S5) becomes

$$\nabla G_{\pm}(r) = (-ik_{\pm} + \frac{1}{r})G_{\pm}(r)\hat{u}_r \approx \frac{1}{r}G_{\pm}(r)\hat{u}_r$$

$$\nabla \nabla G_{\pm}(r) = [(ik_{\pm} + \frac{1}{r^2})G_{\pm}(r)\hat{u}_r\hat{u}_r - (ik_{\pm} + \frac{1}{r^2})\frac{1}{r}(I - \hat{u}_r\hat{u}_r)]$$

(S6)

where $\hat{u}_r$ is unit vector in $r$ direction, $\hat{u}_r\hat{u}_r$ is dyadic, and can be written as,

$$\hat{u}_r = \frac{1}{r} \begin{pmatrix} x \\ y \\ z \end{pmatrix}, \quad \hat{u}_r\hat{u}_r = \frac{1}{r^2} \begin{pmatrix} x^2 & xy & xz \\ yx & y^2 & yz \\ zx & zy & z^2 \end{pmatrix}.$$  

Thus the dyadic Green functions becomes

$$\overline{G}_{ee+}(r) = \frac{-i\eta}{2} \left[ k_{+}G_{+}(r) - \frac{1}{r}G_{+}\hat{u}_r \times \frac{3}{r^2}k_{+}\hat{u}_r\hat{u}_r \right] - \frac{1}{r^2}G_{+}I,$$

$$\overline{G}_{ee-}(r) = \frac{-i\eta}{2} \left[ k_{-}G_{-}(r) + \frac{1}{r}G_{-}\hat{u}_r \times \frac{3}{r^2}k_{-}\hat{u}_r\hat{u}_r \right] - \frac{1}{r^2}G_{-}I.$$  

(S7)

Implementing $k_{\pm}r \ll 1$, we derive

$$\overline{G}_{ee+}(r) = \frac{-i\eta}{2r^2k} [3k_{+}\hat{u}_r\hat{u}_r - G_{+}I],$$

$$\overline{G}_{ee-}(r) = \frac{-i\eta}{2r^2k} [3k_{-}\hat{u}_r\hat{u}_r - G_{-}I],$$  

(S8)

where $k_{\pm} = k_{0}(1 \pm \frac{\kappa}{n_c})$. Since $\kappa$ is of the order of $10^{-5}$, we can approximate $k_{\pm} = k$. Finally, we derive the dyadic Green functions for near field

$$\overline{G}_{ee}(r) = \frac{-i\eta}{2r^2k} e^{-ikr \cos\theta} \frac{k}{n_c} \left[ 3\hat{u}_r\hat{u}_r - I \right],$$

$$\overline{G}_{em}(r) = \frac{-i\eta}{2r^2k} e^{-ikr \sin\theta} \frac{k}{n_c} \left[ 3\hat{u}_r\hat{u}_r - I \right] \approx 0.$$  

(S9)

The polarization strength of chiral molecule is given by [5]

$$\mathbf{P}_{\pm} = n_{mol}\alpha \mathbf{E}_{\pm}^c - n_{mol}\frac{i\kappa}{c} \mathbf{B}_{\pm}^c,$$  

(S10)

where $n_{mol}$ is the number density of molecule. The electric current can be calculated by $\mathbf{J}_{\pm} = \frac{\partial}{\partial t} \mathbf{P}_{\pm}$. Thus, the dipole radiated electric field is

$$\mathbf{E}_{\pm}^d = i\omega n_{mol}\alpha \overline{G}_{ee} \cdot \mathbf{E}_{\pm}^c + \omega n_{mol}\frac{\kappa}{c} \overline{G}_{em} \cdot \mathbf{B}_{\pm}^c.$$  

(S11)

The induced $CD_{ind}$ in the nanostructure is

$$CD_{ind} = \frac{\omega}{2} \text{Im}(\varepsilon_{ns}) \int_{V_{ns}} (\mathbf{E}_{+}^0 + \mathbf{E}_{+}^d)^* \cdot (\mathbf{E}_{+}^0 + \mathbf{E}_{+}^d) dV - \frac{\omega}{2} \text{Im}(\varepsilon_{ns}) \int_{V_{ns}} (\mathbf{E}_{-}^0 + \mathbf{E}_{-}^d)^* \cdot (\mathbf{E}_{-}^0 + \mathbf{E}_{-}^d) dV$$

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

(S12)

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

$$CD_{ind} = \frac{\omega}{2} \text{Im}(\varepsilon_{ns}) \int_{V_{ns}} 2\text{Re} \left( \mathbf{E}_{+}^0 \cdot \mathbf{E}_{+}^{dx} - \mathbf{E}_{-}^0 \cdot \mathbf{E}_{+}^{dx} \right) dV.$$  

(S13)
Replace $E^d$ to calculate $E^0_+ \cdot E^d_+$

\[
(E^0_+ \cdot E^d_+) = E^0_+ \cdot \left\{ \frac{\omega \mu_{\text{mol}} \alpha}{2\pi^2k} \left[ 3(G_+ + G_-) \hat{u}_z \hat{u}_r - (G_+ + G_-) \right] \cdot E^d_+ + \frac{-i\omega \mu_{\text{mol}} \kappa}{c} \left[ 3(G_+ + G_-) \hat{u}_z \hat{u}_r - (G_+ + G_-) \right] \cdot B^d_+ \right\}
\]

\[
= \frac{3\omega \mu_{\text{mol}} \alpha}{2\pi^2k} \left[ 3(G_+ + G_-) \right] \cdot E^d_+ + \frac{-3i\omega \mu_{\text{mol}} \kappa}{c} \left[ 3(G_+ + G_-) \hat{u}_z \hat{u}_r - (G_+ + G_-) \right] \cdot B^d_+ + \frac{3\omega \mu_{\text{mol}} \alpha}{2\pi^2k} \left( G_+ + G_- \right) E^d_+ \cdot E^d_+ - \frac{-3i\omega \mu_{\text{mol}} \kappa}{c} \left( G_+ + G_- \right) E^d_+ \cdot B^d_+ + \frac{3\omega \mu_{\text{mol}} \alpha}{2\pi^2k} \left( G_+ + G_- \right) E^d_+ \cdot E^d_+
\]

If we suppose that the incident light propagates along $z$ direction and the achiral nanostructure is symmetric to $y$ or $x$-axis (any achiral structure has a symmetry axis), then near field in the $xy$-plane has properties $E^d_+(x, y) = E^d_c(x, y)$, $E^d_+(x, y) = -E^d_c(x, y)$, $E^d_-(x, y) = E^d_c(x, y)$, $B^d_c(x, y) = -B^d_0(x, y)$, $B^d_c(x, y) = B^d_0(x, y)$, $B^d_0(x, y) = -B^d_0(x, y)$. Applying coordinate transformations $y \to -y$ or $x \to -x$ (depending on the axis of symmetry) to $\frac{1}{r^2} E_+^{0 \alpha x}(x', y') E_+^{cx}(x, y)$, we derive

\[
\frac{1}{r^2} E_+^{0 \alpha x}(x', y') E_+^{cx}(x, y) = \frac{1}{r^2} E_+^{0 \alpha x}(x', y') E_+^{cx}(x, y).
\]

Eq. (15) means the term $\frac{1}{r^2} E_+^{0 \alpha x}(x', y') E_+^{cx}(x, y)$ is equal for RCP and LCP illumination. Using the same coordinate transformations to other terms, we finally derive

\[
E^0_+ \cdot E^d_+ - E^0_+ \cdot E^d_+ = \frac{-3i\omega \mu_{\text{mol}} \kappa}{r^2k} \left( G_+ + G_- \right) \left( \cos^2(\alpha) E^d_+ B^d_+ + \sin^2(\alpha) E^d_+ B^d_+ \right)
\]

(16)

Eq(16) is general form of field intensity difference generated by chiral molecular dipole. The $z$ component of near field is much smaller than $x$ and $y$ components, neglecting the $z$ component we derive

\[
E^0_+ \cdot E^d_+ - E^0_+ \cdot E^d_+ = \frac{-3i\omega \mu_{\text{mol}} \kappa}{r^2k} \left( G_+ + G_- \right) \left( \cos^2(\alpha) E^d_+ B^d_+ + \sin^2(\alpha) E^d_+ B^d_+ \right)
\]

(17)

For planar nanostructure (in our GNM-chiral molecule composed system, $x$ and $y$ are infinite while $z$ is up to 100 nm) the the distance $r$ can approximated as $r = (x^2 + y^2)^{1/2}$, then Eq. (17) becomes

\[
E^0_+ \cdot E^d_+ - E^0_+ \cdot E^d_+ = \frac{-3i\omega \mu_{\text{mol}} \kappa}{r^2k} \left( G_+ + G_- \right) \left( \cos^2(\alpha) E^d_+ B^d_+ + \sin^2(\alpha) E^d_+ B^d_+ \right)
\]

(18)

where $\alpha$ is angle between $r$ and $x$-axis. If the chiral molecule is uniformly arranged around the achiral structure, taking average values of $\sin^2(\alpha)$ and $\cos^2(\alpha)$, Eq(19) becomes

\[
E^0_+ \cdot E^d_+ - E^0_+ \cdot E^d_+ = \frac{-3i\omega \mu_{\text{mol}} \kappa}{r^2k} \left( G_+ + G_- \right) \left( E^d_+ B^d_+ + E^d_+ B^d_+ \right)
\]

(19)

Finally, we derive the CD induced generated by a molecular strength $P$ in the nanostructure

\[
CD_{\text{ind}} = \frac{\eta \mu_{\text{mol}} \omega}{4} \Im(\varepsilon_n) \int_{V_{\text{ns}}} \frac{1}{r^3} \Re\{ -i\kappa e^{-i\kappa r} [E^d_+ B^d_+ + E^d_+ B^d_+] \} dV.
\]

(20)
II. ANALYZING AND APPORIXISMATIONS

A. Symmetric properties of near field around achiral structure

Suppose if the incident light propagates along z direction, the near field in xy-plane has properties $E_x^+(x,y) = E_x^-(x,-y)$, $E_y^+(x,y) = -E_y^-(x,-y)$, $B_x^+(x,y) = -B_x^-(x,-y)$, $B_y^+(x,y) = B_y^-(x,-y)$, $B_z^+(x,y) = -B_z^-(x,-y)$. Figure S1 shows the electric field and magnetic flux distribution of GNM at mode II. The average electric and magnetic field values satisfy $E_x^+ = E_x^-$, $E_y^+ = -E_y^-$, $E_z^+ = E_z^-$, $B_x^+ = -B_x^-$, $B_y^+ = B_y^-$, $B_z^+ = -B_z^-$, $E_z^+ \ll E_x^+$, $E_y^+$ and $B_z^+ \ll B_x^+$, $B_y^+$. Figure S2 shows the average electric field and magnetic flux spectra of GNM.

B. Symmetry breaking of electric field in achiral nanostructure

The racemic mixture of chiral molecule has no chirality and it does not induce CD$_{ind}$. The pure chiral molecule breaks the electric field symmetry in achiral structure and induces the CD$_{ind}$ signal. The electric field radiated by the racemic mixture is

$$E_{rd}^\pm = \frac{\omega n \mu n_{mol}}{2\gamma^2 k} \left[ (G_+ + G_-)(3\hat{u}_r\hat{u}_r - I) \right] \cdot E_{rc}^\pm. \tag{S21}$$

The averaged electric field in achiral nanostructure and molecular region has symmetry $E_{0x}^+ = E_{0x}^-$, $E_{0y}^+ = -E_{0y}^-$, $E_{0z}^+ = E_{0z}^-$, and $E_{rc}^\pm$ satisfies the constitutive relations

$$D_{rc} = \varepsilon_0 \varepsilon_r E_{rc},$$
$$B_{rc} = \mu_0 \mu_r H_{rc}.$$

The symmetry of radiated electric field is the same as that of original electric field in achiral nanostructure and the symmetry is not broken.

If we calculate the difference of $E_{rd}^\pm$ and $E_{rc}^\pm$, we derive

$$\Delta E_{xz}^\pm = E_{xz}^d - E_{xz}^d = \frac{\omega n \mu n_{mol}}{2\gamma^2 k} \left[ (G_+ + G_-)(3\hat{u}_r\hat{u}_r - I) \right] \cdot B_{xz}^+. \tag{S22}$$

Calculating the electric field difference $\Delta E_{xz}^\pm$ which is generated by dipole moment $n_{mol}\mu P_x\hat{u}_x$ along x axis, we derive

$$\Delta E_{xz}^\pm = \frac{i\omega n n_{mol} \mu}{4\gamma^3} B_{xz}^+ e^{-ikr}. \tag{S23}$$
FIG. S2. Average $x$, $y$ and $z$ components of (a-c) electric field $E^c$, (d-f) magnetic field $B^c$ and (g-i) electric field $E^{ns}$.

Since $B^{cx}$ is antisymmetric for RCP and LCP, therefore $x$ components of magnetic field in the chiral molecular region for RCP and LCP are opposite to each other. As a result, the increments of dipole radiated electric fields compared to the racemic mixture are also opposite. But $x$ components of electric field in nanostructure are same for RCP and LCP. Thus, electric field difference has generated inside in nanostructure and leads to $x$ component of CD$_{ind}$. Similarly, the electric field difference of $y$ and $z$ components are generated. Figure S3 is schematic illustration the origin of electric field strength difference in achiral nanostructure. Generally, the increments in electric field by chiral molecule as compared to racemic mixture are opposite to each other for RCP and LCP, if the electric field increments for RCP is positive, the electric field increments for LCP will be negative. Figure S4 shows the electric field increments in GNM, the results are same as analytical result.

FIG. S3. Schematic of electric field symmetry breaking in achiral nanostructure.
According to Eq. (S20), there is a linear relationship between $\text{CD}^{\text{ind}}$ and $e^{i \kappa r} r^3$. But the electric fields $E^{\text{ns}}$ and magnetic fields $B^c$ change with $h_r$, thereby affecting the $\text{CD}^{\text{ind}}$. Thus we analyzed the $E^{\text{ns}} \cdot B^c$, $E^{\text{ns},}\frac{B^c}{r^3}$ and $\text{CD}^{\text{ind}}$. Figure S5 shows the $E^{\text{ns}} \cdot B^c$, $E^{\text{ns},}\frac{B^c}{r^3}$ and $\text{CD}^{\text{ind}}$ spectra. From $E^{\text{ns}} \cdot B^c$ and $E^{\text{ns},}\frac{B^c}{r^3}$ spectra it can be observed that the influence of distance $r$ is obvious, and the $E^{\text{ns},}\frac{B^c}{r^3}$ spectra are more close to $\text{CD}^{\text{ind}}$ spectra. Another inconvenience to do the linear fitting is that the peak wavelengths are different for different $h_r$. Therefore, for excluding the effect of $E^{\text{ns}}$ and $B^c$, we chose the $18\,\text{nm}$, $19\,\text{nm}$ and $20\,\text{nm}$ for linear fitting. These are off-resonance wavelengths which are off-resonance wavelength and electric and magnetic field strengths are almost the same for different $h_r$.

D. Effect of electric field in nanostructure and magnetic field in molecular region on induced CD

The electric field strength $E^m$ and magnetic field density $B^c$ also affect the $\text{CD}^{\text{ind}}$. For the purpose of investigating the effects of $E^m$ and $B^c$ on the $\text{CD}^{\text{ind}}$, we slightly change the structure to regulate these factors. One additional hole is set at the center of every four adjacent original holes. The radius of additional holes is changed from $20\,\text{nm}$ to $100\,\text{nm}$ by step of $20\,\text{nm}$. The Figure S6 shows the charge distribution at three modes with changing of $R_\alpha$. The Figure S6 shows SPR strength gradually decreases at mode I, and increases at mode II and III. Thereby, the electric field strength $E^m$ and magnetic flux density $B^c$ decreases at mode I, and increases at mode II and I.
FIG. S6. The charge distribution of GNM with different radius $R_\alpha$.

III. CALCULATION OF CHIRAL MEDIUM BY FEM

We used COMSOL software for the simulation. For calculation of chiral medium we changed part of equations through implementing chiral constitutive relation\(^6\). To confirm our calculation results, we applied these modifications to solve for the transmission coefficients for a chiral slab and compared the simulation results of the co- and cross-components of the transmission coefficients at normal incidence from a slab with thickness $L$. The results are:

$$T_{co} = \frac{2\eta_0^2 \cos(kL) \cos(\kappa k L)}{2\eta_0 \cos(kL) + i(\eta_0^2 + \eta^2) \sin(kL)}$$ \hspace{1cm} (S24)

$$T_{cr} = \frac{2\eta_0 \cos(kL) \sin(\kappa k L)}{2\eta_0 \cos(kL) + i(\eta_0^2 + \eta^2) \sin(kL)}$$

Figure S7 depicts infinite chiral slab in air and calculation result. The simulated and analytical results are in a good agreement.

For anisotropic chiral medium, the constitutive relations are expressed by tensorial chirality parameter. The unitary transformations are used for rotation of orientation, for example, for rotation about $y$ axis the constitutive relations are modulated as

$$\begin{pmatrix} D^x \\ D^y \\ D^z \end{pmatrix} = \varepsilon_0 \varepsilon_c \begin{pmatrix} E^x \\ E^y \\ E^z \end{pmatrix} - \frac{i}{c} \begin{pmatrix} \cos(\alpha) & 0 & -\sin(\alpha) \\ 0 & 1 & 0 \\ \sin(\alpha) & 0 & \cos(\alpha) \end{pmatrix} \begin{pmatrix} \kappa_{xx} & \kappa_{xy} & 0 \\ \kappa_{yx} & \kappa_{yy} & 0 \\ 0 & 0 & \kappa_{zz} \end{pmatrix} \begin{pmatrix} \cos(\alpha) & 0 & \sin(\alpha) \\ 0 & 1 & 0 \\ -\sin(\alpha) & 0 & \cos(\alpha) \end{pmatrix} \begin{pmatrix} B^x \\ B^y \\ B^z \end{pmatrix}$$ \hspace{1cm} (S25)

$$\begin{pmatrix} B^x \\ B^y \\ B^z \end{pmatrix} = \mu_0 \mu_c \begin{pmatrix} H^x \\ H^y \\ H^z \end{pmatrix} + \frac{i}{c} \begin{pmatrix} \cos(\alpha) & 0 & -\sin(\alpha) \\ 0 & 1 & 0 \\ \sin(\alpha) & 0 & \cos(\alpha) \end{pmatrix} \begin{pmatrix} \kappa_{xx} & \kappa_{xy} & 0 \\ \kappa_{yx} & \kappa_{yy} & 0 \\ 0 & 0 & \kappa_{zz} \end{pmatrix} \begin{pmatrix} \cos(\alpha) & 0 & \sin(\alpha) \\ 0 & 1 & 0 \\ -\sin(\alpha) & 0 & \cos(\alpha) \end{pmatrix} \begin{pmatrix} E^x \\ E^y \\ E^z \end{pmatrix}$$ \hspace{1cm} (S26)
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