

Supporting Information

S1. Verifying the correctness of the numerical results using the coupled multipole method

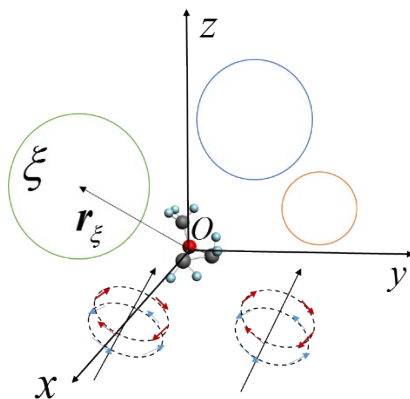


Fig. S1. Geometry and coordinate of a hybrid system consisting of a chiral molecule and a finite cluster of NPs. The system is scattered by an incident polarized light; \mathbf{r}_ξ is the position vector of the ξ^{th} NP. The molecule is put at the origin of the coordinate.

Here we present a coupled multipole method which can be used to verify the correctness of the T-matrix method provided in the main text. Consider the semi-classical hybrid system discussed in the main text. The chiral molecule is put at the origin of the coordinate, in the vicinity of a cluster of NPs, as shown in Fig. 1. The molecule-NP system is excited by the circular polarized light. Rewrite equations (6-8) in the main text, the *induced* electric dipole, magnetic dipole, and electric quadrupole of the chiral molecule are given by

$$\mu_a^{(E1)} \approx \mathcal{D}_{ab}^{\%} E_{tot b}^{(0)} + \mathcal{G}_{ab}^{\%} B_{tot b}^{(0)} + \frac{1}{3} \mathcal{A}_{a,bc}^{\%} \partial_b E_{tot c}^{(0)} \quad (S1)$$

$$m_a^{(M1)} \approx -\mathcal{G}_{ba}^{\%} E_{tot b}^{(0)} \quad (S2)$$

$$\Theta_{ab}^{(E2)} \approx \mathcal{A}_{c,ab}^{\%} E_{tot c}^{(0)} \quad (S3)$$

in the Cartesian coordinate.

On the other hand, considering the size of the system is much smaller than the wavelength of

the incident wave, the NPs can also be modeled as point electric dipoles, magnetic dipoles and electric quadrupoles. For the ξ^{th} NP, the electric dipole $\boldsymbol{\mu}^{(\xi)}$, magnetic dipole $\mathbf{m}^{(\xi)}$, and electric quadrupole $\hat{\Theta}^{(\xi)}$ are proportional to the local electric field $\mathbf{E}_{loc-\xi}$, magnetic field $\mathbf{B}_{loc-\xi}$, and electric field gradient $\nabla\mathbf{E}_{loc-\xi}$, respectively:

$$\boldsymbol{\mu}^{(\xi)} \approx \alpha_{\xi} \mathbf{E}_{loc-\xi} \quad (S4)$$

$$\mathbf{m}^{(\xi)} \approx G_{\xi} \mathbf{B}_{loc-\xi} \quad (S5)$$

$$\hat{\Theta}^{(\xi)} \approx A_{\xi} \left[\frac{\nabla\mathbf{E}_{loc-\xi} + (\nabla\mathbf{E}_{loc-\xi})^T}{2} \right]. \quad (S6)$$

Here α_{ξ} , G_{ξ} and A_{ξ} are the electric dipole, magnetic dipole, and electric quadrupole polarizabilities of the ξ^{th} NP. The local electric field $\mathbf{E}_{loc-\xi}$ is equal to the total fields in space except the scattering fields of the ξ^{th} NP :

$$\mathbf{E}_{loc-\xi} = \mathbf{E}_{tot} - \mathbf{E}_{s-\xi} \quad (S7)$$

with

$$\mathbf{E}_{s-\xi} = \mathbf{E}_{s-\xi}^{(0)} + \mathbf{E}_{s-\xi}^{(E1)} + \mathbf{E}_{s-\xi}^{(M1)} + \mathbf{E}_{s-\xi}^{(E2)}. \quad (S8)$$

Here $\mathbf{E}_{s-\xi}^{(X)}$ ($X=0; E1; M1; E2$) denotes the scattering field of the ξ^{th} NP, which are caused by the external incident field $\mathbf{E}_{inc}^{(0)}$, the *induced* electric dipole $\boldsymbol{\mu}^{(E1)}$, the *induced* magnetic dipole $\mathbf{m}^{(M1)}$, and the *induced* electric quadrupole $\hat{\Theta}^{(E2)}$ of the chiral molecule, respectively.

When the radii of NPs are small compared with the wavelength of the incident wave, absorption rates of the molecule and the NPs are given by: ^{S1, S2}

$$Q_{mol} = \frac{\omega_0}{2} \text{Im} \left[\boldsymbol{\mu}^{(E1)} \cdot \mathbf{E}_{tot}^{(0)*} + \mathbf{m}^{(M1)} \cdot \mathbf{B}_{tot}^{(0)*} + \frac{1}{6} \hat{\Theta}^{(E2)} : \left(\nabla\mathbf{E}_{tot}^{(0)} + (\nabla\mathbf{E}_{tot}^{(0)})^T \right)^* \right], \quad (S9)$$

$$Q_{NP} = \sum_{\xi=1}^{NP} Q_{NP-\xi}, \quad (S10)$$

with $Q_{NP-\xi}$ being the absorption rate of the ξ^{th} NP:

$$Q_{NP-\xi} = \frac{\omega}{2} \text{Im} \left[\boldsymbol{\mu}^{(\xi)} \cdot \mathbf{E}_{loc-\xi}^* + \mathbf{m}^{(\xi)} \cdot \mathbf{B}_{loc-\xi}^* + \frac{1}{6} \hat{\Theta}^{(\xi)} : \left[\nabla \mathbf{E}_{loc-\xi} + (\nabla \mathbf{E}_{loc-\xi})^T \right]^* \right]. \quad (\text{S11})$$

In deriving equation (S9) and equation (S11), the relation *Absorption* \approx *Extinction* has been used when the size of the system is much smaller than the wavelength. The sign ‘:’ in these equations denotes the double scalar product between the tensors.

Following the works by Bouř et. al. ^{S3, S4}, we define the generalized multipole moment (\mathbf{M}_0), polarizability (\mathbf{P}_0) for the molecule as

$$\mathbf{M}_0 = \begin{pmatrix} \boldsymbol{\mu}^{(E1)} \\ \mathbf{m}^{(M1)} \\ \mathbf{t} \\ \Theta^{(E2)} \end{pmatrix} \quad \mathbf{P}_0 = \begin{pmatrix} \mathcal{G} & \mathcal{G} & \mathbf{t}^T / 3 \\ -\mathcal{G} & \mathbf{0} & \mathbf{0} \\ \mathbf{t} & \mathbf{0} & \mathbf{0} \\ A & \mathbf{0} & \mathbf{0} \end{pmatrix}, \quad (\text{S12})$$

where $\mathbf{t}^{(E2)} = \left[\Theta_{u=1}^{(E2)} \quad \Theta_{u=2}^{(E2)} \quad \mathbf{L} \quad \Theta_{u=5}^{(E2)} \right]^T$, A is a 5×3 matrix with $A_{ij} = \mathcal{A}_{j,u=i}^{\mathcal{A}}$, and A^{tT} is a 3×5 matrix with $[A^{tT}]_{ij} = \mathcal{A}_{i,u=j}^{\mathcal{A}} + \mathcal{A}_{i,\bar{u}=j}^{\mathcal{A}} (1 - \delta_{j,4} - \delta_{j,5}) - (\delta_{j,4} + \delta_{j,5}) \mathcal{A}_{i,33}^{\mathcal{A}} \cdot \delta_{x,y}$ is the Kronecker delta function. $u = j$ and $\bar{u} = j$ are introduced here for simplification of the notation, and the conventions $u = 1/u = 2/u = 3/u = 4/u = 5$ stand for 12/13/23/11/22 and $\bar{u} = 1/\bar{u} = 2/\bar{u} = 3/\bar{u} = 4/\bar{u} = 5$ represent 21/31/32/11/22, respectively. Likewise, the general multipole moment (\mathbf{M}_ξ) and polarizability (\mathbf{P}_ξ) for the ξ^{th} NP are defined as

$$\mathbf{M}_\xi = \begin{pmatrix} \boldsymbol{\mu}^{(\xi)} \\ \mathbf{m}^{(\xi)} \\ \mathbf{t} \\ \Theta^{(\xi)} \end{pmatrix} \quad \mathbf{P}_\xi = \begin{pmatrix} \alpha_\xi \bar{\bar{I}}(3) & \mathbf{0} & \mathbf{0} \\ \mathbf{0} & G_\xi \bar{\bar{I}}(3) & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & A_\xi \bar{\bar{I}}(5) \end{pmatrix}, \quad (\text{S13})$$

where $\bar{\bar{I}}(x)$ is an unit matrix with a dimension of x . In the absence of the molecule, the multipole moments of the NPs can be solved by the following equation:

$$\mathbf{M}(0) = P_{inc} \left[\bar{\bar{I}} - X P_{inc} \right]^{-1} F_{inc}, \quad (\text{S14})$$

where $\mathbf{M}(0) = [\mathbf{M}_0^T(0) \quad \mathbf{M}_1^T(0) \quad \mathbf{M}_2^T(0) \quad \mathbf{L} \quad \mathbf{M}_{NP}^T(0)]^T$, F_{inc} is determined by the fields of external waves $(\mathbf{E}_{inc}^{(0)}, \mathbf{B}_{inc}^{(0)})$, and X is matrix which describes the interaction between

NPs in the system. Specifically, F_{inc} is given by $F_{inc} = [F_{inc-mol}^T \quad F_{inc-1}^T \quad \mathbf{L} \quad F_{inc-N}^T]^T$

where

$$\mathbf{F}_{inc-\zeta} = \begin{pmatrix} \mathbf{E}_{inc}(\mathbf{r}_\zeta) \\ \mathbf{B}_{inc}(\mathbf{r}_\zeta) \\ \mathbf{t} \nabla \mathbf{E}_{inc}(\mathbf{r}_\zeta) \end{pmatrix} \quad (\text{S15})$$

with $\mathbf{t} \nabla \mathbf{E}_{inc}(\mathbf{r}_\zeta)$ being a 5×1 matrix, of which elements are given by

$$[\mathbf{t} \nabla \mathbf{E}_{inc}(\mathbf{r}_\zeta)]_i = \frac{[\nabla \mathbf{E}_{inc}(\mathbf{r}_\zeta) + [\nabla \mathbf{E}_{inc}(\mathbf{r}_\zeta)]^T]_{u=i}}{2}. \quad P_{inc} \text{ is assembled by the general}$$

polarizability of the NPs, and can be expressed as

$$\mathbf{P}_{inc} = \begin{pmatrix} \mathbf{0} & \mathbf{0} & \dots & \mathbf{0} \\ \mathbf{0} & \mathbf{P}_1 & \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{P}_{NP} \end{pmatrix}. \quad (\text{S16})$$

At last, the expression of the matrix X is given by

$$\mathbf{X} = \begin{pmatrix} 0 & \mathbf{X}_{01} & \mathbf{L} & \mathbf{X}_{0N} \\ \mathbf{X}_{10} & 0 & \mathbf{L} & \mathbf{X}_{1N} \\ \mathbf{L} & \mathbf{L} & \mathbf{L} & \mathbf{L} \\ \mathbf{X}_{N0} & \mathbf{X}_{N1} & \mathbf{L} & 0 \end{pmatrix}, \quad (\text{S17})$$

where \mathbf{X}_{ij} is determined by the radiation fields at \mathbf{r}_i which are caused by the unit electric dipole,

magnetic dipole and electric quadrupole situated at \mathbf{r}_j . The specific express of \mathbf{X}_{ij} is given by

$$\mathbf{X}_{ij} = \begin{pmatrix} U_E^{(e)}(\mathbf{r}_i - \mathbf{r}_j) & U_E^{(m)}(\mathbf{r}_i - \mathbf{r}_j) & U_E^{(\ominus)}(\mathbf{r}_i - \mathbf{r}_j) \\ U_B^{(e)}(\mathbf{r}_i - \mathbf{r}_j) & U_B^{(m)}(\mathbf{r}_i - \mathbf{r}_j) & U_B^{(\ominus)}(\mathbf{r}_i - \mathbf{r}_j) \\ U_{\nabla E}^{(e)}(\mathbf{r}_i - \mathbf{r}_j) & U_{\nabla E}^{(m)}(\mathbf{r}_i - \mathbf{r}_j) & U_{\nabla E}^{(\ominus)}(\mathbf{r}_i - \mathbf{r}_j) \end{pmatrix} \quad (\text{S18})$$

with $U_Z^{(Y)}(\mathbf{r}_i - \mathbf{r}_j) = \begin{pmatrix} Z_x^{(Y-x)} & Z_x^{(Y-y)} & Z_x^{(Y-z)} \\ Z_y^{(Y-x)} & Z_y^{(Y-y)} & Z_y^{(Y-z)} \\ Z_z^{(Y-x)} & Z_z^{(Y-y)} & Z_z^{(Y-z)} \end{pmatrix}$ when $(Z \rightarrow E \text{ or } Z \rightarrow B)$ and $(Y \rightarrow e \text{ or } Y \rightarrow m)$.

$Z_m^{(Y)}$ denotes the m component of the electric/magnetic field at \mathbf{r}_i caused by an unit electric/magnetic dipole ($Y \rightarrow e$ for the electric dipole; $Y \rightarrow m$ for the magnetic dipole) situated at \mathbf{r}_j . $U_Z^{(\Theta)}(\mathbf{r}_i - \mathbf{r}_j)$ ($Z \rightarrow E \text{ or } Z \rightarrow B$) is a 3×5 matrix, of which elements are given by

$$[U_Z^{(\Theta)}]_{i,j} = \begin{cases} Z_i^{(\Theta_{u=j})} + Z_i^{(\Theta_{\bar{u}=j})} & (j = 1 \text{ or } 2 \text{ or } 3) \\ Z_i^{(\Theta_{u=j})} - Z_i^{(\Theta_{33})} & (j = 4 \text{ or } 5) \end{cases}. \quad (\text{S19})$$

The definition of $Z_m^{(\Theta_{u=j})}$ is similar to $Z_m^{(Y)}$, but it represents the field emitted by an unit electric quadrupole. $U_{\nabla E}^{(Y)}(\mathbf{r}_i - \mathbf{r}_j)$ ($Y \rightarrow e \text{ or } Y \rightarrow m$) is a 5×3 matrix, of which elements are given by $[U_{\nabla E}^{(Y)}]_{j,i} = \left([\nabla E^{(Y-i)}]_{u=j} + [\nabla E^{(Y-i)}]_{\bar{u}=j} \right) / 2$, which are assembled by electric field gradients caused by unit multipoles. At last, $U_{\nabla E}^{(\Theta)}(\mathbf{r}_i - \mathbf{r}_j)$ is a 5×5 matrix, of which elements are given by

$$[U_{\nabla E}^{(\Theta)}]_{i,j} = \begin{cases} \left([\nabla E^{(\Theta_{u=j})}]_i + [\nabla E^{(\Theta_{u=j})}]_{\bar{i}} + [\nabla E^{(\Theta_{u=\bar{j}})}]_i + [\nabla E^{(\Theta_{u=\bar{j}})}]_{\bar{i}} \right) / 2 & (j = 1, 2, 3) \\ \left([\nabla E^{(\Theta_{u=j})}]_i + [\nabla E^{(\Theta_{u=j})}]_{\bar{i}} - [\nabla E^{(\Theta_{33})}]_i - [\nabla E^{(\Theta_{33})}]_{\bar{i}} \right) / 2 & (j = 4, 5) \end{cases}. \quad (\text{S20})$$

The fields generated by multipole momenta $\mathbf{M}(0)$ together with the EM field of the incident wave can induce the electric dipole $\boldsymbol{\mu}^{(E1)}$, magnetic dipole $\mathbf{m}^{(M1)}$ and electric quadrupole $\hat{\Theta}^{(E2)}$ inside the molecule. They can be expressed as

$$M_{mol1} = P_{mol} [I - XP_{inc}]^{-1} F_{inc}, \quad (\text{S21})$$

where

$$\mathbf{P}_{mol} = \begin{pmatrix} \mathbf{P}_0 & \mathbf{0} & \cdots & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{0} \end{pmatrix}. \quad (\text{S22})$$

Besides, $\boldsymbol{\mu}^{(E1)}$, $\mathbf{m}^{(M1)}$ and $\hat{\Theta}^{(E2)}$ can generate multipoles M_{mol2} inside the NPs, of which expressions are given by

$$M_{mol2} = P_{inc} [I - XP_{inc}]^{-1} X \cdot M_{mol1}. \quad (\text{S23})$$

The total multipole of the molecule-NP system is the sum of M_{mol1} , M_{mol2} and $M(0)$:

$$M = M_{mol1} + M_{mol2} + M(0). \quad (\text{S24})$$

After calculating the multipole momenta of the system, the absorption rate and CD can be readily gotten according to equations (S9-S11).

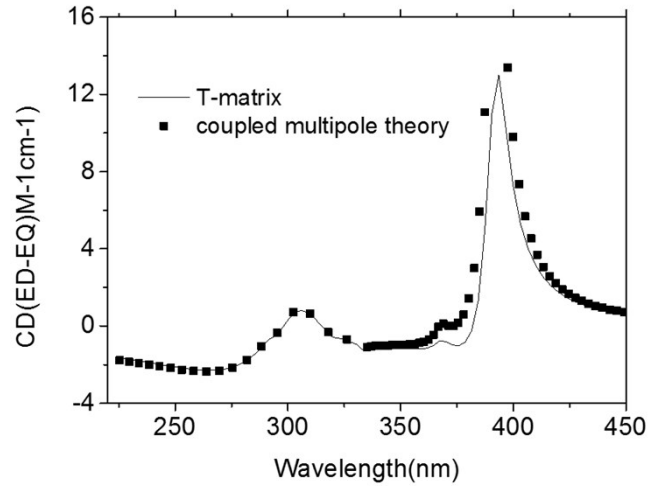


Fig. S2. CD(ED-EQ) for the system discussed in Fig.1. (a) calculated using the T-matrix method (black line) and the coupled multipole theory (black square).

To verify the correctness of the T-matrix formula developed in the main text, the CD(ED-EQ) for the system discussed in Fig. 1a is recalculated using the coupled multipole method discussed in this part. The calculated results are marked by black squares, as shown in Fig. S2. By comparing with the result from the T-matrix method (black line), one can find that there is perfect agreement

between these two methods, except the result by the coupled multipole method is a little larger at 375 nm. Since only the electric dipole and the electric quadrupole of the NP are considered in the coupled multipole method, the discrepancy should be caused by the higher order multipoles of the NP.

S2. Enhanced chiroptical effect by a cylindrical nanoparticle.

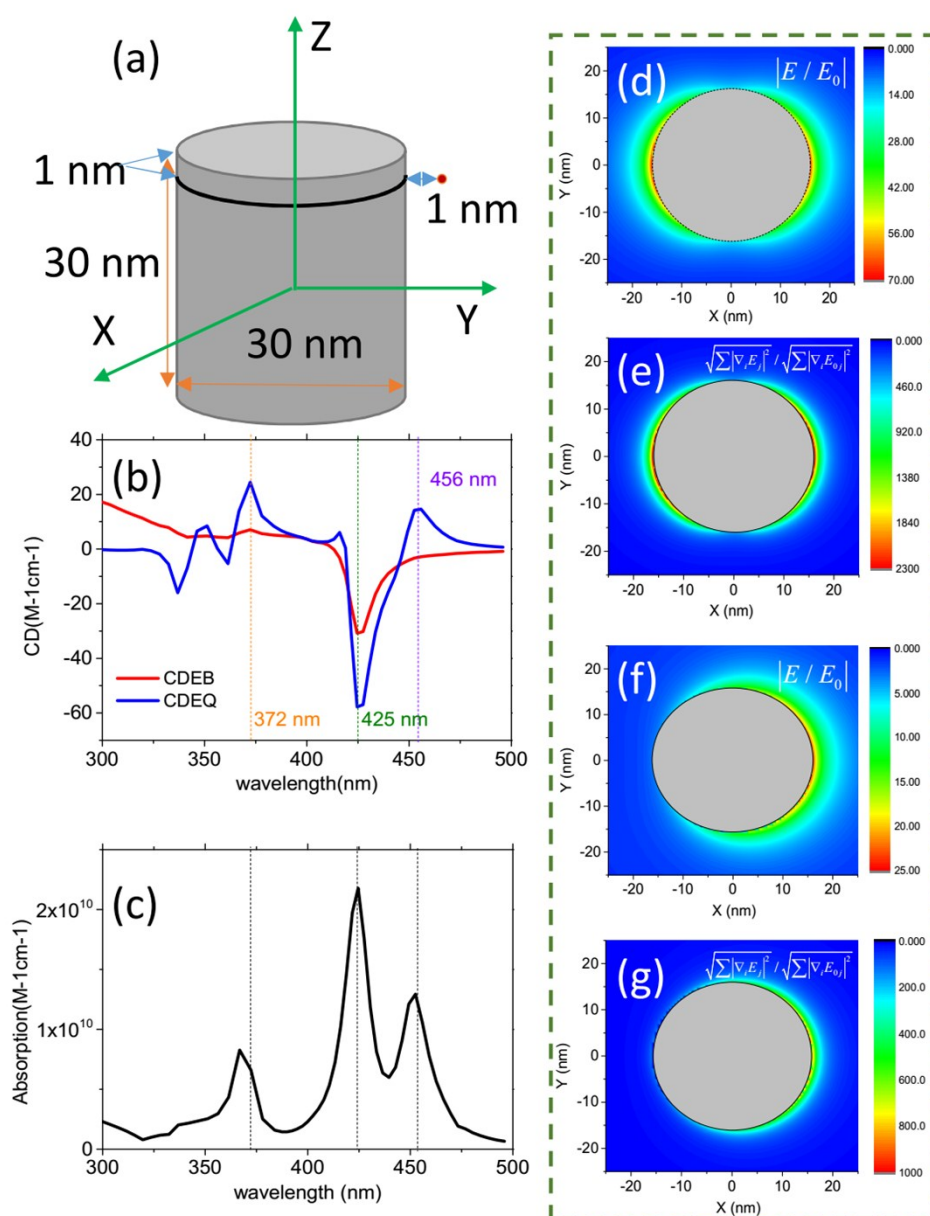


Fig. S3. CD enhanced by a cylindrical particle. (A) Geometry and coordinate of a hybrid system consisting of a chiral molecule and a cylindrical silver nanoparticle. The diameter and the length of the silver cylinder are both 30 nm. The center of the cylinder is put at the origin. The chiral molecule is located at $(x,y,z)=(0, 16, 14)$ nm with multipole momenta set to be equal with those in Fig.1. The whole system is embedded in water with refraction index $n=1.33$. (B) Calculated CD signals for the molecule-NP complex. (C) The orientation averaged absorption cross section of the silver cylinder. (D and F) and (E and G) correspond to the electric fields and the electric field gradients evaluated at the plane $z=14$ nm for the cylinder at the wavelength of 425 nm. The silver cylinder is excited by X-polarized plane wave propagating in Z direction and Z-polarized plane wave propagating in Y direction for Fig. S3 d and e and Fig. S3 f g, respectively.

In this part, we discuss the CD of a chiral molecule put in the vicinity of a silver cylindrical nanoparticle. The parameters of the molecule nanoparticle system are given in Fig. S3 a. For such a system, the electric field and the electric field gradient are significantly enhanced near the bottom and the top of the cylinder, due to the existing of sharp edges. The blue line and red line in Fig. S3 b correspond to the CD(ED-EQ) and CD(ED-MD), respectively. By comparison, we found that CD(ED-EQ) is much larger than CD(ED-MD) in many spectral regions, e.g. the value of CD(ED-EQ) is 2 times larger than that of CD(ED-MD) at 425 nm. Fig. S3 c shows the absorption cross section of the nano cylinder. By comparing, we found that the peak values of CD(ED-EQ) appear not only at the spectral regions where absorption cross sections are large (e.g. at 372 nm, 425 nm, 456 nm), but also in some regions with the small absorption (e.g. at 336 nm). This phenomenon is caused by the excitation of the optical dark modes of the cylinder by the E2 radiation of the molecule. For bright mode regions (eg. 372 nm and 425 nm), the large CD(ED-EQ)

signal are caused by dramatically enhanced electric fields gradient. Fig. S3 d and f show the electric field distributions of the cylinder (evaluated at the plane $z=14$ nm) excited by the plane wave at 425 nm, which are calculated using the method of BEM^{S5,S6}. The directions and polarizations of the incident plane wave are given by the caption. As can be seen, the electric field is strongly enhanced around the circumference of the cylinder. The electric field enhancement factor can be as large as 70 and 25 when the electric field of the plane wave is perpendicular and parallel with the z axis of the coordinate, respectively. The large electric fields near the cylinder are evanescent and rapidly decay away from the surface, which causes large electric field gradient. The corresponding electric fields gradients for the cases discussed in Fig.S3 d and f are plotted in Fig.S3 e and Fig.S3 g. From Fig.S3 e, it can be found that the electric field gradient enhancement factor can be as large as 10^3 , which is even greater than the ‘hotspot’ cases discussed in the main text.

S3. Derivation of equation 14

In this part, we provide the details of the derivation of equation (14) in the main text. Following ref. S7, the incident wave can be expanded as a series of vector spherical function (VSF) in a coordinate system of which origin is set to the position of the molecule:

$$\mathbf{E}_{inc}^{(0)}(\mathbf{r}) = \sum_{\nu, \nu'=0}^{\infty} \begin{bmatrix} a_{\nu}^{\sigma\pm} & b_{\nu}^{\sigma\pm} \end{bmatrix} R_{\nu\nu'}(-\gamma, -\beta, -\alpha) \begin{bmatrix} \mathbf{M}_{\nu'}^{(1)}(\mathbf{r} - \mathbf{r}_d) \\ \mathbf{N}_{\nu'}^{(1)}(\mathbf{r} - \mathbf{r}_d) \end{bmatrix}, \quad (\text{S25})$$

where $a_{\nu}^{\sigma\pm}$ and $b_{\nu}^{\sigma\pm}$ are the expansion coefficients of the left- ($\sigma +$) or right ($\sigma -$) handed circular polarized wave propagating along the z direction. $R_{\nu\nu'}$ is the 2×2 rotation block matrix which has been given in ref. S8. Absorption of waves in any direction can be calculated by changing the Euler angles α , β and γ . $\nu = (m, n)$ is introduced here for simplification of the

notation, and the convention $\nu = 1, 2, \dots$, when $n = 1, 2, \dots$, and $m = -n, \dots, n$ are used.

Likewise, the EM field $(\mathbf{E}_s^{(0)}, \mathbf{B}_s^{(0)})$ scattered by NPs in the absence of the molecule, are related

to the incident wave by the T-Matrix, and they can be expressed as

$$\mathbf{E}_s^{(0)}(\alpha, \beta, \gamma) = \sum_{\nu, \nu', \nu''=1}^{\infty} \begin{bmatrix} a_{\nu}^{\sigma\pm} & b_{\nu}^{\sigma\pm} \end{bmatrix} R_{\nu\nu''}(-\gamma, -\beta, -\alpha) \sum_{\xi=1}^{NP} \begin{bmatrix} T_{\nu'\nu''}^{11}(\xi) & T_{\nu'\nu''}^{12}(\xi) \\ T_{\nu'\nu''}^{21}(\xi) & T_{\nu'\nu''}^{22}(\xi) \end{bmatrix}^T \begin{bmatrix} \mathbf{M}_{\nu'}^{(3)}(\mathbf{r} - \mathbf{r}_{\xi}) \\ \mathbf{N}_{\nu'}^{(3)}(\mathbf{r} - \mathbf{r}_{\xi}) \end{bmatrix}, \quad (\text{S26})$$

$$\mathbf{B}_s^{(0)}(\alpha, \beta, \gamma) = [-i\sqrt{\varepsilon\varepsilon_0\mu\mu_0}] \sum_{\nu, \nu', \nu''=1}^{\infty} \begin{bmatrix} a_{\nu}^{\sigma\pm} & b_{\nu}^{\sigma\pm} \end{bmatrix} R_{\nu\nu''}(-\gamma, -\beta, -\alpha) \sum_{\xi=1}^{NP} \begin{bmatrix} T_{\nu'\nu''}^{11}(\xi) & T_{\nu'\nu''}^{12}(\xi) \\ T_{\nu'\nu''}^{21}(\xi) & T_{\nu'\nu''}^{22}(\xi) \end{bmatrix}^T \begin{bmatrix} \mathbf{N}_{\nu'}^{(3)}(\mathbf{r} - \mathbf{r}_{\xi}) \\ \mathbf{M}_{\nu'}^{(3)}(\mathbf{r} - \mathbf{r}_{\xi}) \end{bmatrix}, \quad (\text{S27})$$

where \mathcal{E} (μ) and ε_0 (μ_0) are the relative and absolute permittivity (permeability) in the space

and vacuum, respectively. \mathbf{r}_{ξ} is the position vector of the ξ th NP, as shown in Fig.S1, $T_{\nu\nu'}^{ij}(\xi)$

are elements of the coupled T-matrix ($T(\xi)$) of the ξ th NP, and they are related to the general

single particle T-Matrix \mathbf{T}_p by the following equation:

$$T(\xi) = \left(\sum_{p=1}^{NP} \mathbf{A}^{\xi p} \mathbf{T}_p S_{p0}^{rt} \right) \quad (\text{S28})$$

with $\mathbf{A} = A^{-1}$, and the block-matrix components of A are written as $A^{\xi\xi} = I$ ($\xi = 1, 2, \dots, NP$)

and $A^{\xi p} = -\mathbf{T}_{\xi} S_{\xi p}^{rt}$ ($\xi \neq p$). Here $S_{\xi p}^{rt}$ and S_{p0}^{rt} are transformation matrices for the coordinate

systems defined in Appendix B of ref. S7, and I is the identity matrix.

Inserting equations (S25-S27) into equation (3), the absorption rate of the molecule can be

written as:

$$\mathcal{Q}_{mol} = \frac{1}{V} \sum_{\substack{\nu_1, \nu_1'', \nu_1', \\ \nu_2, \nu_2'', \nu_2'=0}}^{\infty} \begin{bmatrix} a_{\nu_1}^{\sigma\pm} & b_{\nu_1}^{\sigma\pm} \end{bmatrix} R_{\nu_1\nu_1''}(-\gamma, -\beta, -\alpha) U_{\nu_1''\nu_1'} [U_{\nu_2'\nu_2''}]^{\dagger} [R_{\nu_2\nu_2''}(-\gamma, -\beta, -\alpha)]^{\dagger} \begin{bmatrix} a_{\nu_2}^{\sigma\pm} \\ b_{\nu_2}^{\sigma\pm} \end{bmatrix}^* \quad (\text{S29})$$

with

$$U_{\nu''\nu'} = U_{\nu''\nu'}(\boldsymbol{\mu}_{21}) + U_{\nu''\nu'}(\mathbf{m}_{21}) + U_{\nu''\nu'}(\boldsymbol{\Theta}_{21}), \quad (\text{S30})$$

$$U_{v''v'}(\boldsymbol{\mu}_{21}) = \sum_{\xi=1}^{NP} \left\{ \begin{bmatrix} T_{v''v''}^{11}(\xi) & T_{v''v''}^{12}(\xi) \\ T_{v''v''}^{21}(\xi) & T_{v''v''}^{22}(\xi) \end{bmatrix}^T \begin{bmatrix} \mathbf{M}_{v'}^{(3)}(\mathbf{r}_d - \mathbf{r}_\xi) \cdot \boldsymbol{\mu}_{21} \\ \mathbf{N}_{v'}^{(3)}(\mathbf{r}_d - \mathbf{r}_\xi) \cdot \boldsymbol{\mu}_{21} \end{bmatrix} \right. \\ \left. + \begin{bmatrix} \delta_{v''v''} & 0 \\ 0 & \delta_{v''v''} \end{bmatrix} \begin{bmatrix} \mathbf{M}_{v'}^{(1)}(\mathbf{r}_d) \cdot \boldsymbol{\mu}_{21} \\ \mathbf{N}_{v'}^{(1)}(\mathbf{r}_d) \cdot \boldsymbol{\mu}_{21} \end{bmatrix} \right\}, \quad (\text{S31})$$

$$U_{v''v'}(\mathbf{m}_{21}) = -i\sqrt{\varepsilon\varepsilon_0\mu\mu_0} \left\{ \sum_{\xi=1}^{NP} \begin{bmatrix} T_{v''v''}^{11}(\xi) & T_{v''v''}^{12}(\xi) \\ T_{v''v''}^{21}(\xi) & T_{v''v''}^{22}(\xi) \end{bmatrix}^T \begin{bmatrix} \mathbf{N}_{v'}^{(3)}(\mathbf{r}_d - \mathbf{r}_\xi) \cdot \mathbf{m}_{21} \\ \mathbf{M}_{v'}^{(3)}(\mathbf{r}_d - \mathbf{r}_\xi) \cdot \mathbf{m}_{21} \end{bmatrix} \right. \\ \left. + \begin{bmatrix} \delta_{v''v''} & 0 \\ 0 & \delta_{v''v''} \end{bmatrix} \begin{bmatrix} \mathbf{N}_{v'}^{(1)}(\mathbf{r}_d) \cdot \mathbf{m}_{21} \\ \mathbf{M}_{v'}^{(1)}(\mathbf{r}_d) \cdot \mathbf{m}_{21} \end{bmatrix} \right\}, \quad (\text{S32})$$

$$U_{v''v'}(\Theta_{21}) = \frac{1}{3} \sum_{\alpha\beta} \sum_{\xi=1}^{NP} \left\{ \begin{bmatrix} T_{v''v''}^{11}(\xi) & T_{v''v''}^{12}(\xi) \\ T_{v''v''}^{21}(\xi) & T_{v''v''}^{22}(\xi) \end{bmatrix}^T \begin{bmatrix} [\nabla \mathbf{M}_{v'}^{(3)}(\mathbf{r}_d - \mathbf{r}_\xi)]_{\alpha\beta} \Theta_{21,\alpha\beta} \\ [\nabla \mathbf{N}_{v'}^{(3)}(\mathbf{r}_d - \mathbf{r}_\xi)]_{\alpha\beta} \Theta_{21,\alpha\beta} \end{bmatrix} \right. \\ \left. + \begin{bmatrix} \delta_{v''v''} & 0 \\ 0 & \delta_{v''v''} \end{bmatrix} \begin{bmatrix} [\nabla \mathbf{M}_{v'}^{(1)}(\mathbf{r}_d - \mathbf{r}_\xi)]_{\alpha\beta} \Theta_{21,\alpha\beta} \\ [\nabla \mathbf{N}_{v'}^{(1)}(\mathbf{r}_d - \mathbf{r}_\xi)]_{\alpha\beta} \Theta_{21,\alpha\beta} \end{bmatrix} \right\}. \quad (\text{S33})$$

Here $V = \omega_0 \gamma_{21} / \left(2 \left| \mathbf{h}(\omega - \omega_0) + i\gamma_{21} - G \right|^2 \right)$. Using the T-matrix formula, the fields $\mathbf{E}_{tot}^{(X)}$ (X stands for 0, E1, M1, and E2) can be expressed as:

$$\mathbf{E}_{tot}^{(X)}(\mathbf{r} - \mathbf{r}_\xi) = \mathbf{E}_{s-\xi}^{(X)}(\mathbf{r} - \mathbf{r}_\xi) + \mathbf{E}_{inf}^{(X)}(\mathbf{r} - \mathbf{r}_\xi) \\ = \sum_{v=1}^{\infty} \left\{ \left[c_v^{(X)}(\xi) \mathbf{M}_v^{(3)}(\mathbf{r} - \mathbf{r}_\xi) + d_v^{(X)}(\xi) \mathbf{N}_v^{(3)}(\mathbf{r} - \mathbf{r}_\xi) \right] \right. \\ \left. + \left[a_v^{(X)}(\xi) \mathbf{M}_v^{(1)}(\mathbf{r} - \mathbf{r}_\xi) + b_v^{(X)}(\xi) \mathbf{M}_v^{(1)}(\mathbf{r} - \mathbf{r}_\xi) \right] \right\}, \quad (\text{S34})$$

and

$$\begin{bmatrix} c_v^{(0)}(\xi) \\ d_v^{(0)}(\xi) \end{bmatrix} = \sum_{v', v_0=0}^{\infty} \begin{bmatrix} T_{vv'}^{11}(\xi) & T_{vv'}^{12}(\xi) \\ T_{vv'}^{21}(\xi) & T_{vv'}^{22}(\xi) \end{bmatrix} \left[R_{v_0v'}(-\gamma, -\beta, -\alpha) \right]^T \begin{bmatrix} a_{v_0} \\ b_{v_0} \end{bmatrix}, \quad (\text{S35})$$

$$\begin{bmatrix} c_v^{(E1)}(\xi) \\ d_v^{(E1)}(\xi) \end{bmatrix} = \sum_{a=x,y,z} \mu_a^{(E1)} \begin{bmatrix} \mathfrak{C}_v^{(E1-a)}(\xi) \\ \mathfrak{d}_v^{(E1-a)}(\xi) \end{bmatrix}, \quad (\text{S36})$$

$$\begin{bmatrix} c_v^{(M1)}(\xi) \\ d_v^{(M1)}(\xi) \end{bmatrix} = \sum_{a=x,y,z} m_a^{(M1)} \begin{bmatrix} \mathfrak{C}_v^{(M1-a)}(\xi) \\ \mathfrak{d}_v^{(M1-a)}(\xi) \end{bmatrix}, \quad (\text{S37})$$

$$\begin{bmatrix} c_v^{(E2)}(\xi) \\ d_v^{(E2)}(\xi) \end{bmatrix} = \sum_{a,b=x,y,z} \Theta_{ab}^{(E2)} \begin{bmatrix} \mathfrak{C}_v^{(E2-ab)}(\xi) \\ \mathfrak{d}_v^{(E2-ab)}(\xi) \end{bmatrix}. \quad (\text{S38})$$

Here $(\mathfrak{C}_v^{(E1-a)}(\xi), \mathfrak{d}_v^{(E1-a)}(\xi))$, $(\mathfrak{C}_v^{(M1-a)}(\xi), \mathfrak{d}_v^{(M1-a)}(\xi))$ and $(\mathfrak{C}_v^{(E2-ab)}(\xi), \mathfrak{d}_v^{(E2-ab)}(\xi))$

denote the expansion coefficients of the scattered wave $\mathbf{E}_{s-\xi}^{(X)}(\mathbf{r} - \mathbf{r}_\xi)$ from the ξ^{th} NP being

incident by an *unit* electric dipole along the direction of \mathbf{e}_a , an *unit* magnetic dipole, and an *unit* electric quadrupole with a component of ab . $(\mathcal{C}_v^{(E1-a)}(\xi), \mathcal{d}_v^{(E1-a)}(\xi))$ can be evaluated using the methods described in the supplement information of ref. S9, while $(\mathcal{C}_v^{(M1-a)}(\xi), \mathcal{d}_v^{(M1-a)}(\xi))$ and $(\mathcal{C}_v^{(E2-ab)}(\xi), \mathcal{d}_v^{(E2-ab)}(\xi))$ can be derived in a similar way but the multipole field expansion of the magnetic dipole and the electric quadrupole described in ref. S10 should be used. Using equation (S30) and equation (S31) to substitute $\mu_a^{(E1)}$, $m_a^{(M1)}$ and $\Theta_{ab}^{(E2)}$, equations. (S36-38) can be expressed as:

$$\begin{bmatrix} \mathcal{C}_v^{(E1)}(\xi) \\ \mathcal{d}_v^{(E1)}(\xi) \end{bmatrix} = \left[\sum_{v_0, v_1, v''=0}^{\infty} \frac{-1}{\mathfrak{I}} [a_{v_0}^{\sigma\pm} \quad b_{v_0}^{\sigma\pm}] R_{v_0 v_1}(-\gamma, -\beta, -\alpha) U_{v_1 v''} \right] \begin{bmatrix} \mathcal{C}_v^{(E1)}(\xi) \\ \mathcal{d}_v^{(E1)}(\xi) \end{bmatrix}, \quad (\text{S39})$$

$$\begin{bmatrix} \mathcal{C}_v^{(M1/E2)}(\xi) \\ \mathcal{d}_v^{(M1/E2)}(\xi) \end{bmatrix} = \left[\sum_{v_0, v_1, v''=0}^{\infty} \frac{-1}{\mathfrak{I}} [a_{v_0}^{\sigma\pm} \quad b_{v_0}^{\sigma\pm}] R_{v_0 v_1}(-\gamma, -\beta, -\alpha) U_{v_1 v''}(\boldsymbol{\mu}_{21}) \right] \begin{bmatrix} \mathcal{C}_v^{(M1/E2)}(\xi) \\ \mathcal{d}_v^{(M1/E2)}(\xi) \end{bmatrix}, \quad (\text{S40})$$

where $\mathfrak{I} = \mathbf{h}(\omega - \omega_0) + i\gamma_{21} - G$, $(\mathcal{C}_v^{(E1)}(\xi), \mathcal{d}_v^{(E1)}(\xi))$, $(\mathcal{C}_v^{(M1)}(\xi), \mathcal{d}_v^{(M1)}(\xi))$ and $(\mathcal{C}_v^{(M2)}(\xi), \mathcal{d}_v^{(E2)}(\xi))$ are given by

$$\begin{bmatrix} \mathcal{C}_v^{(E1)}(\xi) \\ \mathcal{d}_v^{(E1)}(\xi) \end{bmatrix} = \sum_{a=x,y,z} \mu_{21a} \begin{bmatrix} \mathcal{C}_v^{(E1-a)}(\xi) \\ \mathcal{d}_v^{(E1-a)}(\xi) \end{bmatrix}, \quad (\text{S41})$$

$$\begin{bmatrix} \mathcal{C}_v^{(M1)}(\xi) \\ \mathcal{d}_v^{(M1)}(\xi) \end{bmatrix} = \sum_{a=x,y,z} m_{21a} \begin{bmatrix} \mathcal{C}_v^{(M1-a)}(\xi) \\ \mathcal{d}_v^{(M1-a)}(\xi) \end{bmatrix}, \quad (\text{S42})$$

$$\begin{bmatrix} \mathcal{C}_v^{(E2)}(\xi) \\ \mathcal{d}_v^{(E2)}(\xi) \end{bmatrix} = \sum_{\substack{a=x,y,z \\ b=x,y,z}} \Theta_{21ab} \begin{bmatrix} \mathcal{C}_v^{(E2-ab)}(\xi) \\ \mathcal{d}_v^{(E2-ab)}(\xi) \end{bmatrix}. \quad (\text{S43})$$

Using the expressions of $\mathcal{C}_v^{(X)}(\xi)$ and $\mathcal{d}_v^{(X)}(\xi)$, the expansion coefficients $a_v^{(X)}(\xi)$ and $b_v^{(X)}(\xi)$ for the incident wave on the ξ^{th} NP can be calculated. They are expressed as

$$\begin{bmatrix} a_v^{(X)}(\xi) \\ b_v^{(X)}(\xi) \end{bmatrix} = \sum_{v, v'=1}^{\infty} \begin{bmatrix} [\mathbf{T}_\xi^{-1}]_{vv'}^{11} & [\mathbf{T}_\xi^{-1}]_{vv'}^{12} \\ [\mathbf{T}_\xi^{-1}]_{vv'}^{21} & [\mathbf{T}_\xi^{-1}]_{vv'}^{22} \end{bmatrix} \begin{bmatrix} \mathcal{C}_{v'}^{(X)}(\xi) \\ \mathcal{d}_{v'}^{(X)}(\xi) \end{bmatrix}. \quad (\text{S44})$$

The absorption rate of the molecule can be calculated using equation (11) in the main text.

Specifically, $Q_{NP}^{(X1-X2)}$ is firstly written into the form as:

$$Q_{NP}^{(X1-X2)} = \frac{1}{(1 + \delta(X1, X2))} \sum_{\xi=1}^N [Q_{\text{ext}}^{(X1-X2)} + Q_{\text{sca}}^{(X1-X2)} + Q_{\text{ext}}^{(X2-X1)} + Q_{\text{sca}}^{(X2-X1)}] \quad (\text{S45})$$

with

$$Q_{\text{ext}}^{(X1, X2)}(\xi) = -\frac{1}{2} \text{Re} \oint_{\Omega_\xi} \mathbf{n} \cdot \mathbf{E}_{s-\xi}^{(X1)}(\mathbf{r} - \mathbf{r}_\xi) \times \mathbf{H}_{\text{inf}-\xi}^{(X2)*}(\mathbf{r} - \mathbf{r}_\xi) + \mathbf{n} \cdot \mathbf{E}_{\text{inf}-\xi}^{(X2)}(\mathbf{r} - \mathbf{r}_\xi) \times \mathbf{H}_{s-\xi}^{(X1)*}(\mathbf{r} - \mathbf{r}_\xi) d\Omega, \quad (\text{S46})$$

$$Q_{\text{sca}}^{(X1, X2)}(\xi) = -\frac{1}{2} \text{Re} \oint_{\Omega_\xi} \mathbf{n} \cdot \mathbf{E}_{s-\xi}^{(X1)}(\mathbf{r} - \mathbf{r}_\xi) \times \mathbf{H}_{s-\xi}^{(X2)*}(\mathbf{r} - \mathbf{r}_\xi) d\Omega. \quad (\text{S47})$$

Using the orthogonality relation of the vector sphere functions, equations (S46) and (S47) can be

written as:

$$Q_{\text{ext}}^{(X1, X2)}(\xi) = -\frac{\pi}{2k^2} \sqrt{\frac{\varepsilon\varepsilon_0}{\mu\mu_0}} \sum_{v=1}^{\infty} \text{Re} \left\{ \begin{bmatrix} c_v^{(X1)}(\xi) & d_v^{(X1)}(\xi) \end{bmatrix} \begin{bmatrix} a_v^{(X2)*}(\xi) \\ b_v^{(X2)*}(\xi) \end{bmatrix} \right\}, \quad (\text{S48})$$

$$Q_{\text{sca}}^{(X1, X2)}(\xi) = -\frac{\pi}{2k^2} \sqrt{\frac{\varepsilon\varepsilon_0}{\mu\mu_0}} \sum_{v=1}^{\infty} \text{Re} \left\{ \begin{bmatrix} c_v^{(X1)}(\xi) & d_v^{(X1)}(\xi) \end{bmatrix} \begin{bmatrix} c_v^{(X2)*}(\xi) \\ c_v^{(X2)*}(\xi) \end{bmatrix} \right\} = Q_{\text{sca}}^{(X2, X1)}(\xi). \quad (\text{S49})$$

Inserting equations (S35), (S39) and (S40) into these equations, we have

$$\begin{aligned} Q_{\text{type}}^{(X1, X2)}(\xi) &= -\frac{\pi}{2k^2} \sqrt{\frac{\varepsilon\varepsilon_0}{\mu\mu_0}} \text{Re} \left\{ \begin{bmatrix} z_{1v}^{(X1)}(\xi) & z_{2v}^{(X1)}(\xi) \end{bmatrix} \begin{bmatrix} s_{1v}^{(X2)*}(\xi) \\ s_{2v}^{(X2)*}(\xi) \end{bmatrix} \right\} \\ &= -\frac{\pi}{2k^2} \sqrt{\frac{\varepsilon\varepsilon_0}{\mu\mu_0}} \sum_{\substack{v_0, v'_0 \\ v_{l_0}, v'_{l_0}}} \text{Re} \left\{ \begin{bmatrix} a_{v_0}^{\sigma\pm} & b_{v_0}^{\sigma\pm} \end{bmatrix} [R_{v_0 v'_0}(-\gamma, -\beta, -\alpha)] \right. \\ &\quad \left. \begin{bmatrix} L_{z \ v'_v}^{(X1)} [L_{s \ v_{l_0} v'}^{(X2)}]^\dagger [R_{v_{l_0} v'_{l_0}}(-\gamma, -\beta, -\alpha)]^\dagger \begin{bmatrix} a_{v_{l_0}}^{\sigma\pm} \\ b_{v_{l_0}}^{\sigma\pm} \end{bmatrix} \right]^* \right\} \end{aligned} \quad (\text{S50})$$

with

$$L_{y \ v'_v}^{(x)} = \sum_{u=1}^{\infty} X_{v'u}(x) P_{uv}(y), \quad (\text{S51})$$

$$X_{v'u}(x) = \begin{cases} \begin{bmatrix} T_{uv'}^{11}(\xi) & T_{uv'}^{12}(\xi) \\ T_{uv'}^{21}(\xi) & T_{uv'}^{22}(\xi) \end{bmatrix}^T & x \rightarrow 0 \\ \frac{-1}{\mathfrak{I}} [U_{v',v''}][c_u^{(E1)}(\xi) \quad d_u^{(E1)}(\xi)] & x \rightarrow \text{E1} \\ \frac{-1}{\mathfrak{I}} [U_{v',v''}(\mu_{21})][c_u^{(x)}(\xi) \quad d_u^{(x)}(\xi)] & x \rightarrow \text{M1 or } x \rightarrow \text{E2} \end{cases}, \quad (\text{S52})$$

$$\mathbf{P}_{uv}(y) = \begin{cases} \begin{bmatrix} \delta_{uv} & 0 \\ 0 & \delta_{uv} \end{bmatrix} & (y = (y_1, y_2) \rightarrow c, d) \\ \begin{bmatrix} [\mathbf{T}_l^{-1}]_{vu}^{11} & [\mathbf{T}_l^{-1}]_{vu}^{12} \\ [\mathbf{T}_l^{-1}]_{vu}^{21} & [\mathbf{T}_l^{-1}]_{vu}^{22} \end{bmatrix}^T & (y = (y_1, y_2) \rightarrow a, b) \end{cases} \quad (\text{S53})$$

Here when 'type' is set to be 'ext' $z = (z_1, z_2) \rightarrow c, d$, and $s = (s_1, s_2) \rightarrow a, b$, while $z = (z_1, z_2) \rightarrow c, d$ and $s = (s_1, s_2) \rightarrow c, d$ for 'sca'.

According to equations (S29) and (S50), the absorption rate of the system can be written as

$$Q_{NP/mol} = \text{Re } \mathbf{a} \mathbf{R}^\dagger(\alpha, \beta, \gamma) \bar{\bar{\mathbf{M}}}_{NP/mol} \mathbf{R}(\alpha, \beta, \gamma) \mathbf{a}^\dagger, \quad (\text{S54})$$

where \mathbf{a} is arranged as $\mathbf{a}(\pm\sigma) = \{a_1^{\sigma\pm} \ b_1^{\sigma\pm} \ \dots \ a_v^{\sigma\pm} \ b_v^{\sigma\pm} \ \dots\}$, while $\bar{\bar{\mathbf{M}}}_{NP/mol}$ is a matrix determined only by the property of the illuminated system, which has the form of

$$\bar{\bar{\mathbf{M}}}_{NP/mol} = \begin{bmatrix} & & \mathbf{M} & & \\ & & [M_{NP/mol}]_{vv'}^{11} & [M_{NP/mol}]_{vv'}^{12} & \\ \mathbf{L} & & [M_{NP/mol}]_{vv'}^{21} & [M_{NP/mol}]_{vv'}^{22} & \mathbf{L} \\ & & & & \\ & & & & \mathbf{M} \end{bmatrix}. \quad (\text{S54})$$

In practice, molecule-NP nanocomposites in the liquid environment have random orientations, averaging over the solid angles of the directions of the samples need to be done. This is equivalent with averaging over the solid angles of the directions of the incident light. Thus, the absorption rate should be rewritten as

$$\langle Q_{NP/mol}(\sigma \pm) \rangle = \mathbf{a}(\sigma \pm) \langle M_{NP/mol} \rangle_\Omega \mathbf{a}^\dagger(\sigma \pm) \quad (\text{S55})$$

with

$$[\langle M_{NP/mol} \rangle_\Omega]_{mn, m_1 n_1}^{ij} = \frac{1}{2n+1} \sum_{m'=-n}^n [M_{NP/mol}]_{m' n, m' n_1}^{ij} \delta_{mm_1} \delta_{nn_1}, \quad (\text{S56})$$

where $\langle \dots \rangle_\Omega$ represents the averaging over the solid angle. By analyzing the properties of $a_v^{\sigma\pm}$ and $b_v^{\sigma\pm}$ under the space inversion, the orientation averaged CD of the system is given by

$$\begin{aligned}
\langle \text{CD}_{NP/mol} \rangle_{\Omega} &= \text{Re} \left[\mathbf{a} \langle M_{NP/mol} \rangle_{\Omega} \mathbf{a}^{\dagger} - \mathcal{D} \langle M_{NP/mol} \rangle_{\Omega} \mathcal{D}^{\dagger} \right] \\
&= 2 \sum_{\nu=1}^{\infty} \text{Re} \left\{ \left[\langle M_{NP/mol} \rangle_{\Omega} \right]_{\nu\nu}^{12} a_{\nu}^{\sigma+} b_{\nu}^{\sigma+*} + \left[\langle M_{NP/mol} \rangle_{\Omega} \right]_{\nu\nu}^{21} b_{\nu}^{\sigma+} a_{\nu}^{\sigma+*} \right\} \quad (\text{S57})
\end{aligned}$$

From the equations above, CD mainly comes from four aspects of contributions: field-field interaction CD(0-0), electric dipole-electric dipole interaction CD(ED-ED), electric dipole-magnetic dipole interaction CD(ED-MD), and electric dipole-electric quadrupole interaction CD(ED-EQ), which are determined by the products of $\boldsymbol{\mu}_{12(21)}$, $\mathbf{m}_{12(21)}$ and $\boldsymbol{Q}_{12(21)}$. When the structure formed by the nano structure is achiral, CD(0-0) is vanished. Furthermore, for the systems discussed in our article, the CD(ED-ED) does not give a contribution due to symmetry. Then, the total CD signal is the sum of CD(ED-MD) and CD(ED-EQ), which can be calculated using equation (S57) and setting $\boldsymbol{Q}_{12(21)}$ or $\mathbf{m}_{12(21)}$ to be zero, respectively.

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