

Supplementary Information for

Quantitatively analyzing the mechanism of giant circular dichroism in extrinsic plasmonic chiral nanostructures by the interplay of electric and magnetic dipoles

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The electric and magnetic dyadic Green's functions

The electric dyadic Green's tensor $\vec{G}_e(r, r_0)$ and magnetic dyadic Green's function tensor $\vec{G}_m(r, r_0)$ are the free space field susceptibility tensors propagator relating an electric dipole source p_e at position r_0 in vacuum to the electric field E and magnetic field H it generates at position r through

$$E(r) = \frac{k^2}{\epsilon_0} \vec{G}_e(r, r_0) p_e \quad (S1)$$

$$H(r) = ck^2 \vec{G}_m(r, r_0) p_e \quad (S2)$$

For the electric and magnetic fields generated by a magnetic dipole p_m ,

$$E(r) = -Z_0 k^2 \vec{G}_m(r, r_0) p_m \quad (S3)$$

$$H(r) = k^2 \vec{G}_e(r, r_0) p_m \quad (S4)$$

With

$$\vec{G}_e(r, r_0) = \frac{e^{ikr}}{r} \left[(\hat{n} \otimes \hat{n} - \vec{I}) + \frac{ikr - 1}{k^2 r^2} (3 \cdot \hat{n} \otimes \hat{n} - \vec{I}) \right] \quad (S5)$$

$$\vec{G}_m(r, r_0) = \frac{e^{ikr}}{r} \left(1 + \frac{i}{kr} \right) (\hat{n} \times \vec{I}) \quad (S6)$$

$$\vec{G}_e(r, r_0)p = \frac{1}{4\pi} \frac{e^{ikr}}{r} \left[(\hat{n} \times p) \times \hat{n} + \frac{ikr - 1}{k^2 r^2} (3 \cdot \hat{n}(\hat{n} \cdot p) - p) \right] \quad (S7)$$

$$\vec{G}_m(r, r_0)p = \frac{e^{ikr}}{r} \left(1 + \frac{i}{kr} \right) (\hat{n} \times p) \quad (S8)$$

Where $r = |r - r_0|$, $k = 2\pi/\lambda$ and $\hat{n} = \frac{r - r_0}{r}$.

The coupled dipole approximation method

Let us consider many three dimensional dipole scatters.

The local field at each dipole can be expressed as[1]

$$p_{e,j} = \varepsilon_0 \vec{\alpha}_j E_{j,total} = \varepsilon_0 \vec{\alpha}_j \left(E_{j,in} + \sum_{k=1, k \neq j}^N \left(\frac{k^2}{\varepsilon_0} \vec{G}_e(r_j, r_k) p_{e,k} - Z_0 k^2 \vec{G}_m(r, r_0) p_{m,k} \right) \right) \quad (S9)$$

$$p_{m,j} = \vec{u}_j H_{j,total} = \vec{u}_j \left(H_{j,in} + \sum_{k=1, k \neq j}^N (ck^2 \vec{G}_m(r_j, r_k) p_{e,k} + k^2 \vec{G}_e(r, r_0) p_{m,k}) \right) \quad (S10)$$

For coupled electric and magnetic dipoles, we have

$$p_e = \varepsilon_0 \vec{\alpha}_1 (E_{1,in} - Z_0 k^2 \vec{G}_m(r_e, r_m) p_m) \quad (S11)$$

$$p_m = \vec{u}_2 (H_{2,in} + ck^2 \vec{G}_m(r_m, r_e) p_e) \quad (S12)$$

We can easily get the self-consistent form of dipole moments

$$p_e = \frac{\varepsilon_0 \vec{\alpha}_1 E_{1,in} - Z_0 k^2 \varepsilon_0 \vec{\alpha}_1 \vec{G}_m(r_m, r_e) \vec{u}_2 H_{2,in}}{\vec{I} + cZ_0 k^4 \varepsilon_0 \vec{\alpha}_1 \vec{G}_m(r_m, r_e) \vec{u}_2 \vec{G}_m(r_e, r_m)} \quad (S13)$$

$$p_m = \frac{\vec{u}_2 E_{2,in} + ck^2 \varepsilon_0 \vec{u}_2 \vec{G}_m(r_e, r_m) \vec{\alpha}_1 E_{1,in}}{\vec{I} + cZ_0 k^4 \varepsilon_0 \vec{u}_2 \vec{G}_m(r_e, r_m) \vec{\alpha}_1 \vec{G}_m(r_m, r_e)} \quad (S14)$$

The extinction is

$$A = \frac{\omega}{2} \text{Im}(E^* \cdot p_e + B^* \cdot p_m) \quad (S15)$$

Radiation power of the dipoles

The radiation power expressions of the electric dipole p_e and magnetic dipole p_m are

$$Q_e = \frac{\omega^4}{12\pi\epsilon_0 c^3} |p_e|^2 \quad (S16)$$

$$Q_m = \frac{\omega^4 Z_0}{12\pi c^4} |p_m|^2 \quad (S17)$$

The polarizability of the ellipsoid dipole

For an ellipsoid the polarizability tensor is

$$\vec{\alpha}(r, \omega) = \vec{\alpha}_0(r, \omega) \left[\vec{I} - \left(\frac{2}{3} \right) i k_0^3 \vec{\alpha}_0(r, \omega) - k^2 / \vec{\alpha}_0 \right]^{-1} \quad (S18)$$

where $\vec{\alpha}_0(r, \omega)$ is the Clausius-Mossotti polarizability

$$\vec{\alpha}_0 = \begin{pmatrix} \alpha_1 & 0 & 0 \\ 0 & \alpha_2 & 0 \\ 0 & 0 & \alpha_3 \end{pmatrix} \quad (S19)$$

$$\alpha_j = 4\pi abc \frac{(\epsilon_{particle} - \epsilon_{medium})}{(3\epsilon_{particle} + 3L_j(\epsilon_{particle} - \epsilon_{medium}))} \quad (S20)$$

$$L_j = \frac{abc}{2} \int_0^\infty \frac{dq}{(x^2 + q)f(q)} \quad (S21)$$

with $j = a, b, c$, $f(q) = [(q + a^2)(q + b^2)(q + c^2)]^{1/2}$ and a, b, c are the axis of the ellipsoid[2].

$\vec{u}$ is obtained in the same way. Because nature material has bad magnetic response in optical frequency and the magnetons in our paper are yield by the plasmon resonance with circular current, we used a fake $u_{particle}$ value to yield the magnetic resonance in optical wavelength range, which is from ϵ_{Au} but with imaginary part divided by 1.5 (the magnetic mode is dark, so the spectrum profile is narrower).

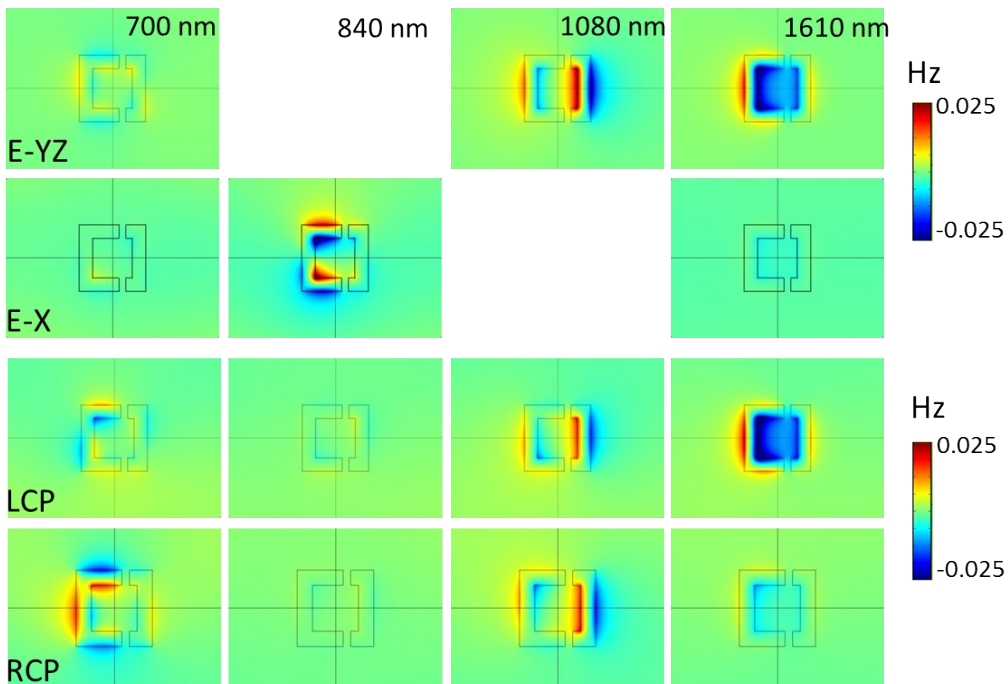


Figure S1. The magnetic field distributions in z direction for the simulations in Figure 2.

References:

1. G. W. Mulholland, C. F. Bohren and K. A. Fuller, "Light-Scattering by Agglomerates - Coupled Electric and Magnetic Dipole Method", *Langmuir* **10**, 2533-2546 (1994).
2. C. F. Bohren and D. R. Huffman, *Absorption and scattering of light by small particles*. (John Wiley & Sons, New York, 1998).