Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Planar Plasmonic Chiral Nanostructures

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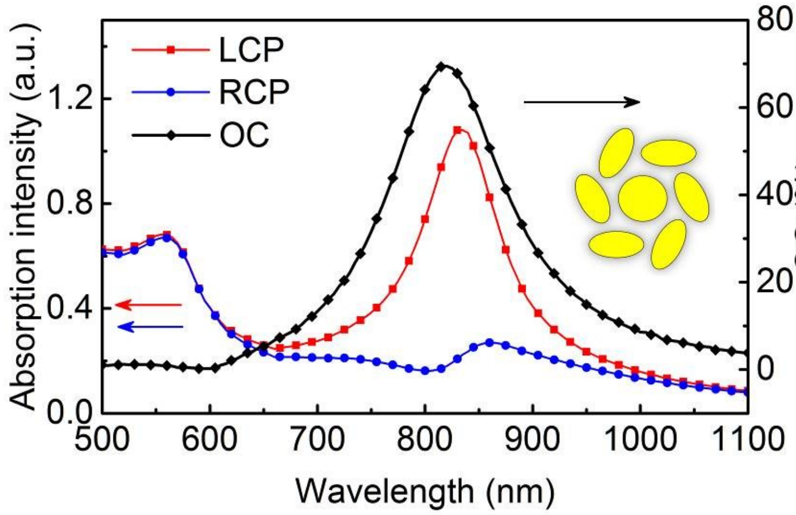


Fig. S1 The absorption cross section of chiral Fano oligomers calculated by FEM simualtion under LCP (red line) and RCP (blue line).

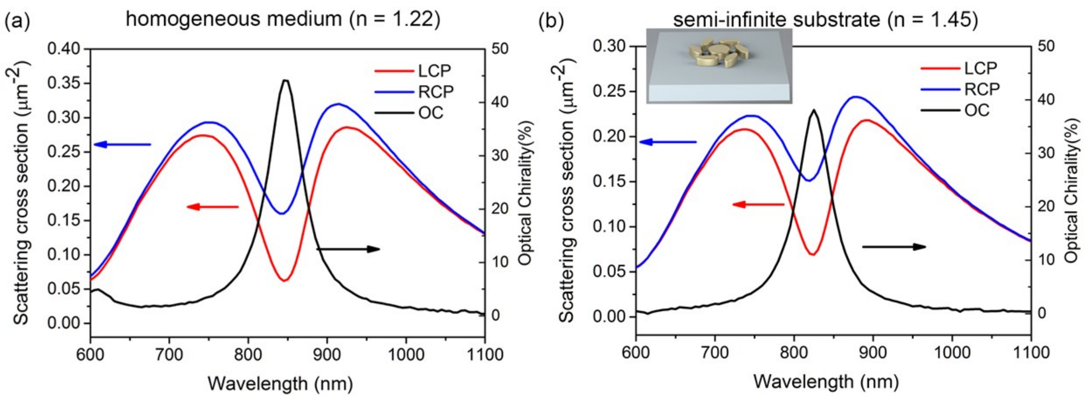


Fig. S2 The FEM simulated scattering cross section of chiral Fano oligomers (a) embedded in homogeneous medium (n = 1.22) and (b) on semi-infinite substrate (n = 1.45) under LCP (red line) and RCP (blue line).

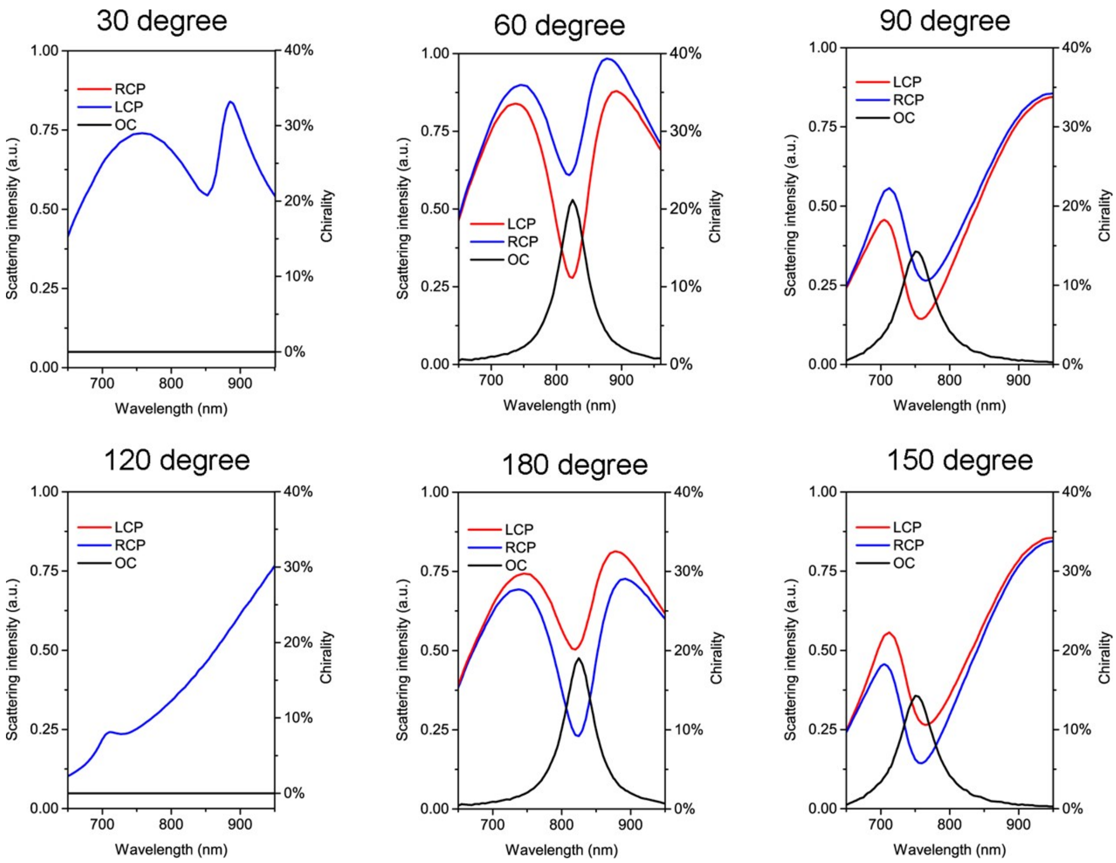
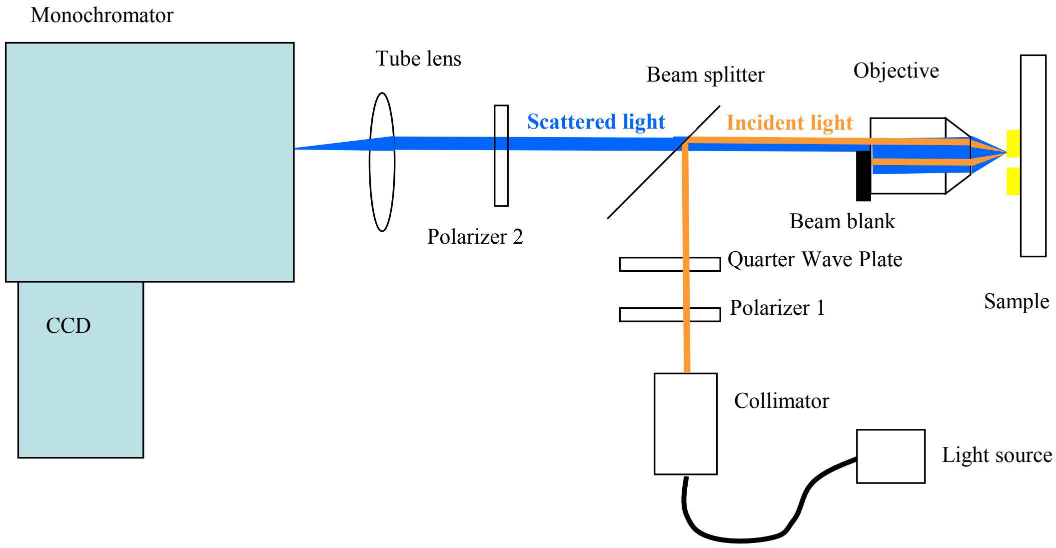


Fig. S3 FEM simulations for the scattering cross section of chiral oligomers with different surrounding ellipse rotation angle. All of the main spectral features have a good agreement with the experimental results, and the difference between the experiment and simulations possibly is induced by the sample nanofabrication..

Text S1. The near-normal incident dark-field optical microscopy

Optical scattering spectra of individual structure were measured using a near-normal incident dark-field microscope [1] (Fig. S3) with an 50* objective with a numerical aperture of 0.42 (MPlanApoNIR, 50*, Mitutoyo). The illumination source was a halogen lamp (Edmund MI-150), collimated by lens pairs and apertures, circular polarized with a linear polarizer (Polarizer 1) and a broadband quarter wave plate. Dark-field illumination is achieved by blocking the reflected incident light collected by the objective on the rear entrance pupil, allowing only scattered light to pass. The scattered light was polarized by a linear polarizer (Polarizer 2) to choose interested polarization. The modified scattered light was collected by a CCD-equipped imaging spectrometer (CCD was Princeton Instruments PIXIS 400, monochromator is Princeton Instruments Acton SP2150), and corrected for the spectral efficiency of the system using a white calibration standard (Edmund).



Schematic view of near-normal incident dark-field optical microscope.

Text S2. Calculation of the polarizabilities of a disk and an ellipse

In the coupled dipole approximation (CDA) model [2-4], the nanoparticle is considered as a dipole. In the far zone, the dipole fields are of the spherical-wave form [5]

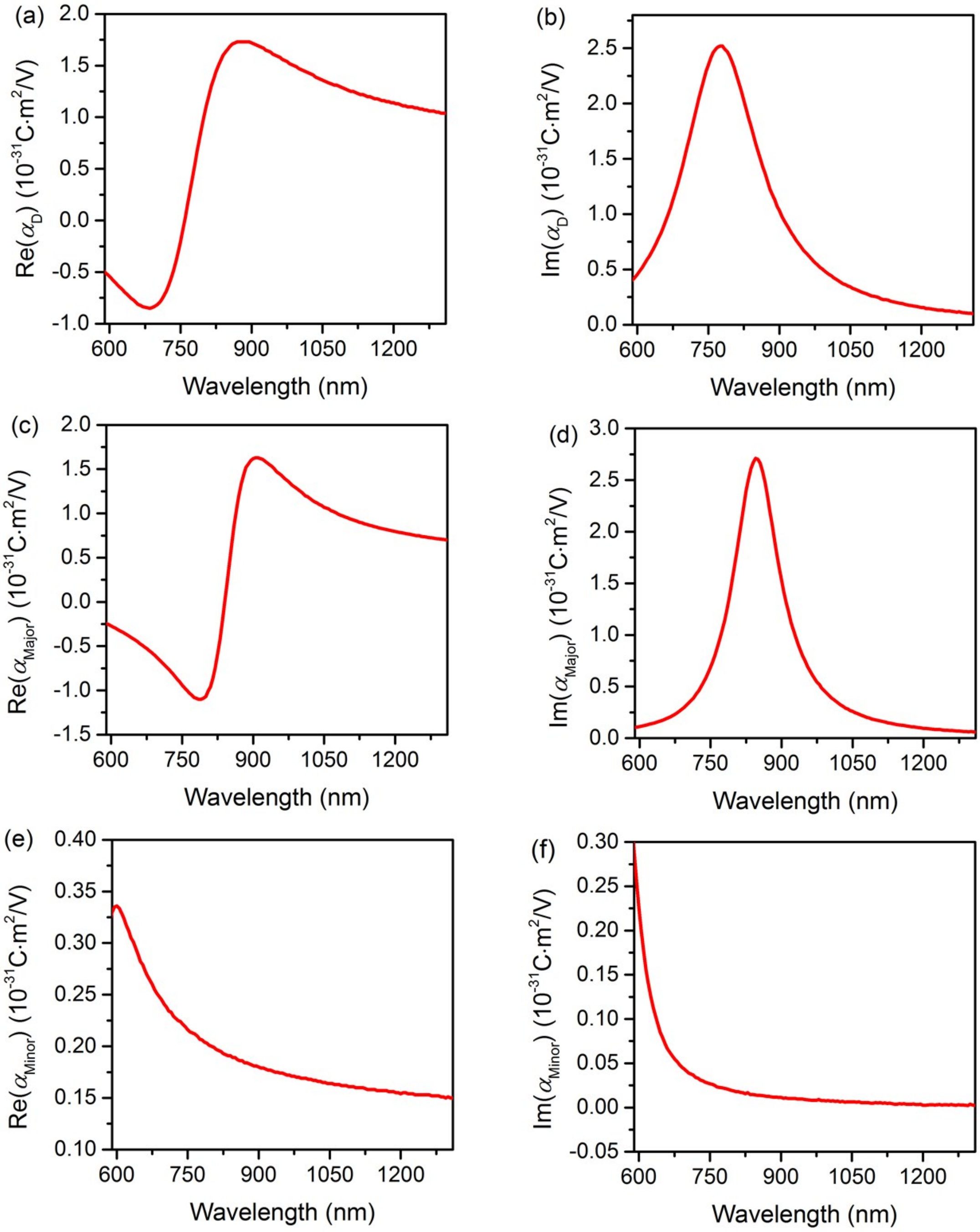
$$\boldsymbol{H}=\frac{ck^2}{4\pi}(\hat{r}\times\boldsymbol{p})\frac{e^{ikr}}{r} \tag{S1}$$

$$\boldsymbol{E}=\sqrt{\frac{\mu_0\mu_r}{\varepsilon_0\varepsilon_r}}(\boldsymbol{H}\times\hat{r}) \tag{S2}$$

where r is the distance from the dipole, $\hat{r}$ is the unit vector, $k=nk_0$ is the wave number in the surrounding (homogeneous) medium with refractive index n, $c=1\big/\sqrt{\mu_0\mu_r\varepsilon_0\varepsilon_r}$ is the light speed in medium, ε_0, μ_0 is the permittivity and permeability in vacuum, and ε_r, μ_r are the relative permittivity and permeability. For a circular disk, the forward scattering electric field $\boldsymbol{E}_F$ can be written as

$$\boldsymbol{E}_F = \frac{k^2 e^{ikr}}{4\pi\varepsilon_0\varepsilon_r r}\boldsymbol{p}_D = \frac{k^2 e^{ikr}}{4\pi\varepsilon_0\varepsilon_r r}\alpha_D \boldsymbol{E}_{ext} \tag{S3}$$

where α_D is the polarizability of the circular disk and $\boldsymbol{E}_{ext}$ is the external electric field. By using the far field calculation in FEM, we can obtain the forward scattering electric field at r = 1.0 m. Thus, we obtain the polarizability of the circular disk α_D. For the ellipse, by forcing the external electric field along major axis and minor axis directions respectively, we can get the polarizability along the major axis α_{maj} and minor axis $\alpha_{\min}$. In Fig. S4, we show the wavelength dependence of $\mathrm{Re}(\alpha_D)$, $\mathrm{Im}(\alpha_D)$, $\mathrm{Re}(\alpha_{maj})$, $\mathrm{Im}(\alpha_{maj})$, $\mathrm{Re}(\alpha_{\min})$ and $\mathrm{Im}(\alpha_{\min})$.



The FEM calculated real and imaginary part of the polarizabilities of the disk (a-b), the major axis of the ellipse (c-d) and the minor axis of the ellipse (e-f).

Text S3. Calculation of extinction, scattering and absorption cross section for coupled dipole system

S3.1 Scattering cross section

For an in-plane electric dipole $\boldsymbol{p}_j = (p_{xj}, p_{yj})$ located at the position (r_{xj}, r_{yj}), its radiation fields at the point (R,θ,φ) in the spherical coordinate are[5]

$$\boldsymbol{E}_j = \frac{k^2 e^{ikr}}{4\pi\varepsilon_0\varepsilon_r r}(\boldsymbol{e_R}\times\boldsymbol{p}_j)\times\boldsymbol{e_R} \tag{S4}$$

$$\boldsymbol{B}_j = -\frac{\boldsymbol{E}_j\times\boldsymbol{e_R}}{c} \tag{S5}$$

where $r = R-\sin\theta\cos(\varphi-\beta_j)a_j$, $\beta_j = \mathrm{atan2}(r_{yj}, r_{xj})$, $a_j = \sqrt{r_{xj}{}^2 + r_{yj}{}^2}$ and the unit vector $\boldsymbol{e_R}$ is given by $\boldsymbol{e_R} = (\sin\theta\cos\varphi, \sin\theta\sin\varphi, \cos\theta)$.

Inserting the expression of the electric dipole $\boldsymbol{p} = p_{xj}\boldsymbol{e_x} + p_{yj}\boldsymbol{e_y}$ into Eq. (S4), the electric field can be rewritten as

$$\begin{aligned}\boldsymbol{E}_j &= \frac{k^2 e^{ikr}}{4\pi\varepsilon_0\varepsilon_r R}[\boldsymbol{e_R}\times(p_{xj}\boldsymbol{e_x}+p_{yj}\boldsymbol{e_y})]\times\boldsymbol{e_R} \\ &= \frac{k^2 e^{ikR}}{4\pi\varepsilon_0\varepsilon_r R} e^{-ik\sin\theta\cos(\varphi-\beta_j)a_j} \\ &\quad \times[(p_{xj}\cos\theta\cos\varphi + p_{yj}\cos\theta\sin\varphi)\boldsymbol{e}_\theta - (p_{xj}\sin\varphi - p_{yj}\cos\varphi)\boldsymbol{e}_\varphi]\end{aligned} \tag{S6}$$

where $\boldsymbol{e_x}$ and $\boldsymbol{e}_y$ are the unit vectors along *x* and *y* directions, and the unit vectors $\boldsymbol{e}_\theta$ and $\boldsymbol{e}_\varphi$ are given by $\boldsymbol{e}_\theta = (\cos\theta\cos\varphi, \cos\theta\sin\varphi, -\sin\theta)$ and $\boldsymbol{e}_\varphi = (-\sin\varphi, \cos\varphi, 0)$, respectively.

The total electric field from the *n* electric dipoles is thus

$$\begin{aligned}\boldsymbol{E} = \sum_{j=1}^{n}\boldsymbol{E}_j = \frac{k^2 e^{ikR}}{4\pi\varepsilon_0\varepsilon_r R}[&\sum_{j=1}^{n} e^{-ik\sin\theta\cos(\varphi-\beta_j)a_j}(p_{xj}\cos\theta\cos\varphi + p_{yj}\cos\theta\sin\varphi)\cdot\boldsymbol{e}_\theta \\ &-\sum_{j=1}^{n} e^{-ik\sin\theta\cos(\varphi-\beta_j)a_j}(p_{xj}\sin\varphi - p_{yj}\cos\varphi)\cdot\boldsymbol{e}_\varphi]\end{aligned} \tag{S7}$$

The time-averaged flux of energy is given by the real part of the complex Poynting vector:

$$\overline{\boldsymbol{S}} = \frac{1}{2}\mathrm{Re}(\boldsymbol{E}^*\times\boldsymbol{H}) = -\frac{1}{2\mu_0\mu_r c}\mathrm{Re}[\boldsymbol{E}^*\times(\boldsymbol{E}\times\boldsymbol{e_R})] = \frac{1}{2\mu_0\mu_r c}|\boldsymbol{E}|^2\boldsymbol{e_R} \tag{S8}$$

So the total scattering power is

$$P = \int |\boldsymbol{S}| R^2 d\Omega$$

$$= \iint \frac{k^4 \sin\theta}{(4\pi\varepsilon_0\varepsilon_r)^2 2\mu_0\mu_r c} \times \left\{ \left| \sum_{j=1}^{n} e^{-ik\sin\theta\cos(\varphi-\beta_j)a_j} (p_{xj}\cos\theta\cos\varphi + p_{yj}\cos\theta\sin\varphi) \right|^2 + \left| \sum_{j=1}^{n} e^{-ik\sin\theta\cos(\varphi-\beta_j)a_j} (p_{xj}\sin\varphi - p_{yj}\cos\varphi) \right|^2 \right\} d\theta d\varphi \quad \text{(S9)}$$

Assuming an incident electric field $\boldsymbol{E}_0 = E_x \boldsymbol{e_x} + E_y \boldsymbol{e_y} = (E_x' + iE_x'')\boldsymbol{e_x} + (E_y' + iE_y'')\boldsymbol{e_y}$, the scattering cross section is

$$\sigma_{sca} = \frac{2P}{c\varepsilon_0\varepsilon_r} \cdot \frac{1}{(|E_x'|^2 + |E_x''|^2 + |E_y'|^2 + |E_y''|^2)} \quad \text{(S10)}$$

For circular polarizations (LCP and RCP), the scattering cross section is

$$\sigma_{sca} = \frac{P}{c\varepsilon_0\varepsilon_r} \quad \text{(S11)}$$

S3.2 Extinction cross section

When $\theta = 0$, the scattering field can be simplified as

$$\boldsymbol{E} = \frac{k^2 e^{ikR}}{4\pi\varepsilon_0\varepsilon_r R} [\sum_{j=1}^{n} (p_{xj}\cos\varphi + p_{yj}\sin\varphi) \cdot \boldsymbol{e}_\theta - \sum_{j=1}^{n} (p_{xj}\sin\varphi - p_{yj}\cos\varphi) \cdot \boldsymbol{e}_\varphi]$$
$$= \frac{k^2 e^{ikR}}{4\pi\varepsilon_0\varepsilon_r R} \left[(\sum_{j=1}^{n} p_{xj}) \cdot \boldsymbol{e}_x + (\sum_{j=1}^{n} p_{yj}) \cdot \boldsymbol{e}_y \right]$$

By using the optical theorem [6], which relates the extinction cross section to its forward scattering amplitude (in our case $\theta = 0$), we can obtain the extinction cross section as

$$\sigma_{ext} = \frac{k}{\varepsilon_0\varepsilon_r} \cdot \frac{1}{(|E_x'|^2 + |E_x''|^2 + |E_y'|^2 + |E_y''|^2)} \mathrm{Im}\left[(E_x' - iE_x'') \cdot \sum_{j=1}^{n} p_{xj} + (E_y' - iE_y'') \cdot \sum_{j=1}^{n} p_{yj} \right] \quad \text{(S12)}$$

For circular polarizations, the extinction cross section is

$$\sigma_{ext} = \frac{k}{2\varepsilon_0\varepsilon_r} \cdot \mathrm{Im}\left\{ \sum_{j=1}^{n} p_{xj} \pm i \sum_{j=1}^{n} p_{yj} \right\} \quad \text{(S13)}$$

where $(+)$ is used for LCP ($\boldsymbol{E}_0 = \boldsymbol{e_x} - i\boldsymbol{e_y}$) and $(-)$ for RCP ($\boldsymbol{E}_0 = \boldsymbol{e_x} + i\boldsymbol{e_y}$).

S3.3 Absorption cross section

The absorption cross section σ_{abs} is the difference between the extinction cross section and the scattering cross section:

$$\sigma_{abs} = \sigma_{ext} - \sigma_{sca} \tag{S14}$$

Text S4. Multipole expansion method

For a current source distribution, the total scattering power can be expanded as the sum of the multipole radiation [7, 8]. In the case of harmonic excitation $\exp(i\omega t)$, it has the form

$$\begin{aligned} I &= \frac{1}{12\pi\varepsilon_0\varepsilon_r c^3}\left[\left(\ddot{\boldsymbol{p}} - \frac{1}{c^2}\dddot{\boldsymbol{T}}\right)^2 + \frac{\left|\ddot{\boldsymbol{m}}\right|^2}{c^2}\right] + \frac{1}{160\pi\varepsilon_0\varepsilon_r c^5}\sum\left|\dddot{Q}_{\alpha\beta}\right|^2 + \frac{1}{160\pi\varepsilon_0\varepsilon_r c^7}\sum\left|\dddot{M}_{\alpha\beta}\right|^2 \\ &= \frac{\omega^4}{12\pi\varepsilon_0\varepsilon_r c^3}\left|\boldsymbol{p}\right|^2 + \frac{\omega^4}{12\pi\varepsilon_0\varepsilon_r c^5}\left|\boldsymbol{m}\right|^2 + \frac{\omega^6}{12\pi\varepsilon_0\varepsilon_r c^7}\left|\boldsymbol{T}\right|^2 - \frac{i\omega^5}{12\pi\varepsilon_0\varepsilon_r c^5}(\boldsymbol{p}^*\cdot\boldsymbol{T} - \boldsymbol{p}\cdot\boldsymbol{T}^*) \\ &+ \frac{\omega^6}{160\pi\varepsilon_0\varepsilon_r c^5}\sum\left|Q_{\alpha\beta}\right|^2 + \frac{\omega^5}{160\pi\varepsilon_0\varepsilon_r c^7}\sum\left|M_{\alpha\beta}\right|^2 \end{aligned} \tag{S15}$$

The first two terms correspond to the electric and magnetic dipole scattering. The third term corresponds to the toroidal dipole scattering and the fourth term accounts for the coupling between the electric and toroidal dipoles. The fifth and sixth terms come from electric and magnetic quadrupoles. The multipole moments in the above equation are defined as:

electric dipole moment: $\boldsymbol{p} = \frac{1}{i\omega}\int \boldsymbol{j}d^3r$

magnetic dipole moment: $\boldsymbol{m} = \frac{1}{2}\int(\boldsymbol{r}\times\boldsymbol{j})d^3r$

electric quadrupole moment: $Q_{\alpha\beta} = \frac{1}{i\omega}\int\left[r_\alpha j_\beta + r_\beta j_\alpha - \frac{2}{3}(\boldsymbol{r}\cdot\boldsymbol{j})\delta_{\alpha\beta}\right]d^3r$

magnetic quadrupole moment: $M_{\alpha\beta} = \frac{1}{3}\int\left[(\boldsymbol{r}\times\boldsymbol{j})_\alpha r_\beta + (\boldsymbol{r}\times\boldsymbol{j})_\beta r_\alpha\right]d^3r$

toroidal dipole moment: $\boldsymbol{T} = \frac{1}{10}\int\left[(\boldsymbol{r}\cdot\boldsymbol{j})\boldsymbol{r} - 2r^2\boldsymbol{j}\right]d^3r$

where c is the speed of light in medium, $\boldsymbol{j}$ is the current density, and $\alpha, \beta = x, y, z$.

In the CDA model, the j:th two-dimensional electric dipole $\boldsymbol{p}_j = (p_{xj}, p_{yj})$ located at (r_{xj}, r_{yj}) is calculated. The electric dipole and its corresponding current density are related by

$$j_{xj} = i\omega p_{xj}\,,\, j_{yj} = i\omega p_{yj}\,. \tag{S16}$$

By using the electric dipole instead of the current density in the expression of the multipole moments, we can rewrite the components of the multipoles as:

electric dipole moment:

$$p_x = \sum_j p_{xj}\,,\quad p_y = \sum_j p_{yj}\,, p_z = 0\,. \tag{S17}$$

magnetic dipole moment:

$$m_x = m_y = 0,\, m_z = \frac{i\omega}{2}\sum_j (r_{xj}p_{yj} - r_{yj}p_{xj})\,. \tag{S18}$$

electric quadrupole moment:

$$Q_{11} = \sum_j \left(\frac{4}{3}r_{xj}p_{xj} - \frac{2}{3}r_{yj}p_{yj}\right),\, Q_{22} = \sum_j \left(\frac{4}{3}r_{yj}p_{yj} - \frac{2}{3}r_{xj}p_{xj}\right),$$

$$Q_{12} = Q_{21} = \sum_j (r_{xj}p_{yj} + r_{yj}p_{xj})\,,\, Q_{33} = Q_{13} = Q_{31} = Q_{23} = Q_{32} = 0\,. \tag{S19}$$

magnetic quadrupole moment:

$$M_{13} = M_{31} = \frac{i\omega}{3}\sum_j r_{xj}(r_{xj}p_{yj} - r_{yj}p_{xj})\,, M_{23} = M_{32} = \frac{i\omega}{3}\sum_j r_{yj}(r_{xj}p_{yj} - r_{yj}p_{xj})\,,$$

$$M_{11} = M_{22} = M_{33} = M_{12} = M_{21} = 0\,. \tag{S20}$$

toroidal dipole moment:

$$T_x = \frac{i\omega}{10}\sum_j (r_{xj}p_{xj} + r_{yj}p_{yj})r_{xj} - 2(r_{xj}^2 + r_{yj}^2)p_{xj}\,,$$

$$T_y = \frac{i\omega}{10}\sum_j (r_{xj}p_{xj} + r_{yj}p_{yj})r_{yj} - 2(r_{xj}^2 + r_{yj}^2)p_{yj}\,,$$

$T_z = 0$. (S21)

Thus the electric dipole $\boldsymbol{p}_j = (p_{xj}, p_{yj})$ in the CDA model can be used for the calculation of the scattering power of the multipoles, as shown in Fig. 4 in the main text.

Reference:

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