Measuring optical activity in the far-field from a racemic nanomaterial: diffraction spectroscopy from plasmonic nanogratings

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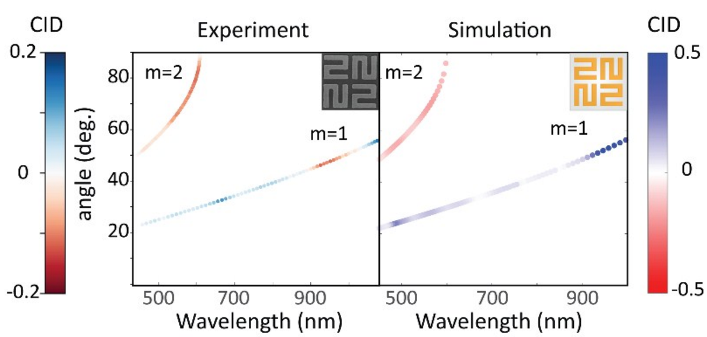
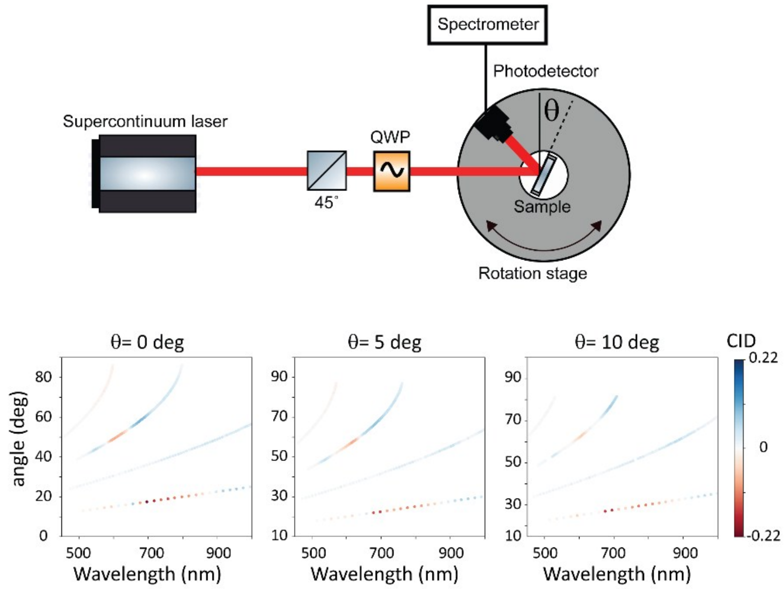
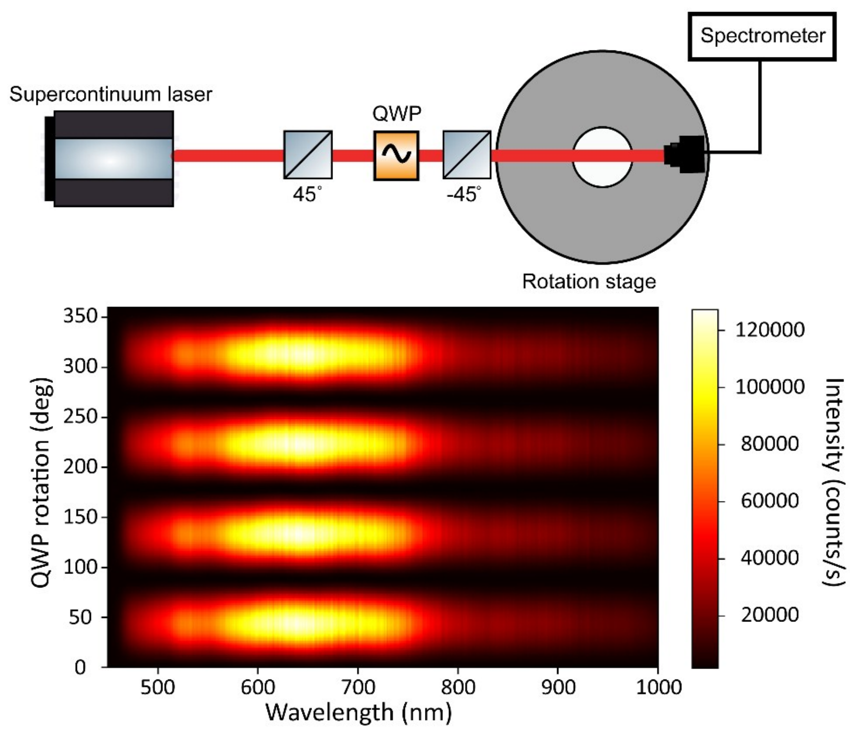
Supplementary Information



Supplementary Figure 1. Diffraction data for mirrored S-shaped nanograting perfectly flips in sign in both experiment and simulation



Supplementary Figure 2. Circular intensity difference measured for racemic nanograting almost identical for increased angles of incidence.



Supplementary Figure 3. Quarter-waveplate (QWP) long pathlength performance test. The top shows the schematic setup. Two linear polarizers are set at 90° to each other with a QWP between them. The distance between the QWP and the collection fiber is identical to the experimental configuration used for all experiments. The QWP is rotated in 1° until fully rotated and for each angle a spectrum is taken. The heatmap shows that the QWP creates circularly polarized light when orientated at 45°, 135°, 225° and 315° respectively, as expected. Thus, we can conclude that the light coupling into the fiber as well as the beam deviation induced by the QWP are negligible.

Supplementary Note 1: Near Field – Far Field Transformation

Here, we follow the general procedure for the near field – far field transformation to evaluate the electromagnetic fields at an observation plane positioned at $z > 0$ above the sample. There are three essential steps.

Part I: Main Equations including Free Electric and Magnetic Current Sources

Maxwell's equations are

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} - \mathbf{M}_f \Rightarrow \nabla \times \mathbf{E} = +i\omega \mathbf{B} - \mathbf{M}_f, \qquad \text{* MERGEFORMAT (1)}$$

$$\nabla \times \mathbf{H} = +\frac{\partial \mathbf{D}}{\partial t} + \mathbf{J}_f \Rightarrow \nabla \times \mathbf{H} = -i\omega \mathbf{D} + \mathbf{J}_f. \qquad \text{* MERGEFORMAT (2)}$$

In * MERGEFORMAT (1) and * MERGEFORMAT (2), J_f and M_f are *free* electric and magnetic currents. They are imposed currents serving as sources for electromagnetic fields E and H. Assume the space is homogeneously filled with a medium whose permittivity and permeability are ε and μ. Therefore, the electric displacement D and the magnetic induction field B are linked with the electric field and the magnetic field via,

$$\mathbf{D} = \varepsilon \mathbf{E},\ \mathbf{B} = \mu \mathbf{H}. \qquad \text{* MERGEFORMAT (3)}$$

Substitute the constitutive relations in * MERGEFORMAT (3) into * MERGEFORMAT (1) and * MERGEFORMAT (2),

$$\nabla \times \mathbf{E} = +i\omega \mathbf{B} - \mathbf{M}_f \Rightarrow \nabla \times \mathbf{E} = +i\omega\mu \mathbf{H} - \mathbf{M}_f, \qquad \text{* MERGEFORMAT (4)}$$

$$\nabla \times \mathbf{H} = -i\omega \mathbf{D} + \mathbf{J}_f \Rightarrow \nabla \times \mathbf{H} = -i\omega\varepsilon \mathbf{E} + \mathbf{J}_f. \qquad \text{* MERGEFORMAT (5)}$$

Taking the divergence of * MERGEFORMAT (5) leads to

$$\nabla \cdot \nabla \times \mathbf{H} = \nabla \cdot \left(-i\omega\varepsilon \mathbf{E} + \mathbf{J}_f\right) \Rightarrow 0 = -i\omega \nabla \cdot \mathbf{D} + i\omega\rho_e \Rightarrow \nabla \cdot \mathbf{D} = \rho_e. \qquad \text{* MERGEFORMAT (6)}$$

Taking the divergence of * MERGEFORMAT (4) leads to,

$$\nabla \cdot \nabla \times \mathbf{E} = \nabla \cdot \left(+i\omega \mathbf{B} - \mathbf{M}_f\right) \Rightarrow 0 = +i\omega \nabla \cdot \mathbf{B} - i\omega\rho_m \Rightarrow \nabla \cdot \mathbf{B} = \rho_m. \qquad \text{* MERGEFORMAT (7)}$$

In * MERGEFORMAT (6) and * MERGEFORMAT (7), we have used 1) that the divergence of the curl is zero and 2) the continuity relation.

<u>Electric Current Source</u>

When there are electric currents but no magnetic currents, equations * MERGEFORMAT (4) - * MERGEFORMAT (7) reduce to

$$\begin{gathered} \nabla \times \mathbf{E} = +i\omega\mu \mathbf{H},\ \nabla \times \mathbf{H} = -i\omega\varepsilon \mathbf{E} + \mathbf{J}_f, \\ \nabla \cdot \mathbf{D} = \rho_e,\ \nabla \cdot \mathbf{B} = 0. \end{gathered} \qquad \text{* MERGEFORMAT (8)}$$

Since the magnetic induction field is divergence free, we can introduce a vector potential,

$$\mathbf{B} = \nabla \times \mathbf{A}. \qquad \text{* MERGEFORMAT (9)}$$

Use * MERGEFORMAT (9) in the first equation of * MERGEFORMAT (8),

$$\nabla \times \mathbf{E} = +i\omega \nabla \times \mathbf{A} \Rightarrow \nabla \times \left(\mathbf{E} - i\omega \mathbf{A}\right) = 0 \Rightarrow \mathbf{E} = i\omega \mathbf{A} - \nabla \varphi_e. \qquad \text{* MERGEFORMAT (10)}$$

Substitute * MERGEFORMAT (9) and * MERGEFORMAT (10) into the second equation of * MERGEFORMAT (8),

$$\nabla^2 \mathbf{A} + k^2 \mathbf{A} = -\mu \mathbf{J}_f,\ \nabla \cdot \mathbf{A} = i\omega\varepsilon\mu\varphi_e. \qquad \text{* MERGEFORMAT (11)}$$

<u>Magnetic Current Source</u>

When there are magnetic currents but no electric currents, equation * MERGEFORMAT (4) - * MERGEFORMAT (7) reduce to

$$\begin{gathered} \nabla \times \mathbf{E} = +i\omega\mu \mathbf{H} - \mathbf{M}_f,\ \nabla \times \mathbf{H} = -i\omega\varepsilon \mathbf{E}, \\ \nabla \cdot \mathbf{D} = 0,\ \nabla \cdot \mathbf{B} = \rho_m. \end{gathered} \qquad \text{* MERGEFORMAT (12)}$$

Since the electric displacement field is divergence free, we can introduce a vector potential,

$$\mathbf{D} = -\nabla \times \mathbf{F}. \qquad \text{* MERGEFORMAT (13)}$$

Use * MERGEFORMAT (13) in the second equation of * MERGEFORMAT (12),

$$\nabla\times\mathbf{H}=i\omega\nabla\times\mathbf{F}\Rightarrow\nabla\times\left(\mathbf{H}-i\omega\mathbf{F}\right)=0\Rightarrow\mathbf{H}=i\omega\mathbf{F}-\nabla\varphi_m .$$ * MERGEFORMAT (14)

Substitute * MERGEFORMAT (13) and * MERGEFORMAT (14) into the first equation in * MERGEFORMAT (12)

$$\nabla^2\mathbf{F}+k^2\mathbf{F}=-\varepsilon\mathbf{M}_f,\ \nabla\cdot\mathbf{F}=i\omega\varepsilon\mu\varphi_m .$$ * MERGEFORMAT (15)

In * MERGEFORMAT (11) and * MERGEFORMAT (15), we have defined a wave number $k=\sqrt{\varepsilon\mu}k_0$ where k_0 is the vacuum wavenumber.

Vector potentials

From * MERGEFORMAT (11) and * MERGEFORMAT (15), the electric and magnetic vector potentials can be expressed in terms of scalar green's function $g(r,r')$ which links the field at an observation point r with the source at a source point r',

$$\mathbf{A}(\mathbf{r})=\frac{\mu}{4\pi}\int_S g(\mathbf{r},\mathbf{r}')\cdot\mathbf{J}_f(\mathbf{r}')ds'=\frac{\mu}{4\pi}\int_S\frac{e^{ik|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|}\cdot\mathbf{J}_f(\mathbf{r}')ds',$$ * MERGEFORMAT (16)

$$\mathbf{F}(\mathbf{r})=\frac{\varepsilon}{4\pi}\int_S g(\mathbf{r},\mathbf{r}')\cdot\mathbf{M}_f(\mathbf{r}')ds'=\frac{\varepsilon}{4\pi}\int_S\frac{e^{ik|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|}\cdot\mathbf{M}_f(\mathbf{r}')ds'.$$ * MERGEFORMAT (17)

In * MERGEFORMAT (16) and * MERGEFORMAT (17), the integration is conducted with respect to a domain S carrying the free electric and free magnetic currents. The domain may be a 1D, a 2D or a 3D region.

Electric and magnetic fields

Electric and magnetic fields can be expressed in terms of vector potentials,

$$\mathbf{E}=i\omega\left[\mathbf{A}+\frac{1}{k^2}\nabla\left(\nabla\cdot\mathbf{A}\right)\right]-\frac{1}{\varepsilon}\nabla\times\mathbf{F},$$ * MERGEFORMAT (18)

$$\mathbf{H}=i\omega\left[\mathbf{F}+\frac{1}{k^2}\nabla\left(\nabla\cdot\mathbf{F}\right)\right]+\frac{1}{\mu}\nabla\times\mathbf{A}.$$ * MERGEFORMAT (19)

Part 2: Scalar and Dyadic Green's Function in the Fourier-transformed Domain for the Free Space Case

The Wely identity expands the scalar green's function in * MERGEFORMAT (16) in terms of plane waves,

$$g(\mathbf{r},\mathbf{r}')=\frac{e^{ik|\mathbf{r}-\mathbf{r}'|}}{4\pi|\mathbf{r}-\mathbf{r}'|}=\frac{i}{8\pi^2}\iint_\infty\frac{e^{ik_x(x-x')+ik_y(y-y')+ik_z(z-z')}}{k_z}dk_x dk_y .$$ * MERGEFORMAT (20)

In * MERGEFORMAT (20), we assume that the observation point is above the source point $z>z'$ and k_z is defined as

$$k_z=\sqrt{k^2-k_x^2-k_y^2}.$$ * MERGEFORMAT (21)

It is always assumed that the imaginary part of k_z is positive to satisfy the radiation boundary condition.

Dyadic Green's Function in the Fourier-transformed Domain

The dyadic green's function is

$$\begin{aligned}\overline{\mathbf{G}}(\mathbf{r},\mathbf{r}') &= \left(\overline{\mathbf{I}}+\frac{1}{k^2}\nabla\nabla\right)g(\mathbf{r},\mathbf{r}') \\ &= \left(\overline{\mathbf{I}}+\frac{1}{k^2}\nabla\nabla\right)\frac{i}{8\pi^2}\iint_\infty e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}\frac{dk_x dk_y}{k_z}.\end{aligned} \qquad \text{* MERGEFORMAT (22)}$$

In * MERGEFORMAT (22), $\overline{I}$ is a unit 3 by 3 identity matrix. Especially,

$$\nabla\nabla\left(e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}\right)=\begin{pmatrix}\frac{\partial^2}{\partial x^2} & \frac{\partial^2}{\partial x\partial y} & \frac{\partial^2}{\partial x\partial z}\\ \frac{\partial^2}{\partial x\partial y} & \frac{\partial^2}{\partial y^2} & \frac{\partial^2}{\partial y\partial z}\\ \frac{\partial^2}{\partial x\partial z} & \frac{\partial^2}{\partial y\partial z} & \frac{\partial^2}{\partial z^2}\end{pmatrix}e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}=\begin{pmatrix}-k_x^2 & -k_xk_y & -k_xk_z\\ -k_xk_y & -k_y^2 & -k_yk_z\\ -k_xk_z & -k_yk_z & -k_z^2\end{pmatrix}e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}=-\mathbf{k}\mathbf{k}e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}. \qquad \text{* MERGEFORMAT (23)}$$

Substitute * MERGEFORMAT (23) into * MERGEFORMAT (22)

$$\overline{\mathbf{G}}(\mathbf{r},\mathbf{r}')=\frac{i}{8\pi^2}\iint_\infty \frac{\overline{\mathbf{I}}k^2-\mathbf{k}\mathbf{k}}{k^2k_z}e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}dk_xdk_y. \qquad \text{* MERGEFORMAT (24)}$$

The Curl of the Dyadic Green's Function in the Fourier-transformed Domain

The curl of the dyadic Green's function in * MERGEFORMAT (24) is

$$\begin{aligned}\nabla\times\overline{\mathbf{G}}(\mathbf{r},\mathbf{r}') &= \frac{i}{8\pi^2}\iint_\infty \nabla\times\left[\frac{\overline{\mathbf{I}}k^2-\mathbf{k}\mathbf{k}}{k^2k_z}e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}\right]dk_xdk_y\\ &= \frac{i}{8\pi^2}\iint_\infty \nabla e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}\times\frac{\overline{\mathbf{I}}k^2-\mathbf{k}\mathbf{k}}{k^2k_z}dk_xdk_y\\ &= \frac{i}{8\pi^2}\iint_\infty i\mathbf{k}\times\frac{\overline{\mathbf{I}}k^2-\mathbf{k}\mathbf{k}}{k^2k_z}e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}dk_xdk_y.\end{aligned} \qquad \text{* MERGEFORMAT (25)}$$

In * MERGEFORMAT (25), it is noticed that

$$\mathbf{k}\times(\mathbf{k}\mathbf{k})=(\mathbf{k}\times\mathbf{k})\mathbf{k}=\mathbf{0}. \qquad \text{* MERGEFORMAT (26)}$$

Therefore, * MERGEFORMAT (25) is reduced to

$$\nabla\times\overline{\mathbf{G}}(\mathbf{r},\mathbf{r}')=\frac{i}{8\pi^2}\iint_\infty \frac{i\mathbf{k}\times\overline{\mathbf{I}}}{k_z}e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}dk_xdk_y. \qquad \text{* MERGEFORMAT (27)}$$

Part 3: Dyadic Green's Function in the transformed space for periodic structures

Poisson Summation

For a 2D lattice, it is well-known that the Poisson summation reads,

$$\sum_{\mathbf{R_p}} e^{-i\mathbf{k}\cdot\mathbf{R_p}} = \frac{4\pi^2}{A}\sum_{\mathbf{K_n}}\delta\left(\mathbf{k}-\mathbf{K_n}\right). \qquad \text{* MERGEFORMAT (28)}$$

In * MERGEFORMAT (28), $R_p = p_1 a + p_2 b$ is a lattice vector in the 2D lattice where a and b are two unit vectors in the xy plane and $p = (p_1, p_2)$ is a two-element tuple. $K_n = n_1 r + n_2 s$ is a lattice vector in the corresponding reciprocal lattice where r and s are two unit vectors in the xy plane and $n = (n_1, n_2)$ is a two-element tuple.

Evaluation of Periodic Greens Function

The periodic sum of the dyadic green's function is

$$\begin{aligned}
&\sum_{\mathbf{R_n}} \overline{\mathbf{G}}\left(\mathbf{r},\mathbf{r}'+\mathbf{R_n}\right)e^{i\mathbf{k_P}\cdot\mathbf{R_n}} \\
&= \sum_{\mathbf{R_n}} \frac{i}{8\pi^2}\iint_\infty \frac{\overline{\mathbf{I}}k^2-\mathbf{kk}}{k^2 k_z} e^{i\mathbf{k}\cdot\left(\mathbf{r}-\mathbf{r}'-\mathbf{R_n}\right)} e^{i\mathbf{k_P}\cdot\mathbf{R_n}}\, dk_x dk_y \\
&= \frac{i}{8\pi^2}\iint_\infty \frac{\overline{\mathbf{I}}k^2-\mathbf{kk}}{k^2 k_z} e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} \sum_{\mathbf{R_n}} e^{-i\left(\mathbf{k}-\mathbf{k_P}\right)\mathbf{R_n}}\, dk_x dk_y \\
&= \frac{i}{8\pi^2}\iint_\infty \frac{\overline{\mathbf{I}}k^2-\mathbf{kk}}{k^2 k_z} e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} \cdot \frac{4\pi^2}{A}\sum_{\mathbf{K_n}}\delta\left(\mathbf{k}-\mathbf{k_P}-\mathbf{K_n}\right)\cdot dk_x dk_y \\
&= \frac{i}{2A}\sum_{\mathbf{K_n}} \frac{\overline{\mathbf{I}}k^2-\mathbf{qq}}{k^2 q_z} e^{i\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}')},
\end{aligned} \qquad \text{* MERGEFORMAT (29)}$$

In * MERGEFORMAT (29), the following definitions are employed,

$$\mathbf{q} = \left(\mathbf{k_P}+\mathbf{K_n}, q_z\right),\ q_z = \sqrt{k^2-\left(\mathbf{k_P}+\mathbf{K_n}\right)\cdot\left(\mathbf{k_P}+\mathbf{K_n}\right)}. \qquad \text{* MERGEFORMAT (30)}$$

It is noticed that in * MERGEFORMAT (29),

$$\overline{\mathbf{I}}k^2 - \mathbf{qq} = \begin{pmatrix} k^2-q_x^2 & -q_x q_y & -q_x q_z \\ -q_x q_y & k^2-q_y^2 & -q_y q_z \\ -q_x q_z & -q_y q_z & k^2-q_z^2 \end{pmatrix}. \qquad \text{* MERGEFORMAT (31)}$$

The periodic sum of the curl of the dyadic green's function is

$$\begin{aligned}
&\sum_{\mathbf{R_n}} \nabla\times\overline{\mathbf{G}}\left(\mathbf{r},\mathbf{r}'+\mathbf{R_n}\right)e^{i\mathbf{k_P}\cdot\mathbf{R_n}} \\
&= \sum_{\mathbf{R_n}} \frac{i}{8\pi^2}\iint_\infty \frac{i\mathbf{k}\times\overline{\mathbf{I}}}{k_z} e^{i\mathbf{k}\cdot\left(\mathbf{r}-\mathbf{r}'-\mathbf{R_n}\right)} e^{i\mathbf{k_P}\cdot\mathbf{R_n}}\, dk_x dk_y \\
&= \frac{i}{8\pi^2}\iint_\infty \frac{i\mathbf{k}\times\overline{\mathbf{I}}}{k_z} e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} \sum_{\mathbf{R_n}} e^{-i\left(\mathbf{k}-\mathbf{k_P}\right)\mathbf{R_n}}\, dk_x dk_y \\
&= \frac{i}{8\pi^2}\iint_\infty \frac{i\mathbf{k}\times\overline{\mathbf{I}}}{k_z} e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} \cdot \frac{4\pi^2}{A}\sum_{\mathbf{K_n}}\delta\left(\mathbf{k}-\mathbf{k_P}-\mathbf{K_n}\right)\cdot dk_x dk_y \\
&= \frac{i}{2A}\sum_{\mathbf{K_n}} \frac{i\mathbf{q}\times\overline{\mathbf{I}}}{q_z} e^{i\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}')}.
\end{aligned} \qquad \text{* MERGEFORMAT (32)}$$

In * MERGEFORMAT (32), the following definitions are employed,

$$\mathbf{q}=\left(\mathbf{k}_{\mathrm{P}}+\mathbf{K}_{\mathbf{n}},q_z\right),\ q_z=\sqrt{k^2-\left(\mathbf{k}_{\mathrm{P}}+\mathbf{K}_{\mathbf{n}}\right)\cdot\left(\mathbf{k}_{\mathrm{P}}+\mathbf{K}_{\mathbf{n}}\right)}. \qquad \text{* MERGEFORMAT (33)}$$

It is noticed that the cross product in * MERGEFORMAT (32) gives

$$\begin{aligned}\mathbf{q}\times\bar{\mathbf{I}}&=\mathbf{q}\times\left(\hat{\mathbf{x}}\hat{\mathbf{x}}+\hat{\mathbf{y}}\hat{\mathbf{y}}+\hat{\mathbf{z}}\hat{\mathbf{z}}\right)\\&=\left(\mathbf{q}\times\hat{\mathbf{x}}\right)\hat{\mathbf{x}}+\left(\mathbf{q}\times\hat{\mathbf{y}}\right)\hat{\mathbf{y}}+\left(\mathbf{q}\times\hat{\mathbf{z}}\right)\hat{\mathbf{z}}\\&=\left[q_z\hat{\mathbf{y}}\hat{\mathbf{x}}-q_y\hat{\mathbf{z}}\hat{\mathbf{x}}\right]+\left[-q_z\hat{\mathbf{x}}\hat{\mathbf{y}}+q_x\hat{\mathbf{z}}\hat{\mathbf{y}}\right]+\left[q_y\hat{\mathbf{x}}\hat{\mathbf{z}}-q_x\hat{\mathbf{y}}\hat{\mathbf{z}}\right]\\&=\begin{pmatrix}0&-q_z&q_y\\q_z&0&-q_x\\-q_y&q_x&0\end{pmatrix}.\end{aligned} \qquad \text{* MERGEFORMAT (34)}$$

In the derivation of * MERGEFORMAT (34), the following identities are used

$$\hat{\mathbf{x}}\times\hat{\mathbf{x}}=0,\ \hat{\mathbf{y}}\times\hat{\mathbf{x}}=-\hat{\mathbf{z}},\ \hat{\mathbf{z}}\times\hat{\mathbf{x}}=\hat{\mathbf{y}}, \qquad \text{* MERGEFORMAT (35)}$$

$$\hat{\mathbf{x}}\times\hat{\mathbf{y}}=\hat{\mathbf{z}},\ \hat{\mathbf{y}}\times\hat{\mathbf{y}}=\mathbf{0},\ \hat{\mathbf{z}}\times\hat{\mathbf{y}}=-\hat{\mathbf{x}}, \qquad \text{* MERGEFORMAT (36)}$$

$$\hat{\mathbf{x}}\times\hat{\mathbf{z}}=-\hat{\mathbf{y}},\ \hat{\mathbf{y}}\times\hat{\mathbf{z}}=\hat{\mathbf{x}},\ \hat{\mathbf{z}}\times\hat{\mathbf{z}}=\mathbf{0}. \qquad \text{* MERGEFORMAT (37)}$$

Electromagnetic fields due to a 2D array of sources

Based on * MERGEFORMAT (18) and * MERGEFORMAT (19), the electromagnetic fields due to a 2D array of phased sources are

$$\begin{aligned}\mathbf{E}(\mathbf{r})&=i\omega\mu\sum_{\mathbf{R_n}}\int_S\bar{\mathbf{G}}\left(\mathbf{r},\mathbf{r}'+\mathbf{R_n}\right)\cdot\mathbf{J}\left(\mathbf{r}'\right)e^{i\mathbf{k}_{\mathrm{P}}\cdot\mathbf{R_n}}ds'-\sum_{\mathbf{R_n}}\int_S\nabla\times\bar{\mathbf{G}}\left(\mathbf{r},\mathbf{r}'+\mathbf{R_n}\right)\cdot\mathbf{M}\left(\mathbf{r}'\right)e^{i\mathbf{k}_{\mathrm{P}}\cdot\mathbf{R_n}}ds'\\&=i\omega\mu\int_S\sum_{\mathbf{R_n}}\bar{\mathbf{G}}\left(\mathbf{r},\mathbf{r}'+\mathbf{R_n}\right)e^{i\mathbf{k}_{\mathrm{P}}\cdot\mathbf{R_n}}\cdot\mathbf{J}\left(\mathbf{r}'\right)ds'-\int_S\sum_{\mathbf{R_n}}\nabla\times\bar{\mathbf{G}}\left(\mathbf{r},\mathbf{r}'+\mathbf{R_n}\right)e^{i\mathbf{k}_{\mathrm{P}}\cdot\mathbf{R_n}}\cdot\mathbf{M}\left(\mathbf{r}'\right)ds',\end{aligned} \qquad \text{* MERGEFORMAT (38)}$$

$$\begin{aligned}\mathbf{H}(\mathbf{r})&=i\omega\varepsilon\sum_{\mathbf{R_n}}\int_S\bar{\mathbf{G}}\left(\mathbf{r},\mathbf{r}'+\mathbf{R_n}\right)\cdot\mathbf{M}\left(\mathbf{r}'\right)e^{i\mathbf{k}_{\mathrm{P}}\cdot\mathbf{R_n}}ds'+\sum_{\mathbf{R_n}}\int_S\nabla\times\bar{\mathbf{G}}\left(\mathbf{r},\mathbf{r}'+\mathbf{R_n}\right)\cdot\mathbf{J}\left(\mathbf{r}'\right)e^{i\mathbf{k}_{\mathrm{P}}\cdot\mathbf{R_n}}ds'\\&=i\omega\varepsilon\int_S\sum_{\mathbf{R_n}}\bar{\mathbf{G}}\left(\mathbf{r},\mathbf{r}'+\mathbf{R_n}\right)e^{i\mathbf{k}_{\mathrm{P}}\cdot\mathbf{R_n}}\cdot\mathbf{M}\left(\mathbf{r}'\right)ds'+\int_S\sum_{\mathbf{R_n}}\nabla\times\bar{\mathbf{G}}\left(\mathbf{r},\mathbf{r}'+\mathbf{R_n}\right)e^{i\mathbf{k}_{\mathrm{P}}\cdot\mathbf{R_n}}\cdot\mathbf{J}\left(\mathbf{r}'\right)ds'.\end{aligned} \qquad \text{* MERGEFORMAT (39)}$$

Using the final results from * MERGEFORMAT (29) and * MERGEFORMAT (32), e.g., in * MERGEFORMAT (38), we find

$$\begin{aligned}\mathbf{E}(\mathbf{r})&=i\omega\mu\int_S\frac{i}{2A}\sum_{\mathbf{K_n}}\frac{\bar{\mathbf{I}}k^2-\mathbf{q}\mathbf{q}}{k^2q_z}e^{i\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}')}\cdot\mathbf{J}\left(\mathbf{r}'\right)ds'-\int_S\frac{i}{2A}\sum_{\mathbf{K_n}}\frac{i\mathbf{q}\times\bar{\mathbf{I}}}{q_z}e^{i\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}')}\cdot\mathbf{M}\left(\mathbf{r}'\right)ds'\\&=i\omega\mu\cdot\frac{i}{2A}\sum_{\mathbf{K_n}}\frac{\bar{\mathbf{I}}k^2-\mathbf{q}\mathbf{q}}{k^2q_z}\cdot\int_S\mathbf{J}\left(\mathbf{r}'\right)e^{-i\mathbf{q}\cdot\mathbf{r}'}ds'\cdot e^{i\mathbf{q}\cdot\mathbf{r}}-\frac{i}{2A}\sum_{\mathbf{K_n}}\frac{i\mathbf{q}\times\bar{\mathbf{I}}}{q_z}\cdot\int_S\mathbf{M}\left(\mathbf{r}'\right)e^{-i\mathbf{q}\cdot\mathbf{r}'}ds'\cdot e^{i\mathbf{q}\cdot\mathbf{r}}\\&=\sum_{\mathbf{K_n}}\mathbf{E}\left(\mathbf{q}\right)e^{i\mathbf{q}\cdot\mathbf{r}}.\end{aligned} \qquad \text{* MERGEFORMAT (40)}$$

In * MERGEFORMAT (40), we have the following definitions,

$$\mathbf{E}\left(\mathbf{q}\right)=\mathbf{G}_{ee}\left(\mathbf{q}\right)\cdot\mathbf{J}\left(\mathbf{q}\right)+\mathbf{G}_{em}\left(\mathbf{q}\right)\cdot\mathbf{M}\left(\mathbf{q}\right). \qquad \text{* MERGEFORMAT (41)}$$

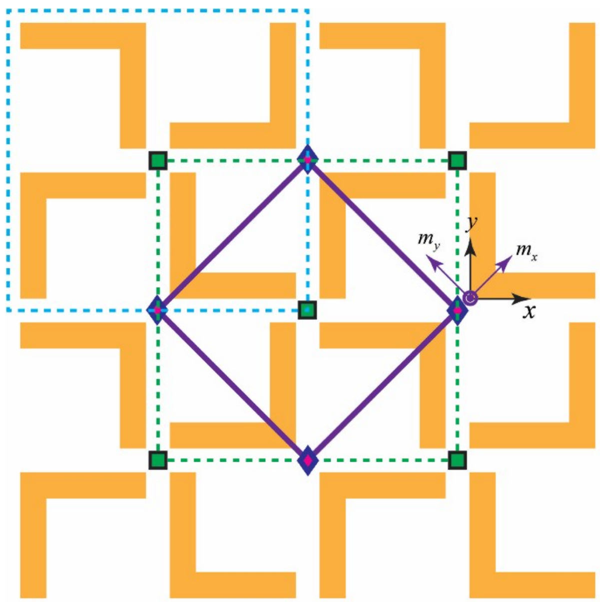
In * MERGEFORMAT (41), G_{ee} and G_{em} are the dyadic Green's function that respectively link electric currents and magnetic currents with electric fields,

$$\mathbf{G}_{ee}(\mathbf{q}) = i\omega\mu \cdot \frac{i}{2A}\frac{\bar{\mathbf{I}}k^2 - \mathbf{q}\mathbf{q}}{k^2 q_z},\ \mathbf{G}_{em}(\mathbf{q}) = -\frac{i}{2A}\frac{i\mathbf{q}\times\bar{\mathbf{I}}}{q_z}. \qquad \text{* MERGEFORMAT (42)}$$

And $J(q)$ and $M(q)$ are the Fourier coefficients,

$$\mathbf{J}(\mathbf{q}) = \int_S \mathbf{J}(\mathbf{r}')e^{-i\mathbf{q}\cdot\mathbf{r}'}ds',\ \mathbf{M}(\mathbf{q}) = \int_S \mathbf{M}(\mathbf{r}')e^{-i\mathbf{q}\cdot\mathbf{r}'}ds'. \qquad \text{* MERGEFORMAT (43)}$$

Supplementary Note 2: Symmetries of the 4L Racemic Structure



Supplementary Figure 4. An illustration of the symmetries in the 4L racemic structure. In the plot, the squares filled by the green colour represent a centre of rotation of order four, while the diamonds filled by the pink colours represent a centre of rotation of order two. Further, the solid purple lines represent two axes of reflection. The cyan and green dashed squares delineate the "normal" and "mirror" 4L unit cells, respectively. Lastly, two possible coordinate frames, that is, the $x-y$ coordinate system and the m_x-m_y coordinate system, are marked by the black and purple colours.

It can be readily seen from each unit cell that the sample holds a four-fold rotational symmetry. Additionally, it is less apparent that there are two reflection axes (see the purple solid lines in Figure S4) and they do not coincide with the rotation centres of the unit cells. These symmetries form a D_4 point group. The point group together with the square lattice form the so-called $p4g$ plane group[1].

Supplementary Note 3: Polarization of Incident Field in Different Coordinate Frame

Assume that there is a plane wave propagating along the negative z direction and being left (right) circularly polarized with an unit amplitude. As shown in Figure S4, in the $x-y$ coordinate frame (i.e., the original frame), the polarization can be expressed as

$$\mathbf{E}_L = \frac{1}{\sqrt{2}}\left(\hat{\mathbf{x}} + i\hat{\mathbf{y}}\right)e^{-ik_z z},\ \mathbf{E}_R = \frac{1}{\sqrt{2}}\left(\hat{\mathbf{x}} - i\hat{\mathbf{y}}\right)e^{-ik_z z}. \qquad \text{* MERGEFORMAT (44)}$$

In the $m_x - m_y$ coordinate frame (i.e. the rotated frame), the polarization can be expressed as

$$\mathbf{E}_L = E_0\left(\hat{\mathbf{m}}_\mathbf{x} + i\hat{\mathbf{m}}_\mathbf{y}\right),\ \mathbf{E}_R = E_0'\left(\hat{\mathbf{m}}_\mathbf{x} - i\hat{\mathbf{m}}_\mathbf{y}\right). \qquad \text{* MERGEFORMAT (45)}$$

In * MERGEFORMAT (44) and * MERGEFORMAT (45), $\hat{x}$ and $\hat{y}$ are the unit vectors in the $x-y$ coordinate frame, while $\hat{m}_x$ and $\hat{m}_y$ being the unit vectors in the $m_x - m_y$ coordinate frame. The unit vectors in the original and rotated frame are related,

$$\hat{\mathbf{m}}_\mathbf{x} = \frac{1}{\sqrt{2}}\hat{\mathbf{x}} + \frac{1}{\sqrt{2}}\hat{\mathbf{y}},\ \hat{\mathbf{m}}_\mathbf{y} = -\frac{1}{\sqrt{2}}\hat{\mathbf{x}} + \frac{1}{\sqrt{2}}\hat{\mathbf{y}}. \qquad \text{* MERGEFORMAT (46)}$$

Therefore, the field components in the rotated frame are,

$$E_0 = \frac{1}{2}(1+i),\ E_0' = \frac{1}{2}(1-i). \qquad \text{* MERGEFORMAT (47)}$$

Therefore, combing * MERGEFORMAT (45) with * MERGEFORMAT (47) shows that,

$$\mathbf{E}_L = \frac{1}{\sqrt{2}}e^{i\pi/4}\left(\hat{\mathbf{m}}_\mathbf{x} + i\hat{\mathbf{m}}_\mathbf{y}\right)e^{-ik_z z},\ \mathbf{E}_R = \frac{1}{\sqrt{2}}e^{-i\pi/4}\left(\hat{\mathbf{m}}_\mathbf{x} - i\hat{\mathbf{m}}_\mathbf{y}\right)e^{-ik_z z}. \qquad \text{* MERGEFORMAT (48)}$$

* MERGEFORMAT (48) tells that the light is still a left (right) circularly polarized in the rotated frame but with a possible phase difference, which is expected because a coordinate transformation does not alter the physical property of the system.

Supplementary Note 4: Symmetries in the Response from the 4L Racemic Structure

For the sake of clarity, we present the main conclusion of this section: <u>the reflection from the 4L Racemic Structure is identical for the LCP incident light and the RCP incident light</u>. In the following, we prove this argument by rigorous mathematical procedures.

<u>Step 1: Volume Integral Equation (VIE) Formulation for the Light-Matter Interaction</u>

Before delving into the details, we would like to clarify two conventions adopted in the derivations. On the one hand, we use the SI units. On the other hand, since we formulate the problem in the frequency domain, we employ the physical time convention, i.e., $e^{-i\omega t}$ where ω is the angular frequency.

As discussed in [2–4] and under the assumption of the *local-response* material model, the interaction of light with metallic nanoscatterers (e.g. the 4L structures in the 2D array) can be described in terms of a volume integral equation (VIE),

$$Z\left(\mathbf{J}_{\mathbf{ind}}\left(\mathbf{r}\right)\right)=\mathbf{E}_{\mathbf{inc}}\left(\mathbf{r}\right), \qquad \text{* MERGEFORMAT (49)}$$

In * MERGEFORMAT (49), by applying the volume equivalence principle [2–4], $J_{ind}(r)$ are induced electric currents flowing in the volume of the nanoscatterers and $E_{inc}(r)$ is an incident light applied onto the scatterers. The Z operator is defined as

$$Z\left(\mathbf{J}_{\mathbf{ind}}\left(\mathbf{r}\right)\right)=\frac{1}{-i\omega\varepsilon_0\left(\varepsilon_r\left(\mathbf{r},\omega\right)-1\right)}\mathbf{J}_{\mathbf{ind}}\left(\mathbf{r}\right)-i\omega\mu_0\int_V \mathbf{G}\left(\mathbf{r},\mathbf{r'},\omega\right)\cdot\mathbf{J}_{\mathbf{ind}}\left(\mathbf{r}'\right)\,dv'. \qquad \text{* MERGEFORMAT (50)}$$

In * MERGEFORMAT (50), we assume that the nanoscatterers are positioned on top of a multilayer structure (e.g., in our experiment, the layers include the SiO2 layer and the Si layer) and immersed in vacuum. The vacuum is characterized by the vacuum permittivity ε_0 and the vacuum permeability μ_0. Then, the nanoscatterers occupying a volume V are assumed to be non-magnetic and are electromagnetically characterized by a frequency dependent permittivity $\varepsilon_r(\omega)$. Lastly, $G(r,r',\omega)$ is the so-called dyadic Green's function, which links an observation point r with a source point r' at a frequency ω. Due to the multilayer structure considered, the dyadic Green's function has two parts, the *direct field* part, $G_0(r,r',\omega)$,and the *reflected field* part, $G_r(r,r',\omega)$**.** Both Green's functions can be compactly written in the following generic form in the reciprocal space,

$$\mathbf{G}_{0/r}\left(\mathbf{r},\mathbf{r}',\omega\right)=\mathbf{G}_{0/r}^{s}\left(\mathbf{r},\mathbf{r}',\omega\right)+\mathbf{G}_{0/r}^{p}\left(\mathbf{r},\mathbf{r}',\omega\right). \qquad \text{* MERGEFORMAT (51)}$$

In * MERGEFORMAT (51), the dyadic Green's function for the direct field case and the reflected field case is split into two parts: the part for the *s* polarized waves and the part for the *p* polarized waves. Each can be further expanded in the spectral domain (or the reciprocal space),

$$\mathbf{G}_{0/r}^{s/p}\left(\mathbf{r},\mathbf{r}',\omega\right)=\frac{i}{8\pi^2}\iint_\infty \mathbf{M}_{0/r}^{s/p}\left(k_x,k_y,z,z'\right)e^{ik_x(x-x')+ik_y(y-y')}dk_x dk_y. \qquad \text{* MERGEFORMAT (52)}$$

In * MERGEFORMAT (52), M is the spectral domain representation of the corresponding dyadic Green's function. It is functions of in-plane wave numbers k_x and k_y and the z coordinate of the observation point and the source point. Their detailed forms can be found in, e.g. [5], and listed here,

$$\mathbf{M}_0^{s}\left(k_x,k_y,z,z'\right)=\frac{1}{k_z k_\rho^2}\begin{pmatrix} k_y^2 & -k_x k_y & 0 \\ -k_x k_y & k_x^2 & 0 \\ 0 & 0 & 0 \end{pmatrix} e^{ik_z|z-z'|}, \qquad \text{* MERGEFORMAT (53)}$$

$$\mathbf{M}_0^{p}\left(k_x,k_y,z,z'\right)=\frac{1}{k_0^2 k_\rho^2}\begin{pmatrix} k_x^2 k_z & k_x k_y k_z & \mp k_x k_\rho^2 \\ k_x k_y k_z & k_y^2 k_z & \mp k_y k_\rho^2 \\ \mp k_x k_\rho^2 & \mp k_y k_\rho^2 & k_\rho^4/k_z \end{pmatrix} e^{ik_z|z-z'|}, \qquad \text{* MERGEFORMAT (54)}$$

$$\mathbf{M}_r^s\left(k_x,k_y,z,z'\right)=\frac{r_s\left(k_\rho\right)}{k_z k_\rho^2}\begin{pmatrix} k_y^2 & -k_x k_y & 0 \\ -k_x k_y & k_x^2 & 0 \\ 0 & 0 & 0 \end{pmatrix} e^{ik_z(z+z')}, \qquad \text{* MERGEFORMAT (55)}$$

$$\mathbf{M}_r^p\left(k_x,k_y,z,z'\right)=\frac{-r_p\left(k_\rho\right)}{k_0^2 k_\rho^2}\begin{pmatrix} k_x^2 k_z & k_x k_y k_z & +k_x k_\rho^2 \\ k_x k_y k_z & k_y^2 k_z & +k_y k_\rho^2 \\ -k_x k_\rho^2 & -k_y k_\rho^2 & -k_\rho^4/k_z \end{pmatrix} e^{ik_z|z-z'|}. \qquad \text{* MERGEFORMAT (56)}$$

In * MERGEFORMAT (53) - * MERGEFORMAT (56), k_ρ and k_z are the lateral and vertical wave numbers and defined as,

$$k_\rho=\sqrt{k_x^2+k_y^2},\ k_z=\sqrt{k_0^2-k_\rho^2}. \qquad \text{* MERGEFORMAT (57)}$$

The square root in the second equation in * MERGEFORMAT (57) always takes a positive imaginary part to ensure the radiation boundary condition.

<u>Step 2: the P_{M_x} operator</u>

Consider the $m_x - m_y$ coordinate frame and especially focus on a symmetry operation in the D_4 group, that is, M_x which mirrors a vector with respect to the m_x reflection axis. A representation of this geometric transformation is a matrix,

$$M_x=\begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \qquad \text{* MERGEFORMAT (58)}$$

Besides, we accordingly introduce a transformation operator[6] corresponding to the M_x operation. The operator works on a function which can be a scalar function (e.g., an electric charge distribution) and a vector function (e.g., an electric field distribution) and is defined as

$$P_{M_x}\left(f\left(\mathbf{r}\right)\right)=f\left(M_x^{-1}\mathbf{r}\right),\ P_{M_x}\left(\mathbf{f}\left(\mathbf{r}\right)\right)=M_x\cdot\mathbf{f}\left(M_x^{-1}\mathbf{r}\right). \qquad \text{* MERGEFORMAT (59)}$$

In * MERGEFORMAT (59), M_x^{-1} represents the inverse of the transformation matrix in * MERGEFORMAT (58).

<u>Step 3: Commutative relation between the P_{M_x} operator and the Z operator</u>

In this section, we prove that the P_{M_x} operator and the Z operator are indeed commutative, that is,

$$P_{M_x}Z\left(\mathbf{J}_{\mathbf{ind}}\left(\mathbf{r}\right)\right)=ZP_{M_x}\left(\mathbf{J}_{\mathbf{ind}}\left(\mathbf{r}\right)\right). \qquad \text{* MERGEFORMAT (60)}$$

Consider the $m_x - m_y$ coordinate frame. On the one hand, we focus on the first term in * MERGEFORMAT (50). Since the permittivity profile is invariant under the symmetry operation,

$$P_{M_x}\left(\varepsilon_r\left(\mathbf{r}\right)\right)=\varepsilon_r\left(M_x^{-1}\mathbf{r}\right)=\varepsilon_r\left(\mathbf{r}\right), \qquad \text{* MERGEFORMAT (61)}$$

applying the transformation operator to the first term in * MERGEFORMAT (50) results in

$$\begin{aligned}
&P_{M_x}\left(\frac{1}{-i\omega\varepsilon_0\left(\varepsilon_r\left(\mathbf{r},\omega\right)-1\right)}\mathbf{J}_{\mathbf{ind}}\left(\mathbf{r}\right)\right)\\
&=M_x\cdot\frac{1}{-i\omega\varepsilon_0\left(\varepsilon_r\left(M_x^{-1}\mathbf{r},\omega\right)-1\right)}\mathbf{J}_{\mathbf{ind}}\left(M_x^{-1}\mathbf{r}\right)\\
&=\frac{1}{-i\omega\varepsilon_0\left(\varepsilon_r\left(M_x^{-1}\mathbf{r},\omega\right)-1\right)}M_x\cdot\mathbf{J}_{\mathbf{ind}}\left(M_x^{-1}\mathbf{r}\right)\\
&=\frac{1}{-i\omega\varepsilon_0\left(\varepsilon_r\left(\mathbf{r},\omega\right)-1\right)}P_{M_x}\left(\mathbf{J}_{\mathbf{ind}}\left(\mathbf{r}\right)\right).
\end{aligned}$$ * MERGEFORMAT (62)

Therefore, the commutative relation is proved. On the other hand, we focus on the volume integral in the second term in * MERGEFORMAT (50). Apply the transformation operator to the integral leads to,

$$\begin{aligned}
&P_{M_x}\left(\int_V \mathbf{G}\left(\mathbf{r},\mathbf{r}',\omega\right)\cdot\mathbf{J}_{\mathbf{ind}}\left(\mathbf{r}'\right)dv'\right)\\
&=M_x\cdot\int_V \mathbf{G}\left(M_x^{-1}\mathbf{r},\mathbf{r}',\omega\right)\cdot\mathbf{J}_{\mathbf{ind}}\left(\mathbf{r}'\right)dv'\\
&=\int_V M_x\cdot\mathbf{G}\left(M_x^{-1}\mathbf{r},\mathbf{r}',\omega\right)\cdot M_x^{-1}\cdot M_x\mathbf{J}_{\mathbf{ind}}\left(\mathbf{r}'\right)dv'.
\end{aligned}$$ * MERGEFORMAT (63)

In * MERGEFORMAT (63), it is noted that the transformation is for the observation coordinates. Substitute the spectral domain representation of the dyadic Green's function in * MERGEFORMAT (53) - * MERGEFORMAT (56) into * MERGEFORMAT (63). Especially, take the direct field dyadic Green's function for the s polarized waves (i.e., * MERGEFORMAT (53)) as an example,

$$\begin{aligned}
&\int_V M_x\cdot\mathbf{G}\left(M_x^{-1}\mathbf{r},\mathbf{r}',\omega\right)\cdot M_x^{-1}\cdot M_x\mathbf{J}_{\mathbf{ind}}\left(\mathbf{r}'\right)dv'\\
&=\int_V M_x\cdot\left[\frac{i}{8\pi^2}\iint_\infty\frac{1}{k_zk_\rho^2}\begin{pmatrix}k_y^2 & -k_xk_y & 0\\ -k_xk_y & k_x^2 & 0\\ 0 & 0 & 0\end{pmatrix}e^{ik_z|z-z'|}e^{ik_x(x-x')+ik_y(-y-y')}dk_xdk_y\right]\cdot M_x^{-1}\cdot M_x\mathbf{J}_{\mathbf{ind}}\left(\mathbf{r}'\right)dv'\\
&=\int_V M_x\cdot\left[\frac{i}{8\pi^2}\iint_\infty\frac{1}{k_zk_\rho^2}\begin{pmatrix}k_y^2 & k_xk_y & 0\\ k_xk_y & k_x^2 & 0\\ 0 & 0 & 0\end{pmatrix}e^{ik_z|z-z'|}e^{ik_x(x-x')+ik_y(y+y')}dk_xdk_y\right]\cdot M_x^{-1}\cdot M_x\mathbf{J}_{\mathbf{ind}}\left(\mathbf{r}'\right)dv'.
\end{aligned}$$ * MERGEFORMAT (64)

In * MERGEFORMAT (64), to reach the second equality, a change of variable $k_\rho \to -k_\rho$ is made. Then, a change of variable $y' \to -y'$ is conducted,

$$\int_V M_x \cdot \left[\frac{i}{8\pi^2} \iint_\infty \frac{1}{k_z k_\rho^2} \begin{pmatrix} k_y^2 & k_x k_y & 0 \\ k_x k_y & k_x^2 & 0 \\ 0 & 0 & 0 \end{pmatrix} e^{ik_z|z-z'|} e^{ik_x(x-x')+ik_y(y+y')} dk_x dk_y \right] \cdot M_x^{-1} \cdot M_x \cdot \mathbf{J_{ind}}(\mathbf{r}')\, dv'$$

$$= \int_V \left[\frac{i}{8\pi^2} \iint_\infty \frac{1}{k_z k_\rho^2} M_x \cdot \begin{pmatrix} k_y^2 & k_x k_y & 0 \\ k_x k_y & k_x^2 & 0 \\ 0 & 0 & 0 \end{pmatrix} e^{ik_z|z-z'|} \cdot M_x^{-1} e^{ik_x(x-x')+ik_y(y-y')} dk_x dk_y \right] \cdot M_x \cdot \mathbf{J_{ind}}(x', -y', z')\, dv'$$

$$= \int_V \left[\frac{i}{8\pi^2} \iint_\infty \frac{1}{k_z k_\rho^2} M_x \cdot \begin{pmatrix} k_y^2 & k_x k_y & 0 \\ k_x k_y & k_x^2 & 0 \\ 0 & 0 & 0 \end{pmatrix} e^{ik_z|z-z'|} \cdot M_x^{-1} e^{ik_x(x-x')+ik_y(y-y')} dk_x dk_y \right] \cdot M_x \cdot \mathbf{J_{ind}}\left(M_x^{-1}\mathbf{r}'\right) dv' \quad \text{* MERGEFORMAT (65)}$$

$$= \int_V \left[\frac{i}{8\pi^2} \iint_\infty \frac{1}{k_z k_\rho^2} M_x \cdot \begin{pmatrix} k_y^2 & k_x k_y & 0 \\ k_x k_y & k_x^2 & 0 \\ 0 & 0 & 0 \end{pmatrix} e^{ik_z|z-z'|} \cdot M_x^{-1} e^{ik_x(x-x')+ik_y(y-y')} dk_x dk_y \right] \cdot P_{M_x}\left(\mathbf{J_{ind}}(\mathbf{r}')\right) dv'.$$

Lastly, it is not hard to see that

$$M_x \cdot \begin{pmatrix} k_y^2 & k_x k_y & 0 \\ k_x k_y & k_x^2 & 0 \\ 0 & 0 & 0 \end{pmatrix} \cdot M_x^{-1} = \begin{pmatrix} k_y^2 & -k_x k_y & 0 \\ -k_x k_y & k_x^2 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \qquad \text{* MERGEFORMAT (66)}$$

Therefore, we prove that

$$P_{M_x}\left(\int_V \mathbf{G}_0^s(\mathbf{r},\mathbf{r}',\omega) \cdot \mathbf{J_{ind}}(\mathbf{r}')\, dv'\right) = \int_V \mathbf{G}_0^s(\mathbf{r},\mathbf{r}',\omega) \cdot P_{M_x}\left(\mathbf{J_{ind}}(\mathbf{r}')\right) dv'. \quad \text{* MERGEFORMAT (67)}$$

The same procedures can be applied to the rest cases (i.e., * MERGEFORMAT (54) - * MERGEFORMAT (56)). Finally, the commutative relation in * MERGEFORMAT (60) is proved.

<u>Step 4: On the Application of the Transformation Operator in * MERGEFORMAT (59) to * MERGEFORMAT (49)</u>

First, we assume a LCP incident light,

$$\mathbf{E}_L(\mathbf{r}) = \frac{1}{\sqrt{2}}\left(\hat{\mathbf{m}}_\mathbf{x} + i\hat{\mathbf{m}}_\mathbf{y}\right) e^{-ik_z z}. \qquad \text{* MERGEFORMAT (68)}$$

Mind the phase difference between the LCP in * MERGEFORMAT (48) and * MERGEFORMAT (68). Through * MERGEFORMAT (49) we find the induced current as,

$$Z\left(\mathbf{J}_L(\mathbf{r})\right) = \mathbf{E}_L(\mathbf{r}). \qquad \text{* MERGEFORMAT (69)}$$

Consider the $m_x - m_y$ coordinate frame and apply the transformation operator in * MERGEFORMAT (59) to both sides of * MERGEFORMAT (69),

$$Z\left(P_{M_x}\mathbf{J}_L(\mathbf{r})\right) = P_{M_x}\mathbf{E}_L(\mathbf{r}). \qquad \text{* MERGEFORMAT (70)}$$

In the derivation of * MERGEFORMAT (70), we use the commutative relation in * MERGEFORMAT (60). It is not hard to see that the right hand side of * MERGEFORMAT (70) is just the right circularly polarized light,

$$ZP_{M_x}\left(\mathbf{J}_L(\mathbf{r})\right)=\mathbf{E}_R(\mathbf{r})\Leftrightarrow Z\left(\mathbf{J}_R(\mathbf{r})\right)=\mathbf{E}_R(\mathbf{r}). \quad \text{* MERGEFORMAT (71)}$$

Therefore, we can conclude that *the induced current due to the LCP can be transformed into the induced current due to the RCP,*

$$\mathbf{J}_R(\mathbf{r})=P_{M_x}\left(\mathbf{J}_L(\mathbf{r})\right). \quad \text{* MERGEFORMAT (72)}$$

<u>Step 4: the Scattered Field, the Poynting Vector and the Reflected Power</u>

In practice, instead of the induced currents flowing in the nanoscatterers, we are more interested the scattered field generated by these currents,

$$\mathbf{E}_s(\mathbf{r})=-i\omega\mu_0\int_V \mathbf{G}(\mathbf{r},\mathbf{r'},\omega)\cdot\mathbf{J}_{\mathbf{ind}}(\mathbf{r}')\,dv',\ \mathbf{H}_s(\mathbf{r})=\frac{1}{i\omega\mu_0}\nabla\times\mathbf{E}_s(\mathbf{r}). \quad \text{* MERGEFORMAT (73)}$$

The volume integral in * MERGEFORMAT (73) is conducted over the volume of all the nanoscatterers and the Green's function is the same as the one in * MERGEFORMAT (52) - * MERGEFORMAT (56). Then, the scattered electric fields generated by the induced currents in * MERGEFORMAT (69) and * MERGEFORMAT (71) are

$$\mathbf{E}_L(\mathbf{r})=-i\omega\mu_0\int_V \mathbf{G}(\mathbf{r},\mathbf{r'},\omega)\cdot\mathbf{J}_L(\mathbf{r}')\,dv',\ \mathbf{E}_R(\mathbf{r})=-i\omega\mu_0\int_V \mathbf{G}(\mathbf{r},\mathbf{r'},\omega)\cdot\mathbf{J}_R(\mathbf{r}')\,dv'. \quad \text{* MERGEFORMAT (74)}$$

Take into account the relation in * MERGEFORMAT (72) and the commutative relation in * MERGEFORMAT (67),

$$\mathbf{E}_R(\mathbf{r})=P_{M_x}\left(\mathbf{E}_L(\mathbf{r})\right). \quad \text{* MERGEFORMAT (75)}$$

For the magnetic fields, it can be proven that

$$\mathbf{H}_R(\mathbf{r})=-P_{M_x}\left(\mathbf{H}_L(\mathbf{r})\right). \quad \text{* MERGEFORMAT (76)}$$

From * MERGEFORMAT (75) and * MERGEFORMAT (76), the z component of the Poynting vector can be derived,

$$S_{L,z}(x,y,z)=E_{L,x}(x,y,z)H^*_{L,y}(x,y,z)-E_{L,y}(x,y,z)H^*_{L,x}(x,y,z). \quad \text{* MERGEFORMAT (77)}$$

$$S_{R,z}(x,y,z)=E_{L,x}(x,-y,z)H^*_{L,y}(x,-y,z)-E_{L,y}(x,-y,z)H^*_{L,x}(x,-y,z). \quad \text{* MERGEFORMAT (78)}$$

Integrate * MERGEFORMAT (77) and * MERGEFORMAT (78) with respect to an infinitely large observation plane at z above the structure,

$$\begin{aligned}P_L(z)&=\int_{S_\infty} S_{L,z}(x,y,z)dxdy\\ &=\int_{S_\infty}\left[E_{L,x}(x,y,z)H^*_{L,y}(x,y,z)-E_{L,y}(x,y,z)H^*_{L,x}(x,y,z)\right]dxdy,\end{aligned} \quad \text{* MERGEFORMAT (79)}$$

$$
\begin{aligned}
P_R(z) &= \int_{S_\infty} S_{R,z}(x,y,z)\,dxdy \\
&= \int_{S_\infty} \left[E_{L,x}(x,-y,z) H_{L,y}^{*}(x,-y,z) - E_{L,y}(x,-y,z) H_{L,x}^{*}(x,-y,z) \right] dxdy \\
&= \int_{S_\infty} \left[E_{L,x}(x,y,z) H_{L,y}^{*}(x,y,z) - E_{L,y}(x,y,z) H_{L,x}^{*}(x,y,z) \right] dxdy.
\end{aligned}
$$ * MERGEFORMAT (80)

Therefore,

$$P_L(z) = P_R(z).$$ * MERGEFORMAT (81)

Eq. * MERGEFORMAT (81) simply points out that the reflected power for the LCP and RCP incident cases is identical. It should be noted that * MERGEFORMAT (79) and * MERGEFORMAT (80) are the reflected power due to the incident field in * MERGEFORMAT (68). For the incident field in * MERGEFORMAT (48) (which is actually used in the experiment), there is an extra phase factor. However, this phase factor disappears due to the complex conjugation in * MERGEFORMAT (79) and * MERGEFORMAT (80).

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