Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information for Diatomic terahertz metasurfaces for arbitrary-to-circular polarization conversion

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Note 1: Normalized magnetic field distributions of different meta-atoms

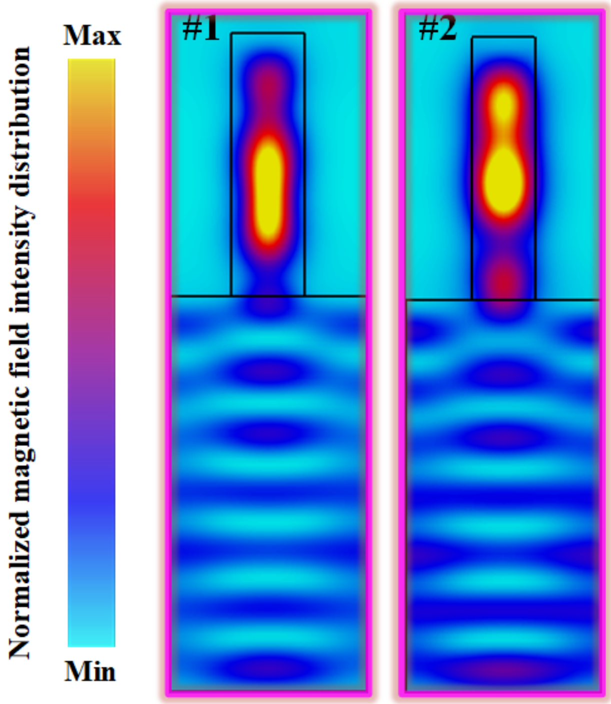

Fig. S1. Normalized magnetic field distributions of meta-atoms with different radii under the illumination of linearly polarized waves.

Note 2: Jones matrix analysis of arbitrary-to-RCP polarization conversion

Here, In the global linear polarization base defined in the *xoy* coordinate system, we can arbitrarily define three polarization states as follows,

Case 1: on the equator of the Poincaré Sphere, namely, linear polarization state α (ψ=5π/8, χ=0),

$$E_{out}=T_{xoy}^{linear}\alpha=e^{i\left(q_1+2\varphi_1+\frac{7}{8}\pi\right)}\begin{bmatrix}1\\-i\end{bmatrix} \tag{S1}$$

And, its orthogonal linear polarization state β (ψ=π/8, χ=0),

$$E_{out}=T_{xoy}^{linear}\beta=e^{i\left(q_1+2\varphi_1+\frac{11}{8}\pi\right)}\begin{bmatrix}1\\-i\end{bmatrix} \tag{S2}$$

Case 2: any other point on the Poincaré sphere, namely, elliptical polarization state, such as α (ψ=5π/8, χ=π/8),

$$E_{out}=T_{xoy}^{linear}\alpha=e^{i\left(q_1+2\varphi_1+\frac{7}{8}\pi\right)}\left(\cos\frac{\pi}{8}-\sin\frac{\pi}{8}\right)\begin{bmatrix}1\\-i\end{bmatrix} \tag{S3}$$

And, its orthogonal elliptical polarization state β (ψ=π/8, χ=-π/8),

$$E_{out}=T_{xoy}^{linear}\beta=e^{i\left(q_1+2\varphi_1+\frac{11}{8}\pi\right)}\left(\cos\frac{\pi}{8}+\sin\frac{\pi}{8}\right)\begin{bmatrix}1\\-i\end{bmatrix} \tag{S4}$$

Case 3: The North and South Points of the Poincaré Sphere, namely, circular polarization state,
North (RCP), α (ψ=5π/8, χ=π/4),

$$E_{out}=T_{xoy}^{linear}\alpha=e^{i\left(q_1+2\varphi_1+\frac{7}{8}\pi\right)}\left(\cos\frac{\pi}{4}-\sin\frac{\pi}{4}\right)\begin{bmatrix}1\\-i\end{bmatrix}=0 \tag{S5}$$

South (LCP), β (ψ=π/8, χ=-π/4),

$$E_{out}=T_{xoy}^{linear}\beta=e^{i\left(q_1+2\varphi_1+\frac{11}{8}\pi\right)}\left(\cos\frac{\pi}{4}+\sin\frac{\pi}{4}\right)\begin{bmatrix}1\\-i\end{bmatrix}=\sqrt{2}e^{i\left(q_1+2\varphi_1+\frac{11}{8}\pi\right)}\begin{bmatrix}1\\-i\end{bmatrix} \tag{S6}$$

The modified polarization conversion efficiencies were calculated and are shown in Fig.1(b). and the corresponding intensity can be described as $T_{L/R}/T_{spacer}=|M_{L/R}|^2/(|M_x|^2+|M_y|^2)$, $T_{L/R}$ is the transmittance of the transmitted LCP or RCP component, and the amplitude is M_L or M_R. T_{spacer} is the background transmittance of the substrate, and the amplitudes of the transmitted *x* and *y* components represent M_x and M_y, respectively. At the pre-designed operating frequency, the incident LCP wave can be maximally converted to its orthogonal polarization mode, namely, RCP, with a theoretical conversion efficiency close to 99%. In contrast, incoming LCP waves are almost completely blocked. It is worth noting that the RCP and LCP components can be separated from arbitrarily polarized waves, so the polarization modes of the transmitted components are both RCP when the proposed design under linearly and elliptically polarized illuminations, respectively. Therefore, we present the modified polarization conversion efficiency curve in Fig. 1(b). It can be clearly seen that, except for the incident mode that is consistent with the desired polarization state, the arbitrary polarized waves are converted into RCP waves as the main component. Therefore, under the incidence of unpolarized waves, periodically arranged meta-molecules mainly generate RCP waves, which is consistent with the results derived using the Jones matrix.

Note 3: Jones matrix analysis of arbitrary-to-LCP polarization conversion

On the other hand, we can assume that the phase difference between both nanofins along the fast axis is q_1-q_2=π/2, and the relative rotation angle is $\Delta\varphi=\varphi_1-\varphi_2=\pi/4$. Eq. 3 can be simplified as,

$$T_{total}^{circular} = e^{i(q_1+\pi)}\begin{bmatrix} 0 & e^{-i2\varphi_1} \\ 0 & 0 \end{bmatrix} \tag{S7}$$

In this state, the proposed metasurface can transmit the THz wave for RCP illumination, while suppressing the LCP incident wave. In other words, the proposed design can be directly operated with unpolarized beams and efficiently converted to LCP beam manipulation without additional polarization equipment,

$$T_{total}^{circular} = e^{i(q_1+\pi-2\varphi_1)}\begin{bmatrix} i & -1 \\ -1 & -i \end{bmatrix} \tag{S8}$$

Thus,

$$E_{out} = e^{i\left(q_1-2\varphi_1+\psi+\frac{3}{2}\pi\right)}\left(\cos\chi+\sin\chi\right)\begin{bmatrix} 1 \\ i \end{bmatrix} = e^{ic_2}\left(\cos\chi+\sin\chi\right)\begin{bmatrix} 1 \\ i \end{bmatrix}$$

$$= \sqrt{2}e^{ic_2}\sin\left(\chi+\frac{\pi}{4}\right)\begin{bmatrix} 1 \\ i \end{bmatrix} \tag{S9}$$

Without loss of generality, that is, when the converted polarization is LCP mode, in the process of the arbitrary polarization state α gradually moving from the equator to the North Pole (RCP) along any meridian on the surface of the Poincaré Sphere, the conversion efficiency of the obtained LCP wave in transmission mode increases monotonically. When $\alpha(2\psi, 2\chi)$ moves to the north pole on the Poincaré Sphere (representing the RCP mode), the conversion efficiency reaches a maximum value. In other words, the proposed design has an excellent circular dichroism (CD) response at the operating frequency. However, its orthogonal state, defined as $\beta(-2\psi, 2(\chi-\pi/2))$, has a monotonically decreasing transition strength during the aforementioned process, as shown in Fig. 2(b). Also, when β moves to the South Pole on the Poincaré Sphere (representing the LCP mode), the conversion intensity reaches a theoretically minimum value of zero. In other words, the proposed design can directly serve as a candidate for a circularly polarized wave generator.

Note 4: Experimental setup for CD spectrum

In the experimental steps, four linear polarizers were placed in a typical THz time-domain spectroscopy system, named P1, P2, P3 and P4, respectively, to form a THz polarization test system. Among them, polarizers P1 and P3 are placed on the side close to the transmitter and receiver, respectively, to ensure that the polarization states of the transmitted and received waves remain unchanged during the measurement process. In addition, P2 and P4 are rotated by 45 degrees (along the propagation direction) as the measured *x*- and *y*-axes, respectively, as shown in Fig. 2(a). After observation, the initial angles of the test system were 10° (P2) and 164° (P4). First, using the experimental test system shown in Fig. 2, background noise was obtained. Secondly, after Fourier transform and data processing, four linear polarization components are obtained, namely t_{xx} (55°, 209°), t_{yx} (325°, 209°), t_{xy} (55°, 119°) and t_{yy} (325°, 119°). Finally, according to Eq. 10, the transmission coefficient under the circularly polarized basis and the corresponding CD spectrum are observed.

Note 5: Sample fabrication

Based on the parameters of the desired meta-atom picked from the geometric library, target samples were generated using MATLAB code. The actual fabrication steps are as follows: deep silicon etch using UV lithography and inductively coupled plasma (ICP) to form the desired array on 500 μm thick high resistance silicon. A 6.8 μm thick patterned positive photoresist (AZ4620) was obtained by UV lithography as a mask. Each ellipse-shaped metaatom was then obtained using ICP deep silicon etching (STS Multiplex ASE-HRM ICPetcher, UK). Finally, the remaining photoresist is washed away to obtain the final sample. The effective etching area of the sample is 1.2 cm × 1.2 cm. It can be seen from Fig. S2 that the sidewalls of the meta-atoms arranged through the above-mentioned process are very steep, which means the processing error is within permission.

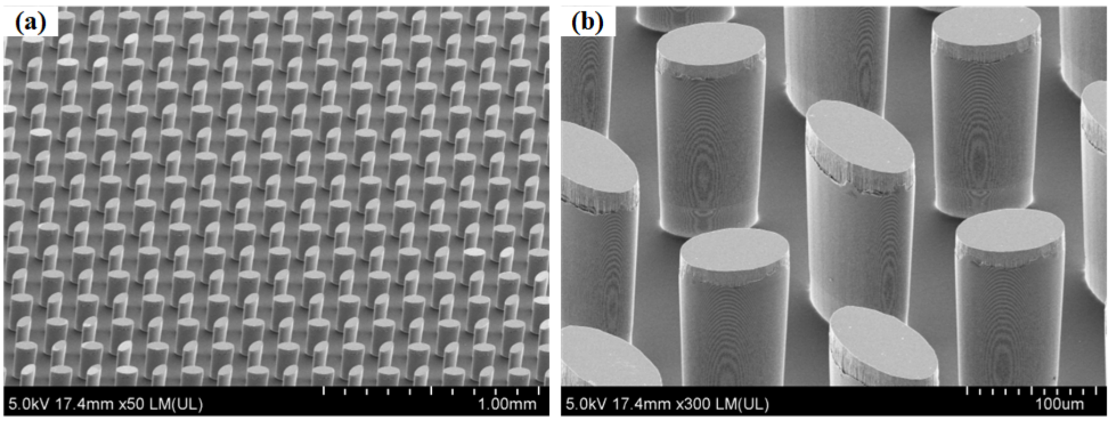

Fig. S2. SEM images of samples at different resolutions (a) 1 mm (b) 100 μm.

Note 6: Origin of the polarization ellipse

From the fluctuation formula of plane simple harmonic electromagnetic waves, the two transverse orthogonal electric field components of the instantaneous optical field propagating along the *z*-axis direction can be calculated as,

$$\begin{cases} E_x(z,t) = E_{0x}\cos(\tau + \delta_x) \\ E_y(z,t) = E_{0y}\cos(\tau + \delta_y) \end{cases} \tag{S10}$$

where $\tau = \omega t - kz$ is the propagation factor, ω denotes the angular frequency of the incident wave, t is the time, k is the number of waves, and z denotes the longitudinal propagation distance. The subscripts $i=x$, y denotes the components in the X and Y directions; E_{0x} and E_{0y} are the maximum amplitudes in the X and Y directions; δ_x and δ_y denote the initial phase responses of the optical field components, respectively. When the light field propagates along the Z-axis, the synthetic vectors of $E_x(z,t)$ and $E_y(z,t)$ form a series of trajectories of points in space, i.e., the spatial trajectories of the light field vectors. Decomposing the trigonometric functions in Eq. S1 yields,

$$\begin{cases} \dfrac{E_x}{E_{0x}} = \cos\tau\cos\delta_x - \sin\tau\sin\delta_x \\ \dfrac{E_y}{E_{0y}} = \cos\tau\cos\delta_y - \sin\tau\sin\delta_y \end{cases} \tag{S11}$$

Hence,

$$\begin{cases} \dfrac{E_x}{E_{0x}}\sin\delta_y - \dfrac{E_y}{E_{0y}}\sin\delta_x = \cos\tau\sin\left(\delta_y - \delta_x\right) \\ \dfrac{E_x}{E_{0x}}\cos\delta_y - \dfrac{E_y}{E_{0y}}\cos\delta_x = \sin\tau\sin\left(\delta_y - \delta_x\right) \end{cases} \tag{S12}$$

By squaring the upper and lower terms in Eq. S2 and then adding them together, the propagation factor τ can be eliminated, which can be further simplified as,

$$\begin{cases} \dfrac{E_x^2}{E_{0x}^2} + \dfrac{E_y^2}{E_{0y}^2} - 2\dfrac{E_x E_y}{E_{0x}E_{0y}}\cos\delta = \sin^2\delta \\ \delta = \delta_y - \delta_x \end{cases} \tag{S13}$$

According to Eq. S4, it can be obtained that the formula of the trajectory of the end of the synthetic polarization vector is an elliptic equation expressed in the cross-section perpendicular to the direction of light propagation. The trajectory of the end of the light field vector is an ellipse, and the ellipse is tangent to rectangles with side lengths $2E_{0x}$ and $2E_{0y}$, respectively, and the sides of the rectangles are parallel to the coordinate axes. The polarization ellipse of the light field characterizes the polarization state of the light field, i.e., the vectorial properties of the light field. We usually refer to the light field with elliptical trajectory at the end of the vector as elliptically polarized light. The superposition of two light waves with the same frequency, vibration direction perpendicular to each other, and a certain phase difference can generally be obtained elliptically polarized light. Further revising Eq. S4 can be obtained,

$$E_y = \frac{E_{0y}}{E_{0x}}E_x\cos\delta \pm \frac{E_{0y}}{E_{0x}}\sqrt{E_{0x}^2 - E_x^2}\sin\delta \tag{S14}$$

Obviously, the shape of the polarization ellipse depends on the amplitude ratio E_{0y}/E_{0x} and phase difference $\delta=\delta_y-\delta_x$ of the two orthogonal electric field components.

Note 7: Calculation steps of multipole decomposition.

The giant spin-selective BCD response originates from the interference effect between orthogonal circular polarization modes by introducing a pair of anisotropic meta-atoms with different phase profiles. Calculations by *COMSOL Multiphysics* show that electric dipole (ED), magnetic dipole (MD), ring dipole (TD), electric quadrupole (EQ), and magnetic quadrupole (MQ) are the scattering properties. The main contribution resonance. The modes that dominate the multipole decomposition of scattered power on the two circularly polarized channels are different due to the overlap of multiple resonant modes. It is worth mentioning that the powerful *COMSOL Multiphysics* post-processing templates facilitate the calculation of multipole decomposition responses of the metasurface arrays. The radiation power of induced multipoles can be calculated using the following formula,

$$I_{ED} = \frac{2\omega^4}{3c^3}\left|\vec{P}\right|^2 \tag{S15}$$

$$I_{MD} = \frac{2\omega^4}{3c^3}\left|\vec{M}\right|^2 \tag{S16}$$

$$I_{TD} = \frac{2\omega^6}{3c^5}\left|\vec{T}\right|^2 \tag{S17}$$

$$I_{Q^{(e)}} = \frac{\omega^6}{5c^5}\sum\left|Q_{\alpha,\beta}^{(e)}\right|^2 \tag{S18}$$

$$I_{Q^{(m)}} = \frac{\omega^6}{40c^5}\sum\left|Q_{\alpha,\beta}^{(m)}\right|^2 \tag{S19}$$

With the ED, MD, TD, EQ, and MQ moment,

$$\vec{P} = \frac{1}{i\omega}\int \vec{j}\, d^3r \tag{S20}$$

$$\vec{M} = \frac{1}{2c}\int\left(\vec{r}\times\vec{j}\right)d^3r \tag{S21}$$

$$\vec{T} = \frac{1}{10c}\int\left[\left(\vec{r}\cdot\vec{j}\right)\vec{r} - 2r^2\vec{j}\right]d^3r \tag{S22}$$

$$Q_{\alpha\beta}^{(e)} = \frac{1}{2i\omega}\int\left[r_{\alpha} j_{\beta} + r_{\beta\alpha} j_{\alpha} - \frac{2}{3}\left(\vec{r}\cdot\vec{j}\right)\delta_{\alpha,\beta}\right]d^3r \tag{S23}$$

$$Q_{\alpha\beta}^{(m)} = \frac{1}{3c}\int\left[\left(\vec{r}\times\vec{j}\right)_{\alpha} r_{\beta} + \left(\vec{r}\times\vec{j}\right)_{\beta} r_{\alpha}\right]d^3r \tag{S24}$$

Where j is the current density, ω is the circular frequency, r is the displacement vector, and c is the speed of light in free space. First, the written operators are saved in a txt file and subsequently added to the list of parameters defined by the component. Finally, another txt file containing the corresponding plotting expressions is added to the onedimensional drawing group.

Note 8: Circularly polarized transmission coefficients of meta-molecules at different rotation angles.

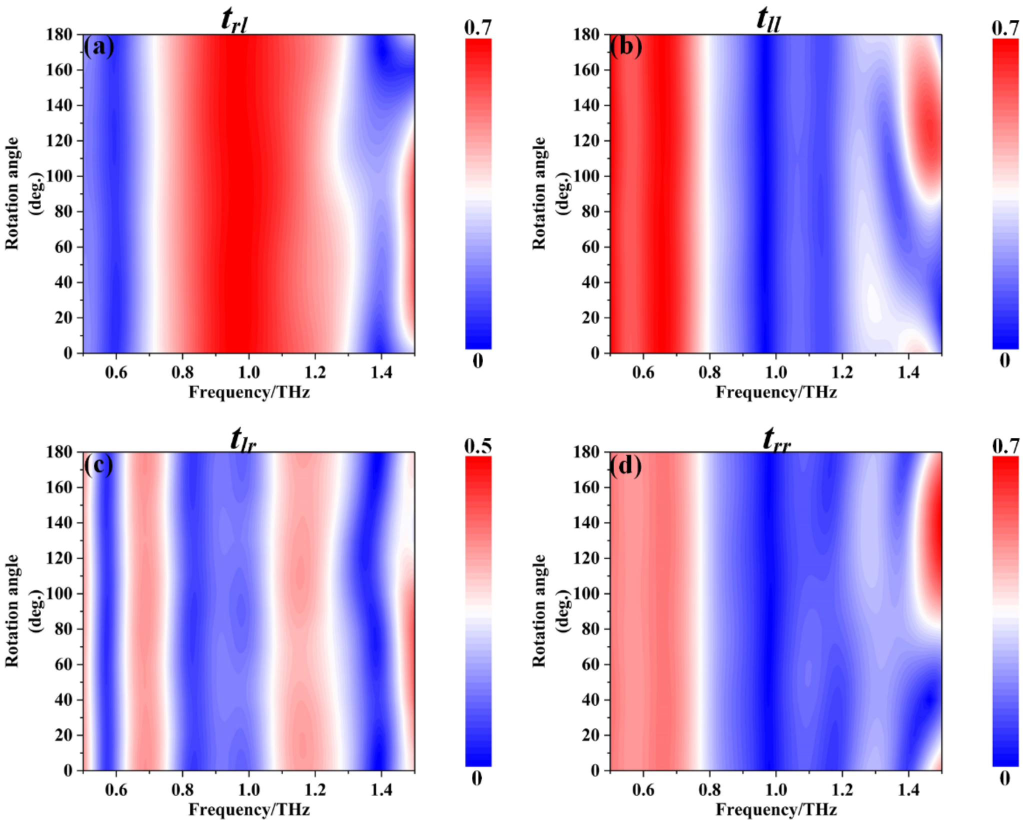

Fig. S3. The magnitude intensity distribution of the four circularly polarized components obtained by applying the geometric phase principle (a) t_{rl}, (b) t_{ll}, (c) t_{lr}, and (d) t_{rr}.