Supporting Information

Title: Spin-decoupled multifunctional metasurface for asymmetric polarization generation

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1. Controllable asymmetric deflection of the ASM

To show our ability in freely controlling the deflection angles of the two circularly polarized waves, another two ASMs are designed at 1.0 THz. One has theoretical phase profiles of P_{rl} = $2\pi x/3P$ and P_{lr} = 0 which deflects LCP output towards 41.81° while it does not deflect the RCP output. The corresponding overall period is $3P$. The other has phase profiles of P_{rl} = $2\pi x/6P$ and P_{lr} = $2\pi x/8P$ which deflects both LCP and RCP outputs towards positive angles at 19.84° and

14.85°, respectively. The corresponding overall period is 24*P*. Figures S1a,c illustrate the theoretical and simulated phase profiles, as well as the corresponding simulated amplitude profiles of the two ASMs, respectively. Here, to make the phase profiles easier to see, we add −2π to both P_{lr} of the two ASMs. The shaded region in Figure S1a represents one overall period of the ASM. Figures S1b,d illustrate the simulated angle-resolved RCP and LCP transmittance profiles of the two ASMs under LCP and RCP incidences, respectively. It is seen that all the simulated results are agreeing well with the theory, including the desired phase distributions and the deflection angles. The corresponding relative asymmetric deflection angles of the two ASMs are Δ = 20.91° and Δ = 17.35°, respectively.

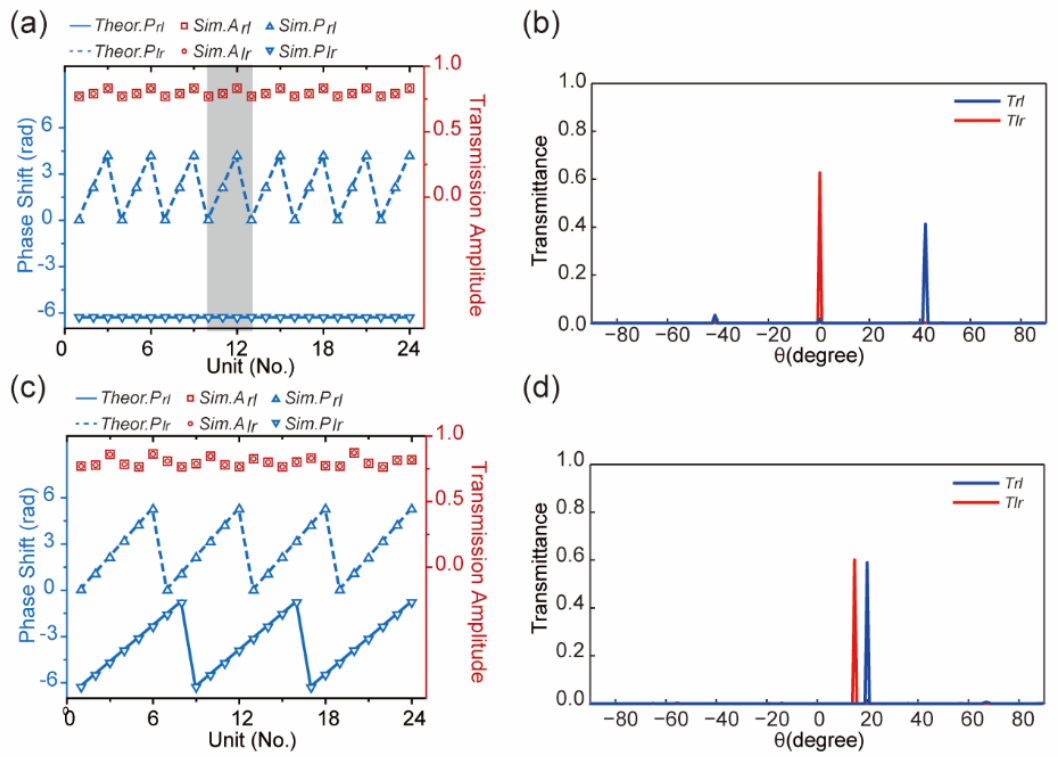

Figure S1. (a,c) Theoretical (solid and dash lines) and simulated (hollow triangles) phase profiles, as well as the corresponding simulated (hollow squares and circles) amplitude profiles of two ASMs for deflecting the RCP and LCP outputs towards positive angle and 0° angle (a), and two different positive angles (c), under the LCP and RCP incidences at 1.0 THz, respectively. The amplitudes are amplitudes of the transmission of the sole structured surface. (b,d) Simulated angle-resolved RCP (blue) and LCP (red) transmittance profiles of the sole structured surface of the ASMs in (a) and (c) under the LCP and RCP normal incidences at 1.0 THz, respectively.

2. Microscope images of the fabricated ASM and MASM

The whole results presented in the manuscript are based on two phase gradients for the two cross-circularly-polarized output waves, which are $2\pi/6P$ and $2\pi/8P$, respectively. Therefore, to realize both phase gradient in one ASM, the overall period should be $24P$, which corresponding to 24 unit cells and an overall length of 3.6 mm. As for the MASM, it is composed by interleaving two ASMs along the *y*-direction. The overall period is also $24P$, but contains 48 unit cells. The microscope image in Figure 3a and Figure 5a only schematically show part of the fabricated samples, where only 8 unit cells are included. Figure S2 illustrates the whole overall period of the fabricated ASM and MASM in a separate way.

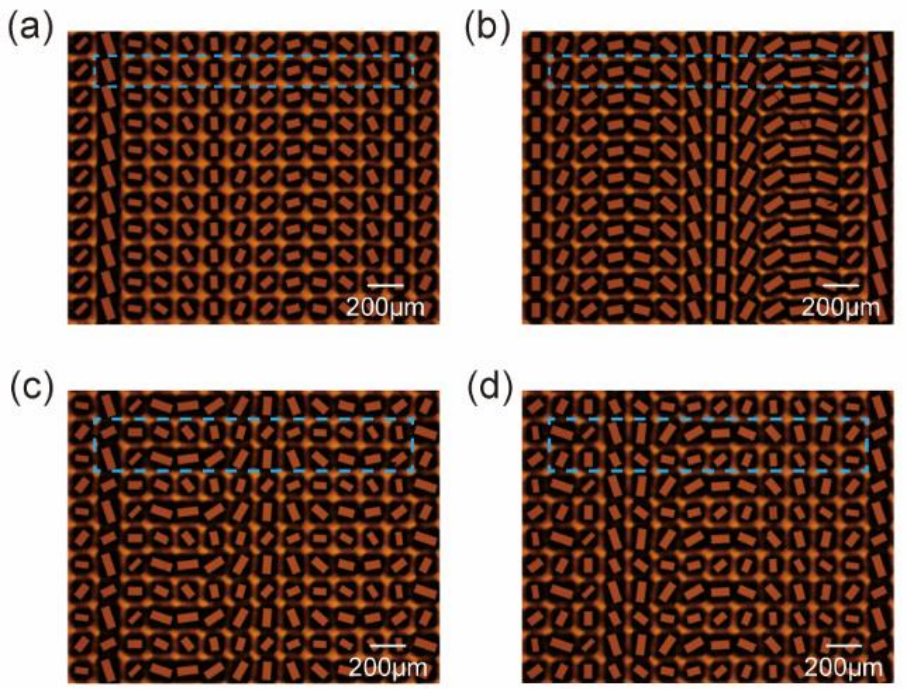

Figure S2. (a,b) Microscope images to show the overall period of the fabricated ASM. The blue dash boxes inset (a) and (b) contain the first 12 and the latter 12 unit cells of the overall period, respectively. (c,d) Microscope images to show the overall period of the fabricated MASM. The blue dash boxes inset (c) and (d) contain the first 24 and the latter 24 unit cells of the overall period, respectively.