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Article

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Planar chiral metasurfaces with maximal and tunable chiroptical response driven by bound states in the continuum

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Abstract:

Optical metasurfaces with high-Q chiral resonances can boost light-matter interaction for various applications of chiral response for ultrathin, active, and nonlinear metadevices. Usually, such metasurfaces require sophisticated depth-resolved nanofabrication to realize subwavelength stereo-nanostructures, posing overwhelming challenges, especially in the short-wavelength range. Here, we suggest a novel planarized design of chiral metasurfaces supporting bound states in the continuum (BICs), and demonstrate experimentally chiroptical responses with record-high Q-factors ($Q=390$) and near-perfect circular dichroism ($CD=0.93$) at optical frequencies. The symmetry-reduced meta-atoms are highly birefringent and support winding elliptical eigen-polarizations with opposite helicity surrounding the BIC polarization singularity, providing a convenient way for achieving maximal planar chirality tuned by either broken in-plane symmetry or changing illumination direction. Such sharply resonant chirality realized in planar metasurfaces promises various practical applications in classical and quantum optics including chiral sensing, enantiomer selection, and chiral quantum emitters.

Keywords: Planar chiral metasurfaces; Bound states in the continuum; Extreme optical chirality; Circular dichroism; Chirality selectivity

Introduction

Chirality refers to the property of an object that cannot be superimposed with its mirror image after rotations or translations¹⁻³. It is quite common in nature, e.g., in organic molecules⁴, quartz crystals⁵, and many others. The study of chirality is fundamentally important in various areas, including analytical chemistry, pharmaceuticals, and even searching for extraterrestrial life. Light interactions with these geometries can induce chiroptical effects, including circular dichroism (CD) and optical activity, manifested by a difference in intensity and phase responses between left/right circularly polarized (LCP/RCP) light illuminations. Optical chirality was first demonstrated in three-dimensional (3D) photonic structures⁶, such as the helices^{6,7}, twisted cross structures^{8,9} and multi-layered structures¹⁰⁻¹², all exhibiting strong CD and optical activity with preserved circular polarization, which however require very demanding 3D nanofabrication techniques. Recently, two-dimensional (2D) or planar structures have been shown to support both intrinsic planar chirality at normal incidence¹³⁻¹⁶ and extrinsic chirality at oblique incidences¹⁷⁻²¹. Being different from 3D chirality with broken mirror symmetry in the propagation direction, although even existing in some stacked structures¹⁵, planar chirality shows a circular polarization conversion between the output and incident light, and the cross-polarization behavior can be further exploited to modulate the geometric phase for arbitrary chiral wavefront shaping²²⁻²⁶. However, based on existing approaches it is challenging to achieve maximal planar chirality combined with ultra-high quality (Q-) factor due to absorption and scattering loss, hindering many applications that rely on strong chiral light-matter interactions.

Importantly, bound states in the continuum (BICs) can provide a feasible solution for the outstanding problems of chiral photonics. BICs are identified as localized states coexisting with extended modes within the light cone, and they have attracted tremendous attention due to their unbounded Q-factors, which may boost light-matter interactions for applications such as surface-emitting lasing²⁷⁻²⁹, biomedical sensors³⁰⁻³², and nonlinear frequency converters³³⁻³⁸. BICs originate from destructive interference of several radiative channels, and hence they cannot be accessed externally. Quasi-BICs (q-BICs) with high-Q resonances are tailored perturbing the symmetry to couple out the resonant mode to free-space radiation³⁹⁻⁴⁹. Recently, chiroptical nanostructures mediated by BICs have been proposed, demonstrating perfect unitary chirality and extremely high Q-factors^{50,51}, and greatly expanding the available platforms to achieve optical chirality⁵¹⁻⁵⁶. However, such chiral q-BIC scheme has only been proposed by complicated stereo-nanostructures, making elusive an experimental realization and application development, especially at optical frequencies.

Here, we suggest and realize experimentally a novel design of planar chiral metasurfaces at optical frequencies with simultaneously mediated maximal chirality and ultrahigh Q-factors. A double-sided scythe (DSS-) shaped α -Si inclusion with in-plane inversion C_2 symmetry but without in-plane mirror symmetry is employed to construct the BIC state. Such BIC state with strong birefringence supports a vortex polarization singularity (V point) surrounded by elliptical eigenstate polarizations with nonvanishing helicity, providing a convenient way to achieve maximum and

high-Q chirality by slightly perturbing the inversion symmetry. With either breaking in-plane inversion symmetry or varying illumination geometry, we realize planar chiral q-BIC states with extremely strong intrinsic or extrinsic chirality, exceeding $CD=0.99$ (in theory) and $CD=0.93$ (in experiment). For the intrinsic chirality, the Q-factor of the CD spectra has an inverse quadratic relation with respect to the geometrical asymmetry, with a nearly unchanged CD peak. For the extrinsic chirality, both value and sign of CD can be flexibly tuned by changing the incident angles encircling the Γ point, which is connected with the intrinsic nonvanishing helicities of the eigenpolarizations near the BIC singularity⁵⁷. We believe that the demonstrated planar chiral metasurfaces governed by the BIC physics may find many applications in chiral sensing of biomolecules, spin-selective emitters, and active chiroptical devices.

Results and Discussion

Previous q-BIC responses in metasurfaces usually did not support polarization effects^{28,43,44} (Fig. 1a) or only supported linear polarization selectivity^{30,39,41}. Although very recently q-BICs with versatile polarization responses (chirality selectivity) were theoretically proposed, they required complicated 3D structures with broken symmetry in the propagation direction^{50,51,58} (Fig. 1b), hindering their implementation within current 2D patterning technology, especially at optical frequencies. Our present work shows that planar metasurfaces with only reduced in-plane symmetry can support q-BICs with both extrinsic and intrinsic planar chirality (Fig. 1c). Figure 1d shows the geometry of interest, a double-sided scythe (DSS) α -Si structure (refractive indexes refer to Supplementary Fig. S2) employed as the unit cell of our proposed planar metasurface. Such DSS structure ($W_1=W_2$ and $L_1=L_2$, but $L_1 \neq W_1$) has in-plane inversion (C_2) symmetry, but lacks any in-plane mirror symmetries. It hosts a BIC at the Γ point, characterized by a vertical magnetic dipole (MD) mode as shown in Figs. 1(e and f). Different from the Γ point BIC state supported by highly symmetric inclusions⁵⁷, which is accompanied by a polarization singularity surrounded by vortex linear polarizations, the BIC state of the DSS structure supports a polarization singularity enclosed by a vortex elliptical eigen-polarization with non-vanishing helicity (see the inset of Fig. 1f and Supplementary Fig. S3). The planar chirality behavior can be predicted by temporal coupled mode theory (TCMT) (Supplementary Notes I & II), which suggests that a perfect planar chiral response can be obtained by tuning the local response to be birefringent using a geometry that has an inversion center, and then breaking inversion symmetry with a suitable perturbation to yield an intrinsic planar chiral q-BIC. Our proposed DSS inclusion with reduced symmetry provides a smart perturbation degree of freedom along the 45° direction, capable of achieving both maximal extrinsic and intrinsic chirality by perturbing a single parameter. As a general remark, in our systems the sign of the CD is determined by the sign of the helicity, while the maximum of CD does not necessarily coincide with circular eigenpolarization with the highest helicity, because there is non-zero background scattering accumulated by Jones matrix elements (see Supplementary Notes I and Supplementary Fig. S4).

Based on such BIC state supported by DSS inclusion with reduced symmetry, one can expect both extrinsic and intrinsic planar chirality by slightly breaking the BIC symmetry. Figures 2(a-d)

show the q-BIC with near-unity extrinsic chirality achieved by illumination symmetry breaking (varying incident angle θ and conical angle φ). Here, the CD is defined as the transmittance difference under right-handed polarization (RCP) and left-handed polarization (LCP) incidence:

$$CD = \frac{(T_{rr}+T_{lr})-(T_{rl}+T_{ll})}{(T_{rr}+T_{lr})+(T_{rl}+T_{ll})} \quad (1)$$

where $T_{ij} = |t_{ij}|^2$ ($i=r, l; j=r, l$; r represents RCP, l represents LCP) is the transmittance of output polarization i from the input polarization j , and all element of t_{ij} construct the Jones matrix under circular polarization basis^{11,59}

$$J_{circ} = \begin{pmatrix} t_{rr} & t_{rl} \\ t_{lr} & t_{ll} \end{pmatrix}. \quad (2)$$

Figure 2b shows the calculated transmission spectra of all Jones matrix elements and the CD spectrum of the DSS metasurface with C_2 symmetry ($W_1=W_2$ and $L_1=L_2$, but $L_1 \neq W_1$) at oblique incidence ($\theta=8^\circ$, $\varphi=90^\circ$). T_{lr} has a sharp peak greater than 0.9, while all the other three Jones matrix elements (T_{rl} , T_{ll} and T_{rr}) exhibit dips close to 0 at the resonant wavelength (1456 nm) of the q-BIC, resulting in an ultra-sharp CD spectrum with a maximum greater than 0.95 (green curves in Fig. 2b). Figure 2c shows the evolution of CD spectra by continuously varying the incident angle θ along the y -direction ($\varphi=90^\circ$) (evolutions of transmission spectra of all Jones matrix elements are shown in Supplementary Fig. S5). The linewidth of the CD spectrum increases from 0 to finite values as θ increases, and the peak CD sustains near-unitary value with finite linewidths, manifesting the transfer process from a non-radiative BIC to a high-Q radiative q-BIC with strong extrinsic chirality. The Q-factor of the extrinsic chiral q-BIC shows the expected inverse quadratic law with the illumination asymmetry parameter $\alpha_I=\sin\theta$ (Fig. 2d), which provides a convenient way to tailor the Q-factor of the chiral response. The evolutions of the extrinsic chiral q-BIC along the x -direction ($\varphi=0^\circ$) are shown in Supplementary Fig. S6, the variation trend is the same as the $\varphi=90^\circ$ case, except that the sign of CD is flipped due to the opposite helicity of eigen-polarization supported by the DSS structure along x - and y -directions (the inset of Fig. 1f). To investigate the full CD flip picture, the evolution of CD spectra with varying conical angle φ at a fixed incident angle $\theta=12^\circ$ is shown in Supplementary Fig. S7. The opposite varying trend of T_{lr} and T_{rl} can be observed as φ is varying, leading to the sign flip of CD at positions near $(2n-1) \times 45^\circ$, ($n=1,2,3,4$), which is consistent with helicity sign flip shown in the inset of Fig. 1f.

Beyond the extrinsic chiral behavior, we can also access intrinsic high-Q chiral behavior under normal incidence. This can be achieved by introducing an in-plane geometrical asymmetry parameter ($\delta=W_2-W_1$) as shown in Fig. 2e. As the DSS structure supports strong birefringence along 45° , an in-plane perturbation that breaks inversion symmetry will provide a degree of freedom to obtain planar chirality, as TCMT suggests in supplementary Note II. Figure 2f depicts the transmission spectra of all Jones Matrix elements and the CD spectra of the metasurface with $\delta=40$ nm ($W_1=191$ nm, $W_2=231$ nm). The ultra-sharp resonant peak of T_{lr} and resonant dips of T_{rl} , T_{ll} and T_{rr} are obtained at the wavelength of 1392 nm, resulting in a CD peak greater than 0.99 and an ultra-narrow linewidth of 1.45 nm. We also confirm the planar chiral q-BIC modes by utilizing multipole

expansions and studying the nearfield electromagnetic patterns of the modes (Supplementary Fig. S8), which further confirm that the high-Q planar chiral q-BIC states are dominated by the MD mode. Figures 2(g-h) show the CD evolution spectra as a function of the asymmetry parameter δ , the Q-factor of the CD spectrum also manifests an inverse quadratic relationship against the relative asymmetry parameter defined by $\alpha_2=\delta/W_1$, which is the signature of symmetry-protected BICs (corresponding spectra for all components of the Jones matrix spectra refer to Supplementary Fig. S9). Different from the intrinsic 3D chirality with preserved helicity between input and output polarizations, the intrinsic planar chirality is always accompanied by circular conversion dichroism (CCD) and asymmetry transmission (AT)^{2,13,60}, as shown in Supplementary Fig. S10, due to the preserved symmetry in the propagation direction. The CCD is produced by the mutual orientation of chiral elements and light transmission direction, and the peak value of CCD is close to 1, which is slightly larger than CD (Supplementary Fig. S11). Perfect unitary circular conversion transmittance T_{lr} and CD can be achieved with a strictly symmetric background as shown in Supplementary Fig. S12. The existence of a practical SiO₂ substrate introduces a perturbation of the background scattering, resulting in reduced transmittance and CD, as presented in Figs. 2(b and f).

Due to the symmetry-protected origin of this phenomenon, our designed DSS metasurface has good tolerance against fabrication imperfections, as tested in Supplementary Fig. S13, which provides a convenient way for experimental realization. We fabricated the DSS metasurfaces with different in-plane asymmetry parameters ($\delta=0, 20, 40, 60$, and 80 nm), and employed a homemade optical setup (Fig. 3a) to measure the Jones matrix elements of the metasurface with a rotating stage. We first examine the extrinsic chiral q-BIC by measuring the DSS metasurface without in-plane geometry asymmetry ($\delta=0$) for varying incident angles (Fig. 3b). Figure 3c shows the comparison between simulated and measured transmission Jones matrix spectra at different incident angles ($\theta=0^\circ, \pm 4^\circ, \pm 8^\circ, \pm 12^\circ$) along the $\varphi=90^\circ$ direction. At normal incidence (middle panel of Fig. 3c), all transmission spectra are smooth without resonant features, because the BIC state at the Γ point is decoupled from any scattering channels of the external environment. At other incident angles, sharp circular conversion peaks of T_{lr} and resonant dips of other three Jones matrix elements (T_{ll}, T_{rl}, T_{rr}) emerge in both simulations and experiments. The measured absolute peak transmittances are greater than 0.78 for all incident angles, which is a little below the simulated 0.9, mainly due to the practical scattering loss of the fabricated sample as well as a limited collection efficiency of the measurement setup. As expected, we can clearly observe the linewidth increment of the q-BIC peak with increasing incident angle in the experiment, consistent with our theory and simulations. From the measured Jones matrix spectra, we extracted the CD spectra for different incident angles corresponding to Eq. (1) in Fig. 3d. As we can see, sharp CD peaks appear at the q-BIC resonant wavelengths, the simulated/measured CD maxima reach 0.8/0.72, 0.95/0.82, and 0.96/0.88 with the simulated/measured Q-factors of 616/258, 143/96, and 58/39 for incident angles $\theta=\pm 4^\circ, \pm 8^\circ, \pm 12^\circ$, respectively, while for normal incidence, the CD is always 0 as the BIC state is not accessed by external excitations. We also experimentally characterized the extrinsic chiral q-BIC along the $\varphi=0^\circ$ direction in supplementary Fig. S14 where sharp transmission peaks of T_{rl} are observed, achieving

opposite signs of simulated/measured CD: -0.89/-0.82, -0.95/-0.88, and -0.97/-0.91 with the simulated/measured Q-factors of 798/312, 196/102, 85/55 at incident angles $\theta=\pm 4^\circ$, $\pm 8^\circ$, $\pm 12^\circ$, respectively, which further confirms the CD flip behavior.

To further experimentally verify the intrinsic chiral q-BIC, we characterized the DSS metasurfaces with different in-plane geometrical asymmetries, as shown in Fig. 4. Figure 4a shows the scanning electron microscope (SEM) images of fabricated samples with different asymmetry parameters δ . From both the top view and slant view, we see that the fabricated DSS silicon pillars are smooth and uniform, in agreement with the theoretical design. Figures 4(b and c) show the simulated and measured transmission Jones matrix and CD spectra of the five fabricated metasurfaces. When $\delta=0$ (lowest panel in Figs. 4b and 4c), there is no resonance with $T_{rl}=T_{lr}$ and $T_{ll}=T_{rr}$ due to reciprocity, and CD=0 crossing the whole frequency band. As the asymmetry parameter δ became non-zero, sharp circular conversion transmission peaks of T_{lr} and CD appear, and the bandwidth increases with the increase of the asymmetry parameter (upper four panels in Figs. 4b and 4c). The simulated/measured CD maxima for the intrinsic planar chirality are 0.98/0.88, 0.99/0.93, 0.99/0.92, and 0.99/0.91 with the simulated/measured Q-factors of 2457/390, 480/121, 198/62, and 128/38 for $\delta=20, 40, 60$, and 80 nm, respectively. Such simultaneously achieved strong chirality and high Q-factors in experiment promise chiroptical functionalities with strong light-matter interactions.

Conclusions

We have proposed and experimentally realized planar chiral metasurfaces with in-plane symmetry-reduced inclusions. Such metasurfaces are governed by the BIC physics with birefringence degrees of freedom, identified as a vortex polarization singularity surrounded by ellipse eigenstate polarizations with non-vanishing helicity. Both extrinsic and intrinsic planar chirality can be realized by symmetry breaking. By illumination symmetry breaking, the BIC state can be transferred to q-BIC with strong extrinsic chirality accompanied by tunable linewidth and signs. By introducing in-plane structure asymmetry, the intrinsic planar chirality can be achieved under normal incidence with maximum CD of 0.99 (theory) and 0.93 (experiment) and Q-factors exceeding 390 (experiment) at optical frequencies. Beyond the chirality based on circular polarizations, we can achieve any elliptical chirality by varying simultaneously the degree of birefringence and the axis angle of the anisotropic inclusion, which suggests a way to spark new research works. Due to the high Q-factor of the planar chiral q-BIC with its accessibility and controllability, our results provide unique opportunities for many applications requiring ultrahigh Q-factors and chirality control, including circular polarization emitter, arbitrary wavefront lasing, and chiral nonlinear optics.

Methods

Simulation of reflection and transmission efficiency: Rigorous coupled wave analysis (S⁴)⁶¹ simulations were carried out to simulate the planar chiral q-BIC metasurface and to calculate

efficiencies for the co-polarization handedness-preserved coefficients, and the circular cross-polarization conversion coefficients transmission spectra. The metasurfaces are designed by α -Si columns based on a SiO_2 substrate.

Fabrication of samples: The metasurfaces were fabricated on a fused quartz substrate by utilizing electron beam lithography and including the processes such as deposition, patterning, lift-off and etching. First, a 350-nm-thick amorphous silicon (α -Si) film was deposited by plasma-enhanced chemical vapor deposition (PECVD) method at 120 °C, during which the flow rates of SiH_4 and Ar are 10 and 475 sccm. And the deposition pressure and RF power are 650 mTorr and 20 W, respectively. Then a PMMA film of 300 nm was spinning coated and covered by PEDOT: PSS film as a conducting layer. The desired structure was patterned by utilizing JEOL 6300FS EBL at a base dose of 1000 $\mu\text{C}/\text{cm}^2$ with an accelerating voltage of 100 kV for 1 hour. After the exposure process, the conducting layer was washed away and the resist was developed in 1:3 MIBK: IPA solution for 40 s and rinsed in IPA for 30 s successively, followed by a deposition of 80 nm Cr using electron beam evaporation deposition (EBD) method. To realize the lift-off process, the sample was immersed in hot acetone of 75 °C and cleaned by ultrasonic. Finally, by using inductively coupled plasma (ICP) reactive ion etching (RIE) method with HBr at room temperature (RT) for 260 s (flow rate of 50 sccm, the pressure of 10 mTorr, RF and ICP power of 50 and 750 W, respectively), the desired structure was transferred from Cr to silicon and the residual Cr was removed by cerium (IV) ammonium nitrate. The five planar chiral q-BIC metasurfaces are composed of 500×500 periods (425 μm ×425 μm) with different cross-sections ($\delta = 0$ nm, 20 nm, 40 nm, 60 nm, and 80 nm), and the final structures are shown in Fig. 4a.

Characterization of samples: A supercontinuum laser (Fianium-WL-SC480) was employed as the broadband light source for the measurement of the transmission spectra of all polarization components under circular polarization incidences. Incident light with a circular polarization was generated by cascading a broadband polarizer and a quarter waveplate from the supercontinuum laser. Then, the incident light was focused on the sample by a lens with a focal length of 5 cm. Subsequently, the polarization components are detected by a quarter waveplate and a polarizer. The transmittances were measured by means of an Ocean spectrometer (flame-NIR). Moreover, the sample was mounted on a rotation stage for varying incident angles in the measurement.

Author Contributions

Z.-L.D. and T.S. conceived the idea. T.S., Z.-L.D. and X.L. designed the experiments. T.S., Z.-L.D. carried out the design and simulation of the metasurface. T.S., Z.-L.D., G. H. A.O and Y. Z conducted the theoretical analysis of the results. G.G. and J.L. fabricated the samples. T.S. performed the measurements. Z.-L.D., Y.K., A.A. and X.L. supervised the project. Z.-L.D., T.S., and X.L. analyzed the data and wrote the manuscript. All authors contributed to the revision and discussion of the manuscript.

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Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Conflict of Interest

The authors declare no conflict of interest.

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Figure 1. Schematics of different BIC states and planar chiral q-BIC metasurface with a polarization singularity enclosed by vortex elliptical eigen-polarizations with non-vanishing helicities. (a-c) Comparison of q-BICs (a) in planar metasurfaces without polarization effect, (b) in stereo-nanostructures with 3D chiral effect in reflection, and (c) in planar metasurfaces with 2D chiral effect in transmission. (d) Schematic of the unit cell of the planar chiral q-BIC metasurface formed by the amorphous silicon (α -Si) DSS structure on a SiO_2 substrate with parameters $P=850$ nm, $R=280$ nm, $L_1=L_2=220$ nm, $W_1=W_2=191$ nm, and $H=350$ nm. (e) Band structure with real parts of the eigen-wavelength of the DSS metasurface near the Γ point. Insets show the field patterns (color: magnetic distributions, arrow: electric vectors) of the three typical modes. (f) Q-factors of the eigenmode in the k -space in the vicinity of the Γ point. Insets show the first Brillouin zone and eigen-polarization profiles. On the eigen-polarization profiles, the polarization states are represented by ellipses of which the red and blue represent the eigen left-handed states and right-handed states, respectively, and the black dot represents the V point (BIC).

Figure 2. Proposed implementation of the DSS structure with both the extrinsic and intrinsic planar chirality by transferring the BIC to q-BICs. (a, e) Schematic of the symmetry-breaking processes that transfer the BIC to planar chiral q-BICs: (a) illumination symmetry breaking by varying incident angle θ , (e) in-plane geometry symmetry breaking by $\delta = W_2 - W_1$. (b, f) Simulated transmission Jones matrix spectra of T_{il} , T_{rr} , T_{rl} and T_{lr} as well as the CD spectrum of the metasurface with (b) the same parameters as Fig. 1d ($\delta = 0$ nm) under oblique incidence ($\theta = 8^\circ$, $\varphi = 90^\circ$) and with (f) an asymmetric structure parameter ($\delta = 40$ nm) for normal incidences. (c, g) The evolution of CD spectra by continuous varying (c) incident angle θ along the $\varphi = 90^\circ$ direction and by (g) the geometry asymmetry parameter δ . (d, h) Dependence of the Q-factors of the planar chiral q-BIC mode on the relative asymmetry parameter (d) $\alpha_1 = \sin\theta$, (h) $\alpha_2 = \delta/W_1$ around the chiral q-BIC state. The solid line shows an inverse quadratic fitting.

Figure 3. Experimental verification of the extrinsic planar chiral q-BIC with illumination symmetry breaking. (a) Experimental setup for the Jones matrix spectra measurement under the circular polarization basis. P₁ and P₂ represent the polarizers, QWP is the quarter-wave plate, L₁, L₂, and L₃ are lenses. (b) Schematic of the metasurface with $\delta=0$ supporting chiral q-BICs through different incident angles along the $\phi=90^\circ$ direction. (c) Simulated and measured transmission Jones matrix spectra (T_{lr} , T_{rl} , T_{rr} and T_{ll}) for different incident angles ($\theta=0^\circ, \pm 4^\circ, \pm 8^\circ, \pm 12^\circ$) along the $\phi=90^\circ$ direction. (d) Simulated and measured CD spectra for different incident angles extracted from the Jones matrix spectra in (c).

Figure 4. Experimental verification of the intrinsic planar chiral q-BIC with various in-plane geometry asymmetry parameters. (a) The top view of the SEM image of five fabricated DSS metasurfaces with asymmetry parameters $\delta=0$ nm, 20 nm, 40 nm, 60 nm, and 80 nm, respectively. The inset shows the slant view of the metasurface. Scale bar: 1 μ m. (b) Simulated and measured transmission Jones matrix spectra (T_{lr} , T_{rl} , T_{rr} and T_{ll}) of the five DSS metasurface samples with different δ . (c) Simulated and measured CD spectra for different δ extracted from the Jones matrix spectra in (b).

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