

Supplementary Information for Observation of Maximum Circular Dichroism in Twisted Hyperbolic van der Waals Homostructures

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Section 1. LB and LD in bound and free electron based systems

The permittivity tensor for a uniaxial anisotropic material can be written as $\begin{bmatrix} \varepsilon_x & & \\ & \varepsilon_y & \\ & & \varepsilon_z \end{bmatrix}$, and the dispersion equation for electromagnetic waves is written as (1):

$$\frac{k_{\parallel}^2}{\varepsilon_z} + \frac{k_z^2}{\varepsilon_{\parallel}} = \frac{\omega^2}{c^2}, \quad (\text{S1})$$

where $k_{\parallel}^2 = k_x^2 + k_y^2$, $\varepsilon_{\parallel}$ stands for ε_x or ε_y , ω is the angular frequency and c is the vacuum light speed. For the normal direction ($k_x = k_y = 0$), the wavenumber is $k_z = \frac{\omega}{c} \sqrt{\varepsilon_{\parallel}}$. The electromagnetic wave is propagating when $\varepsilon_{\parallel} > 0$ and evanescent when $\varepsilon_{\parallel} < 0$. The linear birefringence LB, defined as the difference in phase retardance, can be calculated as:

$$\text{LB} = |\text{real}(\Delta k_z)| \cdot d = k_0 d \cdot |\text{real}(\sqrt{\varepsilon_x} - \sqrt{\varepsilon_y})|, \quad (\text{S2})$$

where d is the sample thickness. The linear dichroism LD, defined as the differential attenuation during propagation, can be calculated as:

$$\text{LD} = |\text{imag}(\Delta k_z)| \cdot d = k_0 d \cdot |\text{imag}(\sqrt{\varepsilon_x} - \sqrt{\varepsilon_y})|. \quad (\text{S3})$$

The dielectric functions of ε_x and ε_y in bound-electron based dielectrics and free-electron based metals can be described by the Drude-Lorentz Model (2):

$$\varepsilon_{\text{bound}}(\omega) = B(\omega_1, \omega_2, \dots, \omega_N; \omega) = \varepsilon_{\infty} + \sum_{i=0}^N \frac{\omega_{p,i}^2}{\omega_i^2 - \omega^2 - i\gamma_i \omega} \quad (\text{S4.1})$$

$$\varepsilon_{\text{free}}(\omega) = F(\omega_{p,0}; \omega) = \varepsilon_{\infty} - \frac{\omega_{p,0}^2}{\omega^2 + i\gamma_0 \omega} \quad (\text{S4.2})$$

To examine the upper bound of permittivity for the two cases, the absolute values are calculated,

$$|\varepsilon_{\text{bound}}(\omega)| = \varepsilon_{\infty} + \left| \sum_{i=0}^N \frac{\omega_{p,i}^2}{\omega_i^2 - \omega^2 - i\gamma_i \omega} \right| = \varepsilon_{\infty} + \sum_{i=0}^N \frac{\omega_{p,i}^2}{\sqrt{(\omega_i^2 - \omega^2)^2 + \gamma_i^2 \omega^2}}, \quad (\text{S5.1})$$

$$|\varepsilon_{\text{free}}(\omega)| = \left| \varepsilon_{\infty} - \frac{\omega_{p,0}^2}{\omega\sqrt{\omega^2 + \gamma_0^2}} \right|. \quad (\text{S5.2})$$

It is clear that $|\varepsilon_{\text{bound}}(\omega)| < \infty$ for any ω and has an upper limit. As for free electrons, the term $1/\omega$ makes $|\varepsilon_{\text{free}}(\omega)|$ divergent for low frequencies. Considering equations S2 and S3, we can infer that both LB and LD for bound-electron based dielectrics have upper limits. For free-electron based metals, LB and LD can be divergent for low frequencies.

As shown in Fig. S6, we have made a comparison of $\text{LB} \cdot \text{LD}$ between a birefringent crystal and a metallic-like hyperbolic material. In conventional birefringent crystals, the anisotropy originates from unequal binding of electrons along different lattice axes (Fig. S6a), leading to anisotropic resonance frequencies and damping rates of their harmonic oscillators, as described by the Lorentz model (Fig. S6b). Both principal axes possess positive permittivity, thus supporting propagating eigenmodes, each with its own phase velocity and attenuation constant. The differences between the real and imaginary parts of refractive indices of the two propagating modes yield the LB and LD. However, the Kramers–Kronig (KK) relationship (3,4) inherently enforces causality between the real and imaginary parts of its dielectric response along each axis (Fig. S6b), and thus prevents the simultaneous enhancement of LD and LB at a single frequency. Meanwhile, since the polarizability in the Lorentz model is inherently bounded due to intrinsic losses in harmonic oscillators, the magnitudes of LB and LD face an upper limit. Consequently, both LB and LD remain small, typically confined to a narrow spectral band near the dipolar transitions (Fig. S6c). To obtain substantial LB or LD, one must rely on large crystal thicknesses.

In hyperbolic systems, the plasma frequencies ω_p of intrinsic electron collective oscillations can differ significantly between crystallographic axes (Fig. S6e), directly leading to permittivity components of opposite signs ($\varepsilon_x \cdot \varepsilon_y < 0$). In such a hyperbolic region (shaded area in Fig. S6e), two distinct electric field modes are supported: one propagating and one evanescent. The evanescent wave yields extreme LD through strong intensity attenuation, while the propagating wave independently produces a colossal LB through its accumulated phase delay. That is, the anisotropic response is no longer between two competitive values, but between an absolute value and a near-zero value (i.e., $\text{LB} = k_0\sqrt{\varepsilon_y}d$, $\text{LD} = k_0\sqrt{-\varepsilon_x}d$). Remarkably, this mechanism enables the simultaneous enhancement of unit-thickness LD and LB by an order of magnitude (Fig. S6f), and their product—often bounded in birefringent crystals—reaches record levels.

Section 2. Experimental extraction of LB and LD in hyperbolic MoOCl₂

The hyperbolic MoOCl₂ ($\epsilon_x < 0$, $\epsilon_y > 0$) can be considered as a metallic/dielectric film along x/y direction. Along the metallic direction, the transmission decays exponentially according to $T_x = \beta_x e^{-2\gamma d} / (1 + \alpha_x e^{-2\gamma d})$, where γ is the metallic loss factor and d is the film thickness. Along the dielectric direction, the transmission is affected by multi-reflections $r = (r_1 + r_2 e^{2i\delta}) / (1 + r_1 r_2 e^{2i\delta})$, where the phase delay $\delta = k_z d$ and $r_{1,2}$ is the reflectivity at the two interfaces. Then, we have $T_y = 1 - rr^*$, and therefore the transmission difference ΔT can be written as:

$$\Delta T = \left| \frac{T_x - T_y}{T_x + T_y} \right| = \frac{1 + \alpha_x^{-1} e^{2\gamma d} - \beta(1 + \alpha_y \cos 2\delta)}{1 + \alpha_x^{-1} e^{2\gamma d} + \beta(1 + \alpha_y \cos 2\delta)}, \quad (\text{S6})$$

As for phase difference $\Delta\varphi$, it follows the definition of LB and can be simplified as a linear function with respect to the MoOCl₂ thickness d :

$$\Delta\varphi = |\text{real}(\Delta k_z)| \cdot d = k_0 |\text{real}(\sqrt{\epsilon_x} - \sqrt{\epsilon_y})| d. \quad (\text{S7})$$

To experimentally extract the LB and LD, we measured the x -/ y -/ 45° - polarized transmission spectra $T_x/T_y/T_{cross}$, and T_{cross} was measured with an analyzer at -45° polarization (Fig. S7a, cross polarization measurement). Suppose the phase retardance along x/y polarization is φ_x/φ_y , we can write the Jones matrix of the MoOCl₂ single layer as:

$$J_{\text{MoOCl}_2} = \begin{bmatrix} \sqrt{T_x} e^{i\varphi_x} & 0 \\ 0 & \sqrt{T_y} e^{i\varphi_y} \end{bmatrix}. \quad (\text{S8})$$

Then the detected transmission after an orthogonal analyzer is

$$T_{cross} = \begin{bmatrix} \frac{1}{2} & -\frac{1}{2} \\ -\frac{1}{2} & \frac{1}{2} \end{bmatrix} \begin{bmatrix} \sqrt{T_x} e^{i\varphi_x} & 0 \\ 0 & \sqrt{T_y} e^{i\varphi_y} \end{bmatrix} \begin{bmatrix} \frac{\sqrt{2}}{2} \\ \frac{\sqrt{2}}{2} \end{bmatrix}. \quad (\text{S9})$$

The experimental phase difference can be retrieved by solving Eq. S9:

$$\Delta\varphi \stackrel{\text{def}}{=} \varphi_x - \varphi_y = \arccos\left(\frac{4T_{cross} - T_x - T_y}{-2\sqrt{T_x}\sqrt{T_y}}\right). \quad (\text{S10})$$

As shown in Fig. S7b-g, the measured transmission spectra $T_x/T_y/T_{cross}$ for MoOCl₂ samples with different thickness agree well with the simulation results. The retrieved ΔT and $\Delta\phi$ are presented in Fig. S7h and Fig. S7i, respectively. Furthermore, we extracted ΔT and $\Delta\phi$ as functions of the MoOCl₂ thickness (Figs. 2d, e in the main text). Equation S6 was used to fit ΔT , yielding the parameters $\alpha_x = 3.04, \alpha_y = -0.222, \beta = 1.71$ and $\gamma = 0.016$ at a represent wavelength of 600 nm. The fitted curve, consistent with the experimental data, exhibits a nearly exponential dependence on flake thickness (Fig. 2d), which is attributed primarily to metallic reflection loss especially at larger sample thicknesses. In addition, the retrieved $\Delta\phi$ increases approximately linearly with flake thickness. This trend is confirmed by a linear fit using Equation S7, which gives a slope of $2.309k_0$ (Fig. 2e). This value agrees well with the refractive index difference of MoOCl₂ at 600 nm, given by $\Delta n = |\text{real}(\sqrt{\varepsilon_x} - \sqrt{\varepsilon_y})| = 2.303$. The similar retrieval process is applicable for other wavelengths.

Figure S8 compares 2D hyperbolic MoOCl₂ with some typical anisotropic crystals at similar thicknesses (5-7), in terms of differential refractive index and extinction coefficient (Δk and Δn). The recently emergent 2D anisotropic materials (*e.g.*, NbOCl₂ and black phosphorus) are also included, clearly highlighting the superior LD and LB properties of MoOCl₂.

Section 3. Jones-matrix-based Mueller Calculus for twisted hyperbolic bilayer

In conventional birefringent crystals, the thin-layer LD-LB interaction is mostly evaluated by the second order Mueller matrix, *i.e.*, $\mathbf{M}_{03}^{(2)}$ (8, 9). However, in our proposed twisted hyperbolic bilayer, LD and LB are both colossal in each layer, and thus the higher-order contributions should also be involved in calculating the LD-LB signal. Here, we present a rigorous and generalized expression for the LD-LB interaction from a Jones-matrix-based Mueller Calculus, which captures the CD response to an infinite order. Firstly, for a hyperbolic single layer (Fig. S9a), giant LD and LB are decoupled onto two orthogonal principal axes, its amplitude and phase manipulation properties can be well described by a diagonalized Jones matrix,

$$J = \begin{bmatrix} e^{-k_0\sqrt{-\varepsilon_x}d} & 0 \\ 0 & e^{ik_0\sqrt{\varepsilon_y}d} \end{bmatrix}, \quad (\text{S11})$$

where $k_0 = 2\pi/\lambda$, ε_x is the negative permittivity at x direction, ε_y is the positive permittivity at y direction, and d is the thickness. We can rewrite the Jones matrix in a manner of LD-LB interaction:

$$J = \begin{bmatrix} e^{-LD} & 0 \\ 0 & e^{iLB} \end{bmatrix} = e^{\frac{LD+iLB}{2}} J_{LD} J_{LB}, \quad (S12)$$

where $J_{LD} = \begin{bmatrix} e^{-LD/2} & 0 \\ 0 & e^{LD/2} \end{bmatrix}$, $J_{LB} = \begin{bmatrix} e^{-iLB/2} & 0 \\ 0 & e^{iLB/2} \end{bmatrix}$, $LD = k_0 \sqrt{-\varepsilon_x} d$ and $LB = k_0 \sqrt{\varepsilon_y} d$.

Next, the Jones matrix for a twisted bilayer (Fig. S9b) with an interlayer twist angle θ is written as:

$$\begin{aligned} J_\theta &= \begin{bmatrix} e^{-LD_2} & 0 \\ 0 & e^{iLB_2} \end{bmatrix} \left\{ R(\theta) \begin{bmatrix} e^{-LD_1} & 0 \\ 0 & e^{iLB_1} \end{bmatrix} R^{-1}(\theta) \right\} \\ &= e^{\frac{-LD+iLB}{2}} J_{LD_2} J_{LB_2} [R(\theta) J_{LD_1} J_{LB_1} R^{-1}(\theta)] \end{aligned} \quad (S13)$$

where $R(\theta) = \begin{bmatrix} \cos \theta & \sin \theta \\ -\sin \theta & \cos \theta \end{bmatrix}$ is the rotation matrix, LD_i, LB_i are the linear dichroism/birefringence of layer i , and $LD = LD_1 + LD_2$, $LB = LB_1 + LB_2$.

The Stokes vector for polarization optics is defined as $S = (I, Q, U, V)^T$, and the elements are calculated as: $I = E_x^2 + E_y^2$, $Q = E_x^2 - E_y^2$, $U = 2E_x E_y \cos \varphi$, $V = 2E_x E_y \sin \varphi$, where E_x and E_y are the amplitudes of the x - and y -components of the electric field and φ is their phase difference, $\varphi \stackrel{\text{def}}{=} \varphi_y - \varphi_x$. The Mueller matrix $\mathbf{M}$ connects output and input Stokes vectors (10):

$$S_{out} = \mathbf{M} S_{in}. \quad (S14)$$

Stokes vector for a left /right circular polarization light is $S = (1, 0, 0, \pm 1)^T$. Therefore, the circular dichroism can be simplified to

$$CD = \frac{t_{lcp} - t_{rcp}}{t_{lcp} + t_{rcp}} = \frac{\mathbf{M}_{03}}{\mathbf{M}_{00}}. \quad (S15)$$

For a deterministic hyperbolic bilayer (Fig. S9b), the 4×4 Mueller matrix $\mathbf{M}_{ij}$ is uniquely determined by its 2×2 Jones matrix J_θ via (5)

$$\mathbf{M}_{ij} = \frac{1}{2} \text{tr} \{ \sigma_i J_\theta \sigma_j J_\theta^{*T} \} \quad (S16)$$

This allows to connect the LD-LB encoded in J_θ to total apparent CD through the Mueller formalism, where $\text{tr}\{\mathbf{A}\}$ is the trace of the matrix $\mathbf{A}$. Substituting equation S13 into equation S16, the related elements of $\mathbf{M}_{03}$ and $\mathbf{M}_{00}$ can be derived as,

$$\mathbf{M}_{03} = e^{-LD} \sin LB_1 \sinh LD_2 \sin 2\theta, \quad (S17)$$

$$\mathbf{M}_{00} = e^{-LD} [\cosh(LD_1) \cosh(LD_2) + \sinh(LD_1) \sinh(LD_2) \cos 2\theta], \quad (S18)$$

$$CD = \frac{\mathbf{M}_{03}}{\mathbf{M}_{00}} = \frac{\sin LB_1 \sinh(LD_2) \sin 2\theta}{\cosh(LD_1) \cosh(LD_2) + \sinh(LD_1) \sinh(LD_2) \cos 2\theta}, \quad (S19)$$

From the expression of $\mathbf{M}_{03}$, we make clear that the dominant contribution to CD arises from a cross term that interferes first layer LB_1 with second layer LD_2 , which is denoted as a nontrivial LB-LD channel (red arrow in Fig. S10a). In such a case, the first layer LB_1 rotates the RCP and LCP Stokes vectors away from the poles along different trajectories, which become two distinct elliptical (or linear) polarization states on the Poincaré sphere. When these two nonequivalent states encounter a subsequent LD_2 , different transmission are resulted to generate a net CD. Remarkably, the hyperbolic dispersion of 2D $MoOCl_2$ decouples and dramatically enhances LD and LB within each layer, so that the nontrivial $LB_1 \rightarrow LD_2$ interaction is substantially boosted, yielding record-high apparent CDs (Eqs. S17, S18).

In contrast, another cross term $LD_1 \rightarrow LB_2$ is a trivial interaction channel, because LD_1 attenuates RCP and LCP identically and the subsequent LB_2 can only rotate Stokes vectors on the Poincaré sphere without changing their magnitudes, therefore generating zero net CD (blue dashed arrow in Fig. S10a). Mathematically, this can be deduced by using a pure LD element and a pure LB element as:

$$J_\theta = J_{LB} \left\{ \begin{bmatrix} \cos \theta & \sin \theta \\ -\sin \theta & \cos \theta \end{bmatrix} J_{LD} \begin{bmatrix} \cos \theta & -\sin \theta \\ \sin \theta & \cos \theta \end{bmatrix} \right\} \quad (S20)$$

$$\mathbf{M}_{03} = \frac{1}{2} \text{tr} \left\{ \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} J_\theta \begin{bmatrix} 0 & -i \\ i & 0 \end{bmatrix} J_\theta^{*T} \right\} = 0 \quad (S21)$$

where J_{LD} and J_{LB} are the Jones matrices for a pure LD element and a pure LB element.

Section 4. Decomposition of symmetric and antisymmetric CD

A hallmark feature distinguishing apparent CD from genuine CD is the sign reversal of CD response upon sample flipping (*II*). However, for our twisted hyperbolic bilayers, the situation is markedly different. When the sample is flipped (or backward illumination, Fig. S10b), the incident

light first passes through the original second layer and then through the first layer. Therefore, the total Jones matrix becomes nonreciprocal,

$$J_{\text{total}}^{\text{flip}} = J_1 J_2 \neq (J_2 J_1 = J_{\text{total}}) \quad (\text{S22})$$

According to the rigorous LB-LD interaction formalism in Section 3, nonzero CD is accumulated by second layer LB₂ and first layer LD₁. We therefore attribute the total apparent CD involving two contributions: *symmetric* CD and *antisymmetric* CD (12),

$$\text{CD}_{\text{total}} = \text{CD}_{\text{sym}} + \text{CD}_{\text{anti}}, \quad (\text{S23})$$

where CD_{sym} is the unchanged component while CD_{anti} changes its sign upon sample flipping. Based on the Jones matrix method (Eq. S19), we can derive CD_{total} before and after flipping. Then, the symmetric CD and antisymmetric CD can be calculated as,

$$\text{CD}_{\text{sym}} = \frac{\text{CD}_{\text{total}} + \text{CD}_{\text{total}}^{\text{flip}}}{2} = \left[\frac{1}{2} + \frac{\sin \text{LB}_2 \sinh(\text{LD}_1)}{2 \sin \text{LB}_1 \sinh(\text{LD}_2)} \right] \text{CD}_{\text{total}}, \quad (\text{S24.1})$$

$$\text{CD}_{\text{anti}} = \frac{\text{CD}_{\text{total}} - \text{CD}_{\text{total}}^{\text{flip}}}{2} = \left[\frac{1}{2} - \frac{\sin \text{LB}_2 \sinh(\text{LD}_1)}{2 \sin \text{LB}_1 \sinh(\text{LD}_2)} \right] \text{CD}_{\text{total}}. \quad (\text{S24.2})$$

As shown in Fig. S11(a, b), when these two layers have same thicknesses ($d_1=d_2$), the two layers are exchangeable for the symmetric component. The stacking maps onto itself upon sample flipping, so that it contributes the same CD response. The *antisymmetric* contribution arises from the unequal thickness part between two layers (Fig. S11c-f), where the roles of LD and LB are exchanged directly, thus leads to different CD upon reversion. When these two layers have different thicknesses ($d_1 \neq d_2$), the magnitude of phase retardance (LB) and attenuation (LD) of the layers are different, that is, $\text{LB}_1 \neq \text{LB}_2$, $\text{LD}_1 \neq \text{LD}_2$. As the incident direction is reversed, the sequence of the optical interaction reverses (i.e., from $\text{LB}_1 \rightarrow \text{LD}_2$ to $\text{LB}_2 \rightarrow \text{LD}_1$). Also, because the dispersion of $\text{LB}_{1,2}$ and $\text{LD}_{1,2}$ in each layer, the CD peak position naturally and deterministically shifts to the specific wavelength where this LD-LB product is mathematically maximized, as marked by the purple shaded area in Fig. S18.

Section 5. Yeh's 4x4 Matrix Method

Jones matrix or Mueller matrix method provides clear physical insight of our system, but it completely neglects the reflection of light. Therefore, we need a more accurate theoretical

calculation model to describe the CD response. Yeh's 4×4 Matrix Method (Yeh's Method) provides the exact solution to Maxwell's equations for layered systems (13). It considers that only the eigenmode of the electromagnetic medium can actually propagate with a corresponding phase factor in a single layer. Layers can be connected with boundary conditions at interface. For a monochromatic electromagnetic wave with frequency ω , the wave equation is written as

$$\mathbf{k} \times (\mathbf{k} \times \mathbf{E}) + \omega^2 \mu \varepsilon \mathbf{E} = 0, \quad (\text{S25})$$

where $\mu = \mu_0$ and ε is the permittivity tensor of the medium. As for a twisted uniaxial crystal, the permittivity tensor is written as $\varepsilon(\theta) = R(\theta) \text{diag}(\varepsilon_x, \varepsilon_y, \varepsilon_z) R^{-1}(\theta)$, where θ is the angle between polarization and crystal axis and $R(\theta)$ is the rotation matrix. Considering the uniformity in the x y directions, the electric field can be written in a form $\mathbf{E} = \mathbf{E}(x, y) \exp(i\gamma z)$, where γ is the phase constant. After a standard procedure to derive the wave equation (S25) to a form of matrix equations, the determinant of the coefficient matrix must be zero for nontrivial solutions, which gives four eigenmodes. Then the electromagnetic field can be expressed as linear combination of all eigen states

$$\mathbf{E} = \sum_{n=1}^4 A_n \mathbf{p}_n \exp[i(\omega t - \gamma_n z)], \quad (\text{S26.1})$$

$$\mathbf{H} = \sum_{n=1}^4 A_n \mathbf{q}_n \exp[i(\omega t - \gamma_n z)], \quad (\text{S26.2})$$

where $\mathbf{p}_n$ and $\mathbf{q}_n$ are the polarization vectors. As shown in Fig. S12, the twisted bilayer of hyperbolic material can be processed with 4 layers and 3 interfaces. Imposing the continuity of E_x, E_y, H_x and H_y at the interfaces, we can write the following matrix equation,

$$\begin{pmatrix} A_1(0) \\ A_2(0) \\ A_3(0) \\ A_4(0) \end{pmatrix} = \mathbf{T} \begin{pmatrix} A_1(s) \\ 0 \\ A_3(s) \\ 0 \end{pmatrix} \quad (\text{S27})$$

where the transfer matrix $\mathbf{T} = \mathbf{D}_{\text{sub}}^{-1}(0) * \mathbf{D}(0) \mathbf{P}(0) \mathbf{D}^{-1}(0) \mathbf{D}(\theta) \mathbf{P}(\theta) \mathbf{D}^{-1}(\theta) \mathbf{D}_{\text{air}}(0)$, $A_{1,3}(0)$ being the incident wave, which is the circular polarization $[1, \sigma i]^T$, $A_{2,4}(0)$ being the reflected wave and $A_{1,3}(\theta)$ is the transmitted wave. Components of $\mathbf{T}$ are calculated as

$$\mathbf{D}(\theta) = \begin{bmatrix} -\sin(\theta) & -\sin(\theta) & \cos(\theta) & \cos(\theta) \\ \frac{-\sqrt{\varepsilon_y}}{\mu} \sin(\theta) & \frac{\sqrt{\varepsilon_y}}{\mu} \sin(\theta) & \frac{\sqrt{\varepsilon_x}}{\mu} \cos(\theta) & \frac{-\sqrt{\varepsilon_x}}{\mu} \cos(\theta) \\ \cos(\theta) & \cos(\theta) & \sin(\theta) & \sin(\theta) \\ \frac{-\sqrt{\varepsilon_y}}{\mu} \cos(\theta) & \frac{\sqrt{\varepsilon_y}}{\mu} \cos(\theta) & \frac{-\sqrt{\varepsilon_x}}{\mu} \sin(\theta) & \frac{\sqrt{\varepsilon_x}}{\mu} \sin(\theta) \end{bmatrix}, \quad (\text{S28})$$

$$\mathbf{P}(\theta) = \begin{bmatrix} \exp(i\sqrt{\varepsilon_y}k_0 dz) & 0 & 0 & 0 \\ 0 & \exp(-i\sqrt{\varepsilon_y}k_0 dz) & 0 & 0 \\ 0 & 0 & \exp(i\sqrt{\varepsilon_x}k_0 dz) & 0 \\ 0 & 0 & 0 & \exp(-i\sqrt{\varepsilon_x}k_0 dz) \end{bmatrix}. \quad (\text{S29})$$

Then the transmittance and reflectance for circular polarization incidence can be calculated as

$$T_\sigma = \frac{|A_1(s)|^2 + |A_3(s)|^2}{2}, \quad (\text{S30})$$

The circular dichroism for transmission is then calculated as

$$\text{CD} = \frac{T_{+1} - T_{-1}}{T_{+1} + T_{-1}}. \quad (\text{S31})$$

Section 6. Parametric dependence of CD

We firstly study the influence of anisotropy in hyperbolic materials on CD using the Yeh's transfer matrix method. Fig. S13a shows a contour plot of CD as a function of $-\varepsilon_x$ and ε_y , where the wavelength is set at 600 nm and the twist angle is 45 degrees. We can see that a larger anisotropy, proportional to $\varepsilon_y - \varepsilon_x$, leads to a larger CD. Then we use the permittivity parameters of MoOCl_2 to conduct the simulations. The dependence of twist angle on CD is shown in Fig. S13. Figures S13(c, d) show CD as a function of twist angle and d_1, d_2 . Figures S13(e, f) correspond to the case when the twist angle is 45 degrees, with comparison between Jones matrix method and the COMSOL simulation result, which validated our derivations and calculations.

For LD/LB-coupling induced apparent CD, the maximum CD is usually expected when the optical axes of the two anisotropic layers are twisted by approximately 45° (14). However, we note that in our practical twisted hyperbolic bilayer system, the experimentally observed and rigorously simulated optimal angles deviate from 45° . This deviation is owing to the normalization by total transmission within such an ultrathin homostructure. Indeed, because an ideal LD-LB system typically ignores the effect of boundary reflection, the absolute CD, $T_{LCP} - T_{RCP}$, associated with the Muller element M_{03} , is proportional to $\sin(2\theta)$ as: $T_{LCP} - T_{RCP} = 2M_{03} = 2e^{-LD} \cdot \sin(LB_1) \sinh(LD_2) \sin(2\theta) \propto \sin(2\theta)$, which will peak at 45° (Fig. S14a). However, in our work, we use the definition of normalized $CD = (T_{LCP} - T_{RCP}) / (T_{LCP} + T_{RCP}) = M_{03} / M_{00}$, where the total transmission $M_{00} = e^{-LD} [\cosh(LD_1) \cosh(LD_2) + \sinh(LD_1) \sinh(LD_2) \cos(2\theta)]$ is also a function of θ . As a result, the dependence of normalized CD deviates from the $\sin(2\theta)$ function, and does not peak at 45° (Fig. S14b). This derivation has also been validated by TMM simulations as shown in Figs. S14(c, d).

Section 7. Comparison of CD within hyperbolic and birefringent regimes

As discussed before, hyperbolic materials generate giant LD and LB due to the sharp contrast between evanescent mode and orthogonal propagating mode, which originates from negative ϵ_x and positive ϵ_y . When twisting two hyperbolic layers, the interlayer LD and LB interference generates extremely large CD. This mechanism is completely different from apparent CD in conventional birefringent crystals (15). For example, when incident frequencies larger than ω_p^x , both in-plane permittivities of MoOCl_2 are positive, performing as a birefringent material. This leads to both propagating eigen-modes and significantly reduced LD and LB. Correspondingly, in our proposed MoOCl_2 bilayer system, the apparent CD decreases to a small value in twisted MoOCl_2 bilayer. As demonstrated below in Fig. S15, the CD response is compared across the dielectric and hyperbolic parameter spaces, respectively.

Section 8. CD in gapped hyperbolic twisted bilayer

We now consider the case where the two layers are separated by an air gap g . Using the Yeh's transfer matrix method, CD of the gapped twisted bilayer can be obtained. We find CD oscillates periodically with the air gap and the period is the half wavelength (Fig. S16d). The observed oscillatory modulation with gap arises because an air gap introduces a Fabry-Perot cavity between

the two anisotropic layers. The condition of Fabry-Pérot interference is $n \cdot \text{gap} = m\lambda/2$, where gap is the gap length, the air refractive index $n = 1$, λ is the fixed wavelength and m is an integer. Because the MoOCl_2 interfaces (especially along the metallic-like axis) are highly reflective, the multiple internal reflections modulate the overall transmission periodically with a period of $\lambda/2$. Fig. S16e displays the full CD spectra as a function of both wavelength and gap distance, showing typical FP interference fringes (16). Fig. S16f further illustrates the CD oscillations as a function of the first-layer thickness (d_1) and wavelength, also exhibiting characteristic FP interference features. These two maps confirm that in the gapped twisted bilayer system, the apparent CD signals are modulated by both intralayer and interlayer reflections, leading to the CD response depending on both layer thickness and gap thickness. It also indicates that the interlayer coupling of electric-magnetic field in such twisted bilayer could be neglected. Consequently, the genuine CD contribution is negligible for our twisted hyperbolic bilayers, because each single layer lacks strong magnetic dipole responses at the operational frequencies and the interlayer coupling is also weak.

Section 9. Angle-independent CD response

The CD response of our twisted hyperbolic bilayer exhibits remarkable robustness against variations in the incident angle (Fig. S21). In deep-subwavelength limit of thin layers, the oblique-incidence-induced phase accumulation (LB) is negligible. On the other hand, the metallic-like and reflection-based LD are only weakly dependent on the incidence angle over a broad angular range. Therefore, their nanoscale LB-LD interaction remains essentially angle-insensitive. This stands in stark contrast to conventional chiral metamaterials, whose performance often degrades rapidly off-normal incidence due to reliance on magnetic resonances. This attribute is desirable for practical devices, ensuring consistent performance under diverse illumination conditions.

Supplementary Figures and Tables

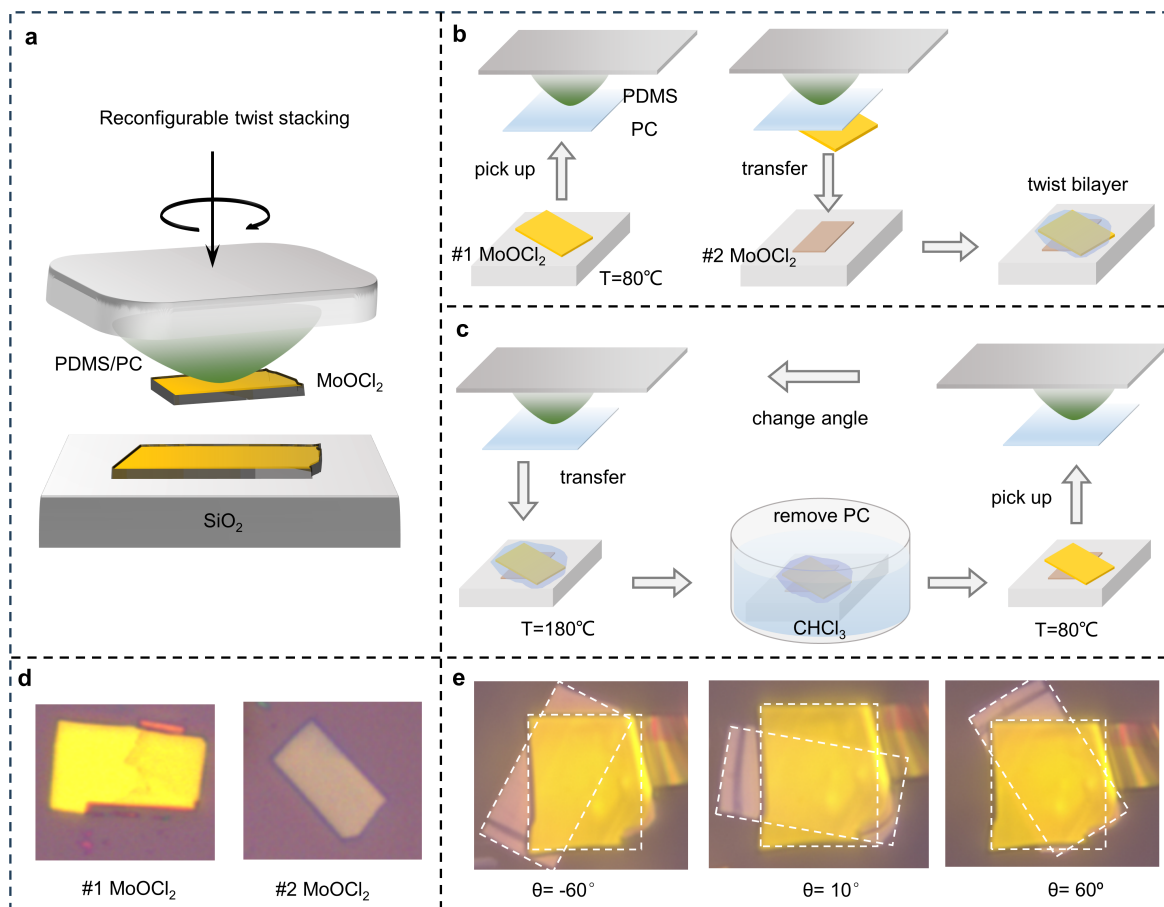


Fig. S1| Fabrication process of twisted MoOCl₂ bilayers. **a**, Schematic of the angle-resolved transfer techniques. **b**, **c**, Detailed peel-and-restack process of twisted MoOCl₂ bilayers. **d**, Optical images of a relatively thick MoOCl₂ layer ($d = 165$ nm) and a thin MoOCl₂ layer ($d = 55$ nm). **e**, Optical images of twisted bilayer samples with different twist angle of $\theta = 60^\circ$, -10° , and -60° , respectively.

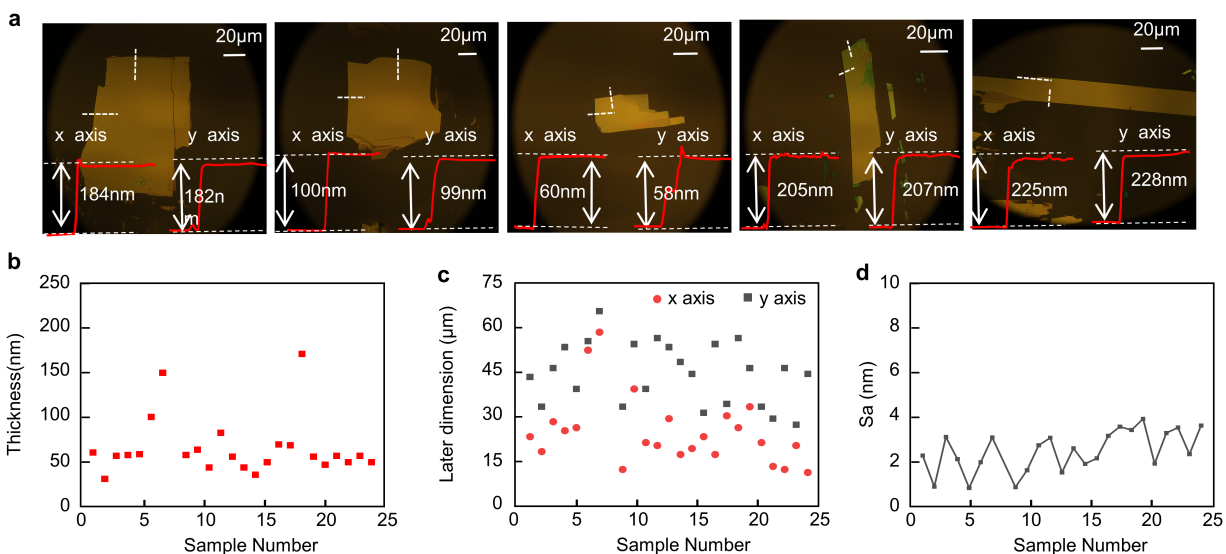


Fig. S2| MoOCl₂ flake uniformity and reproducibility. **a**, Optical images of 5 mechanically exfoliated MoOCl₂ flakes along with AFM height profiles in the x and y directions. **b-d**, Flake thicknesses, lateral-dimensions and surface roughness (thickness uniformity, Sa: arithmetical mean height) of 25 statistic samples.

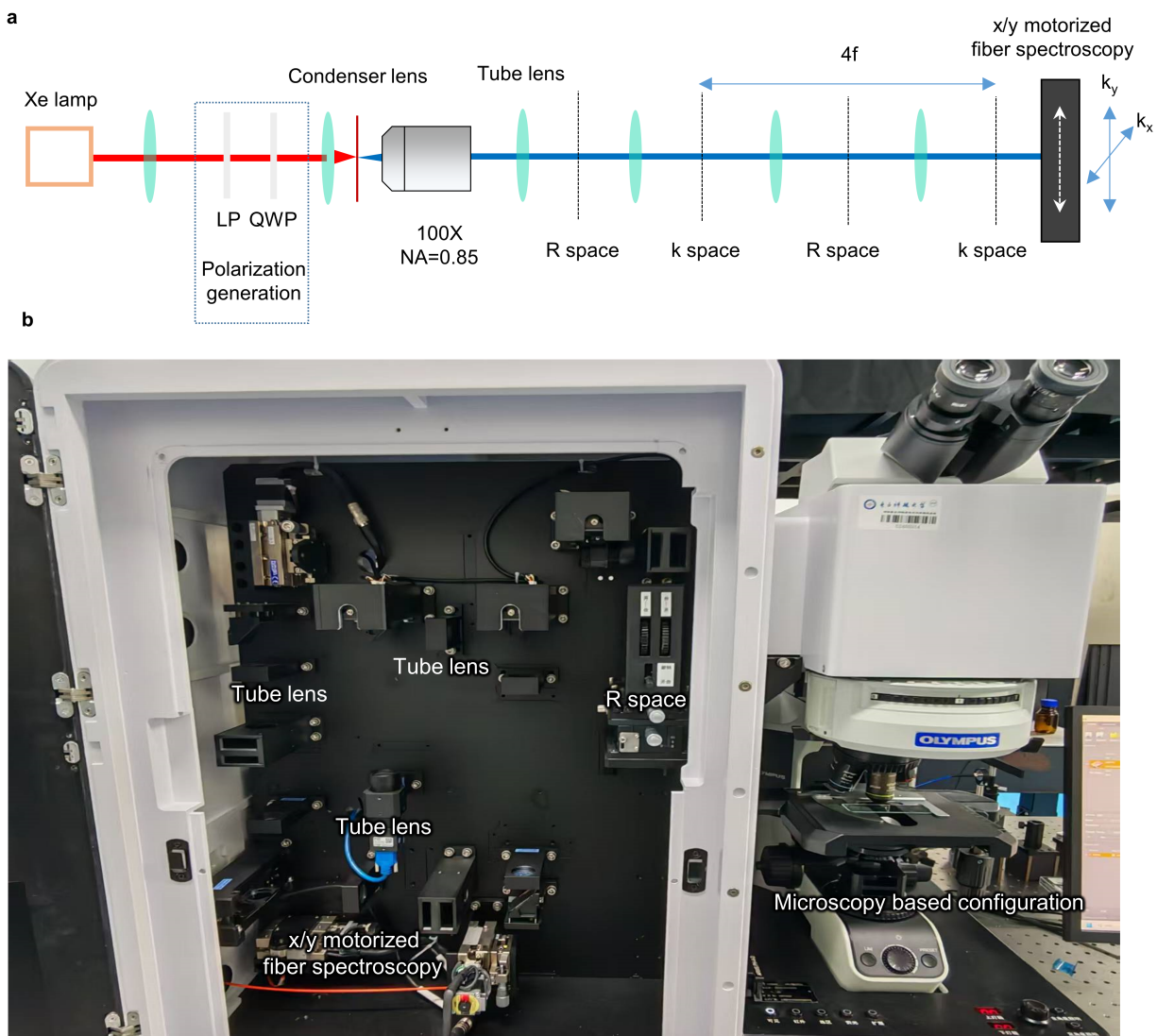


Fig. S3| Angle-Resolved Micro-Spectroscopy (ARMS). The optical set-ups **(a)** and the system photo **(b)** of our commercial ARMS system. The angle-resolved measurement relies on the Fourier imaging principle, where specific exit angles from the focal plane are deterministically mapped to distinct spatial coordinates at back focal plane. By motorized scanning the spectrometer fiber across this conjugated Fourier plane, angle-resolved transmission spectra are simultaneously acquired.

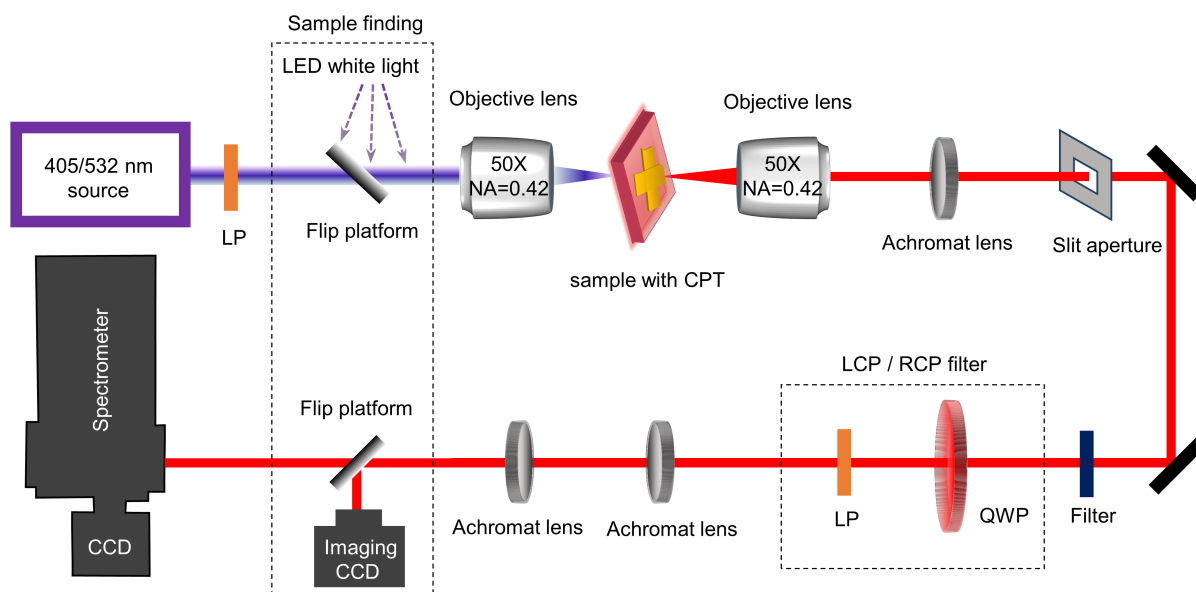


Fig. S4| Optical set-ups for chiral emission measurements. Schematic and detailed optical elements in a real-space optical spectra and imaging measurement setup.

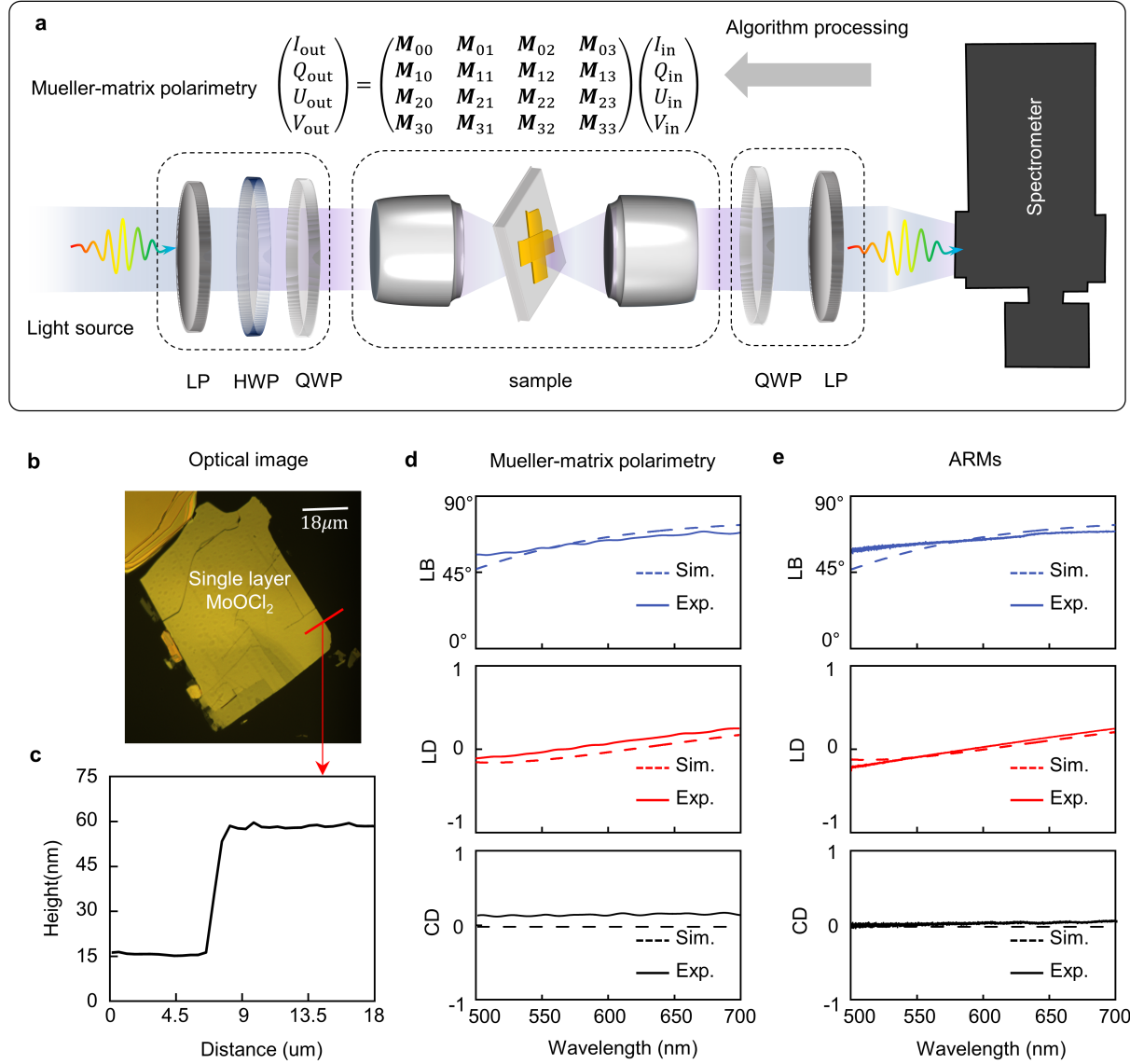


Fig. S5| Calibration on nonchiral controls. **a**, The schematic of a home-built Muller-matrix polarimetry set-up. **b**, **c**, The optical image and AFM line-scan of a single-layer MoOCl_2 . **d**, **e**, The LB, LD and CD results measured by Muller-matrix polarimetry (**d**) and ARMs (**e**).

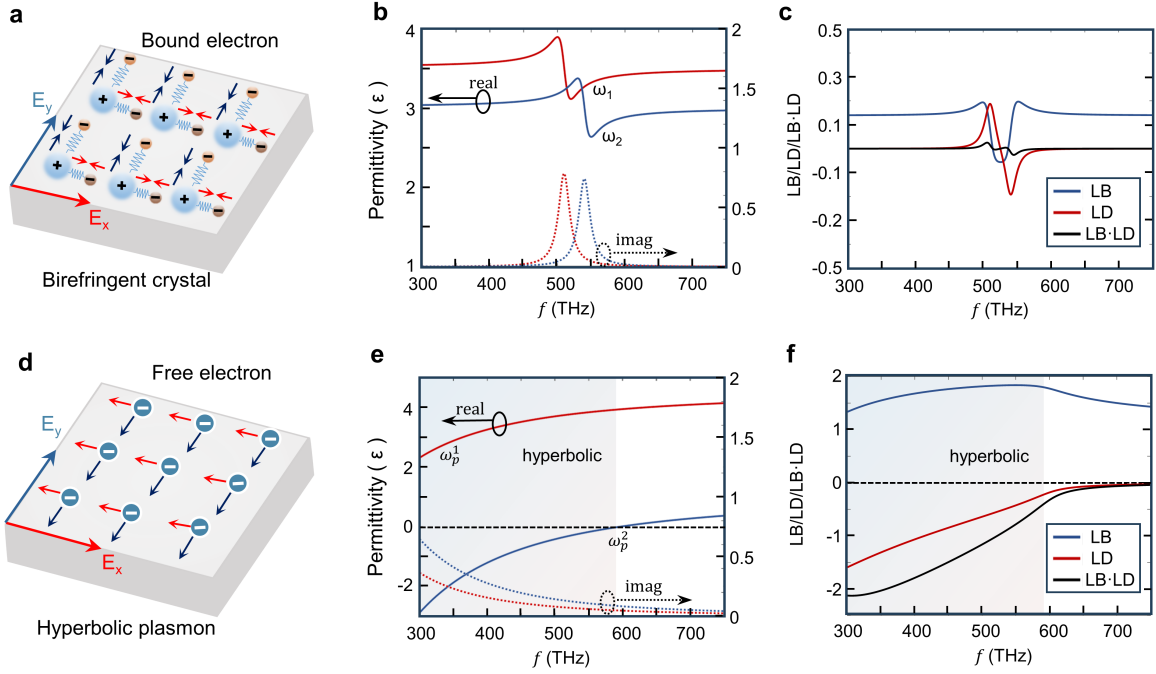


Fig. S6| LB and LD in bound-electron and free-electron based systems. **a, d**, Schematic illustrating the polarization of **(a)** bounded electrons in birefringent crystals and **(d)** collective oscillations of free electrons in hyperbolic metals with external optical fields. **b, e**, The dielectric functions of **(b)** a birefringent crystal, and **(e)** a hyperbolic metal. The fitting parameters for the Drude–Lorentz model are given in Table S1. **c, f**, The unit-thickness LB, LD and LB·LD products in the two systems.

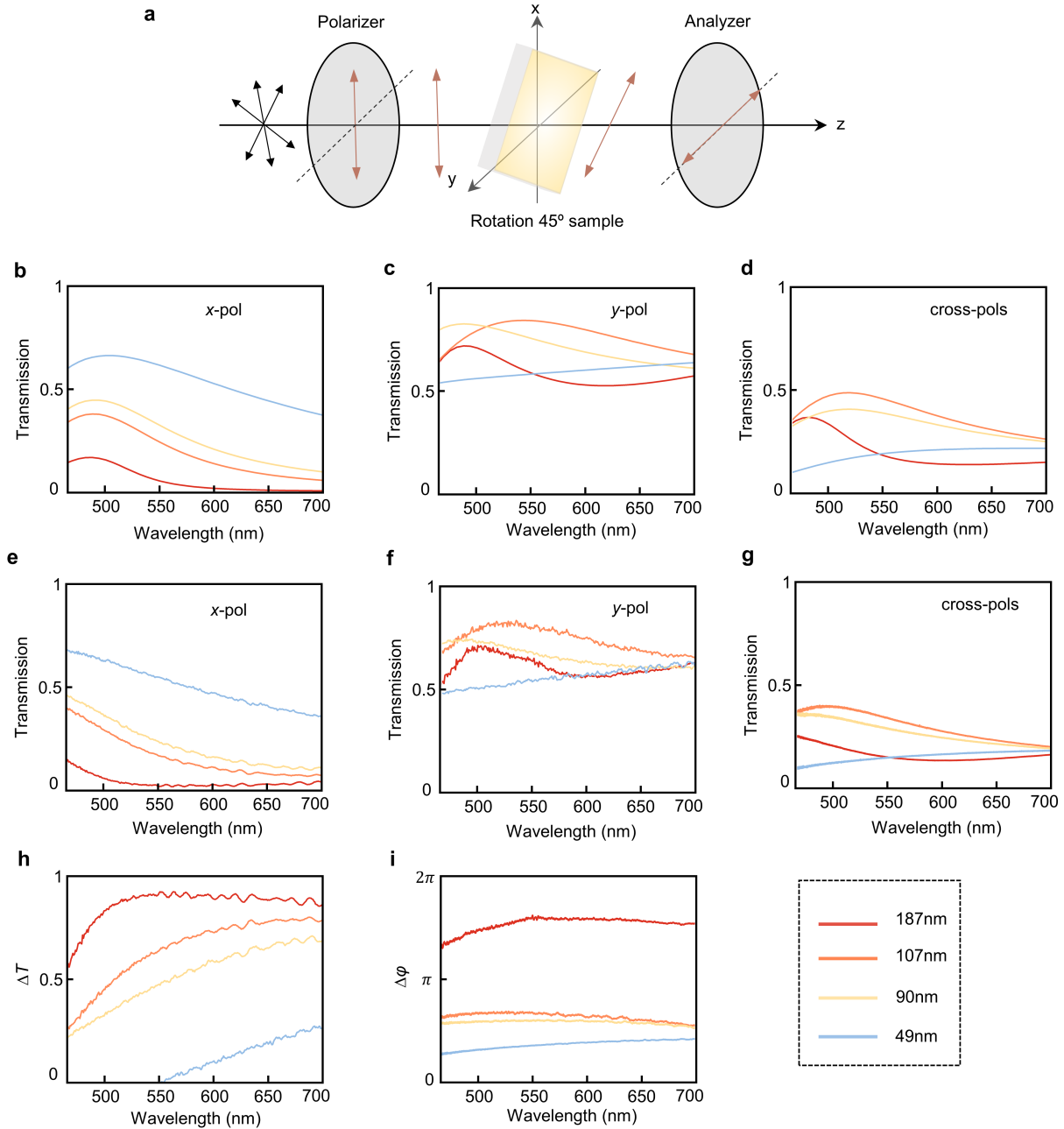


Fig. S7| Coexistence of giant LD and LB in 2D hyperbolic MoOCl_2 . **a**, Schematic of the cross-polarization set-ups. MoOCl_2 flakes are located between a pair of orthogonal linear polarizer and analyzer, the crystal principle axis is set to 45° in relative to both the polarizer and analyzer. **b-d**, Numerical simulated transmission spectra for different thick MoOCl_2 flakes with polarization along the in-plane x-axis, y-axis and cross-pols, respectively. **e-g**, The corresponding experimentally detected transmission spectra. **h**, The normalized transmission difference spectra $\Delta T = |(T_x - T_y)/(T_x + T_y)|$ derived from data in (e, f). **i**, The phase retardance $\Delta\phi$ derived from data in (e-d).

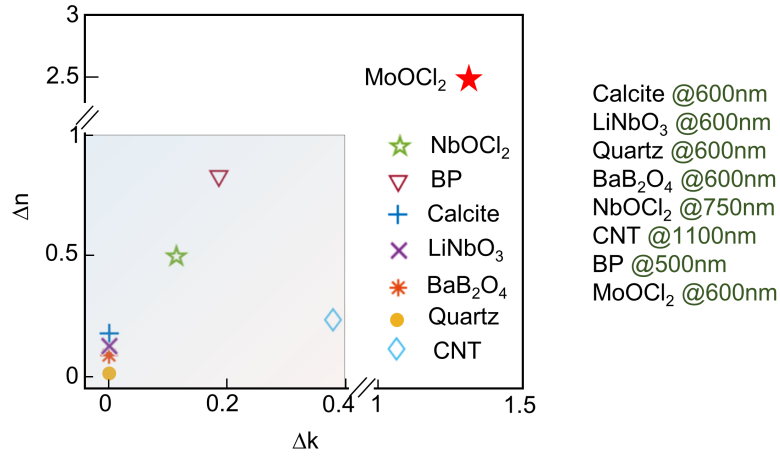


Fig. S8 Comparison between 2D MoOCl_2 and some typical anisotropic materials (5-7) with similar thicknesses in terms of Δn and Δk in their own own relevant operational wavelengths.

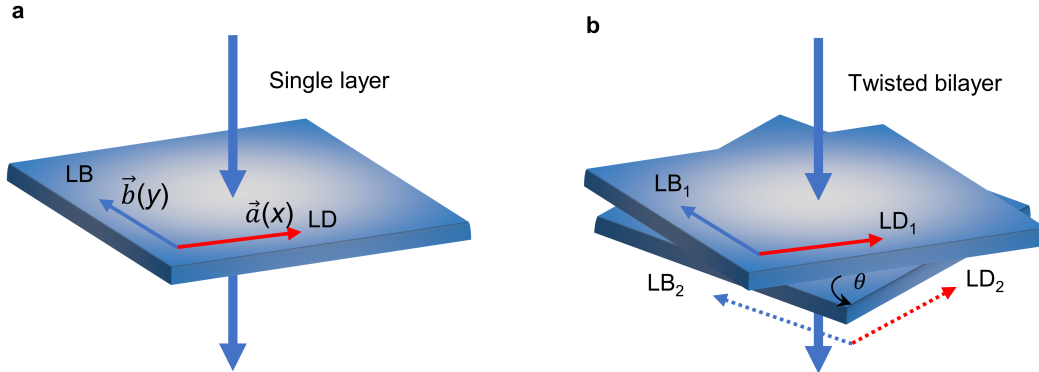


Fig. S9 Schematics of (a) orthogonal LB and LD in a hyperbolic single layer, and (b) interlayer LD and LB interaction in the twisted hyperbolic bilayer.

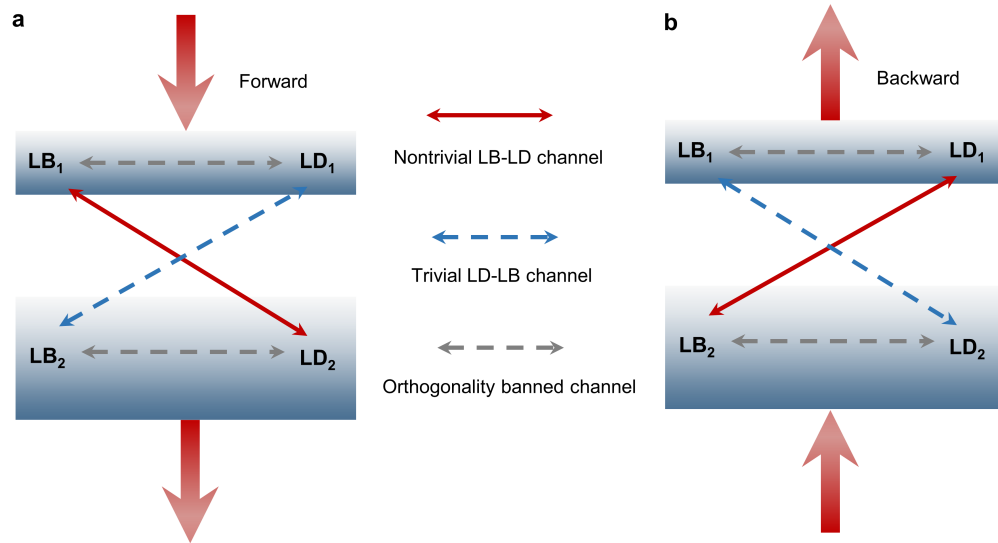


Fig. S10| Twist enabled LB-LD interaction. **a**, LB_1 - LD_2 dominant twisted interlayer interaction for forward illumination. **b**, LB_2 - LD_1 dominant twisted interlayer interaction for backward illumination.

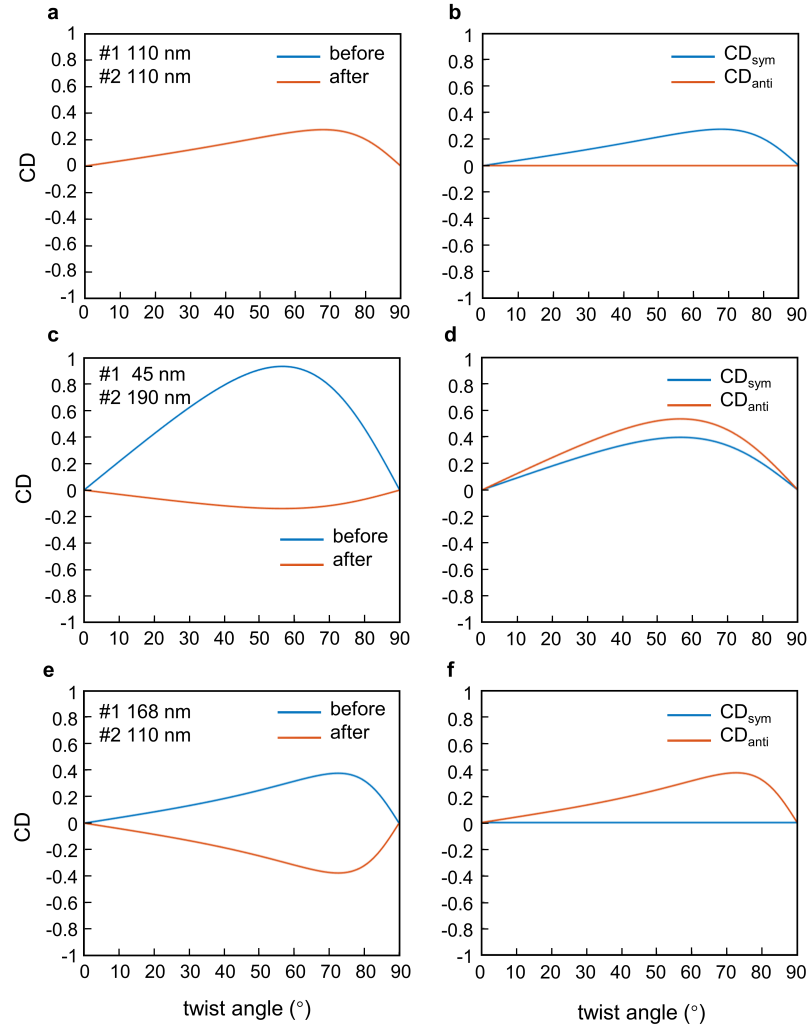


Fig. S11| Symmetric and asymmetric CD with different sample thickness. **a**, CD before and after sample flipping with thickness of $d_1 = 110$ nm and $d_2 = 110$ nm. **b**, Extracted symmetric and asymmetric CD from **a**. **c**, **d**, Flipped and extracted CD with $d_1 = 45$ nm, $d_2 = 190$ nm and **e**, **f** with $d_1 = 168$ nm, $d_2 = 110$ nm. The wavelength is set as $\lambda=600$ nm.

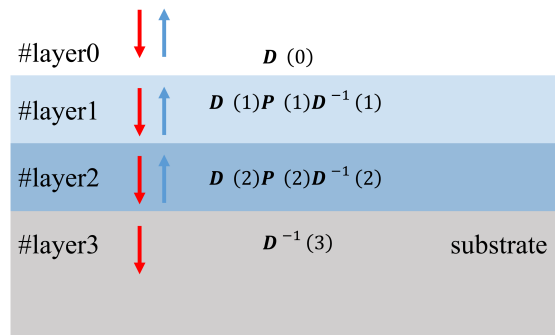


Fig. S12| The schematic of Yeh's 4x4 Matrix Method in layered systems.

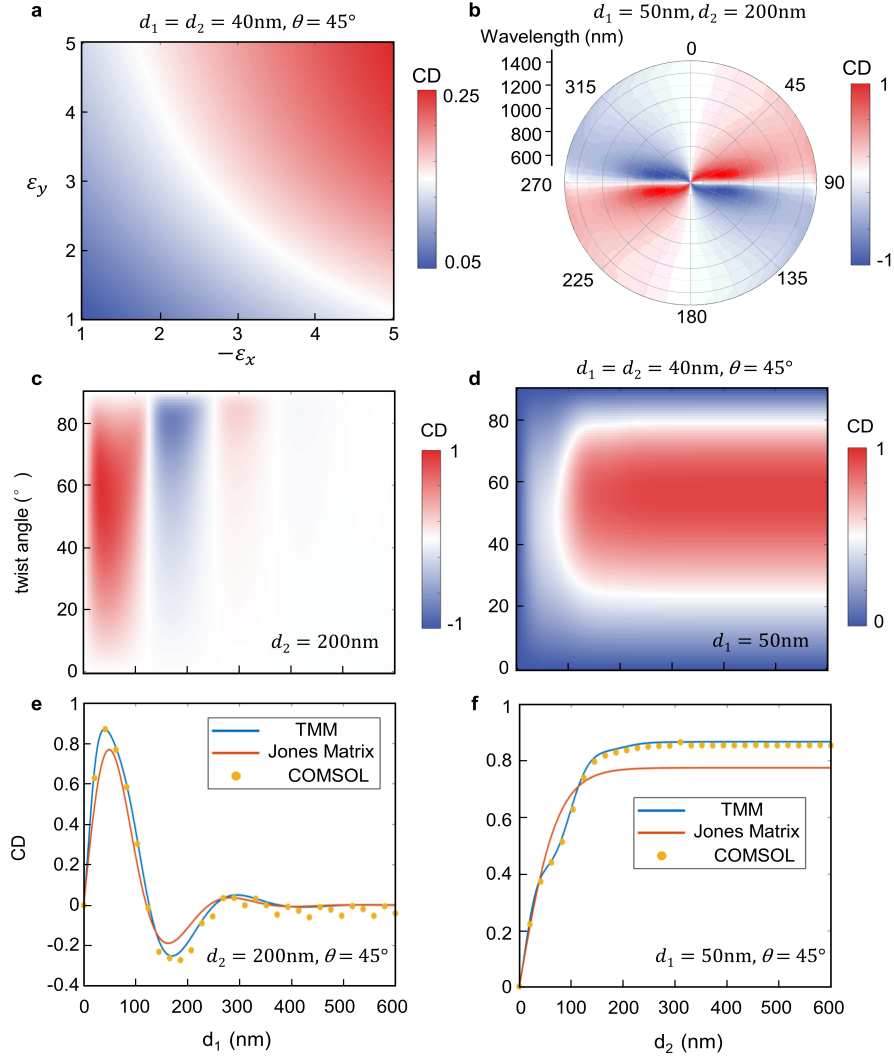


Fig. S13| Parametric dependence of CD in twisted bilayer. **a**, CD as a function of the anisotropic dielectric components $-\epsilon_x$ and ϵ_y . **b**, Polar contour plot of CD as a function of wavelength and the twist angle. **c**, **d**, Contour plot of CD versus twist angle and the layer thickness. **e**, **f**, CD versus d_1 and d_2 with Yeh's TMM Method, Jones Matrix Method and COMSOL simulations. The wavelength is set as $\lambda=600\text{ nm}$ for all results.

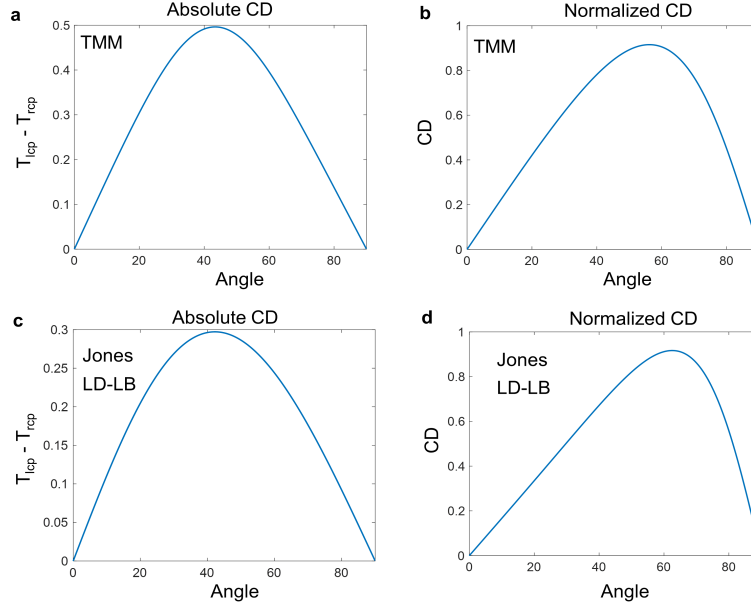


Fig. S14| Absolute and normalized CD response as a function of twist angle. **a, b**, Absolute CD ($T_{LCP} - T_{RCP}$) and normalized CD ($(T_{LCP} - T_{RCP})/(T_{LCP} + T_{RCP})$) as functions of the twisted angle θ calculated by Jones Matrix based LD-LB interaction model. The layer thicknesses are $d_1=50$ nm, and $d_2=190$ nm. The wavelength is set as $\lambda=600$ nm for all results. **c, d**, Absolute CD and normalized CD as functions of twisted angle θ calculated by TMM.

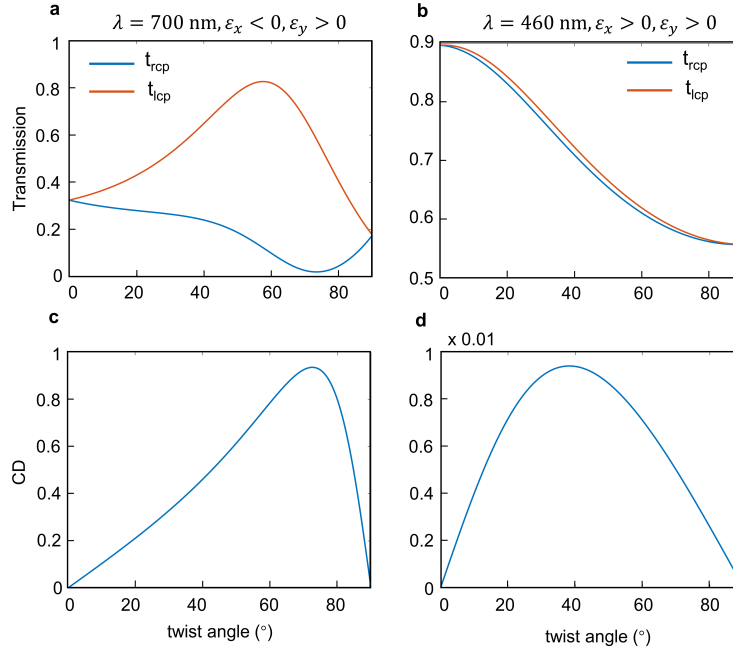


Fig. S15| Comparison of CD in hyperbolic and birefringent regimes. Transmission of RCP and LCP light at **(a)** $\lambda = 700$ nm and **(b)** $\lambda = 460$ nm. CD results at **(c)** $\lambda = 700$ nm and **(d)** $\lambda = 460$ nm. The thickness of two MoOCl_2 layers are $d_1=50$ nm, and $d_2=200$ nm.

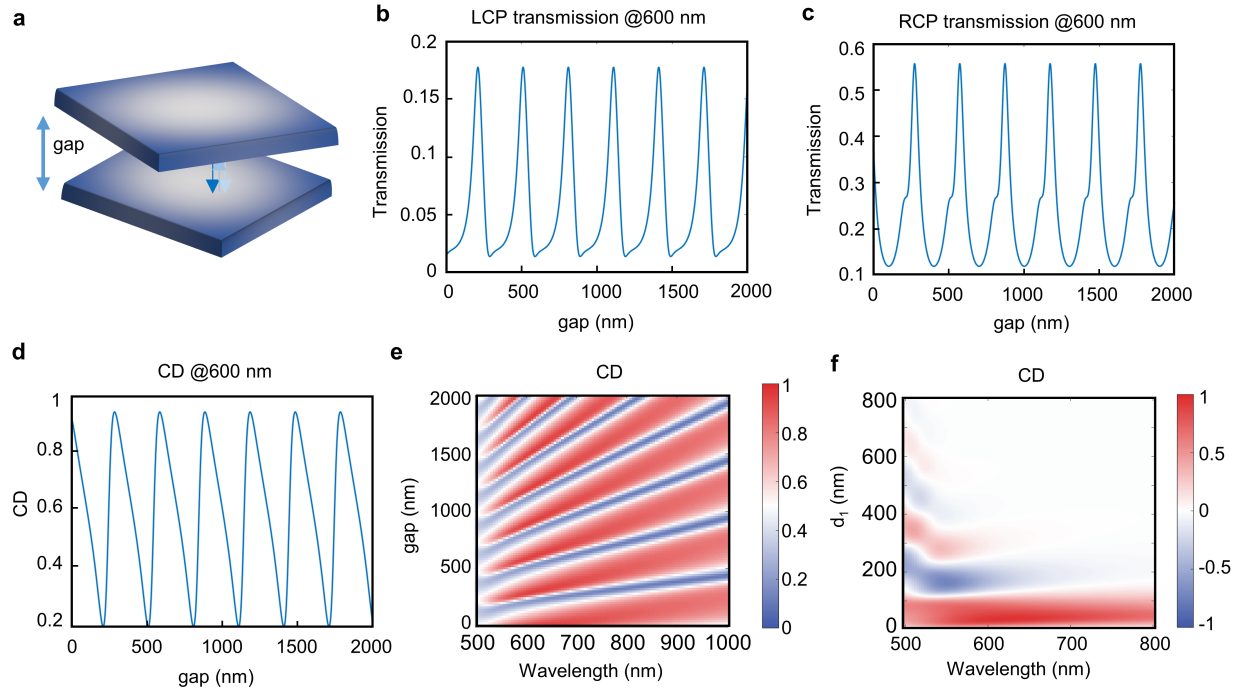


Fig. S16| CD of gapped twisted bilayer. **a**, The schematic of gapped twisted bilayer. Layer thickness: $d_1=45$ nm, and $d_2=190$ nm, the twist angle θ is set as 60° . The LCP (**b**) and RCP (**c**) transmission as a function of the gap length. (**d**) CD as a function of the gap length. The wavelength is set as $\lambda=600$ nm. **e**, CD as a function of wavelength and gap distance. **f**, CD as a function of different first layer thickness d_1 and wavelength, where d_2 is fixed at 190 nm.

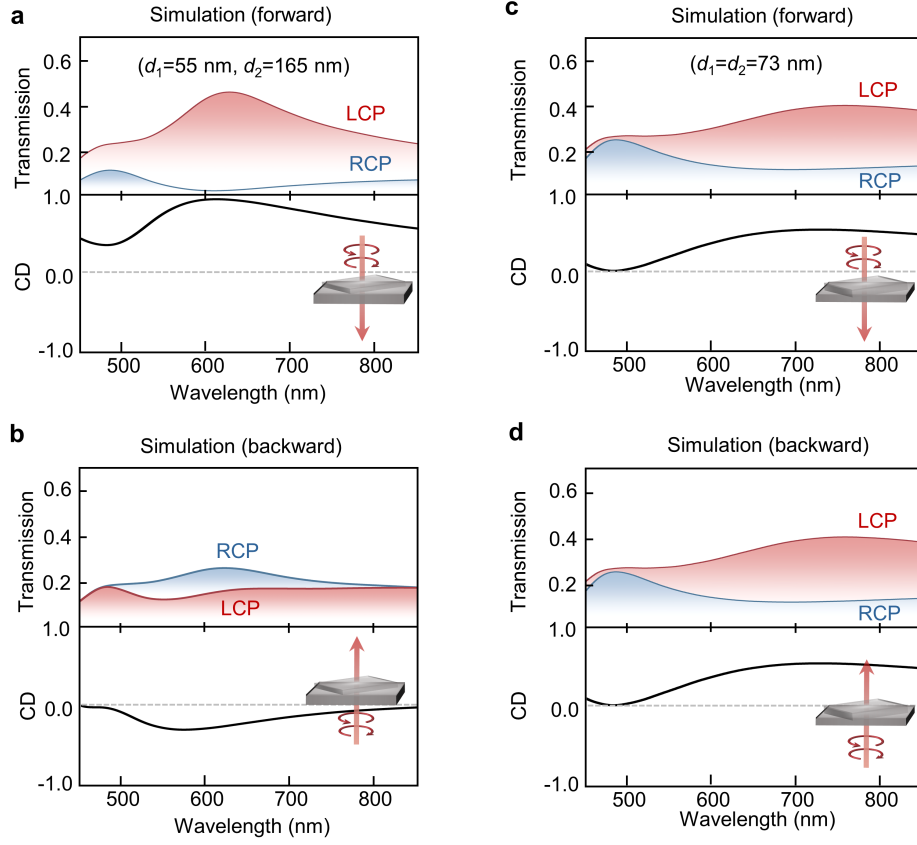


Fig. S17| Simulated apparent CD in two twisted MoOCl₂ homostructures. Simulated transmission spectra under LCP and RCP incidence (top panel), along with the corresponding CD spectra (bottom panel), for the twisted MoOCl₂ bilayers when illuminated from forward and backward direction. **(a, b)** is acquired for the sample ($d_1 = 55$ nm, $d_2 = 165$ nm), while **(c, d)** is obtained for the sample ($d_1 = d_2 = 73$ nm). The twist angle is fixed at $\theta = 60^\circ$.

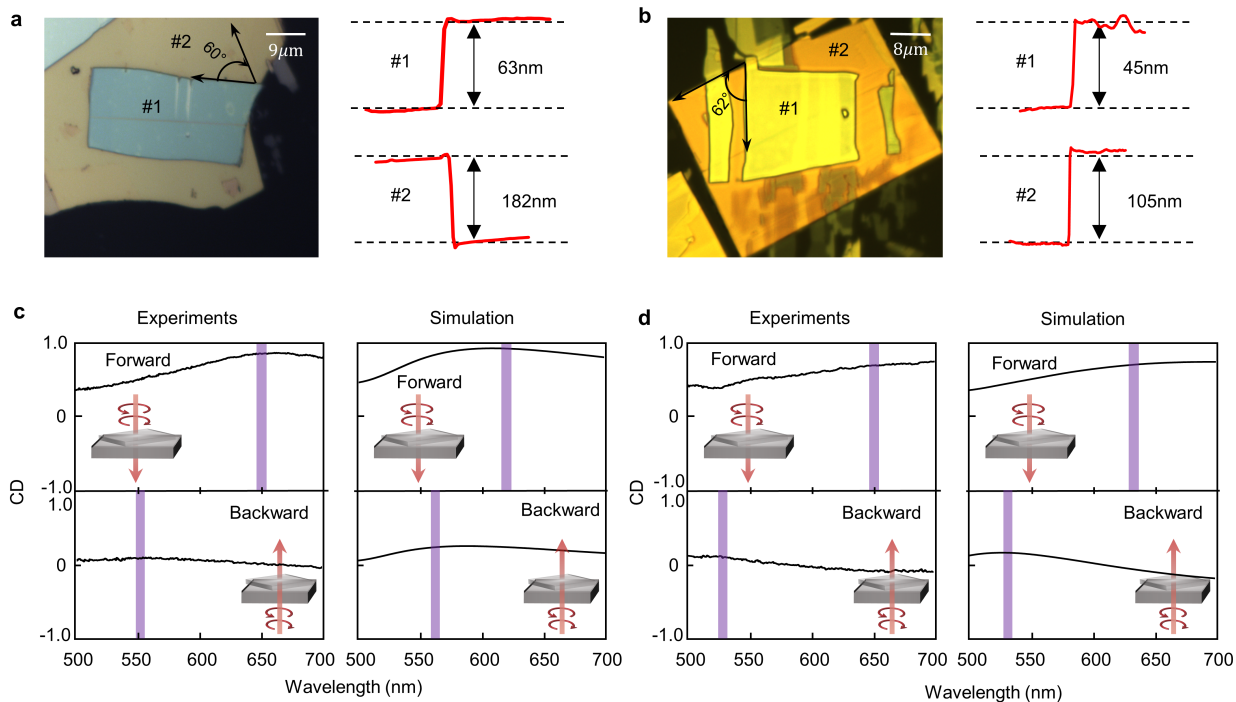


Fig. S18 | Apparent CD in two different twisted MoOCl₂ homostructures. **a, b,** Optical images of two twisted MoOCl₂ bilayer samples with $\theta = -60^\circ$ and 62° , respectively. The thicknesses of the MoOCl₂ layers are measured by AFM (left panel in **a** and **b**). **c, d,** Measured and simulated CD spectra for the twisted MoOCl₂ homostructures when illuminated from forward (top panel) and backward direction (bottom panel).

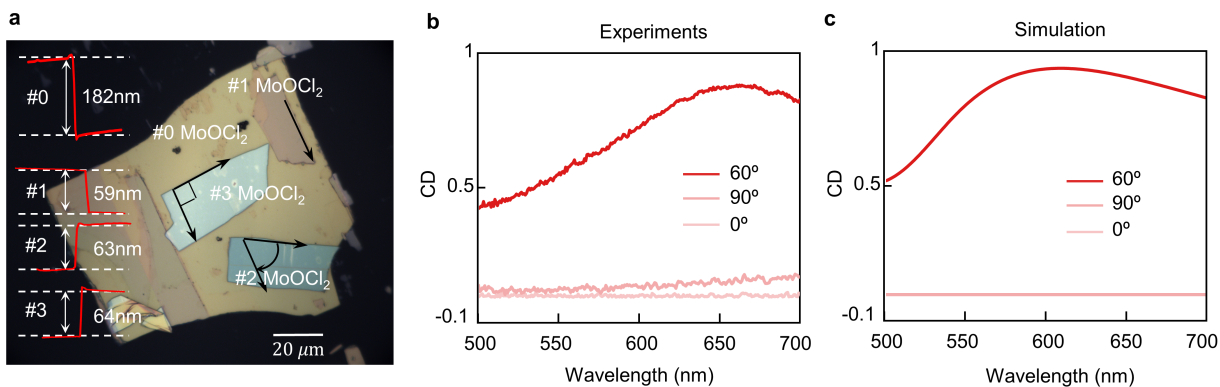


Fig. S19 | CD response at different twisted angle. **(a)** The optical image and AFM results of different twisted MoOCl₂ bilayer samples. The measured **(b)** and simulated **(c)** CD responses.

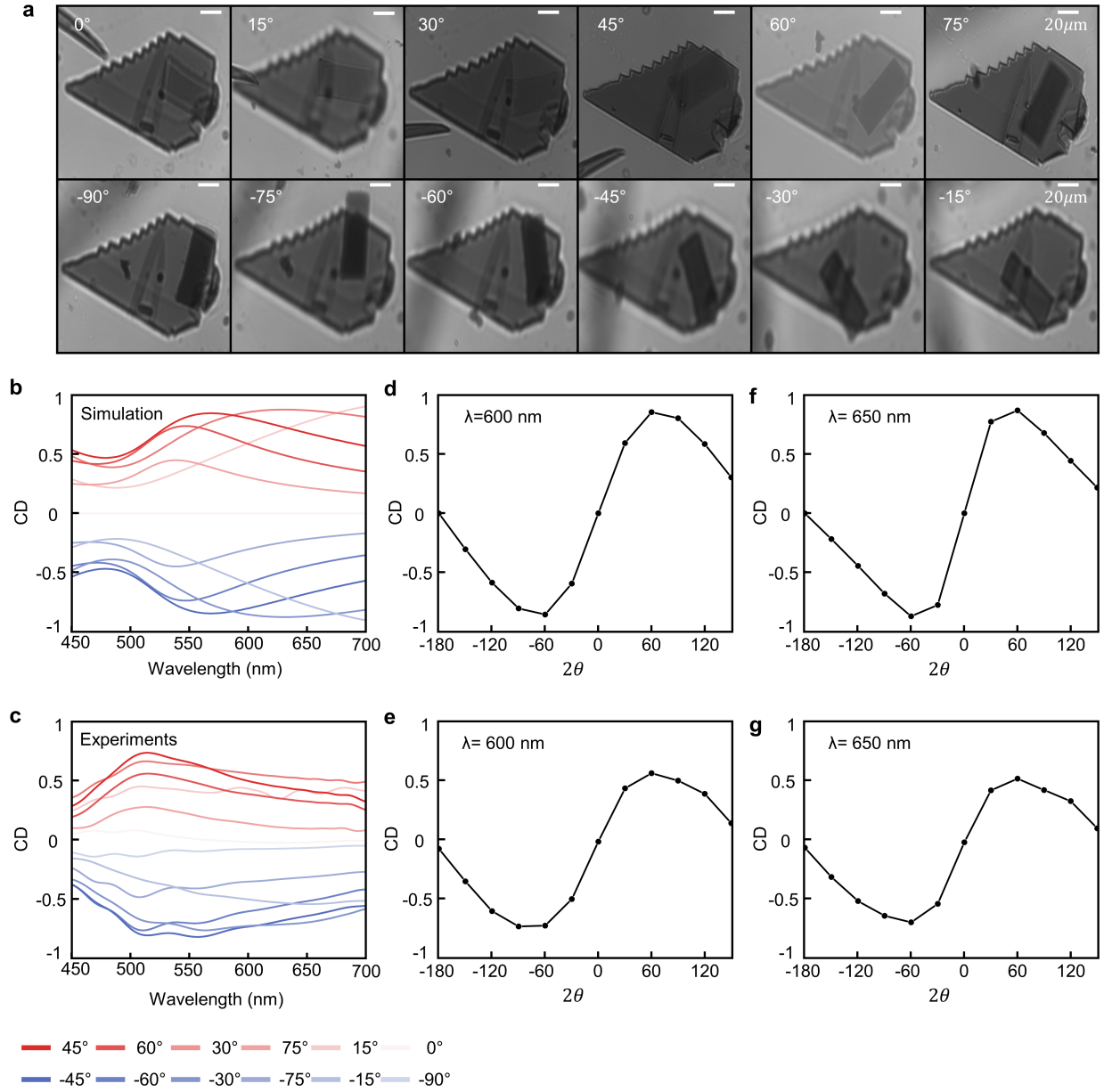


Fig. S20| Reconfiguring CD by the twisted angle. **a**, Optical images for different samples with twisted angles continuously from -90° to 90° at a 15° interval. **b**, **c** Simulated and measured CD spectra for different twisted angles. **d-g**, Simulated and measured CD values as a function of 2θ at incident wavelengths 600 nm and 650 nm, respectively. The layer thicknesses are $d_1 = 70\text{ nm}$, and $d_2 = 230\text{ nm}$.

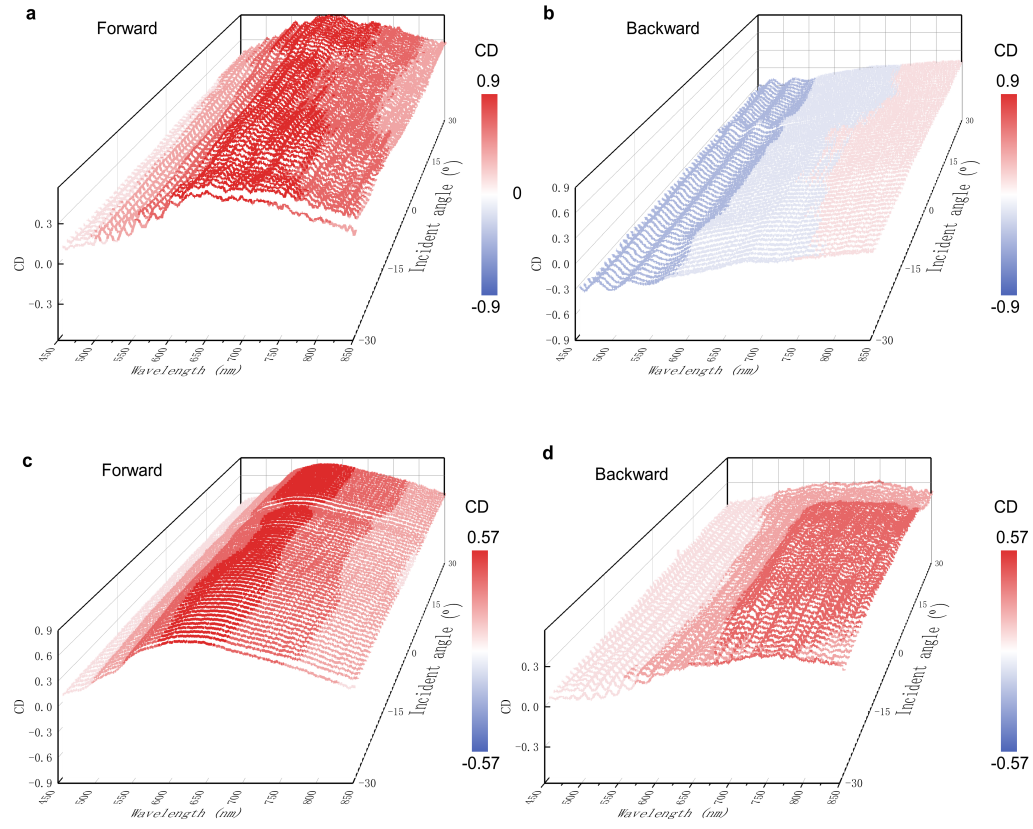


Fig. S21| Experimental angle-resolved CD spectra of the sample **(a, b)** $d_1 = 55$ nm, $d_2 = 165$ nm, and **(c, d)** $d_1 = d_2 = 73$ nm under forward and backward incidence.

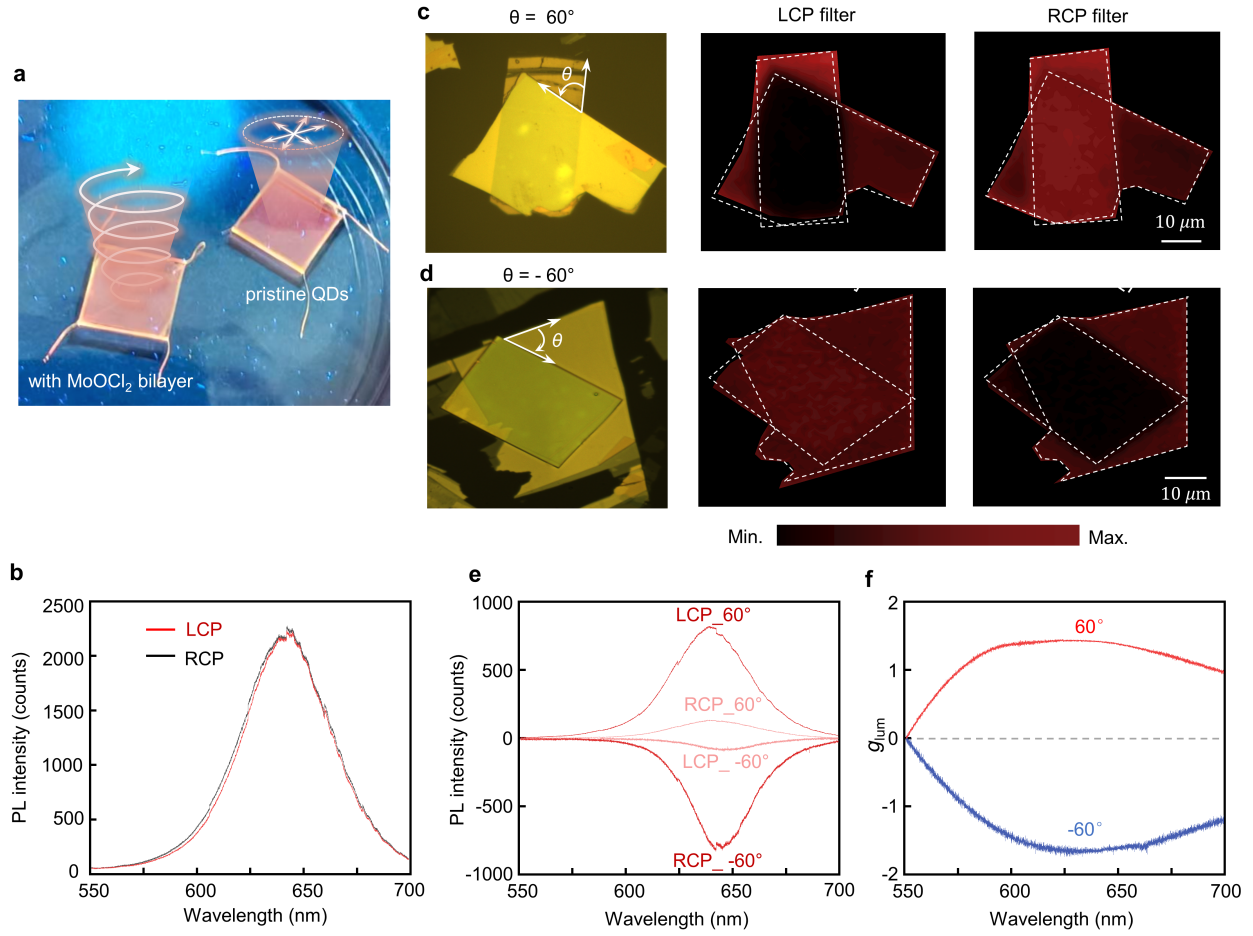


Fig. S22| Circular polarized emission enabled by sticking MoOCl₂ bilayers on QD films. a, Optical image of CsPbBr₂ perovskite quantum dot (QDs) sample with red light emission. **b**, Spin-resolved PL spectra measured from a QDs film, which is pumped by a 532-nm linearly polarized laser. **c, d**, Optical microscopy and PL images of two twisted MoOCl₂ bilayers stuck on QDs films, possessing twist angles of **(c)** $\theta = 60^\circ$ ($d_1 = 60$ nm and $d_2 = 165$ nm) and **(d)** $\theta = -60^\circ$ ($d_1 = 55$ nm and $d_2 = 160$ nm). The PL images are captured after a RCP (middle) and LCP (right) filter, respectively. **e**, Spin-resolved PL spectra measured from QDs films capped by twisted MoOCl₂ bilayers of $\theta = 60^\circ$ and -60° . **f**, Retrieved g_{lum} spectra of the three cases in **(b, e)**.

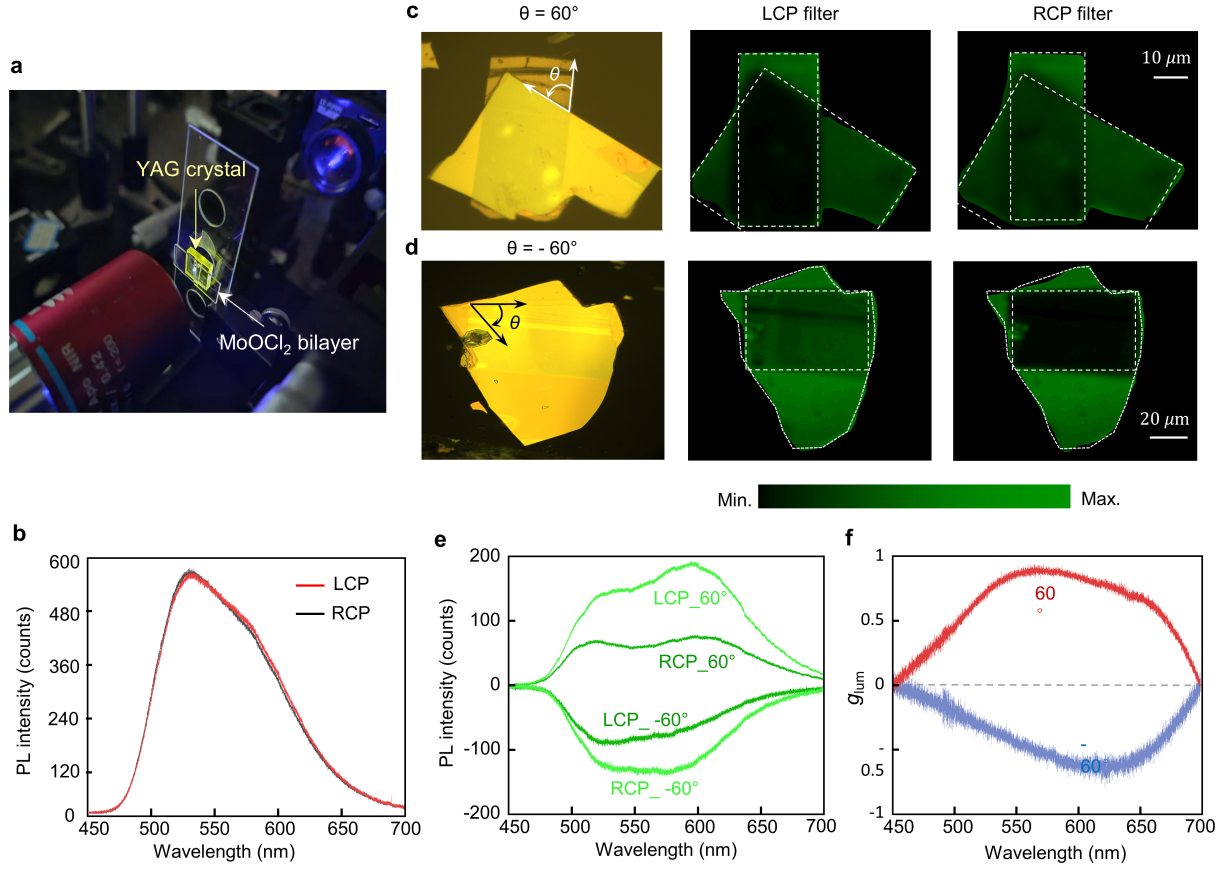


Fig. S23| Circular polarized emission enabled by sticking a MoOCl₂ bilayer on YAG crystal. **a**, Optical image of MoOCl₂ bilayer integrated with YAG:Ce crystals. **b**, Spin-resolved PL spectra measured from a bulk YAG:Ce crystal, which is pumped by a 405-nm linearly polarized laser. **c**, **d**, Optical microscopy and PL images of two twisted MoOCl₂ bilayers stuck on a YAG, possessing twist angles of (**c**) $\theta = 60^\circ$ ($d_1 = 60$ nm and $d_2 = 180$ nm) and (**d**) $\theta = -60^\circ$ ($d_1 = 55$ nm and $d_2 = 160$ nm). The PL images are captured after a RCP (middle) and LCP (right) filter, respectively. **e**, Spin-resolved PL spectra measured from YAG crystal capped by twisted MoOCl₂ bilayers of $\theta = 60^\circ$ and -60° . **f**, Retrieved g_{lum} spectra of the three cases in (**b**, **e**).

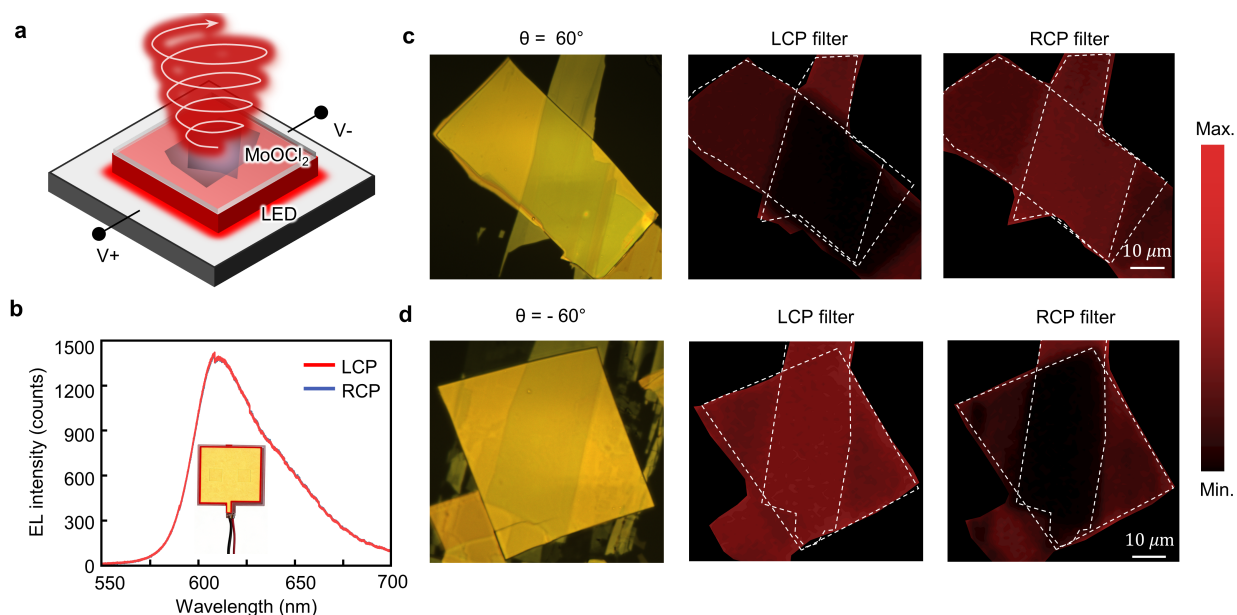


Fig. S24| Electrically pumped circular polarized emission from MoOCl_2 bilayer-LED heterostructure. **a**, Schematic of electrically pumped circular polarized emission of a commercial LED capped by two twisted MoOCl_2 bilayers. **b**, Spin-resolved EL spectra measured from a pristine LED panel. **c**, **d**, Optical microscopy and EL images of two twisted MoOCl_2 bilayers stuck on LED, possessing twist angles of (c) $\theta = 60^\circ$ ($d_1 = 48\ \text{nm}$ and $d_2 = 160\ \text{nm}$) and (d) $\theta = -60^\circ$ ($d_1 = 51\ \text{nm}$ and $d_2 = 181\ \text{nm}$). The EL images are captured after a RCP (middle) and LCP (right) filter, respectively.

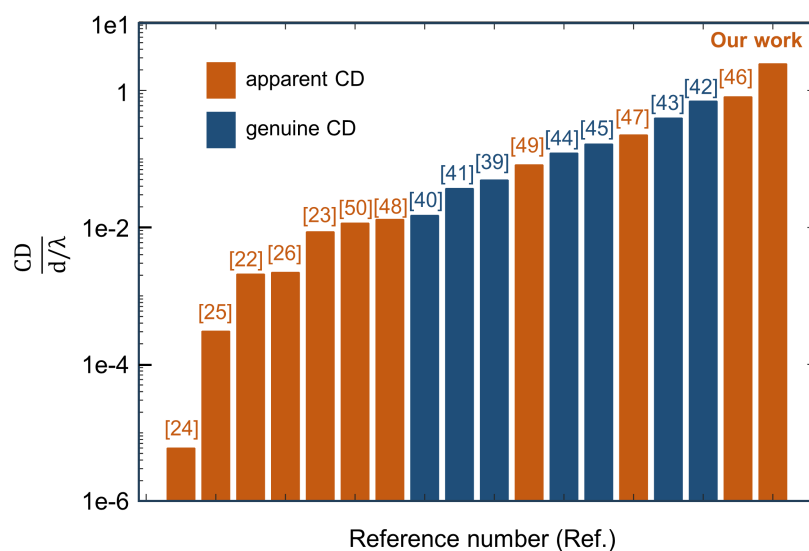


Fig. S25| Comparison of our work with some typical CD-active materials in terms of achievable CD value per unit normalized thickness [$\text{CD}/(\text{thickness}/\text{wavelength})$].

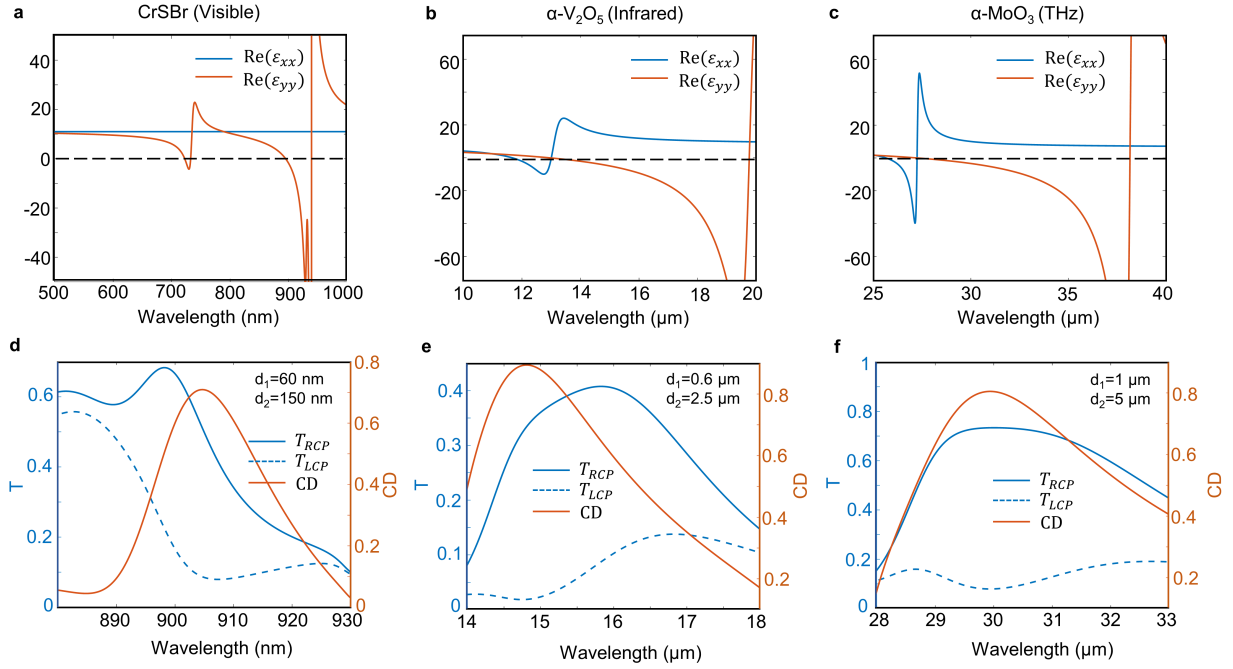


Fig. S26| Colossal CD response in different twisted hyperbolic bilayer. **a-c**, Dispersion of ϵ_{xx} and ϵ_{yy} for different hyperbolic materials. **d-f**, Transmission for LCP/RCP light and CD for each twisted bilayer hyperbolic material. Thickness for layers is labeled in each figure and the twist angle is 60° .

Table S1| Fitting parameters of the Drude–Lorentz model for a birefringent crystal (bound electron), and a hyperbolic metal (unbound free electron) in Fig. S3. The frequency unit is THz.

bound	ϵ_∞	$\omega_{p,0}$	γ_0	$\omega_{p,1}$	ω_1	γ_1
x	3.5	0	0	90	510	20
y	3	0	0	90	540	20
free	ϵ_∞	$\omega_{p,0}$	γ_0	$\omega_{p,1}$	ω_1	γ_1
x	4.5	450	50	0	0	0
y	1	600	50	0	0	0

Table S2| Summary of CD signals of twisted MoOCl₂ hyperbolic bilayer (our work) and other known nano-materials.

Reference	material	CD deg. /norm	thickness	wavelength	LB-LD or Genuine CD
Our work	MoOCl₂	42 / 0.90	220 nm	600 nm	LB LD
[22]	Oligothiophene	0.15 / 0.005	720 nm	300 nm	LB LD
[23]	Li ₂ Co ₃ (SeO ₃) ₄	5 / 0.17	12.5 μm	635 nm	LB LD
[24]	PET/PDMS+Ti ₃ C ₂ T _x	0.273 / 0.01	1 mm	600 nm	LB LD
[25]	PDMS+AuNSs	40 / 0.85	2.5 mm	900 nm	LB LD
[26]	CNC + AuNP	5 / 0.17	100 μm	1300 nm	LB LD
[46]	AuNDs	1 / 0.03	20 nm	540 nm	LB LD
[47]	(S/R-HP1A)PbBr ₃	2 / 0.07	130 nm	417 nm	LB LD
[48]	(R-EBA)PbI ₃	0.04 / 0.0014	40 nm	375 nm	LB LD
[49]	PTPO films	1.2/ 0.042	230 nm	450 nm	LB LD
[50]	CdTe/CdSe MSC	0.16 / 0.006	170 nm	328 nm	LB LD
[39]	triazatruxene	0.475 / 0.02	150 nm	370 nm	Genuine CD
[40]	PAA-b-PS / (R/S)- MA	1 / 0.03	500 nm	250 nm	Genuine CD
[41]	tellurium nanocrystals	0.1 / 0.0035	66 nm	700 nm	Genuine CD
[42]	L/D-dipeptide NP	2.6 / 0.09	90 nm	700 nm	Genuine CD
[43]	Fe ₃ O ₄ /Au	0.35 / 0.0122	20 nm	650 nm	Genuine CD
[44]	black phosphorus	1 / 0.03	160 nm	650 nm	Genuine CD
[45]	AuNP	1.25 / 0.044	200 nm	750 nm	Genuine CD

Table S3| Comparison of our work with the existing works on MoOCl₂.

Reference	Structure	Research field	Theoretical model	CD (Exp.)	Application demonstration
Nat. Comm. 2024, 15, 9727	Single layer	Near-field poalritonics	Hyperbolic dispersion analysis	No CD	No demo.
Nat. Comm. 2025,16, 6172	Single layer	Near-field poalritonics	Hyperbolic dispersion analysis	No CD	No demo.
ACS Nano 2025, 19, 25413	Single layer	Far-field	Only simulation	No CD	Optical fiber polarizer
npj 2D Mater Appl 2026, 10, 42	Twisted bilayer	Far-field	Transfer-matrix method	~0.4	No demo.
Nat. Comm. 2026, 17, 2716	Twisted nanostructure	Far-filed	Only simulation	~0.54	No demo.
Our work	Twisted bilayer	Far-field	Rigorous LB-LD framework	Reconfigurable -0.9 to 0.9	Circular polarized light emission devices

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