

Supplementary Information for Engineering shear polaritons in 2D twisted heterostructures

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1 Supplementary Note 1. The analytical solutions of hyperbolic shear polaritons (HShPs)

Generally, we assume the electric fields of two eigenmodes to be o, e in biaxial medium and s, p in isotropic medium, which are given by ¹:

$$|s\rangle_{\pm} = \frac{1}{q} \begin{pmatrix} -q_y \\ q_x \\ 0 \end{pmatrix} e^{ik_{s\pm}r} \quad (\text{S1})$$

$$|p\rangle_{\pm} = \frac{1}{q} \begin{pmatrix} q_x \\ q_y \\ \frac{q_z}{\mp iq_z} \end{pmatrix} e^{ik_{p\pm}r} \quad (\text{S2})$$

$$|o\rangle_{\pm} = \frac{1}{q} \begin{pmatrix} -q_y(1 - \Delta_1\Delta_z) \\ q_x \\ \mp iq_x q_y q_{oz} \Delta_1 \end{pmatrix} e^{ik_{o\pm}r} \quad (\text{S3})$$

$$|e\rangle_{\pm} = \frac{1}{q} \begin{pmatrix} q_x \frac{\Delta_2 - q_y^2}{\Delta_x^e} \\ q_y \\ \frac{\Delta_2}{\mp iq_{ez}} \end{pmatrix} e^{ik_{e\pm}r} \quad (\text{S4})$$

The parameters in these formulas are as follows:

$$q_{x,y} = \frac{k_{x,y}}{k_0}, q_z = \frac{-ik_z}{k_0}, q = \sqrt{q_x^2 + q_y^2} \quad (\text{S5})$$

where $k_{x,y,z}$ are wave vectors in the xyz directions, respectively. $q_{o,ez}$ is obtained via solving the

Maxwell's equations:

$$q_{o,e,z}^2 = \frac{1}{2} \left(\frac{\varepsilon_x + \varepsilon_z}{\varepsilon_z} q_x^2 + \frac{\varepsilon_y + \varepsilon_z}{\varepsilon_z} q_y^2 - (\varepsilon_x + \varepsilon_y) \right) \pm \frac{1}{2} \sqrt{D} \quad (\text{S6})$$

$$D = \left(\varepsilon_x - \varepsilon_y + \frac{\varepsilon_z - \varepsilon_x}{\varepsilon_z} q_x^2 - \frac{\varepsilon_z - \varepsilon_y}{\varepsilon_z} q_y^2 \right)^2 + 4 \frac{(\varepsilon_z - \varepsilon_x)(\varepsilon_z - \varepsilon_y)}{\varepsilon_z^2} q_x^2 q_y^2 \quad (\text{S7})$$

where $\varepsilon_{x,y,z}$ are the permittivity of biaxial materials in the xyz directions, respectively.

Our structure is depicted as a layered configuration of air/top α -MoO₃/bottom α -MoO₃/SiO₂, as illustrated in Supplementary Figure 1a. We establish a Cartesian coordinate system xyz , aligning the [100], [001], and [010] crystallographic orientations of the bottom layer α -MoO₃ to the x , y , and z axes, respectively. And the angle between the [100] crystallographic orientations of the top and bottom layers of α -MoO₃ is θ . Thus, the general electrical field vectors of top layer are acted by the rotation matrix $\mathbf{F}(\theta)$.

$$\mathbf{F}(\theta) = \begin{pmatrix} \cos \theta & -\sin \theta \\ \sin \theta & \cos \theta \end{pmatrix} \quad (\text{S8})$$

The general electrical field vectors of top layer transform as follows:

$$|y_2^r\rangle = \mathbf{F}(\theta) |y_2\rangle, y = o, e \quad (\text{S9})$$

$$|y_2^r\rangle'_{\pm} = \mathbf{F}(\theta) |y_2\rangle'_{\pm}, y = o, e \quad (\text{S10})$$

By applying boundary condition:

$$\begin{cases} n \times (\mathbf{E}_2 - \mathbf{E}_1) = 0 \\ n \times (\mathbf{H}_2 - \mathbf{H}_1) = \frac{4\pi}{c} \sigma \mathbf{E} \end{cases} \quad (\text{S11})$$

where $n = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}$. We have:

$$\begin{bmatrix} 1 & 0 & -\langle s|o_2^r \rangle \xi_{\text{stop}}^{o_2\uparrow} & -\langle s|o_2^r \rangle \xi_{\text{stop}}^{o_2\downarrow} & -\langle s|e_2^r \rangle \xi_{\text{stop}}^{e_2\uparrow} & -\langle s|e_2^r \rangle \xi_{\text{stop}}^{e_2\downarrow} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & -\langle p|o_2^r \rangle \xi_{\text{stop}}^{o_2\uparrow} & -\langle p|o_2^r \rangle \xi_{\text{stop}}^{o_2\downarrow} & -\langle p|e_2^r \rangle \xi_{\text{stop}}^{e_2\uparrow} & -\langle p|e_2^r \rangle \xi_{\text{stop}}^{e_2\downarrow} & 0 & 0 & 0 & 0 & 0 & 0 \\ Y_1^s & 0 & -\langle s|o_2^r \rangle'_+ \xi_{\text{stop}}^{o_2\uparrow} & -\langle s|o_2^r \rangle'_- \xi_{\text{stop}}^{o_2\downarrow} & -\langle s|e_2^r \rangle'_+ \xi_{\text{stop}}^{e_2\uparrow} & -\langle s|e_2^r \rangle'_- \xi_{\text{stop}}^{e_2\downarrow} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & Y_1^p & -\langle p|o_2^r \rangle'_+ \xi_{\text{stop}}^{o_2\uparrow} & -\langle p|o_2^r \rangle'_- \xi_{\text{stop}}^{o_2\downarrow} & -\langle p|e_2^r \rangle'_+ \xi_{\text{stop}}^{e_2\uparrow} & -\langle p|e_2^r \rangle'_- \xi_{\text{stop}}^{e_2\downarrow} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & -\langle y|o_2^r \rangle & -\langle y|o_2^r \rangle & -\langle y|e_2^r \rangle & -\langle y|e_2^r \rangle & \langle y|o_3 \rangle \xi_{\text{bot}}^{o_3\uparrow} & \langle y|o_3 \rangle \xi_{\text{bot}}^{o_3\downarrow} & \langle y|e_3 \rangle \xi_{\text{bot}}^{e_3\uparrow} & \langle y|e_3 \rangle \xi_{\text{bot}}^{e_3\downarrow} & 0 & 0 \\ 0 & 0 & -\langle x|o_2^r \rangle & -\langle x|o_2^r \rangle & -\langle x|e_2^r \rangle & -\langle x|e_2^r \rangle & \langle x|o_3 \rangle \xi_{\text{bot}}^{o_3\uparrow} & \langle x|o_3 \rangle \xi_{\text{bot}}^{o_3\downarrow} & \langle x|e_3 \rangle \xi_{\text{bot}}^{e_3\uparrow} & \langle x|e_3 \rangle \xi_{\text{bot}}^{e_3\downarrow} & 0 & 0 \\ 0 & 0 & -\langle y|o_2^r \rangle'_+ & -\langle y|o_2^r \rangle'_- & -\langle y|e_2^r \rangle'_+ & -\langle y|e_2^r \rangle'_- & \langle y|o_3 \rangle'_+ \xi_{\text{bot}}^{o_3\uparrow} & \langle y|o_3 \rangle'_- \xi_{\text{bot}}^{o_3\downarrow} & \langle y|e_3 \rangle'_+ \xi_{\text{bot}}^{e_3\uparrow} & \langle y|e_3 \rangle'_- \xi_{\text{bot}}^{e_3\downarrow} & 0 & 0 \\ 0 & 0 & -\langle x|o_2^r \rangle'_+ & -\langle x|o_2^r \rangle'_- & -\langle x|e_2^r \rangle'_+ & -\langle x|e_2^r \rangle'_- & \langle x|o_3 \rangle'_+ \xi_{\text{bot}}^{o_3\uparrow} & \langle x|o_3 \rangle'_- \xi_{\text{bot}}^{o_3\downarrow} & \langle x|e_3 \rangle'_+ \xi_{\text{bot}}^{e_3\uparrow} & \langle x|e_3 \rangle'_- \xi_{\text{bot}}^{e_3\downarrow} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & -\langle s|o_3 \rangle & -\langle s|o_3 \rangle & -\langle s|e_3 \rangle & -\langle s|e_3 \rangle & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & -\langle p|o_3 \rangle & -\langle p|o_3 \rangle & -\langle p|e_3 \rangle & -\langle p|e_3 \rangle & 0 & 1 \\ 0 & 0 & 0 & 0 & 0 & 0 & -\langle s|o_3 \rangle'_+ & -\langle s|o_3 \rangle'_- & -\langle s|e_3 \rangle'_+ & -\langle s|e_3 \rangle'_- & Y_s^4 + \frac{4\pi\sigma}{c} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & -\langle p|o_3 \rangle'_+ & -\langle p|o_3 \rangle'_- & -\langle p|e_3 \rangle'_+ & -\langle p|e_3 \rangle'_- & 0 & Y_p^4 + \frac{4\pi\sigma}{c} \end{bmatrix} \begin{bmatrix} \alpha_s^1 \\ \alpha_p^1 \\ \alpha_o^{2\uparrow} \\ \alpha_o^{2\downarrow} \\ \alpha_e^{2\uparrow} \\ \alpha_e^{2\downarrow} \\ \alpha_o^{3\uparrow} \\ \alpha_o^{3\downarrow} \\ \alpha_e^{3\uparrow} \\ \alpha_e^{3\downarrow} \\ \alpha_s^4 \\ \alpha_p^4 \end{bmatrix} = 0 \quad (\text{S12})$$

The parameters in the above secular equation are as follows:

$$\left\{ \begin{array}{l} |\beta\rangle = |\beta\rangle_{\pm, \text{sub}}, \beta = s, p, o, e \\ |o\rangle'_{\pm} = \mp i q_{oz} (|s\rangle + q_y \Delta_1 |a\rangle), |a\rangle = \frac{1}{q} \begin{pmatrix} \Delta_2 + q_x^2 \\ q_x q_y \end{pmatrix} \\ |e\rangle'_{\pm} = \frac{\Delta_2}{\mp i q_{ez}} |p\rangle \mp i q_{ez} |b\rangle, |b\rangle = \frac{1}{q} \begin{pmatrix} q_x \frac{\Delta_2 - q_y^2}{\Delta_x^e} \\ q_y \end{pmatrix} \\ \xi_{\text{bot}}^{o,e\uparrow\downarrow} = e^{\mp q_{o,e} k_0 d_2} \\ \xi_{\text{top}}^{o,e\uparrow\downarrow} = e^{\mp q_{o,e} k_0 d_1} \\ Y_s^{1,4} = \mp i q_{1,4z} \\ Y_p^{1,4} = \frac{\varepsilon_{1,4}}{\mp i q_{1,4z}} \\ \langle x| = \begin{pmatrix} 1 & 0 \end{pmatrix} e^{-i(k_x x + k_y y)} \\ \langle y| = \begin{pmatrix} 0 & 1 \end{pmatrix} e^{-i(k_x x + k_y y)} \end{array} \right. \quad (\text{S13})$$

where the parameters in the above equations are expressed as:

$$\Delta_1 = \frac{\Delta_x^o - q_x^2}{\Delta_z \Delta_x^o + q_x^2 q_{oz}^2} \quad (\text{S14})$$

$$\Delta_2 = \frac{\Delta_x^e \Delta_y^e - q_x^2 q_y^2}{\Delta_x^e - q_x^2} \quad (\text{S15})$$

$$\Delta_z = \varepsilon_z - q_x^2 - q_y^2 \quad (\text{S16})$$

$$\Delta_x^{o,e} = \varepsilon_x - q_y^2 + q_{o,e}^2 \quad (\text{S17})$$

$$\Delta_y^{o,e} = \varepsilon_y - q_x^2 + q_{o,e}^2 \quad (\text{S18})$$

For this linear equation $\mathbf{TA} = 0$ to have non-trivial solution, the matrix should be invertible:

$$\det(\mathbf{T}) = 0 \quad (\text{S19})$$

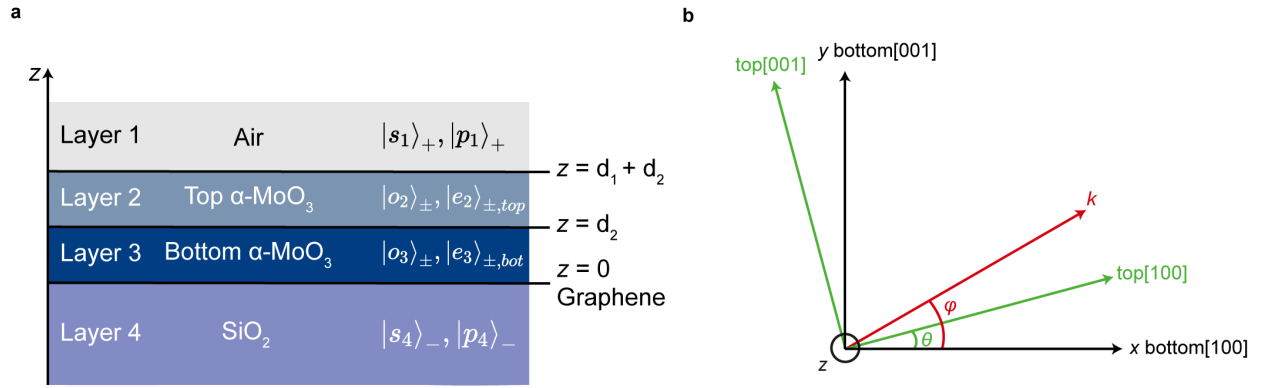
In order to visualize the relationship between the in-plane wavevector $k = \sqrt{k_x^2 + k_y^2}$ of HShPs and the engineering parameters, we assume the angle between the x axis and the direction of in-plane wavevector k is denoted as φ , as shown in Supplementary Figure 1b. Under the large momentum approximation, equation (S19) can be significantly simplified to:

$$k\rho(\varphi)d_2 + \arctan \frac{4\pi\sigma k - i\omega\varepsilon_{\text{sub}}}{i\omega\varepsilon_z\rho(\varphi)} - \arctan \frac{\rho(\varphi)}{\rho(\varphi - \theta)} \tan \left(\arctan \frac{\varepsilon_{\text{air}}}{\rho(\varphi - \theta)\varepsilon_z} - k\rho(\varphi - \theta)d_1 \right) + m\pi = 0 \quad (\text{S20})$$

For the condition without graphene, this equation (S20) simplifies to:

$$k\rho(\varphi)d_2 - \arctan \frac{\varepsilon_{\text{sub}}}{\varepsilon_z\rho(\varphi)} - \arctan \frac{\rho(\varphi)}{\rho(\varphi - \theta)} \tan \left(\arctan \frac{\varepsilon_{\text{air}}}{\rho(\varphi - \theta)\varepsilon_z} - k\rho(\varphi - \theta)d_1 \right) + m\pi = 0 \quad (\text{S21})$$

where $\rho(\varphi) = i\sqrt{\frac{\varepsilon_x \cos^2(\varphi) + \varepsilon_y \sin^2(\varphi)}{\varepsilon_z}}$, $\varepsilon_{x,y,z}$ are the permittivities of $\alpha\text{-MoO}_3$ in the [100], [001] and [010] crystallographic directions, respectively. ε_{sub} is the permittivity of substrate SiO_2 , σ is the conductivity of graphene and ε_{air} is the permittivity of air. d_1 and d_2 are the thicknesses of the top and bottom layers, respectively. In equation (S20), the in-plane wavevector k is determined by the twist angle θ , the thicknesses of the top and bottom layers d_1 and d_2 , and the conductivity of graphene σ . By manipulating these parameters, we can achieve the versatile engineering of HShPs.



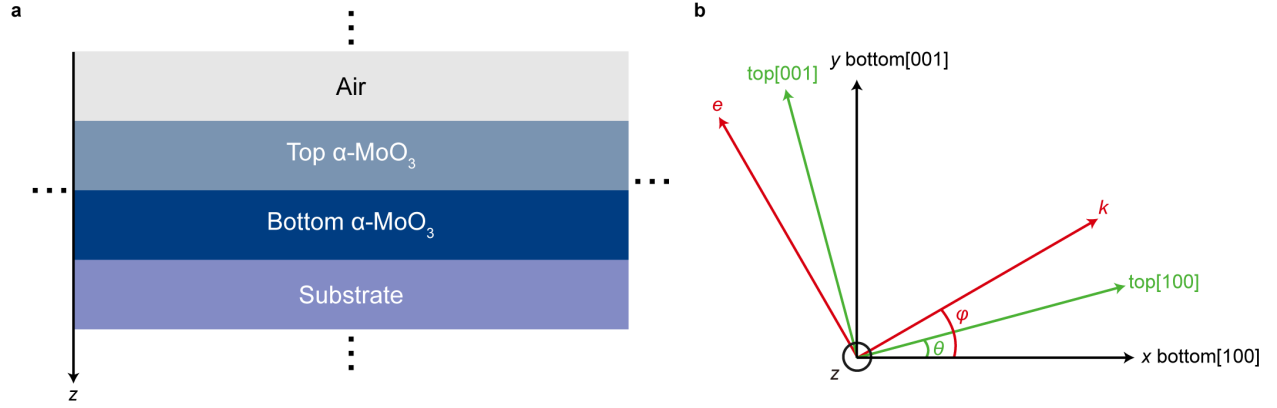
Supplementary Figure 1. Illustration of the structure and Cartesian coordinate systems used in analytical solutions. **a** The structure of the twisted bilayer $\alpha\text{-MoO}_3$ /graphene employed in the analytical solutions. From top to bottom, it represent air, top $\alpha\text{-MoO}_3$ layer, bottom $\alpha\text{-MoO}_3$ layer, graphene and SiO_2 , respectively. **b** The Cartesian coordinate systems utilized in the analytical solutions. Black, green, and red arrows correspond to the crystallographic orientations of the bottom layer $\alpha\text{-MoO}_3$, top layer $\alpha\text{-MoO}_3$, and the direction of the polariton wavevector in momentum space, respectively. θ represents the angle between the $[100]$ crystallographic orientations of the top and bottom layers of $\alpha\text{-MoO}_3$. φ indicates the angle between the k and x axes.

2 Supplementary Note 2. Calculation of the IFCs using mode expansion method

We employ the mode expansion method to streamline the computation of the IFCs of HShPs in twisted bilayer α -MoO₃². Mode expansion method is a well-established technique commonly utilized for characterizing modes in periodic structures. In our study, the twisted bilayer α -MoO₃ is represented by a multilayered structure extending infinitely along the $x - y$ plane. Thus, our model exhibits no periodicity in the real space. To adapt mode expansion method to our framework, we treat the multilayered structure as the fundamental unit and replicate it along the z direction to introduce periodicity a_z into our calculations. The structure unit is shown in Supplementary Figure 2a. Due to the localized nature of hyperbolic shear polariton modes along the z direction, we can disregard inter-unit coupling effects as long as the periodicity a_z is large enough ($a_z = 3000$ nm in our example), even in the absence of additional Perfectly Matched Layer (PML) in the z direction. Thus, the introduction of periodicity in the z direction serves only as a precondition for the application of this method and does not influence the final calculated results.

To simplify the problem, we establish a Cartesian coordinate system xyz , aligning the [100], [001], and [010] crystallographic orientations of the bottom layer α -MoO₃ to the x , y , and z axes, respectively. The angle between the [100] crystallographic orientations of the top and bottom layers of α -MoO₃ is θ . Meanwhile, we establish a Cartesian coordinate system kez in momentum space, in which the k axis represents the direction of the in-plane wavevector within the polariton. The angle between the x axis and the k axis is denoted as φ . The coordinate systems are shown in the Supplementary Figure 2b.

Initially, we normalize the magnetic field intensity $\mathbf{H}$ of the structure unit via the following



Supplementary Figure 2. Illustration of the unit structure and Cartesian coordinate systems used in mode expansion method. **a** The unit structure of the twisted bilayer α - MoO_3 employed in the mode expansion method. From top to bottom, it represent air, top α - MoO_3 layer, bottom α - MoO_3 layer, and SiO_2 , respectively. **b** The Cartesian coordinate systems utilized in the mode expansion method. Black, green, and red arrows correspond to the crystallographic orientations of the bottom layer α - MoO_3 , top layer α - MoO_3 , and the direction of the polariton wavevector in momentum space, respectively. θ represents the angle between the [100] crystallographic orientations of the top and bottom layers of α - MoO_3 . φ indicates the angle between the k and x axes.

equation:

$$\tilde{\mathbf{H}} = i\sqrt{\frac{\mu_0}{\varepsilon_0}}\mathbf{H} \quad (\text{S22})$$

where ε_0 represents the permittivity of vacuum, μ_0 represents the permeability of vacuum. In the derivation of the following equations, we assume that the material is non-magnetic, with $\mu \equiv 1$. In the kez coordinate system, we can employ Bloch theorem and expand the normalized magnetic field intensity $\tilde{\mathbf{H}}$ in the equation S22 along the z axis as follows:

$$\tilde{\mathbf{H}} = e^{ik_{\parallel} \cdot r} \sum_G \tilde{h}_G e^{i(k_z + G) \cdot z} \quad (\text{S23})$$

$$\mathbf{E} = e^{ik_{\parallel} \cdot r} \sum_G e_G e^{i(k_z + G) \cdot z} \quad (\text{S24})$$

where $k_{\parallel}$ and k_z represent the in-plane and out-of-plane Bloch wavevector of polaritons in momentum space, respectively. $G = n2\pi/a_z$ is reciprocal lattice vector, where n is an integer number. $\tilde{h}_G$ and e_G are normalized magnetic and electric expansion coefficients, respectively. The Maxwell's equations in the structure unit are:

$$\nabla \times \tilde{\mathbf{H}} = -k_0 \varepsilon \mathbf{E} \quad (\text{S25})$$

$$\nabla \times \mathbf{E} = k_0 \tilde{\mathbf{H}} \quad (\text{S26})$$

where k_0 is the wavevector of the excitation light in vacuum, ε is the permittivity tensor of materials. According to the coordinate transformations between the kez and xyz coordinate systems, the permittivity tensors of the bottom layer $\alpha\text{-MoO}_3$ in the kez and xyz coordinate systems, denoted as $\varepsilon_{kez, \text{bottom}}$ and $\varepsilon_{xyz, \text{bottom}}$ respectively, are related as follows:

$$[\varepsilon_{kez, \text{bottom}}] = \mathbf{F}_{\text{bottom}}^{-1}(\varphi) [\varepsilon_{xyz, \text{bottom}}] \mathbf{F}_{\text{bottom}}(\varphi), \mathbf{F}_{\text{bottom}} = \begin{pmatrix} \cos \varphi & -\sin \varphi & 0 \\ \sin \varphi & \cos \varphi & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad (\text{S27})$$

The permittivity tensors of the top layer $\alpha\text{-MoO}_3$, denoted as $\varepsilon_{kez, \text{top}}$ and $\varepsilon_{xyz, \text{top}}$ respectively, are related as follows:

$$[\varepsilon_{kez, \text{top}}] = \mathbf{F}_{\text{top}}^{-1}(\varphi - \theta) [\varepsilon_{xyz, \text{top}}] \mathbf{F}_{\text{top}}(\varphi - \theta), \mathbf{F}_{\text{top}} = \begin{pmatrix} \cos(\varphi - \theta) & -\sin(\varphi - \theta) & 0 \\ \sin(\varphi - \theta) & \cos(\varphi - \theta) & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad (\text{S28})$$

where $\mathbf{F}_{\text{bottom}}$ and $\mathbf{F}_{\text{top}}$ are rotation matrices. The isotropic material exhibits rotational symmetry, thus its permittivity tensor remains the same under coordinate transformation between two coordinate systems. In the kez coordinate system, substituting equation (S23-24) into equation (S25-26) yields:

$$\begin{bmatrix} \mathbf{O} & \vdots & \mathbf{I} \\ & \dots & \\ & & \mathbf{P} \end{bmatrix} \begin{bmatrix} \mathbf{E}_e \\ \mathbf{E}_z \\ \widetilde{\mathbf{k}}_k \mathbf{E}_e \\ \widetilde{\mathbf{k}}_k \mathbf{E}_z \end{bmatrix} = \widetilde{\mathbf{k}}_k \begin{bmatrix} \mathbf{E}_e \\ \mathbf{E}_z \\ \widetilde{\mathbf{k}}_k \mathbf{E}_e \\ \widetilde{\mathbf{k}}_k \mathbf{E}_z \end{bmatrix} \quad (\text{S29})$$

where $\mathbf{O}$ and $\mathbf{I}$ are the zero and identity matrix, respectively. $\widetilde{\mathbf{k}}_k = \mathbf{k}_k/\mathbf{k}_0$ is eigenvalue, and the

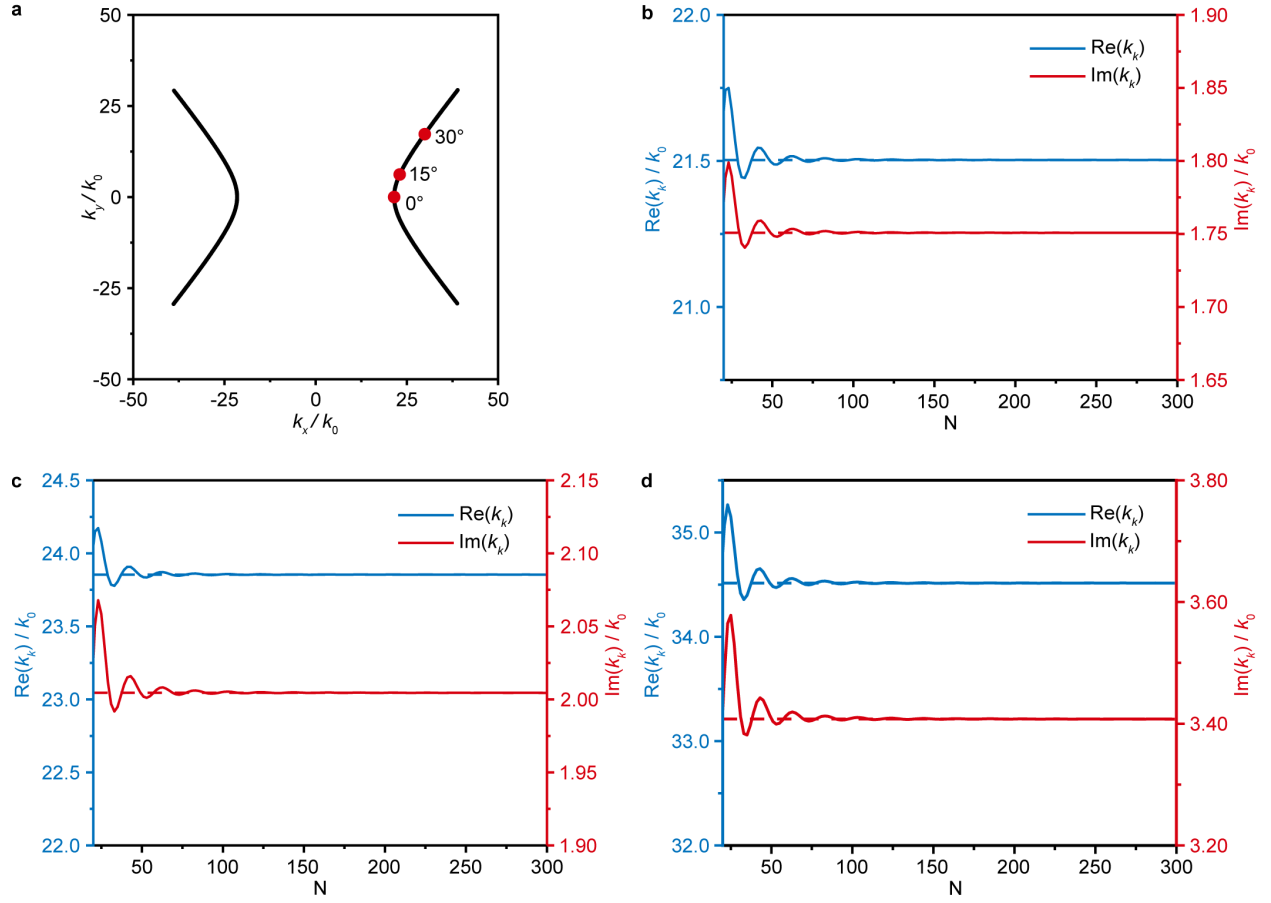
order of eigenfunction $\begin{bmatrix} \mathbf{E}_e \\ \mathbf{E}_z \\ \widetilde{\mathbf{k}}_k \mathbf{E}_e \\ \widetilde{\mathbf{k}}_k \mathbf{E}_z \end{bmatrix}$ is $4N$. N is the number of the set $\{G\}$.

$$\mathbf{P} = \mathbf{M}^{-1} \left(\mathbf{B}_1 \mathbf{S} \left(\mathbf{Q} : \mathbf{D} \right) + \left(\mathbf{B}_2 : \mathbf{C}_1 \mathbf{S}^{-1} \mathbf{Q} \right) \right) \quad (\text{S30})$$

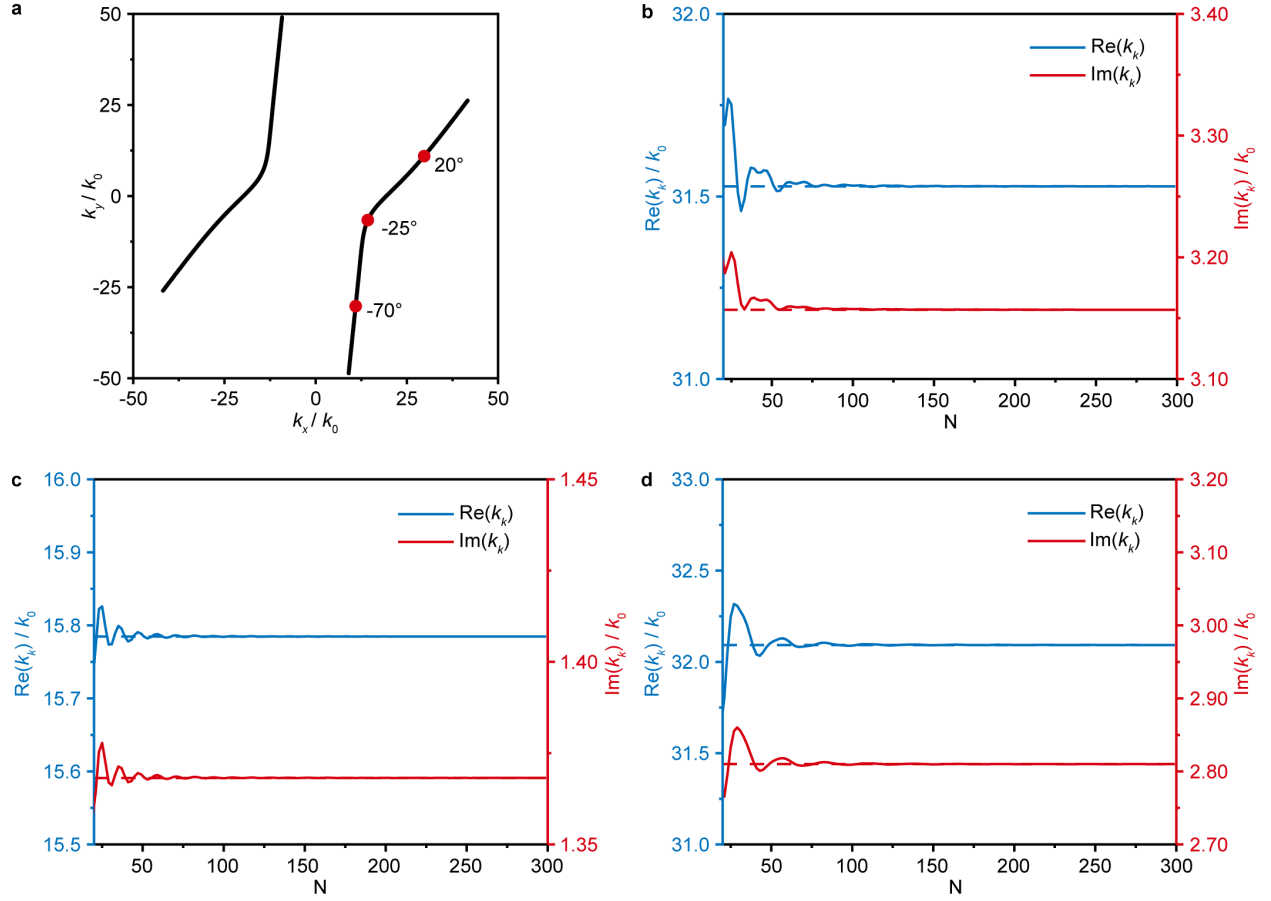
$$\left\{ \begin{array}{l} \mathbf{M} = (\mathbf{I} - \mathbf{C}_1 \mathbf{S}^{-1} \mathbf{D})^{-1} \\ \mathbf{C}_1 = \begin{pmatrix} \widetilde{(\mathbf{k}_z + \mathbf{G})} \\ \widetilde{\mathbf{k}_e} \end{pmatrix} \\ \mathbf{D} = \begin{pmatrix} \widetilde{\mathbf{k}_e} & \widetilde{(\mathbf{k}_z + \mathbf{G})} \end{pmatrix} \\ \mathbf{S} = \widetilde{(\mathbf{k}_z + \mathbf{G})}(\mathbf{k}_z + \mathbf{G}) + \widetilde{\mathbf{k}_e} \widetilde{\mathbf{k}_e} - [\varepsilon_{kk}] \\ \mathbf{Q} = \begin{pmatrix} [\varepsilon_{ke}] & \mathbf{O} \end{pmatrix} \\ \mathbf{B}_1 = \begin{pmatrix} \mathbf{O} \\ [\varepsilon_{ek}] \end{pmatrix} \\ \mathbf{B}_2 = \begin{pmatrix} \widetilde{\mathbf{k}_e}(\mathbf{k}_z + \mathbf{G}) & [\varepsilon_z^{-1}]^{-1} - \widetilde{\mathbf{k}_e} \widetilde{\mathbf{k}_e} \\ [\varepsilon_{ee}] - \widetilde{(\mathbf{k}_z + \mathbf{G})}(\mathbf{k}_z + \mathbf{G}) & \widetilde{(\mathbf{k}_z + \mathbf{G})} \widetilde{\mathbf{k}_e} \end{pmatrix} \end{array} \right. \quad (\text{S31})$$

where $[\varepsilon_z^{-1}]^{-1}$ is Toeplitz matrix, to which we apply inverse rule in order to improve the convergence of the calculation. By solving the eigen matrix $\begin{bmatrix} \mathbf{O} & \vdots & \mathbf{I} \\ \dots & & \\ \mathbf{P} & & \end{bmatrix}$ for its eigenvalues through equation (S29), we can obtain the in-plane wavevector $\mathbf{k}_k$ in any direction at a certain frequency, and consequently derive the IFCs of HShPs.

To validate the accuracy of mode expansion method calculations for IFCs, we compare the computed results with those obtained from analytical solutions. For both single layer and twisted bilayer $\alpha\text{-MoO}_3$, all results obtained through the mode expansion method demonstrate high convergence consistent with the results from analytical solutions, as illustrated in Supplementary Figure 3 and Supplementary Figure 4.



Supplementary Figure 3. Calculation of IFCs for monolayer $\alpha\text{-MoO}_3$. **a** Calculated IFC of monolayer $\alpha\text{-MoO}_3$ with a thickness of 200 nm. The red dots indicate the momenta and their angles used for assessing convergence. **b-d** Convergence calculations at momentum angles of **(b)** 0°, **(c)** 15°, and **(d)** 30°. N is the expansion order. The blue and red lines represent the real and imaginary parts of the momentum, respectively. The solid lines denote the results computed by mode expansion method under different N , while the dashed lines represent the results obtained from analytical solutions. As N increases, the results calculated by the mode expansion method gradually converge to those of the analytical solutions. During the calculation process, we assume that the directions of the real and imaginary parts of the momentum are consistent.



Supplementary Figure 4. Calculation of IFCs for twisted bilayer α -MoO₃. **a** Calculated IFC of twisted bilayer α -MoO₃ with top and bottom layer thicknesses of 150 nm, 200 nm, respectively. And twist angle is 45° . The red dots indicate the momenta and their angles used for assessing convergence. **b-d** Convergence calculations at momentum angles of **(b)** 20° , **(c)** -25° , and **(d)** -70° . N is the expansion order. The blue and red lines represent the real and imaginary parts of the momentum, respectively. The solid lines denote the results computed by mode expansion method under different N , while the dashed lines represent the results obtained from analytical solutions. As N increases, the results calculated by the mode expansion method gradually converge to those of the analytical solutions. During the calculation process, we assume that the directions of the real and imaginary parts of the momentum are consistent.

3 Supplementary Note 3. Calculation of the reciprocal space maps using transfer matrix method (TMM)

We utilize the TMM as numerical approach to compute the reciprocal space maps of HShPs.³ In our theoretical framework, we model the twisted bilayer α -MoO₃ as a heterostructure which is homogenous in the x and y directions and comprise four layers in the z direction (Supplementary Figure 5a). In a similar manner, we model the twisted bilayer α -MoO₃/graphene heterostructure (Supplementary Figure 5b). In the set up of our computations, we model graphene layer as a 2D conductive sheet in TMM, where the boundary condition should be $n \times (\mathbf{H}_2 - \mathbf{H}_1) = \mathbf{J}$. The visualization of polariton dispersion in reciprocal space maps is achieved through the calculation of the imaginary component of the reflection coefficient $\text{Im}(r_{pp})$ for p -polarized incident beams.



Supplementary Figure 5. Illustration of the structures used in transfer matrix method. **a** Layered structure of the twisted bilayer $\alpha\text{-MoO}_3$. From top to bottom, it represent air, top $\alpha\text{-MoO}_3$ layer, bottom $\alpha\text{-MoO}_3$ layer, and SiO_2 , respectively. **b** Layered structure of the twisted bilayer $\alpha\text{-MoO}_3$ /graphene. From top to bottom, it represent air, top $\alpha\text{-MoO}_3$ layer, bottom $\alpha\text{-MoO}_3$ layer, graphene, and SiO_2 , respectively.

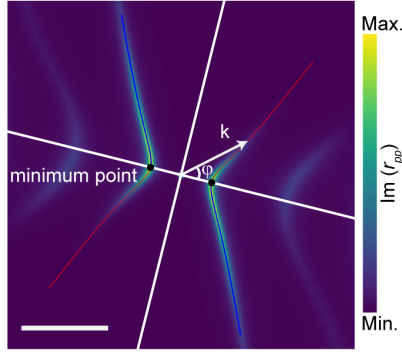
4 Supplementary Note 4. Definition and Calculation of shear factor

To quantify the shear effect of HShPs in relation to the thicknesses and twist angles of the top and bottom layers, we define and calculate the shear factor of HShPs. First, we calculate the IFCs of HShPs through analytical solutions and mode expansion method. Subsequently, by considering the calculated results (k, φ) as a function f , we can determine $k' \in k$ as a global minimum point of function f , where k represents the real part of the wavevector and φ represents the angle of the wavevector. Following this, we connect the two global minimum points symmetric with respect to the origin with a straight line, and then draw a second line perpendicular to the first. Thus, the IFCs of HShPs are divided into red part and blue part, as shown in Supplementary Figure 6.

We evaluate shear factor by conducting line integrals along the path of IFCs of HShPs on the momentum space obtained through analytical calculations, simulations, or experiments, respectively, where the shear factor is defined as follows:

$$\text{Shear factor} = \left| \frac{I_{\text{blue}} - I_{\text{red}}}{I_{\text{blue}} + I_{\text{red}}} \right| \quad (\text{S32})$$

where I_{red} and I_{blue} are line integral intensities along red and blue paths, respectively. The integration limits are defined starting from the global minimum points along the path, extending until the shear factor converges to within 0.01. This ensures that the calculated shear factor achieves a precision of two decimal places.



Supplementary Figure 6. The illustration for shear factor calculation. The IFCs of twisted bilayer α -MoO₃ calculated by TMM with top and bottom layer thicknesses of 130 nm, 200 nm, respectively. And twist angle is 30°. The black dots are global minimum points. The white lines divide the IFCs into four parts. The red and blue curves are line integral paths. k represents the real part of the wavevector and φ represents the angle of the wavevector. The scale bar is $40k_0$.

5 Supplementary Note 5. Optical parameters of graphene and α -MoO₃ and numerical simulations

In our work, the conductivity σ of graphene is determined using the non-approximate Kubo formula and consists of intra-band and inter-band contributions, which reads:⁴

$$\sigma(\omega, \tau, E_F, T) = \sigma_{\text{intra}}(\omega, \tau, E_F, T) + \sigma_{\text{inter}}(\omega, \tau, E_F, T) \quad (\text{S33})$$

$$\sigma_{\text{intra}}(\omega, \tau, E_F, T) = \frac{-ie^2}{\pi\hbar^2(\omega + \frac{i}{\tau})} \int_0^\infty \xi \left(\frac{\partial f_d(\xi)}{\partial \xi} - \frac{\partial f_d(-\xi)}{\partial \xi} \right) d\xi \quad (\text{S34})$$

$$\sigma_{\text{inter}}(\omega, \tau, E_F, T) = \frac{ie^2(\omega + \frac{i}{\tau})}{\pi\hbar^2} \int_0^\infty \frac{f_d(-\xi) - f_d(\xi)}{(\omega + \frac{i}{\tau})^2 - 4(\frac{\xi}{\hbar})^2} d\xi \quad (\text{S35})$$

The conductivity σ depends on the light frequency ω , the Fermi energy E_F , temperature T , and the relaxation time $\tau = \mu E_F / e v_F^2$, where μ is the carrier mobility. Here, $\hbar$ is the reduced Plank constant, e is the electron charge, $f_d(\xi) \equiv 1/(e^{(\xi - E_F)/(k_B T)} + 1)$ is the Fermi-Dirac distribution, and k_B is Boltzmann constant. Supplementary Figure 7a illustrates the variation of the real and

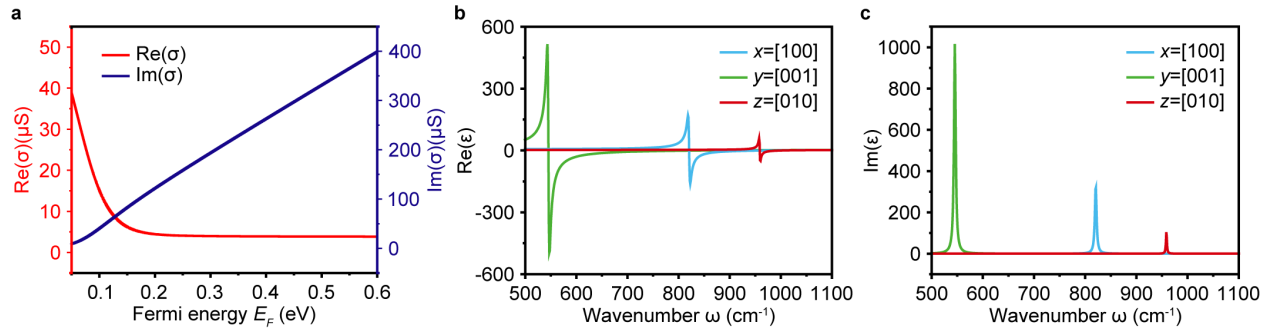
imaginary components of graphene conductivity σ as a function of E_F .

The permittivity of α -MoO₃ can be described by the Lorentz model:

$$\varepsilon_j(\omega) = \varepsilon_j^\infty \left(1 + \frac{\omega_j^{\text{LO}^2} - \omega_j^{\text{TO}^2}}{\omega_j^{\text{TO}^2} - \omega^2 - i\omega\Gamma_j} \right), \quad j = x, y, z \quad (\text{S36})$$

where $\varepsilon_j(\omega)$ represents the principal components of the permittivity tensor $\bar{\bar{\varepsilon}}_{\text{MoO}_3}$ at the light frequency ω . The parameters ε_j^∞ denote the high-frequency dielectric constants, while ω_j^{LO} and ω_j^{TO} are the longitude optical (LO) and transverse optical (TO) phonon frequencies, respectively. Additionally, Γ_j is the damping constant of the material. The x , y , and z axes correspond to the crystallographic orientations [100], [001], and [010] of α -MoO₃, respectively. These parameter values in our work are taken from the literature.⁵ Supplementary Figure 7b, c illustrate the real and imaginary parts of $\varepsilon_j(\omega)$, respectively.

Real-space full-wave simulations of HShPs were performed employing the finite-difference time-domain (FDTD) method, incorporating the aforementioned parameters, and utilizing the commercially available Lumerical FDTD software (version 2020b, <http://www.lumerical.com/tcad-products/fdtd/>). The simulated real-space nanoimages are detailed in Supplementary Figure 9-10 and Supplementary Figure 13. The parameter values used for α -MoO₃ and graphene are presented in Supplementary Tables 1 and 2, respectively. Specific simulation settings are detailed in Supplementary Table 3, where the plane wave is composed of two plane wave sources with perpendicular polarizations to simulate natural light.



Supplementary Figure 7. Optical parameters of graphene and α -MoO₃. **a** Real (red curve) and imaginary (blue curve) parts of graphene conductivity σ as a function of Fermi energy E_F at a frequency of 931 cm⁻¹. **b, c** Real (**b**) and imaginary (**c**) parts of the permittivity of α -MoO₃ along different crystal directions.

Supplementary Table 1: Parameters of α -MoO₃

Orientation	ϵ_∞	ω_{LO} (cm ⁻¹)	ω_{TO} (cm ⁻¹)	γ
[100]	4	972	820	4
[001]	5.2	851	545	4
[010]	2.4	1004	958	2

Supplementary Table 2: Parameters of graphene

Parameter Name	Parameter
Reduced Plank Constant	1.05457×10^{-34} J · s
Boltzmann Constant	1.380649×10^{-23} J/K
Temperature	300 K
Fermi Velocity	1×10^6 m/s

Supplementary Table 3: FDTD simulation parameters

Parameter Name	Parameter
Source	Plane Wave or Dipole
Frequency	931 cm ⁻¹
Temperature	300 K
Boundary Conditions	PML
Mesh Step	20 nm on all dimensions
Simulation Time	20000 fs

6 Supplementary Note 6. Fitting equations of HShPs and Calculation of quality factor Q

In order to better reflect the engineering effects of HShPs, the quality factor Q is defined:

$$Q = \frac{\text{Re}(k)}{\text{Im}(k)} = \frac{1}{\gamma_p} \quad (\text{S37})$$

where k is the in-plane momentum components and γ_p is dissipation rate of HShPs. For deriving γ_p from experimental results, we utilize the following equation ⁶:

$$I_{\text{NF}} = A_p \cdot \frac{e^{-\frac{2\pi\gamma_p}{\lambda_p}x}}{\sqrt{x}} \cdot \sin\left(2\pi\frac{x-x_c}{\lambda_p}\right) + B \cdot \frac{e^{-\frac{4\pi\gamma_p}{\lambda_p}x}}{x} + I_0, A_p > 0, \gamma_p > 0, \lambda_p > 0, B > 0 \quad (\text{S38})$$

where I_{NF} is the intensity of IR s -SNOM line profiles, A_p , γ_p , λ_p , x_c , B and I_0 are fitting parameters. For simulated results, we use the fitting equation:

$$y = A_p \cdot \frac{e^{-\frac{4\pi\gamma_p}{\lambda_p}x}}{\sqrt{x}} \cdot \sin\left(2\pi\frac{x-x_c}{\lambda_p}\right) + y_0, A_p > 0, \gamma_p > 0, \lambda_p > 0 \quad (\text{S39})$$

where A_p , γ_p , λ_p , x_c and y_0 are fitting parameters.

7 Supplementary Note 7. Calculation of Fermi energy E_F from gate voltage V_g

Utilizing gate-dependent electronic transport measurements, we have successfully ascertained the precise Fermi energy E_F of graphene within our experimental setups. Initially, the precise determination of the charge neutrality point (CNP) is important for accurately calculating and determining E_F . Our electric transport experiments yield the graphene resistance curve as a function of back-gate voltage V_g , with the peak value of the curve during the gate-tuning near-field experiment (Supplementary Figure 8). It's noteworthy that when E_F aligns with the CNP, the carrier density in graphene is minimal, resulting in maximum resistance, corresponding to the peak on the resistance curve. Furthermore, our devices are carefully maintained in a controlled dry air environment to minimize any potential influence of atmospheric moisture during electric transport.

The Fermi energy E_F in graphene can be controlled by adjusting the carrier density n . The relationship between E_F and n in graphene is:

$$E_F = \hbar |v_F| \sqrt{\pi n} \quad (\text{S40})$$

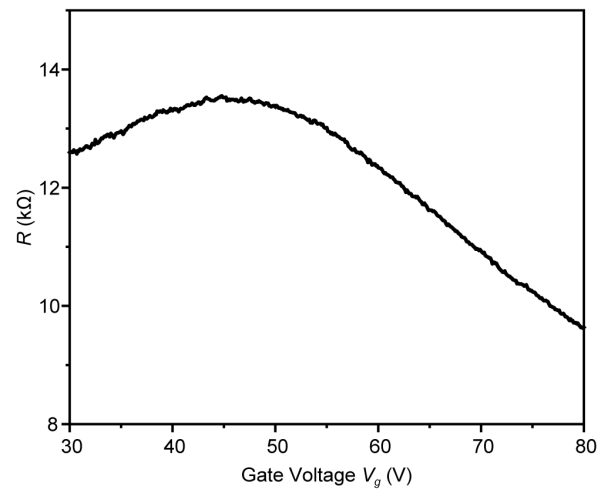
where $v_F = 1.1 \times 10^6$ m/s is the Fermi velocity of graphene, and $\hbar$ is the reduced Planck's constant. For our electric devices, the relationship between E_F and the back-gate voltage V_g is:⁷

$$V_g - V_{\text{CNP}} = \frac{E_F}{e} + \frac{ne}{C_g} \quad (\text{S41})$$

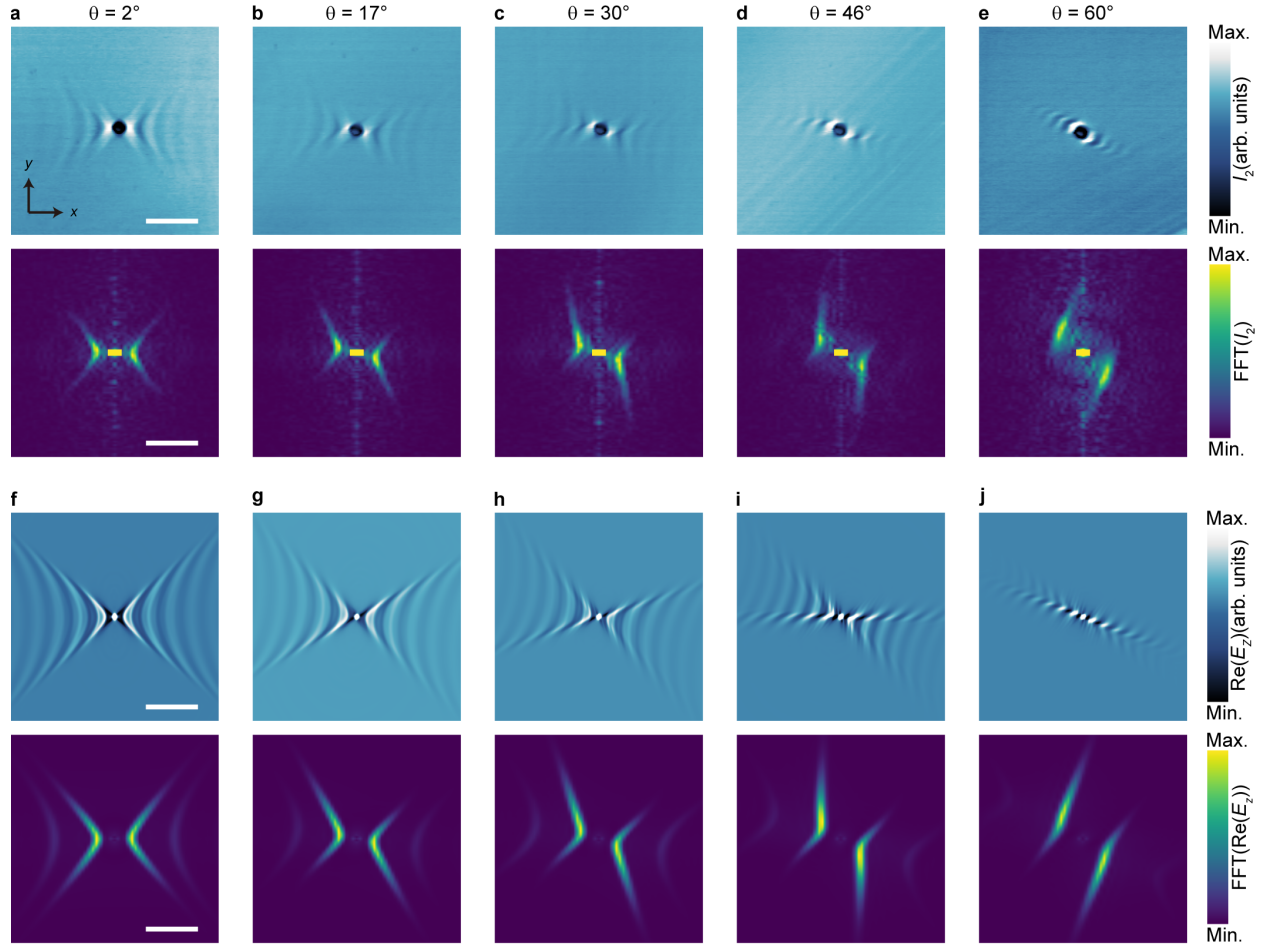
where V_{CNP} is the voltage at the CNP determined from the peak position in the resistance curve, e is the electron charge. The gate capacitance of the field effect transistor C_g is:

$$C_g = \varepsilon \varepsilon_0 / d_g \quad (\text{S42})$$

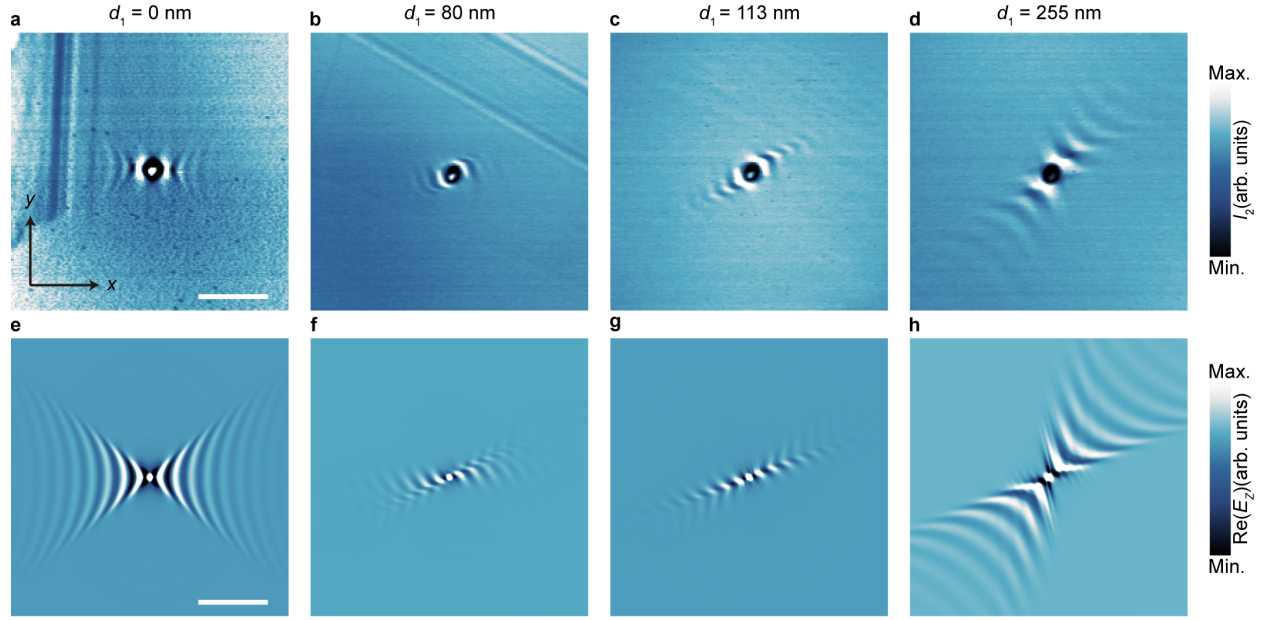
where ε is the dielectric constant of SiO_2 (~ 4), ε_0 is the permittivity of free space, and d_g represent the thickness of the dielectric layer (285 nm SiO_2). For a certain V_g , only E_F and n are unknowns in equations (S40) and (S41). By simultaneously solving these two equations, we can determine the relationship between E_F and V_g .



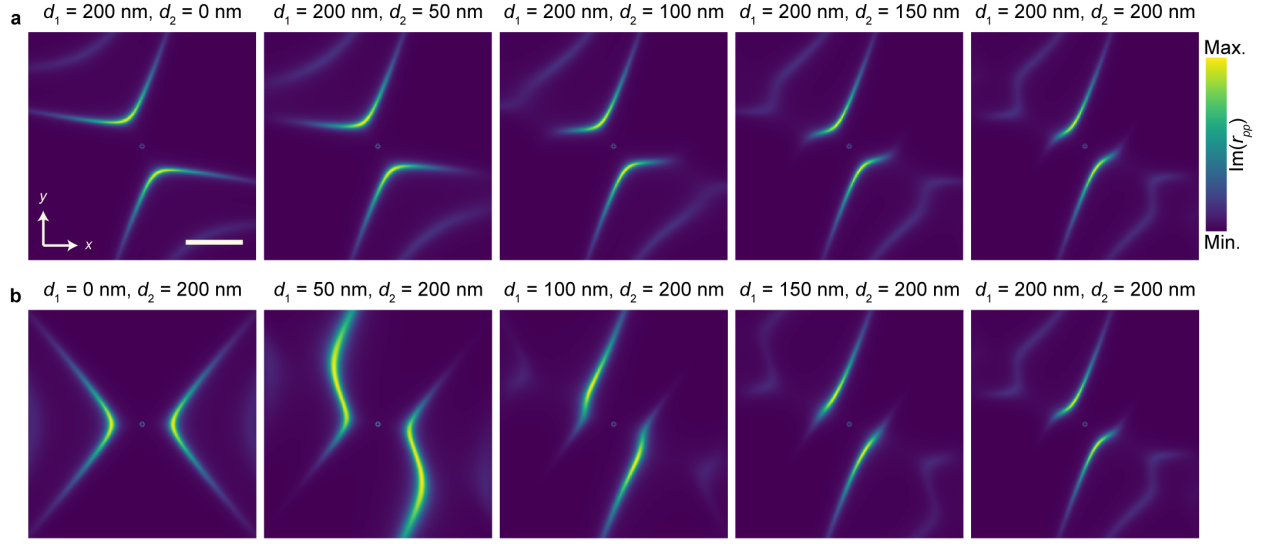
Supplementary Figure 8. Measured dependence of graphene resistance R on V_g . Dependence of graphene resistance on V_g for the twsited bilayer $\alpha\text{-MoO}_3$ /graphene device #1 (Fig. 5 in the main text).



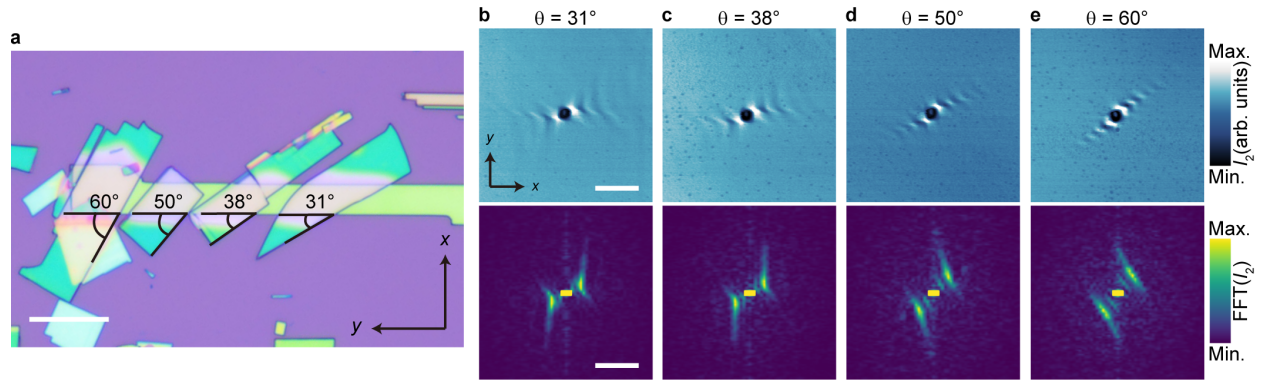
Supplementary Figure 9. Comparison of experimental near-field images Fourier spectra and corresponding simulated results with varying twist angles. **a-e** Experimental near-field images and Fourier spectra. The corresponding parameters are **(a)** $\theta = 2^\circ$ ($d_1 = 133$ nm), **(b)** $\theta = 17^\circ$ ($d_1 = 128$ nm), **(c)** $\theta = 30^\circ$ ($d_1 = 130$ nm), **(d)** $\theta = 46^\circ$ ($d_1 = 134$ nm), and **(e)** $\theta = 60^\circ$ ($d_1 = 135$ nm) with $d_2 = 202$ nm. The x -direction corresponds to the $[100]$ crystallographic orientation of the bottom layer of α -MoO₃. Scale bar: $1\ \mu\text{m}$ (top), $80\ k_0$ (bottom). **f-j** Simulated results excited by the dipole, which correspond to the experimental cases in **a-e**. Scale bar: $2\ \mu\text{m}$ (top), $40\ k_0$ (bottom).



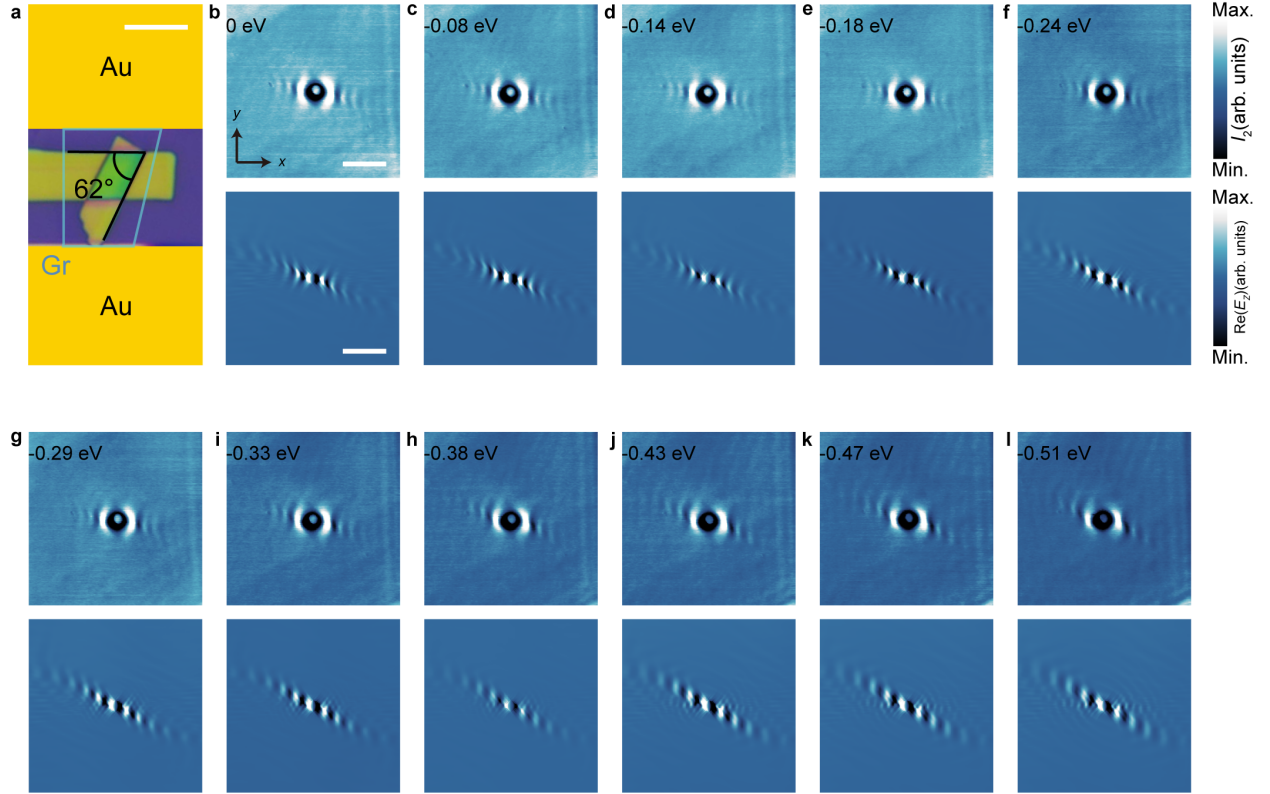
Supplementary Figure 10. Comparison of experimental near-field images and numerical field distributions with varying thicknesses. **a-d** Experimental near-field images, the thickness of the top layer, d_1 , varies among **(a)** 0 nm, **(b)** 80 nm, **(c)** 113 nm, and **(d)** 255 nm, while maintaining $d_2 = 170$ nm and $\theta = 60^\circ$. The scale bar indicates 1 μm . The x -direction corresponds to the [100] crystallographic orientation of the bottom layer of $\alpha\text{-MoO}_3$. **e-h** Numerical field distributions excited by the dipole, which correspond to the experimental cases in **a-d**. The scale bar indicates 2 μm .



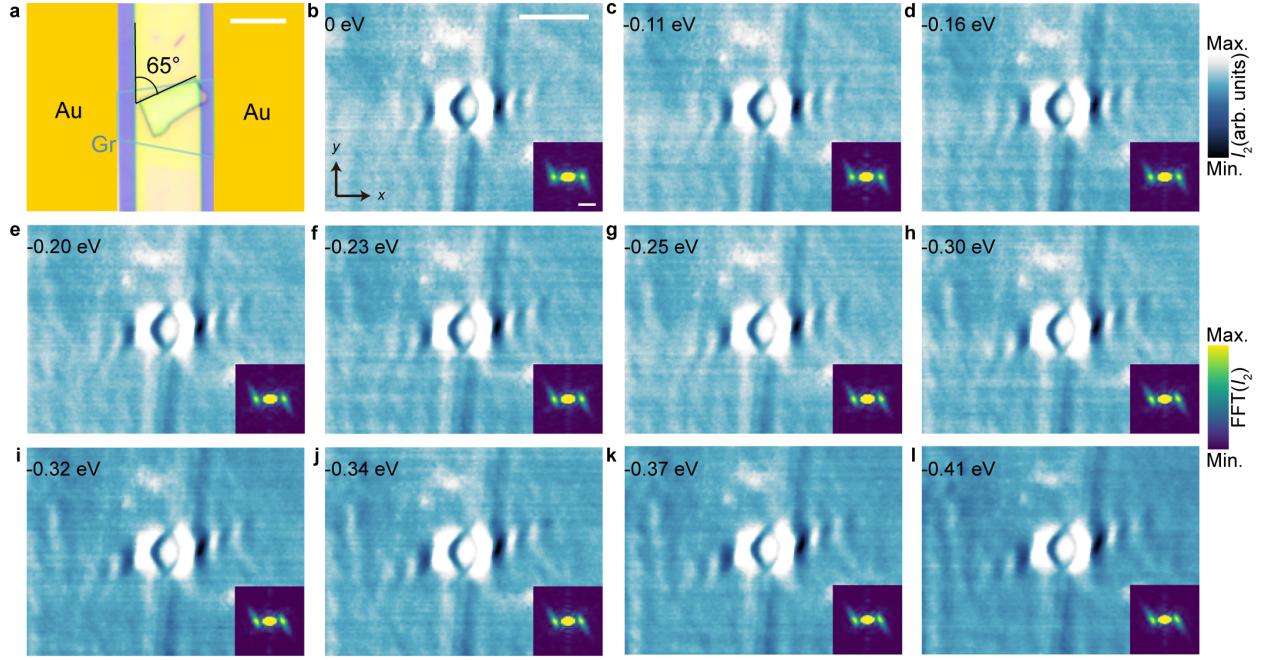
Supplementary Figure 11. Calculated reciprocal space maps of twisted bilayer $\alpha\text{-MoO}_3$ through TMM with varying top layer and bottom layer, respectively. a The top layer $d_1 = 200 \text{ nm}$ and varying bottom layer. The twist angle $\theta = 60^\circ$. The x -direction corresponds to the $[100]$ crystallographic orientation of the bottom layer of $\alpha\text{-MoO}_3$. The scale bar indicates $40 k_0$. **b** The bottom layer $d_2 = 200 \text{ nm}$ and varying top layer. The twist angle $\theta = 60^\circ$.



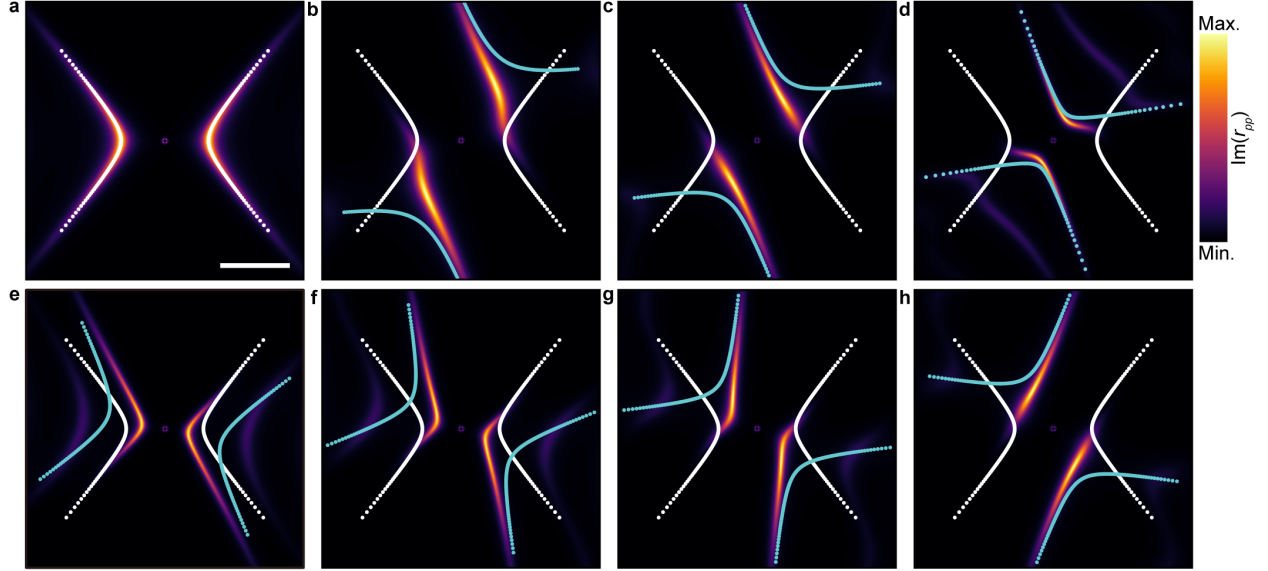
Supplementary Figure 12. Experimental near-field images of another sample with varying twist angles. **a** Optical image of α - MoO_3 flakes with similar thicknesses, each positioned at a varying twist angle on a single α - MoO_3 flake. The scale bar indicates $20\ \mu\text{m}$. **b-e** Near-field images (top panel), along with corresponding Fourier spectra (bottom panel) derived from experimental results for HShPs. The corresponding parameters are **(b)** $\theta = 31^\circ$ ($d_1 = 146\ \text{nm}$), **(c)** $\theta = 38^\circ$ ($d_1 = 148\ \text{nm}$), **(d)** $\theta = 50^\circ$ ($d_1 = 171\ \text{nm}$), and **(e)** $\theta = 60^\circ$ ($d_1 = 158\ \text{nm}$) with $d_2 = 193\ \text{nm}$. The x -direction corresponds to the $[100]$ crystallographic orientation of the bottom layer of α - MoO_3 . The scale bar indicates $1\ \mu\text{m}$ (top panel), and $80\ k_0$ (bottom panel), respectively.



Supplementary Figure 13. Comparison of experimental near-field images of device #1 and corresponding numerical field distributions with varying graphene Fermi energy. **a** Optical image of device #1. Scale: 5 μm . **b-l** Experimental near-field images (top panel) and numerical simulated field distributions (bottom panel). The top and bottom α -MoO₃ layers have thicknesses of $d_1 = 96$ nm and $d_2 = 78$ nm, respectively. The twist angle $\theta = 62^\circ$. The x -direction corresponds to the [100] crystallographic orientation of the top layer of α -MoO₃. The scale bar indicates 500 nm (top panel), and 1 μm (bottom panel), respectively. Numerical field distributions are excited by the point defect illuminated by the plane wave source.



Supplementary Figure 14. Gate-tuning experimental near-field images of device #2. **a** Optical image of device #2. Scale: 5 μm . **b-l** Near-field images, along with corresponding Fourier spectra (inset) derived from experimental results for HShPs. The top and bottom $\alpha\text{-MoO}_3$ layers have thicknesses of $d_1 = 108\text{ nm}$ and $d_2 = 88\text{ nm}$, respectively. The twist angle $\theta = 65^\circ$. The x -direction corresponds to the $[100]$ crystallographic orientation of the top layer of $\alpha\text{-MoO}_3$. The scale bar indicates 500 nm, and $80\ k_0$ (inset), respectively.



Supplementary Figure 15. The IFCs between twisted bilayer α -MoO₃ and relative top and bottom α -MoO₃. **a-d** The thickness of the top layer, d_1 , varies among **(a)** 0 nm, **(b)** 80 nm, **(c)** 113 nm, and **(d)** 255 nm, while maintaining $d_2 = 170$ nm and $\theta = 60^\circ$. The white and blue lines correspond to the IFCs of the bottom and top α -MoO₃ flakes, respectively. The scale bar indicates $40 k_0$. **e-h** The top and bottom α -MoO₃ layers have thicknesses of $d_1 = 130$ nm and $d_2 = 200$ nm, respectively. The twist angles θ are **(e)** 15° , **(f)** 30° , **(g)** 45° , and **(h)** 60° , respectively. The white and blue lines correspond to the IFCs of the bottom and top α -MoO₃ flakes, respectively.

Supplementary Note 8. Effects of graphene doping on the shear factor of twisted bilayer $\alpha\text{-MoO}_3$

The inclusion of graphene and its doping significantly impacts the shear factor in twisted bilayer $\alpha\text{-MoO}_3$. These effects arise through two primary mechanisms: the enhancement of structural asymmetry and the topological transition in the IFCs of the polaritons. Below, we provide a detailed explanation of these mechanisms and their implications.

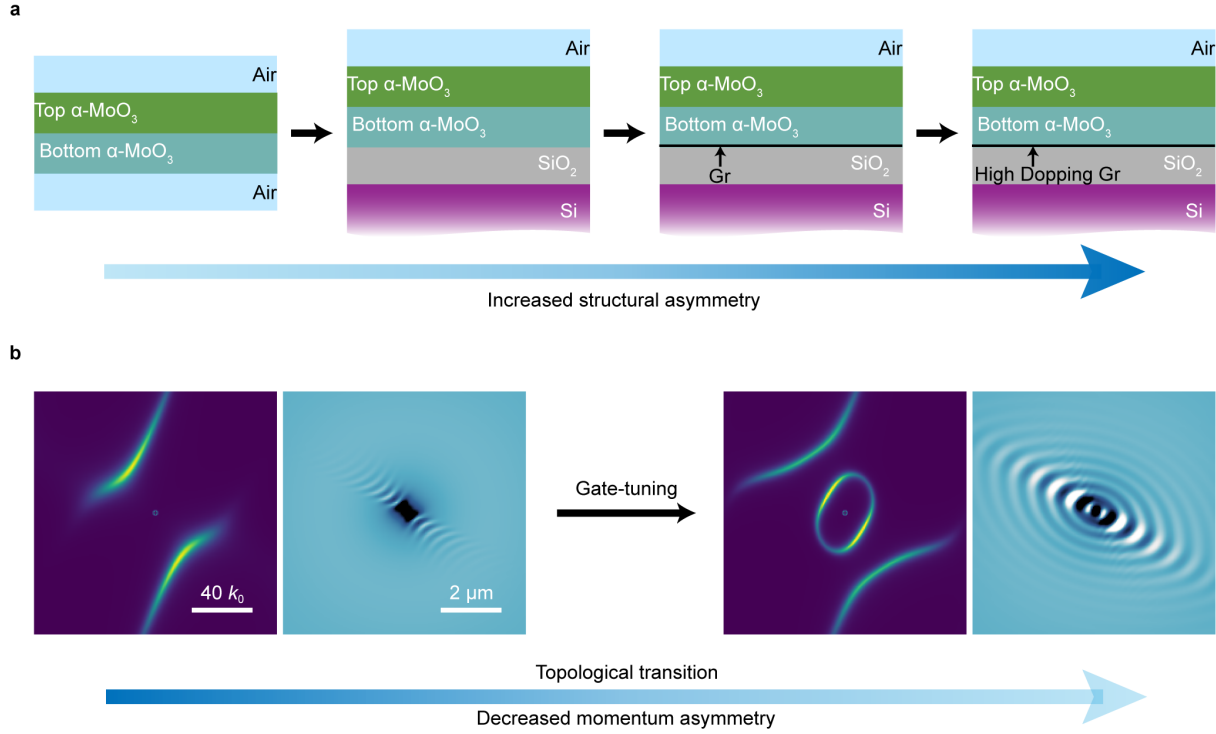
Consider a twisted $\alpha\text{-MoO}_3$ bilayer with equal thicknesses for the top and bottom layers, placed in a symmetric air environment. Such a configuration ensures maximum structural symmetry along the z-direction. However, introducing an Si/SiO₂ substrate disrupts this symmetry by creating an asymmetric dielectric environment, with air above and SiO₂ below the bilayer. This structural asymmetry increases further when a graphene layer is introduced at the bottom layer of the $\alpha\text{-MoO}_3$.

The effect of this asymmetry becomes more pronounced as the graphene's Fermi energy increases. Higher Fermi energy enhances the carrier concentration and conductance of graphene, modifying its dielectric properties and amplifying the contrast between the top (air) and bottom (graphene-Si/SiO₂) interfaces. This enhancement strengthens the shear effect, as the increased asymmetry amplifies the redistribution of energy flow of hyperbolic polaritons. Consequently, the shear factor increases with the inclusion and doping of graphene, as illustrated in Supplementary Figure 16a.

As the graphene doping increases, the system exhibits a topological transition in the IFCs of the polaritons—from hyperbolic to elliptical shapes, as depicted in Supplementary Figure 16b. El-

liptical IFCs are inherently more symmetric compared to hyperbolic ones, reducing the momentum asymmetry within the system. This reduction in asymmetry counteracts the shear effect, thereby decreasing the shear factor.

At low Fermi energy, the system remains in the hyperbolic regime, where asymmetric structure and dielectric environment induced by graphene dominates. In this regime, increasing the graphene doping leads to a continuous rise in the shear factor. However, at sufficiently high Fermi energy, the IFCs approach the elliptical regime, where the higher symmetry reduces the shear factor. In the present experimental setup, the graphene's Fermi energy was not tuned high enough to observe this reduction. Nevertheless, the balance between structural asymmetry and topological transition in IFCs provides methods for controlling the shear factor.



Supplementary Figure 16. Effects of graphene and its doping on the shear factor of twisted bilayer $\alpha\text{-MoO}_3$. **a** Schematic illustration of the structural and dielectric asymmetry induced by the twisted bilayer $\alpha\text{-MoO}_3$ on an Si/SiO_2 substrate, with the incorporation of graphene further amplifying the asymmetry along the z -direction. The increasing Fermi energy and carrier concentration in graphene enhances the contrast between the dielectric environments at the top (air) and bottom (graphene- Si/SiO_2) interfaces, leading to a strengthened shear effect. **b** Topological transition and decreased momentum asymmetry: The IFCs of polaritons shift from hyperbolic to elliptical with increasing graphene doping, reducing momentum asymmetry due to the higher symmetry associated with elliptical IFCs, potentially counteracting the shear factor enhancement.

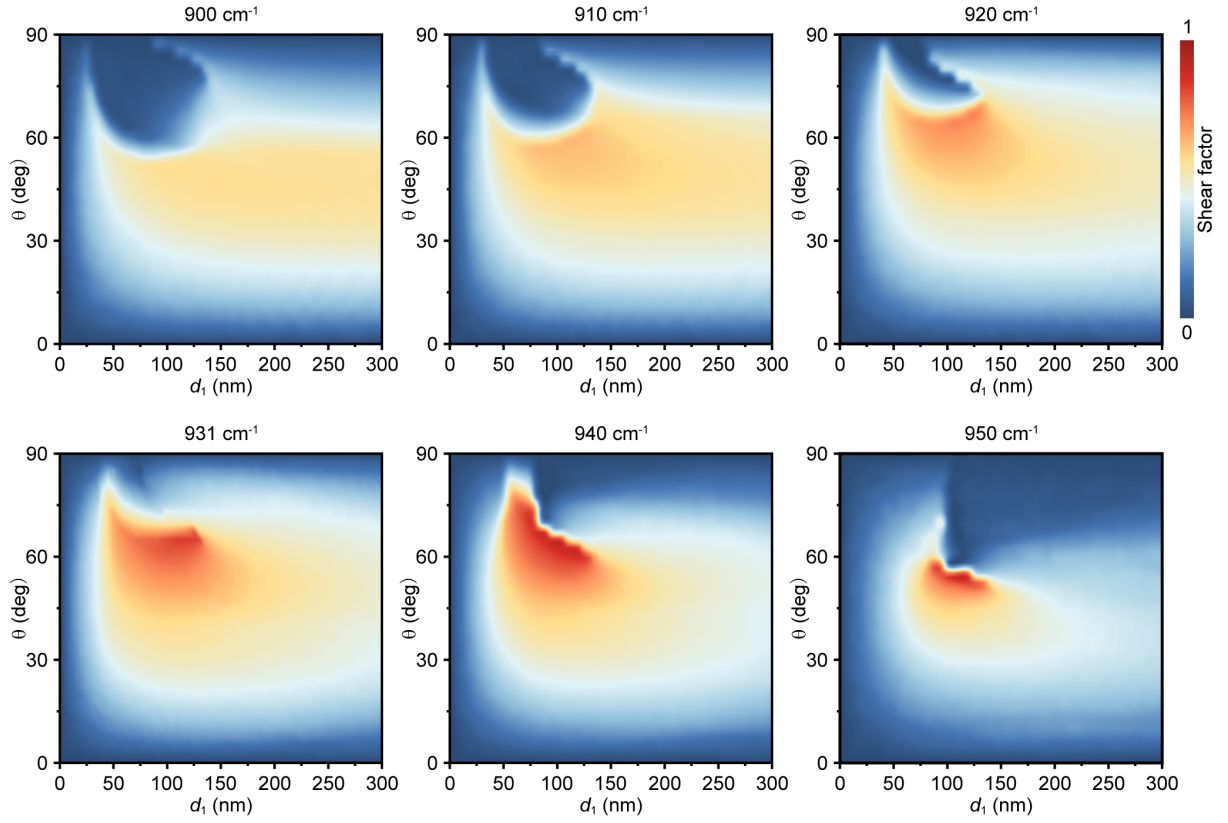
Supplementary Note 9. Frequency dependence of the shear factor

To complement the findings presented in the main manuscript, we have performed additional simulations to investigate the frequency dependence of the shear factor. While the experimental and simulation results in the main text focus on a fixed frequency of 931 cm^{-1} due to experimental constraints, the shear factor is inherently frequency-dependent. Exploring this relationship is essential for optimizing the system's geometrical parameters to achieve maximal shear factor values.

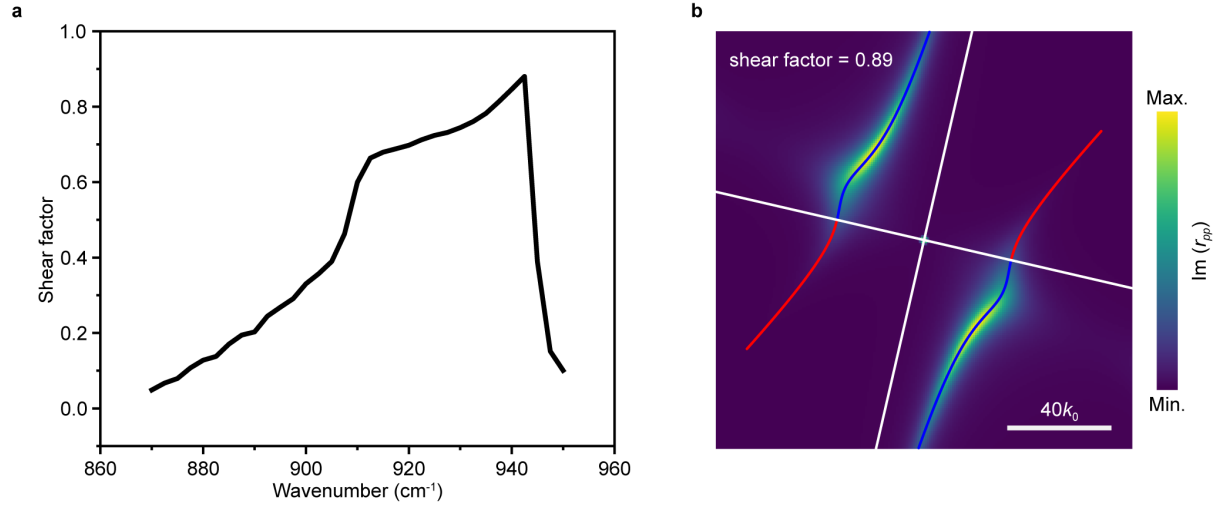
In these simulations, the bottom $\alpha\text{-MoO}_3$ layer thickness was fixed at 200 nm, while the top-layer thickness and the twist angle were systematically varied. This approach was adopted because an asymmetric configuration, achieved by varying only the top-layer thickness, enhances the structural asymmetry between the layers. Based on the identified "sweet spot" parameters, we further calculated the reciprocal space distributions to examine the resulting dispersion characteristics of HShPs. These results provide valuable insights into the interplay between frequency and geometry in controlling the properties of HShPs.

The shear factor exhibits a frequency-dependent trend, initially increasing with frequency and then decreasing at higher frequencies as shown in Supplementary Figure 18. This behavior arises from the intricate balance between topological transitions and interlayer coupling. At lower frequencies within the Reststrahlen Band 2, the critical twist angle required for a topological transition in twisted $\alpha\text{-MoO}_3$ bilayers decreases⁸, making the topological transition more likely happen and reducing the system's overall asymmetry.

In contrast, at higher frequencies, the coupling between the two $\alpha\text{-MoO}_3$ layers weakens due to the stronger field localization at higher frequencies⁵, reducing the shear effects arising from their



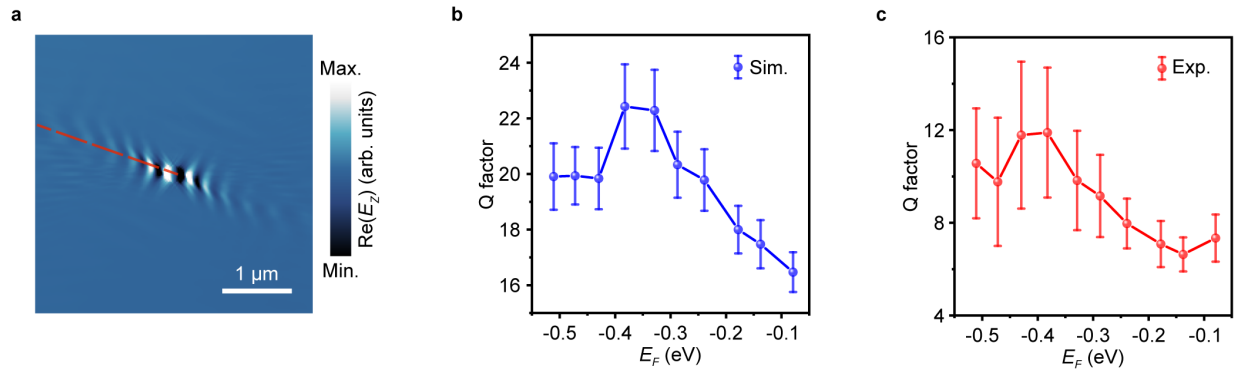
Supplementary Figure 17. Frequency-dependent shear factor calculated for a twisted bilayer $\alpha\text{-MoO}_3$ system. The bottom layer thickness is fixed at 200 nm, while the top-layer thickness and twist angle are varied. The results illustrate the dependency of the shear factor on frequency and reveal the optimal parameters for maximizing its value.



Supplementary Figure 18. Frequency-dependent behavior of the shear factor and momentum-space distribution. **a** Shear factor as a function of excitation frequency. **b** Reciprocal space distribution calculated under the optimal conditions for maximum shear factor. The parameters are a frequency of 940 cm^{-1} , a top-layer thickness of 120 nm, a bottom layer thickness of 200 nm, and a twist angle of 62.5° . The shear factor is 0.89.

interaction. This weakened coupling results in a decline in the shear factor at higher frequencies. The combination of these two opposing trends gives rise to a critical frequency at which the shear factor is maximized. This optimal frequency represents a balance between interlayer coupling and topological transitions, where the shear effect is most pronounced. Our simulation results align with these theoretical expectations, confirming the observed trend.

Supplementary Note 10. Dependence of Q factor on Fermi energy



Supplementary Figure 19. Dependence of Q factor on Fermi energy. **a** Real-space distribution of shear polaritons, with the red dashed line indicating the location where the line profile is extracted for Q factor analysis. **b** Simulated Q factors obtained by fitting the line profiles along the red dashed line in **a**. **c** Experimentally measured Q factors, derived using the same method. The results in **b** and **c** demonstrate the dependence of Q factors on the Fermi energy. The error bars represent the 95% confidence intervals.

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