Title: Polar and quasicrystal vortex observed in twisted bilayer molybdenum disulfide

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Abstract: We report the observation of electric field in twisted bilayer molybdenum disulfide (MoS_2) and elucidate its correlation with local polar domains using four-dimensional scanning transmission electron microscopy (4D-STEM) and first-principles calculations. We reveal the emergence of in-plane topological vortices within the periodic moiré patterns for both commensurate structures at small twist angles and the incommensurate quasicrystal structure that occurs at a 30° twist. The large-angle twist leads to mosaic chiral vortex patterns with tunable characteristics. A twisted quasicrystal bilayer, characterized by its twelve-fold rotational symmetry, hosts complex vortex patterns, and can be manipulated by picometer-scale interlayer displacement. Our findings highlight that twisting 2D bilayers is a versatile strategy for tailoring local electric polar vortices.

One-Sentence Summary:
The twisted molybdenum disulfide bilayers display polar chiral vortex patterns that are contingent upon the twist angle and can be actively controlled.

Main Text:

Bilayer two-dimensional (2D) materials, such as MoS_2, are dominated by a few energy-favorable states. These 2D materials naturally adopt distinct stacking orders: the anti-aligned 2H (P63/mmc) or the aligned 3R (R3m), which are differed by a relative interlayer rotation of 60°(*1*). Recently, twisting of van der Waals (vdW) bilayers and related multilayered homo-or heterostructures has emerged as a transformative approach to access and tailor electronic properties through enhanced quantum coupling effects induced by long-wavelength moiré patterns. For example, moiré superlattices can host charge density waves (*2*), exhibit unconventional superconductivity (*3*), and give rise to Mott insulator states in magic-angle twisted bilayer and trilayer graphene (*4*). Moreover, unexpected atomic reconstructions (*5-8*) and moiré excitons (*9*) have been observed in twisted bilayer transition metal dichalcogenides (TMDCs).

In bilayer 2D materials, interlayer lattice structures undergo notable transformations as the twist angle is varied. At small twist angles ($\theta_t < 3°$), the two layers maintain commensurability (*10-15*). However, at larger angles ($\theta_t > 8°$), the lattice structures enter an incommensurate regime in which the superposition of electronic lattices leads to an approximate moiré supercell, albeit with a discernible loss of the long-range order translational symmetry. Previously, the incommensurate bilayer structures were thought to have a minimal impact on interlayer electronic interactions, primarily because of the lack of electronic phase coherence. However, recent studies have demonstrated nontrivial interlayer coupling in the twisted quasicrystal bilayer, as evidenced by the discovery of Dirac electrons in dodecagonal graphene quasicrystals (*16, 17*) and the identification of van Hove singularities associated with the rich K valley structures in dodecagonal tungsten diselenide quasicrystals (*18*). These findings suggest that overlapping moiré potential wells may show emergent properties.

Unlike conventional ferroelectric materials that host distinct multiple polar states at their free energy minima (*19*), low-dimensional 2D systems may encompass collective local electric dipole moments. These moments give rise to spontaneous polarization domains that emerge from a balance among dimensional constraints, misfit elastic energies, and interfacial charge effects (*20, 21*). As a result, this balance fosters the evolution of non-trivial topological configurations with continuous polar structures, such as skyrmions (*22, 23*), merons (*24*), and vortices (*21, 25*). These local electric fields could be detected, manipulated, and could have device applications. However, the observation of topological polar textures within twisted 2D vdW materials has remained elusive.

In this study, we used four-dimensional scanning transmission electron microscopy (4D-STEM) and first-principles calculations to characterize local topological polar electric fields and establish their relation with the twist angle in twisted bilayer MoS_2. In contrast to the "sliding ferroelectricity" observed in the normal direction of bilayer 2D materials such as hexagonal boron nitride (*h*-BN) (*26, 27*), TMDCs (*28*), and other vdW systems (*22, 23*), the topological vortex fields in twisted 2D bilayer MoS_2 were oriented in-plane and even exhibited tunability of polar patterns over a wide range of twist angle, the intricate results were directly associated with the twist-induced moiré superstructures.

These experimental findings are further supported by our first-principles calculations, in collaboration with recent insights into topological interlayer-driven ferroelectricity within moiré domains at minimal twist angles (*29*) and twisted $BaTiO_3$ freestanding layers (*30*). Moreover, the vdW interaction in atomically smooth 2D materials provides considerable degree of turnability in terms of interlayer sliding and twisting. Beyond the complex topological mosaic vortex configurations discovered at lower twist angles, we have detected even more elaborate topological polar patterns at larger angles, including the polar quasicrystal structure with a motif of clock-like patterns and twelve-fold rotational symmetry, recurring throughout the material matrix. These polar patterns within twisted 2D bilayers could be manipulated through picometer-scale interlayer displacements, which could allow additional control over emergent properties.

Polar vortex field in twisted moiré structures

The as-synthesized twisted bilayer MoS_2 samples (see Materials and Methods) were investigated with 4D-STEM experiment, in which a focused electron beam was systematically rastered across the samples under focused-probe differential phase contrast (DPC) imaging condition (*31-35*). These scanning transmission electron microscopies (STEM) and 4D-STEM techniques were used to map the in-plane atomic polarization fields in the twisted bilayer MoS_2. The distribution of twist angles is analysed through a series of SAED patterns, which can be found in fig. S1. The atomic structure model of the twisted bilayer MoS_2 is shown in Fig. 1A, and the STEM-annular dark field (ADF) images are shown in Fig. 1, B to D. The observed topological polar structures, such as vortices illustrated in Fig. 1, E and F, correspond to the 4D-STEM results in Fig. 1, G and H.

The 4D-STEM analysis is based on the shift in the center of mass (ΔCOM) of the electron beam, which correlates to the deflected trajectory of the incident electrons after interaction with the electrostatic field in ultrathin samples. The details of 4D-STEM method, including the computation of the in-plane vorticity of the measured polarization fields ($-\mathbf{1}\times\nabla\times\boldsymbol{E}_{xy}$), can be found in the Materials and Methods section. The intensity of vorticity is proportional to the curl of polarization field, which signifies the swirling nature of the local field. In this context, the blue and red hues denote the clockwise and anti-clockwise swirling directions, respectively (Fig. 1, G and H).

The STEM-ADF (fig. S2) image revealed hexagonal arrangements with the moiré periodicity (λ_m ~ 1.5 nm, see fig. S3). In addtion, the additional spots appeared in fast Fourier transform (FFT) analysis due to the coherence effect of the STEM-ADF images from two samples (θ_t~8.3° and θ_t~11.6° in fig. S4) also confirmed existence of the moiré pattern and its periodicity. Although slight variations in equilateral periodicities observed were caused by strain influences or experimental drift, they did not impact the measured features. The characteristic periodic vortex patterns aligned well with the moiré lattice, as unveiled in our 4D-STEM observations (Fig. 1, G and H).

The most pronounced vortex feature, associated with local AA stacking regions that exhibit mirror symmetry with respect to the middle plane of bilayer, manifested a lateral periodicity of ~ 1.5 nm in the 11.6° twisted bilayer sample (Fig. 1G). This vortex feature was also found at the AB/BA stacking domains, showing a hexagonal sublattice pattern with six minor vortices (Fig. 1H). These features were in concert with the switchable out-of-plane moiré polarization domains – sliding ferroelectricity in small twisted 2D structure. The vortex polarization fields were also observed in bilayer MoS_2 samples at the twist angle of 1.8°, 2.4°, -4.1° and 8.3° (fig. S5-7), where the positive/negative twist angle denotes a clockwise/anti-clockwise rotation of

the top layer with respect to the bottom layer. This further demonstrates the presence of the in-plane vortex polarization patterns in twisted bilayer MoS_2 across a wide twist angle range.

The out-of-plane polarization within the moiré supercells, which are a consequence of the interlayer twisting, could be quantitatively assessed through density functional theory (DFT) calculations. These calculations distinctly differentiate potential variations in the out-of-plane direction, which indicate the emergence of downward (−P) and upward (+P) polarization states within the AB and BA stacking domains, respectively. In contrast, AA stacking domains did not show out-of-plane polarization (see fig. S8 and S9). By combining these findings, the overall scheme of the polar field distribution can be illustrated (Fig. 1F), where the AB and BA sites exhibited similar in-plane vortex field and opposite out-of-plane polarity, resulting in different chirality.

To elucidate the nature of such non-trivial topological polar textures, it was essential to evaluate their topological charges (*36, 37*) that can be computed by integrating the angular variation of the normalized local field over a two-dimensional cross-section. The winding number, an integral topological invariant, is defined as: $\boldsymbol{N_{sk}} = \frac{1}{4\pi}\int \boldsymbol{n}\cdot\frac{\partial \boldsymbol{n}}{\partial x}\times\frac{\partial \boldsymbol{n}}{\partial y}\boldsymbol{dx\,dy}$ (*20, 38*), where $\boldsymbol{N_{sk}}$ refers to that the number of times that plane surface can wrap around a unit sphere, and $\boldsymbol{n}$ is normalized local dipole moment. The experimental results of in-plane polar field in twisted MoS_2 (Fig. 1, G and H) show 2π-modulation of the in-plane component. However, the absence of z-component measurement in the experimental hinders a direct analysis for these meron-like topologies that commonly seen in a ferroelectric material as characterized by discrete swirling core and spherical domain boundaries(*24*).

The localized polar vortex field which we observed could be ascribed to the moiré periodicity that directly resulted from the twisting of the MoS_2 bilayers. The formation of in-plane vortex polar domains in these twisted bilayers were likely to stem from the twist stacking-induced charge redistribution in combination with weak contribution from small in-plane ionic displacements. This is in stark contrast to the ferroelectricity observed in traditional perovskite oxides, which arises primarily from ionic displacements (*23, 24*), or out-of-plane ferroelectricity in twisted bilayer *h*-BN and TMDs (*5, 26-28, 39*) induced by interfacial charge transfer only. Specifically, the distinctive in-plane polarization in highly twisted bilayers are originated from the local approximant stacking domains. DFT calculations imply small in-plane atomic displacements of within 0.055 Å at the twist angle of 6.0090° upon stacking and tend to further decrease and even diminish with the increasing twist angle (Fig. S10). This is indicative of weak contribution to the in-plane polarization from the in-plane ionic displacement especially in the large twist angle regime. On the other hand, there are noticeable electronic charge redistribution induced by the twist stacking, which are mainly localized around the interfacial S and Mo atoms, as displayed in Fig. S11. Further details on the in-plane polarization estimates are provided in the Supplementary Text and Table S1.

Twist angle dependent polar structures

To explore the impact of twist angles on the topological polarization structures, we undertook a comprehensive spatial evolution analysis. We scrutinized the polar domains in MoS_2 bilayer moiré patterns across a wide range of twist angles, using both 4D-STEM measurement and corroborative image simulations, as illustrated in Fig. 2, A to P. We observed that MoS_2 samples subjected to diverse twist angles—namely 8.3°, 11.6°, -18.4°, and 30°—demonstrated different polar field distributions that ranged from intricate vortex formations to twelve-fold quasicrystal polar domains resembling clock-like structures.

At lower twist angles, ($\theta_t < 3^o$), the interlayer twist modulated the electronic properties through induced moiré superlattice reconstruction (*8, 40*). At higher twist angles ($\theta_t > 8^o$), the bilayers showed complex atomic configurations (Fig. 2C). These structures gave rise to diverse polar textures (Fig. 2G), which were different from those observed at lower twist angles (Fig. 2, E and F). We further discovered both counter-rotational (blue) and anti-counter-rotational (red) vortices around the AA sites (rotation center), highlighted by yellow circles (Fig. 2G). The combination of these rotational vortices led to a pattern of concentric counter-rotating vortices (Fig. 2G), which was consistent with the results obtained from multislice image simulations (Fig. 2O).

In the 30^o twisted quasicrystal bilayer MoS_2, we observe a "clock" motif manifested in both the STEM-ADF and the selected-area electron diffraction (SAED) patterns (Fig. 2D, fig. S12, A and B). Each clock motif could be identified by a dotted circle ring that enclosed an arrangement of 12 lobes or atomic clusters, centering around AA stacking sites exhibiting a 30/90° twist. These clocklike motifs distributed in an aperiodic manner, filling STEM-ADF image of the 30^o twisted MoS_2 and left space for motifs of the 30° twisted AA stacking "hole".

The polar mapping of the 30° twisted MoS_2 (Fig. 2P) unveiled a distinct circular ring formation in the shape of a clock, each featuring twelve lobes that resembled the patterns observed in the STEM-ADF images. Upon closer examination of the atomic structure, it was evident that the atomic columns were closely stacked, which resulted in pairs of blue-red and red-blue lobes relative to the center. These atomic columns exhibited a deviation of ~ 15° from the zigzag and armchair orientations of MoS_2 (fig. S13). Furthermore, the atoms situated at the top and bottom of the nearly aligned atomic column were laterally separated by ~ 25 pm. When the clock motif was oriented at ~ 30°, a slight distortion in the atomic structure influenced the polar map, leading to a weakened representation of the symmetric blue-red and red-blue lobes. The curl map for the quasicrystal displayed weak swirling features, as shown in fig. S14. As a result, only a few of these lobes were clearly distinguishable in experiments, marked by yellow dot lines (Fig. 2H).

We present the polar field distribution across a wide range of twist angles, ranging from 0° to 120° (Fig. 2Q). During the twisting process, a selected area of the AA core region was used as the rotation center. The intensities of the polar fields at these AA cores were notably prominent at twist angles in the vicinity of ~8° and 52°, as well as 68° and 112°, which corresponded to vortex and anti-vortex configurations, respectively. In the case of the commensurate structures observed at 0° and 60° twist angles, a remarkable mirror symmetry was noticeable, manifesting in the vorticity inversion. Interestingly, at certain twist angles such as 30° and 90°, the emergence of a horizontal plateau was observed, indicating the quasicrystal structure formation. These plateaus occurred at high twist angles, underscoring a transition to a more complex structural paradigm. At higher twist angles ($\theta_t > 8°$), a mosaic periodicity emerges, which is often classified as an incommensurate system. This mosaic periodicity involved a periodic variation in polar order spanning 0° to 60° and it became increasingly pronounced within the intermediate twist angles (15° to 22°), as shown in fig. S15.

The presence of this mosaic periodicity in the high twist angle regime raised fundamental questions regarding the stacking configuration and moiré wavelength. A closer examination of the STEM-ADF image (fig. S16) revealed saddle domain walls, where AB/BA domains became obscure and gave way to a dominance of the AA stacking configuration. The difference between AA and AB/BA lies in the interlayer slip or shift along the $(11\bar{2}0)$ crystal plane with

a magnitude of $\frac{\sqrt{3}}{3}a$, where a is the lattice constant. In the intermediate twist angle regime, the overall structure could be described as approximated stacking configuration where AA regions were separated by saddle domain walls, rather than the distinct AA, AB, and BA configurations observed in the low twist angle region. As the twist angle deviated from 30°, we observed the emergence of a distinct circular pattern with the shortest periodicity occurring between 23° and 37°. This pattern exhibited characteristics similar to the clock motif observed in the 30° quasicrystal bilayer. The overall periodicity in the twisting tendency transited from atomic and mosaic structures to a more aperiodic or microscale arrangement, rather than strictly following the rules of the moiré equation or wavelength.

To complement our experimental findings, multislice image simulations were used to model the scattering dynamics of incident electron probes interacting with the twisted bilayer samples (*41*). The simulation results aligned remarkably well with the experimental observation (Fig. 2, A to P). These results also suggested that twist polarization phenomena could occur in other materials. Notably, vortices are observed in various types of rigidly twisted stacking 2D materials, including MoS_2, indium selenide (InSe), and bismuth selenide (Bi_2Se_3), although they are nearly absent in graphene (see fig. S17).

Further explorations into the effects of lateral sizes on MoS_2 reveal intriguing insights. The polarization signals between a single hexagonal MoS_2 unit cell and an extended 5 nm unit cell showed negligible variance (fig. S18). This observation indicates that the detected polarization signals are caused by the twisted lattice and electronic structures, rather than the effects from moiré pseudo-particles. To verify our results and mitigate any potential artifacts introduced by the 4D-STEM setup, we conducted a series of controlled experiments with varied collection angles and beam convergence angles to assess their influence on the observed polar field results (fig. S19 and S20) and ferroelectric structures (fig. S21). These comprehensive investigations ensured the robustness and accuracy of our findings.

Quasicrystal tiling and 12-fold symmetry

The 30° rotation between the two MoS_2 layers led to the formation of a dodecagonal quasicrystal moiré superlattice, an incommensurate structure characterized by its 12-fold rotational symmetry but lacking translational symmetry (*16*). Our observations revealed a distinct signal displaying 12-fold symmetry with two side lobes, similar to the optical phase singularities observed in momentum space (*42*). Using STEM-ADF images obtained with a virtual detector constructed through 4D-STEM, we demonstrated the quasi-periodic ordering of the atomic structure (Fig. 3B). The STEM-ADF images showed the vertices of the dodecagonal framework, constituted by two-hole aligned columns. This STEM result (Fig. 3B and Fig. S12A) is in contrast with the phase-contrast-based high-resolution transmission electron microscopy (HRTEM) images of quasicrystal bilayer graphene (*16*). The dodecagonal arrays in this quasicrystal bilayer MoS_2 are composed of Stampfli tiles such as squares, rhombuses, and triangles, which fill the space without any voids (Fig. 3, A and B) (*43*). Furthermore, the triangular tiling pattern can be classified as four distinct sublattices based on the anisotropic atomic configurations of Mo and S_2.

Although both MoS_2 and graphene possess hexagonal lattices, the lattice sites occupied by Mo and S in MoS_2 are not equivalent to those of the carbon atoms in graphene. This, MoS_2 belongs to the space group P63/mmc, whereas graphene belongs to the space group P6/mmm. In twisted quasicrystal bilayer MoS_2, four distinct types of triangular tiles emerged, each resulting from different stacking combinations of sublattice atoms within the triangular domains. These

combinations can be characterized by the overlapping of atom columns at the center, as depicted in Fig. 3A. We identified these combinations as Mo/S_2, S_2/S_2, S_2/Mo, and Mo/Mo stackings, which are denoted as (i), (ii), (iii), and (iv), respectively (Fig. 3A). Such varied stackings are absent in graphene quasicrystals because the two sublattice are symmetry equivalent.

We further analyzed the divergence in the 4D-STEM electric field measurements in bright field mode (detailed in the Materials and Methods), which allowed us to map the charge density distribution in the quasicrystal bilayer MoS_2. The mapping results are in good agreement with DFT simulations (Fig. 3, C and D). It should be noted that the electronic coupling effect was too weak to10 result in substantial amplitude fluctuations of electron wave functions detectable by STEM technique. However, it could be indirectly inferred through techniques such as angle-resolved photoemission spectroscopy (ARPES) and scanning tunnelling spectroscopy (STS), which can capture photon-electron interactions (*16*) and weak tunnelling current (*17*), respectively, and might offer the necessary sensitivity to observe this effect.

Within the triangular sub-tiles of the quasicrystal structure, the polar fields not only exhibit a noticeable degree of similarity but also a centrosymmetry relative to the center of the tile, as evidenced in Fig. 3A and fig. S22. Notably, even within the same type of triangular tiles, we observed "two side lobes" with different orientations near the tile center. This observation suggested a deviation from the expected 3-fold rotational symmetry within the polar field of the triangular tiles, which could be caused by small lateral displacements in the superimposed atomic columns.

The corresponding grid, atomic structures, and experimental images (Fig. 3A) provide a detailed depiction of each Stampfli tile. In the rhombus tiles, despite the variations in the two sub-tiles created by different stacking sequences, they recurred in an alternating pattern around a 12-fold center. The STEM-ADF images and the associated polar fields of these rhombic sub-tiles were identical (Fig. 3B). Ignoring the stacking order, the structure appeared the same when viewed in projection. For the square tiles, the columns of atoms overlapping near the centers consisted of Mo/S_2, S_2/S_2, S_2/Mo, and Mo/Mo. These configurations gave rise to a self-affine square tiling pattern, which follows the deflation and inflation rule with a scaling factor of $\sqrt{3+\sqrt{2}}$, as marked by the dotted square (Fig. 2A) (*41*).

We emphasize that vorticity was only observed in areas with small rotational displacements where atomic columns were nearly aligned. It disappeared in regions characterized by well-arranged BA/AB and AA stackings. Furthermore, the detailed STEM images did not align perfectly with the Stampfli mapping, possibly because of drifting and distortion during the 4D-STEM data acquisition process. To maintain the integrity of the original data, we did not apply affine transformations to correct these imperfections during data processing. Instead, we chose to showcase the quasi-periodic order through SAED result (fig. S12B). We note the two side lobes forming a clock pattern in the polar field, which correspond to a mesoscale local stacking periodicity of ~ 2 nm perpendicular to the rhombic tile boundary. This configuration aligned well with the joint line connecting the vorticity inversion lobes (fig. S13).

In situ TEM manipulation of bilayer quasicrystal MoS_2

To further manipulate the Stampfli tile patterns of the bilayer MoS_2 quasicrystal, we performed in situ TEM experiments. The pristine 30° twisted bilayer MoS_2 grown by CVD method was free of strain, and then we initially introduced a crack into the twist sample. The crack mediated by the electron beam propagated away from the beam-exposed area, causing substantial shear

and strain in the vicinity of the bilayer MoS_2 crack. No beam-induced defects were observed in these areas. The different crystal orientations of the individual layers caused the crack to propagate along various directions, which resulted in the bifurcation of the crack tip (fig. S23A). This approach allowed us to observe and document the relative tiny interlayer displacement at the atomic scale under interlayer shear strain in the bilayer quasicrystal MoS_2. Such interlayer motion directly impacted the local tessellation pattern of the quasicrystal, as depicted in Fig. 4A, suggesting that these clock polar patterns were closely associated with the vertices of the tiles.

To visualize and analyse the atomic displacements, we plotted false-colored Mo atomic displacements derived from sequential STEM-ADF images through atom tracking method (fig. S23). Arrows are used to signify the direction of the displacements, tracking the transition from the primary (initial) to the secondary (subsequent) image frames (fig. S23C). By subtracting displacements between two successive STEM-ADF image frames (fig. S23, D and F), specifically between the individual layers, we mapped the local interlayer shift of the bilayer quasicrystal (Fig. 4B). As a result, this specific sample area exhibited a maximum lateral interlayer displacement of 80 pm.

The results demonstrated a correlation between the quasicrystal tiling configurations and the relative interlayer displacements. Notable modifications in the quasicrystal tessellation were readily identified in highly distorted regions, which are highlighted by white grid overlays in the STEM-ADF images (Fig. 4, C and D). These modifications were in good agreement with the patterns of atomic displacements in Fig. 4B. In essence, our observations substantiated the modulation both the tiling and polar domain structures through subtle interlayer displacement in bilayer MoS_2. The examination of node components (fig. S24) indicated a consistent distribution of node elements among various groups, despite tiling patterns undergoing considerable lateral shift (1117 pm, see the red arrow in Fig. 4D) after the manipulation. Hence, in the bilayer quasicrystal MoS_2, the tiling pattern could amplify the interlayer displacement by more than 10 times and provide a versatile approach to manipulate the atomic and polar structures in these twisted bilayers. It should be noted that the amplification factor is anisotropic to atomic movement, which results in varied magnitudes and differ from the predicted moiré period (fig. S25).

Conclusion

Our study successfully demonstrated the emergence of in-plane polar vortex in the twisted bilayer MoS_2, highlighting the universal nature of topological chiral polar structures in 2D moiré systems. By combining experimental 4D-STEM measurements with theoretical simulations, we not only clarify the intricate angle-dependent polar structures in twisted bilayer MoS_2, but also uncover the potential to manipulate the vortex polar domains through interlayer displacement. The presence of correlated in-plane polar vortices and local atomic stacking orders provides opportunities for manipulating these chiral polar vortices through external electric field or interlayer sliding and twisting. These discoveries provide useful insights into the complex behavior of polar structures in twisted 2D bilayers, paving a way towards tuning emergent quantum properties at atomic-scale, and thus enabling promising prospects for high density information storage and processing.

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Author contributions:

T.H.L. J.Z., and M.Y. led and supervised this project. T.H.L. conceived the project. C.S.T. conducted STEM, image simulation, formal analysis and wrote the original manuscript. C.S.T and X.Z. conducted 4D-STEM and in situ STEM. C.S.T., H.W., J. H., S. G., and K. H. L. prepared the materials. T.Y., Z.Y., L.W.W., C.-S.L., S.P.L., M.Y., J.Z., and T.H.L. helped data analysis. T.Y. and M.Y. conducted the DFT calculations. All authors read and approved of the paper.

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Supplementary Materials

Materials and Methods

Supplementary Text

Figs. S1 to S25

Tables S1

References (*45-58*)

Fig. 1. 4D-STEM result of polar field in twisted bilayer MoS_2. **(A)** Schematic of moiré structure emergence from the twist stacking of two individual layers of MoS_2. The AB and BA stacking area are highlighted by orange and purple color, respectively. STEM annular dark field (ADF) images of synthesized moiré structures of bilayer MoS_2, with twist angles **(B)** θ_t~7.83°, **(C)** θ_t~11.80°, and **(D)** θ_t~24.57°. **(E)** A scheme of the polar field distribution with clockwise and anti-clockwise vortex, respectively. The arrows represent the in-plane component of the polarization. **(F)** The 3D map of the polar field distribution of twisted bilayer MoS_2 at the AA, AB and BA area. The arrow denotes the direction of polarity. **(G)** The 4D-STEM curl (vorticity) mapping of the polar field $(-1\times\nabla\times \boldsymbol{E}_{xy})_{[001]}$ with polar vectors for bilayer MoS_2 with the twist angle of θ_t=11.6°. The blue color denotes clockwise rotation, while red color denotes anticlockwise rotation. The inset is a STEM-ADF image corresponding to atomic structure at the hexagonal lattice. The dashed line illustrates the boundaries between the AA, AB and BA stacking area in the twisted bilayer. **(H)** Zoomed-in 4D-STEM polar mapping with polar vectors presents noticeable in-plane vortex and anti-vortex structures at the AA, AB and BA stacking area. **(G)** and **(H)** share the same intensity bar.

Fig. 2 Twist-angle dependence of polar field distributions. (**A-D**) The experimental STEM-ADF images of bilayer MoS_2 with different twist angles (θ_t= 8.3°, 11.6°, -18.4° and 30°, respectively). (**E-H**) Corresponding curl (vorticity) mapping of polar field based on the 4D-STEM experimental results. (**I-L**) Multislice simulations of STEM-ADF results with the same twist angles shown in (**A-D**). (**N-P**) Corresponding curl (vorticity) mapping of polar field based on the multislice simulation method. **(Q)** The twist angle-dependent polar field varies across all twist angles in a specified 0.4 nm × 0.4 nm region. This angular dependent polar plot presents the variation in vortex tendency (vorticity). 120 models of MoS_2, each featuring a different twist angle ranging from 0° to 119° with an incremental step of 1°, were created. The chosen 0.4 nm × 0.4 nm region, positioned within the AA stacking area and labelled as the grid mask of MoS_2, serves as the central point for rotation transformation in each twisted sample. The orange line on the plot corresponds to the average vorticity as a function of the twist angle, illustrating how the vorticity evolves with the twist angle. The additional background color is used to indicate vorticity directions—clockwise (blue) and anti-clockwise (red) for better recognition of peaks and trends across a wide twist angle range. This comparison allows for an evaluation of how the polar field varies

with the twist angle in the specifically selected AA stacking region. The four diagrams below represent the unique vortex features at the angles of θ_t~8.3°, θ_t~17.9°, θ_t~30°, and θ_t~ 50.6°, respectively, which show the vorticities of the rotation centre changing from clockwise to anti-clockwise. Scale bars, 1 nm.

Fig. 3 The basic tiles and charge density distribution of 30° twist bilayer MoS_2 quasicrystal. (A) The grid structure, atomic structure, and STEM-ADF image of Stampfli tiles in 30° twisted bilayer quasicrystal MoS_2. **(B)** Experimental STEM-ADF image of 30° twisted bilayer quasicrystal MoS_2 mapped with false-colored 12-fold Stampfli tiles. **(C)** Experimental and **(D)** DFT simulated charge density contour plot in 30° twisted bilayer quasicrystal MoS_2. The superimposed gridlines refer to the perfect Stampfli tiles. Scale bars, 1 nm.

Fig. 4. In situ TEM manipulation of Stampfli tiles in 30° twisted bilayer quasicrystal MoS_2. (A) Schematic for the alteration of Stampfli tiles before and after 100-pm interlayer shift in 30° twisted bilayer quasicrystal MoS_2. A lateral displacement of 100 pm in the top layer relative to the bottom layer results in a notable alteration of the Stampfli tiles, as the pattern shifts towards the right by 636 pm. **(B)** Experimental interlayer displacement mapping for bilayer MoS_2 upon manipulation by in situ STEM technique, corresponding to (**C**) and (**D**). The color bar represents the magnitude of local relative displacements, as determined by difference of Mo atomic positions before and after manipulation. **(C)** The STEM-ADF snapshot of Stampfli tiles in the original 30° twisted bilayer quasicrystal MoS_2. **(D)** The STEM-ADF snapshot of Stampfli tiles at the same area as shown in (**C**) after in situ STEM manipulation. The Stampfli tiles are typed as rhombus (light pink), square (green), and triangle (yellow). The gridlines illustrate patterns with (white) and without (black) severe changes after manipulation, respectively. The red arrow in (**D**) highlights the upward shift of a hexagon by 1117 pm after manipulation. Scale bars in (**B-D**) are 1 nm.