

Supplementary information for

Ferromagnetism and correlated insulating states in

monolayer $\text{Mo}_{33}\text{Te}_{56}$

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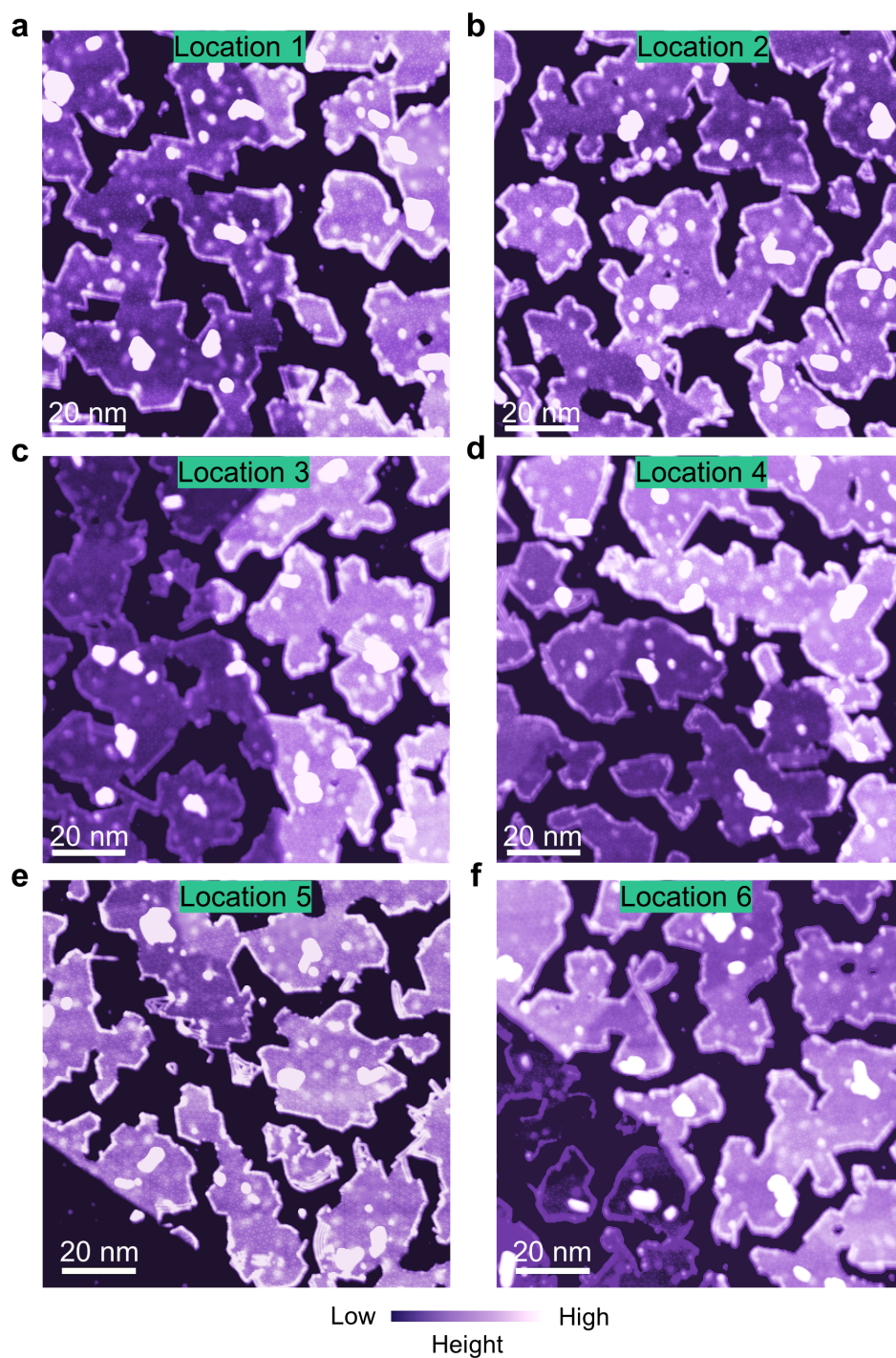


Fig. S1. a-f, High-resolution STM images of monolayer MoTe_{2-x} acquired at six random locations on an as-grown sample [see preparation recipe in the Methods section]. We used the coarse motion motor to ensure large separations (at least by hundreds of micrometers) among these six locations. In all images, the superlattice with a uniform 1.91-nm periodicity is observed as an overwhelming majority. Among the various Mo-Te phases¹, only the Mo_6Te_6 wires appear at the very edge of 2D flakes; all other ones

34 (*e.g.*, MTBs with different sizes, Mo_5Te_8 , MoTe_2) are nearly absent in this sample. The
35 bright spots are amorphous residuals on top of ML- MoTe_{2-x} . The above observations
36 confirmed the relative thermal preference of the $\text{Mo}_{33}\text{Te}_{56}$ phase, and there shall be an
37 accessible growth window to prepare the scalable $\text{Mo}_{33}\text{Te}_{56}$ sample. Scanning
38 parameters: $V_{\text{bias}} = 0.8 \text{ V}$, $I_t = 20 \text{ pA}$; size: $120 \times 120 \text{ nm}^2$.

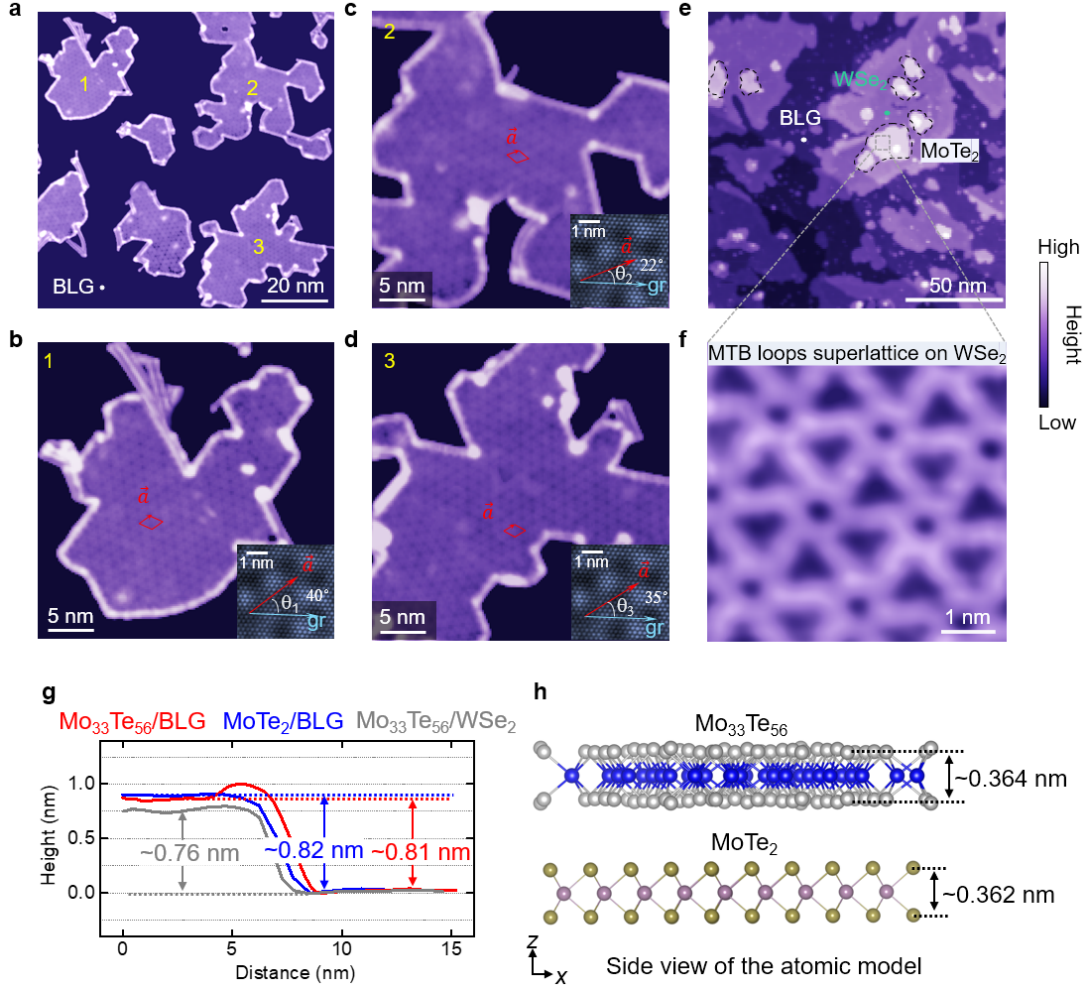


Fig. S2. The evidence to rule out the possibility of a moiré pattern. We note that some recent works identified the similar structures in MoTe₂ as a moiré pattern². Here, we provide experimental evidence to rule out this possibility. **a**, A large-scale STM image of the ordered MTB superlattice. Three islands are marked as 1-3. **b-d** are close-up images of islands 1-3 with the supercell vectors $\mathbf{a}$ marked as shown. The insets in **b-d** are the atomically resolved images taken on the adjacent graphene. The blue arrows represent one zigzag direction of the graphene honeycomb. The twist angles between the MTB superlattice and the graphene atomic lattice can thus be determined as $\theta_1 \sim 40^\circ$ (island 1), $\theta_2 \sim 22^\circ$ (island 2), and $\theta_3 \sim 35^\circ$ (island 3), respectively. **e**, Large-scale STM image of the ML-MoTe₂ grown on ML-WSe₂. Dashed black lines encircle the MoTe₂ islands. The growth parameters are the same as those of ML-MoTe₂ grown on graphene. **f** is the close-up image taken on MoTe₂/WSe₂ (corresponding to the grey

square in e), exhibiting the same superstructures. These results prove the observed super periodicity ($a = 1.91$ nm) is independent of the twist angle and the mismatch in lattice constants, and thus, cannot be a moiré pattern. Scanning parameters: (a-d) $V_{\text{bias}} = 1$ V, $I_t = 50$ pA. (e) 3 V, 10 pA. (f) 2 V, 50 pA. g, Height profiles of $\text{Mo}_{33}\text{Te}_{56}/\text{BLG}$, MoTe_2/BLG , and $\text{Mo}_{33}\text{Te}_{56}/\text{WSe}_2$. In the red curve, the slightly elevated bump near the edge of the island is attributed to the formation of one-dimensional Mo_6Te_6 nanowires. h, Side views of theoretical models of $\text{Mo}_{33}\text{Te}_{56}$ and MoTe_2 . The distance between the centres of top- and bottom-layer Te atoms is labeled as shown.

Fig. S2g presents the STM measurements of the height difference between the $\text{Mo}_{33}\text{Te}_{56}$ phase and the underlying BLG substrate, which is approximately 0.81 nm (red curve). For the ML- MoTe_2 phase, the height difference is measured to be 0.82 nm (blue curve). This value is consistent with previous reports for monolayer MoTe_2 on BLG, which is typically slightly greater than 0.8 nm²⁻⁴. This consistency further supports the reliability of our STM height measurements.

When the substrate is replaced with WSe_2 , the height of the $\text{Mo}_{33}\text{Te}_{56}$ phase is slightly reduced to approximately 0.76 nm (grey curve in Fig. S2g). Since the pure MoTe_2 phase was not intentionally grown on WSe_2 substrates, corresponding height data are not available. Given the differing chemical potentials of substrates, such minor variations in height between the BLG and WSe_2 substrates fall within the range of expected experimental results. Similar height variations, on the order of tens of picometers, have been reported for monolayer TMDs grown on various substrates^{5,6}.

Our experiments reveal that the heights of $\text{Mo}_{33}\text{Te}_{56}$ and MoTe_2 are nearly identical. This observation can be further understood through theoretical structural models of the two materials. As shown in the side-view structural models in Fig. S2h, the presence of MTBs in $\text{Mo}_{33}\text{Te}_{56}$ induces slight corrugation in the z -direction. However, the overall thickness of $\text{Mo}_{33}\text{Te}_{56}$ shall not differ significantly from that of MoTe_2 . Specifically, when measuring the distance between the centres of the top-layer and bottom-layer Te atoms, we obtain 0.364 nm for ML- $\text{Mo}_{33}\text{Te}_{56}$ (average value) and 0.362 nm for ML- MoTe_2 , highlighting the minor thickness differences between the two phases.

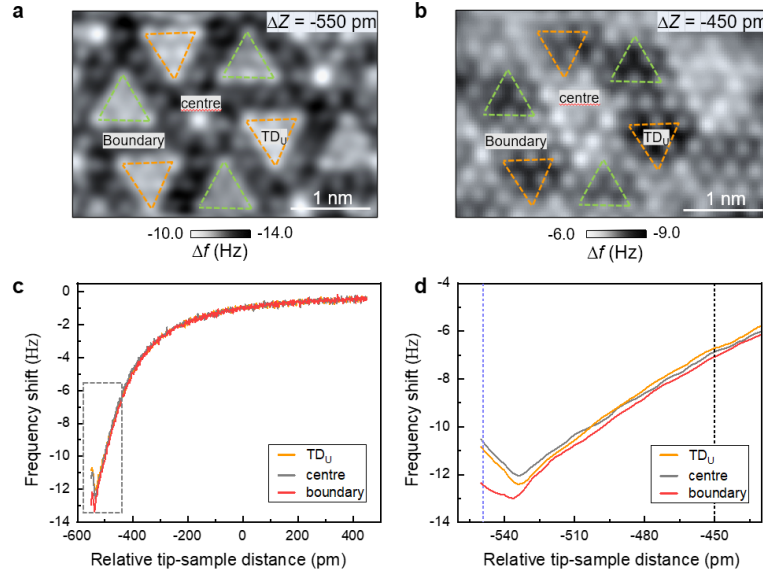


Fig. S3. AFM images and force spectroscopic measurements. **a, b**, Constant height Δf images acquired on the MTB superlattice with different relative tip heights (as marked). **c**, Frequency shift as a function of tip-sample distance measured on the TDU (orange), the centre site (grey), and the boundary site (red). **d**, Zoomed-in spectra recorded at the grey dashed square in **c**. The purple/black dashed vertical line in **d** corresponds to the ΔZ value used to acquire **a/b**, respectively, illustrating the **a/b** was taken with repulsive/attractive force.

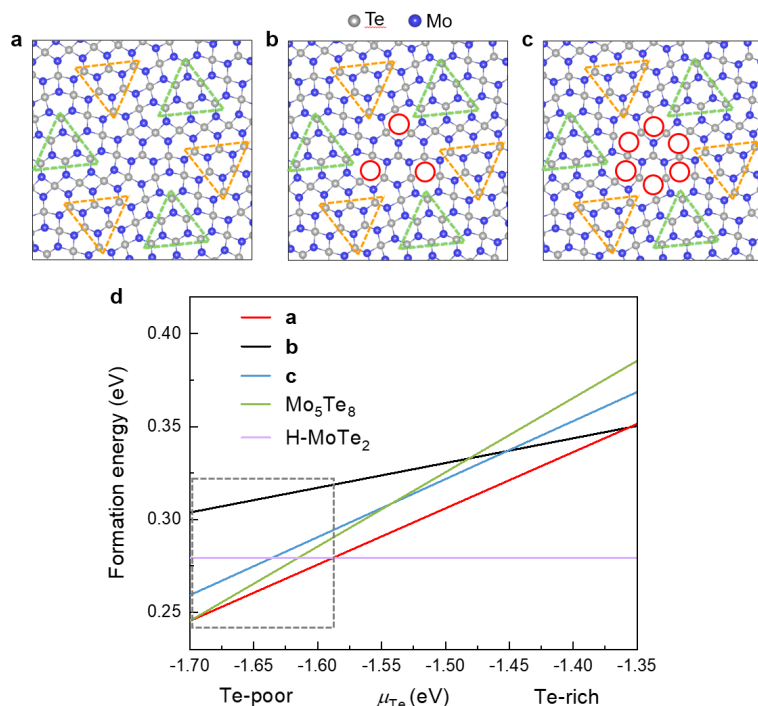


Fig. S4. Formation energies of different Mo-Te compounds. **a-c**, Three atomic models of the “wagon-wheel” structure. They are different in the number of Mo atoms missing near the centre region (indicated by red circles). **a**, No Mo vacancies⁷; **b**, Three Mo vacancies⁸; **c**, Six Mo vacancies⁹. Since the scanning probe microscope (STM/AFM) only images the top layer atoms, one is not able to distinguish them through the atomically resolved imaging as shown in Fig. S3a and b. **d**, DFT calculated the formation energies of the following structures: model (**a**) (red line), model (**b**) (black line), model (**c**) (blue line), Mo_5Te_8 (green line), and pristine H-MoTe₂ (purple line). They are plotted as a function of Te chemical potential μ_{Te} , simulating the uncertainty in experimental growth parameters (*i.e.*, the Te-poor and Te-rich conditions).

The scanning probe measurements (STM&AFM; Fig. 1d and e) display that the as-grown superlattice exhibits a "wagon-wheel" pattern, whose size and concentration are thermally dependent. This pattern is a well-known hallmark of the MTB network structure. The MTB itself has a well-established structural model where the row of chalcogen atoms is centred along the boundary and each chalcogen atom is bound to four metal atoms instead of three. In our work, the $\text{Mo}_{33}\text{Te}_{56}$ superlattice exhibits a periodicity of 1.91 nm (Fig. 1f). The atomic-resolution AFM measurement further

reveals that the triangular domain consists of six surface Te atoms (*i.e.*, three Mo-Te units), with an interatomic distance of 0.365 nm. These findings enable an accurate determination of the atomic structure of the boundary and domain. However, regarding the structure at the centre of the "wagon-wheel" network, there remain uncertainties in the literature lie in the number of Mo vacancies. Depending on the specific fabrication process and the domain size, previous studies have proposed various configurations, including no Mo vacancies (a)⁷, three Mo vacancies (b)⁸, and six Mo vacancies (c)⁹.

Determining which of these three configurations best represents the observed structure is the central issue in confirming the stoichiometry. Experimentally, one of the most prominent features for identifying the presence of Mo vacancies is the distinct contrast at the central region of the "wagon-wheel" pattern, as observed in AFM images. For example, in ref. 9, three out of six Te hollow sites at the centre appear significantly darker than the surrounding positions (also using a standard W tip), a feature attributed to the presence of Mo vacancies. In our AFM experiments, we did not observe similar prominent contrast variations at the central region. This observation strongly suggests that the structure we obtained does not contain Mo vacancies. The simulated frequency-shift AFM image based on the no-Mo-vacancies model closely resembles this experimental feature (as shown in main text Fig. 1d and e).

To understand this experimental observation and further validate the atomic model, we performed a theoretical comparison of the formation energies of three possible structural models as a function of the Te chemical potential, aiming to comprehensively simulate the experimental growth conditions (Fig. S4a-c). Our calculations reveal that, for the observed superlattice size, the model without Mo vacancies (Fig. S4d) exhibits significant lower formation energies than the other two models (as depicted by the red curve versus the black & blue curves). In addition, the consistencies established between our theoretical predictions and experimental observations, particularly regarding the energy-dependent dI/dV spectra (Fig. 1h and Fig. S5), further provide robust support for the validity of our proposed model.

Based on this model, we determined the stoichiometry of the new phase by counting the number of Mo and Te atoms in the unit cell (Fig. S4a and Fig. 1h). This analysis

- 137 reveals 33 Mo atoms and 56 Te atoms per unit cell, identifying the stoichiometry as
- 138 $\text{Mo}_{33}\text{Te}_{56}$.

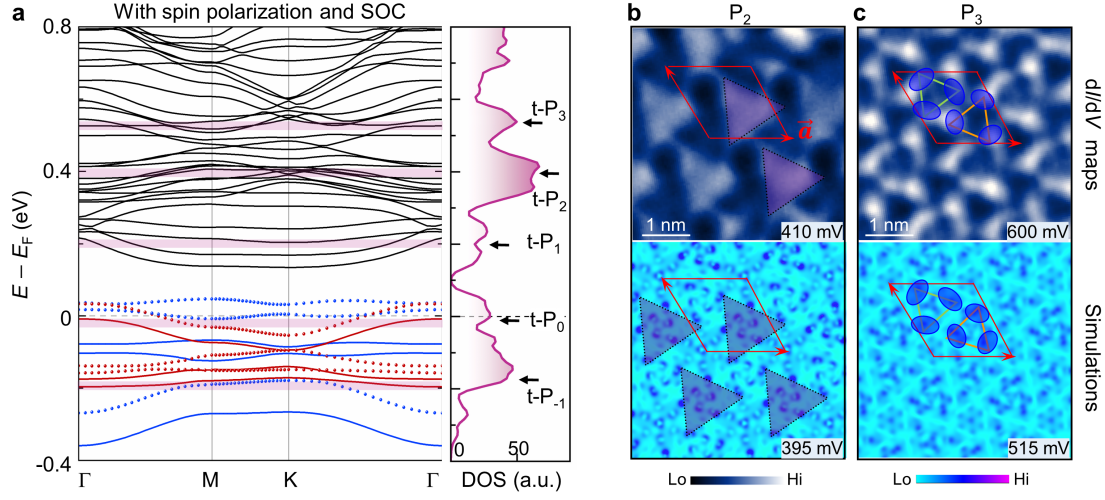


Fig. S5. Spatial distributions of the electronic states at P_2 and P_3 . **a**, Calculated band structures with spin polarization and SOC. The energy ranges used for simulations are set at ± 5 mV (as the pink shadows indicate). The five regions are marked as $t-P_{-1}$ to $t-P_3$. **b**, **c**, Upper panel: measured dI/dV maps at energies of +410 mV (P_2) and +600 mV (P_3). Lower panel: simulated dI/dV maps at +395 mV ($t-P_2$) and +515 mV ($t-P_3$). Other measured and simulated states are shown in the main text, Fig. 2c-e, where the simulation energies are set as -185 meV, -35 meV and 200 meV for P_{-1} , P_0 and P_1 , respectively. The energy ranges utilized in these simulations are set at ± 5 mV. They display the general agreements between the dI/dV maps and the simulations (**b**, **c**). Although a strict resemblance is hampered by the highly mixed band structures at these energy levels, the major experimental features are well captured in the simulations. In **b**, the P_2 state sees that a large triangular protrusion forms, as marked by the purple shadow. In **c**, the electron states of P_3 are mostly distributed at the corners of each triangular domain (highlighted by the blue ovals), while the TD_E centre (green triangles) is slightly higher than the TD_U centre (orange triangles). The present consistencies meet the prevailing standard in the STM&DFT combined studies of new materials. The red rhombus in **b** and **c** labels a supercell. Stabilization parameters in **b**, $I_t = 100$ pA, $V_{\text{mod}} = 6$ mV; **c**, $I_t = 100$ pA, $V_{\text{mod}} = 8$ mV.

Supplementary Note 1.

The uniformly formed superlattice at the mesoscale is of critical importance for future investigations and applications, which is a key advance of our system compared with other proposed materials. For instance, an emergent honeycomb network in the charge-density-wave phase of 1T-TaS₂ was discussed in ref. 10. While there appear to be some similarities between this work and ours, we emphasize the essential differences in the following: (1) In ref. 10, the network state appears along domain walls, which are topological excitations of charge density waves. It is unclear, at least to us, how such topological excitations (subjected to thermal fluctuations and disorder) can form an ordered lattice at the mesoscale in a controlled manner. By contrast, the Mo₃₃Te₅₆ monolayer we discovered here has a thermally stable ordered lattice structure. (2) The STM results in ref. 10 reflected the local physics along domain walls; the emergent honeycomb network was only a theoretical proposal for 1T-TaS₂. By sharp contrast, we unveil that the Mo₃₃Te₅₆ monolayer, as a whole system, behaves as a correlated insulator.

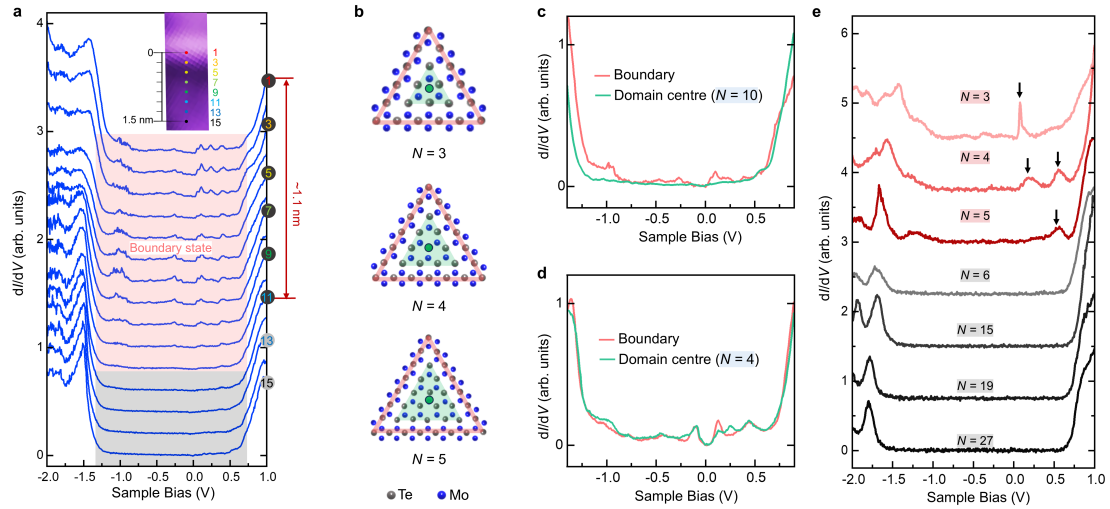


Fig. S6. a, The dI/dV were measurements taken across an MTB with a nearly infinite length. A series of dI/dV spectra were recorded from an MTB to the nearby domain region (along the dotted line in the inset). The total distance from #1 to #15 is 1.5 nm. The boundary states are maintained until spectrum #11 (pink shadow), corresponding to a penetration distance d_p of ~ 1.1 nm. Spectrum #12 – #15 (grey shadow) show nearly the same band structure as pristine MoTe₂. In **b-e**, we illustrate the size-dependent

179 electronic properties of the triangular domains enclosed by MTB loops. **b**, Atomic
 180 models of triangular domains with different sizes ($N = 3, 4$, and 5), where N denotes the
 181 number of Te atoms at the domain (*i.e.*, the green triangles) edges. The Te rows in the
 182 middle of the boundaries are highlighted with pink lines for clarity. **c**, **d**, dI/dV spectra
 183 at the boundary (pink) and centre (green dots) of triangular domains with sizes $N = 10$
 184 (**c**) and $N = 4$ (**d**). When the triangular domain size is sufficiently large in comparison
 185 with d_p (as in the case of $N = 10$ in **c**), the centre of the domain retains the intrinsic
 186 semiconducting band structure. However, as the domain size decreases (e.g., $N = 4$ in
 187 **d**), boundary states interact, leading to extended electronic states even at the domain
 188 centre. **e**, A series of dI/dV spectra recorded at the domain centre with different N
 189 numbers. This illustrates how localized 1D boundary states extend into the inner
 190 domain as the domain size shrinks. The threshold size for entirely extended in-gap states
 191 (black arrows) is $N = 5$. Note that the spectra here were taken at locations without long-
 192 range ordered superperiodicity, causing the line shape for $N = 3$ in **e** to differ from those
 193 in the main text. Scanning parameter in the inset of **a**: $V_{\text{bias}} = 2.0$ V, $I_t = 10$ pA.
 194 Stabilization parameters in **a**: $V_{\text{bias}} = 1.0$ V, $I_t = 200$ pA, $V_{\text{mod}} = 10$ mV; (**c-e**) $V_{\text{bias}} = 1.0$
 195 V, $I_t = 100$ pA, $V_{\text{mod}} = 10$ mV.

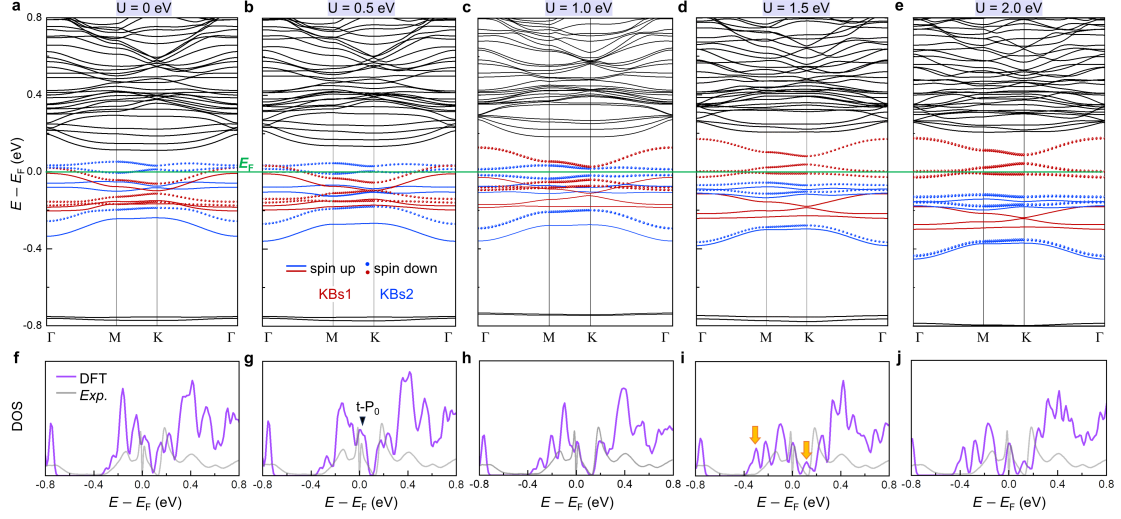


Fig. S7. Discussion for the on-site repulsion energy U . In **a-e**, we present the band structures (with spin polarization and SOC) obtained with Hubbard $U = 0, 0.5$ eV, 1.0 eV, 1.5 eV, and 2.0 eV. Here, U is applied to individual atomic orbitals. The corresponding DOS profiles (purple lines) are displayed in **f-j**, with the experimental spectrum superimposed (grey lines). When $U > 1.0$ eV, a pronounced DOS intensity below -0.2 eV and near $+0.15$ eV range emerge (marked by orange arrows in **i** and **j**). These features contrast significantly with experimental observations, which show a flat and featureless line shape within the two energy ranges (-0.55 V to -0.32 V, and 0.12 V to 0.14 V). Thus, we conclude that a relatively small U value is sufficient for our system, consistent with recent studies on Mo-Te compounds^{11,12}. The calculated results with $U = 0.5$ eV (**b, g**) give the best agreement with the spectroscopic line shape, except for the zero-bias dip.

Note that all calculations presented in our work were performed without accounting for the graphene substrate. The choice of this simplified model was based on the following consideration regarding the potential effects of Mo₃₃Te₅₆-graphene interactions.

1) Interfacial charge transfer: Preliminary calculations for Mo₃₃Te₅₆-interface heterostructure with a rotational angle of 0° . The results indicate a neglectable charge transfer of approximately 0.25 e /supercell (< 0.01 e /Mo), which keeps the kagome band structures of Mo₃₃Te₅₆ intact. A similarly small value was also reported for interactions

between graphene and single-layer chalcogenides, as noted in previous work^{13,14}.

2) In-plane strain: Our experimental observations reveal that the $\text{Mo}_{33}\text{Te}_{56}$ monolayers exhibit randomly distributed twist angles with respect to the graphene substrate. This randomness suggests that no epitaxial relationship is established (see Fig. S2a-d), making it unlikely for the graphene substrate to induce significant in-plane strain on the $\text{Mo}_{33}\text{Te}_{56}$ layers.

3) Dielectric screening: The screening effect introduced by the graphene substrate can be approximated by adjusting the effective on-site Hubbard U value in our calculations. We found that setting $U = 0.5$ eV successfully reproduces the experimental results (Fig. S7b, g), effectively capturing the screening effect without the need for explicit graphene. What's more, the parent state of $\text{Mo}_{33}\text{Te}_{56}$ exhibits a high density of states at the Fermi level (Fig. S7g), a feature that remains largely unaffected by these substrate-related effects.

Supplementary Note 2 Tight-binding (TB) simulation.

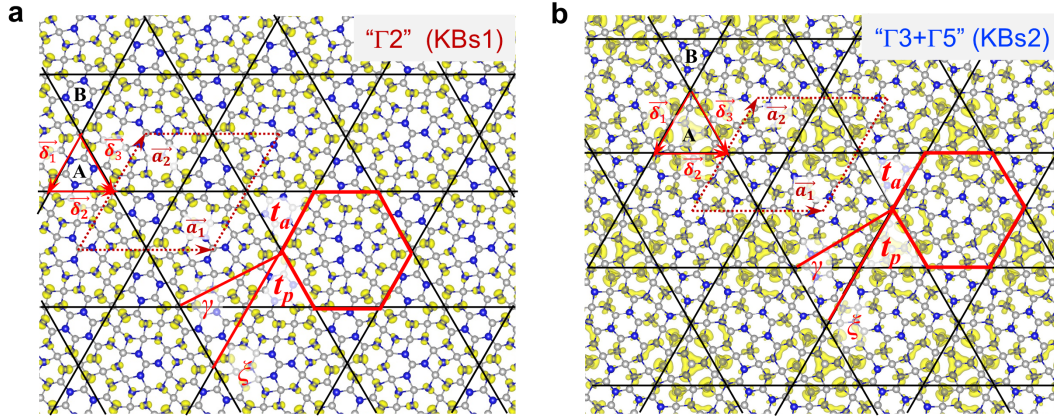


Fig. S8. We choose “ $\Gamma 2$ ” states (KBs1) and “ $\Gamma 3 + \Gamma 5$ ” states (KBs2) as the basis in the following tight-binding simulation of the kagome lattice. For each set of flat bands, the real-space charge densities shall have very similar distributions at all other k points. The in-plane charge densities distributions are displayed in **a** (KBs1) and **b** (KBs2), with the atomic lattice structures superimposed. Here, $\vec{a}_1$ and $\vec{a}_2$ are the lattice vectors, t_p and t_a are the nearest-neighbor (NN) hopping energy inside the triangles A and B, respectively, and γ and ξ are the next-nearest-neighbor (NNN) and third-nearest-neighbor (TNN) hopping strengths, respectively. The black lines illustrate the real-space geometry of the kagome lattices (depicted by the vectors of $\vec{\delta}_{1-3}$). Details are seen in the following discussion. Noted that due to the chiral atomic structure, the charge density profiles also display in-plane chirality (*i.e.*, mirror symmetries breaking).

A classic kagome lattice, consisting of two corner-sharing triangles of identical size, exhibits mirror symmetry in both the x and y directions. However, this symmetry can be broken through specific distortions, while the flatness of the electronic bands is largely preserved. A well-known example of this is the breathing kagome lattice, where the two triangles are unequal in size, leading to partial breaking of mirror symmetry (with respect to the y direction). Further tilting of the two triangles can completely remove the mirror symmetry. However, the MTB superlattice has the threefold rotational C_3 symmetry, the horizontal reflection σ_h and the broken inversion symmetry, but it has no vertical mirror plane σ_v . Therefore, the MTB superlattice has

an in-plane chiral structure. The lattice structure of MTB superlattice with in-plane chiral charge densities derived from KBs1 and KBs2 are plotted in Fig. S8a and b, respectively. The vertical mirror symmetry of the geometrical kagome lattice is broken by the different NN hopping energy t_p and t_a [see following Eq. (2)] and the basis of these chiral charge density orbitals. These lattice-dependent of the geometrical kagome lattice read:

$$\begin{aligned}\vec{a}_1 &= a(1, 0), \vec{a}_2 = a\left(\frac{1}{2}, \frac{\sqrt{3}}{2}\right), \\ \vec{\delta}_1 &= \frac{a}{2}\left(-\frac{1}{2}, -\frac{\sqrt{3}}{2}\right), \vec{\delta}_2 = \frac{a}{2}(1, 0), \vec{\delta}_3 = \frac{a}{2}\left(\frac{1}{2}, -\frac{\sqrt{3}}{2}\right), \\ \vec{\delta}_4 &= \frac{\sqrt{3}a}{2}\left(\frac{\sqrt{3}}{2}, -\frac{1}{2}\right), \vec{\delta}_5 = \frac{\sqrt{3}a}{2}(0, 1), \vec{\delta}_6 = \frac{\sqrt{3}a}{2}\left(\frac{\sqrt{3}}{2}, \frac{1}{2}\right),\end{aligned}\quad (1)$$

where $a = 19.0614\text{\AA}$ is the lattice constant. The TB Hamiltonian reads

$$H = \sum_i \varepsilon_i c_i^\dagger c_i + \sum_{\langle i, j \rangle} t(\mathbf{r}_{ij}) c_i^\dagger c_j + \sum_{\langle\langle i, j \rangle\rangle} \gamma c_i^\dagger c_j + \sum_{\langle\langle\langle i, j \rangle\rangle\rangle} \xi c_i^\dagger c_j, \quad (2)$$

where ε_i is the on-site energy with the same value ε for three sublattices of the kagome unit cell for C_3 symmetry, $\langle i, j \rangle$ denotes the NN interaction, $\langle\langle i, j \rangle\rangle$ and $\langle\langle\langle i, j \rangle\rangle\rangle$ represent the NNN with hopping γ and TNN with hopping ξ , respectively, and t_p and t_a correspondingly represent the NN hoppings inside the triangles A and B. For simplicity, we only consider the TB model without spin-orbital coupling (see Fig.

2d in the main text). In $\vec{k}$ space, the Hamiltonian $H_{wo}(\vec{k})$ reads

$$H_{wo}(\mathbf{k}) = \begin{bmatrix} \varepsilon + 2\xi\phi(k_1, k_2, k_3) & t_p e^{ik_1} + t_a e^{-ik_1} + 2\gamma\cos(k_4) & t_p e^{ik_3} + t_a e^{-ik_3} + 2\gamma\cos(k_6) \\ t_p e^{-ik_1} + t_a e^{ik_1} + 2\gamma\cos(k_4) & \varepsilon + 2\xi\phi(k_1, k_2, k_3) & t_p e^{ik_2} + t_a e^{-ik_2} + 2\gamma\cos(k_5) \\ t_p e^{-ik_3} + t_a e^{ik_3} + 2\gamma\cos(k_6) & t_p e^{-ik_2} + t_a e^{ik_2} + 2\gamma\cos(k_5) & \varepsilon + 2\xi\phi(k_1, k_2, k_3) \end{bmatrix} \quad (3)$$

where $k_i = \vec{k} \cdot \vec{\delta}_i$ with $i = 1, \dots, 6$ and $\phi(k_1, k_2, k_3) = \cos(2k_1) + \cos(2k_2) + \cos(2k_3)$. At Γ point the Hamiltonian H_{wo}^Γ reads

$$H_{wo}^\Gamma = \begin{bmatrix} \varepsilon + 6\xi & t_p + t_a + 2\gamma & t_p + t_a + 2\gamma \\ t_p + t_a + 2\gamma & \varepsilon + 6\xi & t_p + t_a + 2\gamma \\ t_p + t_a + 2\gamma & t_p + t_a + 2\gamma & \varepsilon + 6\xi \end{bmatrix}, \quad (4)$$

Then we write the eigen energy at Γ point as

$$\begin{aligned}
E_1^\Gamma &= \varepsilon + 6\xi + 2(t_p + t_a + 2\gamma), \\
E_2^\Gamma &= \varepsilon + 6\xi - (t_p + t_a + 2\gamma), \\
E_3^\Gamma &= \varepsilon + 6\xi - (t_p + t_a + 2\gamma).
\end{aligned} \tag{5}$$

At K point with $K = \frac{2\pi}{a}(\frac{2}{3}, 0)$, we have $k_1 = -\frac{\pi}{3}, k_2 = \frac{2\pi}{3}, k_3 = \frac{\pi}{3}, k_4 = \pi, k_5 = 0$, and $k_6 = \pi$. The Hamiltonian H_{wo}^K reads

$$H_{wo}^K = \begin{bmatrix} \varepsilon - 3\xi & t_p e^{-i\frac{\pi}{3}} + t_a e^{i\frac{\pi}{3}} - 2\gamma & t_p e^{i\frac{\pi}{3}} + t_a e^{-i\frac{\pi}{3}} - 2\gamma \\ t_p e^{i\frac{\pi}{3}} + t_a e^{-i\frac{\pi}{3}} - 2\gamma & \varepsilon - 3\xi & t_p e^{i\frac{2\pi}{3}} + t_a e^{-i\frac{2\pi}{3}} + 2\gamma \\ t_p e^{-i\frac{\pi}{3}} + t_a e^{i\frac{\pi}{3}} - 2\gamma & t_p e^{-i\frac{2\pi}{3}} + t_a e^{i\frac{2\pi}{3}} + 2\gamma & \varepsilon - 3\xi \end{bmatrix} \tag{6}$$

Then we write the eigen energy at K point as

$$\begin{aligned}
E_1^K &= \varepsilon - 3\xi + 2t_p - t_a - 2\gamma, \\
E_2^K &= \varepsilon - 3\xi - t_p + 2t_a - 2\gamma, \\
E_3^K &= \varepsilon - 3\xi - t_p - t_a + 4\gamma.
\end{aligned} \tag{7}$$

After solving the energy spectrum equations from the TB model in Eqs. (4) and (6) and the DFT calculation results at Γ and K points, we have $\varepsilon = -0.11956$ eV, $t_p = 0.03584$ eV, $t_a = 0.01978$ eV, $\gamma = 0.00251$ eV, and $\xi = 0.001215$ eV for KBs1, and $\varepsilon = -0.07463$ eV, $t_p = -0.0118$ eV, $t_a = -0.07093$ eV, $\gamma = -0.00583$ eV, and $\xi = -0.003378$ eV for KBs2. As shown in main text Fig. 2a, we see that the TB kagome lattice model with in-plane chirality reproduces mostly the DFT energy spectrum.

In the main text (Fig. 2c), we employed a line graph representation of the boundary network to provide a simple and easily understandable schematic illustration in real space. In TB fitting, there is a degree of flexibility in lattice site selection that does not affect the main conclusions. The lattice sites need not align with the maximum DOS locations. Instead, we adopt a more general approach by first creating a standard kagome lattice in real space (as shown by the black grid in Fig. S8 with the lattice site near the boundary centre). From this standard structure, we obtain the breathing pattern ($t_a \neq t_p$) by fitting the hopping parameters. This method, grounded in the atomic

294 structure's symmetry, effectively captures the essential features of the electronic
295 kagome pattern.

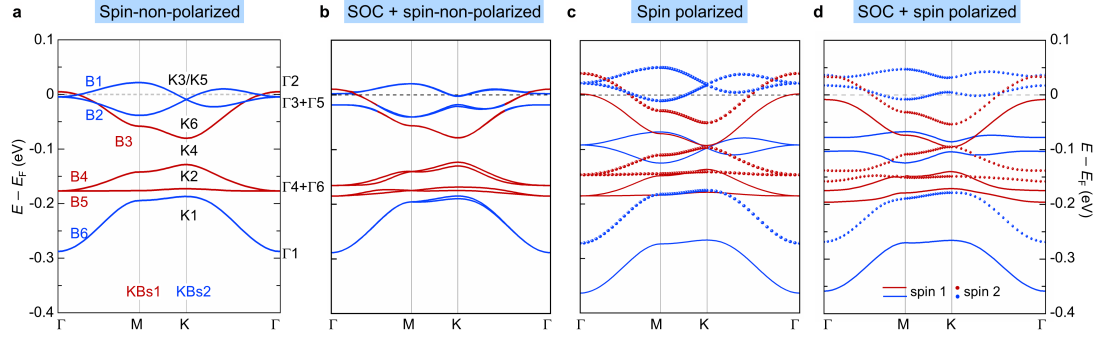


Fig. S9. Systematic calculation results illustrate the roles of the spin-orbit coupling and spin polarization on the band structures. The spin-non-polarized band structure for the $\text{Mo}_{33}\text{Te}_{56}$ monolayer is plotted in **a** (the same one as in Fig. 2a). Inclusion of SOC slightly lifts the degeneracy of the two spins in band sets KBs1 and KBs2 (comparing **b** with **a**), but the Γ points are degenerate between the Dirac band and the flat band still. With spin polarization included (**c**), the spin exchange splits the two spin components by an order of 0.1 eV (see the Γ point of B6), and the system now contains net spin polarization (*i.e.*, $n_{\text{up}} \neq n_{\text{down}}$). The most significant splitting was observable for bands B1 and B2 from set KBs2 residing near the Fermi level where the two bands open gaps at points Γ (21 meV) and K (33 meV). Additional consideration of SOC in **d** does not qualitatively change the band structures near the Fermi level, but the originally partially filled B1 in **c** becomes entirely unoccupied and the originally fully occupied B2 turns into partially occupied.

The relative positions of bands B1, B2 and B3 are highly U -dependent, which slightly modifies the filling status of bands B1 and B2. For $U = 1$ eV, the Fermi level is right across the gap between bands B1 and B2 (see Fig. S9c, width ranging from 19 to 55 meV). Although, this result could, perhaps, well explain the experimentally observed bandgap, we do not prefer to overclaim this result. Higher level calculation methods, *e.g.*, DFT+DMFT or +DMRG should be used to verify this speculation, which is, however, beyond the scope of this work.

The Stoner criterion calculation indicates the spin-polarized bands sitting near the Fermi level is complying with the spontaneous magnetization of partially filled (nearly) flat-bands originated from the kagome-variant structure. Although bands B1 and B2

should, in principle, be one nominal flat-band and one nominal Dirac band, their shapes and dispersions are nearly mirror symmetrized to an energy level slightly off the Fermi level. Thus, we prefer to conclude that either B1 or B2 is partially comprised of the flat-band and the Dirac band, which differentiates the spontaneous magnetization found in the present work from that found in twisted bilayer graphene where the spontaneous magnetization was resulted from flattened Dirac bands.

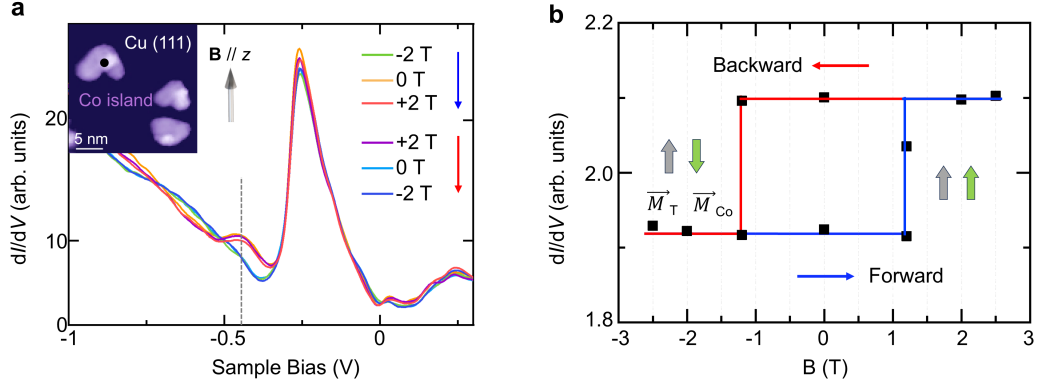


Fig. S10. Calibration of the spin-polarized Cr-coated W tip on Co/Cu (111). **a**, Spin-polarized tunnelling spectra measured on the Co islands at magnetic fields of ± 2 T and 0 T using the Cr-coated W tip. Inset: an STM image showing the topography of the Co islands on Cu (111). **b** shows the ferromagnetic hysteresis loop of the dI/dV signal at $V = -490$ mV (grey dashed marker in **a**). This loop feature of the states at ~ 500 mV is consistent with the well-known properties of the Cr-coated W tip on Co/Cu (111), as reported in previous studies^{15,16}, where the Co island has a coercive field of around 1 T. By doing so, we demonstrate that the tip magnetization $\vec{M}_T$ (grey arrow) is along the $+z$ direction, and remains unchanged in a sufficiently large field (compare to the $\mathbf{B}$ used in Fig. 3). Red (blue) arrows indicate that the magnetic field was swept from positive to negative (negative to positive) values, *i.e.*, in a backward (forward) direction. Scanning parameters in the inset of **a** are $V_{\text{bias}} = 1.0$ V, $I_t = 20$ pA. The stabilization parameters in **a** are $V_{\text{bias}} = 1$ V, $I_t = 200$ pA, and $V_{\text{mod}} = 8$ mV.

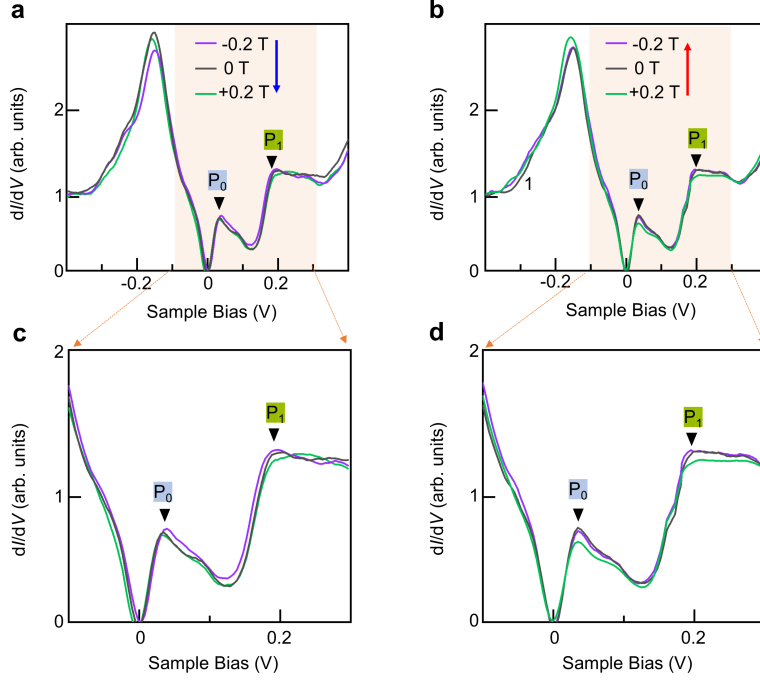


Fig. S11. Additional data of P_0 and P_1 at varied magnetic fields. For P_{-1} , we performed magnetic field sweeps with very fine step sizes, as discussed in the main text. For P_0 and P_1 , we checked the spectra at -0.2 T, 0 T, $+0.2$ T, and back to 0 T and -0.2 T. **a, b**, Tunnelling spectra obtained on the boundary site at magnetic fields of ± 0.2 T and 0 T, with the magnetic field swept from negative to positive (**a**), and positive to negative values (**b**), respectively. Their corresponding zoomed-in tunnelling spectra were displayed on **c** and **d**. Despite the coarser sampling, the observed phenomena for P_0 and P_1 are generally consistent with a hysteresis behaviour, where the 0 T spectrum aligns with the -0.2 T ($+0.2$ T) spectrum in the forward (backward) sweep.

We observed that the conductance of P_0 and P_1 at positive magnetic field ($+\mathbf{B}$) are lower than those at negative magnetic field ($-\mathbf{B}$), a behaviour opposite to that of the P_{-1} state. Given that the tip magnetization is oriented along the $+z$ direction (Fig. S10), we infer that the P_0 and P_1 states exhibit a minority spin-down component, whereas the P_{-1} state is characterized by a majority spin-up component¹⁶. Stabilization parameters in **a-d** are $V_{\text{bias}} = 0.4$ V, $I_t = 200$ pA, and $V_{\text{mod}} = 4$ mV.

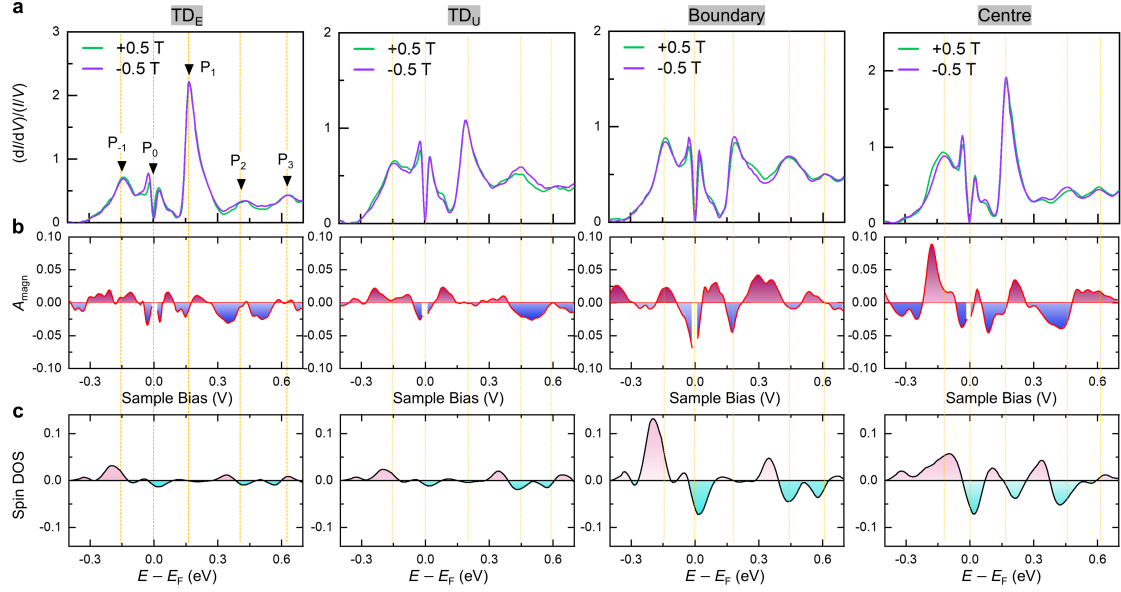


Fig. S12. Spatially resolved dI/dV and A_{magn} spectrum with spin-polarized tip. In Fig. 3, we show the averaged dI/dV and A_{magn} spectrum; here we display the data on the four typical sites. **a**, The normalized spin-polarized tunnelling spectra measured on four representative sites (TD_E , TD_U , boundary, and centre) at magnetic fields of ± 0.5 T. **b**, Magnetic asymmetry (A_{magn}) calculated from the experimental curves in **a** and the theoretical spin density of states (**c**) projected on each site. The $P_{-1} - P_3$ are marked by the orange-dash lines. We notice that the calculated net-spin DOS (**c**) shows a general agreement with the experimental A_{magn} (**b**). The stabilization parameters in **a** are $V_{\text{bias}} = 0.8$ V, $I_t = 200$ pA, and $V_{\text{mod}} = 8$ mV.

The magnitude of A_{magn} (**b**) illustrates spatial differences among the four sites, being particularly notable on the boundary and centre sites, which aligns well with the spin distribution shown in Fig. 3f. Despite these variations in magnitude, the spin polarizations extend across the entire primitive cell, displaying delocalized behaviour. This marks an important distinction between our system and other monolayers, such as the 1T-TaSe₂ family with 'Star-of-David' CDW¹⁷, which host correlated magnetic insulating states. These CDW phases are typically governed by Mott-Hubbard physics and exhibit a local moment picture where magnetic moments primarily reside in the SoD centre, with a smaller localization length than the inter-moment distance. Our recent STM work confirmed this localization in monolayer 1T-NbSe₂¹⁸.

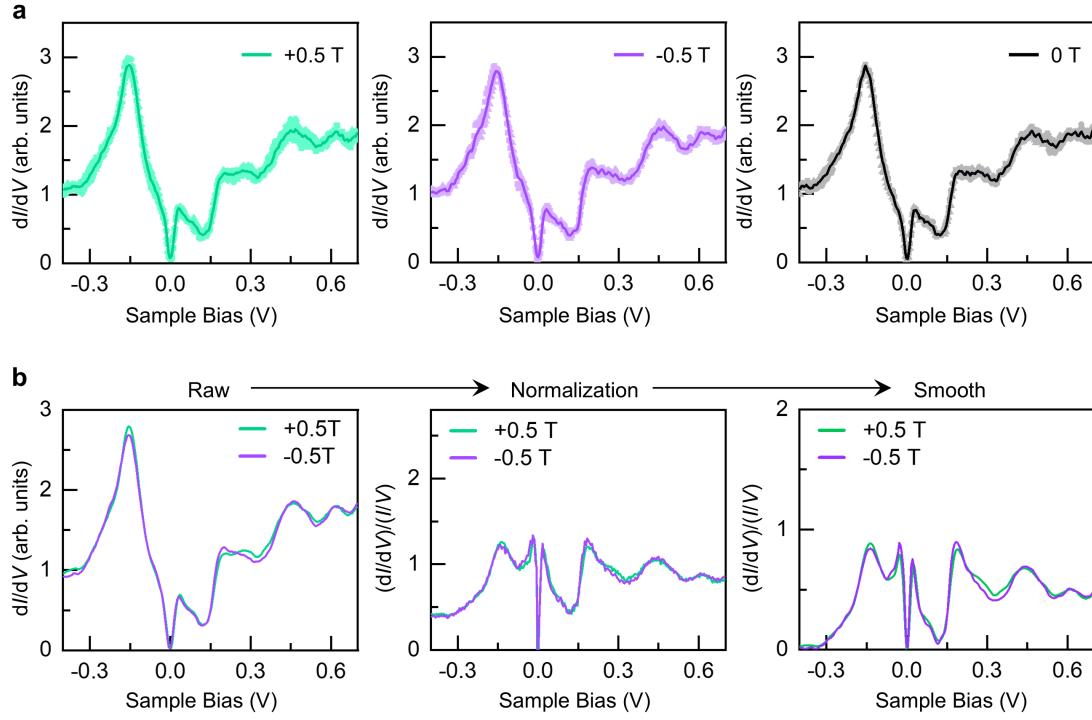


Fig. S13. a, Raw data at three magnetic fields (+0.5 T, -0.5 T, 0 T), including individual sweeps (shaded areas) and the averaged spectra (solid lines). **b**, Left panel is the average spectra from (a), normalized by total conductance (I/V) and smoothed using a 15-point Savitzky–Golay filter with a smoothing window of 10 mV. The boundary site serves as an example here. The right panel in (b) corresponds to the boundary panel in Fig. S12a.

The spectra presented in the main text have undergone smoothing and averaging processes. Below, we provide a detailed explanation of the data processing procedures.

1) Multiple sweeps and averaging: In our experiments, each raw spectrum is obtained by averaging multiple sweeps performed under identical parameters, typically with more than ten sweeps. In Fig. 13a, we display all the individual sweeps and the corresponding averaged spectra at three different magnetic fields (+0.5 T, -0.5 T, 0 T). This averaging process enhances the signal-to-noise ratio and leads to the smooth appearance of the raw data.

2) Normalization: The raw dI/dV spectra are normalized by the total conductance (I/V) to eliminate the influence of bias-dependent tunneling probability on the spectral weight (Fig. 13b)^{19,20}. The total conductance (I/V) is also averaged in the same manner as described above. This approach is commonly employed in STS data processing to achieve better consistency with the theoretical DOS (E).

395 3) Smoothing: To further improve data quality, the normalized spectra were
396 smoothed using a 15-point Savitzky–Golay filter with a smoothing window of 10 mV.
397 This process preserves the main features of the spectra while reducing noise.

398 4) Spatial averaging: The spectrum in Fig. 3b is the average of the four spectra from
399 Fig. S12a, obtained from all four typical sites and processed according to the procedures
400 described above. A straightforward numerical averaging was employed here, without
401 considering the weighting of different spatial locations.

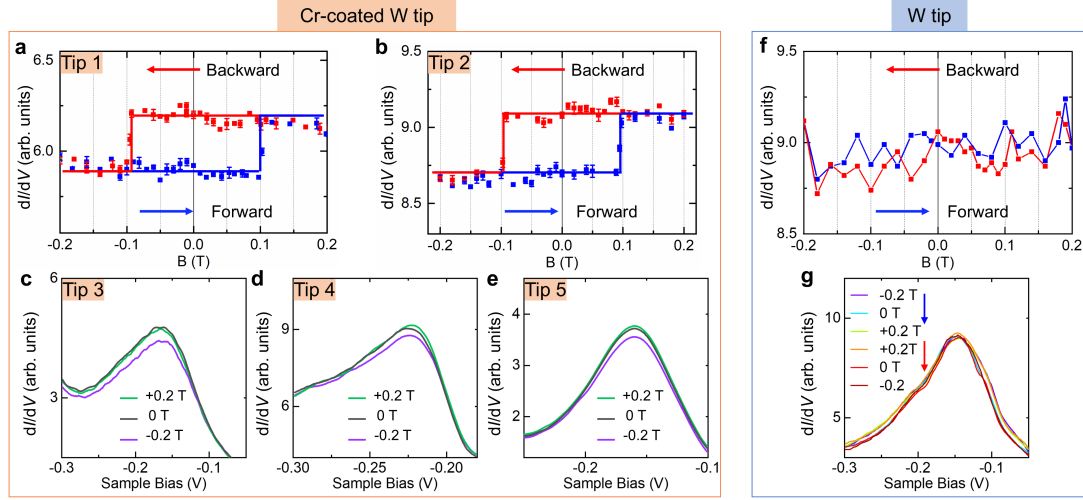


Fig. S14. Consistent magnetic measurements using multiple tips. **a-e**, SP-STM data taken by five additional Cr-coated W tips, all exhibiting consistently reproducible ferromagnetic hysteresis behaviours, in agreement with the result presented in the main text. **f, g**, A nonmagnetic W tip was used for a control measurement. As thoroughly discussed in the previous report²¹, minor shifts were applied in **a** and **b** to correct the residual stray field of the tips, which are 0.02 T (**a**) and 0.05 T (**b**) (0.04 T for Fig. 3e). The stabilization parameters in **c-e, g** are $V_{\text{bias}} = 0.4$ V, $I_t = 200$ pA, and $V_{\text{mod}} = 6$ mV.

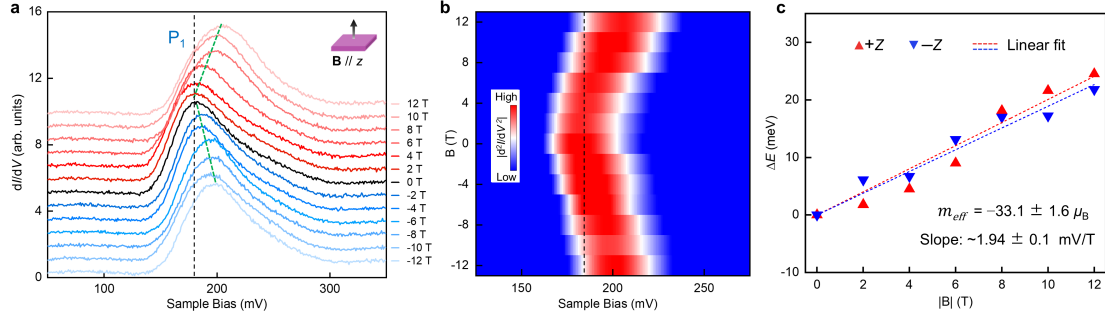


Fig. S15. Negative orbital magnetism of the P_1 state. **a**, dI/dV spectra of the P_1 state in a magnetic field perpendicular to the sample surface, ranging from -12 T to 12 T, taken with a non-magnetic tip. Green dashed lines depict the shift of the peak. **b**, Intensity plot of the second derivative of the field-dependent tunnelling spectra (*i.e.*, $|d^2I/dV^2|$). The black dashed lines in **a** and **b** indicate the energy locations of the peaks at $\mathbf{B} = 0$ T. **c**, Energy shifts of P_1 ($\Delta E = E(\mathbf{B}) - E(0\text{T})$) as functions of the external field. The stabilization parameters in **a** are $V_{\text{bias}} = 0.35$ V, $I_t = 200$ pA, and $V_{\text{mod}} = 3$ mV.

We further investigated the magnetism nature of our system by measuring the magnetic field response using a normal W tip. Our experimental results reveal a magnetization-polarized Zeeman effect of P_1 , where its energy level linearly shifts towards higher energies regardless of the magnetic field direction (Figs. S15a and b). Furthermore, the values of the effective magnetic moment (m_{eff}) of P_1 can be extracted from the linear fitting of the Zeeman shift $\Delta E = -\mathbf{m}_{\text{eff}} \cdot \mathbf{B}$. With linear fitting, we determined m_{eff} to be $-33.1 \pm 1.6 \mu_B$ (Fig. S15c).

This magnetization-polarized Zeeman shift with negative moments is consistent with observations in 3D kagome ferromagnets such as $\text{Co}_3\text{Sn}_2\text{S}_2$, TbV_6Sn_6 ²²⁻²⁴. This phenomenon occurs because the state's magnetic moments are always aligned with the applied magnetic field, making the energy shift independent of the field direction. The non-trivial Berry phase in the flat band contributes to this negative orbital magnetic moment through spin-orbit coupling in the kagome lattice²²⁻²⁴. The notably large m_{eff} in our system may result from the enlarged lattice size compared with the atomic kagome lattice in former systems.

Supplementary Note 3 Thermal broadening simulation.

To demonstrate that the temperature dependence of the gap is not just an effect of thermal broadening, we have performed a thermal broadening simulation

$$\frac{dI}{dV}(eV) \propto \int_{-\infty}^{\infty} \rho(E) \left(\frac{d}{dV} F(E - eV, T) \right) dE$$

where $\rho(E)$ is the zero-temperature density of states and $F(E, T)$ is the Fermi distribution function. In our simulation, As the thermal broadening at 0.35 K is negligible, we took the spectrum taken at 0.35 K as $\rho(E)$. Then, the red curve in Fig. 4c was generated by using the $F(E, T)$ at $T = 20$ K. Moreover, the simulated thermal broadening of the gap Δ_N and the experimentally measured values as a function of temperature are shown in Fig. 4d. Here, the gap Δ_N is defined by the midpoints of the edge slopes (Fig. S18a), *i.e.*, the maximum point in the $|d^2I/dV^2|$ spectra²⁵.

Supplementary Note 4 Excluding the Possibility of a Coulomb Gap

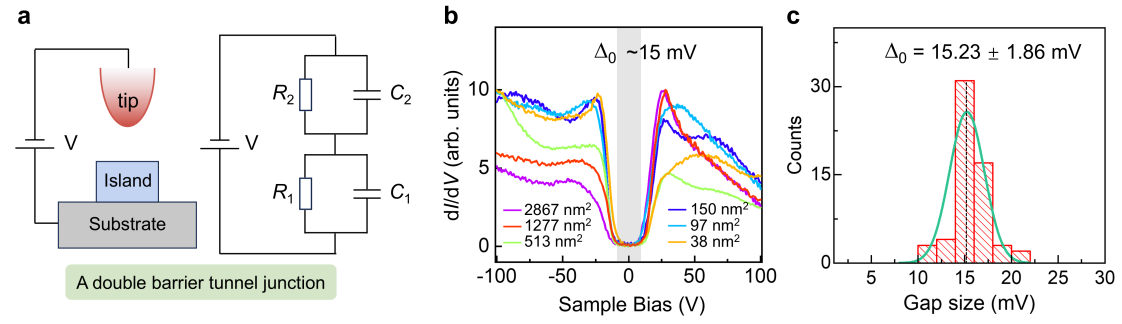


Fig. S16. **a**, Left panel: a schematic diagram of the tunnelling setup; right panel: the equivalent model of a double barrier tunnel junction (DBTJ) circuit. The two barriers correspond to the tip-island junction and the island-substrate junction, each consisting of a capacitor (C_2/C_1) and a resistor (R_2/R_1) connected in parallel. These parameters are relevant to the island size. The capacitance (resistance) increases (decreases) as the size of the island increases. The dynamic Coulomb blockade (DCB) and Coulomb blockade (CB) differ in the parameter regime of R & C , which is beyond the scope of the present discussion. **b**, dI/dV spectra measured on islands with areas ranging from 38 nm² to 2867 nm². All measurements exhibit a hard gap with nearly identical gap size (Δ_0) regardless of the island size. Stabilization parameters in **b**, $V_{\text{bias}} = 100$ mV, $I_t = 200$ pA, $V_{\text{mod}} = 2$ mV. **c**, Statistical analysis of the gap sizes. The green line is the fitting distribution curve which indicates an average value ($\Delta_0 = 15.23$ mV). Measurements were randomly taken from various islands over the sample surface. While local structural variations (such as defects) may contribute to some deviations, the Gaussian fitting of the gap histogram yields a statistical result of $\Delta_0 = 15.23 \pm 1.86$ mV.

Here, we elaborate on the exclusion of the Coulomb gap in our observation on the basis of the following two critical characteristics:

(1) The Coulomb gap was first named by Efros and Shklovskii (*i.e.*, the E - S model)²⁶. They investigated the Coulomb interactions between localized electrons on disordered systems (*e.g.*, amorphous or doped crystalline semiconductors), in which the electronic states are localized close to the Fermi level. The Coulomb gap can be understood as a result of the excitonic attraction of electrons and holes near the Fermi level, which

depletes the one-particle DOS there²⁷. The DOS in the two-dimensional case can be qualitatively given as:

$$N_{ES}(E) \propto \left(\frac{2}{\pi e^4}\right) |E|$$

Therefore, the gap in this model is expected to be linear in energy (namely, a “V”-shaped gap). This “V-shaped” (so-called a “soft gap”) feature has been widely used to identify the Coulomb gap in tunnelling microscopic studies of 2D systems, such as observations of Be films²⁸ and monolayer 1T'-WTe₂^{26,29}. Our measurements exhibit a “U-shaped” line shape (namely, a “hard gap”), which discriminates the present insulating states from the picture depicted by the E - S model.

(2) In STS studies of thin films, there is another case in which the gap opening near the Fermi level was sometimes named the Coulomb gap as well. This effect is often referred to as dynamic Coulomb blockade (DCB) or Coulomb blockade (CB).

This effect arises from single electrons exchanging energy with the environment as they tunnel through the barriers³⁰. Generally, the STS setup can be modelled with a circuit consisting of a double barrier tunnel junction (DBTJ)³¹, as sketched in Fig. S16a. When a single electron transfers through the DBTJ, it must overcome the Coulomb charging energy $E_C = e^2/C_\Sigma$. C_Σ is the total capacitance of the system, which is proportional to the island area³². Thus, in such a process, the suppression of the differential conductance near E_F is closely dependent on the island area; the smaller the island is, the larger the gap becomes.

In practical experimental research, the island-size dependence of the gap size has been acknowledged as a ‘smoking gun’ to identify this type of “CB gap” in various 2D systems (*e.g.*, Pb islands^{30,33}, ML-NbSe₂ on HOPG³⁴, ML-Bi₂Te₃ on Bi³⁵). Former studies illustrate the significant enhancement of the gap as the island size shrinks.

In Fig. S16b, we measured the gap widths on islands of different sizes. We found that although the delicate line shapes show slight differences from sample to sample, the hard gap was preserved even at the maximum area (2867 nm², which is comparable to the former studies^{30,34}), and the gap size ($\Delta_0 \sim 15$ mV) shows little variation along

with the change in island areas.

Thus, we have sorted out the key characteristics used in STM/S identifications of the 2D Coulomb gap. The spectral shape and island-size dependence allow us to rule out the possibility of the Coulomb gap in our case. A recent paper (ref. 36) excluded the Coulomb gap in the STS of a 1T/1H-TaS₂ vdW heterostructure following the same methodology.

In addition to the aforementioned scenarios, a U-shaped gap has been observed in the Landau levels of graphene systems, attributed to Coulomb interactions³⁷⁻³⁹. According to this scenario, “the electrons are treated as classical point charges at fixed positions in space with no overlap of the electronic wave functions”³⁹. Consequently, a minimum energy ($E_c \sim e^2/\epsilon l_b$) is required for electron tunneling between Landau orbitals. Here, l_b is the magnetic length (*i.e.*, the localization length), which varies from 10 to 100 nm within the experimental **B** range. In contrast, the wavefunction overlap (*i.e.*, inter-site hopping) is a significant characteristic of kagome materials. This overlap results in the presence of flat and Dirac bands, suggesting an absence of significant localization on the length scale observed in graphene's Landau orbitals. Therefore, it is less likely that the observed gap in our system is due to this mechanism.

Nevertheless, our primary conclusion remains unchanged: the observed gap at the Fermi energy (E_F) is attributed to electron correlation effects, leading to a correlated insulating state rather than a band insulator.

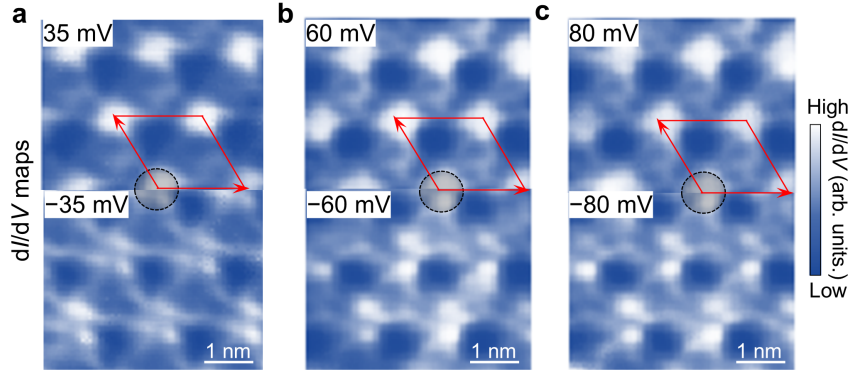
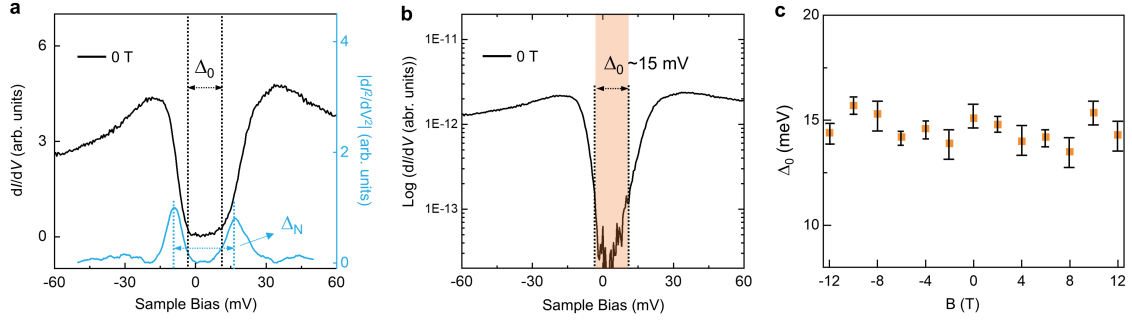


Fig. S17. Exclusion of the CDW patterns by bias-dependent STM imaging. **a-c** are STM differential conductance maps measured at the sample bias of ± 35 mV, ± 60 mV, and ± 80 mV. These images were taken near the edges of the observed gap. No appearance of charge modulations can be identified beyond the original periodicity of the atomic structure. Moreover, the contrasts in conductance images persist well when the sign of sample bias is reversed (*i.e.*, centre $> \text{TD}_U > \text{TD}_E$). Thus, the most typical characteristic of a CDW gap, that the charge intensity reversal between the electron and hole^{40,41}, is not seen in our system. The red rhombus labels a supercell. Scale bars are 1 nm. Stabilization parameters in **a-c**, $I_t = 100$ pA, lock-in modulation $V_{\text{mod}} = 1$ mV.



528

529 **Fig. S18. Determination of Δ_0 and Δ_N , and exclusion of superconductivity. a, b,**

530 Tunnelling spectra obtained with a W tip at 0 T, displayed on linear and logarithmic

531 scales, respectively. The gap size Δ_0 defines the energy range of the zero-conductance

532 region, as illustrated in **b**. The gap size Δ_N (blue dashed lines) is determined by the

533 midpoints of the edge slopes, *i.e.*, the maximum point in the $|d^2I/dV^2|$ spectra²⁵, as

534 illustrated in **a**. **c**, The gap size Δ_0 as a function of the external field B , indicating that

535 the gap remains unchanged by applying magnetic fields up to 12 T. Similar

536 measurements were performed on various locations to exclude the formations of

537 vortices. Therefore, we exclude the possibility that the observed gap is a

538 superconducting gap³⁶. The stabilization parameters in **a** are $V_{\text{bias}} = 0.1$ V, $I_t = 200$ pA,

539 and $V_{\text{mod}} = 2$ mV.

Table S1. The character table of Γ/K -little group in space group $P\bar{6}$ which is used to determine the irreducible representations (IRs) for the energy bands at Γ/K of the MTB superlattice in the IRVSP program⁴². This character table is obtained from Bilbao Crystallographic Server⁴³.

Symmetry operation	1	2	3	4	5	6
Seitz symbol	$\{1 t_1, t_2, t_3\}$	$\{3_{001}^+ 0,0,0\}$	$\{3_{001}^- 0,0,0\}$	$\{m_{001} 0,0,0\}$	$\{\bar{6}_{001}^- 0,0,0\}$	$\{\bar{6}_{001}^+ 0,0,0\}$
$\Gamma1/K1$	1.00+0.00i	1.00+0.00i	1.00+0.00i	1.00+0.00i	1.00+0.00i	1.00+0.00i
$\Gamma2/K2$	1.00+0.00i	1.00+0.00i	1.00+0.00i	-1.00+0.00i	-1.00+0.00i	-1.00+0.00i
$\Gamma3/K3$	1.00+0.00i	-0.50+0.87i	-0.50-0.87i	1.00+0.00i	-0.50+0.87i	-0.50-0.87i
$\Gamma4/K4$	1.00+0.00i	-0.50+0.87i	-0.50-0.87i	-1.00+0.00i	0.50-0.87i	0.50+0.87i
$\Gamma5/K5$	1.00+0.00i	-0.50-0.87i	-0.50+0.87i	1.00+0.00i	-0.50-0.87i	-0.50+0.87i
$\Gamma6/K6$	1.00+0.00i	-0.50-0.87i	-0.50+0.87i	-1.00+0.00i	0.50+0.87i	0.50-0.87i
$\Gamma7/K7$	1.00+0.00i	-1.00+0.00i	-1.00+0.00i	0.00-1.00i	0.00+1.00i	0.00-1.00i
$\Gamma8/K8$	1.00+0.00i	-1.00+0.00i	-1.00+0.00i	0.00+1.00i	0.00-1.00i	0.00+1.00i
$\Gamma9/K9$	1.00+0.00i	0.50-0.87i	0.50+0.87i	0.00-1.00i	-0.87-0.50i	-0.87+0.50i
$\Gamma10/K10$	1.00+0.00i	0.50-0.87i	0.50+0.87i	0.00+1.00i	0.87+0.50i	0.87-0.50i
$\Gamma11/K11$	1.00+0.00i	0.50+0.87i	0.50-0.87i	0.00-1.00i	0.87-0.50i	0.87+0.50i
$\Gamma12/K12$	1.00+0.00i	0.50+0.87i	0.50-0.87i	0.00+1.00i	-0.87+0.50i	-0.87-0.50i

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