

Supplementary Information for

Atomic-scale spin sensing of a 2D d-wave altermagnet

***via* helical tunneling**

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Supplementary Note 1: Schematic diagrams of collinear magnetic phases and spin-polarized STM

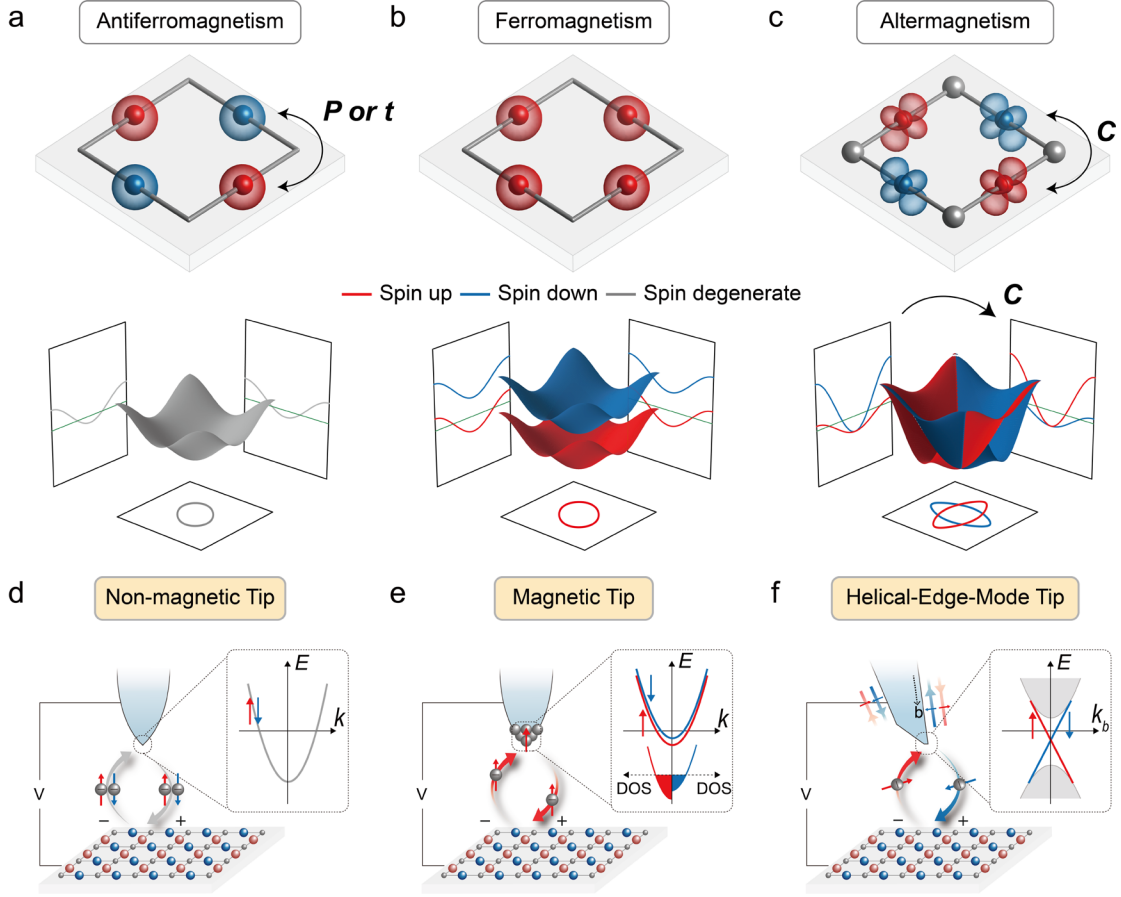


Figure S1 | Schematic diagrams of three collinear magnetic phases and spin-polarized STM. **a-c**, Real-space lattice and spin densities (top), k -space band structure (middle) and Fermi surface (bottom) in conventional AFM (**a**), FM (**b**), and d -wave altermagnetism (**c**). P , t , and C refer to inversion, translation, and crystal (mirror or rotation) symmetry, respectively. Red, blue, and grey colors denote spin-up, spin-down, and spin-degenerate states, respectively. **d-f**, Schematics of STM tunneling with a spin-degenerate metallic tip (**d**), a magnetic tip coated with ferromagnetic clusters (**e**) and a helical-edge-mode tip (**f**). The latter two enable spin-selective tunnelling through different mechanisms. For a ferromagnetic tip, the magnetization leads to spin-polarized electronic states of the tip, and the tunneling current depends on the relative alignment of the local magnetizations of the tip and sample that are usually controlled by external magnetic fields (**e**). For a helical-edge-mode tip, spin-momentum locking relates counter-propagating edge states with opposite spin expectation values. Selective coupling to these states can produce tunnelling-direction-dependent spin polarization, controlled by reversing the sample-bias polarity (**f**).

Supplementary Note 2: Topological classification and helical edge modes of α -Bi₄Br₄

One-dimensional helical edge modes have been proposed in two-dimensional (2D) quantum spin Hall insulators (QSHIs), weak topological insulators and higher-order topological insulators (HOTIs), but the number of materials realizing these topologies is still quite limited [S1-S14]. Because these edge modes are 1D objects that are strongly confined to the boundaries, experimental studies have largely focused on STM and ballistic transport signatures of these modes. In [Supplementary table 1](#), we summarize the STM results on the helical edge modes.

An alternative route to achieve helical edge channels in a 3D crystal, i.e., *via* a 3D QSHI, has been recently proposed and verified [S14, S15]. This spin-resolved topology is characterized by nonzero spin Chern numbers on all constant- k_z planes and is not fully captured by conventional symmetry indicators. As a result, both α -Bi₄Br₄ with inversion $\mathbb{Z}_4 = 2$ and α -Bi₄I₄ with trivial symmetry indicator could be classified as 3D QSHIs [S14, S15]. The key feature here is the weak interlayer coupling due to the spatial confinement of the edge modes and the large thickness of a Bi₄X₄ layer. When the interlayer coupling is weak and S_z is approximately conserved, the spin S_z Hall conductivity can be nearly quantized of $3.62 \times e/4\pi$ per bulk unit cell [S14]. The robustness of helical edge channels at multilayer boundaries could serve as the hallmark of a 3D QSHI ([Extended Data Figure 1](#)), though some edge states might be slightly gapped if not protected by symmetry. In [Supplementary table 2](#), we show the topological indices for strong TI, weak TI, Helical HOTI and 3D QSHI. In [Supplementary Fig. S2](#), we show the potential corresponding locations of the *gapless* and slightly gapped helical edge modes in multilayer Bi₄Br₄ following this discussion. In principle, both two types of edge modes can contribute to the quantum tunneling of electrons in our experiments.

	Type	Edge state	Decay length / Spatial extent
WTe₂	Single layer step ^[1]	√	2.5 nm
	Monolayer film ^[2]	√	1.2 nm
	Bilayer film ^[3]	×	-
WSe₂	Monolayer film ^[4]	√	2 nm
ZrTe₅	Single layer step ^[5,6]	√	1.5nm
Bi*	Monolayer film ^[7]	√	2 nm
	Single layer step ^[8]	√	-
Bi₁₄Rh₃I₉	Single layer step ^[9]	√	0.83 nm (FWHM)
Bismuthene on SiC	Monolayer film ^[10]	√	0.4 nm
Stanene on Bi	Monolayer film ^[11]	√	Less than 4nm (edge A) 5 nm (edge B)
Bi₄Br₄	Single layer step ^[12]	√	2.1 nm
	Bilayer step ^[12]	√ (Selectively)	-
	Single layer step (this work)	√	2.49 nm
	Bilayer step (this work)	√	
Bi₄I₄	Monolayer film ^[13]	√	1.5 nm
	Single layer step ^[14]	√	1.48nm
	Bilayer step ^[14]	√	

Table S1 | 1D helical edge modes in various topological systems. (* for Bi, one QSHI layer contains two bismuth atomic layers)

	STI	WTI	Helical HOTI	3D QSHI	α -Bi ₄ I ₄	α -Bi ₄ Br ₄
Z_2	1	0	0	/	0	0
$Z_{2w,w=1,2,3}$	/	$\neq 0$	0	/	0	0
Z_4	/	/	2	/	0	2
C_s^+ (for all k_z planes)	/	/	/	$\neq 0$	2	2

Table S2 Topological indices for strong TI (STI), weak TI (WTI), Helical HOTI, 3D QSHI and their application to α -Bi₄I₄ and α -Bi₄Br₄. $Z_2 \equiv 1/2 \sum_{K \in TRIMs} n_K^-$ is the strong topological index, $Z_{2w,w=1,2,3} \equiv 1/2 \sum_{K_l=\pi} n_K^-$ are three weak topological indices, $Z_4 \equiv 1/4 \sum_{K \in TRIMs} (n_K^+ - n_K^-)$ is the symmetry-based indicator in centrosymmetric space group, and C_s^+ is the spin Chern number of the positive spin sector.

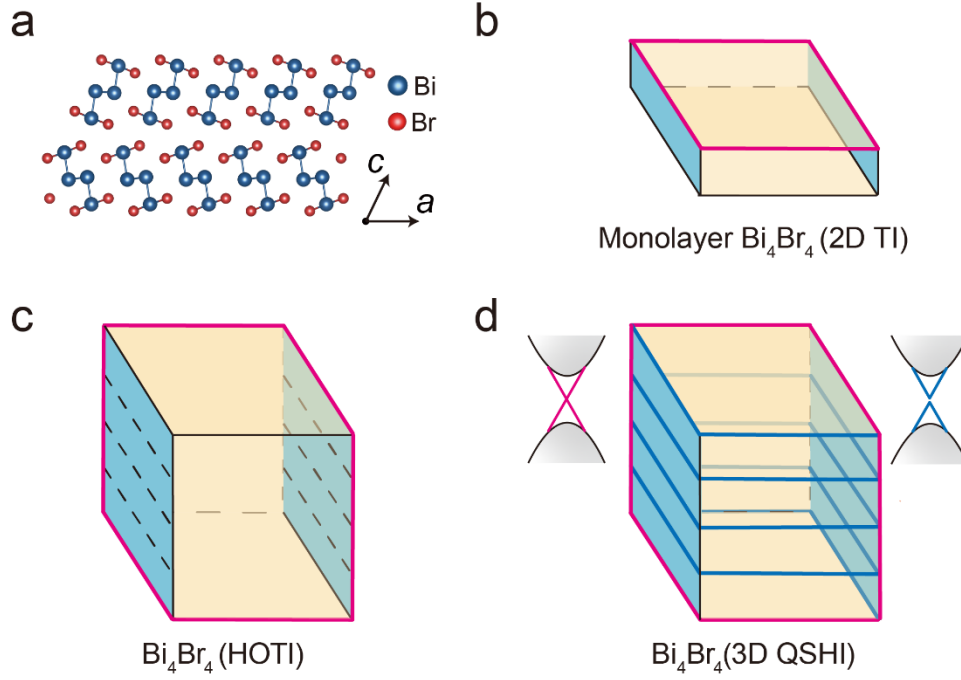


Figure S2 | Helical edge states in Bi_4Br_4 . **a**, Crystal structure of $\alpha\text{-Bi}_4\text{Br}_4$. **b-d**, Schematic of 1D helical states in monolayer Bi_4Br_4 (**b**, 2D TI), bulk $\alpha\text{-Bi}_4\text{Br}_4$ as a HOTI (**c**) and bulk $\alpha\text{-Bi}_4\text{Br}_4$ as a 3D QSHI (**d**). The pink-colored edge modes denote *gapless* edge states enforced by symmetry indicators, and the blue ones correspond to *slightly gapped* edge states in the case of a 3D QSHI.

Supplementary Note 3: Nanofabrication process and magnetic-field-dependent spectra of Bi₄Br₄ tips

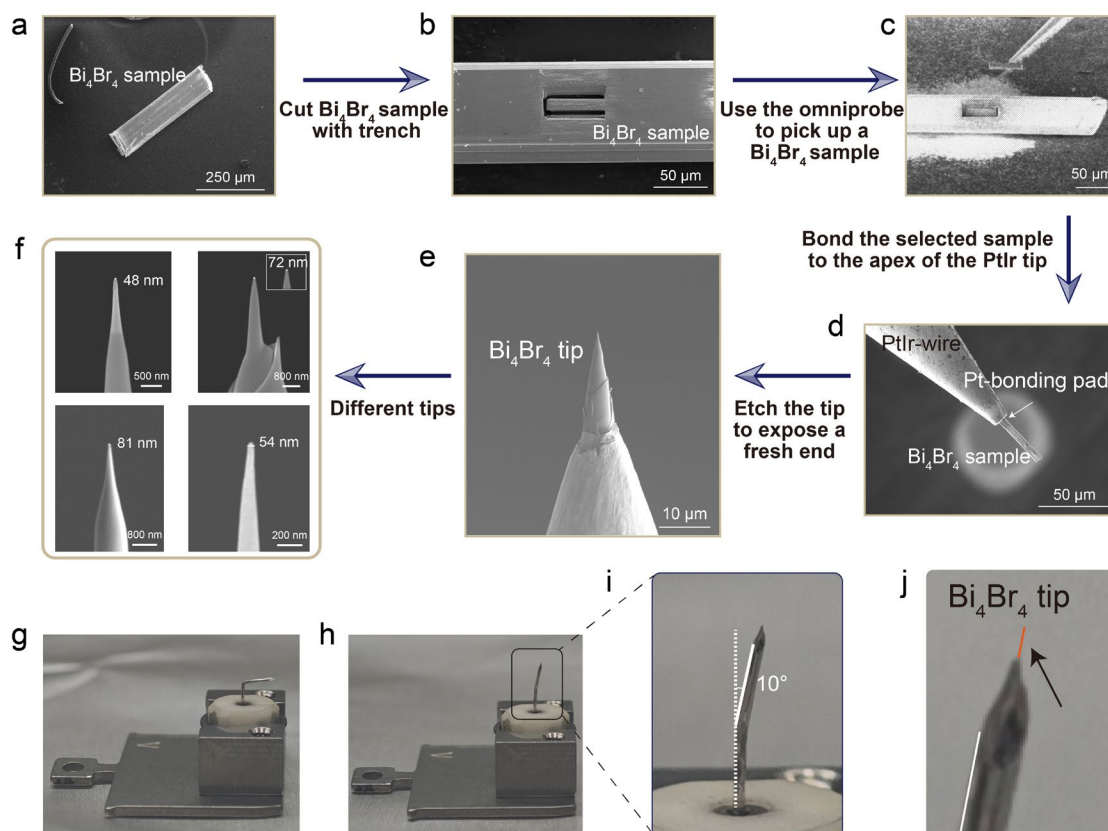


Figure S3 | Nanofabrication process and scanning electron microscopy (SEM) images of the Bi₄Br₄ tips. **a**, SEM image of a bulk Bi₄Br₄ typically several micrometres thick, used as the starting material. **b**, Formation of a slender Bi₄Br₄ strip by gallium-ion-beam milling of the bulk crystal. **c**, Detachment and pickup of the Bi₄Br₄ strip using the micromanipulator probe of the FIB system. **d**, Flattening the apex of a PtIr tip *via* ion-beam milling and then attaching the Bi₄Br₄ strip to this the flattened apex through Pt deposition. **e**, Sharpening the end of the Bi₄Br₄ strip with gentle ion-beam milling. **f**, SEM images of four Bi₄Br₄ tips used in this work for KV₂Se₂O. The apparent tip apex diameter is on the sub-100 nm scale. **g-j**, Schematics of tip-handling procedure, illustrating a typical residual tilt of approximately 10° relative to the sample surface normal. The Bi₄Br₄ strip is aligned approximately parallel to the distal segment of the PtIr support wire.

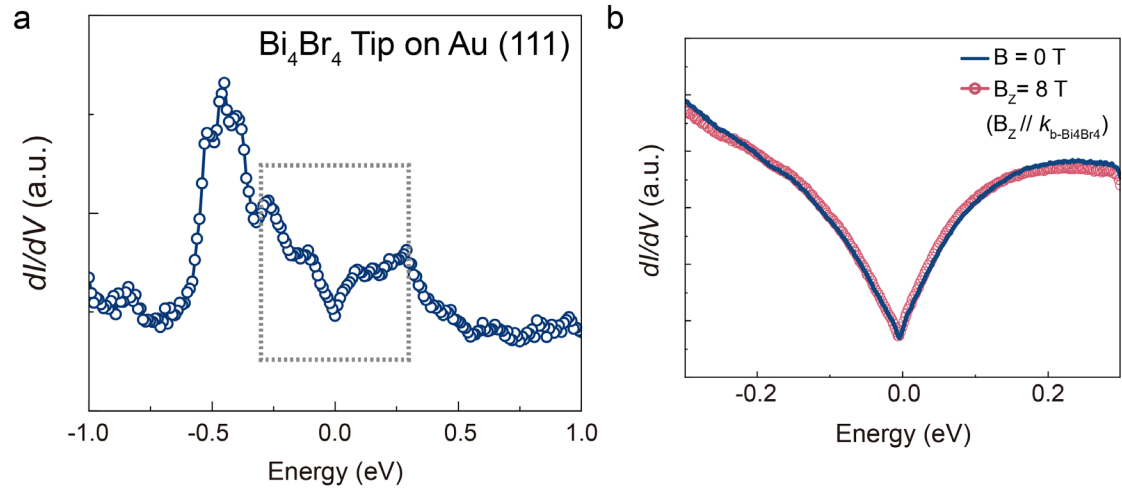


Figure S4 | Magnetic-field-dependent spectra of the Bi₄Br₄ tip. **a**, dI/dV spectrum acquired on Au (111) using a Bi₄Br₄ tip. The dotted lines mark the energy range enlarged in **(b)**. **b**, Comparison of the low-energy spectra measured with the same tip at 0 T and 8 T, with the magnetic field parallel to the crystallographic b axis of Bi₄Br₄. No discernible change was observed at 4.2 K due to the tiny pure Zeeman gap (field direction parallel to the momentum of edge channels) of 8 T.

Supplementary Note 4: Imaging the magnetic structure of Fe_{1.02}Te

Iron telluride (Fe_{1+x}Te) is a prototypical antiferromagnet with bicollinear magnetic order with a unidirectional period of $2a_{Te}$ [S16]. For spin-summed tunneling with PtIr tips, the topography solely exhibits atomic modulation of the Te lattice (Fig. S5a, b). Notably, an additional stripe modulation related to the bicollinear magnetic order can be immediately observed in topographies when Bi₄Br₄ tips or Fe-coated tips are used. Figure S5 c-f present the data obtained with two different Bi₄Br₄ tips. The spin-polarized local DOS undergoes a π -phase shift (contrast reversal) at opposite bias voltages, as observed in both the dI/dV maps and topographic images (Figure S6). When a Fe-coated PtIr tip was used, the signatures of the bicollinear AFM order could be still observed, however, the contrast reversal is clearly absent upon reversing the bias-voltage polarity (Figure S7).

We note that there are additional features primarily stemming from atomic-scale modulations (Bragg modulation) that manifest as local kinks in the dI/dV maps at negative bias (Figure S8c and d). By applying a q -space filter to suppress these Bragg modulations (with the cutoff indicated by the gray shaded region in Figure S8d), a nearly symmetric mirrored pattern is recovered, as shown in Figure S8e. The $2a$ AFM modulations in the dI/dV maps are visible at low energies but diminish significantly near 80 meV; conversely, atomic modulations become more pronounced at higher bias voltages (Figure S6c, f). It is important to note that the AFM stripe contrast persists in topographic images even at higher energies, as topography reflects the integrated DOS from E_F to the set bias. We have included a series of topographic images from +100 meV to -100 meV in Figure S6, which consistently exhibit contrast reversal upon switching the bias polarity. We also acknowledge that Fe_{1+x}Te single crystals exhibit intrinsic nanoscale inhomogeneity, likely due to interstitial Fe atoms. These local variations may contribute to the additional spectral features observed in both topographies and dI/dV maps.

We have further performed a sum-and-difference analysis of the entire dI/dV maps shown in Fig. 2f. To ensure a physically meaningful comparison between different energy scales, we first normalized each map by its spatial average: $\tilde{g}(r, eV) = [g(r, eV) - \langle g(eV) \rangle] / \langle g(eV) \rangle$, where $g(r, eV)$ is the dI/dV at position r and bias V , and $\langle g(eV) \rangle$ denotes the spatial average over the FOV. Using these normalized maps, we computed the difference map, $\tilde{g}(+50 \text{ mV}) - \tilde{g}(-50 \text{ mV})$ (Figure S9a), and the sum map, $\tilde{g}(+50 \text{ mV}) + \tilde{g}(-50 \text{ mV})$ (Figure S9b). The line-profiles extracted from these maps (Figure S9c and d) reveal that the difference map is dominated by a robust stripe pattern with a $2a$ periodicity, characteristic of the bicollinear AFM order. In contrast, the sum map primarily retains the atomic lattice information. This collective analysis suggests that the contrast reversal is an overall spectroscopic signature across the entire FOV rather than a localized artifact.

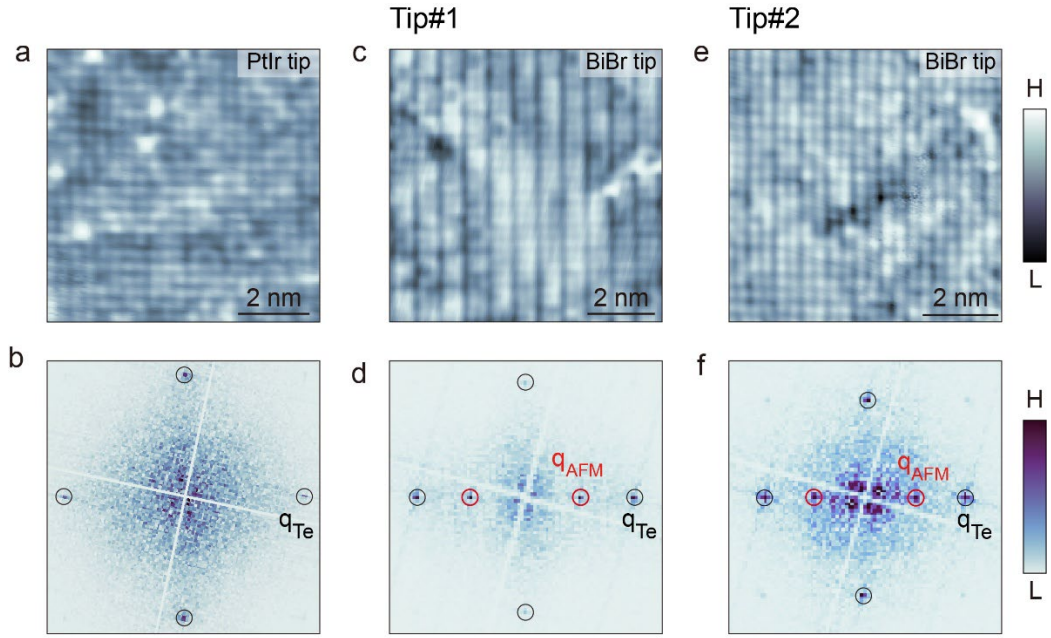


Figure S5 | Topographic images of $\text{Fe}_{1.02}\text{Te}$ acquired with PtIr and Bi_4Br_4 tips. **a**, Topographic image acquired with a PtIr tip. **b**, The corresponding FT of **a**. The Bragg peaks are marked by black circles. **c**, Topographic image obtained with a helical-edge-mode Bi_4Br_4 tip. **d**, The corresponding FFT of **c**. The Bragg peaks and AFM vectors are marked by black and red circles, respectively. **e-f**, STM topography and corresponding FT acquired with a second Bi_4Br_4 tip.

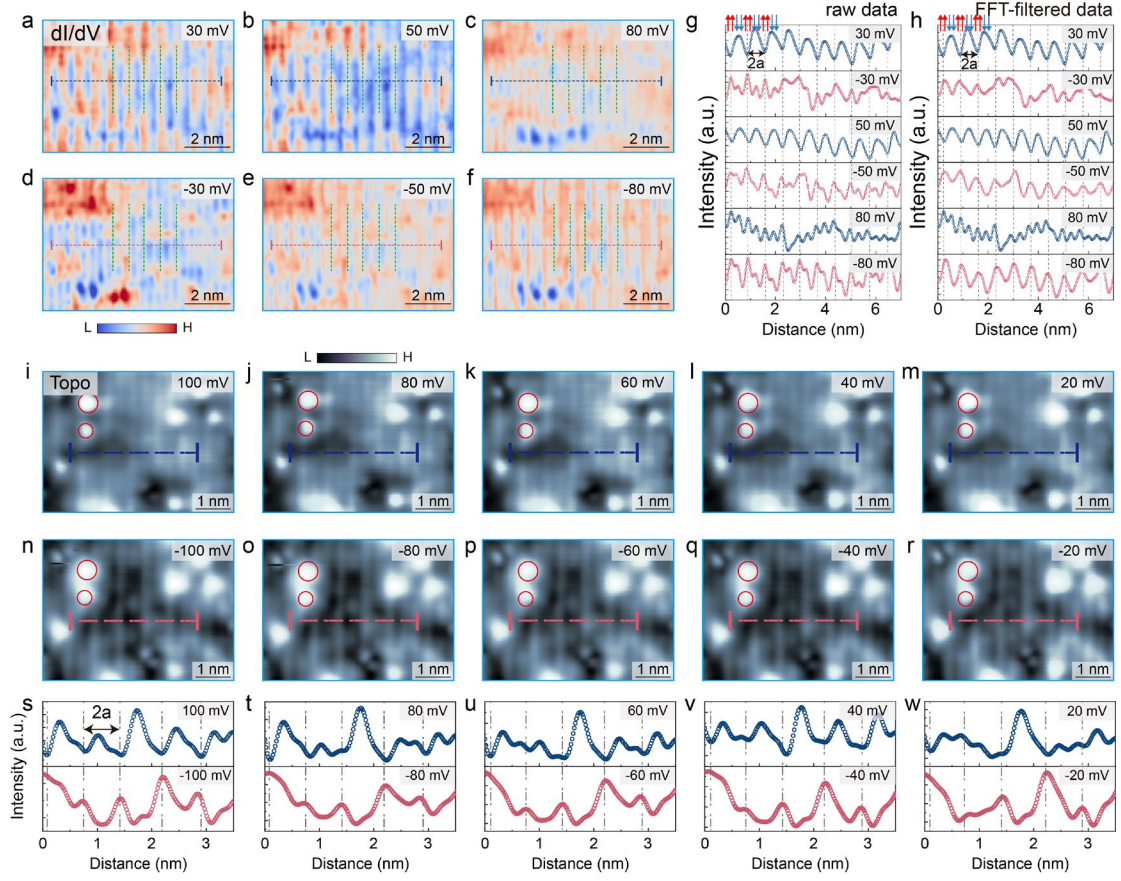


Figure S6 | Bicollinear AFM order in Fe_{1+x}Te imaged by the Bi_4Br_4 tips. a-f, dI/dV maps and i-r, topographic images. In both cases, the spin contrast of the $2a$ modulation exhibits a contrast reversal at opposite bias voltages, as shown in g-h and s-w. In these linecuts we apply a q -space filter to remove the Bragg contribution for the dI/dV maps at negative biases in h. A smooth cubic polynomial background was subtracted for topographic linecuts in s-w. To show the overall tendency, the linecuts in g and s-w are averaged over 30 and 20 pixels perpendicular to the linecut direction, respectively.

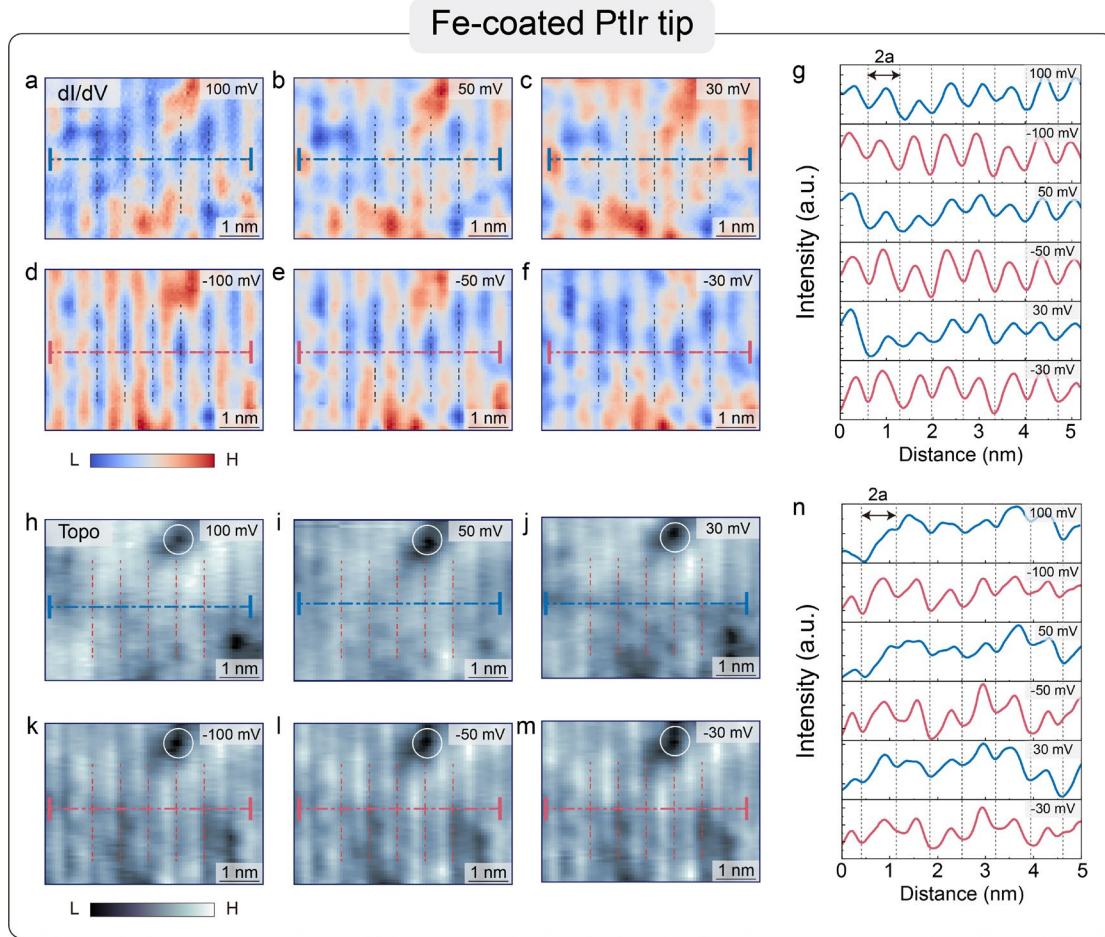


Figure S7 | Bicollinear AFM order in Fe_{1+x}Te imaged by the Fe-coated PtIr tips. **a-f**, dI/dV maps and **h-m**, topographic images. In both cases, the contrast reversal of the $2a$ modulation is absent at opposite bias voltages (**g** and **n**). To show the overall tendency, the linecuts in **g** and **n** are averaged over 30 pixels perpendicular to the linecut direction.

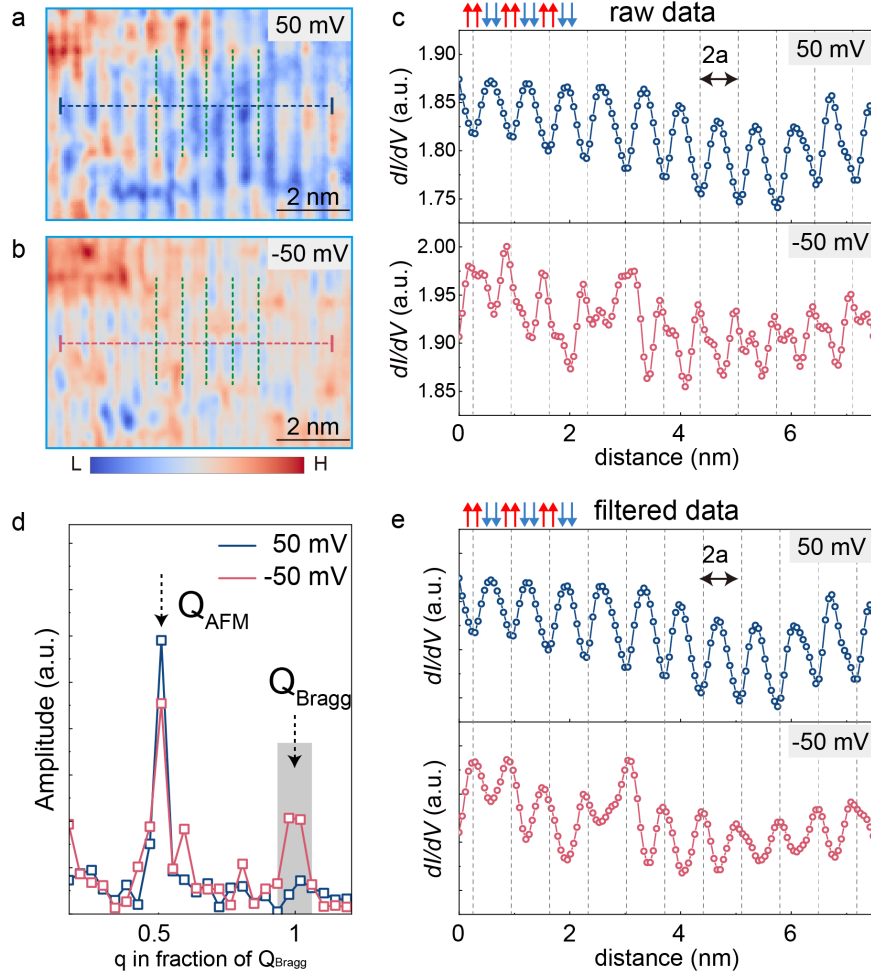


Figure S8 | Bragg modulation in the dI/dV maps at negative biases with Bi_4Br_4 tips. **a, b**, dI/dV maps obtained at 50 meV and -50 meV. **c**, Linecut profile of the dI/dV value. To show the overall tendency, the linecuts are averaged over 30 pixels perpendicular to the linecut direction. **d**, FFT for the linecut profile in **c**, where two peaks at AFM vector and Bragg vector can be observed at -50 meV. The gray shaded region denotes the q -space window for Fourier filter. **e**, A nearly mirrored pattern is recovered after the Fourier filter process.

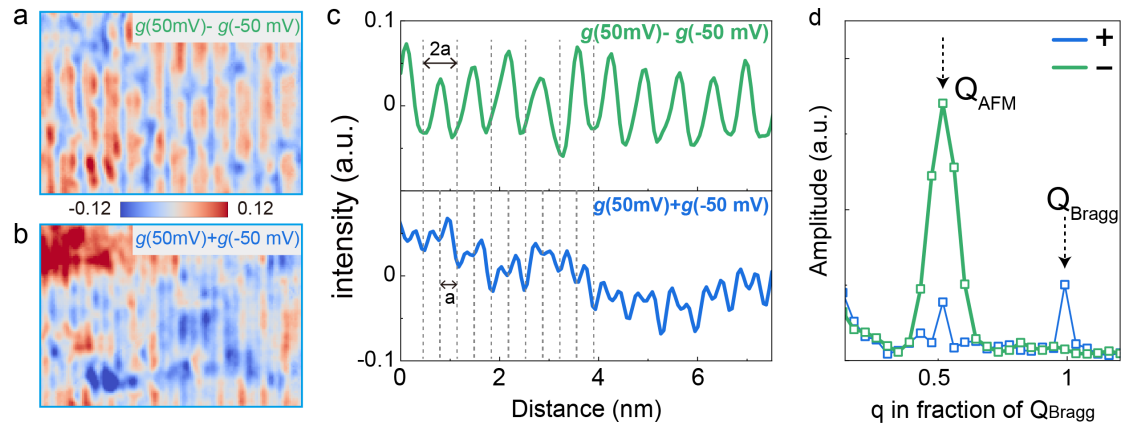


Figure S9 | Difference map and sum map of the dI/dV maps acquired with Bi_4Br_4 tips. **a, b,** The difference (**a**) and sum (**b**) maps of the dI/dV maps obtained at -50meV and 50meV. Each raw map was first normalized by its spatial averaged value. **c,** Line-profiles extracted from the difference and sum maps. To show the overall tendency, the linecuts are averaged over 30 pixels perpendicular to the linecut direction. **d,** FFT for the linecut profile shown in **c**. The peaks of the AFM vector are more pronounced in the difference map, whereas the sum map primarily retains the atomic lattice information.

Supplementary Note 5: Characterization of $\text{KV}_2\text{Se}_2\text{O}$ and STM data with PtIr tips

$\text{KV}_2\text{Se}_2\text{O}$ single crystals were obtained through a three-step synthesis (see [Methods](#)). The sharp XRD peaks suggest the high quality of the single crystals. For these crystals, the average K: V: Se atomic ratio is displayed in [Figure S10d](#). The concentration of V vacancies (defects) was determined to be 0.5% by counting the atomic defects in multiple FOVs. This information together with the EDS data, suggests that the K:V:Se atomic ratio is 0.975:1.990:2.023, which is very close to 1:2:2. The temperature-dependent in-plane resistivity $\rho(T)$ shows weak insulating behavior at low temperatures ([Figure S10b](#)), consistent with our STM measurements that revealed an energy gap at E_F ([Fig. 4d](#)).

In this Note, we also present the STM data obtained with PtIr tips. [Figures S11 and S12](#) exhibit some close-up views of the topographic images which allow us to establish the spatial correlations between these four types of V-site defects. In [Fig. S13](#), we display a line-cut of dI/dV spectra to show the spatial variations in local density of states near E_F . [Figures S14 and S15](#) show more QPI data with different energies acquired with PtIr tips.

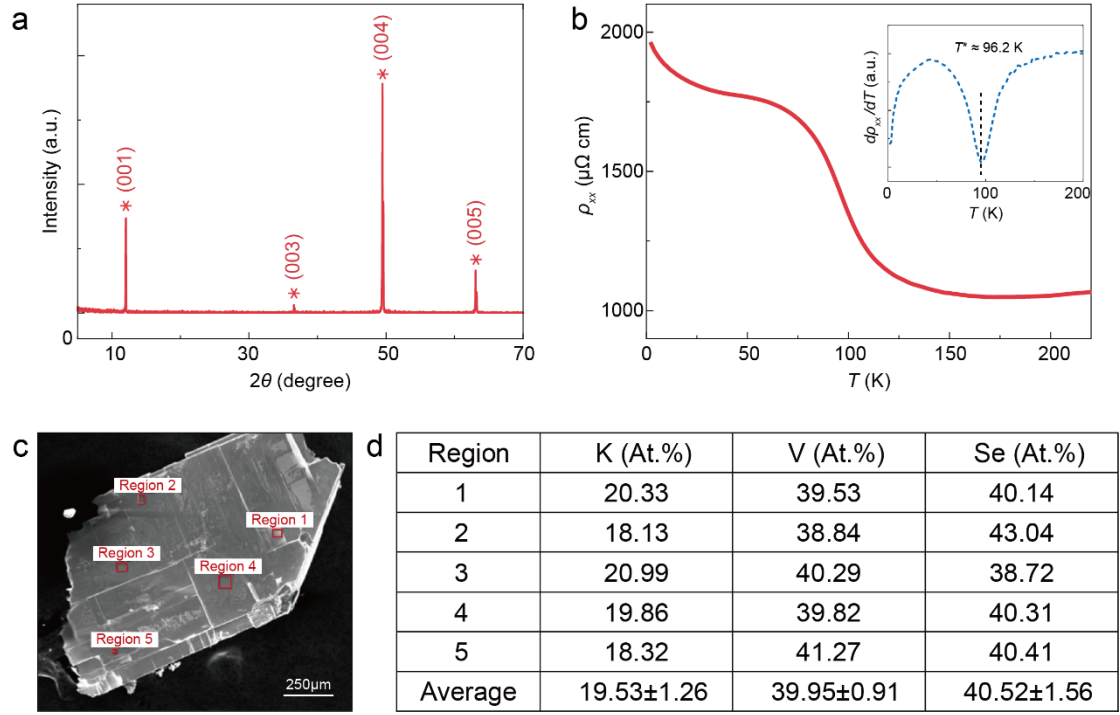


Figure S10 | Characterization of the $\text{KV}_2\text{Se}_2\text{O}$ single crystals. **a**, Single-crystal X-ray diffraction (XRD) pattern measured at room temperature, with Miller indices (001) marked by stars. **b**, Temperature-dependent in-plane resistivity $\rho_{xx}(T)$ from 2 to 220 K, showing weak insulating behavior at low temperatures. Inset: Temperature-derivative of $\rho_{xx}(T)$, highlighting an anomaly at $T^* \approx 96.2$ K due to the density-wave transition. **c**, SEM image of a cleaved $\text{KV}_2\text{Se}_2\text{O}$ sample. **d**, Atomic percentages (at %) of K, V, and Se measured by energy dispersive X-ray (EDX) spectroscopy in the corresponding regions shown in **c**. Combining this information with the concentration of the V vacancies (0.5%) determined via the counting in STM topographies, the average K : V : Se atomic ratio is 0.975 : 1.995 : 2.023, which is very close to 1:2:2.

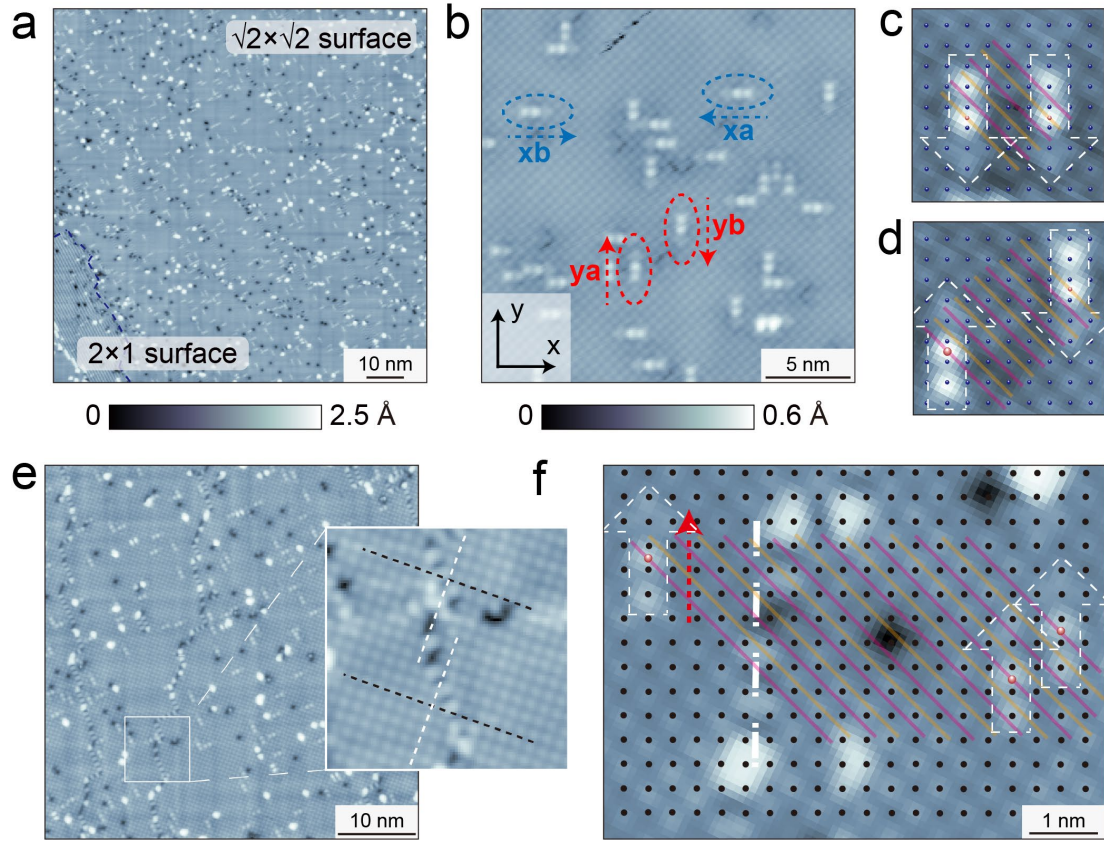


Figure S11 | Relationship between the V-site defects and the proposed $\sqrt{2} \times \sqrt{2}$ superstructure measured with a PtIr tip. **a**, Topographic image of an ordered surface on a 100-nm-square FOV, containing both 2×1 and $\sqrt{2} \times \sqrt{2}$ reconstructed K surfaces. **b**, Atomically resolved topography showing four different types of arrow-shaped defects. **c**, **d**, Spatial correlation among these defects. Two inequivalent sites (V_{y_a} and V_{y_b}) are alternately distributed along the diagonal direction and are marked by pink (V_{y_a}) and orange (V_{y_b}) lines, respectively. **e**, Topographic image of a $\sqrt{2} \times \sqrt{2}$ reconstructed surface. Inset: Close-up view of two domain walls across which the surface $\sqrt{2} \times \sqrt{2}$ K lattice shifts along both surface lattice directions, as marked by the dashed lines. **f**, Enlarged view of a region with V_{y_a} -type V-site defects near a surface-reconstruction domain wall. The spatial correlation of these defects according to the underlying superstructure remains consistent regardless of the surface-reconstruction domain.

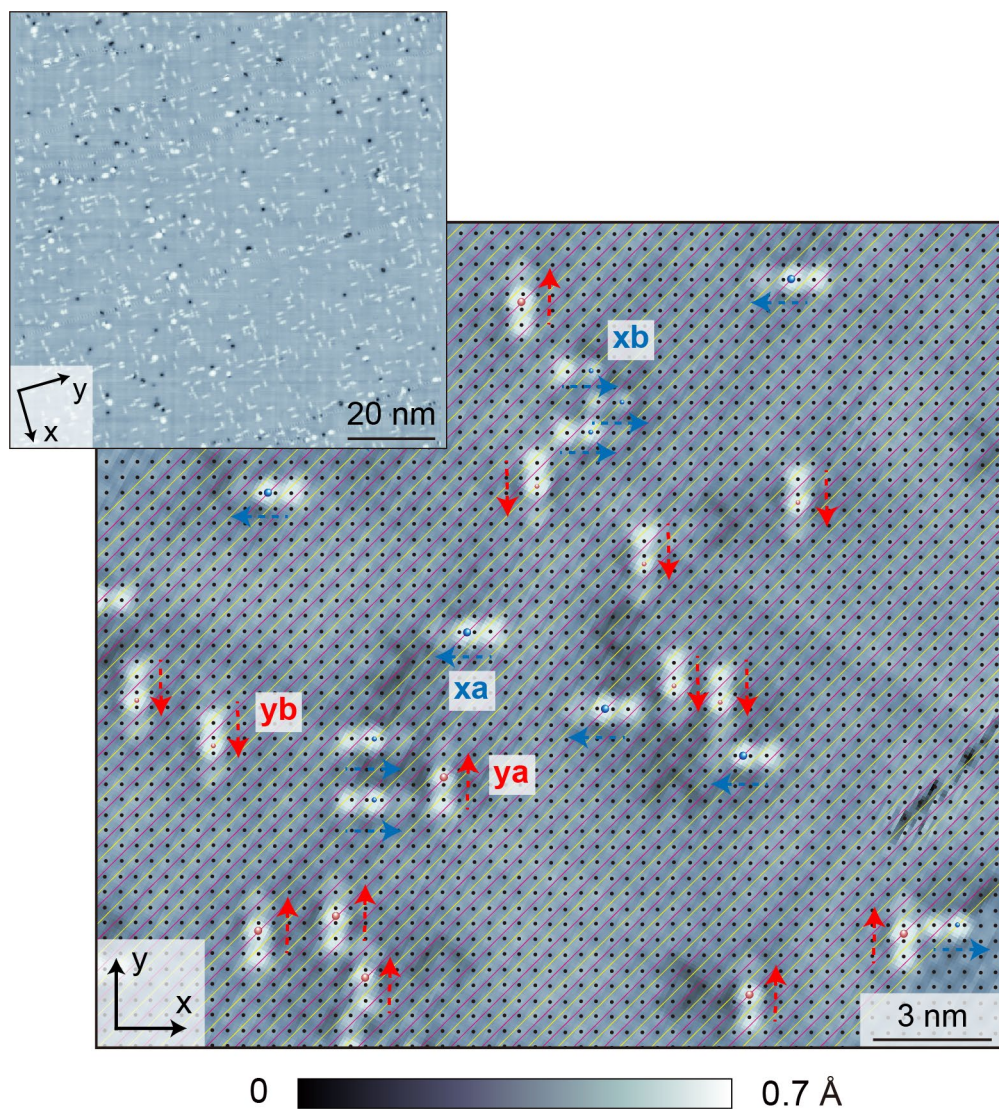


Figure S12 | Atomic-registered topography in a large FOV, showing the long-range spatial correlation among four types of arrow-shaped defects. Inequivalent sites ($V_{ya}(V_{xa})$ and $V_{yb}(V_{xb})$) are alternately distributed along the diagonal direction and are marked by pink ($V_{ya}(V_{xa})$) and orange ($V_{yb}(V_{xb})$) lines, respectively.

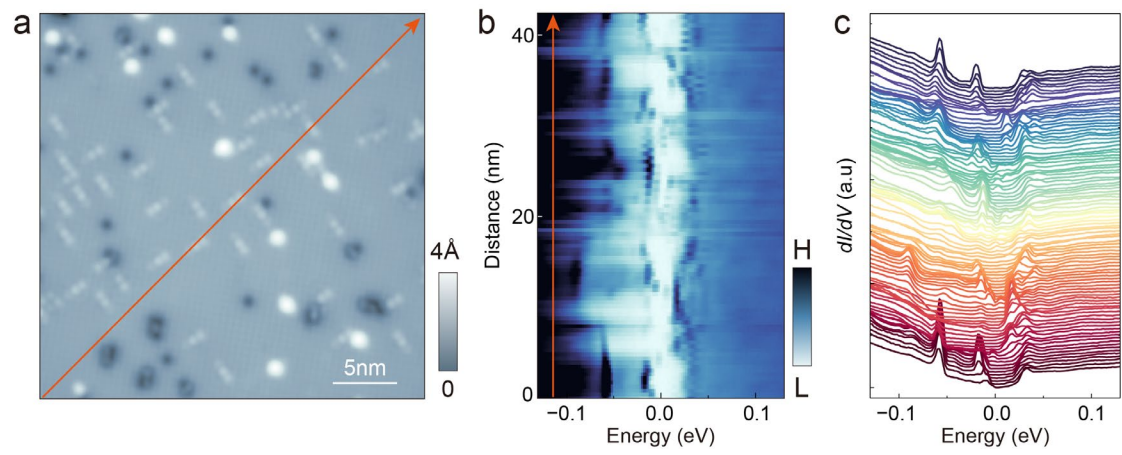


Figure S13 | Inhomogeneous electronic states in $\text{KV}_2\text{Se}_2\text{O}$. **a**, STM topography of a $\sqrt{2} \times \sqrt{2}$ reconstructed K surface. **b**, **c**, dI/dV linecuts along the red arrow in panel **a**.

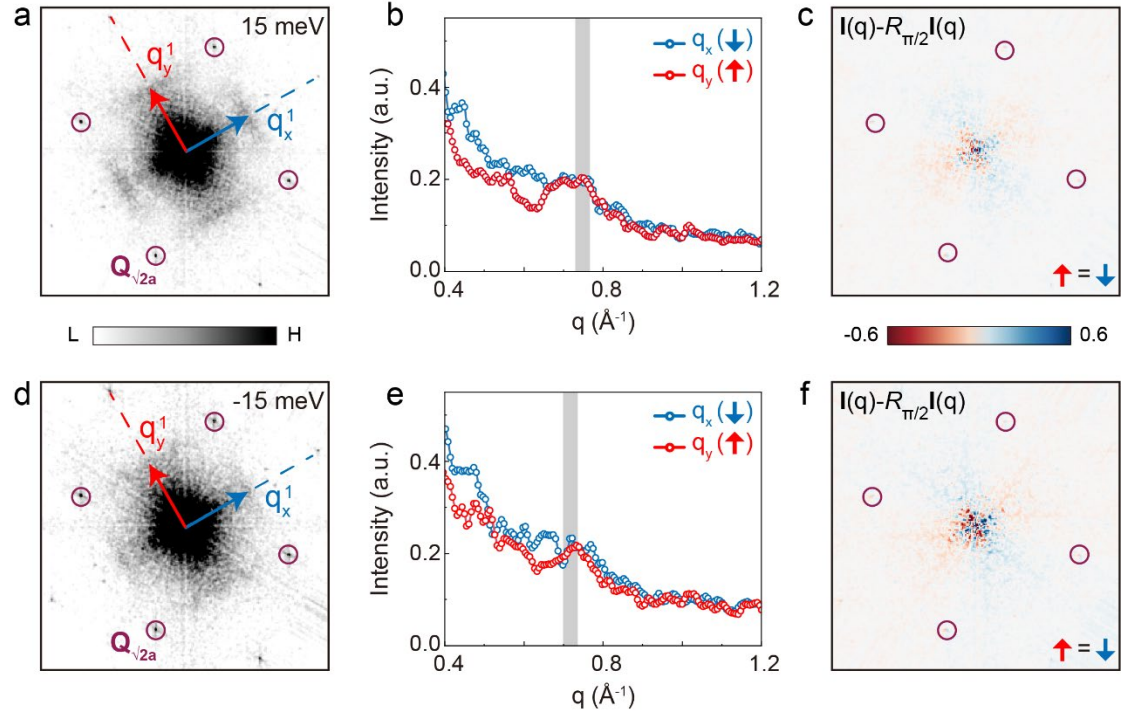


Figure S14 | Nearly isotropic QPI patterns measured with the non-spin-sensitive PtIr tips. **a**, QPI pattern at 15 mV measured with PtIr tip. **b**, FT linecuts along the dashed lines in **a**. **c**, The corresponding “contrast” map $M(\mathbf{q}, eV)$ derived from the QPI image shown in **a**, normalized by Bragg peak intensity. **d-f**, Similar analysis as in **a-c**, but data at -15 mV.

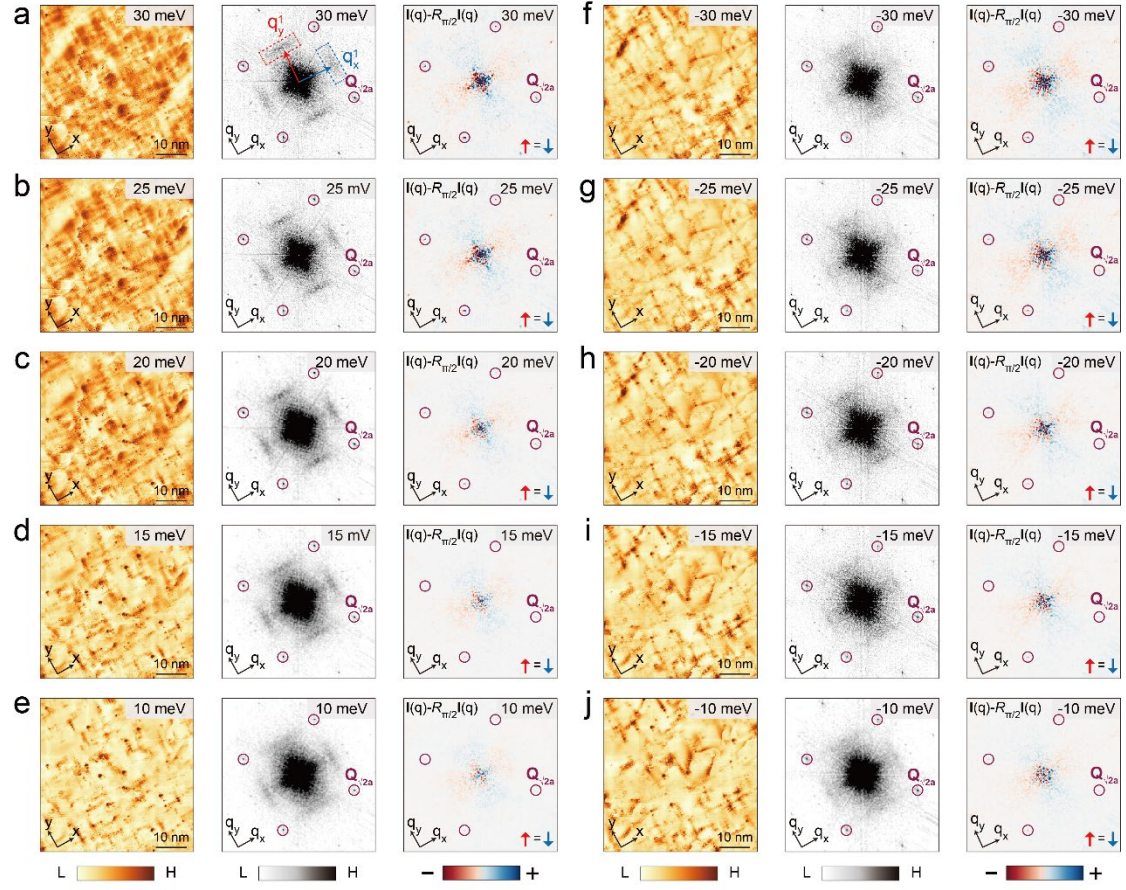


Figure S15 | More QPI images acquired with different PtIr tips. a-j, dI/dV maps (left), their FTs (middle), and the corresponding “contrast” map $M(q, eV)$ (defined in the main text) at different biases.

Supplementary Note 6: STM data on the 2×1 reconstructed surface

While both the $\sqrt{2} \times \sqrt{2}$ and 2×1 reconstructed K surfaces are charge-neutral, we have focused our analysis on the $\sqrt{2} \times \sqrt{2}$ reconstruction for the following two critical reasons.

First, the surface corrugation for the 2×1 reconstruction is approximately 40 pm, which is approximately six times that of the $\sqrt{2} \times \sqrt{2}$ surface (~ 7 pm; [Figure S16](#)). This substantial height modulation tends to mask intrinsic electronic signatures on the 2×1 surface, such as high-resolution impurity shapes, scattering patterns and quasiparticle excitations ([Figure S17](#)), compared to those on the $\sqrt{2} \times \sqrt{2}$ surface. Consequently, the resulting QPI patterns on this surface appear blurred, particularly at negative bias voltages ([Figure S17c–e](#)).

Second, the reconstruction-induced structural anisotropy can complicate the interpretation of the electronic signatures. The 2×1 reconstruction is intrinsically unidirectional and introduces a strong, extrinsic structural anisotropy along either the x or y axis ([Figure S17c–e](#)). Given that the proposed altermagnetic spin polarization in $\text{KV}_2\text{Se}_2\text{O}$ occurs between the X and Y valleys, such a dominant structural anisotropy would severely complicate the disentanglement of intrinsic altermagnetic signatures from reconstruction-induced effects.

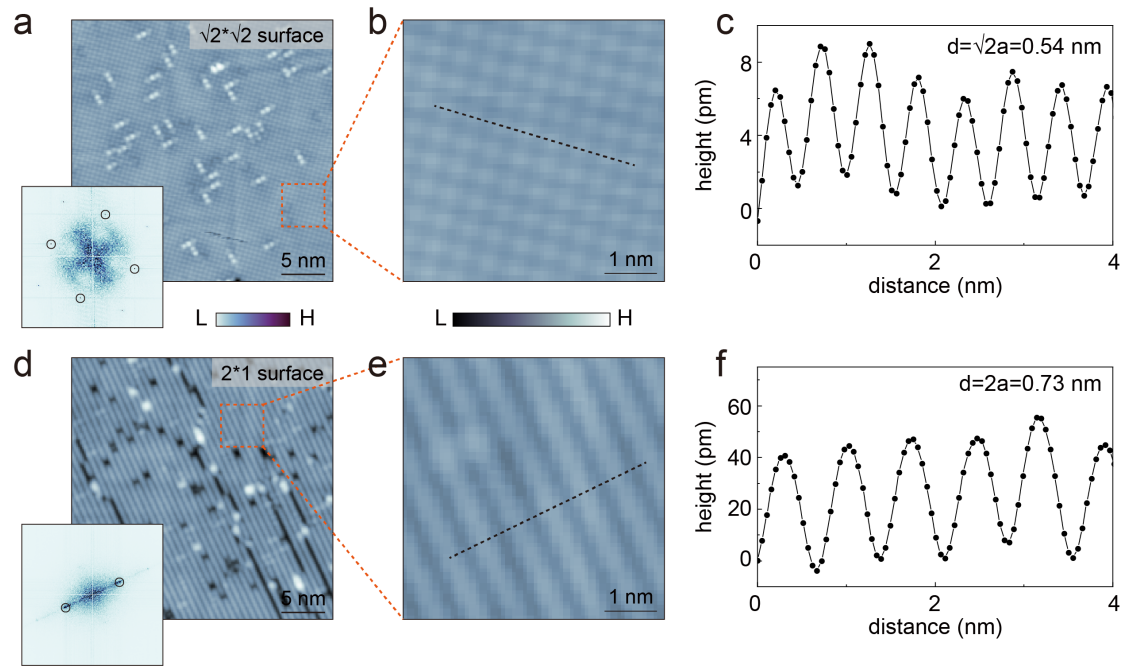


Figure S16 | $\sqrt{2} \times \sqrt{2}$ and 2×1 reconstruction of the K surfaces. **a**, Topographic image and its FT obtained on the $\sqrt{2} \times \sqrt{2}$ reconstructed surfaces. **b**, Zoomed-in topography over the small orange square in panel **a**. **c**, Linecuts taken along the surface lattice direction. **d**, Topography and its FT obtained on the 2×1 reconstructed surfaces. **e**, Zoomed-in topography over the small orange square in panel **d**. **f**, Linecuts taken along the line in **e** showing a periodicity of $2a$.

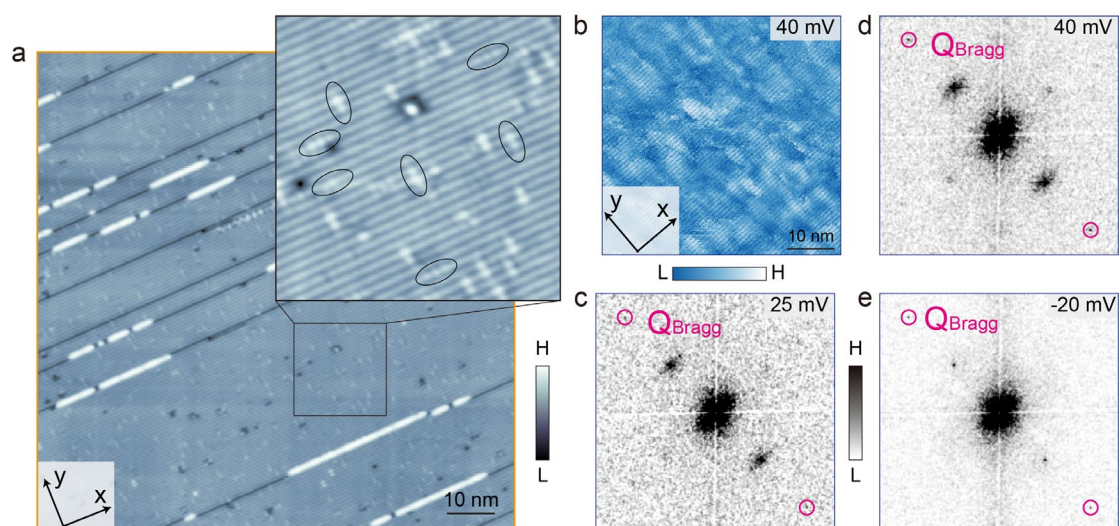


Figure S17 | Topographic image and QPI data on the 2×1 reconstructed surfaces. **a**, Topographic images of the 2×1 reconstructed surface in a large FOV. The inset shows a zoomed-in and the dark ellipses denote the V-site defects. **b**, dI/dV maps obtained on this kind of surface showing large modulation of the reconstruction. **c-e**, FFT of dI/dV maps at several energies.

Supplementary Note 7: Field-induced energy shifts of the in-gap excitations

These V-site defects in $\text{KV}_2\text{Se}_2\text{O}$ induce in-gap quasiparticle excitations that extend preferentially along either the x or y direction. As shown in Figure S18, multiple isolated defects exhibit opposite Zeeman shifts for the x - and y -direction excitations under out-of-plane magnetic fields B_z . Furthermore, we identified a single defect that simultaneously induces in-gap excitations along both the x and y direction (Figure 4i). The two directional excitations still exhibit strictly opposite Zeeman shifts under B_z . Since both originate from the same defect, this response is unlikely to be trivially explained by a defect-localized moment. Their modulation period is close to π/k_F , indicating that they carry information about the underlying Fermi surface and support the spin-polarized electronic structure of $\text{KV}_2\text{Se}_2\text{O}$. These reproducible shifts provide a real-space signature of the opposite spin polarization associated with the two directions.

To determine the primary spin orientation of these excitations, we performed field-orientation-dependent Zeeman spectroscopy at 30 mK. Fields of identical magnitude, ± 1.5 T, were applied along B_x , B_y , and B_z (Figure S19). To quantitatively address the anisotropic magnetic response, we performed additional field-dependent measurements in multiple regions. For each dataset, the peak energies were extracted by fitting the same spectral feature using an identical energy window and line-shape procedure. We define the field-induced shift per *unit field* as $|E(+B_i) - E(-B_i)|/(2|B_i|)$. Averaged over eight regions, the extracted shifts are 0.145 ± 0.021 meV/T for B_z , 0.0084 ± 0.0056 meV/T for B_x , and 0.0124 ± 0.0091 meV/T for B_y , where the quoted uncertainties denote one standard deviation across the measured regions rather than the uncertainty of an individual fit (Figure S20). The mean B_x and B_y responses are therefore 5.8% and 8.5% of the mean B_z response, respectively. The largest individual B_y response is close to 20% of the mean B_z response, consistent with the referee's visual estimate, whereas the largest B_x response is about 12%. These results demonstrate a strongly anisotropic magnetic-field response dominated by B_z , while allowing a smaller in-plane response.

Finally, we discuss how this conclusion is linked to the QPI measurements with Bi_4Br_4 tips. If the approximate ratio of field-induced shifts reflects the corresponding sample spin-polarization components, we can consider the geometric projections of both the in-plane and out-of-plane sample polarizations for the QPI data using Bi_4Br_4 tips. We use the measured mean response ratios as approximate proxies and consider a tilting angle of 10 degree for the tip, the projected contributions scale as $8.5\% * \cos 10^\circ \approx 0.084$ for the y direction and $5.8\% * \cos 10^\circ \approx 0.057$ for the x direction. By comparison, an out-of-plane component projects onto the tilted tip axis with a factor $\cos 80^\circ = \sin 10^\circ \approx 0.17$. Thus, the contributions estimated from the mean in-plane responses are smaller than, but not negligible relative to, the

geometrically projected out-of-plane contribution. Moreover, for the largest individual B_y response of approximately 20%, the corresponding illustrative projection is $20\% * \cos 10^\circ \approx 0.2$, which is comparable to the out-of-plane projection.

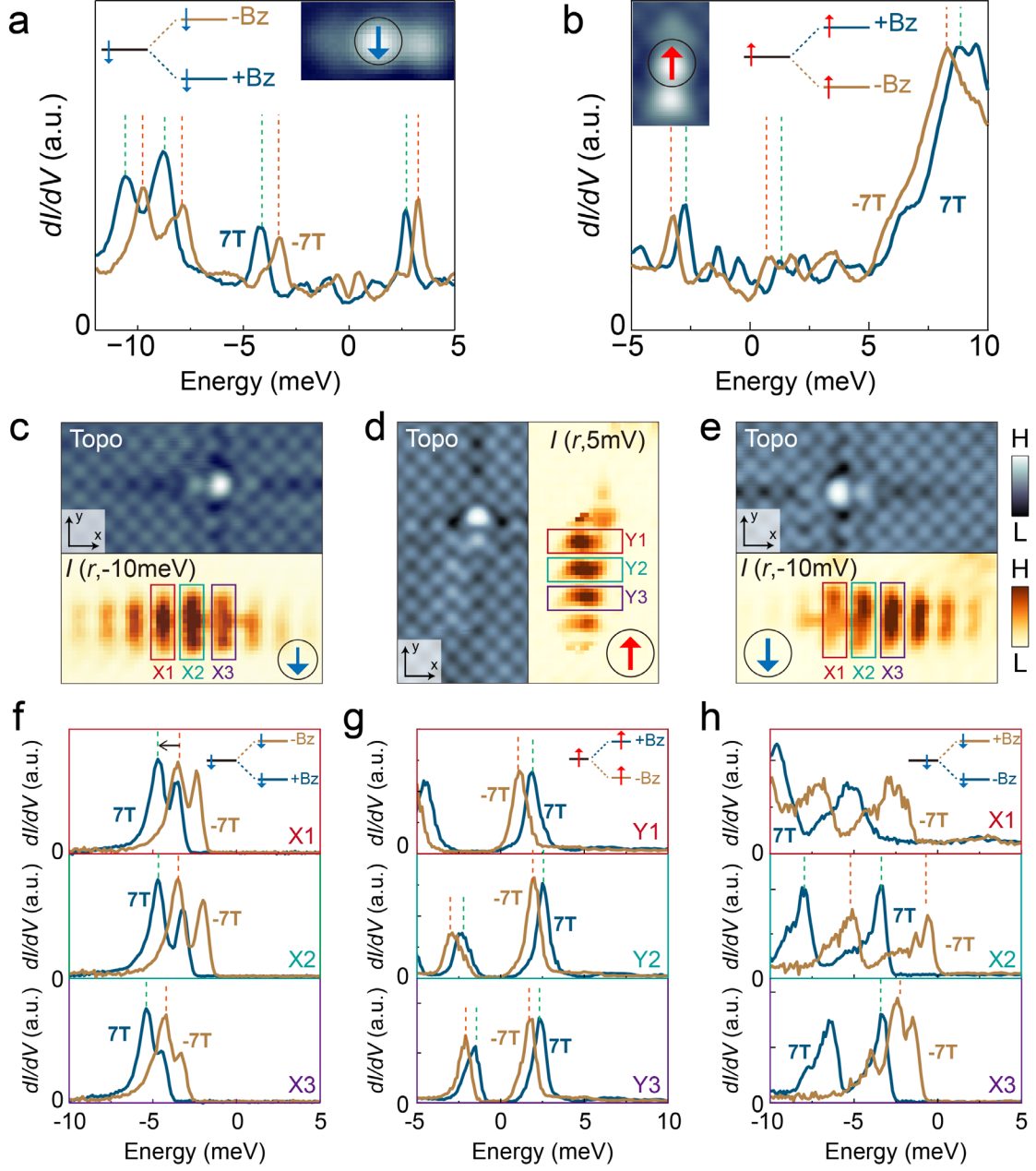


Figure S18 | More data showing Zeeman shifts of these in-gap excitations. **a, b**, Field-dependent peak shifts under B_z , for V_{xb} (**a**) and V_{yb} (**b**) defects. **c-e**, Topographic images and the current maps of three V-site defects that induce in-gap states along the y direction (**d**) and the x direction (**c, e**). **f-h**, Magnetic-field-induced Zeeman shifts under B_z for in-gap states along y -direction (**d**) and x -direction (**c, e**). The spectra are averaged and color coded for each region shown in **c-e**.

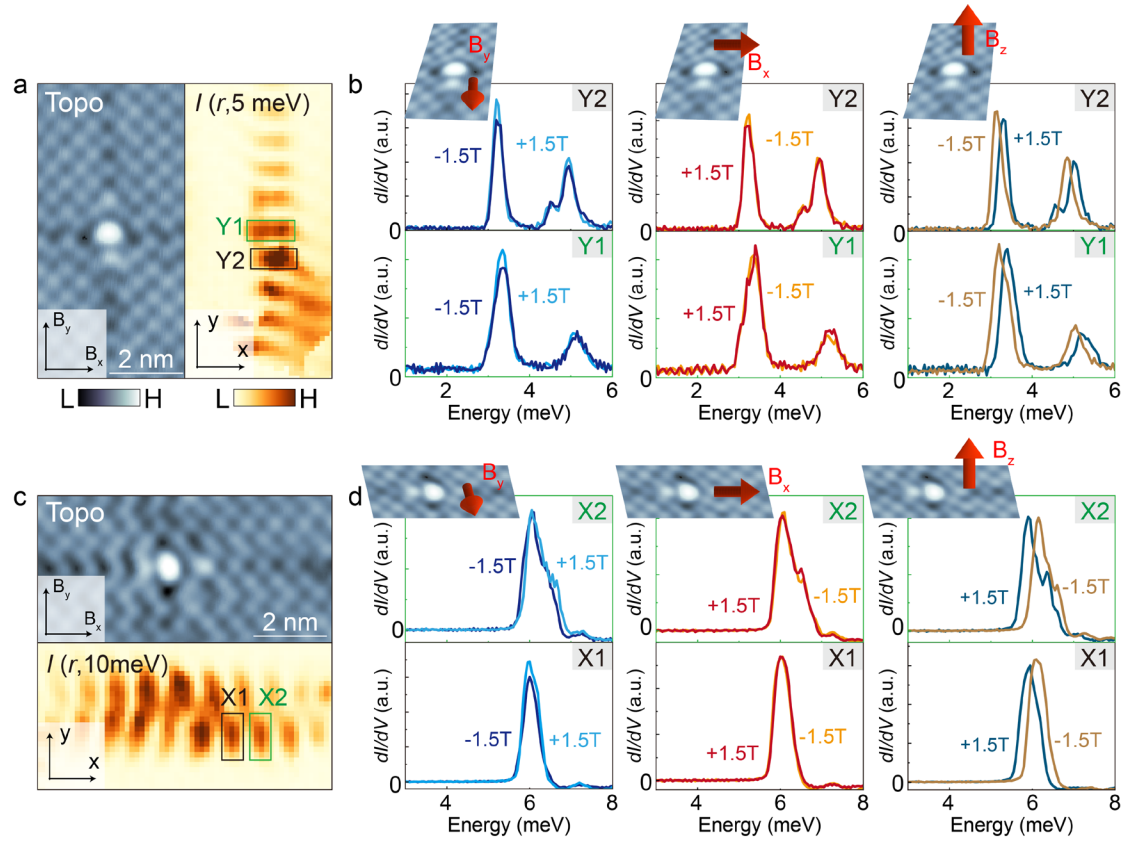


Figure S19 | Anisotropic field response of defect-induced in-gap states. **a, c,** Topographies (left panel) and the corresponding current maps (right panel) of isolated V-site defects that induce in-gap states extending along either the y (**a**) or x direction (**c**). **b, d,** dI/dV spectra acquired at 30 mK under magnetic fields of $\pm 1.5 \text{ T}$ applied along B_x , B_y , and B_z . The spectra are averaged and color coded for each region shown in **a** and **c**.

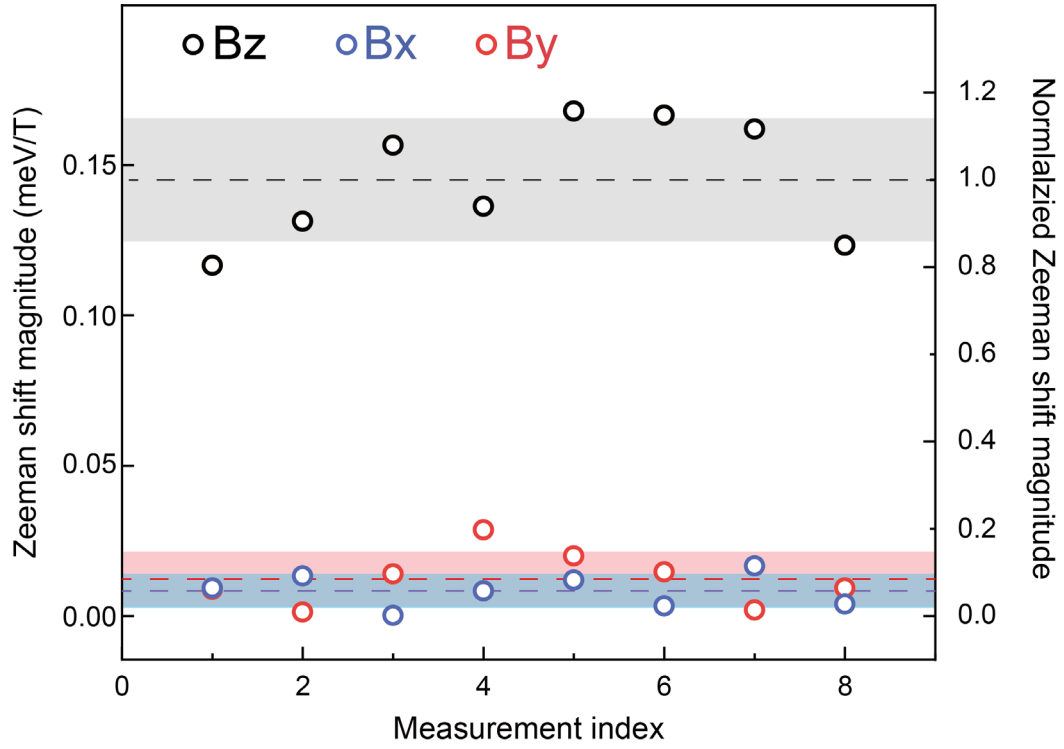


Fig. S20 | Quantitative anisotropy of the magnetic-field response of defect-associated excitations in $\text{KV}_2\text{Se}_2\text{O}$. Field-induced energy shifts per unit field extracted from multiple regions for fields applied along B_z (black), B_x (blue), and B_y (red). For each region, the shift is obtained from fits to the same spectral feature in the $+B$ and $-B$ spectra. Open circles represent individual regions, dashed lines indicate the averages, and shaded bands show one standard deviation across regions. The right axis is normalized to the mean B_z response.

Supplementary Note 8: Extended QPI analysis for KV₂Se₂O

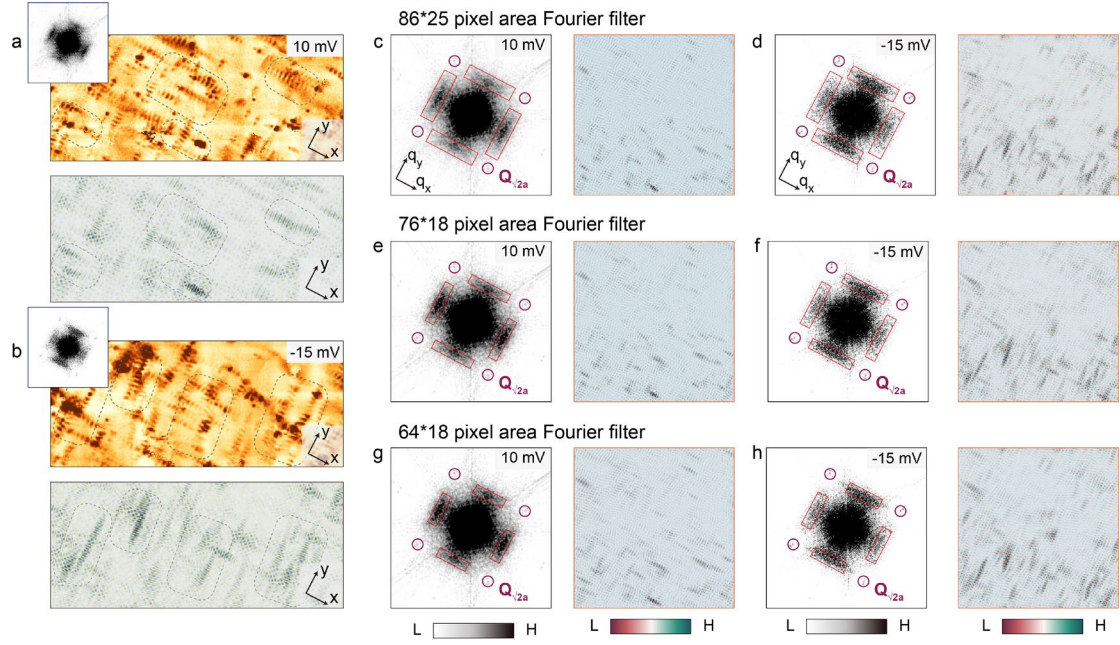
First, the process of inverse FT (Fig. 5g and i) needs special caution to avoid artifacts. We have performed a sensitivity analysis to confirm that our inverse FT results are not biased by the choice of integration windows. As demonstrated in Figure S21, we varied the q -space integration window size (ranging from a narrow window tightly bounding the QPI signatures (64 * 18 pixels) to a significantly larger window (86 * 25 pixels). We note that we always use symmetric windows with same size and shape for q_x and q_y . The resulting real-space modulations remain remarkably consistent across these variations, ruling out selection bias. Moreover, as shown in Figure S21a-b, this rotation of anisotropy at opposite biases can be identified in the raw dI/dV maps without any data processing: the real-space stripes along the y -direction are visibly more pronounced at negative bias compared to positive bias (marked by the dashed boxes), corroborating the inverse FT analysis.

Moreover, the reproducibility of this rotation of anisotropy has been verified across different samples and with different Bi₄Br₄ tips. In Figure S22, we present an independent dataset where the raw dI/dV maps, the QPI patterns, and the IFFT-filtered modulations all coherently support the claimed rotation of anisotropy.

Second, we note that the intensity of the $\sqrt{2}$ spots in the QPI maps basically follow the energy dependence of the spatially averaged dI/dV spectrum in the grid measurement, as shown in Figure S23. Physically, this is expected: the intensity of any spatial modulation is also weighted by the local DOS at a given energy. In the extreme case of a hard superconducting or semiconducting gap, the modulation intensity would vanish within the gap due to the absence of states, regardless of the underlying atomic structure. For KV₂Se₂O, it has a particle-hole-asymmetric energy gap near E_F . Consequently, the dI/dV intensity (and thus the modulation amplitude) is naturally suppressed near E_F . As shown in the comparative data in Figure S23, this energy-dependent trend is consistently observed using both conventional PtIr tips and our Bi₄Br₄ tips. The fact that the structural spots exhibit similar behavior across different tip species further validates the stability of our tunneling junction and the intrinsic nature of the observed electronic structure. Additionally, the $\sqrt{2} \times \sqrt{2}$ density wave (Q^*) folds the original bands to two branches and opens an energy gap of approximately 20 meV in the energy spectrum of k -space eigenstates. The two branches of band dispersions are shown in Figure S24.

Finally, we examine the possible anisotropy introduced by the density-wave order. We constructed a theoretical model incorporating both the AM and density-wave orders within the enlarged $\sqrt{2} \times \sqrt{2}$ unit cell. Although the density-wave order folds the bands, its energy scale of 0.02 eV is much smaller than that of the AM exchange field (−0.9 eV). As shown in Fig. S25a–d, the calculated density-wave gaps along the Γ –M1 and Γ –M2 directions are nearly

identical, with a difference below 0.1 meV. Consistently, the QPI dispersions extracted along q_x^1 and q_y^1 show no appreciable difference (Fig. S25e, f). These results demonstrate that the density-wave-induced band-folding anisotropy is unlikely to account for the pronounced QPI anisotropy observed with Bi₄Br₄ tips.



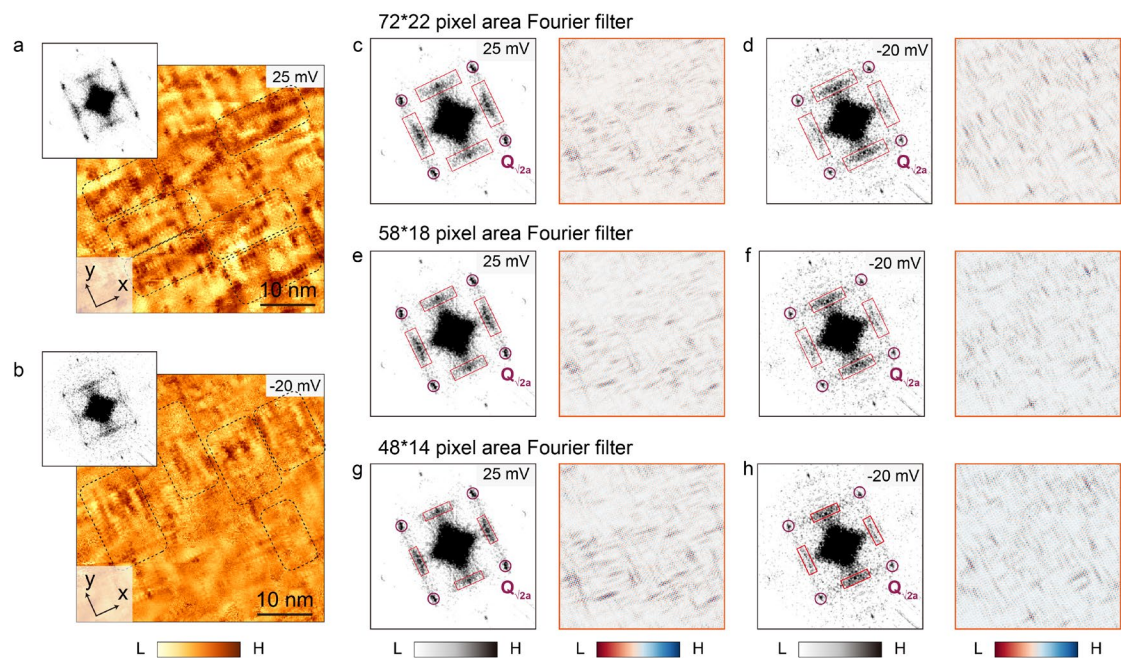


Figure S22 | The raw dI/dV maps and Fourier-filtered real-space QPI patterns on a second sample. **a, b**, Raw dI/dV maps in the same field of view of a grid measurement, acquired at 25 and -20 mV. **c-h**, inverse FT results obtained using different sizes for q -space integration window. Here, symmetry-related windows of identical size and shape centered at q_x^1 and q_y^1 are used.

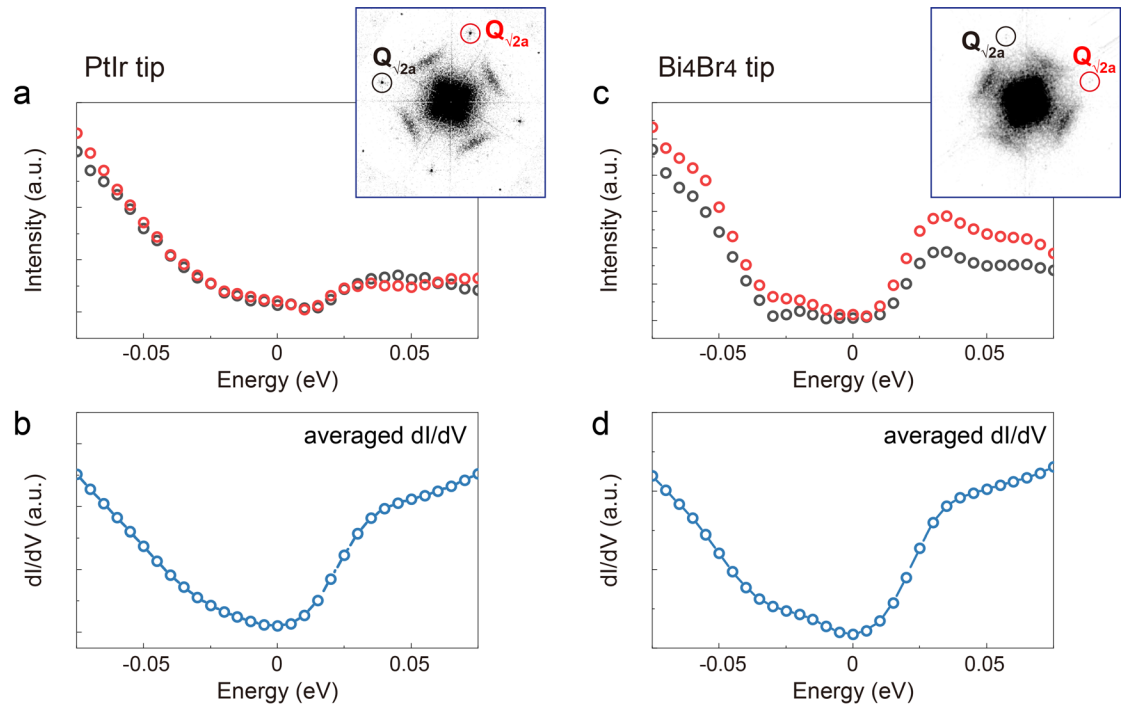


Figure S23 | Energy-dependent intensity of the $\sqrt{2}$ spots in the FFT. **a**, The intensities of the two $\sqrt{2}$ spots in the FT as a function of energy, using a PtIr tip. **b**, Spatially averaged dI/dV spectrum obtained from the same spectroscopic grid dataset. The intensities of the $\sqrt{2}$ spots closely follow the energy dependence of the spatially averaged dI/dV spectrum. **c-d**, Similar analysis for data taken with a Bi_4Br_4 tip. In both cases, the intensities of the $\sqrt{2}$ spots in the QPI maps basically follow the energy dependence of the spatially averaged dI/dV spectrum obtained from the same grid measurement.

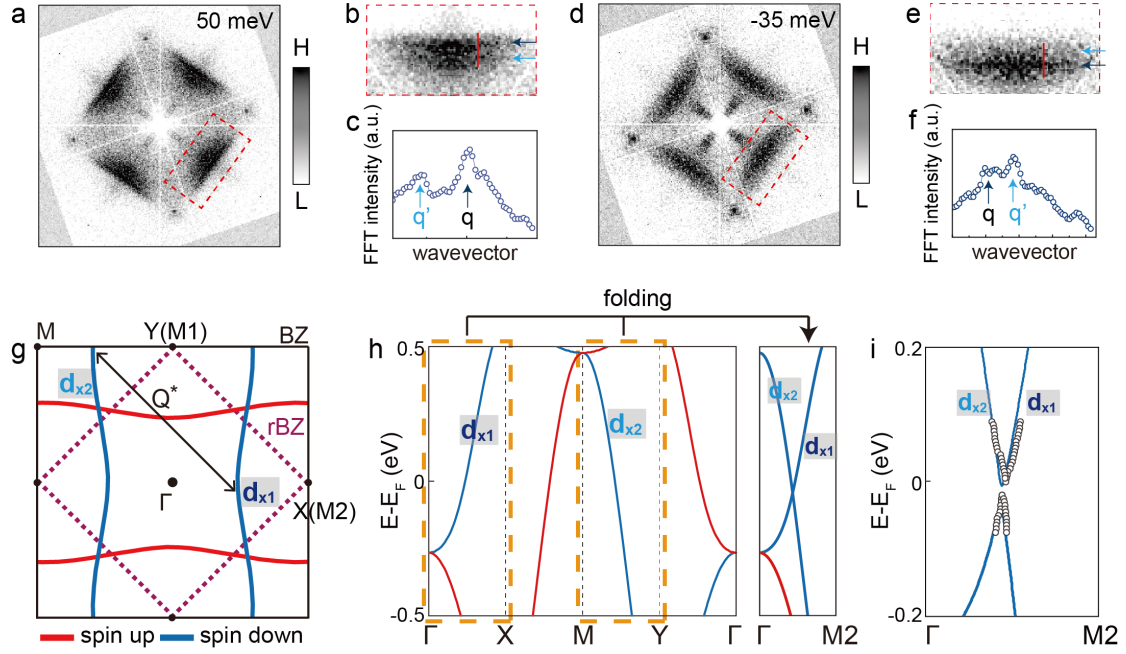


Figure S24 | The band folding feature in QPI images. **a**, QPI patterns at 50 meV. **b**, Enlarged view of the dashed region in panel **a**. **c**, FT linecut along the red line in **b**, showing two scattering features associated with band folding. **d-f**, Similar analysis as in **a-c**, but measured at -35 meV. **g**, Schematic of the pristine Brillouin zone (BZ, black) and the reduced Brillouin zone (rBZ, purple) generated by the density-wave reconstruction. The red and blue curves denote the spin-up and spin-down Fermi surface sheets, respectively. The bands labeled d_{x1} and d_{x2} are folded into the same momentum region by the reconstruction wavevector Q^* . **h**, Pristine band structure along the Γ -X-M-Y- Γ path (left), and its folded counterpart along the Γ -M2 direction of the rBZ (right). Along Γ -M2, the pristine Γ -X and M-Y bands are folded onto one another. **i**, Folded band structure along Γ -M2, showing the hybridization gap arising from coupling between the folded d_{x1} and d_{x2} bands. Open circles denote the experimentally extracted dispersion.

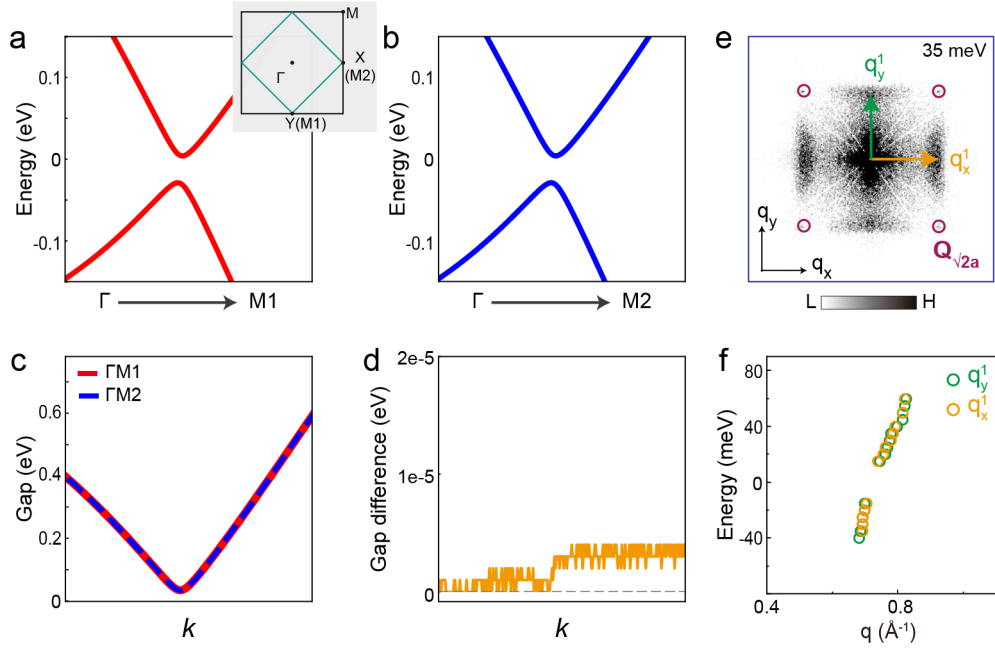


Figure S25 | Possible anisotropy introduced by the density-wave order. **a, b,** Calculated band dispersions along the Γ -M1 (Γ -Y in the original BZ) and Γ -M2 (Γ -X) directions, respectively. **c,** Calculated density-wave gaps along the two directions, which remains below 0.1 meV. **d,** Difference between the calculated density-wave gaps along the two directions, which remains below 0.1 meV. **e,** Representative QPI map acquired at 35 meV, with the two scattering vectors q_x^1 and q_y^1 indicated. **f,** Energy dependence of q_x^1 and q_y^1 extracted from the QPI data.

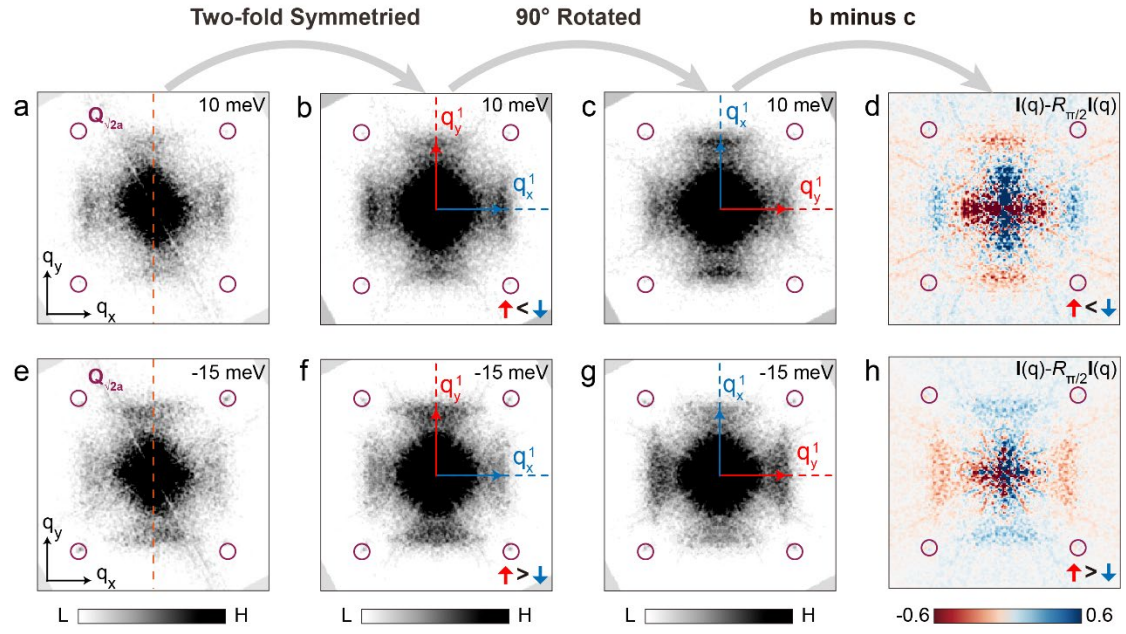


Figure S26 | Procedures to obtain the 2D contrast maps $M(\mathbf{q}, eV)$. **a**, Unprocessed $\mathbf{q}$ -space QPI patterns at 10 meV. **b**, Two-fold symmetrization of **a**. **c**, 90° clockwise rotation of **b**. **d**, Contrast map $M(\mathbf{q}, eV)$, defined as difference between **b** ($I(\mathbf{q}, eV)$) and **c** ($R_{\pi/2} I(\mathbf{q}, eV)$), which is further normalized by the intensity of simultaneously obtained surface Bragg peak, $I(Q_{Bragg}, eV)$. **e-h**, Same process as shown in **a-d** for images at -15 meV.

Supplementary Note 9: Magnetic order calculations for KV₂Se₂O

Here we perform DFT calculations to identify the magnetic ground state of KV₂Se₂O. The DFT calculated energy per unit cell for non-magnetic (NM), ferromagnetic (FM), and antiferromagnetic (AM) states are shown in Fig. S27. The AM state is found to have the lowest energy. Meanwhile, this ground state result is further corroborated by the band dispersions, where the AM bands match well with the experimental data (see the main text and extended data Fig. 7 for details).

We further examine three different antiferromagnetic configurations within the ab plane, including Néel, stripy, and zigzag types (Fig. S28). Among these, the Néel-type AFM (AM state) exhibits the lowest energy and thus the most stable configuration.

Moreover, we examine the magnetic order along the c direction. Figure S29 shows three types of out-of-plane AFM configurations (G-, C-, and A- type) and their corresponding energy per unit cell. The inset of Fig. S29d shows the near degeneracy in energy of G-type and C-type AFM states, with an energy difference in the order of meV, due to the weak interlayer interaction in KV₂Se₂O.

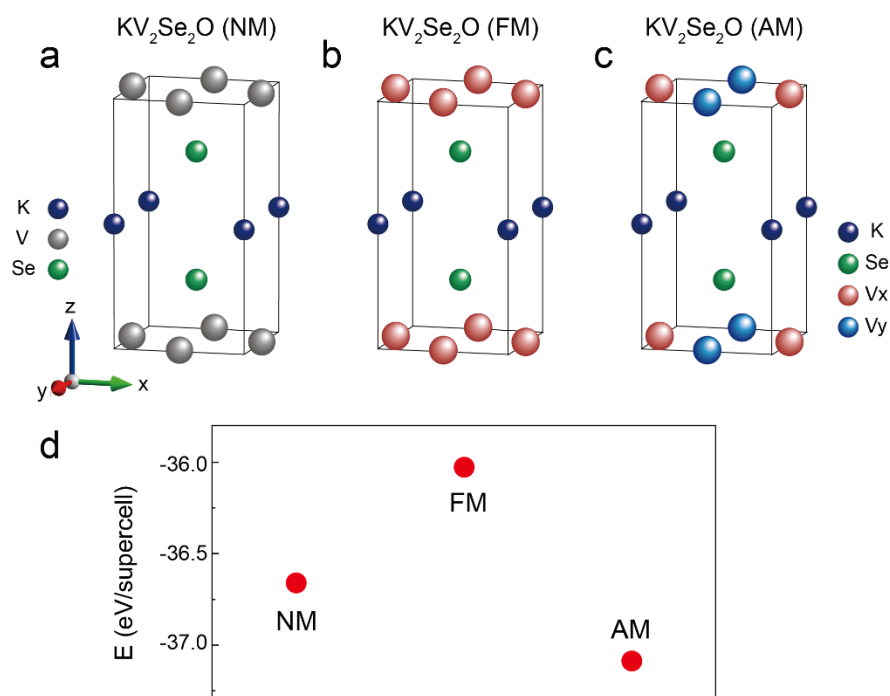


Figure S27 | Magnetic configurations and energy comparison of different magnetic states for unit cell. **a–c**, Magnetic structures of the three types of magnetic states, with nonmagnetic, spin-up, spin-down V atoms are colored by gray, red and blue, respectively. **d**, Energy calculations of the three types of magnetic orders, where the altermagnetic configuration exhibits the lowest energy.

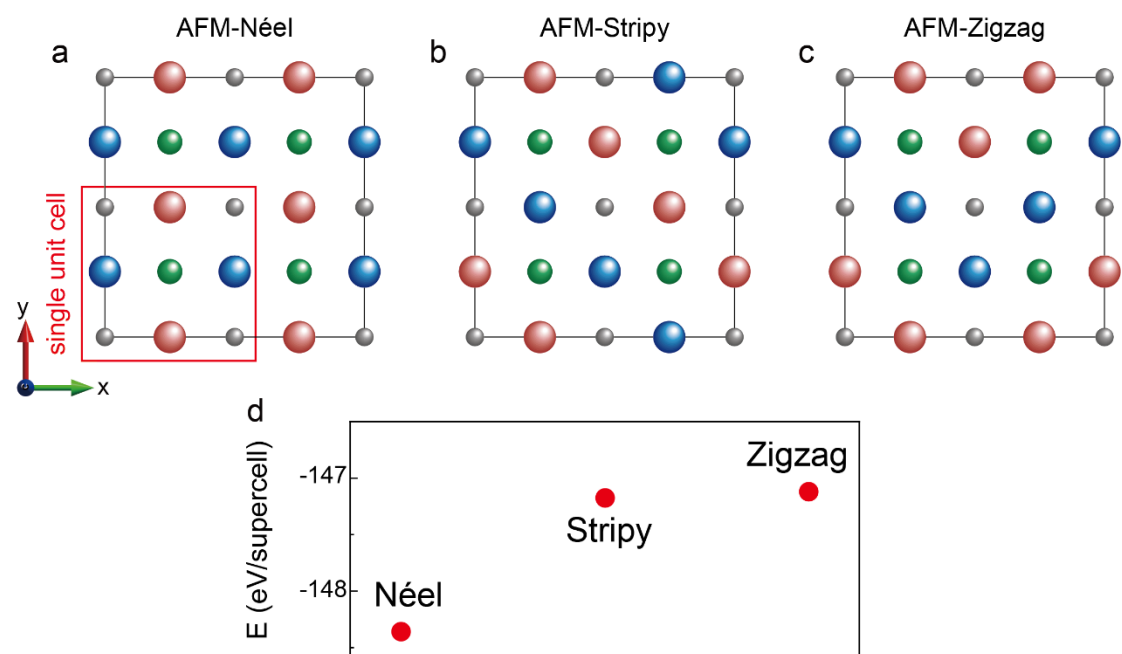


Figure S28 | Magnetic configurations and energy comparison for three *ab*-plane AFM orders. **a–c**, Magnetic structures of the three types of AFM orders, with spin-up and spin-down V atoms colored red and blue, respectively. **d**, Energy calculations of the three types of magnetic orders. The Néel-type (antiferromagnetic) configuration exhibits the lowest energy.

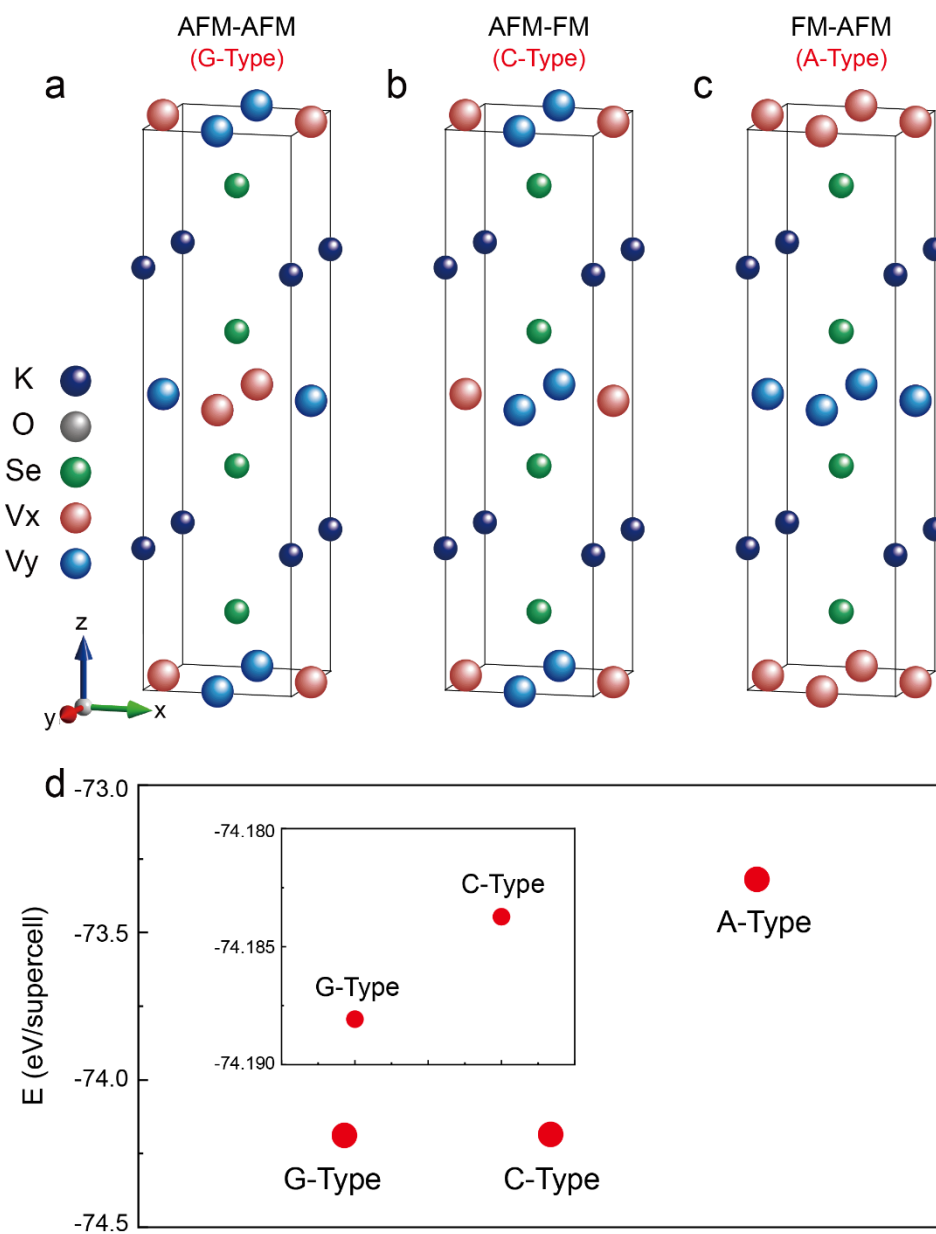


Figure S29 | Magnetic configurations and energy comparison for three *out-of-plane* AFM orders. **a–c**, Schematic $1 \times 1 \times 2$ supercells for the G-type, C-type, and A-type AFM orders, respectively. Red and blue spheres represent V atoms with spin-up and spin-down polarization, respectively. **d**, Calculated energy per supercell for the three AFM configurations. *Inset*: A zoomed-in view for the G-type and C-type AFM states, which are nearly degenerate, with a very small energy difference on the order of meV.

Supplementary Note 10: Spin-polarized calculations for $\text{KV}_2\text{Se}_2\text{O}$

Here we present the spin-polarized calculations for electronic properties of $\text{KV}_2\text{Se}_2\text{O}$, including the anisotropic spin-density distribution in real space (Figure S30), the spin-polarized Fermi surfaces at different energy cuts in reciprocal space (Figure S31), the spin-polarized band structures in the presence of SOC (Figure S32), and the orbital-resolved band structures along high-symmetry lines (Figure S33).

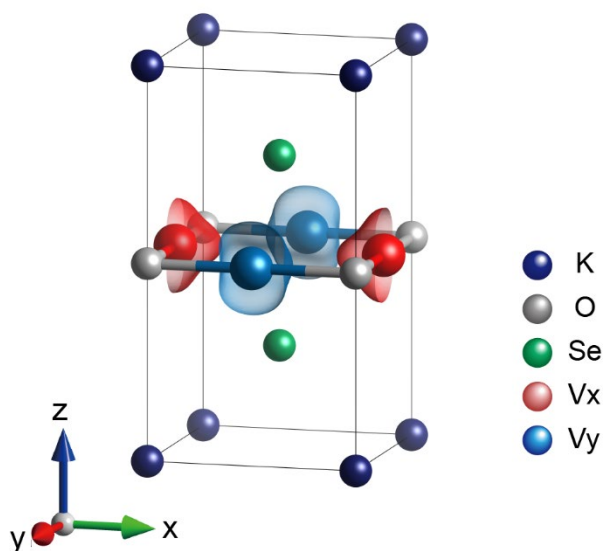


Figure S30 | Real space spin density distribution. Blue and red isosurfaces represent the spin-up and spin-down charge densities, respectively. The spin-up (spin-down) density exhibits strong anisotropy along the x and y directions, while being connected to each other by crystal symmetry C_4 and mirror.

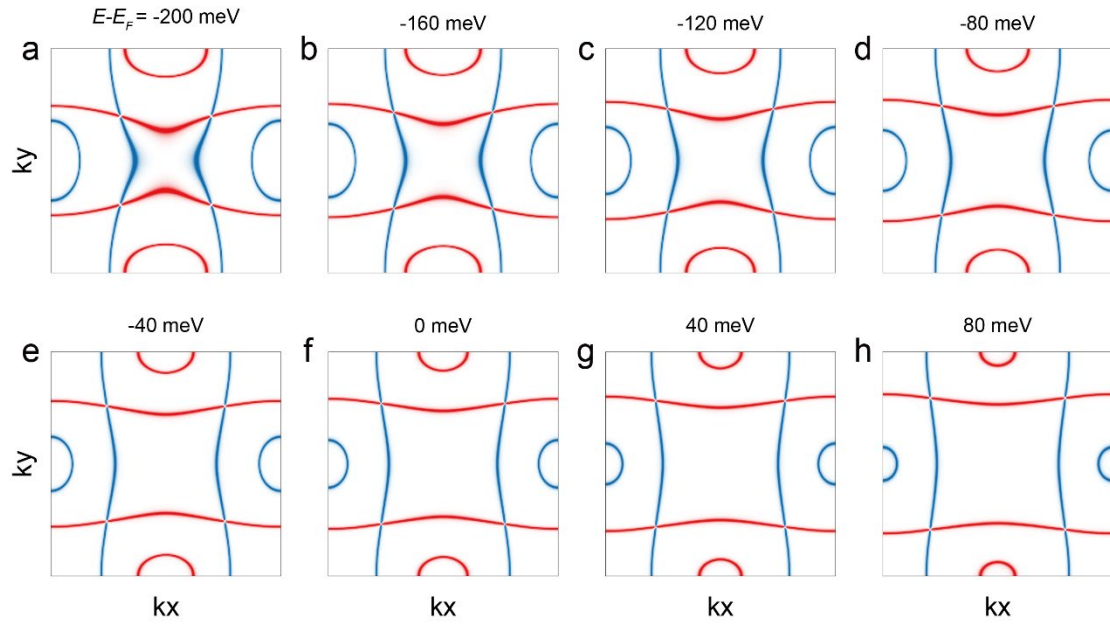


Figure S31 | Spin-polarized Fermi-surface in the $k_z = 0$ plane for different energy cuts.

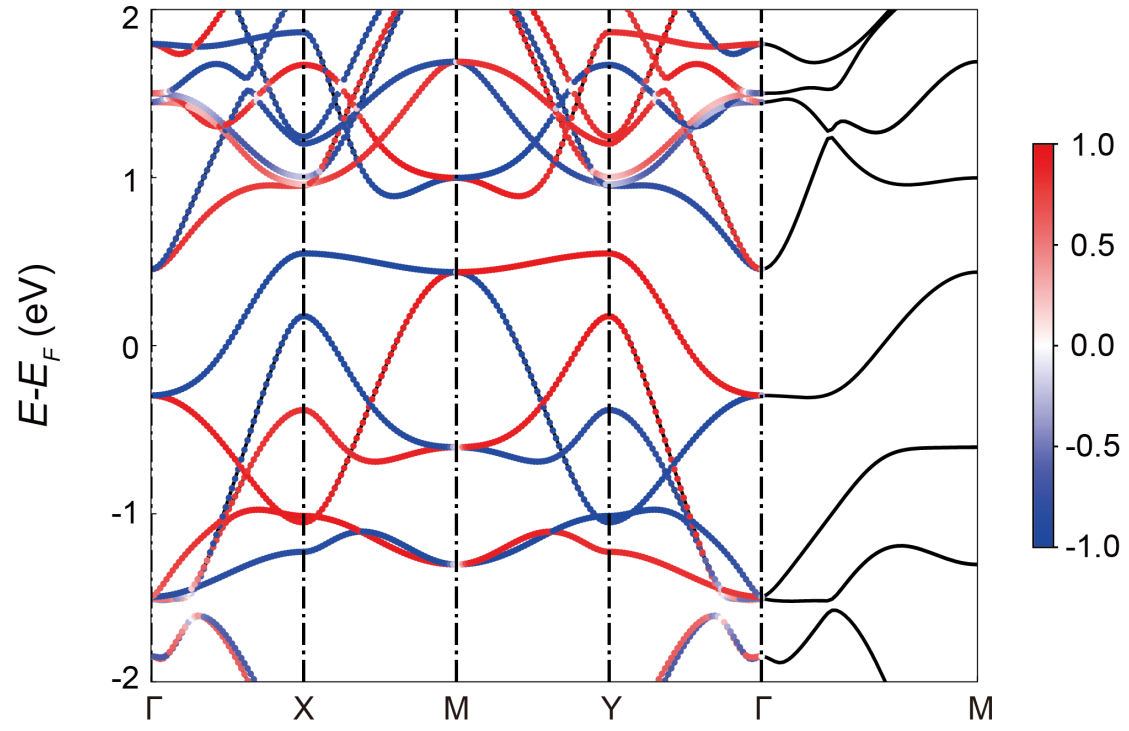


Figure S32 | Band structures and spin-polarization with spin-orbit coupling (SOC). The band structure and spin-polarization S_z remain largely unaffected in the presence of SOC.

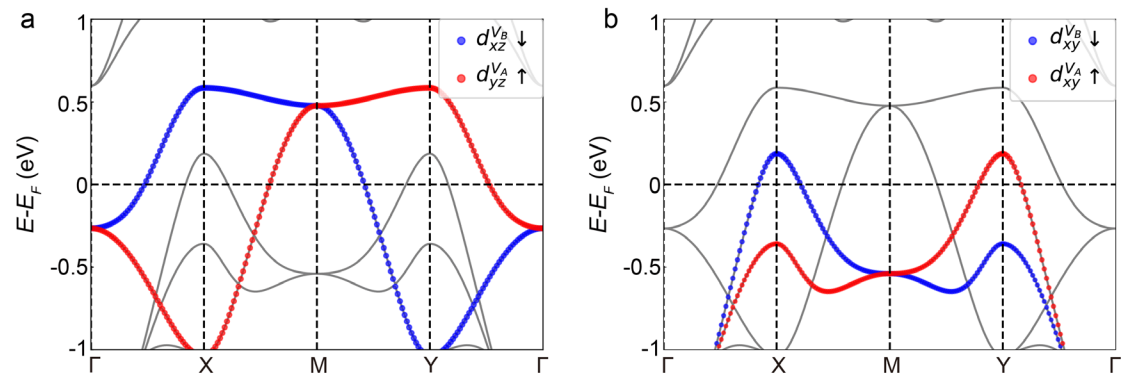


Figure S33 | Calculated orbital- and spin-resolved band structures along high-symmetry lines. Red and blue dots represent spin-up and spin-down bands. The size of the dots scales the projection of orbital components.

Supplementary Note 11: Discussion on the orbital character of QPI pattern in Sr_2RuO_4 .

The Fermi surface of Sr_2RuO_4 has been extensively studied by theoretical calculations and ARPES measurements in the past decade [S17-S20], so that its main orbital character has been established. To further illustrate the orbital feature in QPI patterns, we performed orbital-resolved QPI calculations based on the published three-orbital tight-binding model of [S21]. The model includes the Ru d_{xz} , d_{yz} , and d_{xy} orbitals, together with interorbital hybridization and spin-orbit coupling, and the QPI patterns were calculated using a T-matrix approach for the weak-scattering potential case.

In the calculation (Fig. S34b), the principal q_x^1 feature arises predominantly from intraband scattering between the nearly vertical, quasi-one-dimensional segments of the β band, which have mainly d_{xz} character. The d_{yz} -dominated segments can also generate a very weaker scattering contribution along the same direction. Correspondingly, the principal q_y^1 feature is predominantly associated with the horizontal d_{yz} -dominated segments, with a weaker contribution from d_{xz} . The calculated orbital-resolved QPI signals that we are focusing on here (q_x^1 and q_y^1) are broadly consistent with those reported by Kreisel *et al.* in [S22] (Fig. S34c). Thus, the calculation supports a dominant (but not orbitally pure) d_{xz} and d_{yz} assignment of the q_x^1 and q_y^1 features.

The pronounced axial intensity maxima may be attributed to a concentration of scattering processes at similar wave vectors due to the nearly parallel constant-energy segments of β band. We repeated the QPI anisotropy analysis using three integration-window choices: the full outer QPI segments, the off-axis arc portions with the axial maxima excluded, and the axial maxima alone (Fig. S35). In all three cases, the anisotropy remains below approximately 3% and does not reverse sign with bias polarity.

Therefore, different modelling conditions could give rise to different quantitative details in the QPI calculations and a detailed quantitative modeling analysis is difficult to achieve at the current stage, but the main orbital features for the QPI signatures we focused on here is consistently obtained in these models. Although potential orbital mixing and material-dependent tunnelling matrix elements prevent this comparison from providing a quantitative calibration of orbital sensitivity, the results argue against a generic dominant orbital origin of the observed contrast in $\text{KV}_2\text{Se}_2\text{O}$.

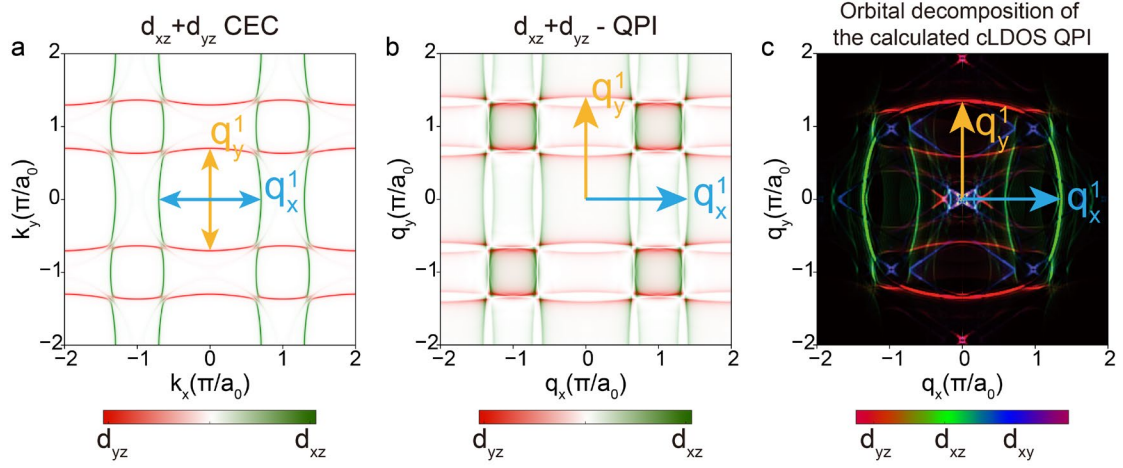


Figure S34 | Main orbital characters of quasiparticle interference in Sr_2RuO_4 . **a**, Orbital-resolved spectral density at the Fermi level for the d_{xz} and d_{yz} states, shown in green and red, respectively. Here, we neglect the contribution from the in-plane d_{xy} orbital, which is typically difficult to detect in STM-based QPI measurements owing to its weak coupling to the STM tip [S19, S21, S23]. The arrows indicate the main scattering vectors q_x^1 and q_y^1 connecting approximately parallel contour segments. **b**, Calculated QPI patterns, with the same color convention and scattering vectors as in **a**. The model is following that in Ref. S21. **c**, Orbital-decomposition QPI pattern with different modeling that is reproduced from Fig. 5 of Kreisel *et al.*[S22]. It shows the QPI obtained from the calculated continuum local density of states (cLDOS) at $E=2$ meV; red, green, and blue represent d_{yz} , d_{xz} , and d_{xy} character, respectively. The main orbital features at q_x^1 and q_y^1 are broadly consistent for these results in **b** and **c**.

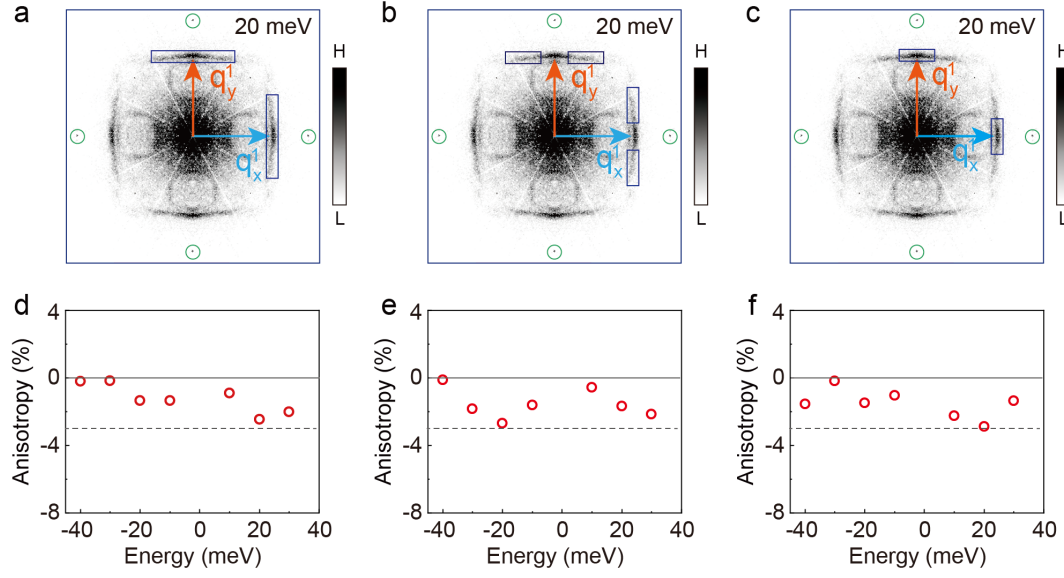


Figure S35 | Robustness of the Sr_2RuO_4 QPI anisotropy against integration-window selection. **a–c**, QPI maps at 20 meV with black rectangles marking the full outer QPI segments (**a**), off-axis portions excluding the axial maxima (**b**), and axial maxima alone (**c**). **d–f**, Corresponding energy-dependent anisotropies between the q_x^1 and q_y^1 features. All three window choices yield similarly small anisotropies, with no contrast reversal upon changing the bias polarity. Dashed lines indicate zero anisotropy.

Supplementary Note 12: Proposed mechanisms for net spin polarization via helical tunneling in Bi₄Br₄ tips

The spin-polarization capability for a Bi₄Br₄ tip via helical tunneling is still a fundamental question. The inherent presence of two counter-propagating edge channels with opposite spins, which form closed loops circulating along the tip boundaries, can cancel each other and lead to reduced net spin polarization. In principle, the measured effective spin polarization is dictated by the microscopic details of how these helical edge-state electrons tunnel through the vacuum barrier to yield a net (non-zero) spin contrast. While a comprehensive theoretical framework for this foundational tunneling problem remains to be fully developed, we propose several plausible mechanisms grounded in our experimental observations.

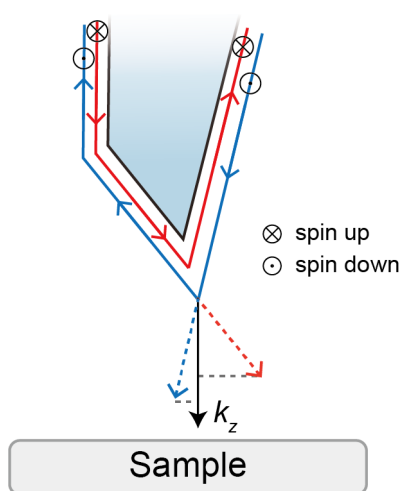


Figure S36 | Quasi-momentum description of the tunneling process.

First, the observed spectral line-shape of the Bi₄Br₄ tip on Au surface closely resembles that of the edge states found on the bulk crystal. This consistency corroborates the existence of edge channels near the tip apex and may imply that the effective tip extremity maintains a finite width (> 5 nm) rather than being atomically sharp (Extended Data Fig. 1d). In this case, even if the strict conservation of quasi-momentum could be relaxed at the localized tip apex, the effective “ k_z ” components (which govern the vacuum tunneling matrix elements) are expected to differ between the two adjacent edges (Fig. S36). This spatial and momentum asymmetry can result in the tunneling current being dominated by one specific edge channel, thereby generating a net spin resolution. However, the concurrent contribution from the secondary channel naturally reduces the overall effective spin-polarization ratio.

Second, if we consider the tip apex as a point-like object, concept of “quasi-momentum” becomes ill-defined and invalid. In this case, the precise mechanism by which the tunneling of helical states generates spin resolution becomes less intuitive. For example, for a tunneling

current directed towards the sample, spin-up electrons can take the path on the right edge while spin-down electrons may take the left edge (Fig. S37a). To achieve a net spin resolution, these two paths *must be inequivalent*. This becomes possible because the unidirectional current can break the time reversal symmetry of the helical edges. We propose two plausible mechanisms to account for this symmetry breaking.

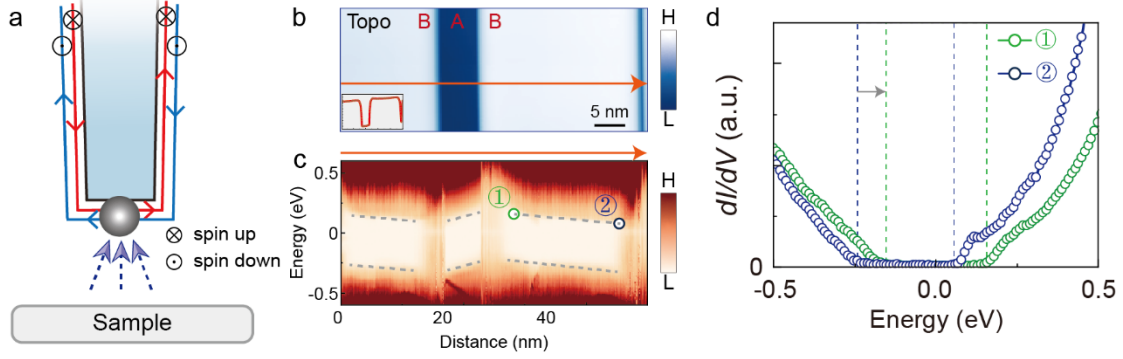


Figure S37 | Asymmetric spectral feature of edge modes on two sides of a terrace. **a**, Sketch for the tunneling condition with a point-like tip apex. **b**, STM topography including one terrace and one step edge of monolayer Bi_4Br_4 . **c**, Linecut profile of dI/dV spectra across these terraces. A pronounced band-bending effect near the step edges can be observed. Interestingly, the bulk gap shifts towards lower and higher energies on opposite sides of a single layer. **d**, Two dI/dV spectra taken at two locations near the edge to show the relative energy shift on the opposite edges.

a) Asymmetric Band Bending. Experimentally, we found that Bi_4Br_4 exhibits a pronounced band-bending effect near the step edges. The bulk gap shifts by ~ 50 meV towards lower and higher energies on opposite sides of a single layer (Fig. S37b-d). This implies that the helical edge states on the left and right sides experience a relative energy shift. Because the tip is grounded (fixing the Fermi level for both sides during measurements), the LDOS contributing to the tunneling process can be inherently different for these two paths, thereby yielding a net spin-polarized current.

b) Nonlinear Current-Voltage Response. The essential experimental signal (dI/dV) that shows resolution of real-space spin texture has two major properties:

- (1) the signal flips sign when the applied voltage is reversed;
- (2) the signal has opposite sign when the tip is aligned with lattice sites with opposite local spin orders.

For small applied voltage, the first property indicates that the linear in V term is most relevant to the leading order, i.e., $\frac{dI}{dV} = \alpha V$. The second property can be fulfilled if $\alpha \propto m_{\text{local}}$. We can rewrite this signal in terms of current, which then has the form:

$$I = \frac{1}{2} \alpha V^2$$

with $\alpha \propto m_{local}$. Such a second order current has the same symmetry property with the nonlinear Hall effect [S24, S25]. The latter is usually discussed in the diffusive regime [S24-S26]. The present experiment signal can then be viewed as the ballistic version of the nonlinear Hall effect. The generalization of the nonlinear Hall effect from the diffusive regime to the ballistic regime has been discussed previously using Boltzmann theory [S27] or the Landauer-Büttiker formula [S28]. Therefore, we expect that the intrinsic nonlinear Hall effect in the ballistic regime can well capture the antiferromagnetic order, as shown in our series of experiments.

We would like to note that a complete microscopic theory of tunneling from topological helical states into a magnetic substrate remains an open and foundational question. Our work establishes the experimental phenomenology of this novel probe, and we anticipate that these findings will stimulate further theoretical investigations into the quantum transport properties of topological tunneling junctions.

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