

Atomic-scale spin sensing of a 2D *d*-wave altermagnet via helical tunneling

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This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Referee #1

(Remarks to the Author)

Zhuying Wang et al. present in their manuscript “Atomic-scale spin sensing of a 2D *d*-wave altermagnet via helical tunneling” two interesting advances: They create an STM tip from the topological insulator α -Bi₄Br₄, and use it to resolve the spin splitting in the altermagnet candidate KV₂Se₂O in real space and momentum space. As this extensive and detailed work combines topological systems with unconventional magnetism, a new method and timely physics, I believe it has potential to be of interest to several communities.

As there are already spin-resolved ARPES measurements on the compound (Ref. 40), the novelty for the material is rather the STM real space resolution of the magnetic order, which has been convincingly achieved in Fig. 4 of the present manuscript using magnetic fields. The authors chose an altermagnetic candidate compound, where due to an extra transition within the magnetic phase, surface reconstruction, and non-stoichiometry the magnetic and crystalline symmetries are subject to uncertainty. Given such complications their result is important.

To make the manuscript suitable for Nature, I believe the method claim has to stand beyond doubt. My central questions concern the interpretation of the topological tip and the size of spin resolution:

1. Reference 40 showed that spin-resolved ARPES indicates *d*-wave altermagnetism, albeit only with a low 10% spin polarization, which might be due to the magnetic domain size. Thus, a finer probe like SP-STM should be a good approach to improve the spin-resolution. For the tip material, the supplement of Ref. 60 mentions a bulk spin *S_z* polarization of as little as 3° deviation, though I failed to find a statement about the spin polarization of the surface states. Compared to these results and the authors’ claim of nearly spin-quantized surface states, the present manuscript reports only 12% of effective spin polarization which varies between tips. The authors suggest the tip anisotropy as optimization criterion, which follows the principle shown in Fig. 1c, but at least from extended Fig. 2 this conclusion is not clear, as the four used tips do not seem particularly asymmetric. This makes it unclear whether just more engineering or a physical improvement is necessary to optimize the spin resolution.

2. Connected to the question of spin polarization: As the authors point out that “... the spin orientation for the edge modes is ‘horizontal’ (Fig. 1c), whereas that for the AFM order in KV₂Se₂O is out-of-plane”. If the near perfect “*S_z*” polarization of the tip is 90° rotated to the substrate, then this seems to mean to me that there should be no spin selectivity as an *S_z* > 0 state of the substrate should hop with equal probability to, for example, *S_x* > 0 and *S_x* < 0 of the tip. This has to be cleared up before the main claim becomes believable.

3. The TI exhibits helical edge states that may slightly gap out due to interlayer hybridization leaving still topological hinge

states. Often for hinge states it is important that the macroscopic sample is invariant under the protecting symmetry. The symmetry requirements for hinge-state protection in a conical tip geometry remain unclear. How do the authors rationalize the hinge mode? Connected to this, the manuscript does not clearly explain the orientation of the helical surface states of the 3D QSHI phase: In Fig. 2a it is rather clear that these propagate on the sides of the (010) layers, but the sample orientation in Fig. 2e or Fig. 1c is less clear. Why do the helical edge modes just end? As extended Fig. 1c, d shows the tips would be wide enough to stabilize the 3D QSHI modes running through the tip.

4. Figure S8b, e seems to have a similar anisotropy between x and y for some values of q as Fig. 5g, i. Why is that? Also regarding anisotropies, if the tip states of α -Bi₄Br₄ are quasi 1D, would that not also affect tunneling probabilities besides the spin, especially since helical and substrate spin axes are orthogonal?

Smaller questions:

- When the bias changes sign the contributions of q_1x and q_1y momenta change sign, as far as I can tell the intensity at smaller momenta does not do that. Is there a reason? Does the JDOS in Fig. 5e change with the offset?

- In Fig. 2h the stripe pattern is stated to reverse with reversed bias. What is the reason for the deviation from a perfectly mirrored pattern? Besides differences in the shape of curves they are also shown with an offset. Is this offset just by presentation or measured?

- In line 159 the authors speak of "twofold defects", in what sense are they twofold?

Referee #2

(Remarks to the Author)

The authors introduce a spin-sensing method for functionalizing STM tips using the bulk crystal of a HOTI α -Bi₄Br₄. The central premise is that the helical edge modes of the HOTI, which possess spin-momentum locking, can act as a spin filter without the need for magnetic tip coatings or external magnetic fields. They apply this helical tunneling technique to KV₂Se₂O, a candidate for 2D d-wave altermagnetism, and claim to observe QPI patterns that rotate by $\pi/2$ upon reversing the bias polarity. They interpret this rotation as "smoking gun" evidence for altermagnetic order.

The concept of exploiting topological edge modes as a metrological tool for spin sensing is interesting, and the target of identifying d-wave altermagnetism is high-impact in current condensed matter physics. However, similar idea of spin sensor by STM has been validated before (Science 377, 1218–1222, 2022), thus reducing its originality. Further, for a claim of this magnitude, the experimental evidence for the altermagnetism must be unambiguous. Upon close inspection, I find the execution and data analysis insufficient to support the high claims made in the manuscript. The data is overselling and the distinction between intrinsic topological spin-filtering and extrinsic artifacts (such as orbital effects or tunneling matrix anisotropy) has not been robustly established. Consequently, I am not convinced this constitutes the "experimental milestone" claimed in the abstract and cannot recommend publication in Nature.

Below are my detailed comments:

1. The verification of the tip's spin-polarization tunneling on Fe_{1.02}Te raises several questions regarding the fidelity of the probe. In Figure 2h, both spin-up and spin-down channels exhibit local peaks or shoulders. This complexity contrasts with the idealized schematic in Figure 1e, which implies a cleaner selection rule. Why do these specific features appear, and do they persist over a broader energy range?
2. Since the contrast reversal is demonstrated only at selected voltages. To be convincing, the authors should show that this pattern persists or evolves consistently over a wider bias window.
3. While the tip is non-magnetic, the sample is. How does the system respond to reversed external magnetic fields? A robust characterization of the helical nature requires more comprehensive controls than what is currently presented.
4. Given intrinsic altermagnetism of KV₂Se₂O, why this result depends on the $\sqrt{2} \times \sqrt{2}$ surface? What about the 2×1 surface?
5. The central claim of d-wave altermagnetism rests on the QPI analysis in Figure 5, specifically the rotation of the scattering intensity anisotropy when switching from positive to negative bias. However, a visual inspection of the FTs reveals that the anisotropy is extremely subtle. The peaks in the line profiles are barely distinguishable from the background noise. Also, the filtered modulations are not surprising when choosing different FT spots. Consequently, the claimed anisotropy appears to rely heavily on aggressive data processing. The filtered modulations are susceptible to selection bias depending on the specific integration windows chosen in reciprocal space.
6. I note that the intensities of the $\sqrt{2}a$ spots are very different between the +10 and -15 meV datasets. If the tunneling condition is stable, these structural peaks should be comparable. This large variation further deepens my skepticism about the results.
7. Even for the magnetic contrast map in Figure 5j,k, The mathematical procedure is highly susceptible to artifacts, particularly if the tip apex has any geometric asymmetry (which is guaranteed in a FIB-fabricated tip). Since the orbital asymmetries in the tip apex are inevitable, which is expected for a tip made of a low-symmetry crystal like Bi₄Br₄, the tip's orbital transmission coefficients will be anisotropic. Reversing the bias polarity probes tunneling into versus out of these anisotropic tip orbitals. This change in tunneling matrix elements can naturally mimic a rotation in the QPI pattern, even in the absence of magnetic scattering. The authors have not provided a robust method to distinguish genuine spin-flip scattering from these inevitable orbital-dependent tunneling effects.

8. Since there are two types of Bi₄Br₄ tips: one with band gap and the other one with helical-edge-mode. The authors should compare these two results on the same KV₂Se₂O surfaces and defects. A side-by-side QPI comparison is definitely essential.
9. The analysis of Zeeman shifts in Figure 4 relies heavily on arrow-shaped defects of V-site vacancies. However, the tunneling spectra exhibit many unidentified in-gap peaks. The authors appear to select only the specific peaks that satisfy the expected Zeeman splitting relations while ignoring others. Such an arbitrary data selection process is scientifically unsound and unacceptable.
10. Since the surface is rich in Vanadium, it is a common occurrence in STM for the tip to pick up a V atom or cluster during scanning or pulsing. This would instantly create a standard, albeit weak, spin-polarized STM tip. The authors have not effectively ruled out the possibility that their helical contrast is simply conventional spin-polarized tunneling from a V-contaminated apex. Given the similar effective spin polarization (~12%) as previously reports by spin-polarized STM measurements, accidental functionalization seems a plausible alternative explanation.
11. The interpretation that the arrow-shaped defects reveal the intrinsic magnetic ground state is debatable. It is equally plausible that the electronic structure in the vicinity of the vacancy is dominated by the strong impurity potential itself. This could induce a local symmetry breaking (local magnetic moment formation or orbital ordering) that does not necessarily reflect the long-range altermagnetic order of the pristine crystal. Distinguishing between defect-induced physics and bulk altermagnetism in such a disordered landscape requires more rigorous validation.
12. The effective polarization ratio shown in Figure 5l exhibits a significant asymmetry between positive and negative bias for a single tip. This is physically inconsistent with the authors' model. Altermagnetism is characterized by spin splitting that typically spans a large energy range (often on the eV scale). Consequently, the spin-dependent contrast should be robust and relatively constant over a wide bias window. However, the contrast reversal reported here appears highly sensitive to the specific bias voltage selected. This strong energy dependence suggests the contrast arises from energy-dependent orbital artifacts or density-of-states features rather than the broad-band spin-momentum locking of a helical edge mode.
13. The organization of Figure 5 is chaotic. The current arrangement makes it extremely difficult for the reader to follow up the logical progression.

Referee #3

(Remarks to the Author)

Summary of the key results

The manuscript "Atomic-scale spin sensing of a 2D d-wave altermagnet via helical tunneling" by Wang and co-workers describes spin-resolved scanning tunneling microscopy (STM) and spectroscopy (STS) measurements as well as quasiparticle interference (QPI) data obtained with conventional PtIr and novel Bi₄Br₄ tips on cleavage planes of KV₂Se₂O. After an introductory paragraph, the manuscript discusses two major results: (a) the utilization of edge states of the higher-order topological insulator (HOTI) Bi₄Br₄ for spin-resolved measurements and (b) the investigation of KV₂Se₂O in real and reciprocal space.

In (a) the successful application is convincingly demonstrated by "benchmarking" experiments on the well known antiferromagnet FeTe. The observation of a characteristic striped pattern caused by spin-polarized tunneling is beyond doubt, even though a few details regarding the presentation of the data might be improved (see list of comments below). In contrary, as pointed out below I am not fully convinced of the data presented in the context of part (b).

Originality and significance: if not novel, please include reference

As stated above, I largely convinced by the data presented in part (a) for Bi₄Br₄-functionalized tips. However, a very similar experimental concept has already been demonstrated by Aishwarya et al. in Ref. 62. Therefore, I do not understand why the authors decided to include part (a) in the main manuscript rather than describing it in either the extended data or the supplemental material.

Data & methodology: validity of approach, quality of data, quality of presentation

Overall, the data are of very high quality and the general approach of investigating the spin state of a potential layered d-wave altermagnet with a Bi₄Br₄-functionalized tip is definitely worth to be pursued. However, I do have some concerns regarding some details of the data analysis and the conclusions drawn (see below). It is quite possible that the authors will be able to refute the criticism expressed with appropriate arguments.

Appropriate use of statistics and treatment of uncertainties

The manuscript doesn't describe or discuss statistics methods or uncertainties. I don't consider this a weakness since a systematic treatment of uncertainties in STM-related experiments is very unusual.

Conclusions: robustness, validity, reliability

My main criticism concerns the presentation and interpretation of the data shown in Fig. 3 (see below). Fig. 1 is used to introduce the topic. Fig. 2 convincingly shows that Bi₄Br₄ tips are capable of mapping the spin structure on the atomic scale. Figs. 4 and 5

Suggested improvements: experiments, data for possible revision

See below.

References: appropriate credit to previous work?

See above my criticism regarding Ref. 62.

Clarity and context: lucidity of abstract/summary, appropriateness of abstract, introduction and conclusions
Good.

List of comments (minor and major):

- page 2, line 45: add appropriate reference after "... and the second one utilizes nonrelativistic Zeeman splitting as in ferromagnets with broken time-reversal symmetry."
- page 2, line 73: add appropriate reference after "electrons injected into or launched from a HOTI Bi₄Br₄ tip carry opposite spins enforced by their helical nature"
- Overall, the tone related to the experiments performed with Bi₄Br₄ functionalized tips is a bit too enthusiastic, since similar experiments which also involve spin-momentum-locked helical states have already been performed in Ref. 62. I suggest to us a more moderate wording which appreciates the merits of earlier work. The authors shall also consider to move this part (although definitely impressive) to the extended data section or even the supplemental material.
- page 3, lines 110-113: "Owing to the weak interlayer coupling, α -Bi₄Br₄ also belongs to the family of 3D QSHIs, which hold helical edge modes encircling each layer despite a tiny hybridization gap (see Methods; 60, 61). These multiple helical edge modes give rise to large electron tunneling channels." To my opinion, it remains unclear why multiple layers shall contribute to the tunneling process in a local probe experiment with an exponentially decaying tunneling probability.
- Fig. 2h: Data are very noisy and it appears questionable whether the opposite corrugation suggested by the line sections in the lower panel is an overall observation (which it should be). Therefore, instead of presenting selected data only, the authors shall present the sum and the difference of the dI/dV images taken at +50mV (left) and -50mV (right).
- page 4, lines 156-157: In the sentence "this gap becomes narrower somewhere and eventually collapses with the emergence of subgap states" the meaning of the word "somewhere" remains unclear.
- Fig. 3: To my opinion the difference between the Vanadium sites caused by the (2x2) reconstruction of the 50% filled K layer, such as V_{ya} and V_{yb}, is not introduced and explained well in the manuscript. The sketches presented in the lower left of Fig. 3a with 50% encircled K atoms and Fig. 3g with large and small red and blue balls representing V_{ya}, V_{yb} and V_{xa}, V_{xb} is not really obvious. Here some (graphical and didactic) adjustments might help.
- page 5, line 170-171: Related to the previous comment. You write "This explicit arrangement doubles the original unit cell to a new $\sqrt{2} \times \sqrt{2}$ supercell (Fig. 3g)". It would be helpful to also show the unit cell as it appears without lifting degeneracy of a and b type sites.
- Fig. 3 b and c: To evaluate any potential correlation between the local structural properties and the gap width, the topography of the same location shown in Fig. 3c shall be presented as well. The reference to Fig. S6 is not helpful as this data set doesn't show the corresponding map of the local gap width.
- page 5, lines 171-178: It remains unclear which data and analysis led the authors to conclude that "this supercell represents an intrinsic modulation of the V lattice but is not a simple consequence of the surface K reconstruction". The authors refer to the Extended Data Fig. 5 and claim the observation that "the spatial correlation is long ranged regardless of the surface domains and appears even on the 2×1 reconstructed surfaces", but I was unable to verify these claims based on the given material, i.e. the extended manuscript in pdf or docx format. (In either case the resolution was too poor! This may not be caused by the data themselves but may be caused by the figure production process.) Therefore, the attribution of the observed supercell to a density wave remains somewhat unsupported by appropriate data.
- page 5, lines 183-185: The authors state that "each (scattering vector) consists of multiple parallel stripes in q-space that reflect the morphology and dispersion of the constant-energy contour (CEC) in k-space (Fig. 4b)." I cannot identify these "multiple parallel stripes in q-space". The authors shall specify what they refer to.
- Fig. 4d: Why do the experimental data show two branches, whereas DFT predicts one linearly dispersing state only?
- Fig. 5 and related text: Why do the authors (somewhat indirectly) conclude the antiferromagnetic order from an opposite magnetic field-induced shift of the spectra shown Fig. 5g and h rather than mapping the antiferromagnetic order directly?

Version 1:

Reviewer comments:

Referee #1

(Remarks to the Author)

Zhuying Wang et al. have provided considerable additional characterization of their Bi₄Br₄ STM tips. Together with additional data on KV₂Se₂O, this improves the robustness of the experimental conclusions.

To my understanding one central logical step still remains unresolved: The spin polarization of topological surface states is mostly orthogonal to the local moments of the alternating magnetic order. The authors state that "Our experiments suggest that the Bi₄Br₄ tip is more sensitive to magnetic structures with in-plane spin orientations". The authors note further that "Our calculations indicate that the edge state electrons also mainly possess an out-of-plane spin orientation ($S_z \sim \pm 0.85 \times \hbar/2$; see Fig. R1-1). In the geometry of the STM measurement, this spin orientation lies "in-plane" with respect to the target sample".

In response to my original comment that a tip spin polarization at a 90° angle to the substrate would remove any spin sensitivity, the authors mention "an unavoidable geometric tilt (approximately 10°) of the tip relative to the surface normal in our experimental setup." as an explanation for an out-of-plane spin contrast.

Qualitatively, this can be true, but an angle of 10° is rather small. To my understanding this would imply for simple spin-conserving tunneling that the spin contrast for out-of-plane moments is proportional to $\cos(80^\circ) \sim 0.17$. The effective spin resolution is reported as 12%, and thus it seems that the tip channel selectivity would need to be ~ 0.7 even if there are no

other losses of spin selectivity. Here, a value of 1 would be a complete selection of a single direction of spin-polarized helical channel.

Conversely, a deviation from the expected out-of-plane altermagnetic order, i.e., an in-plane moment on the sample, would be detected with $\cos(10^\circ) \sim 0.98$, so even a modest channel selectivity of ~ 0.12 would lead to 12% of effective spin signal. If in the vicinity of the measured defects and the QPI there is any deviation from the z-aligned order, the Bi₄Br₄ tip would be sensitive to it.

I think it needs to be explicitly discussed whether deviations from the z-aligned altermagnetic order can be excluded. Maybe an orientation dependence of the external-field-induced shift may resolve this question. Alternatively, also simulations of the defects themselves could exclude local in-plane polarizations.

While I think the use of a topological STM tip is a fascinating development, I believe the manuscript has to clarify why the experiments are not interpreted in terms of in-plane polarizations, to which the setup seems to be more sensitive.

Additional comments:

1. To argue for the absence of orbital effects, the authors argue in terms of a p_y orbital that sticks out of a straight termination of Bi₄Br₄. They conclude "Because this extending orbital is nearly normal to the surface, it maintains an isotropic tunneling matrix element with respect to the in-plane dxz and dyz orbitals of the sample". There are three issues:

- The theoretical modeling considers a defect-free rectangular lattice, yet to my understanding a point-like tip apex is such a defect, for which in principle the orbital configuration may change.
- Stating that the "orbital is nearly normal to the surface" is not an obvious way to exclude orbital effects, when the spin resolution derives from just this 10° deviation from the normal alignment. Although the orbital orientation is not identical to the spin-selective tunneling axis, the manuscript should explain why the same 10° tip tilt is important for spin contrast but negligible for orbital-selection effects.
- The authors state that "the anisotropy stemming from purely structural effects does not rotate when switching the bias polarity", while this seems true, what about the orbital character? Generally, topological phases exhibit a band inversion in energy between bands of different symmetry. It is not obvious to me whether this can be excluded as source of anisotropy.

2. The FeGe measurement exhibits uniform color shifts on both sides of the steps when comparing ± 1 T and ± 8 T, respectively. What do they imply about how the external field affects the topological states on the tip?

3. Why does for FeSn only the middle layer appear to be strongly polarized in the contrast between positive and negative bias?

4. To argue for spin sensitivity, the authors consider possible causes for why there is an asymmetry in how well the active part of the tip connects to the edge states to achieve spin sensitivity:

- a band-bending effect, which seems reasonable in principle.
- a non-linear voltage response of the AFM surface, which I do not understand from the given discussion. The connection between a non-linear charge current response of the tip and the spin texture is unclear. The authors mention a previous study but do not reference it at this point.

5. The authors mention an "effective k_z component" but they labeled only k_b in their figure. Also, they show a vector decomposition of tip tangents in figure S29 which does not seem connected to the provided explanation of tunneling probability.

Referee #2

(Remarks to the Author)
see my attachment

Referee #3

(Remarks to the Author)

I read the revised manuscript and the very (with 38 manuscript pages and 61 letter pages maybe even too) elaborate reply the authors wrote as a rebuttal to the comments of the three referees. I definitely appreciate that the authors put a lot of work in order to better explain and discuss their scientific results, even by performing further experiments on a wide variety of what they consider magnetic reference materials. The vast majority of answers to the questions of all three referees are —to my opinion— fully convincing.

Yet, there are two main issues which led me to speak out in favor of rejecting the revised version of the manuscript. These issues are:

(1) I still don't agree with the authors that the part on SP-STM using helical edge states shall be part of the main part of this manuscript. To my opinion, a strikingly similar concept has been used in Ref. 50 (Science 377, 1218 (2022)). The

modification of this approach used in the manuscript under discussion here does not justify publication in a highly ranked journal like Nature. I read with interest that a similar statement was made by referee 2 in his/her introductory sentences: "However, similar idea of spin sensor by STM has been validated before (Science 377, 1218–1222, 2022), thus reducing its originality."

(2) In my first review I asked the question, why the authors not simply image the AFM spin structure rather than concluding it indirectly from an "opposite magnetic field-induced shift of the spectra shown Fig. 5g and h". The authors respond by claiming an experimental difficulty, i.e. "the Neel vector (QAFM) coincides exactly with either the atomic Bragg modulation or the second-harmonic frequency of the surface structural reconstruction. Consequently, the magnetic signals in q-space are spectrally overlapped with structural contributions. Given that both signals can exhibit complex and potentially energy-dependent changes, it is difficult to unambiguously disentangle the intrinsic magnetic contrast from structural artifacts using real-space mapping techniques."

I have to admit that this explanation doesn't resonate with me. Magnetism induced modifications of the total current are usually verified by inverting the magnetization direction of the magnetic tip, as convincingly shown in Fig. R3-7. For their special SP-STM method with helical edge states this would respond to a comparison of data acquired at small positive and negative bias (around +/- 30 to 50mV).

Overall, my comment is very similar to the criticism #11 raised by referee 2 in the first round of review. Here, he/she wrote the introductory sentence: "The interpretation that the arrow-shaped defects reveal the intrinsic magnetic ground state is debatable." The comment I made in the preceding paragraph touches exactly the same point, but approaches it from a more technical perspective.

There are further, probably minor points which should be clarified before the manuscript is published:

(a) "The extracted energy dispersion well matches the theoretical calculations with in-plane Néel AFM order (Fig. 4d; Extended Data Fig. 8)".

It remains unclear for which band and at which point in the BZ the claimed good agreement is obtained

(b) How does the calculated band structure presented in Fig. S20 correspond to the band structure plotted in Fig. S28? I cannot see how the features along M—Gamma—M of Fig. S20 derive from S28.

(c) I explicitly thank the authors for their figure R3-11 which shows a much smaller (structural) signal in the added signal that in the larger (spin) difference signal. Yet, magnetic contrasts still appear to be masked by additional contributions, which may originate from crystalline imperfections or other effect, such as those discussed by referee B. One particular apparent example are the line sections presented in Fig. R2-1e for +30mV and -30mV. While the modulation with a 2a periodicity is nicely visible at +30mV, the data obtained at -30mV show a strongly distorted signal at distances 3-7nm (x axis). In summary it appears that numerous Bi4Br4 tips are capable of detecting a spin-related signal, as convincingly shown in Fig. R4-1, but it remains questionable if other parameters, such as the orbital character of the tip and sample states which potentially complicate data interpretation substantially, also play an important role.

In summary, I cannot support publication in Nature. I suggest scaling back the scientific scope somewhat, specifically shifting from the direct (atomic-scale) observation of the antiferromagnetic structure of an antiferromagnet to the observation of spin order in the vicinity of structural defects.

Version 2:

Reviewer comments:

Referee #1

(Remarks to the Author)

I would like to thank the authors for extensively considering my comments and requests. My main concern of the previous round aimed at characterizing which component of spin is mainly measured, in-plane or out-of-plane. The authors now provide field-dependent measurements in Fig. R1. Indeed, the data support the measurement of a predominantly out-of-plane magnetic response, which exhibits opposite signs of the field-induced shifts for defect-associated states extending along x and y. This strengthens the interpretation that the studied features are indeed mainly spin-polarized as claimed. Quantitatively, I am not convinced because, as the FeGe study with spin-flop contrast showed, the spin sensitivity is much stronger for an in-plane polarization. The authors state „applying an identical field magnitude within the in-plane (B_x and B_y) yields no resolvable Zeeman shift whatsoever.“, but in Fig. R1d the X2 feature does seem to shift for applied B_y . This would need fits to make a more precise statement.

When I manually read out the shift, it appears to be 0.2 of the response to the B_z -aligned field. If this approximate ratio of field-induced shifts reflects the corresponding sample spin-polarization components, the stated tip geometry would make their contributions to the tunnelling contrast comparable: 0.2 is similar to the geometric factor $\cos(80^\circ) \sim 0.17$ that reduces the out-of-plane component. The data therefore do not yet establish that an in-plane contribution is negligible in the measured tunnelling contrast. This requires a quantitative bound on the in-plane magnetic response and an explanation of how this bound applies to the states contributing to the q_x and q_y QPI features.

I am not sure I fully understand their quantitative argument. One reading would be that there is a $\cos(80^\circ) \sim 0.17$ geometric factor with a 0.85 tip S_z polarization and additionally a $\cos(10^\circ) \sim 0.98$ geometric factor with roughly a 0.15 tip in-plane spin

polarization. The total is ~ 0.29 , which would match their 20-30%. However, this assumes that the lack of S_z tip polarization translates into an in-plane polarization available to enhance the spin contrast, which is generally not true because the total spin polarization does not need to vectorially add up to a quantized value.

Using Sr_2RuO_4 , whose Fermi-surface segments have pronounced orbital character, the authors find a d-wave QPI anisotropy with a magnitude of at most approximately 3%, compared with 10–18% in $\text{KV}_2\text{Se}_2\text{O}$. The authors provide in figure R2a a suggestive nesting that highlights the similarity to $\text{KV}_2\text{Se}_2\text{O}$. Since the warping of Sr_2RuO_4 appears smaller except from the hybridization points, one could also consider a continuum of scattering vectors parallel to q_x but connecting along blue Fermi surface instead of red ones. This might imply that the QPI receives contributions from more than one orbital. The authors do not seem to provide a QPI calculation to show its orbital character. Such a calculation would establish the orbital contrast between the compared QPI features and help assess the tip's orbital sensitivity.

To get an impression, what is known on Sr_2RuO_4 , one may consider [Fig. 5, Kreisel et al., <https://doi.org/10.1038/s41535-021-00401-x>], which reports on the orbital-textured QPI based on a tight-binding model with surface reconstruction.

However, the calculated arcs do not appear to reproduce the pronounced axial intensity maxima seen in the rebuttal. In [Fig. 7, Bhattacharyya et al., <https://doi.org/10.1103/PhysRevB.107.144505>] a central intensity peak at q_x in the dx_z QPI feature has also a contribution from dy_z . Of course, these references are at different modelling conditions and can only give a qualitative idea. So, the orbital contrast used to extract the rebuttal's anisotropy remains unquantified. From my visual inspection of the outer regions of the arcs no clear anisotropy is visible.

Thus, even though the orbital texture of QPI is not quantified and might be not fully comparable to $\text{KV}_2\text{Se}_2\text{O}$, I agree that Sr_2RuO_4 provides meaningful evidence against a generic dominant orbital sensitivity of Bi_4Br_4 tips.

In conclusion, an in-plane polarization of the QPI signal is potentially picked up with a signal strength comparable to that of the out-of-plane component. As the defect-associated resonances are indeed responsive to B_z and the orbital selectivity has been established as less likely, I would regard some sensitivity to the out-of-plane spin component with a transparent discussion as sufficient. But as of now a "direct experimental evidence" claim applies mostly to the d-wave pattern and not to the proposed out-of-plane spin texture.

Referee #2

(Remarks to the Author)

The authors have addressed the major points raised in my previous report. In particular, most of my concerns regarding the experimental observations, data interpretation, and the underlying physical picture have now been adequately mentioned. I note that some aspects of the authors' interpretation may still admit alternative views, and I do not necessarily agree with all of the arguments presented in the response and the revised manuscript. In my opinion, however, these remaining issues mainly concern the interpretation of the results rather than the validity of the experimental observations themselves.

Although there have appeared several similar studies after this work, however, in my view, these subsequent reports do not provide evidence that is as comprehensive and convincing as that presented in the present manuscript. Nevertheless, I consider such differences in interpretation to be scientifically constructive rather than detrimental to the manuscript. The results presented here may stimulate further discussion and experimental and theoretical efforts in this field.

Overall, I believe that the authors have satisfactorily addressed my major concerns, and that the remaining issues do not preclude publication.

Referee #3

(Remarks to the Author)

In order to come to an appropriate advice how to deal with the manuscript "Atomic-scale spin sensing of a 2D d-wave altermagnet via helical tunneling" by Wang and co-workers I read their detailed rebuttal letter to the three referee reports. I appreciate the enormous amount of data and extra analysis the authors invested to deal with the criticism raised by the referees. Overall, I come to the conclusion that the amount of data and the insight provided by the data now justifies publication in Nature since it will —to my opinion— have an impact on the scientific debate and very likely trigger future studies in experiment and theory. Even if some of the interpretations offered in the manuscript might be revised based on future results, the breadth and quality of the data justifies their publication in a highly visible journal.

One side remark: To my opinion, large parts of what is written in the paragraph entitled „Functionalization of the tip with helical edge modes“ should be moved to the Methods section, the Extended Data or the Supplementary Material. But I leave the decision to the editorial team which will certainly safeguard Nature's guidelines.

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We express our deepest gratitude to the referees for their thorough examination of our work and for providing us with constructive comments. We have incorporated their valuable suggestions and made significant improvements to our manuscript. We address individual comments point-by-point below (all major changes to the main text and Supplementary Information are left in blue font).

- [Response to Referee #1: Pages 1-16;](#)
- [Response to Referee #2: Pages 17-42;](#)
- [Response to Referee #3: Pages 43-61;](#)

Response to Referee #1

Zhuying Wang et al. present in their manuscript “Atomic-scale spin sensing of a 2D d-wave altermagnet via helical tunneling” two interesting advances: They create an STM tip from the topological insulator α -Bi₄Br₄, and use it to resolve the spin splitting in the altermagnet candidate KV₂Se₂O in real space and momentum space. As this extensive and detailed work combines topological systems with unconventional magnetism, a new method and timely physics, I believe it has potential to be of interest to several communities.

As there are already spin-resolved ARPES measurements on the compound (Ref. 40), the novelty for the material is rather the STM real space resolution of the magnetic order, which has been convincingly achieved in Fig. 4 of the present manuscript using magnetic fields. The authors chose an altermagnetic candidate compound, where due to an extra transition within the magnetic phase, surface reconstruction, and non-stoichiometry the magnetic and crystalline symmetries are subject to uncertainty. Given such complications their result is important.

Reply: We sincerely thank the referee for endorsing the significance of our work, and his/her constructive suggestions which are really helpful to improve our manuscript.

To make the manuscript suitable for Nature, I believe the method claim has to stand beyond doubt. My central questions concern the interpretation of the topological tip and the size of spin resolution:

Reply: We fully agree with the referee that methodology employed here (utilizing STM tips with helical edge states for spin-resolved measurements) is still in its early stages, and that particular caution is warranted before drawing definitive conclusions.

Since the previous submission, we have fabricated more than 30 Bi₄Br₄ tips, characterized four distinct classes of compounds with different magnetic structures, conducted three types of control experiments (under applied magnetic fields, rotation of the sample, and using a conventional magnetic tip), and performed calculations of the orbital and spin characters of the Bi₄Br₄ edge state. These efforts have yielded a more detailed and comprehensive understanding of the spin-polarization capability of this unconventional “topological” tip. Our experiments suggest that the Bi₄Br₄ tip is more sensitive to magnetic structures with in-plane spin orientations, and further validate its distinct characteristics relative to a conventional magnetic tip. In light of these findings, we first provide a summary before addressing the individual points raised.

The spin orientation of edge-state electrons from theoretical perspective. α -Bi₄Br₄ exhibits a bulk spin polarization along the S_z direction (nearly quantized as $\pm 0.97 \times \hbar/2$). Our calculations indicate that the edge state electrons also mainly possess an out-of-plane spin orientation ($S_z \sim \pm 0.85 \times \hbar/2$; see Fig. R1-1). In the geometry of the STM measurement, *this spin orientation lies “in-plane” with respect to the target sample.*

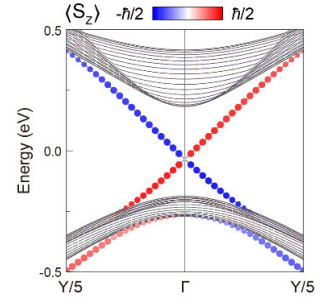


Fig.R1-1 S_z as the major spin orientation for the edge states in Bi₄Br₄

To gain a more comprehensive understanding of its spin-polarization capability, *we have measured four distinct classes of compounds with different magnetic structures:* Fe_{1+x}Te, kagome magnet FeSn, FeGe and finally KV₂Se₂O. *We also performed three types of control experiments* (under magnetic fields, rotated the sample and using conventional magnetic tips and PtIr tips). **This systematic study demonstrates that the spin polarization of the Bi₄Br₄ tip is intrinsic and more sensitive to in-plane spin components, and further verifies its distinct characteristics relative to a conventional magnetic tip.** In the following, we describe several key experimental findings in detail.

1. Fe_{1+x}Te with bicollinear AFM order.

- **A contrast reversal at opposite bias voltages using Bi₄Br₄ tips.** We confirm that the spin-polarized local DOS (with the bicollinear AFM order) *undergoes a contrast reversal at opposite bias voltages using Bi₄Br₄ tips*, (the additional kink at negative biases corresponds to atomic modulations, as shown later in Reply 1-6), which can be observed in *the dI/dV maps and our newly acquired topographic images for a large energy range* (Fig. R1-2).

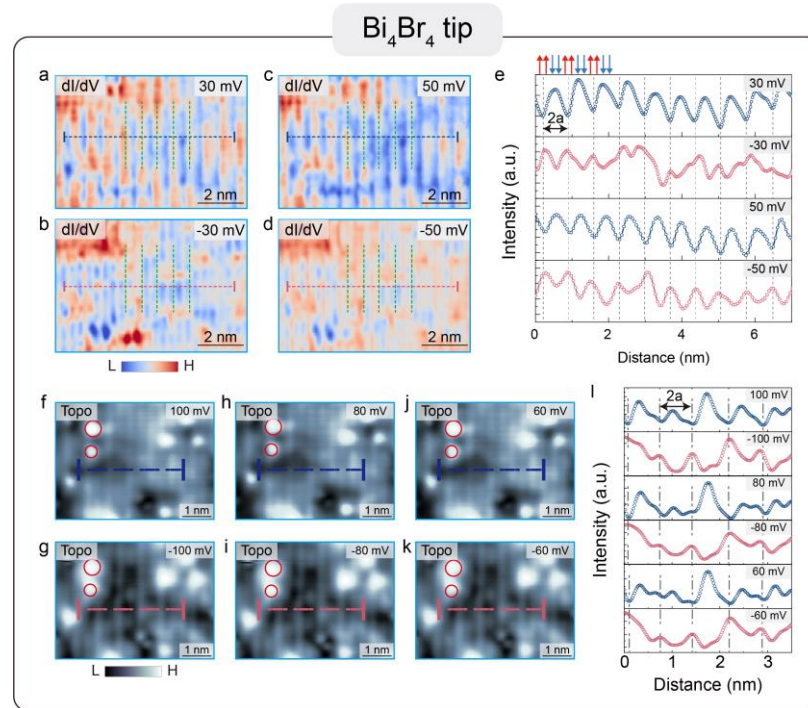
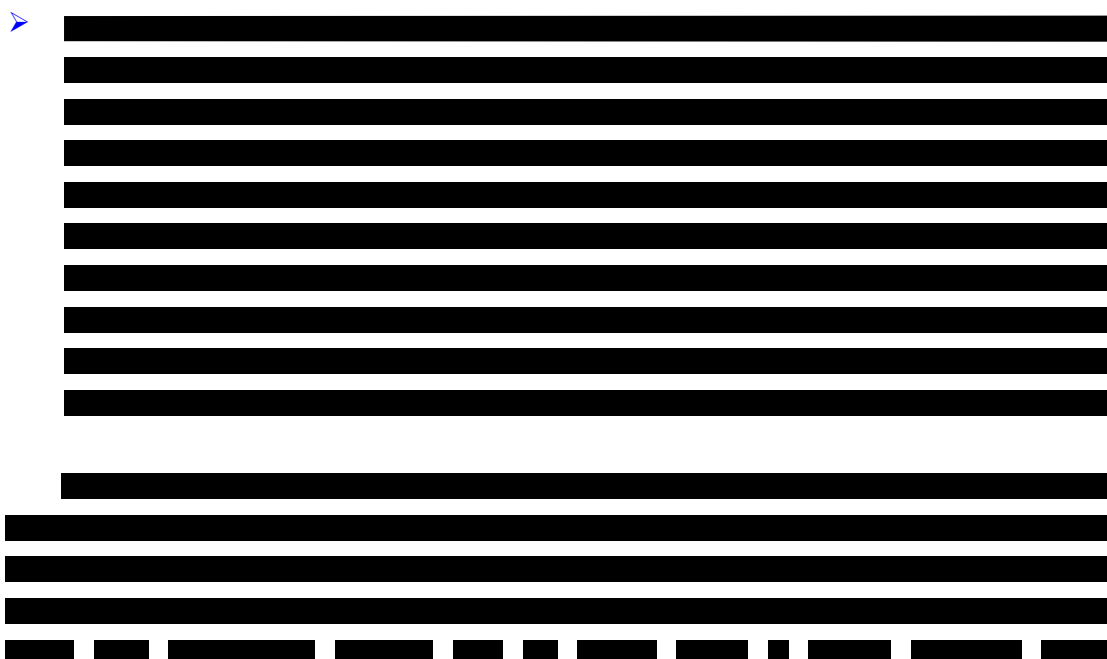


Fig. R1-2 Bicollinear AFM order in Fe_{1+x}Te imaged by the Bi₄Br₄ tips. **a-d**, dI/dV maps and **f-k**, topographic images. In both cases, the spin-polarized local DOS of 2a modulation exhibits a π -phase shift (contrast reversal) at opposite bias voltages, as shown in **e** and **i**. To show the overall tendency, the linecuts in **e** and **i** are averaged over 30 and 20 pixels perpendicular to the linecut direction, respectively.

- ### Fe-coated PtIr tip
-
- Figure 1 displays differential conductance (dI/dV) and topographic (Topo) images of a Fe-coated PtIr tip on a Si(111) surface. The images are arranged in a grid, showing the spatial distribution of dI/dV and Topo signals at various bias voltages (100 mV, 50 mV, 30 mV, -100 mV, -50 mV, -30 mV). The color scale for dI/dV ranges from Low (L) to High (H), and the grayscale scale for Topo ranges from Low (L) to High (H). The images show the characteristic oscillatory behavior of the dI/dV signal, which is used to identify the atomic structure of the tip and the surface. The Topo images show the surface topography, which is used to identify the atomic structure of the surface. The line profiles (Intensity vs. Distance) are shown for each dI/dV map, with a scale bar of 2a indicated for the 100 mV profile.

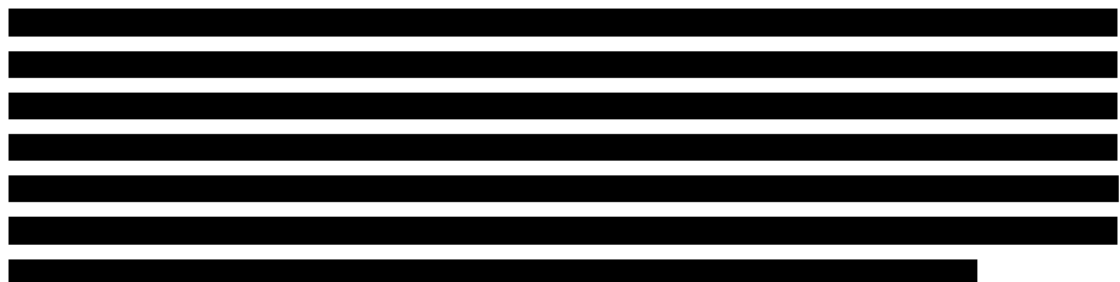
Fig. R1-3 Bicollinear AFM order in Fe_{1-x}Te imaged by the Fe-coated PtIr tips. **a-f**, dI/dV maps and **h-m**, topographic images. In both cases, the spin-polarized local DOS of $2a$ modulation exhibit a same phase (without contrast reversal) at opposite bias voltages. To show the overall tendency, the linecuts in **g** and **n** are averaged over 30 pixels perpendicular to the linecut direction, and no contrast reversal is observed.

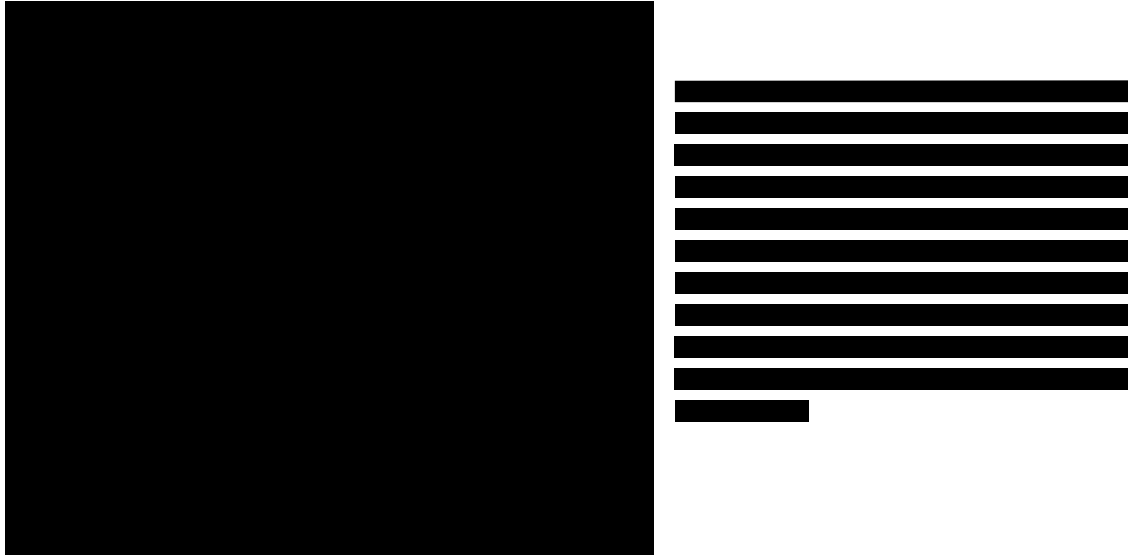




These experiments in Fe_{1+x}Te demonstrate **the intrinsic difference between Bi_4Br_4 tips and magnetic tips**, suggests that the orbital feature of Fe_{1+x}Te is unlikely to be the origin for the contrast reversal at opposite bias voltages seen by Bi_4Br_4 tips, and further suggest that the spin orientation on the Bi_4Br_4 tips more likely lies in one “in-plane” direction with respect to the target sample.

2. Kagome metal FeSn with A-type AFM order. For an A-type AFM order, spins are ferromagnetically aligned within one layer but antiferromagnetically coupled between adjacent layers. *This magnetic configuration provides an ideal platform to verify the spin resolution of the Bi_4Br_4 tip, as additional contributions from orbital effects and tunneling matrix elements should be largely suppressed between these two layers with same atomic constitutions.*





3. Kagome metal FeGe with A-type AFM order: another key compound

FeGe hosts an A-type AFM order *with canted spins mainly oriented out-of-plane at 0T* (Fig. R1-6a; Nature 609, 490 (2022)). With an external out-of-plane magnetic field exceeding 7T, the system undergoes a spin-flop transition, reorienting the spins into in-plane direction (Fig. R1-6b; Nature 609, 490 (2022)). Consequently, magnetic-field-dependent STM measurements are highly valuable for further elucidating the spin resolution of the Bi₄Br₄ tip.

- **Bi₄Br₄ tips are more sensitive to in-plane spin components.** Figure R1-6c shows an STM topographic image of a cleaved FeGe surface featuring a single-unit-cell step edge. The two sides of the edge are thus identical (except spin). Under zero or weak ($\pm 1\text{T}$ or $\pm 2\text{T}$) magnetic fields, the spin orientation of FeGe remains predominantly out-of-plane, whereas the spin polarization of the tip is in-plane (enforced by the large energy scale of SOC in Bi₄Br₄ so that magnetic field of a few Tesla cannot change the spin direction on the tip); therefore, the dI/dV maps exhibit very weak contrast between adjacent layers (Fig. R1-6d). However, *when an external field of $\pm 8\text{ T}$ is applied to induce the spin-flop transition, a pronounced contrast emerges between the two layers* (Fig. R1-6 e, f). This arises because the spins of both the sample and the tip are now aligned in-plane, yielding a finite overlapping component between them. Upon reducing the external field to $\pm 1\text{T}$, this contrast correspondingly diminishes again (Fig. R1-6g-h).

This series of experiments were carried out in the same area with the same STM tip, and the most obvious change is the spin direction on the sample (but with the contribution from orbitals, spin-independent tunneling-matrix elements largely unchanged). Thus, we believe *it provides compelling evidence that the spin polarization of the helical tunneling channels is intrinsic, and the spins on tip are “in-plane” relative to the coordinate system of the target sample.*

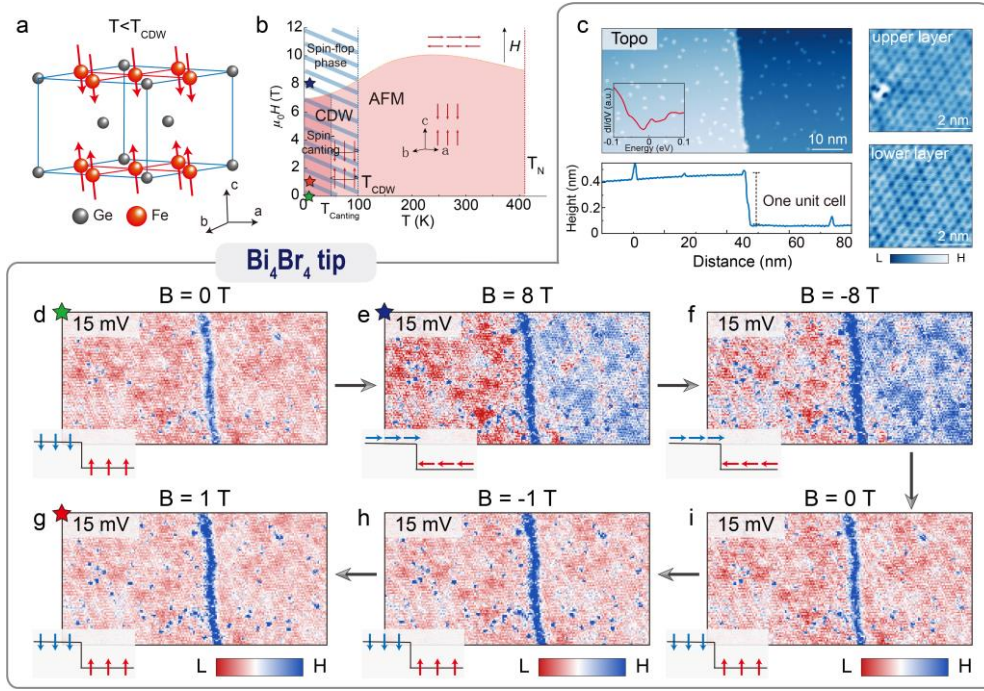


Fig. R1-6 **a**, Magnetic structure of FeGe at 0 T. **b**, Phase diagram of FeGe with field. **c**, STM topography including one single-unit-cell step edge. The right panels show atomic images of the upper and lower layers. **d-i**, dI/dV maps recorded at 15 meV with varying the magnetic field. A large enhancement of the contrast is found at ± 8 T under which the spins flop.

- **The spin resolution behavior of Bi₄Br₄ tip is again different from that of magnetic tip.** For a standard magnetic tip, a small external field of 1 T or 2 T is typically sufficient to fully polarize its apex out-of-plane. Given that the spin orientation of FeGe is also predominantly out-of-plane under these conditions, a distinct spin-polarized contrast should be readily observable at 1 T or 2 T. To experimentally verify this picture, we have prepared Fe-coated tips. The resulting data reveal a pronounced spin contrast at 1 T (or 2 T), as shown in Fig. R1-7. This conventional magnetic-field dependence is fundamentally distinct from the behavior observed with the Bi₄Br₄ tip, further corroborating the non-trivial, in-plane spin resolution of the helical tunneling channels in the Bi₄Br₄ tip.

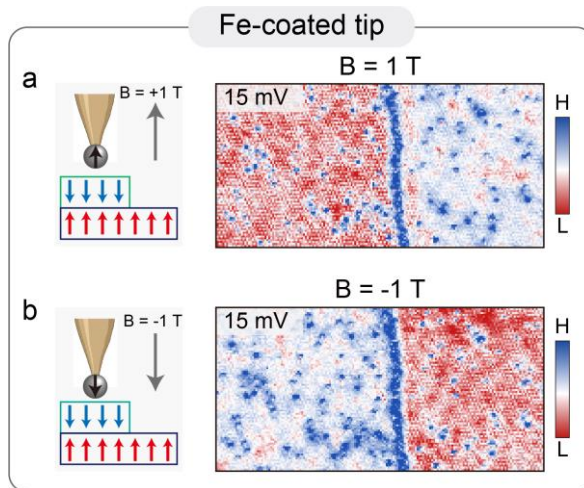


Fig. R1-7. dI/dV maps at 15 meV under magnetic fields of 1 T (**a**) and -1 T (**b**), in the same area but measured with a Fe-coated STM tip. An external field of 1 T is typically sufficient to fully polarize the tip's apex spin to out-of-plane, therefore a clear contrast can be observed. Reversing this smaller field flips the spin direction on the tip but keeps these in the sample, thus the contrast reverses between layers at ± 1 T.

4. KV₂Se₂O with d-wave checkerboard AFM order. The opposite Zeeman shifts of the in-gap states confirm their opposite spin polarization along the x- and y- directions (Fig. 4; we updated these data with much higher quality). This is a direct and well-established way to determine the spin texture of these states. Consequently, the observed $\pi/2$ rotation of the QPI anisotropy upon reversing the bias voltage polarity can be fundamentally attributed to the underlying spin texture of these electronic modulations, and **we further confirmed that such rotation is absent when using three spin-summed PtIr tips** (Reply 1-4).

Summary. By investigating four distinct magnetic compounds, we have consistently obtained spin-resolved STM data that reflect their respective magnetic structure, which can rule out the possibility of coincidental artifacts. We hope the reviewer appreciates the experimental efforts we have dedicated to comprehensively addressing this crucial aspect. Furthermore, these collective data consistently indicate that the spins carried by the helical edge channels are more sensitive to in-plane spins on the target sample. We are confident that this new methodology represents a substantial advancement in the fields of topological materials and STM technique, alongside the physics for altermagnetism.

In the revised version, we have included the typical datasets in Extended Data figures and SI notes: Fe_{1+x}Te (Extended Data Figure 4; Methods; Supplementary Note 3), FeGe (Extended Data Figure 5; Methods) and KV₂Se₂O including the control experiments (Methods; Extended Data Figure 10).

1. Reference 40 showed that spin-resolved ARPES indicates d-wave altermagnetism, albeit only with a low 10% spin polarization, which might be due to the magnetic domain size. Thus, a finer probe like SP-STM should be a good approach to improve the spin-resolution. For the tip material, the supplement of Ref. 60 mentions a bulk spin S_z polarization of as little as 3° deviation, though I failed to find a statement about the spin polarization of the surface states. Compared to these results and the authors' claim of nearly spin-quantized surface states, the present manuscript reports only 12% of effective spin polarization which varies between tips. The authors suggest the tip anisotropy as optimization criterion, which follows the principle shown in Fig. 1c, but at least from extended Fig. 2 this conclusion is not clear, as the four used tips do not seem particularly asymmetric. This makes it unclear whether just more engineering or a physical improvement is necessary to optimize the spin resolution.

Reply 1-1: We hope that we have convinced the referee for the spin-polarization capability of Bi₄Br₄ tip. Here, **there are two fundamental factors contributing to the reduction in the spin-polarization ratio: the spin directions (we will discuss in details in Reply 1-2) and the inherent presence of two counter-propagating edge channels** with opposite spins, which form closed loops circulating along the tip boundaries. 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.

First, the observed spectral line-shape of the Bi₄Br₄ tip on Au (111) surface (Extended Data Fig. 3a–d) 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 “kz” components (which govern the vacuum tunneling matrix elements) are expected to differ between the two adjacent edges (Fig. R1-8). 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.

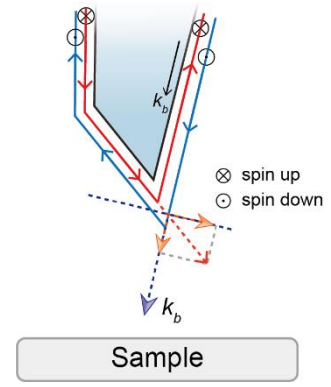


Fig. R1-8. quasi-momentum description of the tunneling process.

We note that the precise microscopic geometry of the tip apex cannot be directly deduced from Figure 2. The structural details that dictate the local tunneling process occur at the nanometer scale, a length scale significantly smaller than the macroscopic shape of the tip resolved in Figure 2.

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. R1-9a). 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.

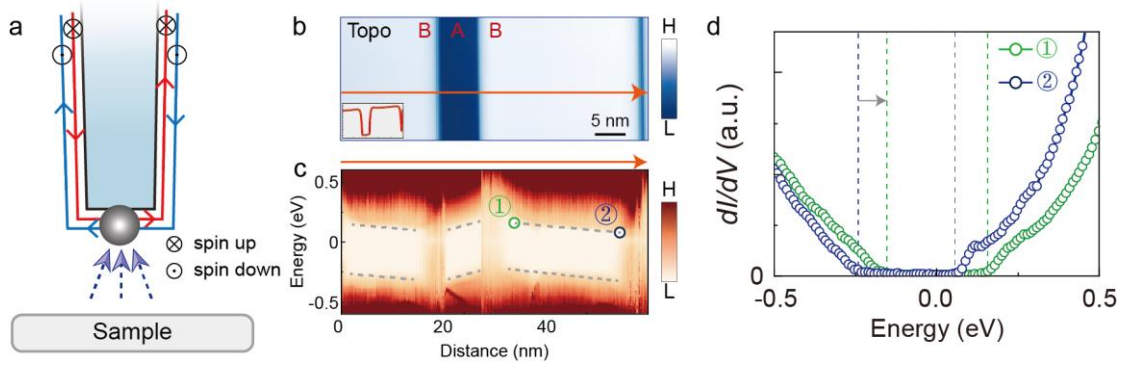


Fig. R1-9 **a**, Sketch for the tunnelling condition with a point-like tip apex. **b**, STM topography including one and a half terraces of monolayer Bi₄Br₄. **c**, Linecut profile of dI/dV spectra across these terraces. A pronounced band-bending effect near the step edges can be observed. Interestingly, 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 Bi₄Br₄ 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. R1-9b-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. Phenomenologically, the relevant dI/dV signal or equivalently the local DOS $\rho(V)$ exhibits a component that is an odd function of the applied bias voltage V . This implies to a second-order current response of the form

$$J = \sigma(N)V^2,$$

where $\sigma(N)$ is the second-order nonlinear conductivity coefficient that depends on the local Neel order N . We require that $\sigma(N) = -\sigma(-N)$. As a result, at two magnetic sites with opposite spin orientations, $\rho(V)$ has different magnitude, or equivalently, $\rho(V) - \rho(-V)$ has opposite signs. This represents the spatial resolution of the local spin order. *Such second-order current can be viewed as a ballistic version of the nonlinear Hall effect.* Based on previous study on the nonlinear Hall effect, the intrinsic contribution from the quantum metric dipole can indeed couple to and capture the antiferromagnetic spin order. We note that extrinsic contributions such as skew scattering and side jump can also contribute.

We sincerely appreciate the referee for raising this insightful point. While a comprehensive microscopic theory elucidating precisely how helical electrons transmit through a vacuum barrier remains an open fundamental question, we believe our present work firmly establishes the crucial experimental foundation. We anticipate that these detailed experimental observations will stimulate broad interest within the theoretical community to further investigate this novel phenomenon.

In the updated version, we have added a brief discussion in the main text (discussion section; page 8) and included this detailed discussion in Supplementary Note 10.

2. Connected to the question of spin polarization: As the authors point out that “... the spin orientation for the edge modes is ‘horizontal’ (Fig. 1c), whereas that for the AFM order in KV2Se2O is out-of-plane”. If the near perfect “Sz” polarization of the tip is 90° rotated to the substrate, then this seems to mean to me that there should be no spin selectivity as an $S_z > 0$ state of the substrate should hop with equal probability to, for example, $S_x > 0$ and $S_x < 0$ of the tip. This has to be cleared up before the main claim becomes believable.

Reply 1-2: The supplementary control experiments on Fe_{1+x}Te (Figure R1-4) and FeGe (Figure R1-6) strongly indicate that the Bi_4Br_4 tip is more sensitive to the in-plane spin configuration of the substrate. Nevertheless, the tip retains the capability to resolve the out-of-plane spin contrast of DOS. This arises from an unavoidable geometric tilt (approximately 10°) of the tip relative to the surface normal in our experimental setup.

As shown in Fig. R1-10b, during the FIB process, the PtIr wire supporting the Bi₄Br₄ tip must be bent by 90° to accommodate the holder geometry within the FIB-SEM system. After the FIB processing, we bent the PtIr wire back with tweezers to make it vertical to fit the STM tip holder. However, due to the mechanical rigidity and short length of the wire, a residual macroscopic tilt angle of approximately 10° is practically inevitable (Fig. R1-10b). Because the Bi₄Br₄ stripe is mounted strictly parallel to the distal end of the PtIr wire, it consequently inherits this ~10° deviation from the surface normal of the sample. This residual tilt is documented in optical photographs taken during the experiment (Fig. R1-11).

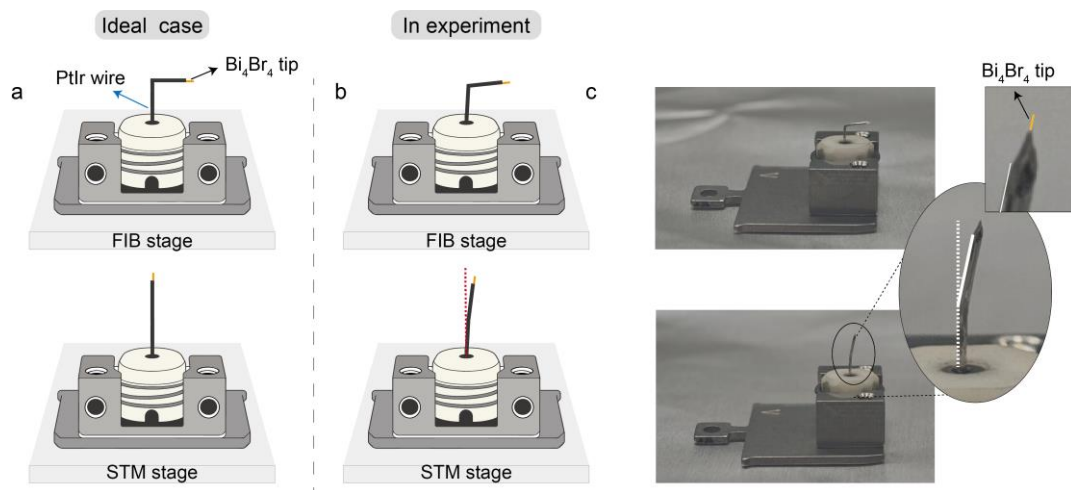


Fig. R1-10 **a**, Sketch showing the ideal case to prepare the Bi₄Br₄ tips, in which the tip is strictly vertical to the surface of sample. **b**, Sketch showing the experimental process in which there is an unavoidable geometric tilt (approximately 10°) of the tip relative to the surface normal. **c**, Photos to show the tilting of the tip. The Bi₄Br₄ stripe is strictly parallel to the end of PtIr wire.

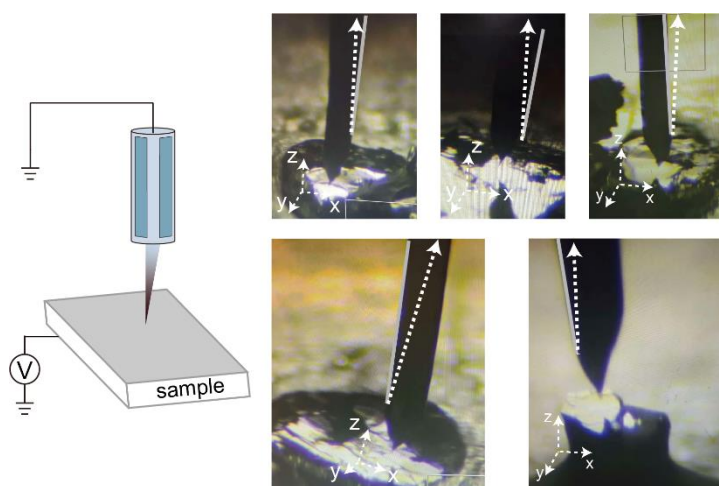


Fig. R1-11. Several optical photographs taken during the experiment to show the relative tilt of the tip (white line) to the surface normal (dashed arrow).

Consequently, the spin polarization of the helical edge modes is not strictly orthogonal to the out-of-plane spins of the substrate. This geometric projection yields a finite, overlapping spin component, allowing for the detection of an effective spin contrast in the dI/dV measurements (similar to TMR). *This could be another reason for the reduction of effective spin-polarization ratio.*

In the updated version, we have added a related discussion in the Methods section, changed Fig. 2e and added related figures to Extended Data Figure 2g-j.

3. The TI exhibits helical edge states that may slightly gap out due to interlayer hybridization leaving still topological hinge states. Often for hinge states it is important that the macroscopic sample is invariant under the protecting symmetry. The symmetry requirements for hinge-state protection in a conical tip geometry remain unclear. How do the authors rationalize the hinge mode? Connected to this, the manuscript does not clearly explain the orientation of the helical surface states of the 3D QSHI phase: In Fig. 2a it is rather clear that these propagate on the sides of the (010) layers, but the sample orientation in Fig. 2e or Fig. 1c is less clear. Why do the helical edge modes just end? As extended Fig. 1c, d shows the tips would be wide enough to stabilize the 3D QSHI modes running through the tip.

Reply 1-3: We sincerely thank the referee for raising this insightful point. The referee is correct that the topological protection of *gapless* hinge states in HOTIs strictly requires specific crystalline symmetries, making their configuration highly sensitive to the sample geometry. However, as theoretically established in Ref. 59 and corroborated by our experimental observations (Figure 2c, d), α -Bi₄Br₄ also belongs to the 3D QSHI with *slightly gapped* (almost invisible in our data) helical edge channels for each layer (also see our previous experiments on α -Bi₄I₄, Ref. 60). It can be viewed as a stacking of 2D QSHI layers (*with tiny gap due to weak interlayer coupling*). The presence of these *slightly-gapped* edge channels is robust and does not rely on special crystalline symmetry. If the Fermi level is placed far away from the tiny hybridization gap, the material behaves like a weakly-coupled stacked QSHI system. To illustrate this layered QSHI feature, we show several dI/dV maps taken at the boundaries of monolayer, bilayer, and trilayer step edges, all of which clearly exhibit the robust presence of these edge channels (Fig. R1-12a-c).

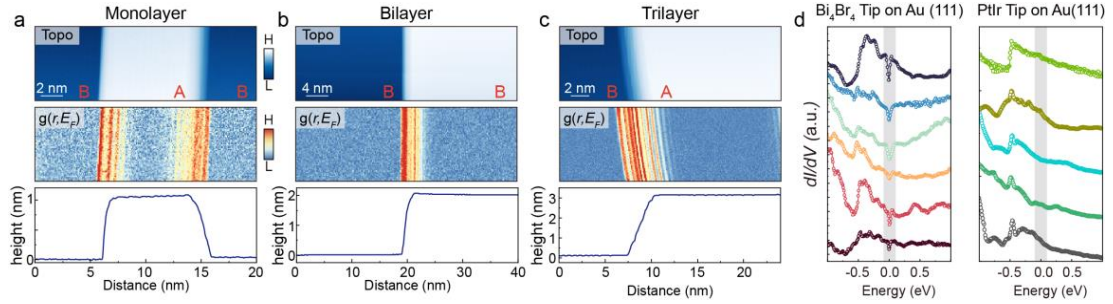
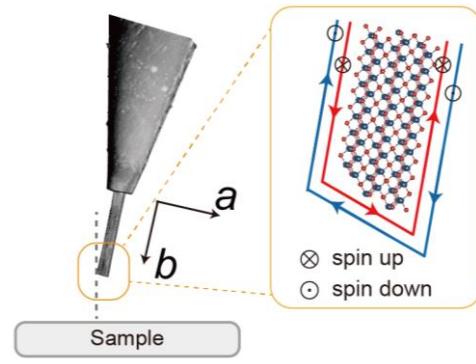


Fig. R1-12 a-c, topographies and the corresponding dI/dV maps at E_F for monolayer, bilayer, and trilayer step edges in Bi₄Br₄ crystal. **d**, comparison of Bi₄Br₄ tips and PtIr tips on Au surface to show the TLL anomaly.

Experimentally, we found that over 60% of the fabricated Bi₄Br₄ tip successfully exhibit the characteristic spectral signatures of these edge states after tip conditioning on an Au (111) surface. This refers to the TLL anomaly near E_F (Figure R1-12d), a feature entirely absent for conventional PtIr tips.

This high success rate strongly implies that the active tunneling states for the Bi_4Br_4 tip *are indeed these slightly gapped, weakly-coupled helical edge channels circulating within each layer*. Furthermore, given the quasi-one-dimensional chain-like crystal structure of Bi_4Br_4 , the physical apex of the tip should be terrace-shaped instead of conical-shaped, with the edge states propagating along the chains. To clarify the spatial orientation of these helical states and eliminate any ambiguity, we have revised Figure 2e (see Figure on the right).



Finally, the referee is completely correct that these edge channels cannot simply terminate abruptly; they must form closed loops circulating along the boundaries of the outmost layer on the tip. In the revised manuscript, we have updated the relevant figures to accurately depict the fully circulating nature of these edge channels (Fig. 1c and Fig. 2e) and have expanded the corresponding discussion in the Methods section to prevent any further misunderstanding.

4. Figure S8b, e seems to have a similar anisotropy between x and y for some values of q as Fig. 5g, i. Why is that? Also regarding anisotropies, if the tip states of $\alpha\text{-Bi}_4\text{Br}_4$ are quasi 1D, would that not also affect tunneling probabilities besides the spin, especially since helical and substrate spin axes are orthogonal?

Reply 1-4: We sincerely thank the referee for this insightful comment. The data presented in Fig. S8 were acquired using a standard non-magnetic PtIr tip. The observed anisotropy in these QPI maps originates purely from the asymmetric geometric shape of the tip apex. between x and y in QPI maps originates from the anisotropy of the tip apex. Such structural anisotropy of tip is ubiquitous and notoriously difficult to control in STM measurements; therefore, one cannot draw any conclusion solely from this observation. Crucially, because this anisotropy is rooted in the geometry shape of the tip, its principal axes remain static and do not rotate upon reversing the bias voltage. Across all three of our control datasets obtained with different PtIr tips, the anisotropy indeed does not rotate when switching the bias polarity, as exemplified in Fig. R13a–c.

To disentangle the intrinsic spin-texture from tip-shape artifacts, a spin-resolving tip is needed. For conventional magnetic tips, this typically requires taking two separate measurements under opposing external magnetic fields to reverse the tip's magnetization, where the differential signal yields the spin-resolved information. However, ensuring that the microscopic tip apex remains

strictly identical when magnetic fields sweeps is usually challenging.

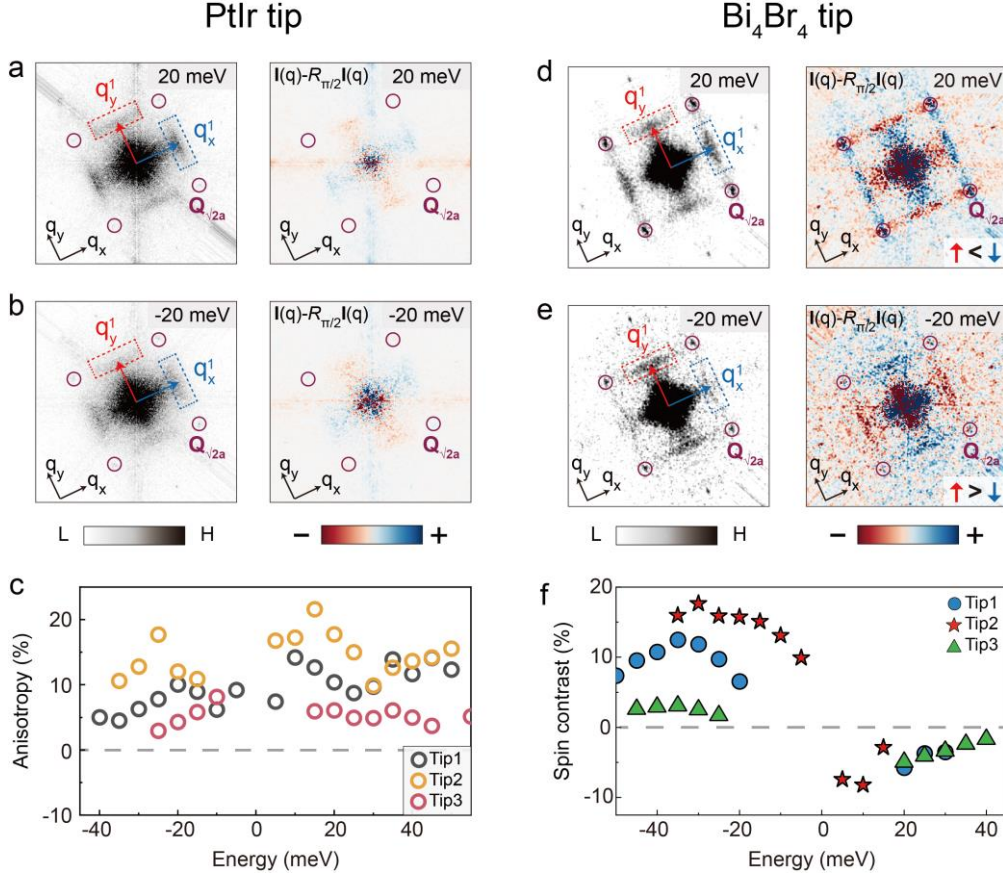


Fig. R1-13 a-b, dI/dV maps and the corresponding contrast map of rotating, at 20 (a) and -20 meV (b) with PtIr tips. **c**, the anisotropy ratio between q_x and q_y as a function of energy, without sign change for all three different PtIr tips. **d-f**, dataset acquired with Bi₄Br₄ tips, which collectively show that the anisotropy axis rotates for opposite bias voltages.

With the Bi₄Br₄ tip, the spin-texture of the sample's electronic states is probed via the tunneling of helical edge states. Because electrons with opposite spins are selectively probed at opposite bias voltages, the intrinsic spin-related QPI anisotropy inherently rotates by $\pi/2$ when switching the bias polarity (as consistently demonstrated across three independent datasets with different samples and different tips in Fig. R1-13d-f). This behavior is totally different from that with the non-magnetic PtIr tips, and reflects the spin-texture of electronic states along x- and y- direction (**a conclusion further corroborated by our magnetic-field-induced Zeeman shift measurements**). Furthermore, this approach extracts the spin information in a single, continuous measurement using an unperturbed tip.

We completely agree with the referee that additional factors (e.g., orbitals that may affect the tunneling probabilities) must be rigorously evaluated to substantiate the claim of true spin resolution. We address this critical concern from both theoretical and experimental standpoints:

First, from a theoretical perspective, we have calculated the orbital characteristics of the helical edge state in Bi₄Br₄. The main results are summarized in Fig. R1-14. For edge channels propagating along the atomic chains, the electronic states are dominated by Bi-p_x orbital (defined within the local

coordinate system of the Bi_4Br_4 crystal; Fig. R1-14b). For channels residing on the lateral facets (tip ending), the dominant contributions equally arise from the Bi-px and Bi-py orbitals (defined within the local coordinate system of the Bi_4Br_4 crystal; see Fig. R1-14c). In our experimental STM configuration, the Bi- p_y orbital lobes extend outward from the tip apex, oriented nearly perpendicular to the measured sample surface (Fig. R1-14a). Consequently, from the perspective of the tunneling junction, the tip effectively acts as a “ p_z ”-like orbital probe relative to the sample plane. Because this extending orbital is nearly normal to the surface, it maintains an isotropic tunneling matrix element with respect to the in-plane d_{xz} and d_{yz} orbitals of the sample (what we measure in $\text{KV}_2\text{Se}_2\text{O}$). Therefore, this geometric configuration is highly unlikely to cause significant, purely orbital-driven differences in the tunneling probabilities.

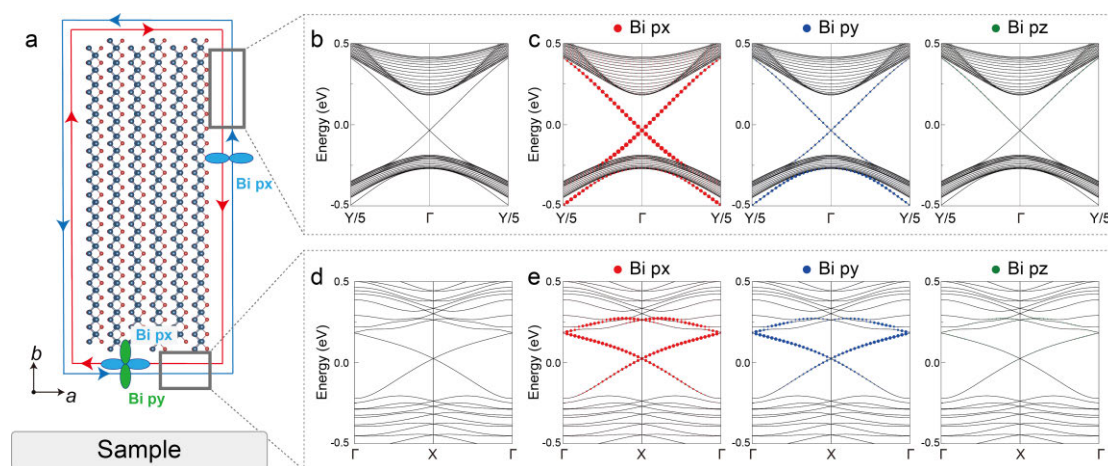


Fig. R1-14 **a**, Sketch for the tunnelling junction in which the orientation of helical edge channels and their orbital characteristics are included. the Bi- p_y orbital lobes extend outward from the tip apex, and consequently, the tip effectively acts as a “ p_z ”-like orbital probe relative to the sample plane. **b-e**, calculated overall dispersion and the orbital characteristics of the helical edge state in Bi_4Br_4 , propagating along the atomic chains (**b**, **c**) and residing on the lateral facets (**d**, **e**).

Second, from an experimental perspective, as detailed in our preceding responses, we have performed spin-polarized STM measurements on *four distinct magnetic compounds* using Bi_4Br_4 tips. In every case, we successfully resolved local density of states (LDOS) features that accurately reflect their predicted magnetic ground states. **This systematic consistency definitively rules out coincidental orbital artifacts.** Most notably, in the kagome magnets FeSn and FeGe , which host A-type AFM orders (Figs. R1-5 and R1-6), we resolved strong spin-related DOS contrast between adjacent layers separated by a single unit-cell step edge. **Because these adjacent layers share identical orbital and chemical environments but possess opposite spin orientations, any extraneous effects from orbital symmetries and spin-independent tunneling matrix elements are effectively cancelled out.** Finally, our magnetic field-dependent control experiments on FeGe (Figure R1-6) provide decisive evidence: **by applying an external magnetic field to induce a spin-flop transition, we observed a dramatic enhancement of the DOS contrast between adjacent unit cells.** Crucially, **these measurements were performed over the exact same area with the identical tip, isolating the magnetic spin reorientation as the sole variable responsible for the signal change.**

We believe that these robust experimental demonstrations, combined with our theoretical analysis of the edge-state orbital symmetries, can largely address the referee’s concern. We also believe these

new and detailed STM experiments establish a rigorous foundation for the utilization of topological quantum materials as advanced spin-polarized scanning probes.

In the revised manuscript, we have included the control experiments using PtIr tips in Extended Data Fig. 10, the calculated orbital feature of Bi_4Br_4 in Extended Data Fig. 1, the control experiments of FeGe in Extended Data Fig. 5, and added related discussion in the main text (page 7) and more detailed information in Methods.

Smaller questions:

- When the bias changes sign the contributions of q_{1x} and q_{1y} momenta change sign, as far as I can tell the intensity at smaller momenta does not do that. Is there a reason? Does the JDOS in Fig. 5e change with the offset?

Reply 1-5: We thank the referee for this insightful observation. Generally, the QPI signal at small momenta is more challenging to analyze quantitatively. Contributions from long-range spatial modulations and the random distribution of impurities (specifically the high concentration of V vacancies in our samples) tend to dominate the spectral weight near $q \sim 0$. In the dataset presented in Fig. 5, the signal-to-noise ratio at low momenta is relatively limited, rendering the expected sign-reversal less discernible compared to the more distinct q_{x1} and q_{y1} features. However, in the data from sample #1 (Extended Data Fig. 9), this sign-change upon reversing the bias polarity becomes more evident (see Fig. R1-15), likely due to a lower background intensity in the small- q regime.

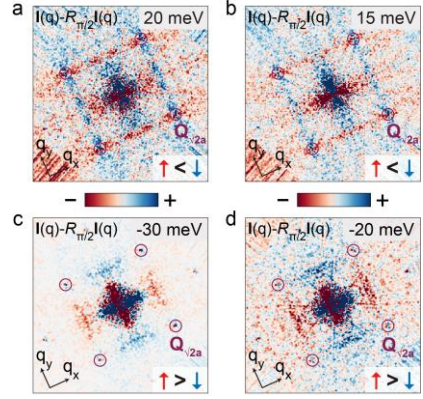


Fig. R1-15. Spin contrast maps at four energies obtained on sample #1 with Bi_4Br_4 tip#1. The sign-reversal in these data (upper and lower panels) becomes more evident at small q .

Furthermore, we have calculated the DOS and confirmed that there is no significant energy-dependent evolution for the spin-up and spin-down channels within the experimental energy window. Consequently, according to the spin-selection rules associated with the Bi_4Br_4 tip, a sign-reversal near $q \sim 0$ is theoretically predicted, which is consistent with our observations in samples where the $q \sim 0$ noise is sufficiently suppressed.

- In Fig. 2h the stripe pattern is stated to reverse with reversed bias. What is the reason for the deviation from a perfectly mirrored pattern? Besides differences in the shape of curves they are also shown with an offset. Is this offset just by presentation or measured?

Reply 1-6: We sincerely thank the referee for this comment. We have carefully checked these data and found that the additional feature at negative biases is the atomic modulation in LDOS (Fig. R1-16b, c). As shown in Fig. R1-16d, the FFT of dI/dV linecut at -50 meV shows both $2a$ modulation (AFM vector) and modulation of a (Bragg modulation). Notably, the latter Bragg modulation is significantly suppressed in the images acquired at positive biases (Fig. R1-16c and d). By applying

a q -space filter to remove the Bragg contribution (with the cutoff marked by the gray shaded region in Fig. R1-16d), a nearly mirrored pattern is recovered, as shown in Fig. R1-16e.

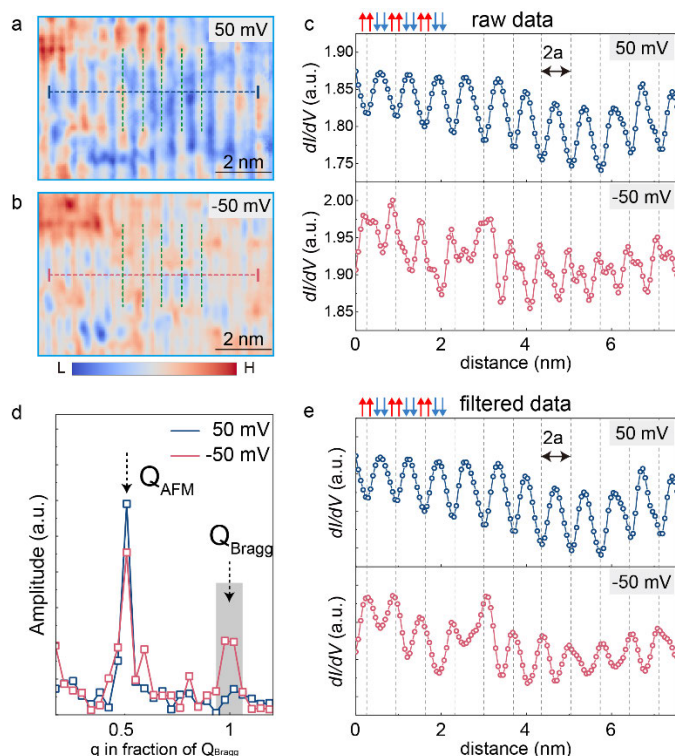


Fig. R1-16. **a, b,** dI/dV maps obtained at 50meV and -50meV. **c,** linecuts of the dI/dV spectra. To show the overall tendency, the linecuts are averaged over 30 pixels perpendicular to the linecut direction. **d,** FFT for the linecut profile shown in **c**, where both peaks at AFM vector and Bragg vector can be observed at -50meV. The gray shaded region denotes the q -space window for Fourier filter. **e,** a nearly mirrored pattern is recovered after the Fourier filter process.

Regarding the presentation in Fig. 2h, the vertical offset between the two curves was initially introduced for visual clarity. We acknowledge that this may have caused ambiguity; therefore, we have updated Fig. 2h in the revised manuscript to avoid any confusion. We have also added a related discussion in Supplementary Note 3.

- In line 159 the authors speak of "twofold defects", in what sense are they twofold?

Reply 1-6: Sorry for the confusion. Here the term "twofold" refers to the local C_2 structural environment for these specific defects, due to the Lieb lattice feature of vanadium-oxygen plane. A schematic illustration of this crystalline environment was previously provided in Fig. 3g to visualize the local coordination. To improve clarity and ensure the term is not misinterpreted, we have revised the corresponding sentence in the manuscript to explicitly state that these defects reside in a site with C_2 symmetry.

Overall Reply: We sincerely thank the referee for the positive assessment of the quality, significance, and broad interest of our work. By incorporating new STM data from additional magnetic compounds and performing carefully designed control experiments, we have established a more coherent and rigorous picture of the proposed methodology. These additions further substantiate our conclusions regarding altermagnetism and enhance the overall robustness of the study. We have revised the manuscript to improve clarity and have included more in-depth discussions addressing the referee's remarks. We believe that this methodology, along with its underlying physics, represents a substantial advancement in the fields of topological physics, scanning tunneling microscopy, and altermagnetism.

Response to Referee #2

The authors introduce a spin-sensing method for functionalizing STM tips using the bulk crystal of a HOTI α -Bi₄Br₄. The central premise is that the helical edge modes of the HOTI, which possess spin-momentum locking, can act as a spin filter without the need for magnetic tip coatings or external magnetic fields. They apply this helical tunneling technique to KV₂Se₂O, a candidate for 2D d-wave altermagnetism, and claim to observe QPI patterns that rotate by $\pi/2$ upon reversing the bias polarity. They interpret this rotation as “smoking gun” evidence for altermagnetic order.

The concept of exploiting topological edge modes as a metrological tool for spin sensing is interesting, and the target of identifying d-wave altermagnetism is high-impact in current condensed matter physics. However, similar idea of spin sensor by STM has been validated before (Science 377, 1218–1222, 2022), thus reducing its originality. Further, for a claim of this magnitude, the experimental evidence for the altermagnetism must be unambiguous. Upon close inspection, I find the execution and data analysis insufficient to support the high claims made in the manuscript. The data is overselling and the distinction between intrinsic topological spin-filtering and extrinsic artifacts (such as orbital effects or tunneling matrix anisotropy) has not been robustly established. Consequently, I am not convinced this constitutes the “experimental milestone” claimed in the abstract and cannot recommend publication in Nature.

Reply: We thank the referee for the positive assessment of the significance and interest of our work. We also appreciate the constructive suggestions, which have been instrumental in improving the manuscript.

We totally understand the referee’s concern on the validity of this relatively new method (utilizing STM tips with helical edge states for spin-resolved measurements), and fully agree that particular caution is warranted before drawing definitive conclusions.

Since our previous submission, we have significantly expanded the scope of our study to reinforce the validity of our conclusions. Specifically, we have fabricated over 30 Bi₄Br₄ tips, characterized four distinct classes of compounds with diverse magnetic structures, conducted three types of control experiments (under applied magnetic fields, rotation of the sample and using a conventional magnetic tip), and performed calculations of the orbital and spin characters of the Bi₄Br₄ edge state. Collectively, these systematic investigations have yielded a more detailed and comprehensive understanding of the spin-polarization capability of this unconventional tip. These experiments further suggest that the Bi₄Br₄ tip is more sensitive to magnetic structures with in-plane spin orientations, and further validate its distinct characteristics relative to a conventional magnetic tip. In light of these findings, we first present a summary of our key findings prior to addressing the specific points raised by the referee, and show how our work advances the application and fundamental understanding of these STM tips beyond the seminal work reported in Science 377, 1218–1222 (2022).

The spin orientation of edge-state electrons from theoretical perspective. Based on the intrinsic spin-resolved topology of 3D insulators, α -Bi₄Br₄ is classified as a 3D QSHI (Nat. Commun. 15, 550 (2024)), exhibiting a bulk spin polarization along the S_z direction (nearly quantized as $0.97 \times \pm \hbar/2$). Our DFT calculations indicate that the edge state electrons also mainly possess an out-of-plane spin orientation ($S_z \sim 0.85 \times \pm \hbar/2$; see Fig. R2-1). In the geometry of the STM measurement, this spin orientation lies “in-plane” with respect to the target sample. Therefore, the spin orientation of the 1D edge states in Bi₄Br₄ could be easier to predict than these 2D surface states with helical spin-texture.

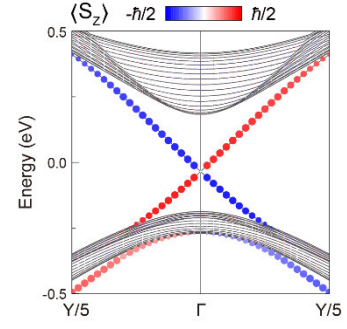


Fig.R2-1 S_z as the major spin orientation for the edge states in Bi₄Br₄

To gain a more comprehensive understanding of its spin-polarization capability, we have measured four distinct classes of compounds with different magnetic structures: Fe_{1+x}Te, kagome magnet FeSn, FeGe and finally KV₂Se₂O. We also performed three types of control experiments (under magnetic fields, rotated the sample and using conventional magnetic tips and PtIr tips). **This systematic study demonstrates that the spin polarization of the Bi₄Br₄ tip is intrinsic and more sensitive to in-plane spin components, and further verifies its distinct characteristics relative to a conventional magnetic tip.** In the following, we describe several key experimental findings in detail.

1. Fe_{1+x}Te with bicollinear AFM order.

- **A contrast reversal at opposite bias voltages using Bi₄Br₄ tips.** We confirm that the spin-polarized local DOS (with the bicollinear AFM order) *undergoes a contrast reversal at opposite bias voltages using Bi₄Br₄ tips* (the additional kink at negative biases corresponds to

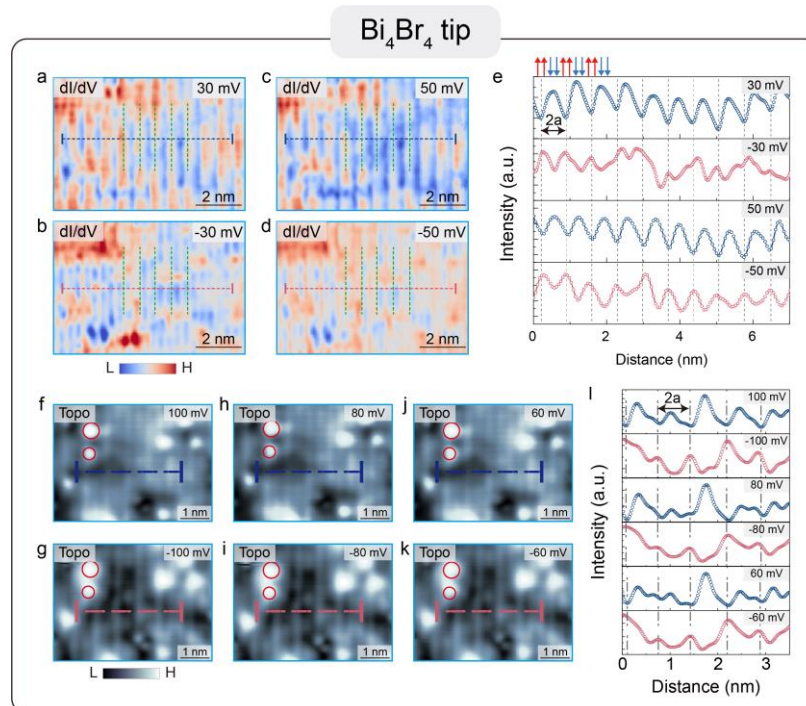


Fig. R2-2 Bicollinear AFM order in Fe_{1+x}Te imaged by the Bi₄Br₄ tips. **a-d**, dI/dV maps and **f-k**, topographic images. In both cases, the spin-polarized local DOS of $2a$ modulation exhibits a π -phase shift (contrast reversal) at opposite bias voltages, as shown in **e** and **i**. To show the overall tendency, the linecuts in **e** and **i** are averaged over 30 and 20 pixels perpendicular to the linecut direction, respectively.

atomic modulations, as shown later in Reply 2-1), which can be observed in *the dI/dV maps and our newly acquired topographic images* (Fig. R2-2).

➤ **Such contrast reversal is absent when using a conventional magnetic tip.** It is necessary to examine whether the orbital/charge distribution of the Fe_{1+x}Te sample itself can produce a similar contrast reversal, before concluding that this *contrast reversal* primarily originates from spin selection by the Bi_4Br_4 tip. We performed spin-polarized LDOS measurements *using Fe-coated PtIr tips*. Although signatures of the bicollinear AFM order could be still observed, with these conventional magnetic tips we found that the contrast reversal was absent upon reversing the bias-voltage polarity (Fig. R2-3). This comparison indicates that the contrast reversal at opposite biases for the Bi_4Br_4 tip should originate from the unique spin selection rule of the helical tunneling.

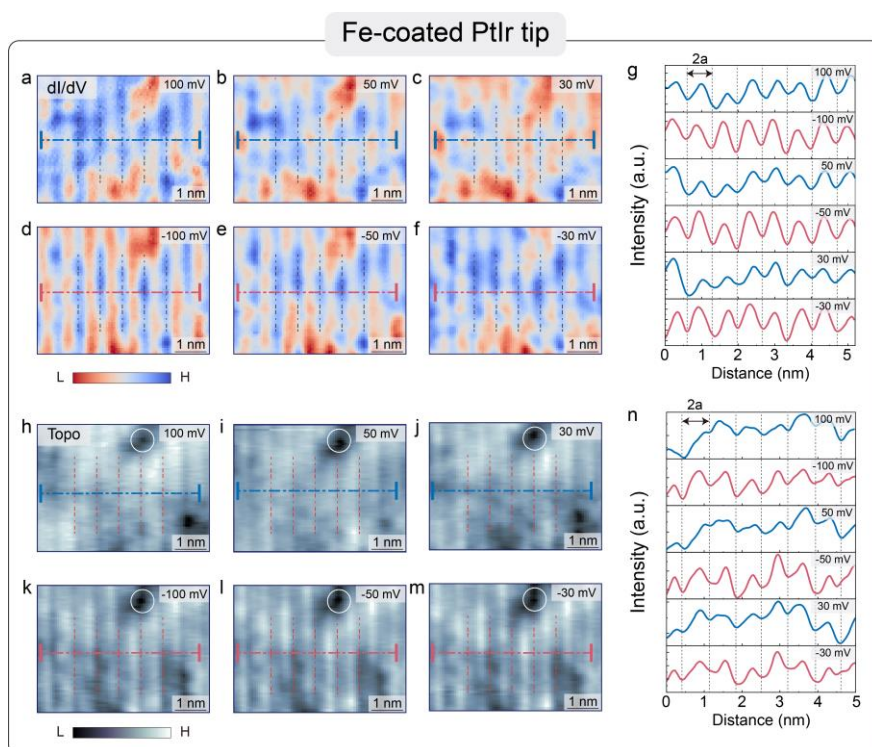
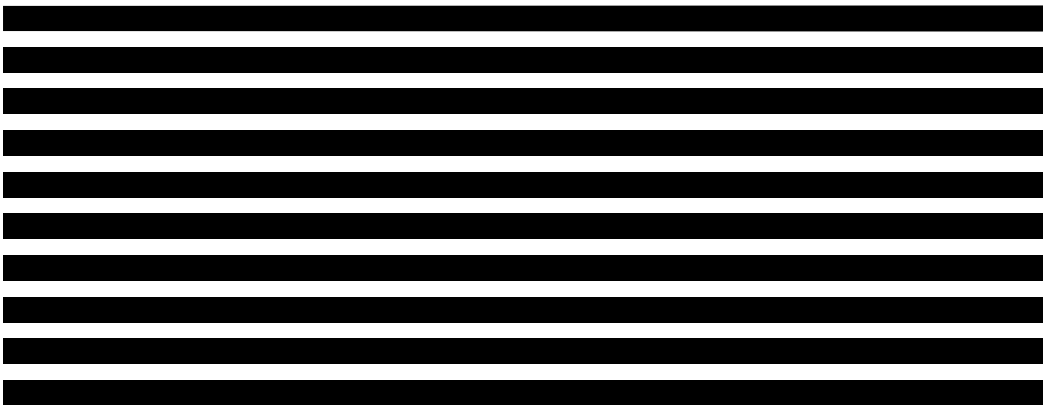
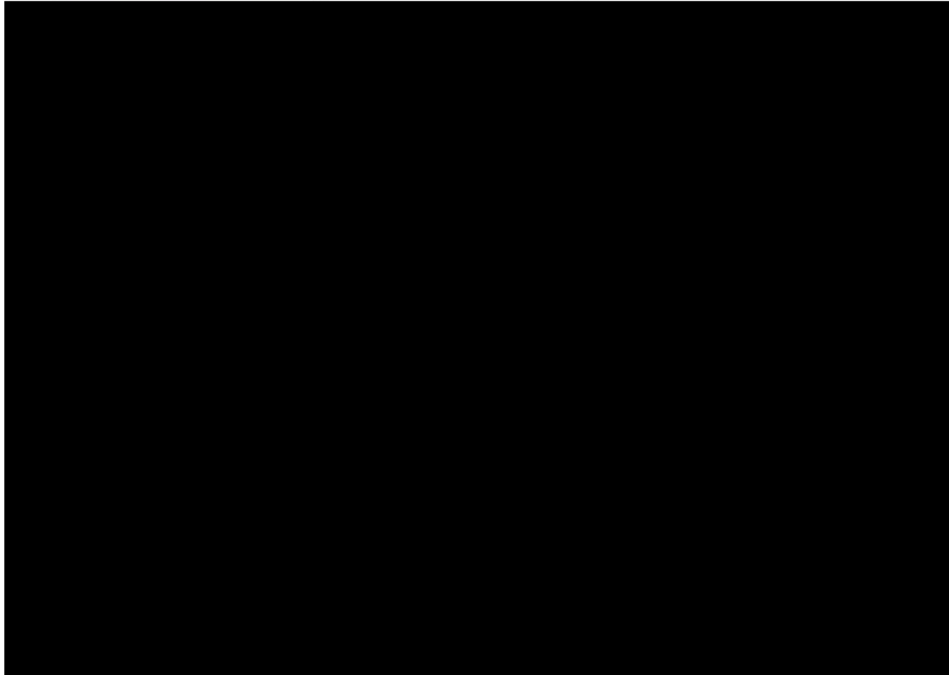


Fig. R2-3 Bicollinear AFM order in Fe_{1+x}Te imaged by the Fe-coated PtIr tips. **a-f**, dI/dV maps and **h-m**, topographic images. In both cases, the spin-polarized local DOS of $2a$ modulation exhibit a same phase (without contrast reversal) at opposite bias voltages. To show the overall tendency, the linecuts in **g** and **n** are averaged over 30 pixels perpendicular to the linecut direction, and no contrast reversal is observed.





[REDACTED]

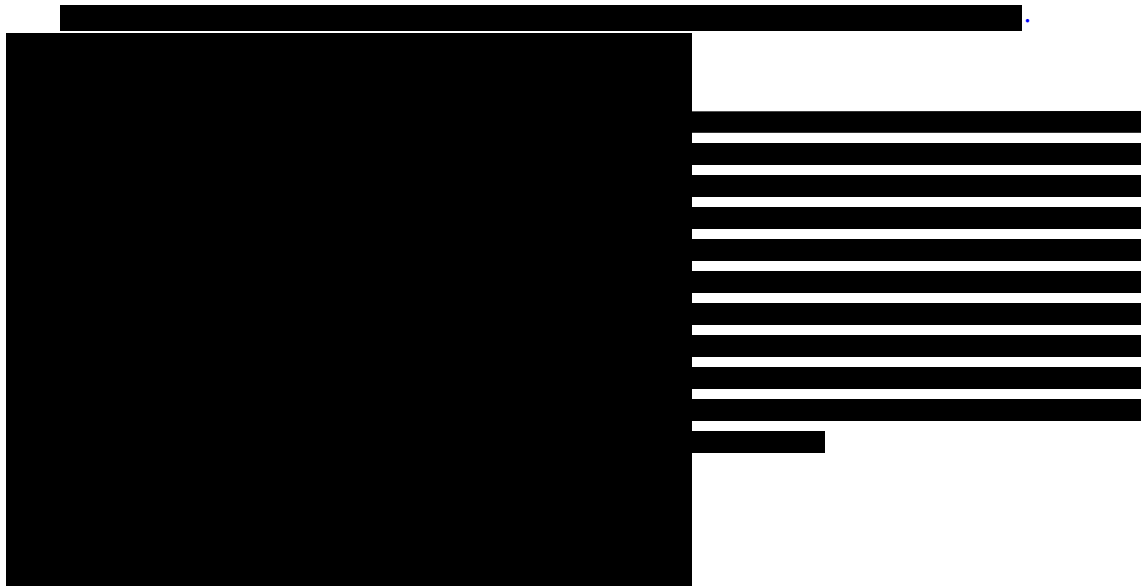


[REDACTED]

These experiments in Fe_{1+x}Te demonstrate **the intrinsic difference between Bi_4Br_4 tips and magnetic tips**, suggests that the orbital feature of Fe_{1+x}Te is unlikely to be the origin for the contrast reversal at opposite bias voltages seen by Bi_4Br_4 tips, and further suggest that the spin orientation on the Bi_4Br_4 tips more likely lies in one “in-plane” direction with respect to the target sample.

2. Kagome metal FeSn with A-type AFM order. For an A-type AFM order, spins are ferromagnetically aligned within one layer but antiferromagnetically coupled between adjacent layers. *This magnetic configuration provides an ideal platform to verify the spin resolution of the Bi_4Br_4 tip, as additional contributions from orbital effects and tunneling matrix elements should be largely suppressed between these two layers with same atomic constitutions.*

[REDACTED]



3. Kagome metal FeGe with A-type AFM order: another key compound

FeGe hosts an A-type AFM order *with canted spins mainly oriented out-of-plane at 0T* (Fig. R2-6a; Nature 609, 490 (2022)). With an external out-of-plane magnetic field exceeding 7T, the system undergoes a spin-flop transition, reorienting the spins into in-plane direction (Fig. R2-6b; Nature 609, 490 (2022)). Consequently, magnetic-field-dependent STM measurements are highly valuable for further elucidating the spin resolution of the Bi₄Br₄ tip.

- **Bi₄Br₄ tips are more sensitive to in-plane spin components.** Figure R2-6c shows an STM topographic image of a cleaved FeGe surface featuring a single-unit-cell step edge. The two sides of the edge are thus identical (except for spins). Under zero or weak (± 1 T or ± 2 T) magnetic fields, the spin orientation of FeGe remains predominantly out-of-plane, whereas the spin polarization of the tip is in-plane (enforced by the large energy scale of SOC in Bi₄Br₄ so that magnetic field of a few Tesla cannot change the spin direction on the tip); therefore, the dI/dV maps exhibit very weak contrast between adjacent layers (Fig. R2-6d). However, *when an external field of ± 8 T is applied to induce the spin-flop transition, a pronounced contrast emerges between the two layers* (Fig. R2-6 e, f). This arises because the spins of both the sample and the tip are now aligned in-plane, yielding a finite overlapping component between them. Upon reducing the external field to ± 1 T, this contrast correspondingly diminishes again (Fig. R2-6g-h).

This series of experiments were carried out in the same area with the same STM tip, and the most obvious change is the spin direction on the sample (but with the contribution from orbitals, spin-independent tunneling-matrix elements largely unchanged). Thus, we believe *it provides compelling evidence that the spin polarization of the helical tunneling channels is intrinsic, and the spins on tip are “in-plane” relative to the coordinate system of the target sample.*

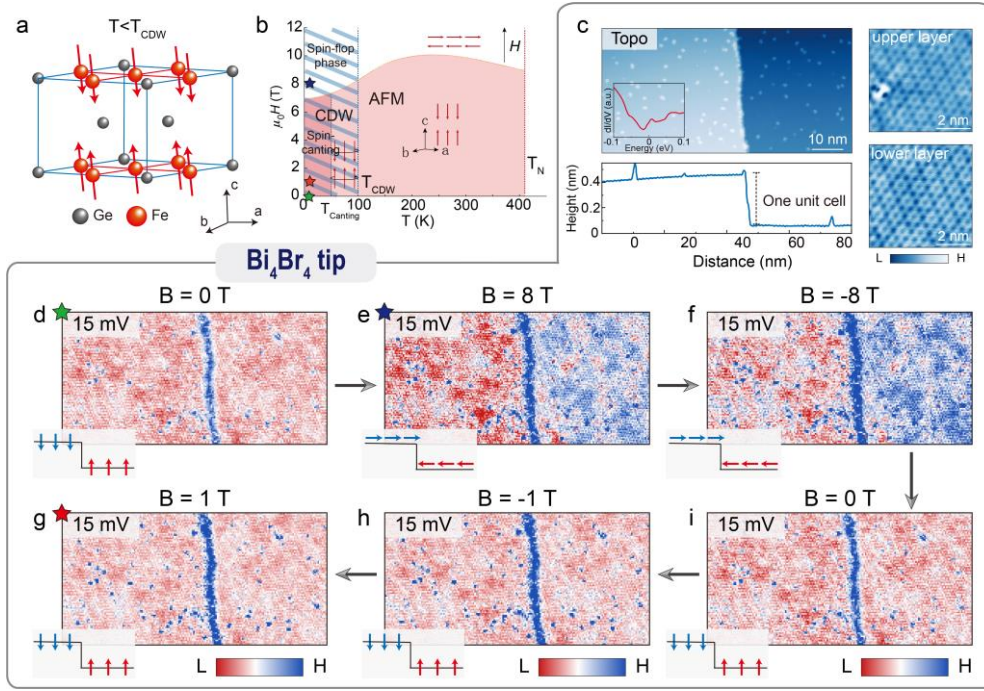


Fig. R2-6 **a**, Magnetic structure of FeGe at 0 T. **b**, Phase diagram of FeGe with field. **c**, STM topography including one single-unit-cell step edge. The right panels show atomic images of the upper and lower layers. **d-i**, dI/dV maps recorded at 15 meV with varying the magnetic field. A large enhancement of the contrast is found at ± 8 T under which the spins flop.

- **The spin resolution behavior of Bi_4Br_4 tip is again different from that of magnetic tip.** For a standard magnetic tip, a small external field of 1 T or 2 T is typically sufficient to fully polarize its apex out-of-plane. Given that the spin orientation of FeGe is also predominantly out-of-plane under these conditions, a distinct spin-polarized contrast should be readily observable at 1 T or 2 T. To experimentally verify this picture, we have prepared Fe-coated tips. The resulting data reveal a pronounced spin contrast at 1 T (or 2 T), as shown in Fig. R2-7. This conventional magnetic-field dependence is fundamentally distinct from the behavior observed with the Bi_4Br_4 tip, further corroborating the non-trivial, in-plane spin resolution of the helical tunneling channels in the Bi_4Br_4 tip.

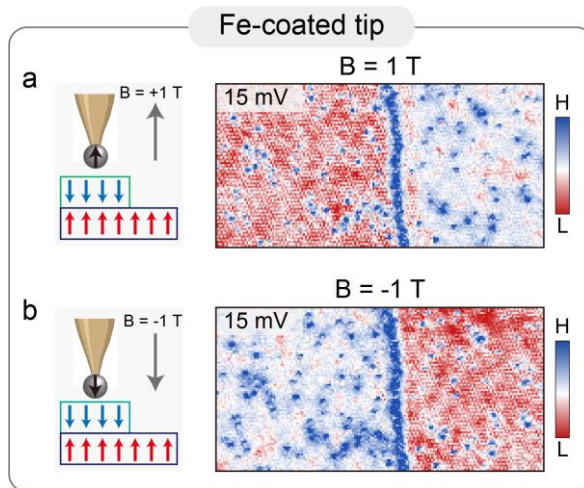


Fig. R2-7. dI/dV maps at 15 meV under magnetic fields of 1 T (**a**) and -1 T (**b**), in the same area but measured with a Fe-coated STM tip. An external field of 1 T is typically sufficient to fully polarize the tip's apex spin to out-of-plane, therefore a clear contrast can be observed. Reversing this smaller field flips the spin direction on the tip but keeps these in the sample, thus the contrast reverses between layers at ± 1 T.

4. KV₂Se₂O with d-wave checkerboard AFM order. The opposite Zeeman shifts of the in-gap states confirm their opposite spin polarization along the x- and y- directions (Fig. 4; we updated these data with much higher quality, see Reply 2-9). This is a direct and well-established way to determine the spin texture of these states. Consequently, the observed $\pi/2$ rotation of the QPI anisotropy upon reversing the bias voltage polarity can be fundamentally attributed to the underlying spin texture of these electronic modulations, and **we further confirmed that such rotation is absent when using three spin-summed PtIr tips** (Reply 2-7).

Summary. By investigating four distinct magnetic compounds, we have consistently obtained spin-resolved STM data *that reflect their respective magnetic structure, which can rule out the possibility of coincidental artifacts. We hope the reviewer appreciates the experimental efforts we have dedicated to comprehensively addressing this crucial aspect.* Furthermore, these collective data consistently indicate that the spins carried by the helical edge channels are more sensitive to in-plane spins on the target sample.

Our work reports the successful fabrication of STM tips from complex bulk quantum materials, demonstrates the presence of robust helical edge channels **on tips** with an exceptionally large QSH gap, and provides systematic experimental elucidation showing that the tunneling of helical electrons yields spin resolution. A primary contribution of this study, which distinguishes it from the pioneering work of SmB₆ nanowires (Science 377, 1218–1222, 2022), *lies in the well-defined nature of the topological boundary states in Bi₄Br₄ compared to the more complex Kondo insulator and discovered that the spins carried by the helical edge channels are more sensitive to in-plane spins on the target sample.* We are confident that this methodology represents a substantial advancement in the fields of topological materials and STM technique, alongside the physics of altermagnetism.

In the revised version, we have included the typical datasets in Extended Data figures and Supplementary Notes: Fe_{1+x}Te (Extended Data Figure 4; Methods; Supplementary Note 3), FeGe (Extended Data Figure 5; Methods) and KV₂Se₂O including the control experiments (Methods; Extended Data Figure 10).

Below are my detailed comments:

1. The verification of the tip's spin-polarization tunneling on Fe_{1.02}Te raises several questions regarding the fidelity of the probe. In Figure 2h, both spin-up and spin-down channels exhibit local peaks or shoulders. This complexity contrasts with the idealized schematic in Figure 1e, which implies a cleaner selection rule. Why do these specific features appear, and do they persist over a broader energy range?

Reply 2-1: We sincerely thank the referee for this comment. Upon a careful re-examination of the data, we attribute the additional features (specifically the shoulders observed at negative biases) to the superposition of atomic lattice modulations in the LDOS. As shown in Fig. R2-8d, the FFT of dI/dV linecut at -50meV contains both a **2a** modulation, corresponding to the antiferromagnetic (AFM) vector, and a modulation at **a** associated with the Bragg periodicity. Notably, the Bragg contribution is significantly suppressed at positive biases. By applying a q-space filter to remove

the Bragg contribution (with the cutoff marked by the gray shaded region in Fig. R2-8d), a nearly mirrored pattern is recovered, as shown in Fig. R2-8e.

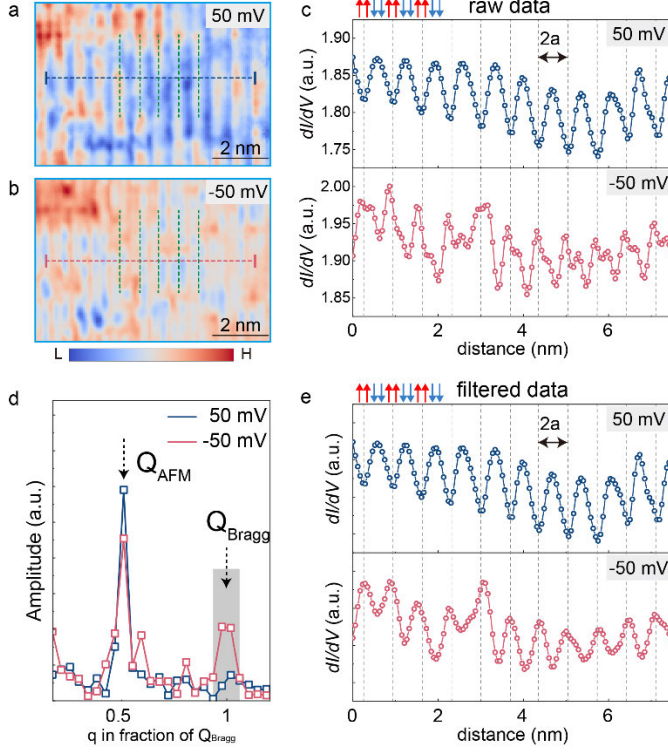


Fig. R2-8. **a, b**, dI/dV maps obtained at 50meV and -50meV. **c**, linecuts of the dI/dV spectra. To show the overall tendency, the linecuts are averaged over 30 pixels perpendicular to the linecut direction. **d**, FFT for the linecut profile shown in **c**, where both peaks at AFM vector and Bragg vector can be observed at -50meV. The gray shaded region denotes the q-space window for Fourier filter. **e**, a nearly mirrored pattern is recovered after the Fourier filter process.

Regarding the energy dependence, the $2a$ AFM modulations in the dI/dV maps are visible at low energies but diminish significantly near 80 meV (see Fig. R2-9); conversely, atomic modulations become more pronounced at higher bias voltages. It is also 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 Fig. R2-10, which consistently exhibit contrast reversal upon switching the bias polarity. Finally, we acknowledge that Fe_{1+x}Te single crystals exhibit intrinsic nanoscale inhomogeneity, likely due to interstitial Fe atoms (Nature 652, 342 (2026)). These local variations may contribute to the additional spectral features observed in both topographies and dI/dV maps. We have clarified these points in the revised manuscript to avoid any confusion between the spin contrast and the secondary lattice effects.

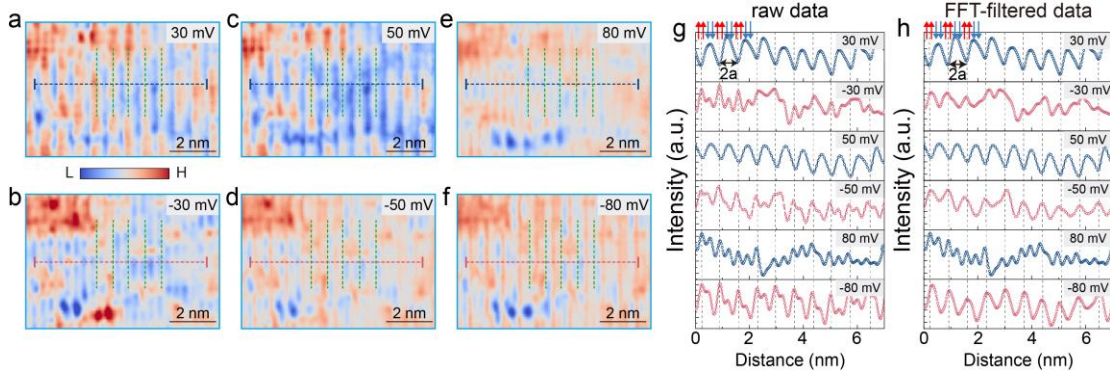


Fig. R2-9 a-f, dI/dV maps obtained at ± 30 , ± 50 and ± 80 meV. **g**, Linecuts of the dI/dV values. To show the overall tendency, the linecuts are averaged over 30 pixels perpendicular to the linecut direction. **h**, the corresponding linecuts of **g** after applying a q-space filter to remove the Bragg contribution for negative biases.

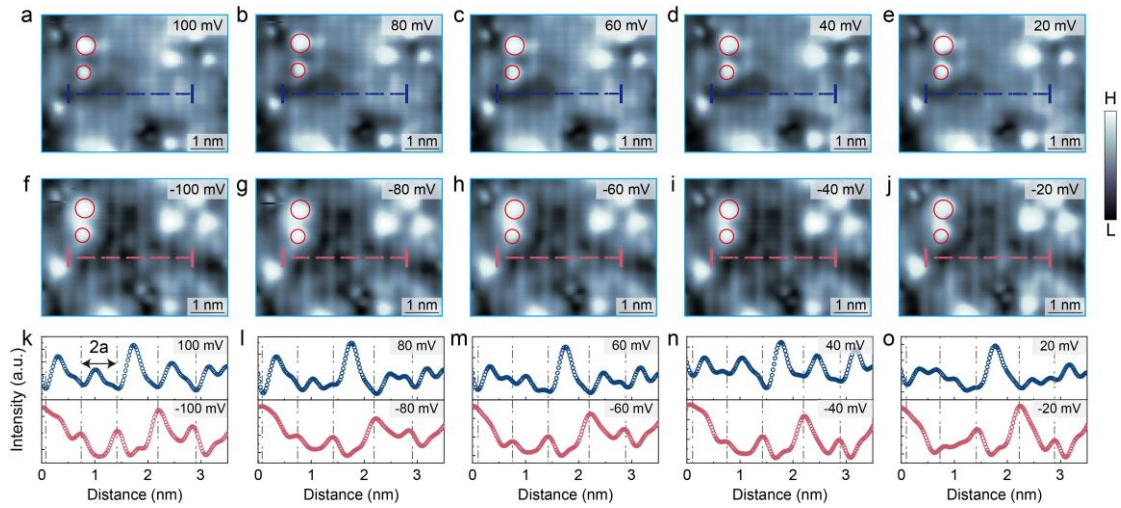


Fig. R2-10 a-j, Bicollinear AFM order in the topographic images of Fe_{1-x}Te imaged by a Bi_4Br_4 tip. **k-o,** Linecuts of the topographic images at different energies. To show the overall tendency, the linecuts are averaged over 20 pixels perpendicular to the linecut direction, and a smooth cubic polynomial background was subtracted. A contrast reversal upon switching the bias polarity can be observed in these topographic images.

2. Since the contrast reversal is demonstrated only at selected voltages. To be convincing, the authors should show that this pattern persists or evolves consistently over a wider bias window.

Reply 2-2: We agree that demonstrating the consistency of contrast reversal across a broader energy range is essential for establishing the robustness of our findings. In response to the referee's comment, we have provided several energy-dependent dI/dV maps and topographic images in Figs. R2-9 and R2-10. These data demonstrate that the contrast reversal remains consistent upon switching the bias polarity across a wide energy window.

Furthermore, we recognize the importance of distinguishing the observed π -phase shift from potential non-spin-related origins. In certain systems, such as those exhibiting CDW or specific orbital textures, a bias-dependent contrast reversal can arise purely from the sample's electronic structure. To rule out this possibility and confirm that the reversal originates from the spin-selection rules of the Bi_4Br_4 tip, we performed a comparative control experiment using a conventional Fe-coated PtIr magnetic tip to measure the same AFM modulation. As shown in Fig. R2-3, the contrast reversal is absent when switching the bias polarity with the Fe-coated magnetic tip. This direct comparison substantiates that the observed contrast reversal at opposite bias polarities is an intrinsic property tied to the spin-resolved tunneling of Bi_4Br_4 tip, rather than an artifact of the sample's local orbital or charge distribution.

In the revised version, we have included the topographic images with Bi_4Br_4 tips, and STM data with Fe-coated tips in Extended Data Figure 4. We have also added more datasets in Supplementary Note 3 and discussions in Methods.

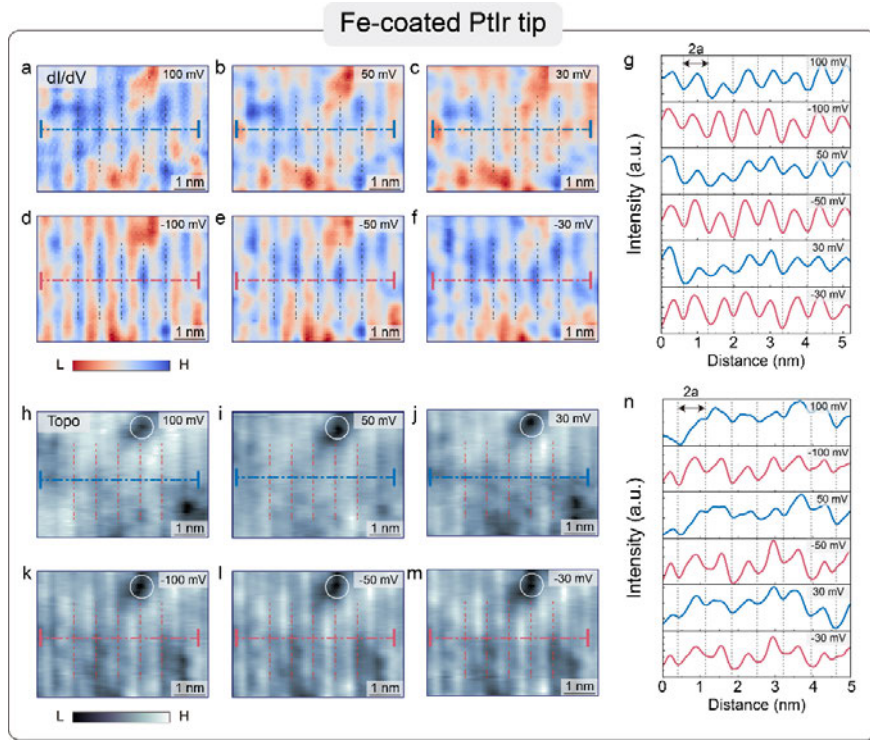


Fig. R2-3 Bicollinear AFM order in Fe_{1+x}Te imaged by the Fe-coated PtIr tips. **a-f**, dI/dV maps and **h-m**, topographic images. In both cases, the spin-polarized local DOS of $2a$ modulation exhibit a same phase (without contrast reversal) at opposite bias voltages. To show the overall tendency, the linecuts in **g** and **n** are averaged over 30 pixels perpendicular to the linecut direction, and no contrast reversal is observed.

3. While the tip is non-magnetic, the sample is. How does the system respond to reversed external magnetic fields? A robust characterization of the helical nature requires more comprehensive controls than what is currently presented.

Reply 2-3: This is an excellent suggestion. Thanks!

First, we did a test of the Bi_4Br_4 tips under magnetic fields on Au (111), to show the influence of magnetic field to the tip. As shown in Fig. R2-11, the low-energy spectra exhibit a TLL-like anomaly, characteristic of tunneling into 1D helical edge channels. Upon applying a vertical magnetic field of 7 T (oriented parallel to the momentum direction but perpendicular to the spin quantization axis), no discernible change was observed in the spectral line-shape. This stability is expected, as a field parallel to the 1D edge induces a pure Zeeman gap that is theoretically predicted to be too small to be resolved within our current experimental resolution (4.2 K).

Moreover, the spin orientation of the topological edge states is dictated by a large SOC energy scale. Consequently, an external field of several Tesla is insufficient to reorient the tip's spin polarization.

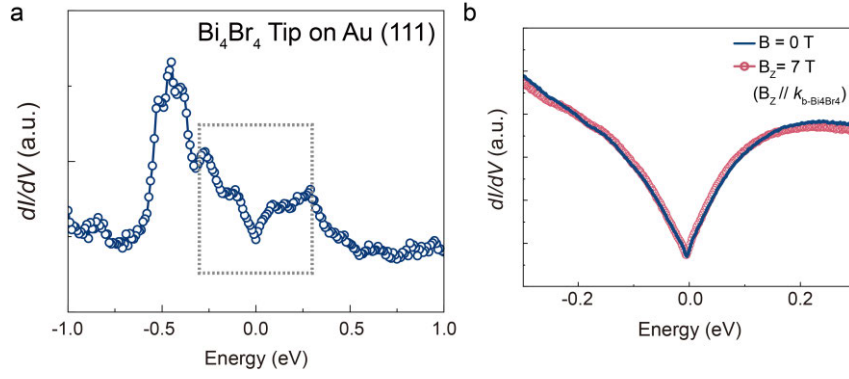
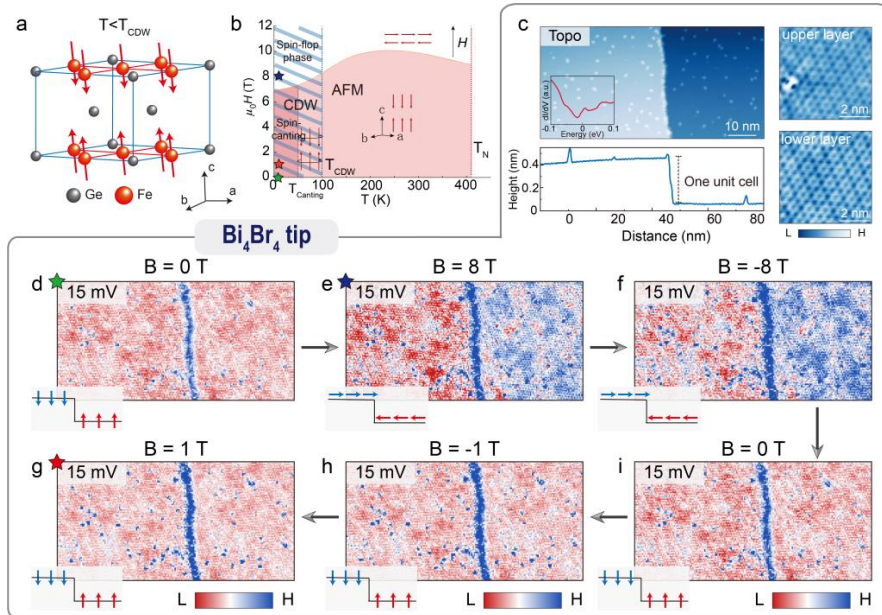


Fig. R2-11 **a**, dI/dV spectrum acquired on the Au (111) surface using a Bi_4Br_4 tip. **b**, zoom-in of the spectra line shape near E_F at 0 T and 7 T with the same tip, no discernible change was observed at 4.2 K due to the tiny pure Zeeman gap.

Third, if the magnetic structure of the target sample can be tuned by magnetic fields, such experiments would provide critical information to characterize the spin-resolution of Bi_4Br_4 tip. In this context, $\text{KV}_2\text{Se}_2\text{O}$ is not a good test system as its Neel order is very robust to magnetic fields of several Tesla (PRB 110, 165151 (2024)). Thus, we chose kagame metal FeGe.

FeGe exhibits A-type AFM order where spins can be flopped from a predominantly out-of-plane orientation to an in-plane direction under an external out-of-plane field exceeding 7 T (Nature 609, 490 (2022)). As shown in Fig. R2-6 (*reproduced below*), at zero or low fields, the contrast between adjacent layers (separated by one unit cell) is minimal, as the sample spins are out-of-plane while the tip spins remain "in-plane". However, when a field of 8 T is applied to flop the FeGe spins into the in-plane direction, a pronounced contrast emerges, reflecting the increased overlap between the sample and tip spin components. Crucially, reducing the field to 1 T causes this contrast to diminish again. This reversible behavior strongly suggests that the spin resolution originates from helical channels with a fixed "in-plane" orientation (due to large SOC) relative to the sample geometry. This is fundamentally distinct from conventional magnetic tips, where the polarization direction typically follows the external field (see Fig. R2-7 for a direct comparison). These data are now included as Extended Data Figure 5.



4. Given intrinsic altermagnetism of $\text{KV}_2\text{Se}_2\text{O}$, why this result depends on the $\sqrt{2} \times \sqrt{2}$ surface? What about the 2×1 surface?

Reply 2-4: We thank the referee for bringing this up. While both the $\sqrt{2} \times \sqrt{2}$ and 2×1 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 nearly six times larger than that of the $\sqrt{2} \times \sqrt{2}$ surface (~ 7 pm; see Fig. S14). This substantial height modulation tends to mask intrinsic electronic signatures on the 2×1 surface, such as impurity shapes, scattering patterns and quasiparticle excitations (Figs. R2-12a and b), compared to these on the $\sqrt{2} \times \sqrt{2}$ surface. The large modulation of 2×1 makes the details of impurity shapes, quasiparticle excitations and standing waves less obvious (Fig. R2-12a and b). Consequently, the resulting QPI patterns on this surface appear blurred, particularly at negative bias voltages (Figs. R2-12c–e).

Second, the intrinsic symmetry of the surface reconstruction should be compatible with the underlying electronic structure. The 2×1 reconstruction is intrinsically unidirectional and introduces a strong, extrinsic structural anisotropy along either the x or y axis. (Figs. R2-12c–e). Given that the momentum-space 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.

In the revised version, we have included the STM data on the 2×1 reconstructed surface in Supplementary Note 5 and added a brief discussion in the main text.

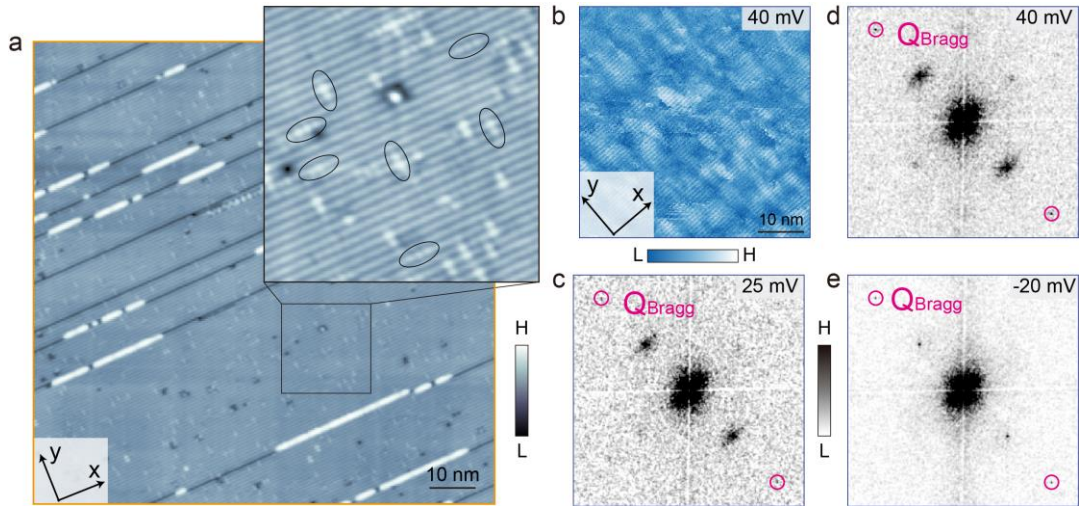


Fig. R2-12 **a**, topographic images of the 2×1 reconstructed surface, in a large field of view. The inset shows a zoom-in. **b**, dI/dV maps obtained on this kind of surface showing large modulation of the reconstruction. **c-e**, FFT of dI/dV maps at several energies.

5. The central claim of d-wave altermagnetism rests on the QPI analysis in Figure 5, specifically the rotation of the scattering intensity anisotropy when switching from positive to negative bias. However, a visual inspection of the FTs reveals that the anisotropy is extremely subtle. The peaks

in the line profiles are barely distinguishable from the background noise. Also, the filtered modulations are not surprising when choosing different FT spots. Consequently, the claimed anisotropy appears to rely heavily on aggressive data processing. The filtered modulations are susceptible to selection bias depending on the specific integration windows chosen in reciprocal space.

Reply 2-5: We appreciate the referee's critical evaluation of our QPI analysis. However, we respectfully disagree that the observed anisotropy is "extremely subtle" or an artifact of aggressive data processing. We provide the following evidence to substantiate the robustness of the scattering anisotropy.

First, the anisotropy is discernible in the raw data prior to extensive processing (Figs. R2-13a and c). To better illustrate the distinction between the q_x and q_y intensities, we have included line profiles extracted from the FFTs (Figs. R2-13b and d). By plotting the raw linecuts (R2-13b, d; upper panels) alongside the background-subtracted versions (R2-13b, d; lower panels), the contrast between the x and y directions becomes evident (the difference ratio is approximately 1/2 in these data). Importantly, the rotation of this intensity anisotropy upon switching the bias polarity is clearly resolved.

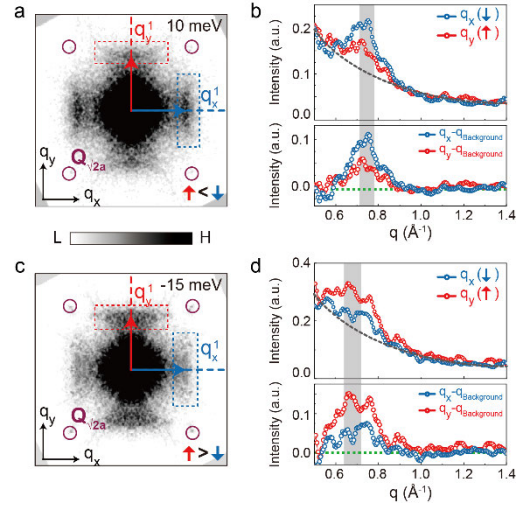


Fig. R2-13 a, c, QPI images at 10 and -15 meV. **b, d**, raw line profiles extracted from the FFTs (upper panels) and the background-subtracted versions (lower panels).

Second, we agree with the referee that the process of inverse FFT needs special caution to avoid artifacts.

We have performed a sensitivity analysis to confirm that our inverse FFT (IFFT) results are not biased by the choice of integration windows. As demonstrated in Figs. R2-14c–h, 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*

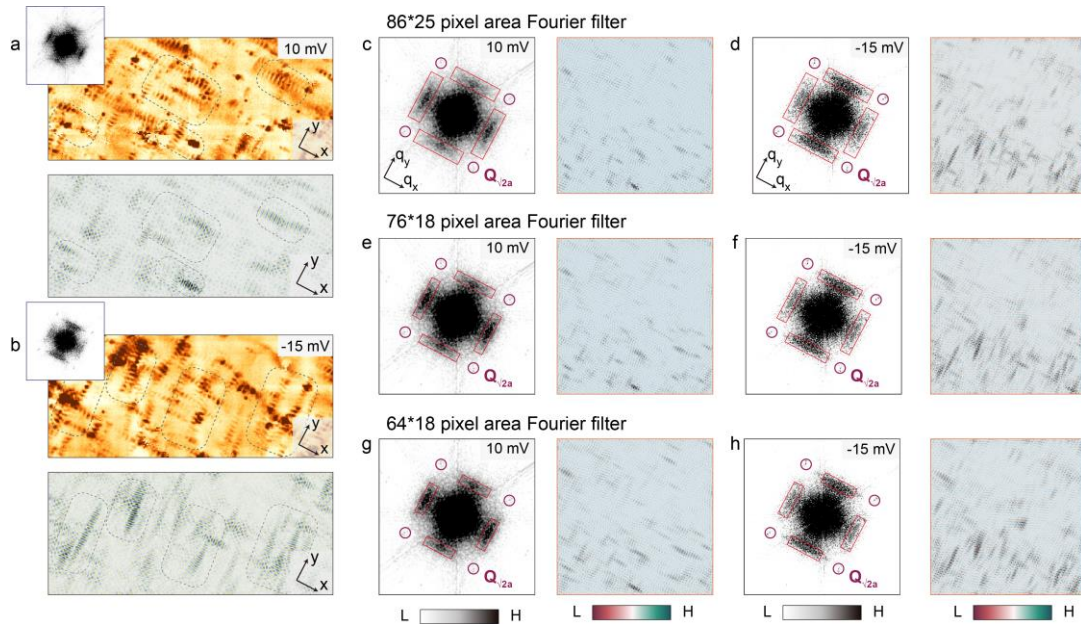


Fig. R2-14 a, b, raw dI/dV maps in the same field of view in a grid, at 10 and -15 meV. The lower panels are the IFFT data using a q-window of 86*25 pixels for q_{1x} and q_{1y}. **c-h**, IFFT process and the results with different q-space integration window size. Here symmetric windows with the same size and shape for q_{1x} and q_{1y} are used.

symmetric windows with the 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 Fig. R2-14a–b, this tendency 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, corroborating the IFFT analysis.

Third, the reproducibility of this effect has been verified across different samples and with different Bi₄Br₄ tips. In Fig. R2-15, 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.

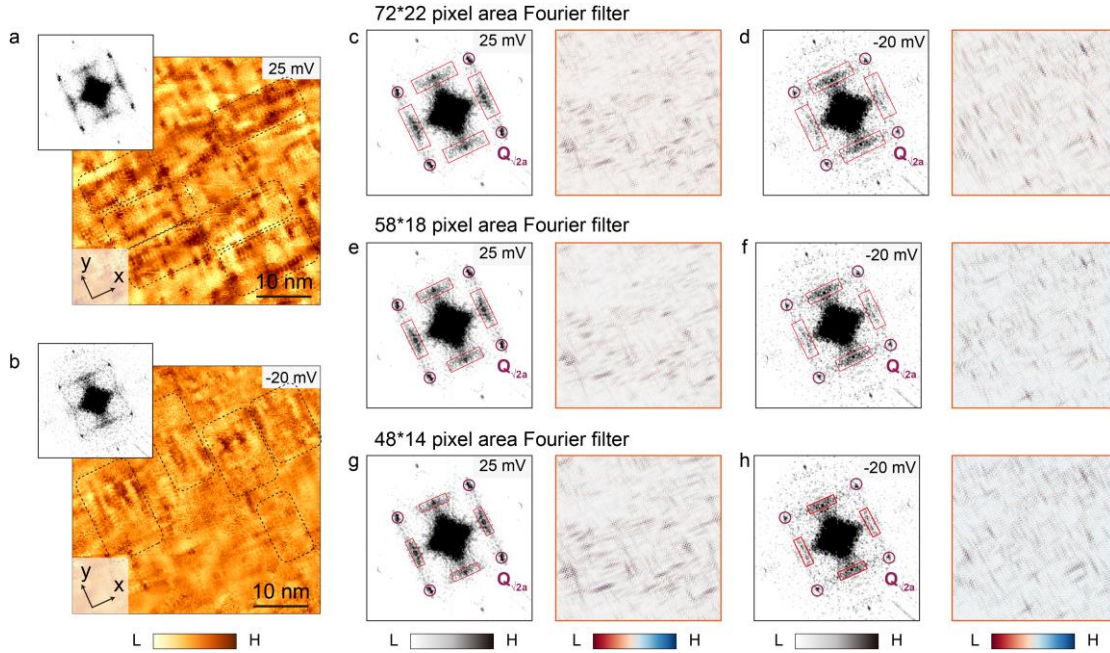


Fig. R2-15 a, b, raw dI/dV maps at 25 and -20 meV on a second sample with different Bi₄Br₄ tip. **c-h**, IFFT process and the results with different q-space integration window size. Here symmetric windows with the same size and shape for q_{1x} and q_{1y} are used.

Finally, we would like to emphasize the technical context of this study. These QPI data were acquired using unconventional STM tips fabricated from bulk quantum materials via FIB milling. This is a relatively nascent methodology in the STM community. We believe the signal-to-noise ratio and the internal consistency of the datasets presented here provide a rigorous basis for our conclusions.

In the revised version, we have added more analyses of the data in Supplementary Note 7 and added a brief discussion in main text and Methods.

6. I note that the intensities of the $\sqrt{2}a$ spots are very different between the +10 and -15 meV datasets. If the tunneling condition is stable, these structural peaks should be comparable. This large variation further deepens my skepticism about the results.

Reply 2-6: We thank the referee for this insightful observation. We have re-examined the data and found that the intensity of the $\sqrt{2}$ spots basically follow the energy dependence of the spatially averaged dI/dV spectrum in the grid measurement, as shown in Fig. R2-16c-d. 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 $\text{KV}_2\text{Se}_2\text{O}$, 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 Fig. R2-16a-b, this energy-dependent trend is consistently observed using both conventional PtIr tips and our Bi_4Br_4 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. In the revised version, we have added this analysis in Supplementary Note 7.

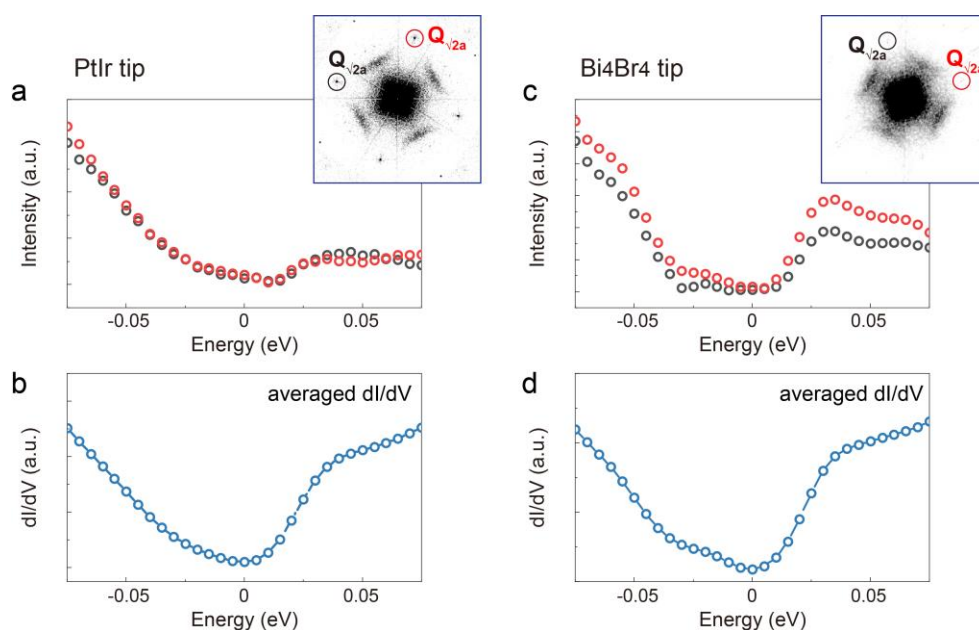


Fig. R2-16 **a**, the intensities of the two $\sqrt{2}$ spots in FFT as a function of energy, using PtIr tips. **b**, spatially averaged dI/dV spectrum of the measurement. One can see that the intensities of the $\sqrt{2}$ spots closely follow the energy dependence of the spatially averaged dI/dV spectrum. **c-d**, similar analysis for data taken with Bi_4Br_4 tip.

7. Even for the magnetic contrast map in Figure 5j,k, The mathematical procedure is highly susceptible to artifacts, particularly if the tip apex has any geometric asymmetry (which is guaranteed in a FIB-fabricated tip). Since the orbital asymmetries in the tip apex are inevitable, which is expected for a tip made of a low-symmetry crystal like Bi_4Br_4 , the tip's orbital transmission coefficients will be anisotropic. Reversing the bias polarity probes tunneling into versus out of these anisotropic tip orbitals. This change in tunneling matrix elements can naturally

mimic a rotation in the QPI pattern, even in the absence of magnetic scattering. The authors have not provided a robust method to distinguish genuine spin-flip scattering from these inevitable orbital-dependent tunneling effects.

Reply 2-7: We thank the referee for raising this critical and highly constructive point. Distinguishing genuine spin-resolved tunneling from potential artifacts arising from tip geometric asymmetry, orbital anisotropy, or bias-dependent tunneling matrix elements is indeed paramount to establishing the fidelity of our topological probes. We address this important concern from both theoretical and experimental perspectives below.

DFT calculations of the tip edge-state orbitals. To evaluate the spatial profile of the tunneling matrix elements, we calculated the orbital characteristics of the helical edge states in α -Bi₄Br₄ (summarized in Fig. R2-17). For edge channels propagating along the chain direction, the electronic states are dominated by the Bi- p_x orbital (defined within the Bi₄Br₄ crystal coordinate system); for the edge channels running along the lateral section, the dominant contributions originate from both Bi- p_x and Bi- p_y orbitals (defined within the Bi₄Br₄ crystal coordinate system). In the STM configuration, the Bi- p_y orbital projects outward from the crystal, aligning nearly vertically (with a minor tilt angle of ~ 10 degrees) to the sample surface. Consequently, this orbital acts effectively as a symmetric p_z -like probe relative to the sample. Such a spatial orientation is highly unlikely to introduce a significant difference in the tunneling matrix elements into the sample's in-plane d_{xz} and d_{yz} orbitals, thereby minimizing spurious, tip-induced orbital anisotropy.

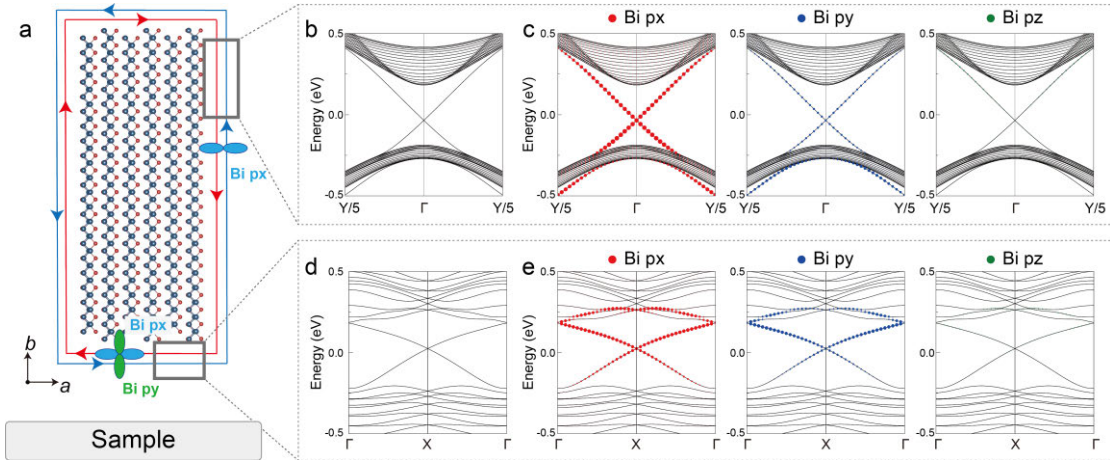


Fig. R2-17 **a**, Sketch for the tunnelling junction in which the orientation of helical edge channels and their orbital characteristics are included, the Bi- p_y orbital lobes extend outward from the tip apex, and consequently, the tip effectively acts as a “ p_z ”-like orbital probe relative to the sample plane. **b-e**, calculated overall dispersion and the orbital characteristics of the helical edge state in Bi₄Br₄, propagating along the atomic chains (**b,c**) and residing on the lateral facets (**d,e**).

Systematic consistency across diverse magnetic compounds. The spin-resolved imaging capability of the Bi₄Br₄ tips has been systematically verified across four distinct classes of magnetic compounds hosting vastly different magnetic structures Fe_{1+x}Te, KV₂Se₂O, FeSn and FeGe. The measured corresponding magnetic contrasts in all four systems strongly argue against a coincidental artifact originating from the tip's orbital or geometric asymmetry. Particularly in the kagome

systems FeSn and FeGe, which exhibit A-type AFM order (Figs. R2-5 and R2-6), the spin-dependent DOS contrast is imaged between adjacent layers separated by a single unit-cell height. Because these two termination layers are structurally and orbitally equivalent (differing only in their spin orientations), any purely geometric or orbital-dependent tunneling matrix effects are intrinsically canceled out.

In-situ magnetic-field-induced spin flop. The most direct evidence ruling out orbital-dependent artifacts comes from our in situ magnetic-field control experiments on FeGe (Fig. R2-6). At low fields, the spin alignment of FeGe is predominantly out-of-plane, which is orthogonal to the in-plane spin orientation of the tip, resulting in a minimal DOS contrast between adjacent layers. Upon applying an out-of-plane magnetic field of 8T to flop the sample spins into the in-plane direction, we observed a pronounced enhancement of the layer-by-layer contrast. Crucially, when the field is reduced back to ± 1 T, the contrast diminishes again. Throughout this process, the measurements were performed on the identical sample area using the same tip apex. The reversible change in contrast must originate mainly from the magnetic orientation of the sample.

Structural tip asymmetry and QPI rotation. Next, we turn to the anisotropy of the tip apex, which is very typical and hard to control in STM measurements. Because such anisotropy is rooted in the shape of the STM tip, *it will not change its direction at different bias voltages*. **In all our three datasets using different PtIr tips, such anisotropy indeed does not rotate when switching the bias polarity (Fig. R2-18a-c.).** For the Bi_4Br_4 tip, the spin-texture of electronic state is revealed by the tunneling of helical edge states, and electrons with opposite spins can be imaged at opposite bias voltages. Therefore, the observed anisotropy in the QPI images rotates $\pi/2$ when switching the bias

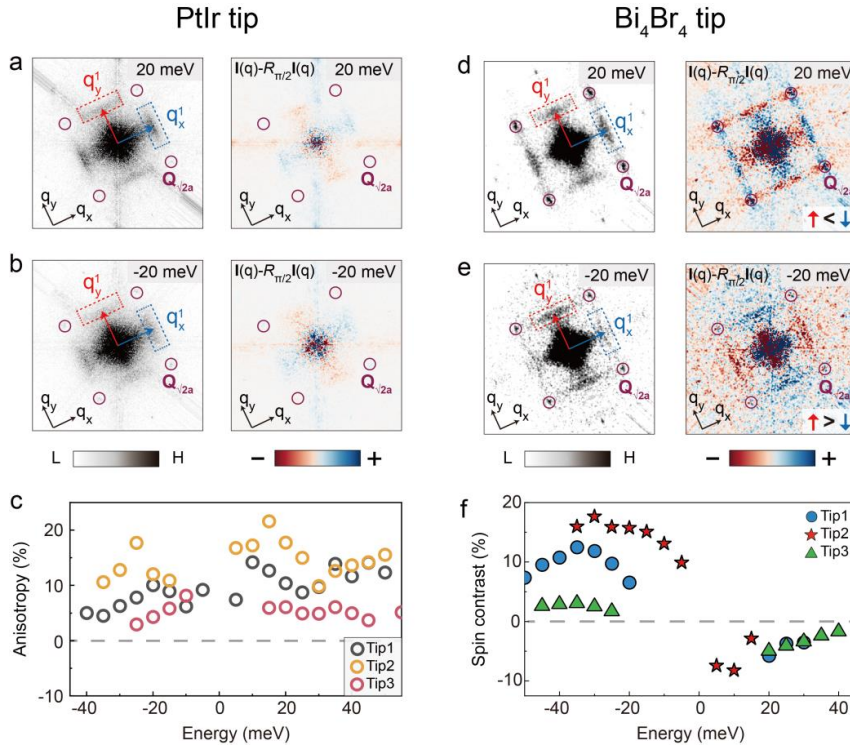


Fig. R2-18 a-b, dI/dV maps and the corresponding contrast map of rotating, at 20 (a) and -20 meV (b) with PtIr tips. **c,** the anisotropy ratio between q_x and q_y as a function of energy, without sign change for all three different PtIr tips. **d-f,** dataset acquired with Bi_4Br_4 tips, which collectively show that the anisotropy axis rotates for opposite bias voltages.

polarity, demonstrated across three independent datasets in Fig. R2-18d-f. This evolution is totally different from that with the non-magnetic PtIr tips, and reflects the spin-texture of electronic states along x- and y- direction (**which has been cross-checked by the magnetic-field induced Zeeman shifts**, see our improved data in Reply 2-9).

We believe that this comprehensive combination of DFT calculations, multi-system replication, in situ magnetic field control, and comparison with conventional tips provides a reliable foundation that distinguishes true spin-resolved tunneling from orbital-dependent artifacts. These are challenging STM experiments and we believe the results can significantly advance the field of using STM tips made of topological materials.

In the revised manuscript, we have included the control experiments using PtIr tips in Extended Data Fig. 10, the calculated orbital feature of Bi₄Br₄ in Extended Data Fig. 1, the control experiments of FeGe in Extended Data Fig. 5, and added related discussion in the main text (page 7) and more detailed information in Methods.

8. Since there are two types of Bi₄Br₄ tips: one with band gap and the other one with helical-edge-mode. The authors should compare these two results on the same KV₂Se₂O surfaces and defects. A side-by-side QPI comparison is definitely essential.

Reply 2-8: We thank the referee for this suggestion. However, we would like to clarify the technical challenges that render a systematic study using "bulk-state" tips on the KV₂Se₂O surface unfeasible at this stage. In our experimental experience, the bulk-state tip (i.e., showing a large semiconducting gap On Au (111)) is less stable for tunneling and it frequently crashed on the surface of KV₂Se₂O during the approaching. This stands in contrast to the "helical-edge-mode" tips, which possess a more robust atomic configuration at the apex that facilitates stable tunneling on these complex magnetic surfaces.

We interpret the referee's underlying concern as a requirement to distinguish the spin-resolved signatures of the helical edge states from those of spin-degenerate states. To address this, we have provided a comparison between results using the conventional PtIr tips and spin-resolved Bi₄Br₄ tips (see Fig. R2-18), where the PtIr tips serve as a spin-degenerate reference. Given that both the bulk state of Bi₄Br₄ and PtIr are non-magnetic and spin-degenerate, this comparison provides useful information to attribute the observed anisotropy rotation to the unique helical nature of the Bi₄Br₄ edge states.

9. The analysis of Zeeman shifts in Figure 4 relies heavily on arrow-shaped defects of V-site vacancies. However, the tunneling spectra exhibit many unidentified in-gap peaks. The authors appear to select only the specific peaks that satisfy the expected Zeeman splitting relations while ignoring others. Such an arbitrary data selection process is scientifically unsound and unacceptable.

Reply 2-9: We take the referee's concern very seriously and appreciate the opportunity to clarify our analysis. We acknowledge that in our initial datasets, a high concentration of V-site vacancies

resulted in spatially overlapping in-gap states, as these states typically extend over several nanometers. Such overlaps can indeed lead to complex spectra with multiple peaks, making the assignment of specific Zeeman shifts challenging.

To provide a more rigorous and unambiguous verification, we have performed an extensive search for regions with significantly lower defect concentrations. This allowed us to identify and measure isolated V-site vacancies, effectively eliminating the spectral complexity induced by multiple nearby defects. The new spectroscopic data are presented in Fig. R2-19. In this cleaner environment, the in-gap states clearly and collectively exhibit Zeeman shifts under an external magnetic field. Specifically, the energy shifts observed for states associated with the X- and Y- sublattices are opposite in sign. This collective behavior of the entire spectral set (rather than selected peaks) provides direct and robust evidence of the underlying spin texture. These findings also serve as a decisive cross-check that corroborates our QPI analysis.

In the revised manuscript, we have updated these data in Figure 4 and added more data in Supplementary Note 6.

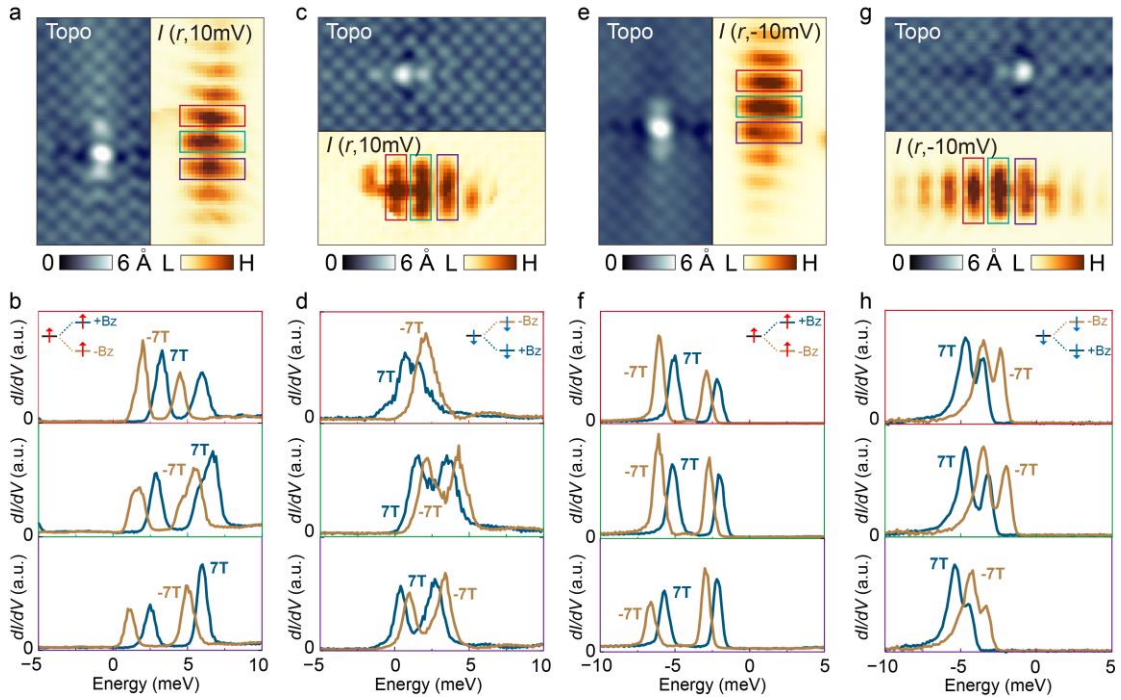


Fig. R2-19 **a, e**, topographies of two V-site defects that induced in-gap states along the y direction. since there could be multiple in-gap states peaked at different energies, here a current map at 10meV is displayed to show the spatial feature of these in-gap state clearly. **b, f**, Magnetic-field-induced Zeeman shifts for in-gap states along y- direction. The spectra are averaged for each colored box shown in **a** and **e**. **c, g** and **d, h**, similar data for in-gap states along the x direction, showing opposite Zeeman shifts under fields.

10. Since the surface is rich in Vanadium, it is a common occurrence in STM for the tip to pick up a V atom or cluster during scanning or pulsing. This would instantly create a standard, albeit weak, spin-polarized STM tip. The authors have not effectively ruled out the possibility that their helical contrast is simply conventional spin-polarized tunneling from a V-contaminated apex. Given the similar effective spin polarization ($\sim 12\%$) as previously reports by spin-polarized STM

measurements, accidental functionalization seems a plausible alternative explanation.

Reply 2-10: We thank the referee for this insightful comment. The possibility of accidental tip picking up magnetic adatoms is a critical consideration. For $\text{KV}_2\text{Se}_2\text{O}$, the cleavage occurs between two adjacent $\text{V}_2\text{Se}_2\text{O}$ sharing a potassium plane. Thus, the resulting surface is typically disordered and dominated by randomly distributed K adatoms (even for low-temperature cleaving; see Fig. R2-20), while the V atoms remain sequestered within the lower $\text{V}_2\text{Se}_2\text{O}$ layer. Throughout our extensive measurements, we have observed no evidence of stable V-atom pickup during routine scanning with both PtIr tips and Bi_4Br_4 tips. Importantly, the rotation of anisotropy in the QPI intensities when switching the bias polarity is unique for the Bi_4Br_4 tip due to the helical tunneling. In contrast, a conventional ferromagnetically polarized tip would exhibit a fixed spin-polarization direction at low bias, resulting in an anisotropy that does not rotate with bias polarity, a fact recently corroborated by independent studies using Cr tips (arXiv:2604.18337).

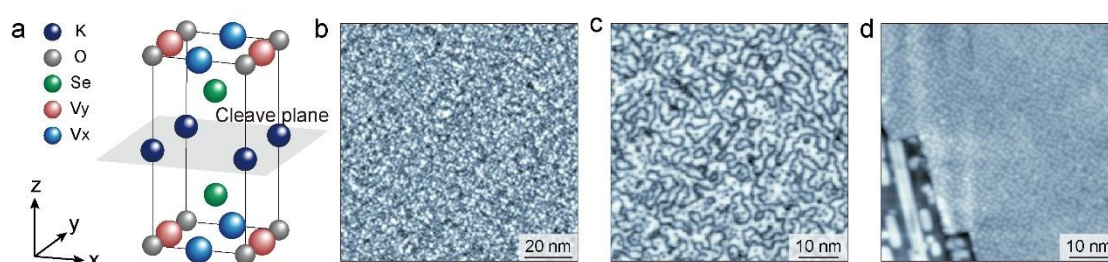


Fig. R2-20 **a**, crystalline structure of $\text{KV}_2\text{Se}_2\text{O}$. **b-d**, topographic images showing the randomly distributed K adatoms, after cleaving at $\sim 30\text{K}$.

Our experiments on four different types of magnetic compounds (Fe_{1+x}Te , $\text{KV}_2\text{Se}_2\text{O}$, FeSn and FeGe) demonstrate that **Bi_4Br_4 tips have intrinsic spin resolution**. Moreover, comparative experiments with Fe-coated tips reveal fundamentally different response behaviors. We have conducted systematic comparisons on two benchmark systems. In Fe_{1+x}Te , a contrast reversal in the dI/dV maps is consistently observed with Bi_4Br_4 tips upon switching bias polarity, yet this reversal is conspicuously absent when using conventional Fe-coated magnetic tips (Fig. R2-3). In FeGe , when using a Bi_4Br_4 tip, the dI/dV contrast between adjacent unit cells is significantly enhanced only after a field 8 T is applied to flop the FeGe spins into the in-plane direction (Fig. R2-6). Conversely, with Fe-coated tips, the contrast is clearly observed at small fields (± 1 T) sufficient to polarize the tip but insufficient to flop the sample spins, and the contrast reversal when changing the field direction (Fig. R2-7). The consistent results demonstrate that the spin-resolved imaging is an intrinsic property of the Bi_4Br_4 topological edge states rather than a result of accidental tip contamination.

11. The interpretation that the arrow-shaped defects reveal the intrinsic magnetic ground state is debatable. It is equally plausible that the electronic structure in the vicinity of the vacancy is dominated by the strong impurity potential itself. This could induce a local symmetry breaking (local magnetic moment formation or orbital ordering) that does not necessarily reflect the long-range antiferromagnetic order of the pristine crystal. Distinguishing between defect-induced physics and bulk antiferromagnetism in such a disordered landscape requires more rigorous validation.

Reply 2-11: We thank the referee for this insightful comment. We acknowledge the importance of

distinguishing intrinsic long-range order from local, impurity-induced symmetry breaking. However, our findings suggest that while the impurity potential localizes electronic states, their spatial and spin characteristics are fundamentally dictated by the bulk altermagnetism. We address this concern through the following two perspectives:

First, the spatial correlation of these arrow-shaped defects (left, right or up, down) strictly follows the same periodicity regardless the surface domains. If the "arrow-shaped" features were solely driven by local impurity potentials, one would expect their orientations to be randomly distributed. However, as shown in the large-scale topography (60 nm by 60 nm, Fig. R2-21), the orientation of every observed V-site vacancy strictly adheres to the $\sqrt{2} \times \sqrt{2}$ density wave modulation. This rigorous spatial correlation across the entire field of view (independent of surface reconstruction domains) provides evidence that these defects act as local probes for a robust, long-range electronic order rather than being independent sources of symmetry breaking.

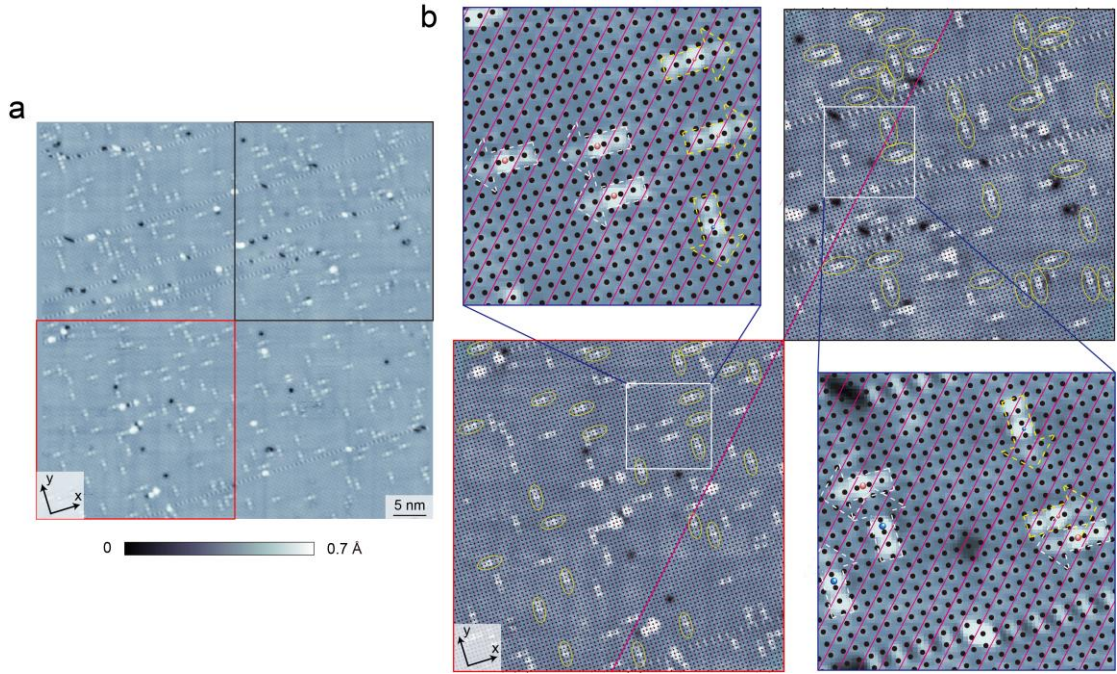


Fig. R2-21 **a**, topographic image of the reconstructed surface with several surface domains. **b**, the long-range spatial correlation of the arrow-shaped impurities' orientations. The pink lines track one type of the two non-equivalent V sites in the $\sqrt{2} \times \sqrt{2}$ density wave state, and all defects on these lines have the same orientation for X and Y sublattices, regardless the surface domain.

Second, the spin texture of the impurity-induced states exhibits a collective Zeeman response consistent with altermagnetic symmetry. Our DFT calculations (Fig. R2-22) indicate that V-site vacancies on the X and Y sublattices possess local moments of approximately $-2\mu_B$ and $+2\mu_B$, respectively, in the AFM state, whereas vacancies at non-magnetic oxygen sites exhibit negligible moments. Experimentally, we systematically characterized the Zeeman shifts of in-gap states for over ten isolated V-site defects. *Crucially, without exception, the electronic modulations along the x and y directions consistently exhibit opposite Zeeman shifts. This coherent spin-texture persists even for defects separated by over 100 nm.* This combined information indicates that the spin texture of these impurity states is also globally correlated in real-space that reflects the underlying AFM order.

Such global correlation in both real and spin space is highly unlikely to arise from isolated impurity physics and instead points to the underlying AFM sublattice symmetry as the primary driver. In summary, the defects do not create the observed symmetry breaking; rather, they pin and reflect the intrinsic bulk antiferromagnetic order through their globally correlated responses.

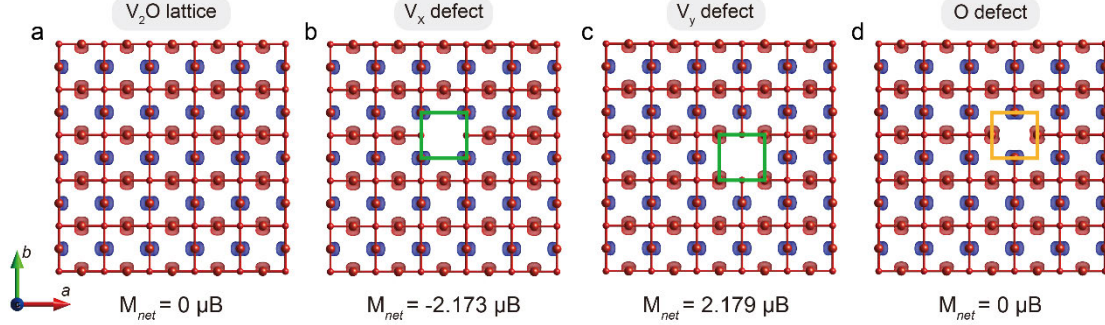


Fig. R2-22 Calculated net magnetization of the V_2O lattice in the AFM phase. **a**, net magnetization is zero without any defect. **b**, net magnetization of approximately $-2.17\mu_B$ with a V-site vacancy on the X sublattice. **c**, net magnetization of approximately $2.17\mu_B$ with a V-site vacancy on the Y sublattice. **d**, net magnetization of zero with an O-site vacancy.

In the revised manuscript, we have modified Extended Data Figure 7 to highlight the long-range correlation of these defects, added a large field of view image in Fig. S10, added more defects data in Supplementary Note 6 and modified the wording in the main text.

12. The effective polarization ratio shown in Figure 5l exhibits a significant asymmetry between positive and negative bias for a single tip. This is physically inconsistent with the authors' model. Antiferromagnetism is characterized by spin splitting that typically spans a large energy range (often on the eV scale). Consequently, the spin-dependent contrast should be robust and relatively constant over a wide bias window. However, the contrast reversal reported here appears highly sensitive to the specific bias voltage selected. This strong energy dependence suggests the contrast arises from energy-dependent orbital artifacts or density-of-states features rather than the broad-band spin-momentum locking of a helical edge mode.

Reply 2-12: We thank the referee for this critical and fundamental question regarding the energy dependence of the effective spin-polarization ratio.

First, we address the asymmetry and energy sensitivity from a measurement perspective. Our analysis relies on the measured QPI patterns. In QPI measurements, the coherent scattering intensity of quasiparticles typically reduces rapidly at higher energies due to the reduced quasiparticle lifetime τ . Unlike ARPES, which directly probes the band structure, QPI signatures in many quantum materials are only discernible within a narrow energy window near E_F . Once the signal-to-noise ratio of the QPI patterns drops, deriving the spin-contrast ratio becomes difficult.

Second, any residual geometric asymmetry of the tip apex (as noted for PtIr tips in Fig. R2-18) contributes a static background anisotropy. While this background is bias-independent in orientation, its magnitude can vary with energy (see Fig. R2-18c; this is true for conventional PtIr tips), potentially leading to the additional asymmetry between positive and negative bias.

These are two direct reasons that can address the concerns regarding the energy dependence of the effective spin-polarization ratio.

Moreover, there are two fundamental factors contributing to the reduction in the spin-polarization ratio: the spin directions (see above) and the inherent presence of two counter-propagating edge channels with opposite spins, which form closed loops circulating along the tip boundaries. 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.

First, the observed spectral line shape of the Bi_4Br_4 tip on Au(111) surface (Extended Data Fig. 3a–d) 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 ($> 5\text{nm}$) 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. R2-23). 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.

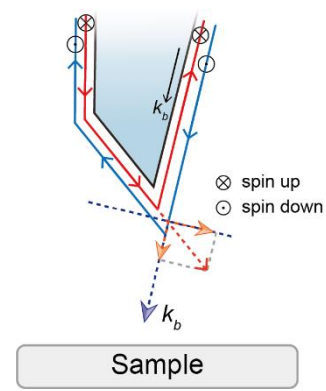


Fig. R2-23. quasi-momentum description of the tunneling process.

Second, if we consider the tip apex as a point-like object, the 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. R2-24a). 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.

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. R2-24b-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.

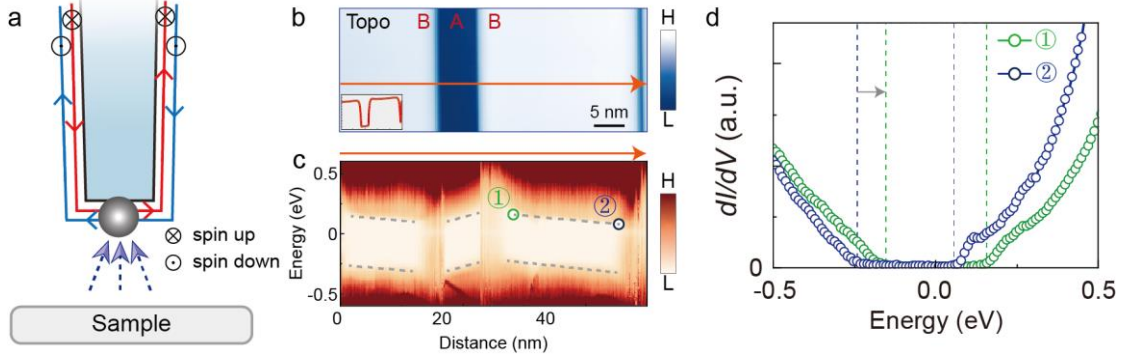


Fig. R2-24 **a**, Sketch for the tunnelling condition with a point-like tip apex. **b**, STM topography including one and a half terraces of monolayer Bi_4Br_4 . **c**, Linecut profile of dI/dV spectra across these terraces. A pronounced band-bending effect near the step edges can be observed. Interestingly, 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.

b) Nonlinear Current-Voltage Response. Phenomenologically, the relevant dI/dV signal or equivalently the local DOS $\rho(V)$ exhibits a component that is an odd function of the applied bias voltage V . This implies to a second-order current response of the form

$$J = \sigma(N)V^2,$$

where $\sigma(N)$ is the second-order nonlinear conductivity coefficient that depends on the local Neel order N . We require that $\sigma(N) = -\sigma(-N)$. As a result, at two magnetic sites with opposite spin orientations, $\rho(V)$ has different magnitude, or equivalently, $\rho(V) - \rho(-V)$ has opposite signs. This represents the spatial resolution of the local spin order. *Such second-order current can be viewed as a ballistic version of the nonlinear Hall effect*. Based on previous study on the nonlinear Hall effect, the intrinsic contribution from the quantum metric dipole can indeed couple to and capture the antiferromagnetic spin order. We note that extrinsic contributions such as skew scattering and side jump can also contribute.

Finally, the supplementary control experiments on Fe_{1+x}Te (Figure R2-4) and FeGe (Figure R2-6) strongly indicate that the Bi_4Br_4 tip is more sensitive to the in-plane spin configuration of the substrate. Nevertheless, the tip retains the capability to resolve the out-of-plane spin contrast of DOS. This arises from an unavoidable geometric tilt (approximately 10°) of the tip relative to the surface normal in our experimental setup.

As shown in Fig. R2-25b, during the FIB process, the PtIr wire supporting the Bi_4Br_4 tip must be bent by 90° to accommodate the holder geometry within the FIB-SEM system. After the FIB processing, we bent the PtIr wire back with tweezers to make it vertical to fit the STM tip holder. However, due to the mechanical rigidity and short length of the wire, a residual macroscopic tilt

angle of approximately 10° is practically inevitable (Fig. R2-25b). Because the Bi_4Br_4 stripe is mounted strictly parallel to the distal end of the PtIr wire, it consequently inherits this $\sim 10^\circ$ deviation from the surface normal of the sample. Consequently, the spin polarization of the helical edge modes is not strictly orthogonal to the out-of-plane spins of the substrate. This geometric projection yields a finite, overlapping spin component, allowing for the detection of an effective spin contrast in the dI/dV measurements (similar to TMR). *This could be another reason for the reduction of effective spin-polarization ratio.*

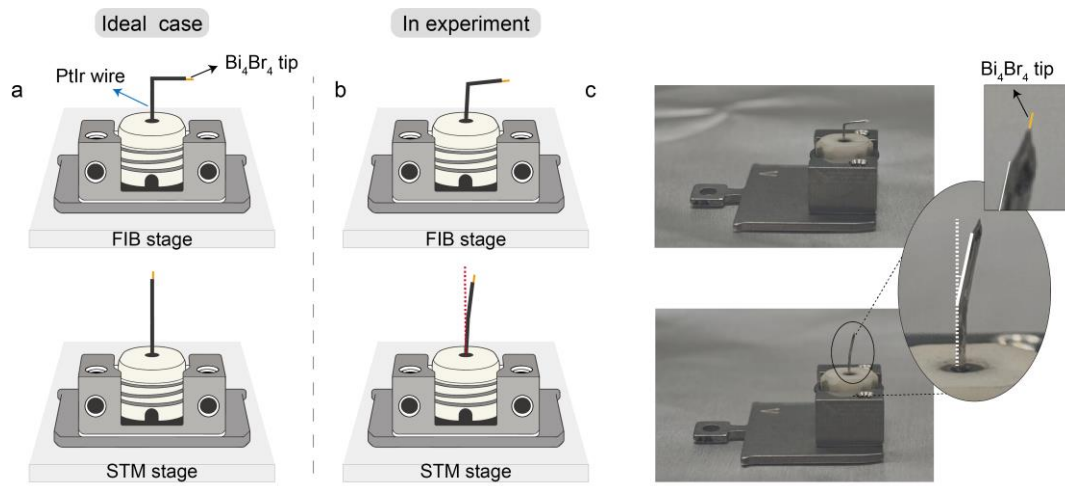


Fig. R2-25 a, Sketch showing the ideal case to prepare the Bi_4Br_4 tips, in which the tip is strictly vertical to the surface of sample. **b**, Sketch showing the experimental process in which there is an unavoidable geometric tilt (approximately 10°) of the tip relative to the surface normal. **c**, Photos to show the tilting of the tip. The Bi_4Br_4 stripe is strictly parallel to the end of PtIr wire.

We agree 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.

In the updated version, we have added a brief discussion in the main text (Discussion Section; page 8) and included these detailed discussion in Supplementary Note 10.

13. The organization of Figure 5 is chaotic. The current arrangement makes it extremely difficult for the reader to follow up the logical progression.

Reply 2-13: We sincerely thank the referee for this critical feedback regarding the presentation of Figure 5. In the revised manuscript, we have reorganized Figure 5 to more effectively communicate the core findings of our QPI analysis.

Overall Reply: We sincerely thank the referee for the constructive comments and critical insights, which have been instrumental in significantly improving the quality and clarity of our manuscript. By incorporating extensive new STM data across a broader range of magnetic compounds and performing additional systematically designed control experiments, we have established a more

coherent and rigorous physical framework for our proposed methodology. These enhancements further substantiate our findings and reinforce the robustness of our study on altermagnetism.

Our primary objective throughout this revision has been to ensure the solidity of our results and to provide a clear, evidence-based elucidation of the underlying phenomena. We believe that this methodology, along with the physics it reveals, represents a substantial advancement in the fields of topological materials, scanning tunneling microscopy, and the emerging physics of altermagnetism.

Response to Referee #3

Summary of the key results

The manuscript "Atomic-scale spin sensing of a 2D d-wave altermagnet via helical tunneling" by Wang and co-workers describes spin-resolved scanning tunneling microscopy (STM) and spectroscopy (STS) measurements as well as quasiparticle interference (QPI) data obtained with conventional PtIr and novel Bi₄Br₄ tips on cleavage planes of KV₂Se₂O. After an introductory paragraph, the manuscript discusses two major results: (a) the utilization of edge states of the higher-order topological insulator (HOTI) Bi₄Br₄ for spin-resolved measurements and (b) the investigation of KV₂Se₂O in real and reciprocal space.

In (a) the successful application is convincingly demonstrated by "benchmarking" experiments on the well-known antiferromagnet FeTe. The observation of a characteristic striped pattern caused by spin-polarized tunneling is beyond doubt, even though a few details regarding the presentation of the data might be improved (see list of comments below).

In contrary, as pointed out below I am not fully convinced of the data presented in the context of part (b).

Originality and significance: if not novel, please include reference

As stated above, I largely convinced by the data presented in part (a) for Bi₄Br₄-functionalized tips. However, a very similar experimental concept has already been demonstrated by Aishwarya et al. in Ref. 62. Therefore, I do not understand why the authors decided to include part (a) in the main manuscript rather than describing it in either the extended data or the supplemental material.

Data & methodology: validity of approach, quality of data, quality of presentation

Overall, the data are of very high quality and the general approach of investigating the spin state of a potential layered d-wave altermagnet with a Bi₄Br₄-functionalized tip is definitely worth to be pursued. However, I do have some concerns regarding some details of the data analysis and the conclusions drawn (see below). It is quite possible that the authors will be able to refute the criticism expressed with appropriate arguments.

Reply: We sincerely thank the referee for the positive assessment of our data quality and for the constructive feedback regarding the manuscript's structure and the contextualization of our work within the field.

First of all, we fully acknowledge the pioneering contribution of Aishwarya et al. (Ref. 62), who first demonstrated the concept of spin-resolved tunneling using the topological surface states of SmB₆ nanowires. In the revised manuscript, we have updated the text to ensure that this seminal work is appropriately credited, explicitly highlighting its foundational role in developing topological STM probes.

While we respect the referee's suggestion to move the characterization of Bi₄Br₄ tips to the Supplemental Information, we believe that keeping this section in the main text is essential for the following reasons. First, based on our interactions with the STM community, there is a strong demand for a detailed elucidation of how these unconventional tips are fabricated and characterized.

Presenting this in the main text ensures that the methodology is clear before we apply the technique to the complex case of altermagnetism. Second, compared to the topological Kondo insulator SmB_6 , $\alpha\text{-Bi}_4\text{Br}_4$ is a van der Waals material with a large bulk gap ($\sim 0.28\text{eV}$) and robust helical edge states; This provides a more predictable platform for topological tunneling. It is important to demonstrate a second, distinct topological platform to realize helical tunneling besides SmB_6 and to advance this field. Moreover, since our initial submission, we have acquired significant new datasets that advance the fundamental understanding of these relatively new probes. This includes a systematic study of four distinct magnetic structures and several control experiments. These results delineate that the Bi_4Br_4 tip is more sensitive to magnetic structures with in-plane spin orientations, a technical detail not presented in the pioneering SmB_6 work. We list the new experiments (on the basis of SmB_6 tip) in the following.

The spin orientation of edge-state electrons from theoretical perspective. $\alpha\text{-Bi}_4\text{Br}_4$ falls within the classification of a 3D QSHI, exhibiting a bulk spin polarization along the S_z direction (nearly quantized as $\pm 0.97 \times \pm \hbar/2$). Our calculations indicate that the edge-state electrons also mainly possess an out-of-plane spin orientation ($S_z \sim 0.85 \times \pm \hbar/2$; see Fig. R3-1). In the geometry of the STM measurement, this spin orientation lies “in-plane” with respect to the target sample.

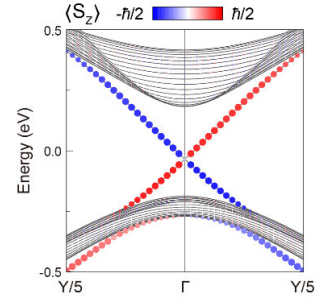


Fig.R3-1 S_z as the major spin orientation for the edge states in Bi_4Br_4

To gain a more comprehensive understanding of its spin-polarization capability, we have measured four distinct classes of compounds with different magnetic structures: Fe_{1+x}Te , kagome magnet FeSn , FeGe and finally $\text{KV}_2\text{Se}_2\text{O}$. We also performed three types of control experiments (under magnetic fields, rotated the sample and using conventional magnetic tips and PtIr tips). **This systematic study demonstrates that the spin polarization of the Bi_4Br_4 tip is intrinsic and more sensitive to in-plane spin components, and further verifies its distinct characteristics relative to a conventional magnetic tip.** In the following, we describe several key experimental findings in detail.

1. Fe_{1+x}Te with bicollinear AFM order.

A contrast reversal at opposite bias voltages using Bi_4Br_4 tips. We confirm that the spin-polarized local DOS (with the bicollinear AFM order) *undergoes a contrast reversal at opposite bias voltages using Bi_4Br_4 tips* (the additional kink at negative biases corresponds to atomic modulations), which can be observed in *the dI/dV maps and our newly acquired topographic images for a large energy range* (Fig. R3-2).

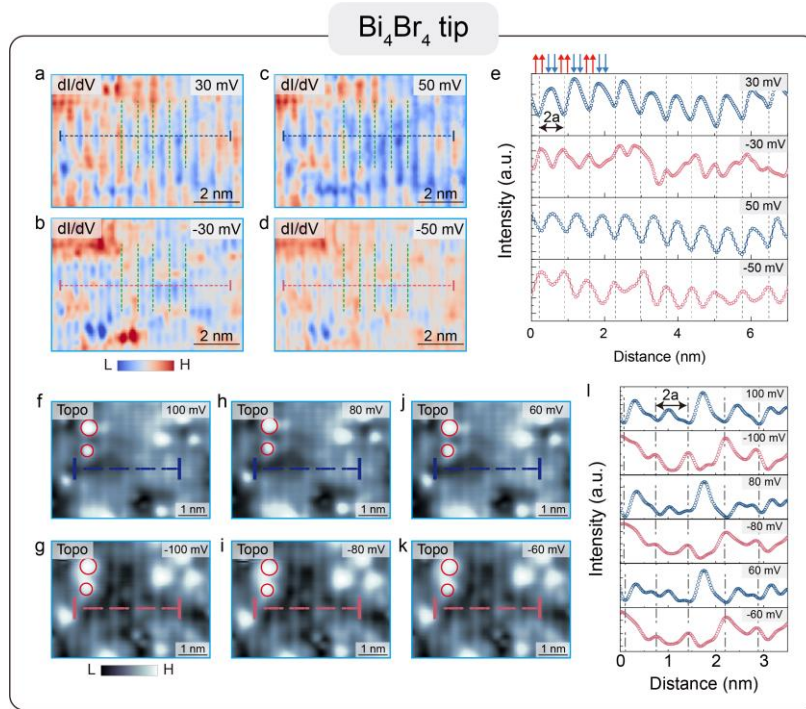


Fig. R3-2 Bicollinear AFM order in Fe_{1+x}Te imaged by the Bi₄Br₄ tips. **a-d**, dI/dV maps and **f-k**, topographic images. In both cases, the spin-polarized local DOS of **2a** modulation exhibits a π -phase shift (contrast reversal) at opposite bias voltages, as shown in **e** and **l**. To show the overall tendency, the linecuts in **e** and **l** are averaged over 30 and 20 pixels perpendicular to the linecut direction, respectively.

- **Such contrast reversal is absent when using a conventional magnetic tip.** We performed spin-polarized LDOS measurements using *Fe-coated PtIr tips*. Although signatures of the bicollinear AFM order could be still observed, with these conventional magnetic tips we found that the contrast reversal was absent upon reversing the bias-voltage polarity (Fig. R3-3). This comparison indicates that the contrast reversal at opposite biases for the Bi₄Br₄ tip should originate from the unique spin selection rule of the helical tunneling.

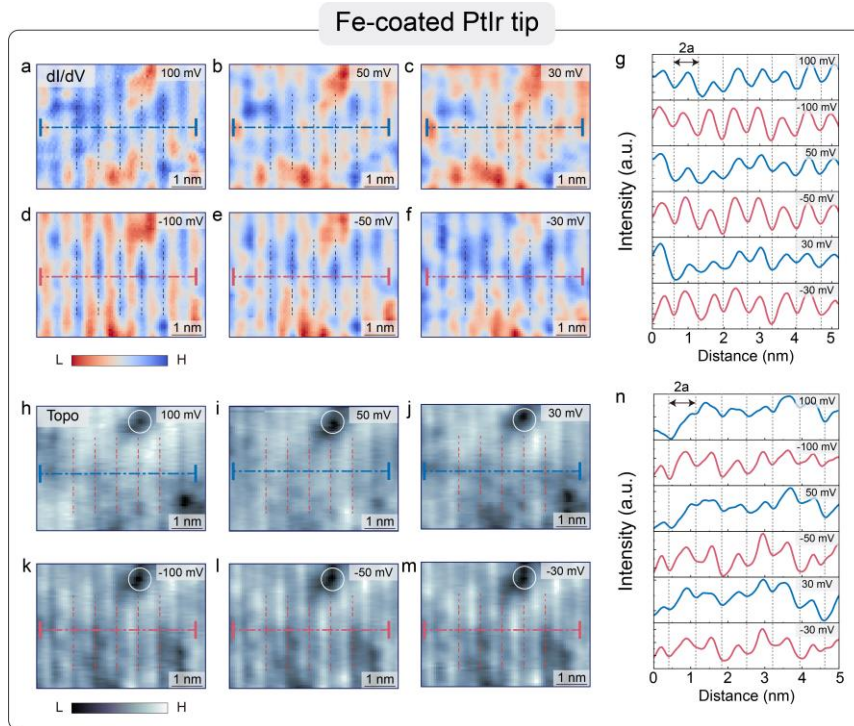


Fig. R3-3 Bicollinear AFM order in Fe_{1+x}Te imaged by the Fe-coated PtIr tips. **a-f**, dI/dV maps and **h-m**, topographic images. In both cases, the spin-polarized local DOS of **2a** modulation exhibit a same phase (without contrast reversal) at opposite bias voltages. To show the overall tendency, the linecuts in **g** and **n** are averaged over 30 pixels perpendicular to the linecut direction, and no contrast reversal is observed.



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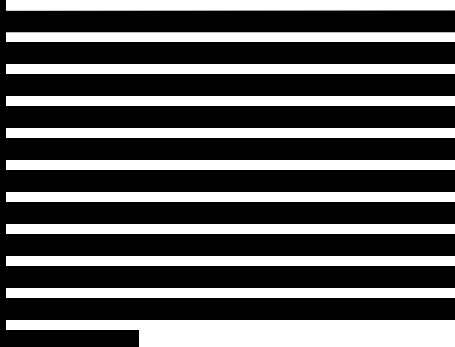
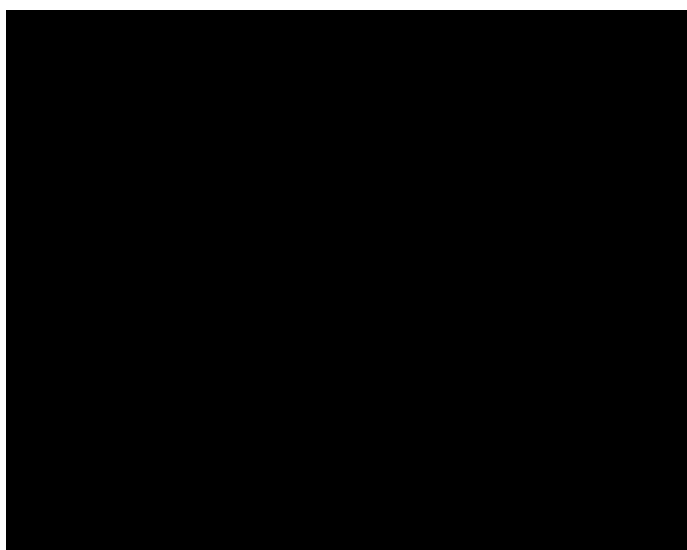
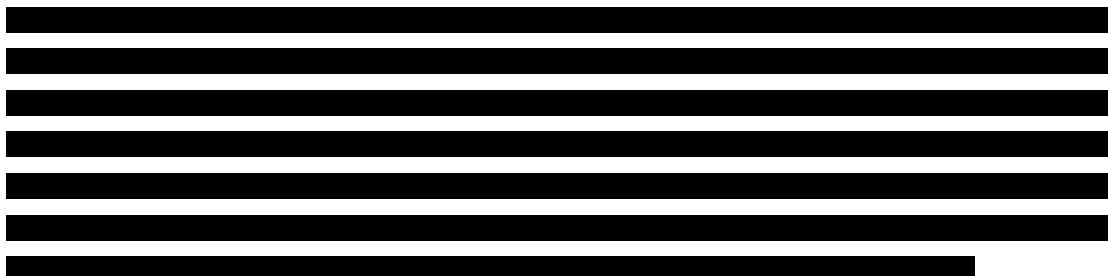
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These experiments in Fe_{1+x}Te demonstrate **the intrinsic difference between Bi_4Br_4 tips and magnetic tips**, suggests that the orbital feature of Fe_{1+x}Te is unlikely to be the origin for the contrast reversal at opposite bias voltages seen by Bi_4Br_4 tips, and further suggest that the spin orientation on the Bi_4Br_4 tips more likely lies in one “in-plane” direction with respect to the target sample.

2. Kagome metal FeSn with A-type AFM order. For an A-type AFM order, spins are ferromagnetically aligned within one layer but antiferromagnetically coupled between adjacent layers. *This magnetic configuration provides an ideal platform to verify the spin resolution of the Bi₄Br₄ tip, as additional contributions from orbital effects and tunneling matrix elements should be largely suppressed between these two layers with same atomic constitutions.*



3. Kagome metal FeGe with A-type AFM order: another key compound

FeGe hosts an A-type AFM order with canted spins mainly oriented out-of-plane at 0T (Fig. R3-6a; Nature 609, 490 (2022)). With an external out-of-plane magnetic field exceeding 7T, the system undergoes a spin-flop transition, reorienting the spins into in-plane direction (Fig. R3-6b; Nature 609, 490 (2022)). Consequently, magnetic-field-dependent STM measurements are highly valuable for further elucidating the spin resolution of the Bi₄Br₄ tip.

- **Bi₄Br₄ tips are more sensitive to in-plane spin components.** Figure R3-6c shows an STM topographic image of a cleaved FeGe surface featuring a single-unit-cell step edge. The two sides of the edge are thus identical (except for spins). Under zero or weak (± 1 T or ± 2 T) magnetic fields, the spin orientation of FeGe remains predominantly out-of-plane, whereas the spin polarization of the tip is in-plane (enforced by the large energy scale of SOC in Bi₄Br₄ so that magnetic field of a few Tesla cannot change the spin direction on the tip); therefore, the dI/dV maps exhibit very weak contrast between adjacent layers (Fig. R3-6d). However, *when an external field of ± 8 T is applied to induce the spin-flop transition, a pronounced contrast emerges between the two layers* (Fig. R3-6 e, f). This arises because the spins of both the

sample and the tip are now aligned in-plane, yielding a finite overlapping component between them. Upon reducing the external field to ± 1 T, this contrast correspondingly diminishes again (Fig. R3-6g-h).

This series of experiments were carried out in the same area with the same STM tip, and the most obvious change is the spin direction on the sample (but with the contribution from orbitals, spin-independent tunneling-matrix elements largely unchanged). Thus, we believe *it provides compelling evidence that the spin polarization of the helical tunneling channels is intrinsic, and the spins on tip are “in-plane” relative to the coordinate system of the target sample.*

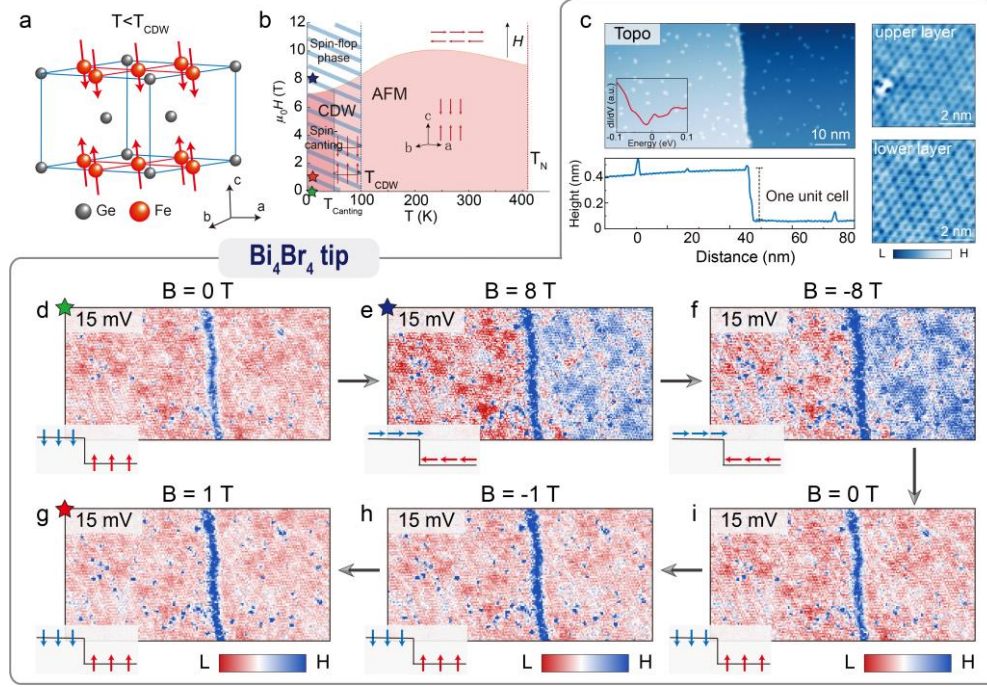


Fig. R3-6 **a**, Magnetic structure of FeGe at 0 T. **b**, Phase diagram of FeGe with field. **c**, STM topography including one single-unit-cell step edge. The right panels show atomic images of the upper and lower layers. **d-i**, dI/dV maps recorded at 15 meV with varying the magnetic field. A large enhancement of the contrast is found at ± 8 T under which the spins flop.

- **The spin resolution behavior of Bi_4Br_4 tip is again different from that of magnetic tip.** For a standard magnetic tip, a small external field of 1 T or 2 T is typically sufficient to fully polarize its apex out-of-plane. Given that the spin orientation of FeGe is also predominantly out-of-plane under these conditions, a distinct spin-polarized contrast should be readily observable at 1 T or 2 T. To experimentally verify this picture, we have prepared Fe-coated tips. The resulting data reveal a pronounced spin contrast at 1 T (or 2 T), as shown in Fig. R3-7. This conventional magnetic-field dependence is fundamentally distinct from the behavior observed with the Bi_4Br_4 tip, further corroborating the non-trivial, in-plane spin resolution of

the helical tunneling channels in the Bi₄Br₄ tip.

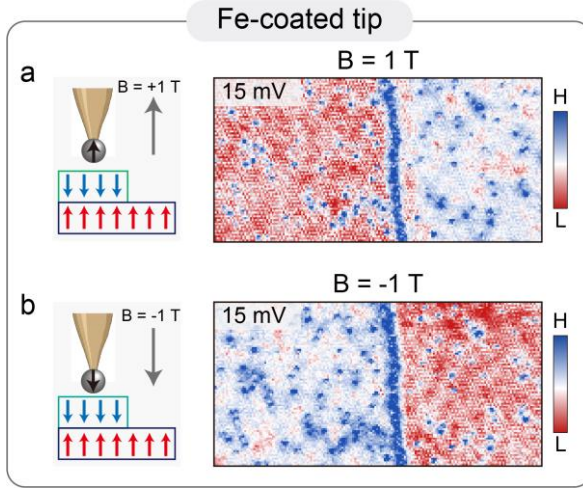


Fig. R3-7. dI/dV maps at 15 meV under magnetic fields of 1 T (a) and -1 T (b), in the same area but measured with a Fe-coated STM tip. An external field of 1 T is typically sufficient to fully polarize the tip's apex spin to out-of-plane, therefore a clear contrast can be observed. Reversing this smaller field flips the spin direction on the tip but keeps these in the sample, thus the contrast reverses between layers at ± 1 T.

4. KV₂Se₂O with d-wave checkerboard AFM order. The opposite Zeeman shifts of the in-gap states confirm their opposite spin polarization along the x- and y- directions (Fig. 4; we updated these data with much higher quality). This is a direct and well-established way to determine the spin texture of these states. Consequently, the observed $\pi/2$ rotation of the QPI anisotropy upon reversing the bias voltage polarity can be fundamentally attributed to the underlying spin texture of these electronic modulations, and **we further confirmed that such rotation is absent when using three spin-summed PtIr tips.**

Finally, the supplementary control experiments on Fe_{1+x}Te and FeGe strongly indicate that the Bi₄Br₄ tip is more sensitive to the in-plane spin configuration of the substrate. **Nevertheless, the tip retains the capability to resolve the out-of-plane spin contrast of DOS. This arises from an unavoidable geometric tilt (approximately 10°) of the tip relative to the surface normal in our experimental setup.**

As shown in Fig. R3-8b, during the FIB process, the PtIr wire supporting the Bi₄Br₄ tip must be bent by 90° to accommodate the holder geometry within the FIB-SEM system. After the FIB processing, we bent the PtIr wire back with tweezers to make it vertical to fit the STM tip holder. However, due to the mechanical rigidity and short length of the wire, a residual macroscopic tilt angle of approximately 10° is practically inevitable (Fig. R3-8b). Because the Bi₄Br₄ stripe is mounted strictly parallel to the distal end of the PtIr wire, it consequently inherits this $\sim 10^\circ$ deviation from the surface normal of the sample. Consequently, the spin polarization of the helical edge modes is not strictly orthogonal to the out-of-plane spins of the substrate. This geometric projection yields a finite, overlapping spin component, allowing for the detection of an effective spin contrast in the dI/dV measurements but with reduced effective spin-polarization ratio.

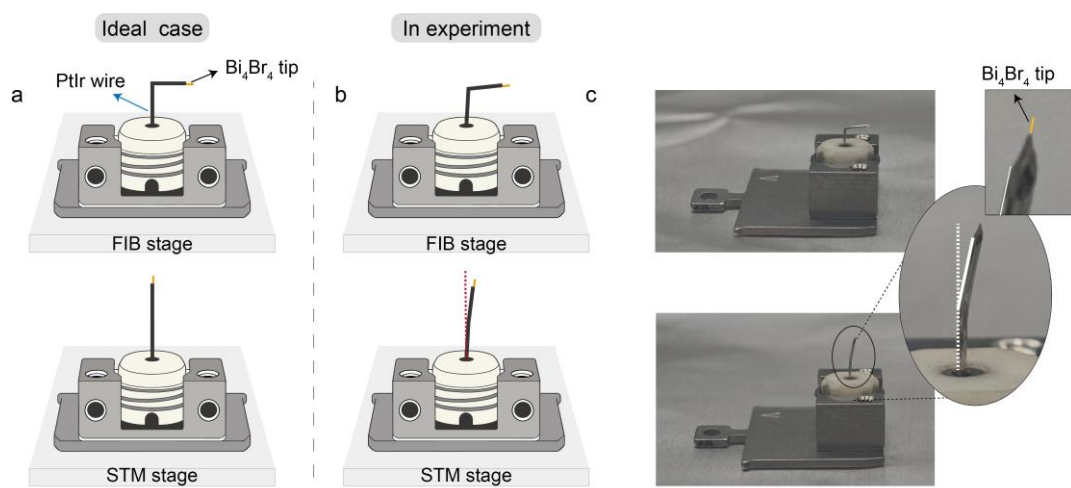


Fig. R3-8 a, Sketch showing the ideal case to prepare the Bi_4Br_4 tips, in which the tip is strictly vertical to the surface of sample. **b**, Sketch showing the experimental process in which there is an unavoidable geometric tilt (approximately 10°) of the tip relative to the surface normal. **c**, Photos to show the tilting of the tip. The Bi_4Br_4 stripe is strictly parallel to the end of PtIr wire.

Summary. The comprehensive set of observations detailed above firmly establishes the spin-resolving capability and robust performance of the Bi_4Br_4 tip. By investigating four distinct magnetic compounds, we have consistently obtained spin-resolved STM data that reflect their respective magnetic structures, which can rule out the possibility of coincidental artifacts. Furthermore, **these collective data consistently indicate that the spins carried by the helical edge channels are more sensitive to in-plane spins on the target sample. This might be one main contribution of this work that advances the field of “topological” tip on the basis of the pioneer work of SmB_6 nanowires (Ref. 62).**

In the revised version, we have included the typical datasets in Extended Data figures and SI notes: Fe_{1+x}Te (Extended Data Figure 4; Methods; Supplementary Note 3), FeGe (Extended Data Figure 5; Methods) and $\text{KV}_2\text{Se}_2\text{O}$ including the control experiments (Methods; Extended Data Figure 10).

Appropriate use of statistics and treatment of uncertainties

The manuscript doesn't describe or discuss statistics methods or uncertainties. I don't consider this a weakness since a systematic treatment of uncertainties in STM-related experiments is very unusual. Conclusions: robustness, validity, reliability

My main criticism concerns the presentation and interpretation of the data shown in Fig. 3 (see below). Fig. 1 is used to introduce the topic. Fig. 2 convincingly shows that Bi_4Br_4 tips are capable of mapping the spin structure on the atomic scale. Figs. 4 and 5

Suggested improvements: experiments, data for possible revision

See below.

Reply: We sincerely thank the referee for these comments. We will address these individual concerns in the following responses.

References: appropriate credit to previous work?

See above my criticism regarding Ref. 62.

Reply: We fully acknowledge the pioneering contribution of Aishwarya et al. (Ref. 62), who first

demonstrated the concept of spin-resolved tunneling using the topological surface states of SmB_6 nanowires. In the revised manuscript, we have updated the text to ensure that this seminal work is appropriately credited, explicitly highlighting its foundational role in developing topological STM probes (“Recently, a new type of SP-STM has been demonstrated in which the 2D topological surface states of SmB_6 nanowires were utilized to image the SP local density of states (LDOS) via their helical tunneling (50). Here, we extend this concept to the 1D topological helical edge modes...”).

Clarity and context: lucidity of abstract/summary, appropriateness of abstract, introduction and conclusions

Good.

List of comments (minor and major):

- page 2, line 45: add appropriate reference after “... and the second one utilizes nonrelativistic Zeeman splitting as in ferromagnets with broken time-reversal symmetry.”

Reply 3-1: We thank the referee for this suggestion. In the revised version, we have included the following foundational review to support the discussion on spin-polarized band splitting in systems with broken time-reversal symmetry:

Žutić, I., Fabian, J. & Sarma, S. D., Spintronics: Fundamentals and applications. Rev. Mod. Phys. 76, 323 (2004). *This review delineates the theoretical framework of exchange-driven spin splitting in ferromagnetic systems.*

- page 2, line 73: add appropriate reference after “electrons injected into or launched from a HOTI Bi_4Br_4 tip carry opposite spins enforced by their helical nature”

Reply 3-2: We thank the referee for this suggestion. In the revised version, we have cited Ref. 62 (the first work that introduces helical tunneling) and Nano Lett. 14, 4767–4771 (2014) (first theoretical prediction of the helical edge states in Bi_4Br_4) here.

- Overall, the tone related to the experiments performed with Bi_4Br_4 functionalized tips is a bit too enthusiastic, since similar experiments which also involve spin-momentum-locked helical states have already been performed in Ref. 62. I suggest to us a more moderate wording which appreciates the merits of earlier work. The authors shall also consider to move this part (although definitely impressive) to the extended data section or even the supplemental material.

Reply 3-3: We sincerely appreciate the referee’s constructive criticism regarding the tone of our manuscript and the contextualization of our work relative to Ref. 62. In the revised manuscript, we have moderated the wording throughout the section introducing the Bi_4Br_4 tips. We have also explicitly cited the pioneering work of Aishwarya et al. (Ref. 62) at the outset of our discussion on helical tunneling, ensuring that its original contribution to the development of topological STM probes is clearly highlighted and appreciated.

On the other hand, we believe that the utilization of edge states in Bi_4Br_4 tips for spin-resolved measurements *not only confirms the generality of the results in ref. 62, but also advances the related field (on basis of SmB_6 nanowires) in several aspects*: 1) a new topological material with larger bulk gap that is insensitive to FIB processes; 2) fabrication of sharp tips directly from complex single crystals; 3) extending 2D topological surface states with helical spin texture to 1D helical modes with nearly S_z quantization; and importantly 4) *experiments suggesting its sensitivity to in-plane spins* (see Fig. R3-6).

Under the premise of fully acknowledging the merits of earlier work, we believe that keeping these results in the main text provides a necessary and coherent foundation for the subsequent study of $\text{KV}_2\text{Se}_2\text{O}$. We have added further discussions to make this information clearer (Discussion section).

• page 3, lines 110-113: “Owing to the weak interlayer coupling, $\alpha\text{-Bi}_4\text{Br}_4$ also belongs to the family of 3D QSHIs, which hold helical edge modes encircling each layer despite a tiny hybridization gap (see Methods; 60, 61). These multiple helical edge modes give rise to large electron tunneling channels.” To my opinion, it remains unclear why multiple layers shall contribute to the tunneling process in a local probe experiment with an exponentially decaying tunneling probability.

Reply 3-4: We apologize for the ambiguity in our previous phrasing. We would like to clarify that our statement does not imply multiple layers contribute simultaneously to a single tunneling event, which would indeed contradict the principle of exponentially decaying tunneling probability in a local probe experiment. Instead, we intended to emphasize that the presence of helical edge states in every layer of the 3D QSHI significantly enhances the probability and robustness of activating topological tunneling channels, regardless of the specific microscopic geometry of the tip apex. Our reasoning is based on the following considerations.

In HOTIs, the presence of *gapless* hinge state on certain hinges relies on specific crystalline symmetries which is hard to be guaranteed at the tip end. However, as theoretically pointed out by ref. 59 and supported by our experimental data shown in Fig.2c, d, $\alpha\text{-Bi}_4\text{Br}_4$ belongs to the 3D QSHI with *slightly gapped* (nearly invisible in our data) helical edge channels for each layer (characterized by the spin Chern number; ref. 59, also see our previous experiments on $\alpha\text{-Bi}_4\text{I}_4$, Ref. 60). *The presence of these helical edge channels is robust without special crystalline symmetry*. As long as the Fermi level is positioned away from the tiny hybridization gap, the system effectively acts as a stack of decoupled 2D QSHIs. To substantiate this layered QSHI feature, we present dI/dV maps acquired at the boundaries of monolayer, bilayer, and trilayer step edges in Fig. R3-9a–c, where the presence of edge channels can be observed in all cases.

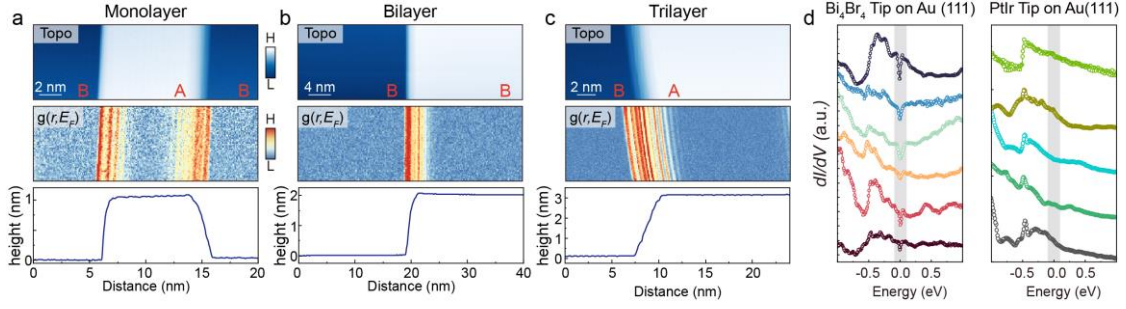


Fig. R3-9 a-c, topographies and the corresponding dI/dV maps at E_F for monolayer, bilayer, and trilayer step edges in Bi_4Br_4 crystal. **d**, comparison of Bi_4Br_4 tips and PtIr tips on Au surface to show the TLL anomaly.

Experimentally, we found that over 60% of the fabricated Bi_4Br_4 tip successfully exhibit the characteristic spectral signatures of these edge states (TLL-like anomaly) after tip conditioning on an Au (111) surface (Fig. R3-9d). This high success rate strongly implies that the active tunneling states for the Bi_4Br_4 tip are indeed these slightly gapped, weakly-coupled helical edge channels circulating within each layer. Consequently, the "multiple helical edge modes" mentioned in the text refer to the high density of available topological states across the tip structure, ensuring that the end of the tip (whichever layer it belongs to) can readily contribute a helical mode. We have revised the manuscript to state this more precisely in the Methods section. We also modified the related sentence to "These robust modes significantly enhance the probability of activating their corresponding tunneling channels, irrespective of the specific microscopic geometry of the tip apex".

- Fig. 2h: Data are very noisy and it appears questionable whether the opposite corrugation suggested by the line sections in the lower panel is an overall observation (which it should be). Therefore, instead of presenting selected data only, the authors shall present the sum and the difference of the dI/dV images taken at +50mV (left) and -50mV (right).

Reply 3-5: We thank the referee for this nice comment. Following this suggestion, we have improved the data presentation to illustrate their overall tendency.

First, we improved the signal-to-noise ratio of the line-profiles through spatial averaging. Rather than using single-pixel line-cuts, each profile was averaged over a 30-pixel width perpendicular to the line-cut direction. As shown in the updated Fig. R3-10, these averaged profiles clearly demonstrate a consistent contrast reversal between opposite bias polarities. We also clarify that the "noisy" features at negative bias primarily stem from atomic-scale modulations (Bragg peaks) that manifest as local kinks (Fig. R3-10c and d). By applying a q -space filter to suppress these Bragg modulations (with the cutoff indicated by the gray shaded region in Fig. R3-10d), a nearly symmetric mirrored pattern is recovered, as shown in Fig. R3-10e.

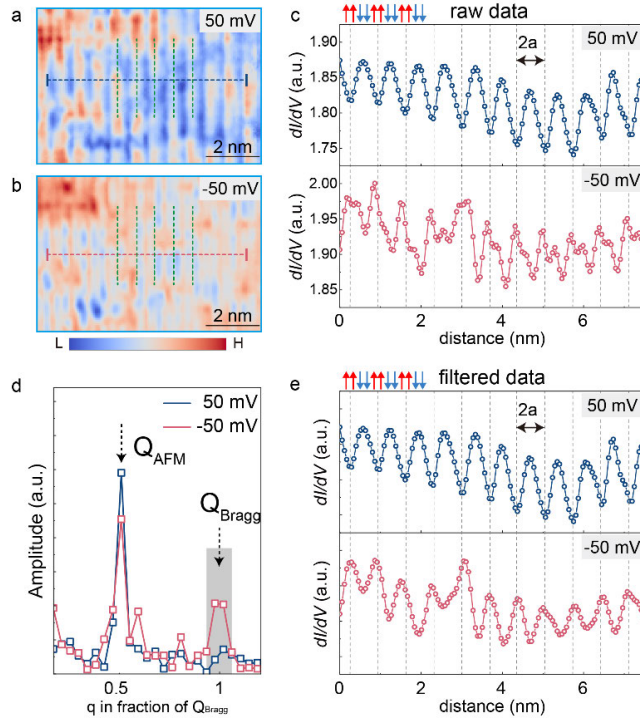


Fig. R3-10. **a, b,** dI/dV maps obtained at 50meV and -50meV. **c,** linecuts of the dI/dV spectra. To show the overall tendency, the linecuts are averaged over 30 pixels perpendicular to the linecut direction. **d,** FFT for the linecut profile shown in **c**, where both peaks at AFM vector and Bragg vector can be observed at -50meV. The gray shaded region denotes the q -space window for Fourier filter. **e,** a nearly mirrored pattern is recovered after the Fourier filter process.

Second, we have performed a sum-and-difference analysis of the entire dI/dV maps. 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, $\tilde{g}(+50 \text{ mV}) - \tilde{g}(-50 \text{ mV})$ (Fig. R3-11a), and the sum map, $\tilde{g}(+50 \text{ mV}) + \tilde{g}(-50 \text{ mV})$ (Fig. R3-11b). The line-profiles extracted from these maps (Fig. R3-11c, d) reveal that the difference map is dominated by a robust stripe pattern with a $2a$ periodicity, characteristic of the AFM order. In contrast, the sum map primarily retains the atomic lattice information. This collective analysis confirms that the contrast reversal is an overall spectroscopic signature across the entire FOV rather than a localized artifact.

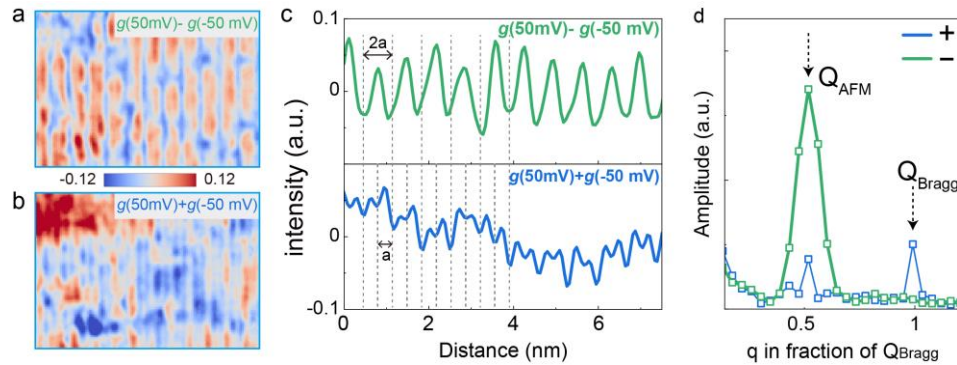


Fig. R3-11. **a, b,** the difference map **(a)** and sum map **(b)** of the dI/dV maps at -50meV and 50meV. Each raw map was first normalized by its spatial average. **c,** line-profiles extracted from these difference and sum maps. To show the overall tendency, the linecuts are averaged over 30 pixels perpendicular to the linecut direction. **d,** FFT for the linecut profile shown in **(c)**.

In the revised version, we have updated Fig. 2h with data after averaging, and included more datasets and analyses in Extended Data Fig. 4 and Supplementary Note 3, as well as discussions in Methods.

• page 4, lines 156-157: In the sentence “this gap becomes narrower somewhere and eventually collapses with the emergence of subgap states” the meaning of the word “somewhere” remains unclear.

Reply 3-6: We thank the referee for pointing out this ambiguous phrasing. The term “somewhere” was intended to denote the specific spatial coordinates in the proximity of V-site defects. To address this, we have overlaid the extracted positions of the V-site defects onto the energy gap map in the revised Fig. 3c (also see Fig. R3-12). This visualization clearly demonstrates that the regions exhibiting a narrowed or collapsed gap (characterized by the emergence of prominent sub-gap states) coincide with the atomic locations of these defects.

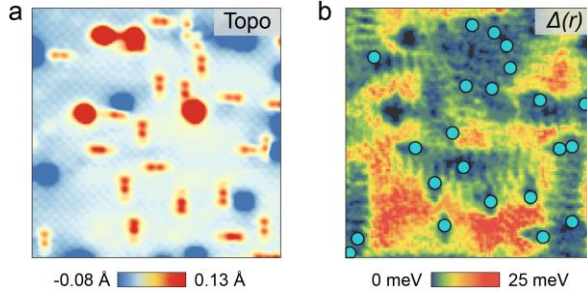
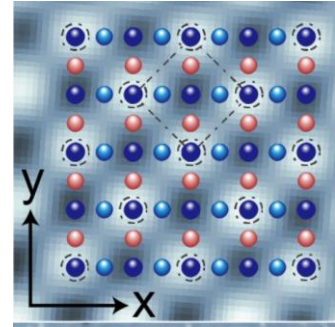


Fig. R3-12. **a**, topographic image showing the location of V-site impurities, **b**, the extracted gapmap in the same FOV, overlaid with the position of impurities (blue spots).

In the revised manuscript, we have replaced “somewhere” with a more precise description, explicitly attributing the localized gap closing to the impurity-induced states at the V-site defect positions.

• Fig. 3: To my opinion the difference between the Vanadium sites caused by the (2x2) reconstruction of the 50% filled K layer, such as V_{ya} and V_{yb} , is not introduced and explained well in the manuscript. The sketches presented in the lower left of Fig. 3a with 50% encircled K atoms and Fig. 3g with large and small red and blue balls representing V_{ya} , V_{yb} and V_{xa} , V_{xb} is not really obvious. Here some (graphical and didactic) adjustments might help.

Reply 3-7: We thank the referee for pointing out the ambiguity regarding the inequivalent Vanadium sites. In fact, the two inequivalent sites V_{xa} and V_{xb} (V_{ya} and V_{yb}) for the V_x (V_y) sublattice are not related to the $\sqrt{2} \times \sqrt{2}$ K surface reconstruction. We have updated the inset of Fig. 3a and now the relative position of atoms can be easily identified (see the new figure on the right).



The lifting of degeneracy within each sublattice is evidenced by the “arrow-shaped” electronic signatures of the V-site defects (Fig. 3d). These defects point either left (V_{xa}) or right (V_{xb}), revealing two inequivalent sites within the V_x sublattice. This symmetry breaking is attributed to the emergence of an intrinsic density wave within the V lattice possessing a $\sqrt{2} \times \sqrt{2}$ periodicity. Although this periodicity coincides with that of the K surface reconstruction, the V-lattice order is a distinct electronic phenomenon. Crucially, as shown in Figs. 3e and f, all V_{xa} and V_{xb} impurities remain globally aligned across large fields of view ($\sim 70\text{nm}$), transcending the domains of the surface K reconstruction (see Reply 3-10 for further evidence).

To avoid confusion, we have revised the schematics in Figs. 3d–f. We have removed the surface K lattice layer from these panels and instead plotted unit-cell lattice, which directly highlights the

correspondence between the defect orientations and the intrinsic order of the Vanadium sublattices.

- page 5, line 170-171: Related to the previous comment. You write ``This explicit arrangement doubles the original unit cell to a new  $\sqrt{2} \times \sqrt{2}$  supercell (Fig. 3g)``. It would be helpful to also show the unit cell as it appears without lifting degeneracy of a and b type sites.

Reply 3-8: We sincerely thank the referee for this suggestion. We have included the original unit cell in Fig. 3 in the new version, which allows a direct comparison of the original and new $\sqrt{2} \times \sqrt{2}$ unit cells.

- Fig. 3 b and c: To evaluate any potential correlation between the local structural properties and the gap width, the topography of the same location shown in Fig. 3c shall be presented as well. The reference to Fig. S6 is not helpful as this data set doesn't show the corresponding map of the local gap width.

Reply 3-9: We fully agree with the reviewer that a direct spatial comparison between the topography and the gap map is essential for establishing their correlation. In the revised Fig. 3c, we have overlaid the identified coordinates of the V-site defects directly onto the gap map to provide a clear visual correspondence. We have also included the topography image (Fig. R3-13a) for the same area in Extended Data Figure 6. This comparison demonstrates that the spatial regions characterized by gap narrowing or collapse coincide precisely with the locations of the V-site defects.

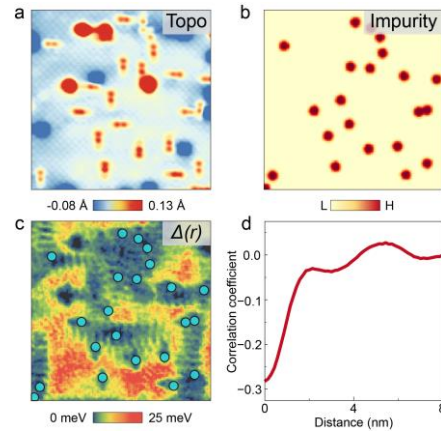


Fig. R3-13. **a**, topographic image showing the location of V-site impurities, **b**, the location of V-site impurities. **c**, the extracted gap map in the same FOV, overlaid with the position of impurities (blue spots). **d**, cross-correlation analysis between the defect distribution map (**b**) and the spatial gap map (**c**).

To provide a quantitative basis for this observation, we performed a 2D cross-correlation analysis between the defect distribution map (Fig. R3-13b) and the spatial gap map (Fig. R3-13c). The resulting correlation coefficient confirms a significant negative correlation, substantiating that the energy gap remains robust in pristine, defect-free regions but systematically suppresses or collapses at the specific coordinates of the V-site defects. We believe these visual and statistical enhancements fully address the referee's concern. These results are now plotted in Extended Data Figure 6.

- page 5, lines 171-178: It remains unclear which data and analysis led the authors to conclude that ``this supercell represents an intrinsic modulation of the V lattice but is not a simple consequence of the surface K reconstruction``. The authors refer to the Extended Data Fig. 5 and claim the observation that ``the spatial correlation is long ranged regardless of the surface domains and appears even on the  $2 \times 1$  reconstructed surfaces``, but I was unable to verify these claims based on the given material, i.e. the extended manuscript in pdf or docx format. (In either case the resolution

was too poor! This may not be caused by the data themselves but may be caused by the figure production process.) Therefore, the attribution of the observed supercell to a density wave remains somewhat unsupported by appropriate data.

Reply 3-10: We sincerely apologize for the clarity issues in the previous version due to image resolution. We have re-produced Extended Data Fig. 7 (reproduced as Fig. R3-14) with a high-resolution rendering and clear annotations to substantiate our conclusions.

In a pristine square lattice of vanadium without additional symmetry breaking, all V_x (V_y) sites are equivalent. However, the emergence of "arrow-shaped" electronic signatures at V-site defects (Fig. 3d) indicates a lifting of this degeneracy. Specifically, defects on the V_x sublattice point either left (V_{xa}) or right (V_{xb}), while those on the V_y sublattice point up (V_{ya}) or down (V_{yb}). This symmetry breaking points to an intrinsic density wave with $\sqrt{2} \times \sqrt{2}$ periodicity within the V lattice, as all these impurities remain globally aligned according to this periodicity (Fig. 3e–f).

To distinguish this intrinsic V-lattice modulation from the coincidentally $\sqrt{2} \times \sqrt{2}$ K surface reconstruction, we provide threefold evidence.

First, if the impurity anisotropy were driven by the surface reconstruction, the spatial correlation rules would necessarily shift or reset when crossing a K-surface domain wall, where K atoms undergo a lattice shift (see an example in Fig. R3-14b, in which the orientation of two impurities along the same diagonal line should change when crossing a surface domain). We present a topographic map spanning multiple surface domains (Fig. R3-14a). Our analysis reveals that the spatial correlation of the impurities remains continuous and long-ranged across the entire field of view. As shown in Figs. R3-14c–d, left-pointing arrows consistently occupy V_{xa} sites (orange lines) while right-pointing arrows occupy V_{xb} sites (purple lines), completely regardless of the surface domain boundaries.

Second, we find that the inequivalence of V sites persists even on the 2×1 reconstructed surfaces (Fig. R3-14e). The fact that defects still exhibit directional symmetry breaking on a structurally different surface pattern demonstrates that the modulation is intrinsic to the V_2Se_2O plane and decoupled from specific surface K arrangements.

Third, as shown in Fig. R3-14 f and g, a similar energy gap is observed on both 2×1 and $\sqrt{2} \times \sqrt{2}$ reconstructed surfaces. This energy-scale consistency supports the conclusion that the gap arises from a bulk-like density-wave transition rather than a surface-specific reconstruction.

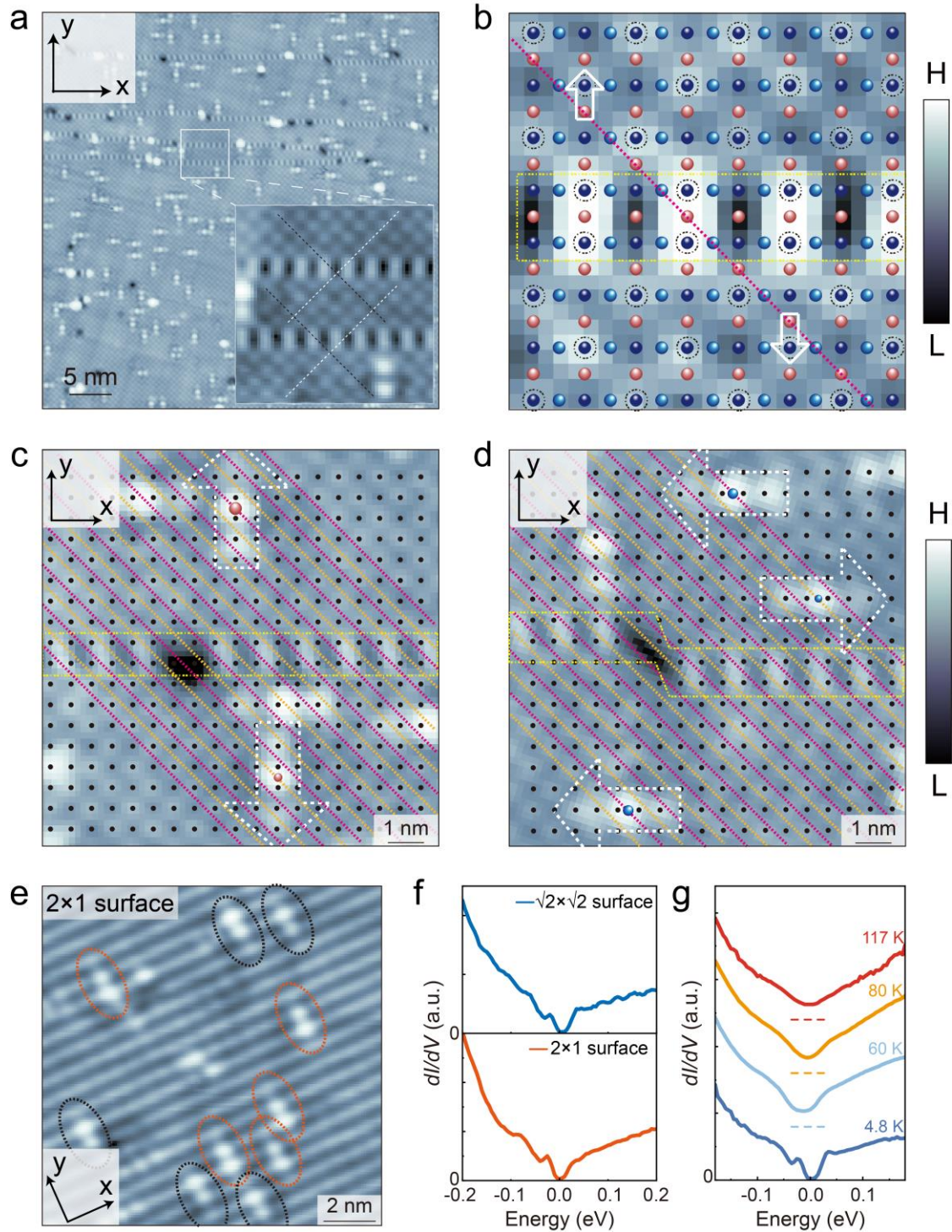
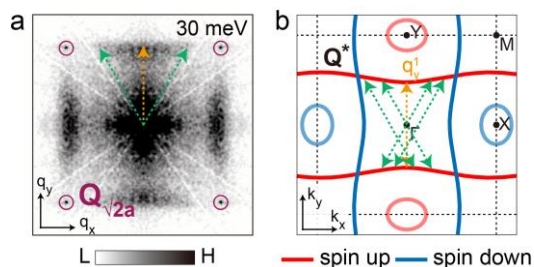


Fig. R3-14 **a**, atomic resolved topographic map in large FOV spanning multiple surface domains. **b**, assumption for a relation between defects' shape and the surface K position (dashed circles) in the $\sqrt{2} \times \sqrt{2}$ surface reconstruction. In this case, the orientation of two impurities along the same diagonal line should change when cross a surface domain (yellow box). **c-d**, continuous and long-ranged spatial correlation of the impurities, regardless of the surface domain. **e**, the inequivalence of V sites persists even on the 2×1 reconstructed surfaces. **f**, a similar energy gap on both 2×1 and $\sqrt{2} \times \sqrt{2}$ reconstructed surfaces. This energy-scale is consistent with the bulk density-wave transition. **g**, Temperature-dependent dI/dV spectra of the $\sqrt{2} \times \sqrt{2}$ surface. The gap feature disappears above 100 K.

• page 5, lines 183-185: The authors state that ``each (scattering vector) consists of multiple parallel stripes in  $q$ -space that reflect the morphology and dispersion of the constant-energy contour (CEC) in  $k$ -space (Fig. 4b).'' I cannot identify these ``multiple parallel stripes in q -space''. The authors shall specify what they refer to.

Reply 3-11: We thank the reviewer for pointing out this issue. The term "multiple parallel stripes" refers to the extended, quasi-1D-like QPI features observed in q -space, rather than discrete, point-like scattering spots. Physically, this morphology is a direct consequence of the CECs in KV_2Se_2O , which host prominent quasi-1D segments derived from the dxz and dyz orbitals. In such a system, the JDOS is dominated by scattering between these flat or nested segments. Beyond the primary scattering channels along the high-symmetry ΓX and ΓY directions, additional slanted scattering paths between these segments are allowed, yielding the broadened or stripe-like QPI signatures (see figure on the right).



In the revised manuscript, we have refined the text to replace "multiple parallel stripes" with more precise terminology as "quasi-1D scattering features", to better describe these features.

• Fig. 4d: Why do the experimental data show two branches, whereas DFT predicts one linearly dispersing state only?

Reply 3-12: We thank the reviewer for this insightful question. Such a difference comes from the fact that the experimental data were acquired in the $\sqrt{2} \times \sqrt{2}$ density wave phase with band-folding and gap opening. In contrast, the initial DFT results presented in the previous version were calculated for the pristine phase, which does not account for the density-wave-induced reconstruction. Specifically, the $\sqrt{2} \times \sqrt{2}$ density wave effectively reduces the Brillouin zone, folding the original bands and resulting in the appearance of two distinct dispersing branches. To verify this, we have performed updated DFT calculations incorporating the $\sqrt{2} \times \sqrt{2}$ supercell to account for the density wave. As shown in Fig. R3-15b, the calculated energy dispersion now resolves two branches, which are in excellent quantitative agreement with our experimental

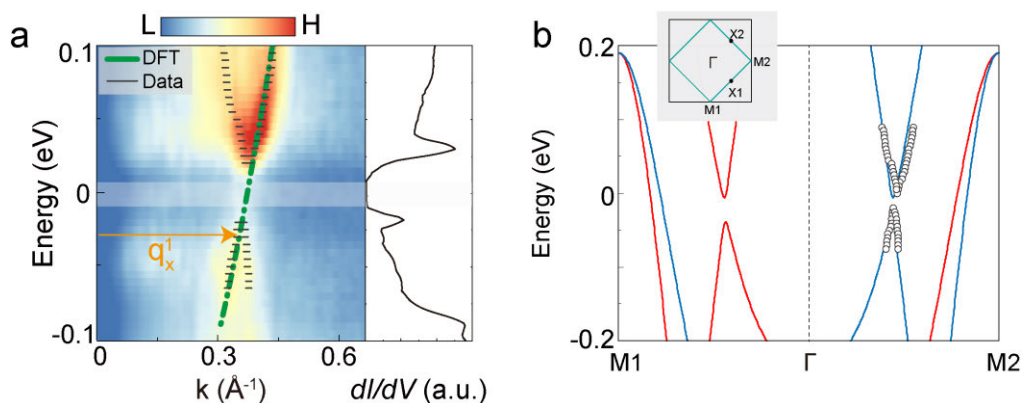


Fig. R3-15 **a**, experimental dispersion and calculated band of the pristine phase. **b**, these for the density wave phase.

observations.

We have revised the manuscript to explicitly state that the two branches of energy dispersion originate from the band folding due to the $\sqrt{2} \times \sqrt{2}$ density wave, and added the related analysis in Fig. S20 (cited in main text).

• Fig. 5 and related text: Why do the authors (somewhat indirectly) conclude the antiferromagnetic order from an opposite magnetic field-induced shift of the spectra shown Fig. 5g and h rather than mapping the antiferromagnetic order directly?

Reply 3-13: This is a good question. We totally agree with the referee that directly mapping the magnetic order can be an important goal for spin-STM, performing such measurement and analysis, however, is difficult for $\text{KV}_2\text{Se}_2\text{O}$. Specifically, the Neel vector (Q_{AFM}) coincides exactly with either the atomic Bragg modulation or the second-harmonic frequency of the surface structural reconstruction. Consequently, the magnetic signals in q -space are spectrally overlapped with structural contributions. Given that both signals can exhibit complex and potentially energy-dependent changes, it is difficult to unambiguously disentangle the intrinsic magnetic contrast from structural artifacts using real-space mapping techniques. This situation contrasts with systems like Fe_{1+x}Te , where the magnetic vector is distinct from the lattice Bragg peaks, allowing for straightforward magnetic imaging.

To circumvent this ambiguity, we rely on the magnetic-field-induced Zeeman shift as a robust alternative. In the revised manuscript, we have updated these data by performing spectroscopic measurements in regions with low vacancy concentrations. This approach allows us to isolate the in-gap states of individual V-site defects and precisely track their evolution under an external field. As shown in Fig. R3-16, the peaks associated with excitations along the x and y directions exhibit collective Zeeman shifts of opposite signs. This spectroscopic response provides a direct and well-established determination of the local spin texture, serving as a decisive cross-check for the

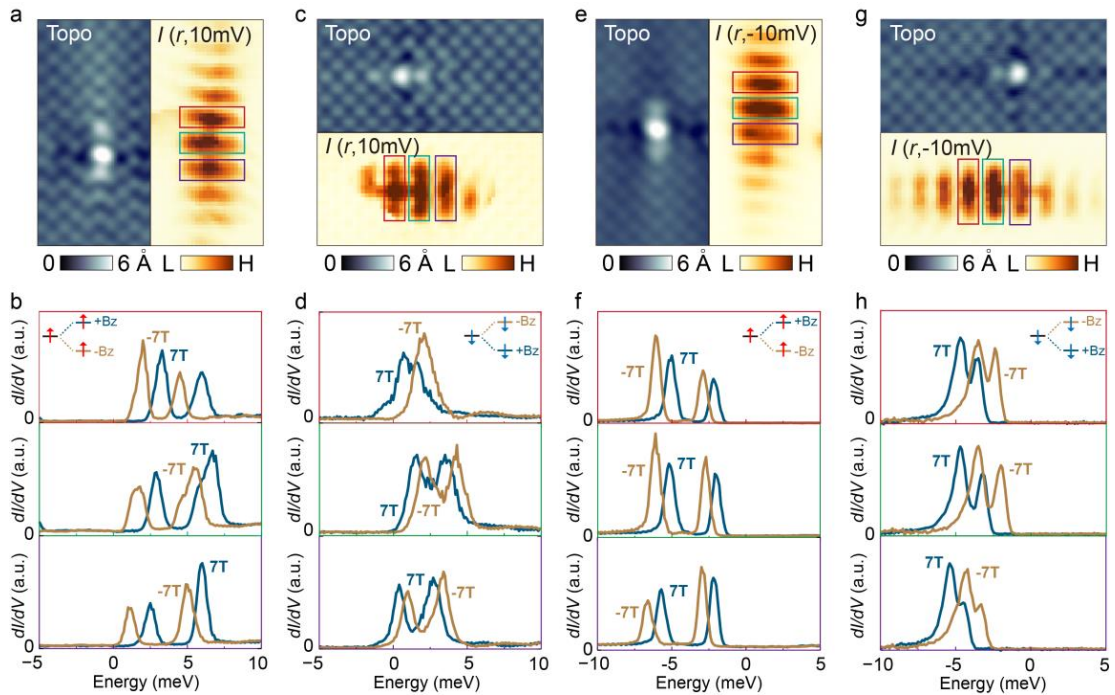


Fig. R3-16 a, e, topographies of two V-site defects that induced in-gap states along the y direction. since there could be multiple in-gap states peaked at different energies, here a current map at 10meV is displayed to show the spatial feature of these in-gap state clearly. **b, f**, Magnetic-field-induced Zeeman shifts for in-gap states along y- direction. The spectra are averaged for each colored box shown in **a** and **e**. **c, g** and **d, h**, similar data for in-gap states along the x direction, showing opposite Zeeman shifts under fields.

quasiparticle interference (QPI) results presented in Fig. 5.

Overall Reply: We sincerely thank the referee for his/her thoughtful evaluation and for recognizing the high quality, scientific significance, and broad interest of our study. We are also grateful for the constructive suggestions which have certainly improved the manuscript. In this revised version, we have carefully addressed each of the referee's remarks by enhancing the clarity of our presentation and incorporating more in-depth physical discussions. We believe these revisions have substantially strengthened the manuscript and hope the referee finds our responses and the improved paper suitable for publication.

Summary of some key responses to all referees

We sincerely thank the referees for their rigorous evaluation and constructive feedback. Before addressing the specific comments point by point, we wish to highlight the new experiments and analyses incorporated into the revised manuscript.

i. Out-of-plane spin polarization in $\text{KV}_2\text{Se}_2\text{O}$

We measured the response of in-gap quasiparticle states to *vector magnetic fields* with *identical magnitude* applied along three orthogonal axes (B_x , B_y , and B_z), with ultra-high energy-resolution at 30 mK. Our data reveal a highly anisotropic Zeeman response: the quasiparticle states are highly sensitive to out-of-plane fields yet exhibit no observable Zeeman shifts under in-plane fields (**Fig. R1**; see below). This strong anisotropy demonstrates that the predominant spin polarization in $\text{KV}_2\text{Se}_2\text{O}$ is out-of-plane. This finding is consistent with bulk neutron-scattering measurements (Sun *et al.*, *Phys. Rev. B* **112**, 184416 (2025)), indicating that our spectroscopic results capture the intrinsic AFM spin structure in $\text{KV}_2\text{Se}_2\text{O}$.

ii. The dominant role of spin polarization over orbital effects

We understand the referees' caution regarding potential orbital contributions, particularly given the introduction of a novel probe methodology in this work. To address this, we have assembled a comprehensive and coherent set of experimental facts (rather than a single experiment), which can disentangle the major role of spin polarization from potential orbital effects for our observations.

From a theoretical perspective, we have carefully analyzed the orbital contribution of both Bi_4Br_4 -tip edge states (Extended Data Fig. 1g-i) and the $\text{KV}_2\text{Se}_2\text{O}$ samples (Fig. S34). We find that the orbital textures of both the Bi_4Br_4 edge states and the $\text{KV}_2\text{Se}_2\text{O}$ sample remain uniform within the low-energy window near E_F where the QPI rotation is observed. Thus, the commonly observed robust bias-polarity-dependent changes in the experiments are difficult to understand with solely orbital effects. On the other hand, it is more physically sound and natural to attribute the current-direction-dependent signals to the spin-momentum locking of the Bi_4Br_4 -tip edge states, which is further demonstrated by our new experiments.

First, we provide a new direct, non-magnetic control experiment. We measured the QPI patterns of another tetragonal oxide, Sr_2RuO_4 , using the Bi_4Br_4 tips. Its well-known Fermi surface features a quasi-1D pocket derived from dxz/dyz orbitals, similar to that of $\text{KV}_2\text{Se}_2\text{O}$, but without AFM order and band spin-splitting. If the $\pi/2$ rotation in $\text{KV}_2\text{Se}_2\text{O}$ were an inherent consequence of dxz/dyz orbital matrix elements, it would necessarily manifest in the QPI data of Sr_2RuO_4 . However, our experimental results on Sr_2RuO_4 exhibit neither pronounced QPI anisotropy between the dxz/dyz orbitals nor any bias-polarity-dependent contrast rotation (**Fig. R2**). This rules out the orbital-selection effect as the primary driver.

Second, we exploit a magnetic field-driven spin-flop transition to show that the measured contrast is sensitive to changes of spin structure. In the A-type AFM compound FeGe , we applied an 8 T magnetic field to flop the spins from a predominantly out-of-plane to in-plane, which resulted in a clear, concurrent change in the LDOS contrast. Given that the pure orbital features of these two layers are identical, this contrast change is unambiguously tied to the magnetic structure of FeGe (**Extended Data Fig. 4**).

Third, we demonstrate universal spin-selectivity across diverse material platforms. We have successfully used the Bi₄Br₄ tips to resolve spin-dependent responses across three distinct magnetic systems: (1) the π -phase shift of spin stripes in bicollinear Fe_{1+x}Te, (2) the inter-layer contrast in A-type AFM FeGe, and (3) the highly anisotropic X- and Y-QPI responses in the d-wave altermagnet KV₂Se₂O. Given that these compounds possess different point group symmetries, crystalline lattices, and orbital configurations, this universal efficacy constitutes compelling evidence for the intrinsic spin-selective nature of the Bi₄Br₄ tip.

Finally, our findings are independently supported by Zeeman spectroscopy shown in Fig. 4 and R1.

Taken together, the spin sensitivity of the Bi₄Br₄ probe is not reliant on a single isolated observation, but is firmly established through a highly consistent, multi-material benchmark experiments. *We do not claim a complete absence of the orbital effects, but from the full dataset it is clear that the spin polarization plays a more dominant role.*

iii. Direct imaging of the AFM order in KV₂Se₂O

Inspired by Reviewer #3's suggestion, we have now obtained real-space images that potentially reflect the AFM order in KV₂Se₂O. This is possible since our QPI data reveal the contribution of vanadium-d electrons in tunneling, though the V lattice is located underneath the top layer. When using a Bi₄Br₄ tip, we found that the FT intensities at the AFM ordering wavevectors also exhibit pronounced anisotropy that systematically reverses upon switching the bias polarity (**Fig. R3 a-b**), mirroring the behavior of QPI patterns. Again, this anisotropy reversal cannot be reproduced with non-magnetic PtIr tips. These findings suggest that while the Bragg modulation and AFM order are located at the same vectors, the latter could contribute to the overall signatures and can be probed with spin-selective Bi₄Br₄ tips. By inverse Fourier transforming all these AFM (Bragg) wavevectors (checked with different windows) and then constructing an averaged supercell, we obtain unidirectional real-space modulations: the intensities at V_x sites are higher at positive biases, whereas those at V_y sites become dominant at negative biases (**Fig. R3**). Thus, the difference map between positive and negative biases can directly reflect the spatial modulation of spin-resolved V_x and V_y sublattices that disappears in spin-summed measurement (**Fig. R3 k-t**), thus reflecting the real-space patterns of AFM order.

Furthermore, we identified a specific type of single defects that can simultaneously induces quasiparticle excitations along both the x and y directions (**Fig. R4 and R5**). These orthogonal excitations also exhibit opposite Zeeman shifts. Such a dichotomy cannot be trivially explained by a localized magnetic moment pinned at the defect; given their π/k_F spatial period that reflects the Fermi surface, it dictates a spin-polarized electronic structure of the host material rather than the defect itself.

Finally, we have added theoretical calculations and new experimental data to reveal the minor role of density wave in generating the anisotropy, and added comparison with non-magnetic PtIr tips to exclude the spin-independent density waves and structural anisotropy as origin of our key observations. The spin polarization of the electronic states is independently supported by Zeeman spectroscopy shown in Fig. 4 and R1.

Summary. Taken together, our revised manuscript now presents the full set of evidence for altermagnetism: *crystalline symmetry (Fig.3), the real-space mapping of AFM order (Extended data*

Fig. 10), the momentum-space band spin-splitting (Fig. 5), and the spin-polarized quasiparticle excitations (Fig. 4). The impact and timeliness of this work are further underscored by the fact that two other independent STM groups have recently adopted well-established SmB_6 tips and Cr tips to study related altermagnetic compounds--work directly inspired by our pioneering framework. Moreover, we have also introduced a totally new topological tip with known spin-polarization direction through extensive experimental efforts in this work, which is a significant plus.

We have tempered our wording to ensure they strictly align with the presented evidence as suggested by Reviewer #2. We firmly believe that these critical new results completely dismantle the remaining reservations, fully substantiating the robust scientific scope required for the broad, high-impact readership of Nature.

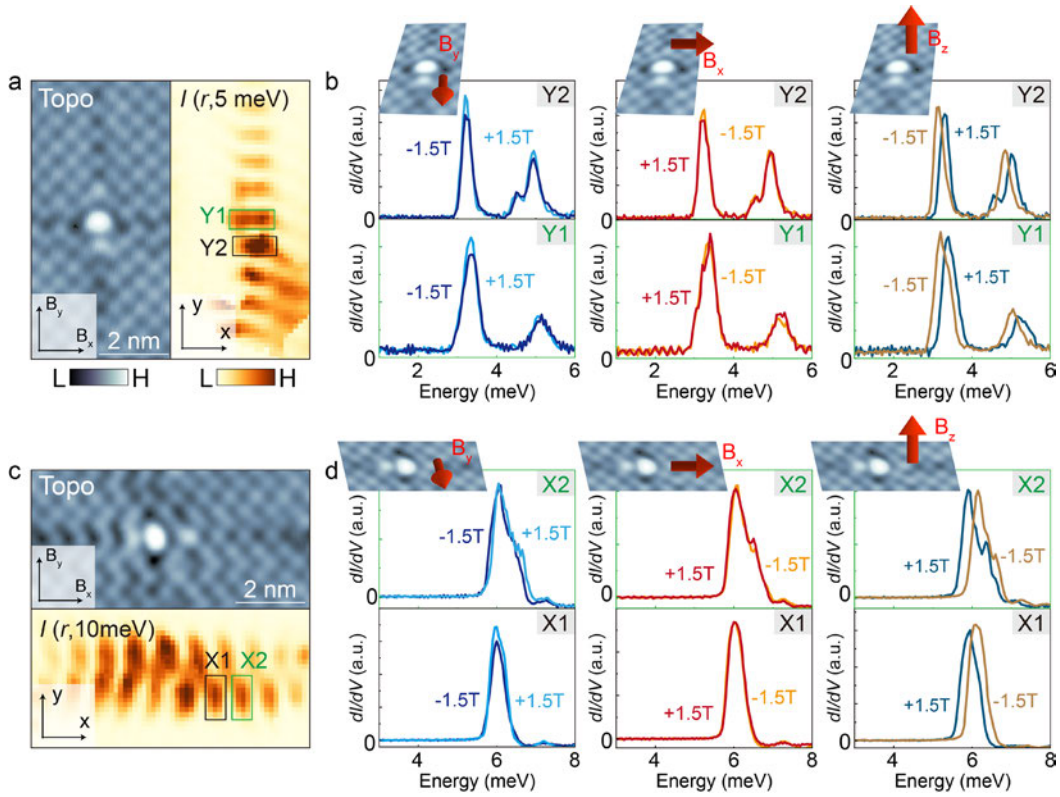


Fig. R1 **a, c**, Topographies and the corresponding current maps of two isolated V-site defects that induce in-gap states extending along the y (**a**) and x (**c**) direction, respectively. **b, d**, dI/dV spectra acquired at 30 mK under magnetic fields of $\pm 1.5 \text{ T}$ applied along the B_x , B_y , and B_z directions. The spectra are spatially averaged within the color-coded rectangles shown in **a** and **c**. The in-gap states show no resolvable shift under the in-plane fields B_x and B_y , whereas clear Zeeman shifts are observed under the out-of-plane field B_z .

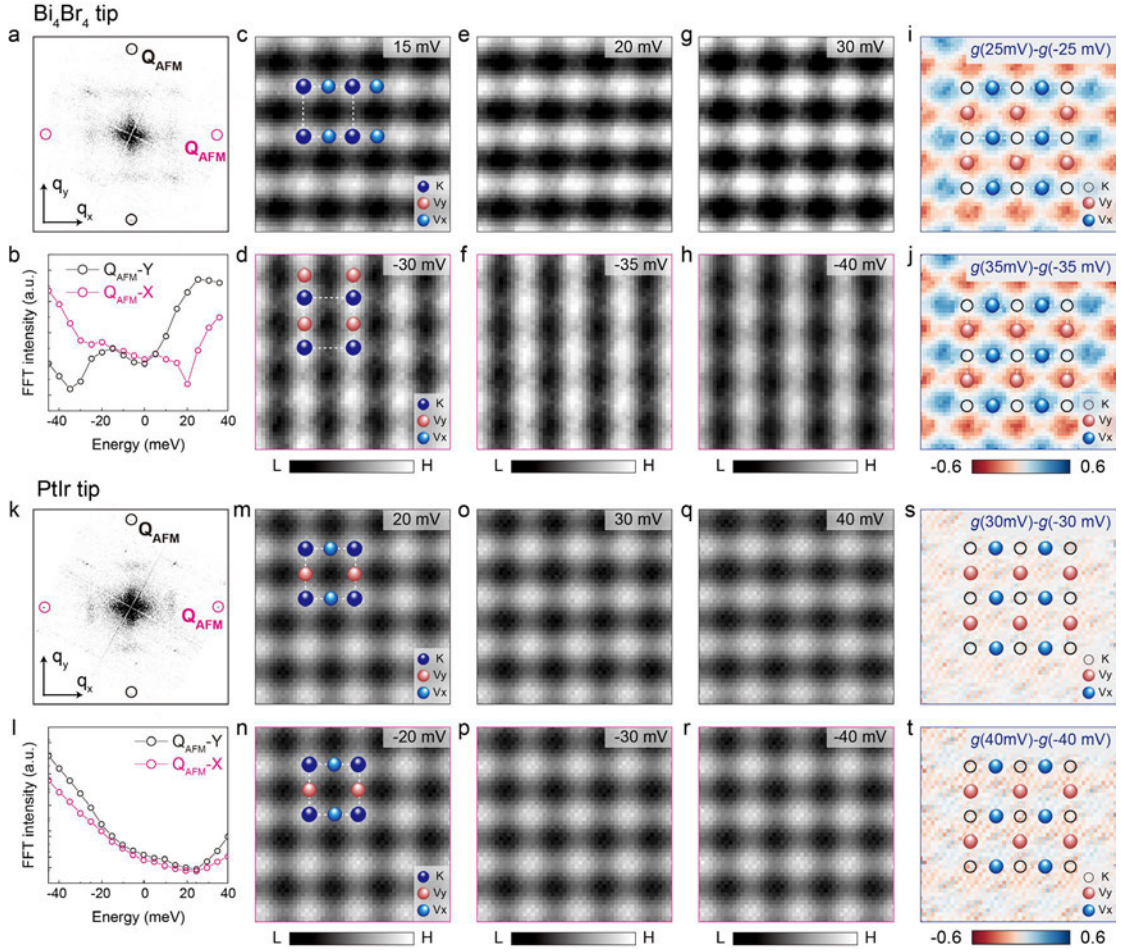
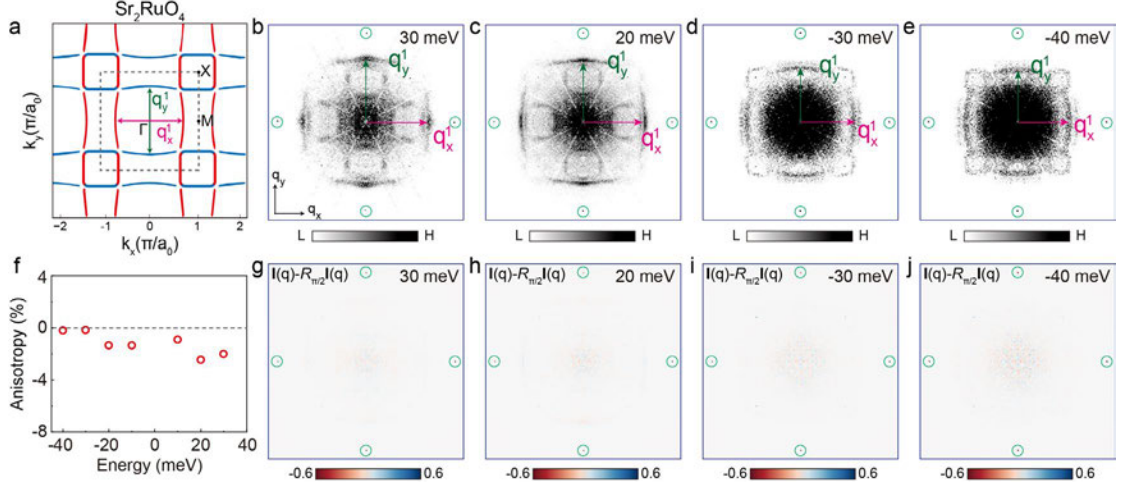


Fig. R3 Potential real-space mapping of the AFM order in $\text{KV}_2\text{Se}_2\text{O}$. **a, b**, FT and the bias-dependent intensities at the AFM wave vectors $Q_{\text{AFM-X}}$ and $Q_{\text{AFM-Y}}$, measured with a Bi_4Br_4 tip. **c-h**, Inverse FT of both the AFM peaks $Q_{\text{AFM-X}}$ and $Q_{\text{AFM-Y}}$, at representative positive and negative biases. The filtered maps were subsequently averaged over 4×4 supercells using an average super-cell algorithm. The dominant contrast switches from the V_x sublattice

at positive bias to the V_y sublattice at negative bias. **i, j**, Difference map between two maps at ± 25 mV (**i**) and ± 35 mV (**j**). Before subtraction, each real-space map was independently normalized by its maximum value. These difference maps provide a real-space visualization of the AFM order. **k-l**, FT and AFM Fourier intensities measured with a PtIr tip. **m-r**, similar to these in **c-h**, but with a PtIr tip, showing no bias-polarity-dependent reversal. **s, t**, Difference maps at ± 30 and ± 40 mV, respectively. No visible AFM contrast is observed with the PtIr tip.

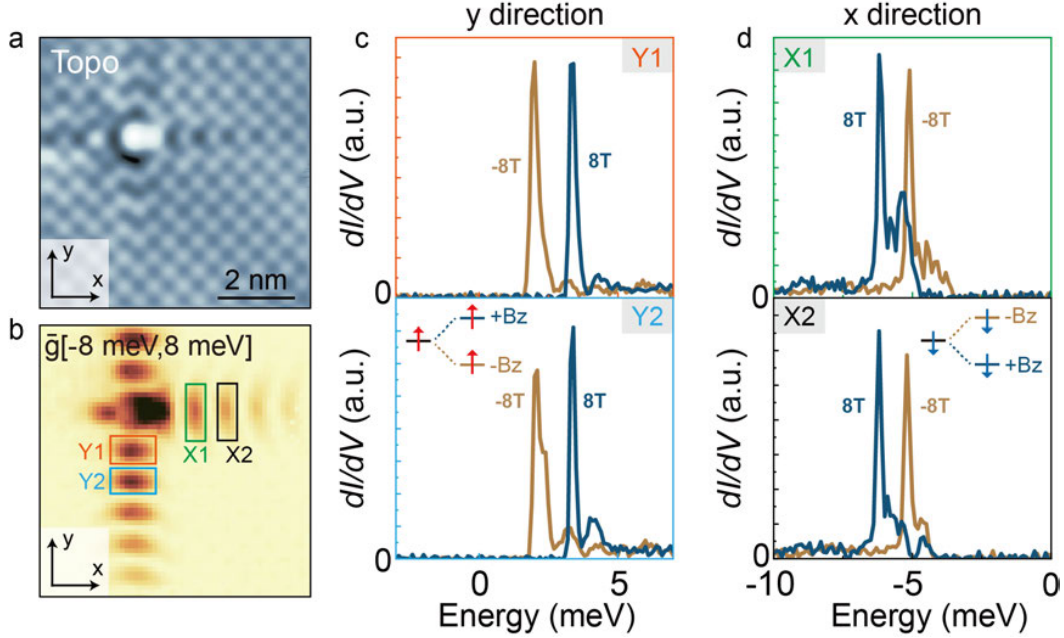


Fig. R4 Opposite Zeeman shifts near a single defect. **a**, topography of a defect that induces in-gap states along both x and y directions. **b**, Energy-averaged conductance map showing quasiparticle excitations induced simultaneously along both the x and y directions. The colored rectangles mark the locations at which the spectra in (**c**) and (**d**) were acquired. **c, d**, Magnetic-field-dependent dI/dV spectra of the in-gap states along the x and y directions, respectively, showing opposite Zeeman shifts upon out-of-plane magnetic fields.

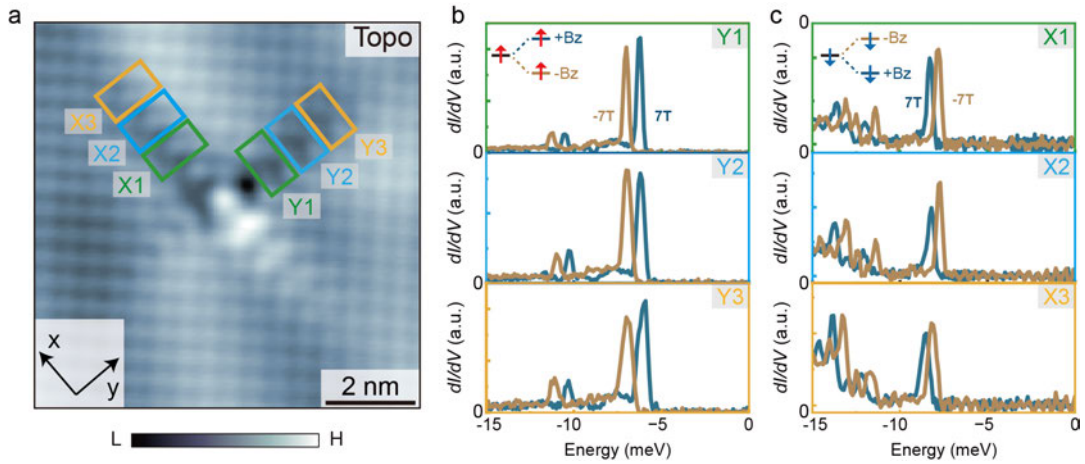


Fig. R5 Another defect showing similar results to those shown in Fig. R4.

Response to Referee #1

Zhuying Wang et al. have provided considerable additional characterization of their Bi₄Br₄ STM tips. Together with additional data on KV₂Se₂O, this improves the robustness of the experimental conclusions.

Reply: We sincerely thank the reviewer for the positive assessment and the constructive comments to improve this work.

To my understanding one central logical step still remains unresolved: The spin polarization of topological surface states is mostly orthogonal to the local moments of the altermagnetic order. The authors state that “Our experiments suggest that the Bi₄Br₄ tip is more sensitive to magnetic structures with in-plane spin orientations”. The authors note further that “Our calculations indicate that the edge state electrons also mainly possess an out-of-plane spin orientation ($\frac{\sigma_z}{\tau_z} \sim \pm 0.85 \times \hbar/2$; see Fig. R1-1). In the geometry of the STM measurement, this spin orientation lies “in-plane” with respect to the target sample”.

In response to my original comment that a tip spin polarization at a 90° angle to the substrate would remove any spin sensitivity, the authors mention “an unavoidable geometric tilt (approximately 10°) of the tip relative to the surface normal in our experimental setup.” as an explanation for an out-of-plane spin contrast.

Qualitatively, this can be true, but an angle of 10° is rather small. To my understanding this would imply for simple spin-conserving tunneling that the spin contrast for out-of-plane moments is proportional to $\cos(80^\circ) \sim 0.17$. The effective spin resolution is reported as 12%, and thus it seems that the tip channel selectivity would need to be ~ 0.7 even if there are no other losses of spin selectivity. Here, a value of 1 would be a complete selection of a single direction of spin-polarized helical channel.

Conversely, a deviation from the expected out-of-plane altermagnetic order, i.e., an in-plane moment on the sample, would be detected with $\cos(10^\circ) \sim 0.98$, so even a modest channel selectivity of ~ 0.12 would lead to 12% of effective spin signal. If in the vicinity of the measured defects and the QPI there is any deviation from the z-aligned order, the Bi₄Br₄ tip would be sensitive to it.

I think it needs to be explicitly discussed whether deviations from the z-aligned altermagnetic order can be excluded. Maybe an orientation dependence of the external-field-induced shift may resolve this question. Alternatively, also simulations of the defects themselves could exclude local in-plane polarizations.

While I think the use of a topological STM tip is a fascinating development, I believe the manuscript has to clarify why the experiments are not interpreted in terms of in-plane polarizations, to which the setup seems to be more sensitive.

Reply 1: We thank the referee for raising this insightful point. We fully agree that given the geometric sensitivity of the Bi₄Br₄ tip to in-plane spin components, it is crucial to determine whether the observed spin signatures can stem from an in-plane deviation (canting) of the spins in KV₂Se₂O.

To provide a direct and unequivocal test, we performed field-orientation-dependent Zeeman spectroscopy on these in-gap states. We selected two representative defects and measured their in-

gap states under vector magnetic fields of *identical magnitude* applied along the three orthogonal axes (B_x , B_y , and B_z) (Fig. R1-1). The measurements were carried out in a dilution-refrigerator-based STM at a base temperature of 30 mK, to achieve ultra-high energy resolution. An out-of-plane field of $B_z = \pm 1.5$ T can induce a pronounced Zeeman shift, and in sharp contrast, applying an identical field magnitude within the in-plane (B_x and B_y) yields no resolvable Zeeman shift whatsoever. This anisotropic response dictates that the quasiparticle states are predominantly spin-polarized along the c axis. This conclusion is consistent with bulk neutron-scattering measurement (Sun *et al.*, *Phys. Rev. B* **112**, 184416 (2025)). Therefore, we can exclude the presence of a sizable in-plane spin polarization within our experimental resolution; the observed response directly reflects the intrinsic z -aligned AFM order in bulk KV_2Se_2O .

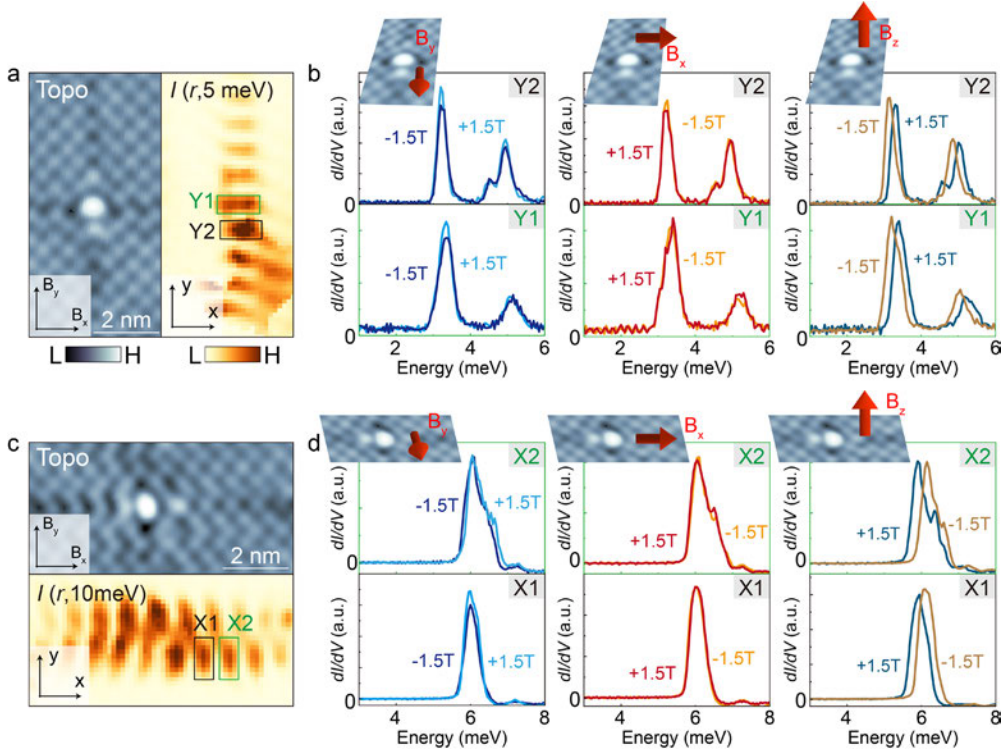


Fig. R1-1 **a, c**, Topographies and the corresponding current maps of two isolated V-site defects that induce in-gap states extending along the y (**a**) and x (**c**) direction, respectively. **b, d**, dI/dV spectra acquired at 30 mK under magnetic fields of ± 1.5 T applied along the B_x , B_y , and B_z directions. The spectra are spatially averaged within the color-coded rectangles shown in **a** and **c**. The in-gap states show no resolvable shift under the in-plane fields B_x and B_y , whereas clear Zeeman shifts are observed under the out-of-plane field B_z .

We would like to note that our work does not aim to provide a quantitative description of the effective polarization ratio. In fact, our estimation is based on the anisotropy of QPI patterns which is indirect. Even for conventional magnetic-tip-based STM, extracting effective polarization ratio is a complex and challenging task: It depends not only on the relative spin orientation, but also on the details of polarized electronic structures of both the tip and sample, as well as the spin-dependent tunneling matrix elements beyond a purely geometrical argument [Wiesendanger, *Rev. Mod. Phys.* **81**, 1495 (2009)]. A quantitative description of the present junction would therefore require a detailed microscopic analysis of the tip, sample and junction beyond this work.

However, qualitatively, an effective polarization ratio of $\sim 10\%$ in our case may not be surprising, if we consider both the tilting angle ($\cos(80^\circ) \sim 17\%$) and the inherent deviation from S_z quantization (15%) of the edge channels on the tip. These two factors, taken together natively, can give an upper

bound of roughly 20-30% for the polarization ratio. Thus, an estimated effective polarization of approximately 12% is reasonable.

In summary, our ultra-low-temperature vector-field measurements unequivocally demonstrate that the in-gap quasiparticle states are nearly c-axis polarized. These results show that the observed contrast is physically consistent with the out-of-plane AFM spin structure of bulk $\text{KV}_2\text{Se}_2\text{O}$. We have explicitly added the new data to Fig. 4 and related discussions in the revised manuscript to clarify this important nuance.

Additional comments:

1. To argue for the absence of orbital effects, the authors argue in terms of a p_y orbital that sticks out of a straight termination of Bi_4Br_4 . They conclude “Because this extending orbital is nearly normal to the surface, it maintains an isotropic tunneling matrix element with respect to the in-plane p_{xy} and p_{yz} orbitals of the sample”. There are three issues:

- The theoretical modeling considers a defect-free rectangular lattice, yet to my understanding a point-like tip apex is such a defect, for which in principle the orbital configuration may change.

Reply 1-1: We thank the referee for this important comment. In this work, the major role of spin polarization over orbital effects is established by a comprehensive and coherent set of experimental facts. The theoretical modeling and arguments of orbital textures of Bi_4Br_4 aim to provide additional support to understand these phenomena.

For Bi_4Br_4 , if the width of the termination were too narrow (specifically, less than 6 nm), the edge channels from opposite boundaries would strongly hybridize, opening a detectable finite-size gap (as shown in Extended Data Fig. 1d). For the Bi_4Br_4 tips used in this work, we routinely check the energy spectrum on Au and confirm that it has no detectable hybridization gap. This means that the effective apex of Bi_4Br_4 tips is larger than 6 nm (roughly 10 Bi_4Br_4 chains). We then calculated a Bi_4Br_4 ribbon with reduced width to 6 nm (10 chains), and the resulting spin and orbital characters of these edge states are essentially identical to those obtained for the wider ribbon (Fig. R1-2).

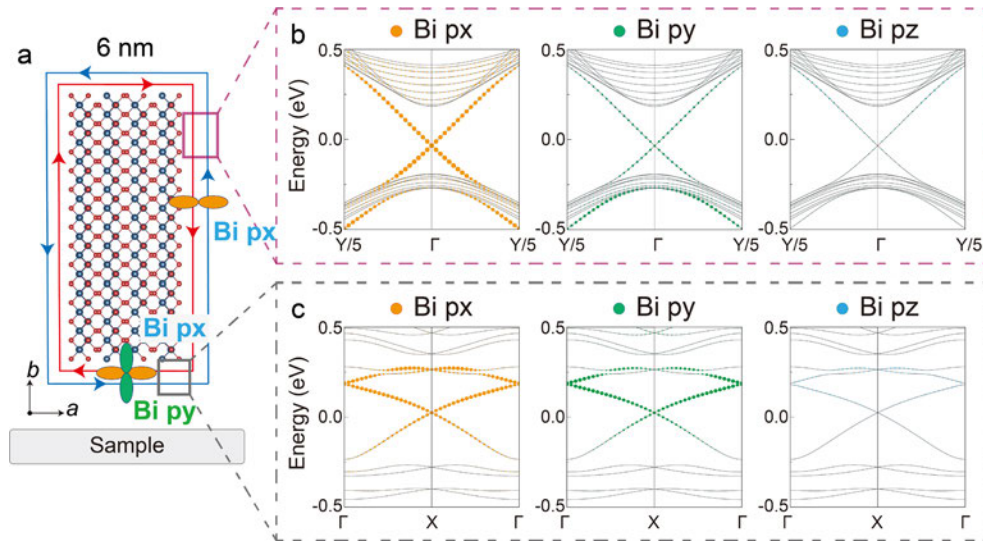


Fig. R1-2 Sketch of the tunneling junction incorporating the orientation of the helical edge channels and their orbital characteristics. The calculation is performed for a narrow Bi_4Br_4 ribbon containing approximately ten atomic chains (about 6 nm). The obtained orbital texture is similar to that of wider ribbons.

- Stating that the “orbital is nearly normal to the surface” is not an obvious way to exclude orbital effects, when the spin resolution derives from just this 10° deviation from the normal alignment. Although the orbital orientation is not identical to the spin-selective tunneling axis, the manuscript should explain why the same 10° tip tilt is important for spin contrast but negligible for orbital-selection effects.

- The authors state that “the anisotropy stemming from purely structural effects does not rotate when switching the bias polarity”, while this seems true, what about the orbital character? Generally, topological phases exhibit a band inversion in energy between bands of different symmetry. It is not obvious to me whether this can be excluded as source of anisotropy.

The referee is correct that definitively decoupling spin and orbital effects using purely geometric or theoretical mechanism arguments is highly non-trivial. For this reason, rather than relying solely on the geometric argument, we have designed a comprehensive suite of control experiments to address this point.

While topological phases indeed originate from bulk band inversion, our DFT calculations confirm that the orbital textures of both the Bi_4Br_4 edge state (Extended Data Fig. 1g-i) and the $\text{KV}_2\text{Se}_2\text{O}$ samples (Fig. S34) remain uniform within the low-energy window near E_F where the QPI rotation is observed. Attributing the systematic $\pi/2$ QPI rotation to an abrupt change in orbital texture across E_F would require a highly contrived set of coincidental assumptions, especially since this $\pi/2$ rotation is consistently observed with multiple tips regardless of their crystal orientations and microscopic geometries.

Therefore, we find it much more physically sound to attribute the current-direction-dependent signals to the spin-momentum locking of the tip's edge states. This conclusion is solidified by the following direct experimental facts.

First, we provide a direct, non-magnetic control experiment. We obtained new QPI data on Sr_2RuO_4 using the Bi_4Br_4 tip, which features a quasi-1D pocket derived from dxz/dyz orbitals, strikingly similar to that of $\text{KV}_2\text{Se}_2\text{O}$, but without AFM order and band spin-splitting. Crucially, our results on Sr_2RuO_4 exhibit neither pronounced QPI anisotropy between the dxz/dyz orbitals nor any tunneling-current-direction-dependent contrast rotation (Fig. R1-3). If the $\pi/2$ rotation in $\text{KV}_2\text{Se}_2\text{O}$ were an inherent consequence of dxz/dyz orbital matrix elements, it would necessarily manifest in Sr_2RuO_4 . Its absence rules out the orbital-selection effect as the primary driver.

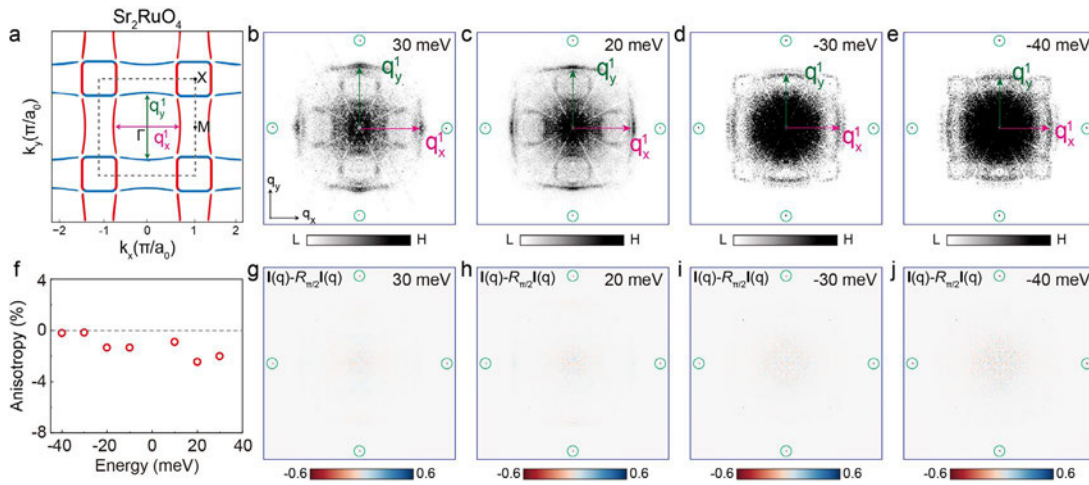


Fig. R1-3 QPI patterns on nonmagnetic Sr_2RuO_4 using a Bi_4Br_4 tip. **a**, CECs of Sr_2RuO_4 near E_F including quasi-1D pockets derived from the d_{xz} and d_{yz} orbitals, whose size and orbital texture are similar to those in $\text{KV}_2\text{Se}_2\text{O}$.

b-e, QPI patterns at representative energies near E_F . **f**, The calculated anisotropy of QPI intensity between d_{xz} and d_{yz} bands, $(I(\mathbf{q}_x^1) - I(\mathbf{q}_y^1))/(I(\mathbf{q}_x^1) + I(\mathbf{q}_y^1))$, as a function of energy. **g-j**, Corresponding contrast maps of rotating $\pi/2$. No pronounced QPI anisotropy reversal is observed, in sharp contrast to the case in KV_2Se_2O .

Second, we exploit a magnetic field-driven spin-flop transition to show that the measured contrast is sensitive to changes in spin structure. In the A-type AFM compound FeGe, applying an 8 T magnetic field flops the spins from a predominantly out-of-plane to an in-plane configuration. This magnetic transition results in a clear, concurrent change in the LDOS contrast. Because the local orbital texture is identical between these two layers, this contrast change is unambiguously tied to the spin structure (Extended Data Fig. 4).

Third, we demonstrate universal spin-selectivity across diverse material platforms. We have successfully deployed the Bi_4Br_4 tips to resolve spin-dependent responses across three distinct magnetic systems: the π -phase shift of spin stripes in bicollinear $Fe_{1+x}Te$, the inter-layer contrast in A-type AFM FeGe, and the highly anisotropic X- and Y-QPI responses in the d-wave altermagnet KV_2Se_2O . Given that these compounds possess different point group symmetries, crystalline lattices, and orbital features, this universal efficacy (but in different behaviors) strongly argues against an orbital-derived mechanism.

Taken together, the spin sensitivity of the Bi_4Br_4 probe is established by the full set of measurements rather than by any single experiment. In FeGe, which has an AFM order but distinct orbital arrangements on hexagonal Kagome lattice, the tip can resolve the layered magnetic contrast that changes with magnetic fields. In contrast, Sr_2RuO_4 has similar orbital textures but no AFM order (no spin splitting), where no QPI anisotropy or contrast reversal at E_F is observed. These findings highlight the major role of spin polarization over orbital effect in our measurements.

We note that we do not claim a complete absence of the orbital effect, but from the full set of data it is clear that the spin polarization plays a more dominant role. We have added these experiments and related discussions in the main text.

2. The FeGe measurement exhibits uniform color shifts on both sides of the steps when comparing $+1T$ and $+8T$, respectively. What do they imply about how the external field affects the topological states on the tip?

Reply 1-2: We thank the referee for raising this perceptive question. The uniform color shifts do not imply large modification of the topological states on the tip. Instead, they are the direct manifestation of a well-documented spin-flop transition intrinsic to the FeGe sample under fields.

The helical edge states in the Bi_4Br_4 are governed by the strong spin-orbit coupling (SOC) inherent to the heavy Bi and Br elements, which is on the energy scale of several hundred meV. In stark contrast, the pure Zeeman energy (field direction parallel to the momentum of edge channels) introduced by a 8 T magnetic field is less than 1 meV. Because of this massive energy scale mismatch, an external field of a few tesla is insufficient to strongly perturb the spin-momentum locking or reorient the spin texture of the tip's edge channels. Experimentally, we also did not find visible changes in the large energy spectra of Bi_4Br_4 tip when calibrated on Au(111) under a 8 T field, the field can open a small gap (1meV) at Dirac point but its energy scale is small. These data have been included in Fig. S4 in the new version.

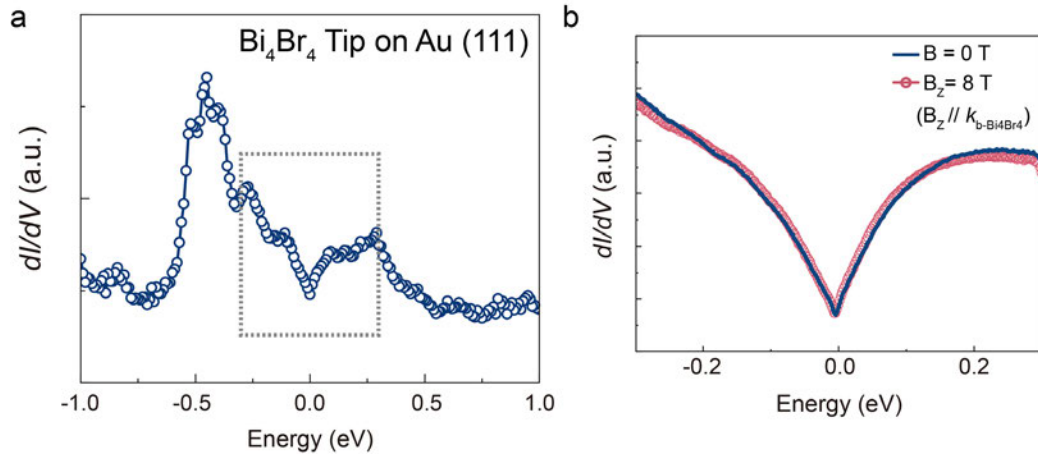


Fig. R1-4 **a**, dI/dV spectrum acquired on the Au (111) surface using a Bi₄Br₄ tip. **b**, zoom-in of the spectral line shape near E_F at 0 T and 8 T with the same tip, no discernible change was observed at 4.2 K due to the tiny pure Zeeman gap.

In contrast, FeGe is an A-type antiferromagnet that undergoes a spin-flop transition under an out-of-plane field of approximately 7 T, with the magnetic moments reorienting from a predominantly out-of-plane direction to in-plane [as confirmed by neutron-scattering in Nature 609, 490 (2022)]. Therefore, the uniform evolution, observed when the field is increased from ± 1 T to ± 8 T, directly reflects the global reorientation of the FeGe spin order, rather than a field-induced modification of the Bi₄Br₄ tip states (Fig. R1-5). Therefore, this provides a direct in-situ measurement to show how the spin orientation in the sample (in the same atomic region) can affect the measured spin resolution, thus directly linking the measured contrast to the spin order of FeGe. We have made this point clearer in the Method section and Extended Data Fig. 4.

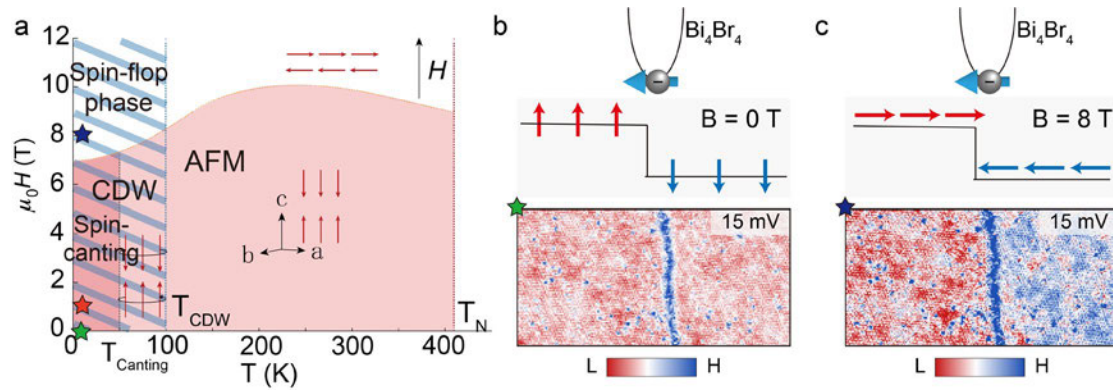
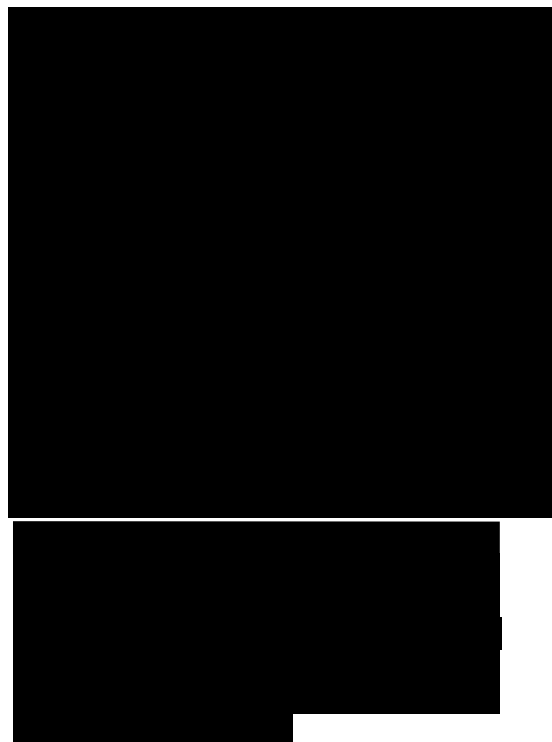


Fig. R1-5 **a**, Phase diagram of FeGe with field. **b-c**, dI/dV maps recorded at 15 meV with varying the magnetic field. A large enhancement of the contrast is found at 8 T under which the spins flop to in-plane in FeGe.

3. Why does for FeSn only the middle layer appear to be strongly polarized in the contrast between positive and negative bias?

Reply 1-3: We thank the referee for pointing out this visual ambiguity. We clarify that the apparent "polarization" confined to the middle layer in the original manuscript was merely a visual artifact caused by an unoptimized color scale. The relevant information is the relative dI/dV contrast between adjacent FeSn layers, rather than their absolute intensities. In the revised figure, we have shown these images with more appropriate color scales, to make this distinction clearer. The middle terrace has the opposite magnetic orientation to the upper and lower terraces, as expected for the A-type AFM order. Consequently, its relative contrast with respect to the two outer terraces reverses between positive and negative bias. Thus, the data reflect the alternating spin-dependent contrast of consecutive layers, rather than a spin polarization confined to the middle layer.



4. To argue for spin sensitivity, the authors consider possible causes for why there is an asymmetry in how well the active part of the tip connects to the edge states to achieve spin sensitivity:

- a band-bending effect, which seems reasonable in principle.
- a non-linear voltage response of the AFM surface, which I do not understand from the given discussion. The connection between a non-linear charge current response of the tip and the spin texture is unclear. The authors mention a previous study but do not reference it at this point.

Reply 1-4: We thank the referee for this question. In the following, we briefly explain non-linear charge current response for the tip and add references to the discussion. We note that this is just one potential scenario to explain the data.

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 signs 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 $\alpha \propto m_{local}$. 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 exactly the same symmetry property with the nonlinear Hall effect [1,2]. The latter is usually discussed in the diffusive regime [1,2,3]. 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 [4] or the Landauer-Büttiker formula [5]. 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.

1. Sodemann and Fu, PRL 115, 216806(2015);
2. Gao, Yang, and Niu, PRL 112, 166601 (2014);
3. Kaplan, Holder, and Yan, PRL 132, 026301(2024);
4. Liu, Liao, Qi, Ma, Geng, Sheng, and Xing, PRB 113, 075424(2026);
5. Wei, Wang, Yu, Xu, and Wang, PRB 105, 115411(2022);

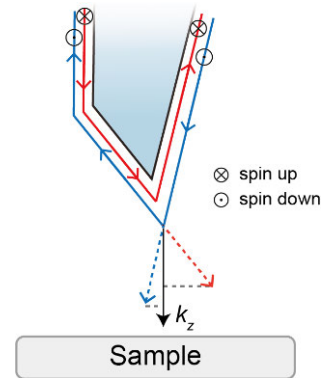
We have modified this discussion in the new version to clarify and added references.

5. The authors mention an "effective k_z component" but they labeled only k_b in their figure. Also, they show a vector decomposition of tip tangents in figure S29 which does not seem connected to the provided explanation of tunneling probability.

Reply 1-5: We thank the referee for pointing out this notational inconsistency. We agree that the label k_b in Fig. S29 was confusing and did not clearly correspond to the “effective k_z component” discussed in the text. Here, k_z denotes the effective momentum component projected along the tunneling direction, i.e., the surface normal of the sample.

We have thus revised Fig. S29 by replacing all k_b labels with k_z . The unnecessary decomposition of the tip tangents has been removed. The revised schematic now displays only the effective normal components for the two adjacent edge channels, and explicitly marks their difference. Consequently, one channel can contribute more strongly to the tunneling current, leading to a finite net spin contrast, while the weaker channel reduces the effective polarization.

We have correspondingly revised the text to clarify that this figure serves as a qualitative schematic to a possible microscopic asymmetry in the tunneling process, rather than a quantitative calculation of the tunneling probability which is more complex.



Response to Referee #2

I would like to thank the authors for their exhaustive and highly constructive rebuttal. In this revision, the authors have substantially expanded the experimental basis of their central methodological claim. They now report more than 30 fabricated Bi₄Br₄ tips, spin-resolved measurements across four distinct magnetic compounds (Fe_{1+x}Te, FeSn, FeGe, KV₂Se₂O), three independent classes of control experiment (applied magnetic field, physical sample rotation, and conventional Fe-coated magnetic tips), and DFT calculations of the orbital and spin character of the Bi₄Br₄ edge states. This is a considerable and commendable body of additional work.

However, while the dataset is extensive, the central claims exceed what is strictly established by the presented evidence. In particular, the interpretation of the Bi₄Br₄ tip as a quantitatively reliable spin filter and the assertion of direct momentum-space visualization of altermagnetism are not fully substantiated.

Reply: We thank the reviewer for the highly positive assessment of our extensive experimental efforts. We also deeply appreciate the referee's rigorous scrutiny regarding the precise boundaries of our central claims. In the revised manuscript, we have added more experimental results and further analyses to clarify our interpretation of the data and strengthen our conclusion, while meticulously recalibrating our wording to ensure they strictly align with the presented evidence.

We agree with the referee that making absolute quantitative claims in SP-STM is challenging. In fact, our current work does not claim a “quantitative” description of the Bi₄Br₄ tips; it aims to provide a coherent dataset for the spin-resolution capability. Thus, we now explicitly frame the tip as a robust, qualitative spin-selective probe, rather than a quantitative spin filter. Meanwhile, although our new data on Zeeman shifts and real-space mapping of AFM order has significantly strengthened our conclusion on the AM order, we have taken the referee's caution and systematically tempered our language throughout the manuscript—for instance, replacing terms like “smoking-gun evidence” with “direct experimental evidence”, and “spin filter” to “spin selective probe”.

We believe that these careful revisions, combined with the critical new experimental data summarized in the opening pages of this response, now firmly align our conclusions with the substantiated evidence. Our detailed, point-by-point responses to your specific concerns are provided below.

Major Comments

1. The control experiments with Fe-coated tips show absence of the reversal of STM contrast under opposite bias polarity when using Bi₄Br₄ tips. However, this does not uniquely isolate spin filtering as the mechanism, since magnetic tips and topological edge-state tips have fundamentally different energy-dependent tunneling characteristics. I thought the AFM signal also changes by rotating the sample for the Fe-coated tip. I cannot understand the bias antisymmetry originates from spin selection rule of the helical tunneling, it may also arise from orbital-selective tunneling from anisotropic edge states.

Reply 2-1: We thank the referee for raising this point. We would like to emphasize that in this work, the major role of spin polarization over orbital effect is established based on a comprehensive set of carefully designed measurements rather than by the single comparison.

The key feature for helical tunneling is the systematic corresponding change depending on the tunneling current direction (therefore, the bias polarity on samples) in one dataset without rotating the sample or modifying the junction. Physically, this arises from the spin-momentum locking of the edge states: reversing the bias polarity reverses the net tunneling current direction, which intrinsically couples to the opposite major spin polarization. *From our calculations, the orbital textures of Bi₄Br₄ tips (Extended Data Fig. 1g-i) and the KV₂Se₂O band (Fig. S34) in the relevant energy window do not show any typical change across E_F. However, the $\pi/2$ QPI anisotropy rotation in KV₂Se₂O (and also the π -shift of spin-stripe modulation in bicollinear Fe_{1+x}Te) across E_F is consistently observed with multiple tips, regardless of the tips' crystalline orientations.* Attributing the systematic evolution to an abrupt orbital texture change across E_F would require a highly contrived set of coincidental assumptions, especially since the $\pi/2$ rotation is consistently observed with multiple tips regardless of their crystal orientations and microscopic geometries.

Therefore, we find it much more physically sound to attribute the current-direction-dependent signals to the spin-momentum locking of the tip's edge states. This conclusion is solidified by the following direct experimental facts.

First, we provide a direct, non-magnetic control experiment. We obtained new QPI data on Sr₂RuO₄ using the Bi₄Br₄ tip, which features a quasi-1D pocket derived from dxz/dyz orbitals, strikingly similar to that of KV₂Se₂O, but without AFM order and band spin-splitting. Crucially, our results on Sr₂RuO₄ exhibit neither pronounced QPI anisotropy between the dxz/dyz orbitals nor any tunneling-current-direction-dependent contrast rotation (Fig. R2-1). If the $\pi/2$ rotation in KV₂Se₂O were an inherent consequence of dxz/dyz orbital matrix elements, it would necessarily manifest in Sr₂RuO₄. Its absence rules out the orbital-selection effect as the primary driver.

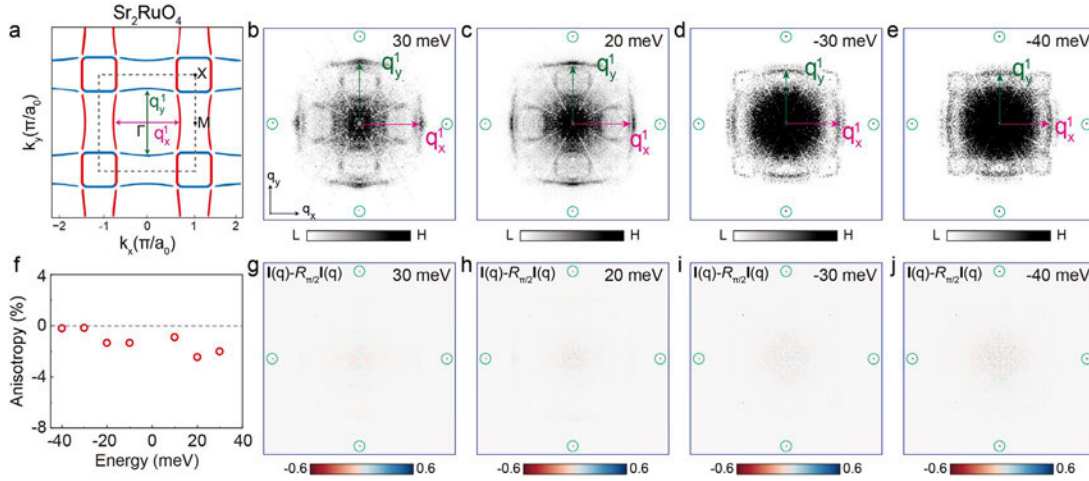


Fig. R2-1 QPI patterns on nonmagnetic Sr₂RuO₄ using a Bi₄Br₄ tip. **a**, CECs of Sr₂RuO₄ near E_F including quasi-1D pockets derived from the d_{xz} and d_{yz} orbitals, whose size and orbital texture are similar to those in KV₂Se₂O. **b-e**, QPI patterns at representative energies near E_F. **f**, The calculated anisotropy of QPI intensity between d_{xz} and d_{yz} bands, $(I(q_x^1) - I(q_y^1))/(I(q_x^1) + I(q_y^1))$, as a function of energy. **g-j**, Corresponding contrast maps of rotating $\pi/2$. No pronounced QPI anisotropy reversal is observed, in sharp contrast to the case in KV₂Se₂O.

Second, we exploit a magnetic field-driven spin-flop transition to show that the measured contrast is sensitive to changes in spin structure. In the A-type AFM compound FeGe, applying an 8 T magnetic field flops the spins from a predominantly out-of-plane to an in-plane configuration. This magnetic transition results in a clear, concurrent change in the LDOS contrast. Given that the pure

orbital arrangements of these two layers are identical, this contrast change is thus tied to the spin structure (Extended Data Fig. 4).

Third, we demonstrate universal spin-selectivity across diverse material platforms. We have successfully deployed the Bi₄Br₄ tips to resolve spin-dependent responses across three distinct magnetic systems: the π -phase shift of spin stripes in bicollinear Fe_{1+x}Te, the inter-layer contrast in A-type AFM FeGe, and the highly anisotropic X- and Y-QPI responses in the d-wave altermagnet KV₂Se₂O. Given that these compounds possess different point group symmetries, crystalline lattices, and orbital features, this universal efficacy (but in different behaviors) strongly argues against an orbital-derived mechanism.

We also note that a similar current-direction-dependent $\pi/2$ QPI anisotropy rotation of KV₂Se₂O has been reported with SmB₆ nanowire tips several months after our work (arXiv:2603.21969). The similar behavior indicates the important role of the spin-momentum locking of helical boundary states, rather than from a coincidental, material-specific orbital effect.

Taken together, the spin sensitivity of the Bi₄Br₄ probe is established by this exhaustive set of experimental cross-checks.

We do not claim a complete absence of the orbital effect in the STM tunneling, but from the full set of data it is clear that the spin polarization plays a more dominant role. [We have added these new experiments and related discussions in the main text.](#)

2. From the results of the two inequivalent V sublattices and defect-state spin inference, the real-space sublattice assignment is not unique, but the origin of strain-induced symmetry breaking is not fully disentangled. Although the $\sqrt{2}\times\sqrt{2}$ surface is less unidirectional than the 2×1 reconstruction, as seen in Figure 3 and 4, it still introduces a strong, extrinsic structural anisotropy. Thus, the data support anisotropic electronic structure consistent with symmetry-allowed altermagnetic scenarios, but do not uniquely establish altermagnetic spin splitting in momentum space. The observed $\pi/2$ rotation in QPI patterns is consistent with symmetry-driven electronic anisotropy but does not constitute a direct measurement of spin-resolved band structure.

Reply 2-2: We thank the referee for raising this point. We fully agree that distinguishing intrinsic spin-split electronic structures from extrinsic structural symmetry breaking is a challenge in STM. However, we can disentangle the altermagnetic spin signature from any structural factors through four independent, decisive lines of experimental evidence.

First, we have mentioned the key feature of helical tunneling in Reply 2-1, that is, the measured contrast undergoes a corresponding change depending on the tunneling current direction. If the $\pi/2$ rotation in QPI at E_F were primarily driven by a structural anisotropy and unrelated to spin, the non-magnetic tip would inherently detect it. However, we found that for a PtIr tip, which has no spin resolution but can equally image this structural anisotropy at atomic scale, cannot detect such $\pi/2$ rotation at all (Extended Data Fig. 9). The fact that the $\pi/2$ rotation is only observed with the spin-polarized Bi₄Br₄ tip dictates that the anisotropy originates from spin-selective tunneling rather than the structural anisotropy. Moreover, in Fe_{1+x}Te, the spin-related signal is not a $\pi/2$ rotation but a π -shift of spin stripe modulation across E_F , following the magnetic structure.

Second, besides the $\pi/2$ rotation of QPI anisotropy, the in-gap quasiparticle excitations along the x

and y directions *always (with no exceptions in more than four measured samples)* exhibiting opposite Zeeman shifts provide a cross-checking for the spin texture which should not be ignored. Structural strain preserves time-reversal symmetry and therefore cannot induce a Zeeman shift under a magnetic field. The real-space modulation period is π/k_F of the d_{xz}/d_{yz} bands, indicating that these excitations carry important information of the related Fermi surface. These opposite Zeeman shifts thus provide an independent real-space probe of opposite spin polarizations in quasiparticle excitations associated with the underlying Fermi surface, which is further supported by our vector-magnetic-field measurements (Fig. R1).

Third, we identified a single defect that simultaneously induces quasiparticle excitations along both the x and y directions. As shown in the new data (Fig. R2-2), these two directional excitations also exhibit strictly *opposite* Zeeman shifts under magnetic fields. Because these excitations originate from the same defect, this orthogonal Zeeman dichotomy cannot be trivially reconciled with a local moment of the defect. Thus, together with their π/k_F modulation period, the opposite Zeeman shifts of these excitations serve as an independent probe, supporting that the excitations inherit the spin-polarized electronic band structure of the host material.

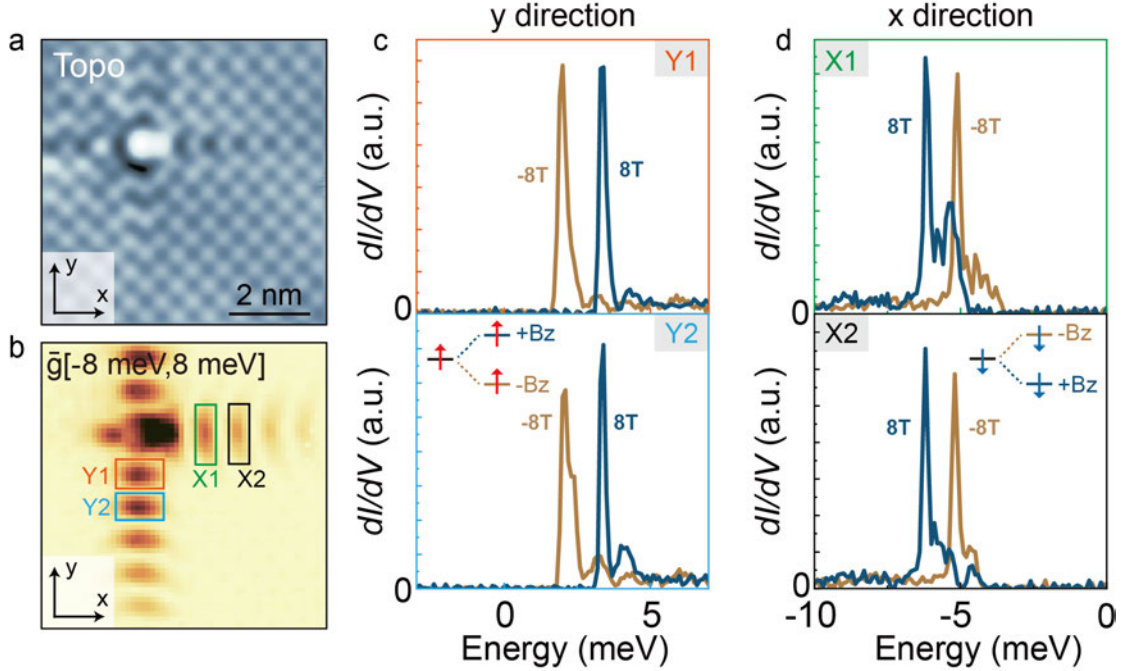


Fig. R2-2 Opposite Zeeman shifts near a single defect. **a**, topography of a defect that induces in-gap states along both x and y directions. **b**, Energy-averaged conductance map showing quasiparticle excitations induced simultaneously along both the x and y directions. The colored rectangles mark the locations at which the spectra in (c) and (d) were acquired. **c**, **d**, Magnetic-field-dependent dI/dV spectra of the in-gap states along the x and y directions, respectively, showing opposite Zeeman shifts upon out-of-plane magnetic fields.

Finally, to eliminate the influence of defect-induced strain and structural anisotropy, we have obtained real-space patterns that reflect the AFM order in defect-free regions with improved data quality. This challenging measurement is made possible because our QPI data demonstrate a substantial spectral weight contribution from the vanadium d -electrons in the tunneling channel. When using a Bi_4Br_4 tip, we found that the FT intensities at the AFM ordering wavevectors also exhibit pronounced anisotropy that systematically reverses upon switching the bias polarity (Fig. R2-3 a-b). Again, this anisotropy reversal cannot be reproduced with non-magnetic PtIr tips. These

findings suggest that while the Bragg modulation and AFM order are located at the same vectors, the latter could contribute to the overall signatures when probed with spin-selective Bi_4Br_4 tips. By inverse Fourier transforming these AFM (Bragg) wavevectors and then constructing an averaged supercell, we obtain unidirectional real-space modulations: the intensities at V_x sites are higher at positive biases, whereas those at V_y sites become dominant at negative biases (**Fig. R2-3**). Thus, the difference map between positive and negative biases can directly reflect the spatial modulation of spin-resolved V_x and V_y sublattices that disappears in spin-summed measurement (**Fig. R2-3 k-t**), thus providing a potential real-space visualization of the AFM order.

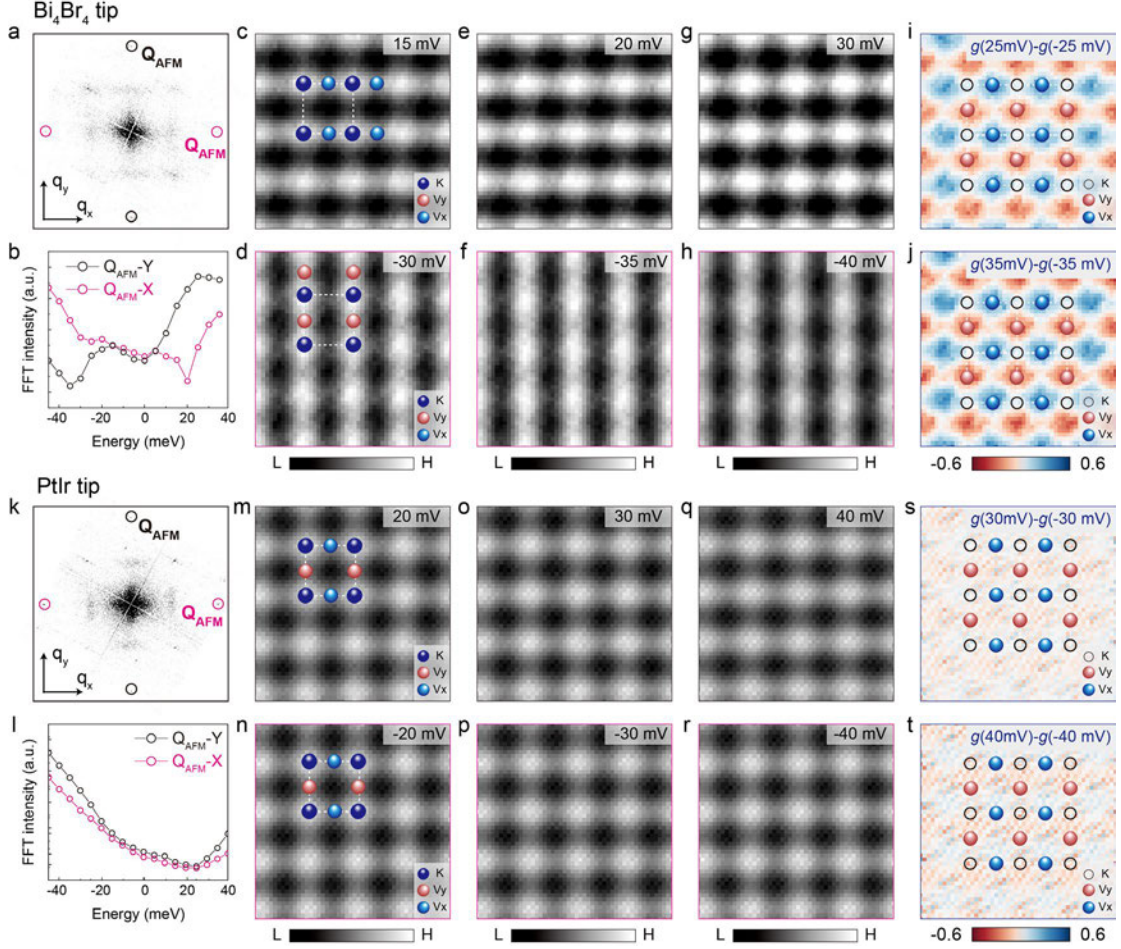


Fig. R2-3 Potential real-space mapping of the AFM order in $\text{KV}_2\text{Se}_2\text{O}$. **a, b**, FT and the bias-dependent intensities at the AFM wave vectors $Q_{\text{AFM-X}}$ and $Q_{\text{AFM-Y}}$, measured with a Bi_4Br_4 tip. **c-h**, Inverse FT of both the AFM peaks $Q_{\text{AFM-X}}$ and $Q_{\text{AFM-Y}}$, at representative positive and negative biases. The dominant contrast switches from the V_x sublattice at positive bias to the V_y sublattice at negative bias. The patterns are obtained with average super-cell algorithm. **i, j**, Difference map between two maps at ± 25 mV (**i**) and ± 35 mV (**j**). **k-l**, FT and AFM Fourier intensities measured with a PtIr tip. **m-r**, similar to these in **c-h**, but with a PtIr tip, showing no bias-polarity-dependent reversal. **s, t**, Difference maps at ± 30 and ± 40 mV, respectively. No visible AFM contrast is observed with the PtIr tip.

In conclusion, while we acknowledge that the defect can introduce a degree of structural asymmetry, our experiments unequivocally isolate the magnetic signature. The absence of $\pi/2$ rotation with PtIr tips, the opposite Zeeman shifts from a single defect, and the real-space mapping in clean regions collectively establish the altermagnetic spin texture in $\text{KV}_2\text{Se}_2\text{O}$ beyond a main structural symmetry-allowed scenario. We have added these new data in Fig. 4 and Extended Data Fig. 10, and added discussions to clarify this important point.

3. I note that the energies studied lie in close proximity to the SDW gap edges near the Fermi level. KV2Se2O hosts a $\sqrt{2} \times \sqrt{2}$ density-wave order, folding the original bands into two branches with a 20 meV gap. Such an order can generate anisotropic constant-energy contours, anisotropic nesting conditions, and hence intrinsically anisotropic, bias-dependent QPI intensities that have nothing to do with spin. Because band folding redistributes spectral weight asymmetrically about E_f , a charge/bond density wave can in principle produce a bias-asymmetric anisotropy in the QPI that superficially resembles the sought-after altermagnetic signature. As a result, the observed contrast and QPI anisotropy may not be purely a manifestation of d-wave spin splitting. So, the authors should disentangle the SDW-induced anisotropy and symmetry-breaking charge channel operating in the same energy window and at overlapping wavevectors from the altermagnetic spin splitting. If the authors quantitatively demonstrate that a spin-independent density-wave phase cannot reproduce the observed signatures, whether the bias-dependent anisotropy survives in the absence of altermagnetic order, or the QPI anisotropy persist over a broad eV-scale window for a d-wave altermagnet, or instead the temperature dependence of the QPI anisotropy above the density-wave transition T_N , then I would recommend that the QPI result be presented as corroborative evidence for the altermagnetic assignment.

Reply 2-3: We thank the referee for this comment. Following the referee's rigorous criteria, we have now conclusively isolated the altermagnetic spin signature from the SDW-induced charge channel through quantitative modeling and a comprehensive suite of control experiments.

Let's first discuss the SDW effect. We constructed a theoretical model incorporating both the AM and SDW orders within the enlarged $\sqrt{2}a \times \sqrt{2}a$ unit cell. Our calculations reveal a massive energy scale mismatch: the AM exchange parameter ($M \approx -0.9$ eV) dominates over the SDW gap ($M_{SDW} \approx -0.02$ eV) by more than an order of magnitude. Consequently, the maximum energy asymmetry introduced by the SDW band folding is extremely small (less than 0.1 meV, see Fig. R2-4a–d), suggesting its minor role in the observed anisotropy. Experimentally, the extracted QPI dispersions along the two directions (q_x^1 and q_y^1) do not exhibit any notable difference between them, which is consistent with this vanishingly small calculated SDW energy difference.

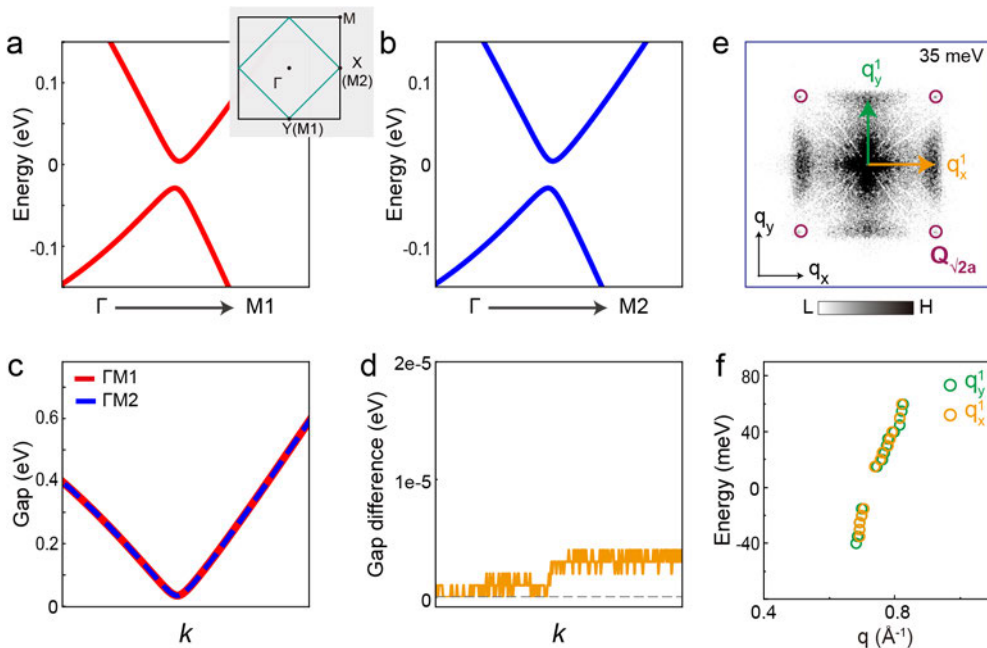


Fig. R2-4 Possible anisotropy introduced by the SDW order. **a, b**, Calculated band dispersions along the Γ -M1 (Γ -Y in the original BZ) and Γ -M2 (Γ -X) directions, respectively. **c**, Calculated SDW gaps along the two directions. **d**, Difference between the two gaps, showing that the SDW-induced energy anisotropy is negligible (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.

Second, we have performed measurements in the same SDW state (with the same temperature) using both Bi₄Br₄ tips and PtIr tips. A generic, spin-independent density-wave phase (which may modulate the spectral weight asymmetrically) therefore should inherently manifest in the local density of states (LDOS) probed by both STM tips. However, the current-direction dependent QPI anisotropy only appears when using the Bi₄Br₄ tips, and is totally absent for the non-magnetic PtIr tips. This suggests that even though the SDW opens a small gap in the altermagnetic bands, it provides a common reconstructed electronic background and the current-direction dependent QPI anisotropy (q_x^1/q_y^1) reflects the spin-selective tunneling channel with the Bi₄Br₄ tips.

Third, the current-direction-dependent QPI anisotropy (q_x^1/q_y^1) uniquely with the Bi₄Br₄ tip consistently persists up to at least $\pm 45\text{meV}$, see Fig.5, Extended Data Fig. 8 and 9. This energy scale extends well beyond the characteristic SDW gap edges ($\sim \pm 15\text{meV}$). This indicates that the SDW modulation is not a dominant source for the measured anisotropy between q_x and q_y .

Fourth, in all the measured samples, we found that the in-gap quasiparticle excitations (k_F oscillations) along the x and y directions always exhibit strictly opposite Zeeman shifts under external magnetic fields. This robust spin texture can be directly connected to the altermagnetic band structure, instead of a generic, spin-independent density wave order.

Finally, we turn to an additional set of control experiments. We would like to note that to measure high-quality, conclusive QPI data up to eV-scale, or at temperatures above the density-wave transition (110K) is in general not realistic for complex quantum materials as accepted by the STM community. Thus, we follow the referee's suggestion to check "whether the bias-dependent anisotropy survives in the absence of altermagnetic order". To do this, we performed analogous QPI measurements on another tetragonal oxide, Sr₂RuO₄. This material shares a strikingly similar quasi-1D dxz/dyz orbital texture and Fermi surface topology, but lacks AFM order and band spin-splitting (Fig. R2-1). Probed with the Bi₄Br₄ tip, Sr₂RuO₄ exhibits neither pronounced QPI anisotropy nor the current-direction-dependent $\pi/2$ rotation, in sharp contrast to the spin-splitting case of KV₂Se₂O. This negative control experiment satisfies the referee's criterion, and together with the opposite Zeeman shifts, confirming that the anisotropy requires genuine spin splitting.

We have added these new experiments and related discussions in the main text, and our calculations in supplementary Fig. S25.

4. The authors argue emphatically that the Bi₄Br₄ tip is most sensitive to in-plane spins, while the KV₂Se₂O moments are out-of-plane. The only $\sim 12\%$ effective polarization, and poor contrast genuinely sitting only marginally above background, naturally question the "smoking-gun" language, which is stronger than the demonstrated signal-to-noise warrants.

Reply2-4: We thank the reviewer for this comment. First of all, we accept the reviewer's suggestion to soften the language: we have tempered our language throughout the revised manuscript, replacing the absolute term "smoking-gun evidence" with the more objective and precise "direct experimental evidence."

However, from the perspective of spin-polarized STM, we must respectfully clarify that an effective spin contrast of $\sim 12\%$ is by no means "marginal" or "poor." As summarized in the SP-STM literature [e.g., Wiesendanger, *Rev. Mod. Phys.* **81**, 1495 (2009)], this value is comparable to, and in many cases exceeds, the magnetic asymmetries typically obtained in SP-STM measurements. The experimental signal itself is robust, physically consistent with the geometric projection of the out-of-plane AFM order, and strongly validated by our comprehensive data sets (e. g., Zeeman shifts).

Moreover, in our work, this contrast is achieved using a newly developed Bi₄Br₄ topological tip and is supported by a series of complementary controls across different magnetic systems. We therefore consider the demonstrated spin resolution and its sensitivity to in-plane spins to be an important advance in the STM probe development.

Minor Comments

1. The authors should state explicitly and prominently, in both abstract and main text, the distinction between the firmly demonstrated 2D monolayer d-wave altermagnetic order and the inferred bulk order, given the unresolved c-axis stacking.

Reply: We thank the referee for this precise and constructive suggestion. We fully agree that maintaining a rigorous distinction between the unambiguously resolved 2D in-plane order and a 3D bulk order is crucial for scientific accuracy. Because STM is a surface-sensitive local probe, our spectroscopic and real-space measurements directly and firmly establish the d-wave altermagnetic order within a 2D monolayer. We have now revised both the abstract and the main text to state this explicitly and prominently. We have also carefully clarified the language in our discussion to reflect that while the 2D monolayer order is directly visualized, the global 3D bulk order remains inferred due to the currently unresolved out-of-plane stacking.

2. Retain the moderated crediting of Ref. 62 already introduced, and ensure the four claimed advances over the SmB₆ platform are stated in the main text rather than only in the rebuttal.

Reply: We thank the reviewer for the suggestion. We have retained the moderated wording regarding Ref. 62 and have revised the main text to state the advances of the present platform over the SmB₆ platform.

Response to Referee #3

I read the revised manuscript and the very (with 38 manuscript pages and 61 letter pages maybe even too) elaborate reply the authors wrote as a rebuttal to the comments of the three referees. I definitely appreciate that the authors put a lot of work in order to better explain and discuss their scientific results, even by performing further experiments on a wide variety of what they consider magnetic reference materials. The vast majority of answers to the questions of all three referees are—to my opinion— fully convincing.

Yet, there are two main issues which led me to speak out in favor of rejecting the revised version of the manuscript. These issues are:

Reply: We deeply appreciate the referee's time and dedication in thoroughly reviewing our extensive rebuttal. We are highly encouraged by the referee's recognition of our immense experimental efforts, and are particularly grateful for the assessment that the vast majority of our previous answers are fully convincing.

We also take these two specific concerns very seriously. By incorporating highly targeted new experimental data and improving our presentation, we have comprehensively addressed both of these remaining points. We sincerely hope that these final, definitive clarifications will fully alleviate the referee's concerns and successfully reverse the recommendation in favor of publication.

(1) I still don't agree with the authors that the part on SP-STM using helical edge states shall be part of the main part of this manuscript. To my opinion, a strikingly similar concept has been used in Ref. 50 (Science 377, 1218 (2022)). The modification of this approach used in the manuscript under discussion here does not justify publication in a highly ranked journal like Nature. I read with interest that a similar statement was made by referee 2 in his/her introductory sentences: ``However, similar idea of spin sensor by STM has been validated before (Science 377, 1218–1222, 2022), thus reducing its originality. ``

Reply 3-1: We thank the referee for this comment. We fully acknowledge the pioneering work in Ref. 50, which first demonstrated the use of spin-momentum-locked topological states as an STM spin sensor. We completely agree with the referee's underlying scientific philosophy: for a highly ranked journal like Nature, the core novelty must be driven by a profound condensed matter physics breakthrough rather than merely an instrumental modification.

The novelty of this work does not only rely on the introduction of a QSH edge tip; the central claim and the fundamental novelty of this manuscript also lie in reporting the first direct, atomic-scale measurement of the spin-split band structure in a layered d-wave altermagnet. The profound impact and timeliness of this physics are further underscored by the fact that two other independent STM groups have recently adopted the well-established SmB_6 tips and Cr tips to study related altermagnetic compounds, and reached similar conclusions (arXiv:2603.21969 and arXiv:2604.18337 appearing several months after our work).

Following this specific advice, we have drastically streamlined the main text. The extensive discussions regarding the fabrication, characterization, and systematic calibration of the Bi_4Br_4 tips have been mainly moved to the Methods and Supplementary Information. The main text now retains only the absolute minimum information required to establish the working principle and the spin-quantization axis of the probe, ensuring that the primary narrative focuses squarely on the intrinsic

altermagnetic properties of $\text{KV}_2\text{Se}_2\text{O}$.

Moreover, owing to the quasi-one-dimensional character of Bi_4Br_4 and the nearly conserved S_z of its edge states, the spin polarization of the active tunneling channel has a well-defined orientation with respect to the crystallographic axes. Combining our calculations with systematic measurements on several magnetic materials, we can determine the spin direction to which the Bi_4Br_4 tip is predominantly sensitive (mainly in-plane direction). This is an advancement for a spin resolved probe.

We believe that by restructuring the manuscript as you suggested, the pioneering altermagnetic physics of $\text{KV}_2\text{Se}_2\text{O}$ now takes its rightful place, fully justifying its consideration for Nature.

(2) In my first review I asked the question, why the authors not simply image the AFM spin structure rather than concluding it indirectly from an ``opposite magnetic field-induced shift of the spectra shown Fig. 5g and h''. The authors respond by claiming an experimental difficulty, i.e. ``the Neel vector (QAFM) coincides exactly with either the atomic Bragg modulation or the second-harmonic frequency of the surface structural reconstruction. Consequently, the magnetic signals in q-space are spectrally overlapped with structural contributions. Given that both signals can exhibit complex and potentially energy-dependent changes, it is difficult to unambiguously disentangle the intrinsic magnetic contrast from structural artifacts using real-space mapping techniques.''

I have to admit that this explanation doesn't resonate with me. Magnetism induced modifications of the total current are usually verified by inverting the magnetization direction of the magnetic tip, as convincingly shown in Fig. R3-7. For their special SP-STM method with helical edge states this would respond to a comparison of data acquired at small positive and negative bias (around ± 30 to 50mV).

Reply3-2: We deeply appreciate the referee's persistence and profound physical insight on this point. We must admit that the referee's proposed methodology, i. e., leveraging the bias-dependent contrast inversion to separate the magnetic signal from the structural background, provided one key to overcoming our previous hesitation. Encouraged by your specific suggestion, we pushed our experimental data analysis further and have now successfully achieved the potential, direct real-space visualization of the AFM order.

Following the referee's intuition, we carefully examined the real-space data, particularly within the suggested energy window of ± 30 to 50 mV , where our QPI data indicate a substantial spectral weight contribution from the vanadium d-electrons.

Indeed, when utilizing the spin-polarized Bi_4Br_4 tip, the Fourier intensities at the AFM ordering wavevectors exhibit a pronounced anisotropy between the x and y directions. Crucially, exactly as the referee astutely predicted, this anisotropy systematically inverts upon reversing the bias polarity. By inverse Fourier transforming the peaks at AFM vectors (checked with different FT windows) and constructing an averaged supercell, we can now directly visualize the unidirectional real-space modulations: the modulation intensity is dominant along the x-direction at positive biases, and dominantly along the y-direction at negative biases, because the signatures of V_x and V_y sublattices are alternately contributed more to the tunneling (Fig. R3-1). Furthermore, we confirm that this signature is indeed maximized at biases around $\pm 30\text{ mV}$, in perfect agreement with the referee's expectations.

To rule out the structural artifacts we previously concerned ourselves with, we performed the identical measurement and FFT filtering utilizing a non-magnetic PtIr tip. The bias-dependent spatial anisotropy is entirely absent. This stark comparison identifies that the bias-dependent contrast inversion arises from the spin-selective tunneling into the alternating V_x and V_y sublattices.

We are incredibly grateful to the referee; the specific guidance has enabled us to provide this direct real-space imaging of the AFM order, which stands as one additional piece of evidence in the revised manuscript. These data have now been added to Extended Data Fig. 10, and we have added a thoughtful discussion in the main text.

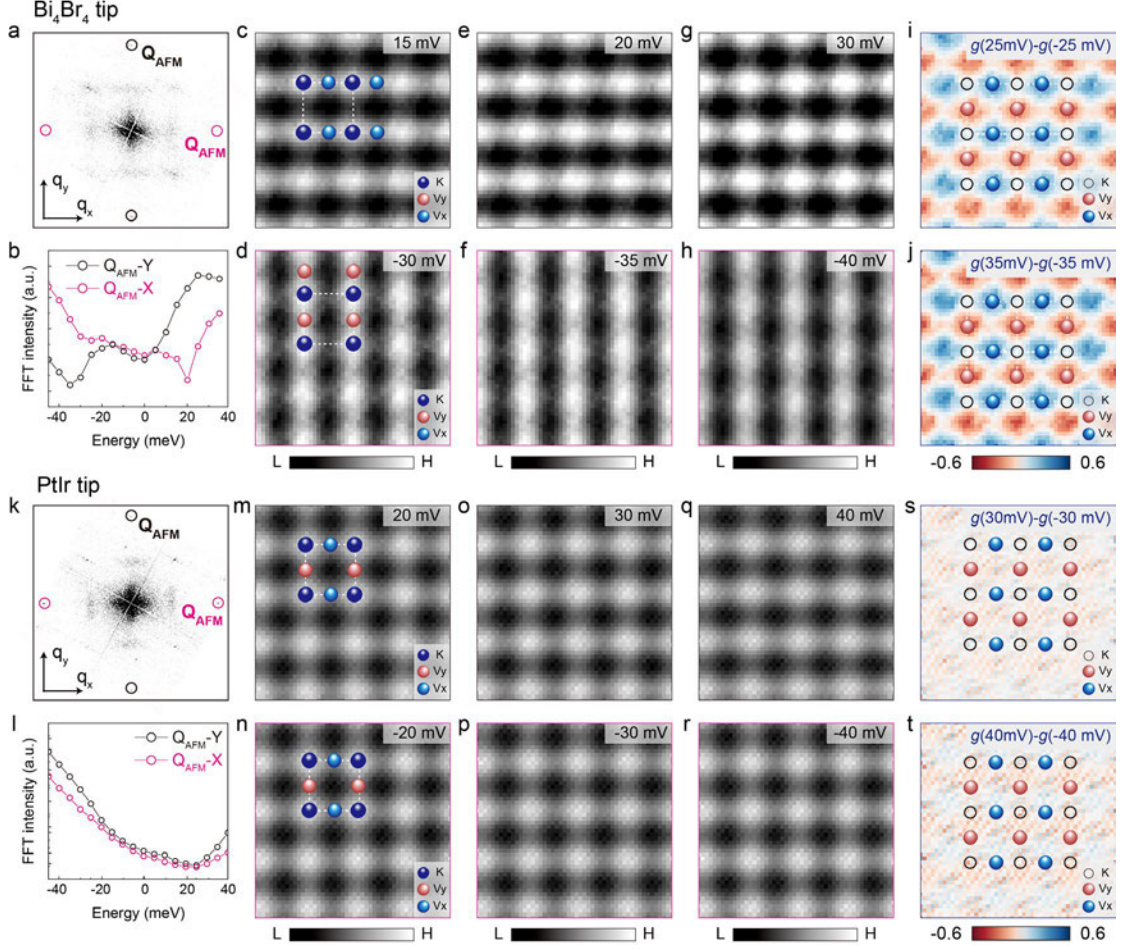


Fig. R3-1 Potential real-space mapping of the AFM order in $\text{KV}_2\text{Se}_2\text{O}$. **a, b**, FT and the bias-dependent intensities at the AFM wave vectors $Q_{\text{AFM}}-X$ and $Q_{\text{AFM}}-Y$, measured with a Bi_4Br_4 tip. **c-h**, Inverse FT of both the AFM peaks $Q_{\text{AFM}}-X$ and $Q_{\text{AFM}}-Y$, at representative positive and negative biases. The dominant contrast switches from the V_x sublattice at positive bias to the V_y sublattice at negative bias. The patterns are obtained with average super-cell algorithm. **i, j**, Difference map between two maps at ± 25 mV (**i**) and ± 35 mV (**j**). **k-l**, FT and AFM Fourier intensities measured with a PtIr tip. **m-r**, similar to these in **c-h**, but with a PtIr tip, showing no bias-polarity-dependent reversal. **s, t**, Difference maps at ± 30 and ± 40 mV, respectively. No visible AFM contrast is observed with the PtIr tip.

Overall, my comment is very similar to the criticism #11 raised by referee 2 in the first round of review. Here, he/she wrote the introductory sentence: "The interpretation that the arrow-shaped defects reveal the intrinsic magnetic ground state is debatable." The comment I made in the preceding paragraph touches exactly the same point, but approaches it from a more technical

perspective.

Reply: Besides the direct imaging of AFM order in defect-free regions as mentioned above, we would like to provide another piece of evidence to address the referee's concern.

Besides the observed QPI anisotropy with Bi₄Br₄ tip, we also focus on the in-gap quasiparticle excitations whose real-space modulation period strictly follows π/k_F of the d_{xz}/d_{yz} bands. This characteristic period indicates that these are Friedel-like quasiparticle oscillations that carry crucial information about the underlying Fermi surface. They always exhibit direction-dependent, opposite Zeeman shifts in four measured samples, thus providing an independent real-space probe of opposite spin polarizations in quasiparticle excitations associated with the underlying Fermi surface.

Crucially, we have isolated a single defect that simultaneously induces these quasiparticle excitations along both the x and y directions (Fig. R3-2). When subjected to an out-of-plane magnetic field, these orthogonal directional excitations exhibit strictly opposite Zeeman shifts.

This specific orthogonal Zeeman dichotomy highly unfavors a defect-localized magnetic origin. A single localized moment is difficult to simultaneously produce opposite spin-polarizations in orthogonal spatial directions without alternating spin-splitting along X and Y. Together with their π/k_F modulation period, it supports that these excitations reflect the spin-polarized electronic structure of the host material.

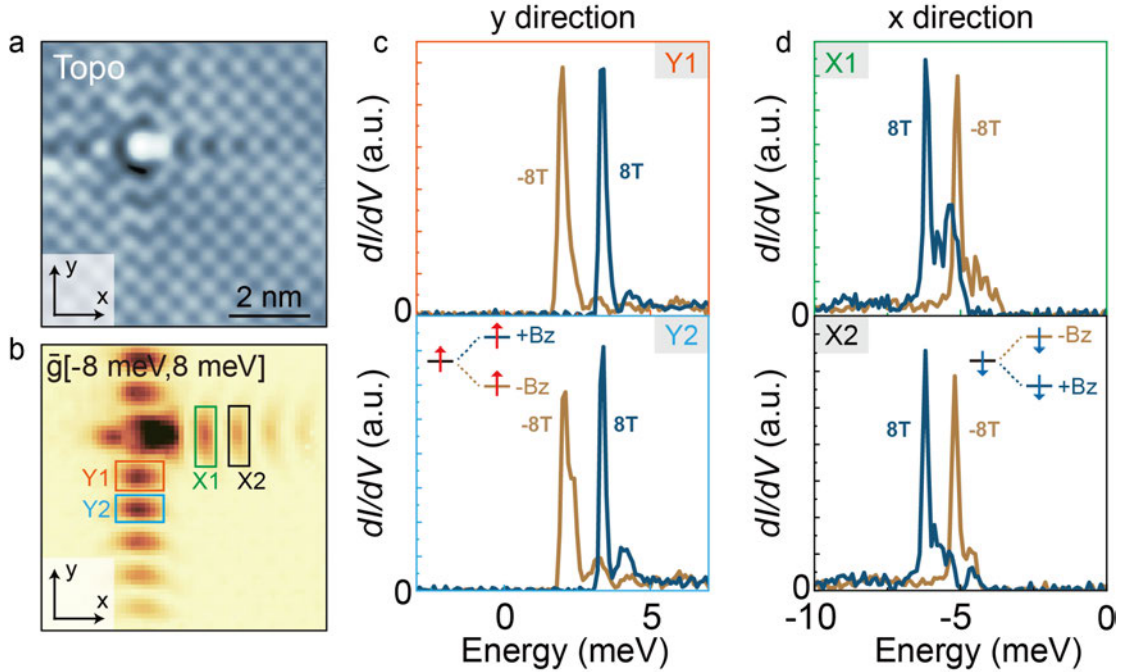


Fig. R3-2 Opposite Zeeman shifts near a single defect. **a**, topography of a defect that induces in-gap states along both x and y directions. **b**, Energy-averaged conductance map showing quasiparticle excitations induced simultaneously along both the x and y directions. The colored rectangles mark the locations at which the spectra in (c) and (d) were acquired. **c**, **d**, Magnetic-field-dependent dI/dV spectra of the in-gap states along the x and y directions, respectively, showing opposite Zeeman shifts upon out-of-plane magnetic fields.

In conclusion, combining the clean-region real-space imaging of the AFM order with the single-defect orthogonal Zeeman spectroscopy, our full dataset provides a robust, multi-faceted validation of the intrinsic altermagnetic ground state in KV₂Se₂O. We have added these new data in Fig. 4 and

[explicitly added this clarifying discussion to the revised manuscript to address this important point.](#)

There are further, probably minor points which should be clarified before the manuscript is published:

(a) ``The extracted energy dispersion well matches the theoretical calculations with in-plane Néel AFM order (Fig. 4d; Extended Data Fig. 8)``.

It remains unclear for which band and at which point in the BZ the claimed good agreement is obtained.

Reply: We thank the referee for pointing out this omission. The experimental energy dispersion presented in Fig. 4d directly corresponds to the quasi-1D V-dxz orbital-dominated band dispersing along the high-symmetry Γ -X direction of the Brillouin zone (as illustrated in Fig. R3-3). In our theoretical model of the in-plane Néel AFM state, this V-dxz-derived band forms a quasi-one-dimensional, electron-like Fermi-surface pocket centered at Γ along the Γ -X direction. The corresponding V-dyz-derived band produces an analogous feature along the orthogonal Γ -Y direction.

[To ensure absolute clarity, we have revised the main text and updated the caption of Fig. 4d to explicitly state that the extracted QPI dispersion traces the quasi-1D V-dxz \(dyz\) dominated Fermi pocket along the \$\Gamma\$ -X\(Y\) direction.](#)

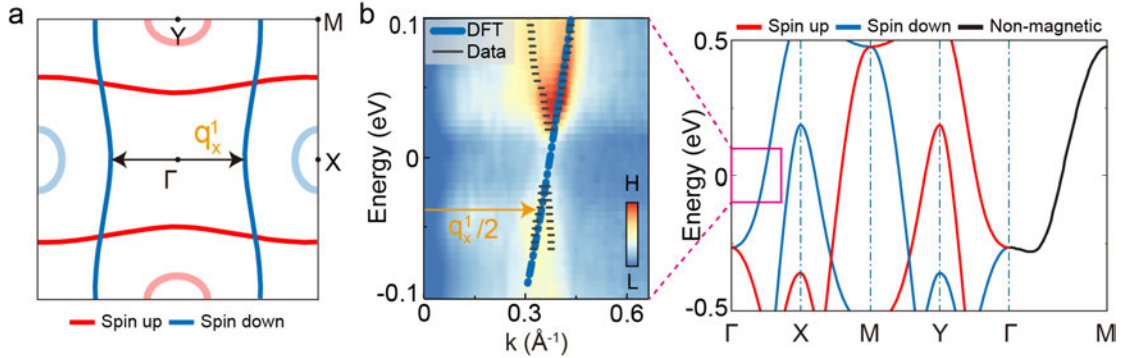


Fig. R3-3 **a**, Schematic of the pristine Brillouin zone and spin-split Fermi-surface contours. The scattering vector q_x^1 connects the V-dxz-dominated bands along the Γ -X direction. **b**, Experimental QPI dispersion along Γ -X direction, overlaid with the calculated dispersion of the quasi-one-dimensional V-dxz-dominated band (blue line). The right panel shows the corresponding calculated band structure along the high-symmetry directions, with spin-up, spin-down, and nonmagnetic bands indicated in red, blue, and black, respectively.

(b) How does the calculated band structure presented in Fig. S20 correspond to the band structure plotted in Fig. S28? I cannot see how the features along M—Gamma—M of Fig. S20 derive from S28.

Reply 3-2: We thank the referee for pointing out that this point. The core difference is that Fig. S28 displays the band structure in the pristine (unreconstructed) Brillouin zone (BZ), whereas Fig. S20 presents the electronic states within the reduced Brillouin zone (rBZ) resulting from the $2a \times \sqrt{2}a$ density wave reconstruction.

The band structure along the $M_1 - \Gamma - M_2$ path of the reduced Brillouin zone in Fig. S20 is derived from the pristine band structure (Fig. S28) with folding induced by the $\sqrt{2}a \times \sqrt{2}a$ SDW reconstruction (as illustrated in Fig. R3-4). Specifically, along the $\Gamma - M_2$ direction in the reduced BZ (Fig. S20), the pristine $\Gamma - X$ and $M - Y$ paths (as marked by orange rectangles in Fig. R3-4b) are folded onto the same momentum region. Consequently, the two bands of d_{x1} (along $\Gamma - X$) and d_{x2} (along $M - Y$) (see Fig. R3-4b) cross each other and open a gap. Likewise, the symmetry-related $\Gamma - M_1$ direction contains the folded contributions from the pristine $\Gamma - Y$ and $M - X$ paths.

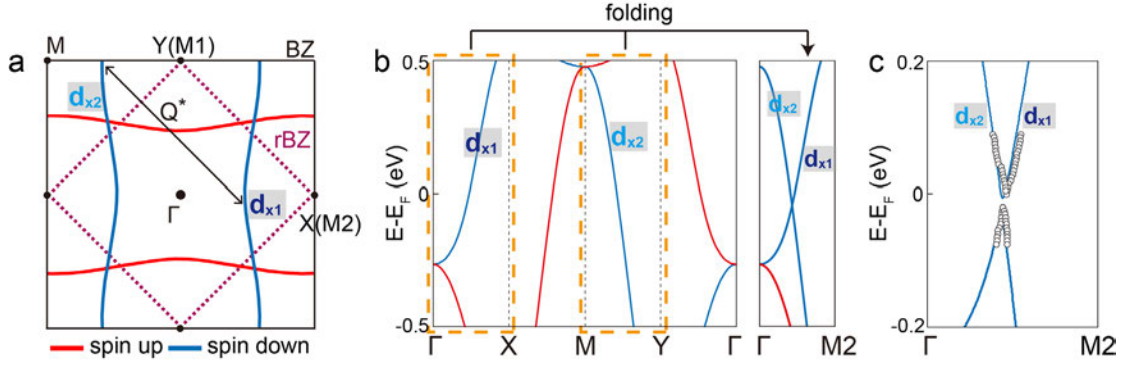


Fig. R3-4 **a**, Schematic of the pristine Brillouin zone (BZ, black) and the reduced Brillouin zone (rBZ, dashed purple lines) generated by the density-wave reconstruction. The red and blue curves denote the spin-up and spin-down Fermi-surface sheets, respectively. The bands labeled d_{x1} and d_{x2} are folded into the same momentum region by the reconstruction wavevector Q^* . **b**, Pristine band structure along the $\Gamma-X-M-Y-\Gamma$ path (left), and its folded counterpart along the $\Gamma-M_2$ direction of the rBZ (right). Along $\Gamma-M_2$, the pristine $\Gamma-X$ and $M-Y$ bands, are folded onto one another. **c**, Folded band structure along $\Gamma-M_2$, showing the hybridization gap opened by the crossing of the folded d_{x1} and d_{x2} bands. Open circles denote the experimentally extracted dispersion.

To eliminate any ambiguity for the readers, we have revised the Supplementary Information to include an explicit zone-folding schematic. This new diagram clearly visualizes the momentum-space mapping, providing a step-by-step derivation of how the reconstructed, gapped bands in Fig. S20 originate from the pristine dispersions in Fig. S28.

(c) I explicitly thank the authors for their figure R3-11 which shows a much smaller (structural) signal in the added signal that in the larger (spin) difference signal. Yet, magnetic contrasts still appear to be masked by additional contributions, which may originate from crystalline imperfections or other effect, such as those discussed by referee B. One particular apparent example are the line sections presented in Fig. R2-1e for +30mV and -30mV. While the modulation with a $2a$ periodicity is nicely visible at +30mV, the data obtained at -30mV show a strongly distorted signal at distances 3-7nm (x axis). In summery it appears that numerous Bi4Br4 tips are capable of detecting a spin-related signal, as convincingly shown in Fig. R4-1, but it remains questionable if other parameters, such as the orbital character of the tip and sample states which potentially complicate data interpretation substantially, also play an important role.

Reply: We highly appreciate the referee's meticulous inspection of our raw data and the positive feedback on our difference-signal analysis. The referee raises an excellent point regarding the potential complications arising from sample imperfections and the potential orbital effect.

First, regarding the distortion observed in the $Fe_{1+x}Te$ line profile: This distortion is more likely an intrinsic spatial inhomogeneity of the sample itself. As recently established by high-resolution STM

studies (Nature 652, 342–348 (2026)), the AFM order and its associated spin contrast in Fe_{1+x}Te are highly sensitive to the local concentration of interstitial Fe atoms. These interstitial defects can inherently induce strong spatial electronic inhomogeneities at the nanoscale.

Second, regarding the broader role of orbital character: We certainly do not claim that orbital tunneling matrix elements are totally absent. However, our comprehensive dataset unambiguously dictates that spin polarization, rather than orbital character, is the dominant driver of the observed contrast reversals. We decouple the spin and orbital contributions through three highly constrained lines of evidence.

First, we provide a direct, non-magnetic control experiment. We obtained new QPI data on another tetragonal oxide, Sr_2RuO_4 , using the Bi_4Br_4 tip. Its well-known Fermi surface features a quasi-1D pocket derived from dxz and dyz orbitals, strikingly similar to that of $\text{KV}_2\text{Se}_2\text{O}$, but without AFM order and band spin-splitting. Crucially, our results on Sr_2RuO_4 exhibit neither pronounced QPI anisotropy between the dxz/dyz orbitals nor any tunneling-current-direction-dependent contrast rotation (Fig. R3-5). If the $\pi/2$ rotation in $\text{KV}_2\text{Se}_2\text{O}$ were an inherent consequence of dxz/dyz orbital matrix elements, it would necessarily manifest in Sr_2RuO_4 . Its absence rules out the orbital-selection effect as the primary driver.

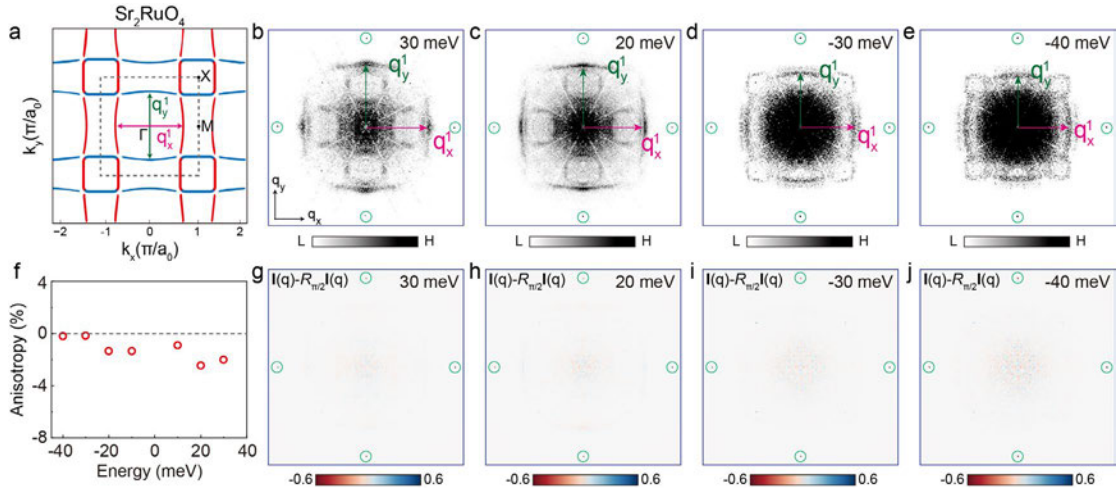


Fig. R3-5 QPI patterns on nonmagnetic Sr_2RuO_4 using a Bi_4Br_4 tip. **a**, CECs of Sr_2RuO_4 near E_F including quasi-1D pockets derived from the d_{xz} and d_{yz} orbitals, whose size and orbital texture are similar to those in $\text{KV}_2\text{Se}_2\text{O}$. **b-e**, QPI patterns at representative energies near E_F . **f**, The calculated anisotropy of QPI intensity between d_{xz} and d_{yz} bands, $(I(q_x^1) - I(q_y^1))/(I(q_x^1) + I(q_y^1))$, as a function of energy. **g-j**, Corresponding contrast maps of rotating $\pi/2$. No pronounced QPI anisotropy reversal is observed, in sharp contrast to the case in $\text{KV}_2\text{Se}_2\text{O}$.

Second, we exploit a magnetic field-driven spin-flop transition to show that the measured contrast is sensitive to changes in spin structure. In the A-type AFM compound FeGe , we applied an 8 T magnetic field to flop the spins from a predominantly out-of-plane to an in-plane configuration, which resulted in a clear, concurrent change in the LDOS contrast. Given that the pure orbital features of these two layers are identical, this contrast modulation is thus tied to the magnetic structure of FeGe (Extended Data Fig. 4).

Third, we demonstrate universal spin-selectivity across diverse material platforms. We have successfully deployed the Bi_4Br_4 tips to resolve spin-dependent responses across three distinct magnetic systems: the π -phase shift of spin stripes in bicollinear Fe_{1+x}Te , the inter-layer contrast in A-type AFM FeGe , and the highly anisotropic X- and Y-QPI responses in the d-wave altermagnet

KV₂Se₂O. Given that these compounds possess different point group symmetries, crystalline lattices, and orbital features, this universal efficacy strongly argues against an orbital-derived mechanism. Instead, it constitutes compelling evidence for the intrinsic spin-selective nature of the tip. We have added these experiments and related discussions in the main text.

In summary, I cannot support publication in Nature. I suggest scaling back the scientific scope somewhat, specifically shifting from the direct (atomic-scale) observation of the antiferromagnetic structure of an antiferromagnet to the observation of spin order in the vicinity of structural defects.

Reply: We thank the referee for their rigorous evaluation and for pushing us to achieve an even higher standard of experimental proof. However, in light of the definitive new data provided in this second revision, we respectfully decline the suggestion to scale back the scientific scope of our manuscript.

The referee's primary reservation—that our observations might merely reflect localized spin order in the vicinity of structural defects—has now been directly addressed by two independent, highly constrained experimental facts.

1. Direct Imaging of AFM order in defect-free regions. By leveraging the bias-dependent contrast inversion, we have now achieved the direct, real-space visualization of the AFM order in defect-free regions. This definitively proves that the alternating spin contrast is a global, intrinsic property of the crystal, not an artifact pinned to or induced by defects.
2. Opposite Zeeman shifts from one defect. The orthogonal Zeeman dichotomy measurements on a single defect rule out a defect-localized magnetic origin. Together with their π/k_F modulation period, it supports that these excitations reflect the spin-polarized electronic structure of the host material.

Therefore, our findings now go beyond the "observation of spin order in the vicinity of structural defects." Supported by extensive multi-material cross-checks, this work constitutes a genuine, atomic-scale visualization of the antiferromagnetic ground state.

We believe that these critical new breakthroughs can dismantle the remaining reservations. We are confident that the robust scientific scope of this manuscript is now substantiated, directly answering the fundamental physics questions appropriate for the broad and high-impact readership of Nature. We sincerely hope these final clarifications will successfully reverse your recommendation in favor of publication.

Response to Referee #1

I would like to thank the authors for extensively considering my comments and requests. My main concern of the previous round aimed at characterizing which component of spin is mainly measured, in-plane or out-of-plane. The authors now provide field-dependent measurements in Fig. R1. Indeed, the data support the measurement of a predominantly out-of-plane magnetic response, which exhibits opposite signs of the field-induced shifts for defect-associated states extending along x and y. This strengthens the interpretation that the studied features are indeed mainly spin-polarized as claimed.

Reply: We thank the referee for carefully considering our revised manuscript and for recognizing that the vector-field measurements support a predominantly out-of-plane magnetic response and strengthen the interpretation that the observed features are mainly spin-polarized. Below, following the referee's constructive suggestions, we address the remaining quantitative questions concerning the spin-polarization direction, provide quantitative constraints on the in-plane magnetic response, and revise our interpretation to acknowledge that even a smaller in-plane component may contribute appreciably, and potentially comparably, to the measured QPI contrast in our Bi₄Br₄-tip STM configuration.

Quantitatively, I am not convinced because, as the FeGe study with spin-flop contrast showed, the spin sensitivity is much stronger for an in-plane polarization. The authors state, applying an identical field magnitude within the in-plane (B_x and B_y) yields no resolvable Zeeman shift whatsoever.“ but in Fig. R1d the X2 feature does seem to shift for applied B_y . This would need fits to make a more precise statement.

Reply: We thank the referee for this careful observation and constructive suggestion. We agree that our previous statement in the rebuttal that there was “no resolvable Zeeman shift whatsoever” under in-plane fields was too categorical. We have now quantified the field-induced shifts over multiple datasets and revised the statement accordingly so that the data are no longer described as excluding an in-plane response.

To address this point quantitatively, we performed additional field-dependent measurements of defect-associated excitations in more 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 (Fig. R1). 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. We have therefore revised the manuscript to describe the

defect-associated excitations as exhibiting a predominantly, rather than exclusively, out-of-plane magnetic response. On Page 5, it reads: *Averaged over multiple measured regions, we find that the mean B_x and B_y responses are approximately 5.8% and 8.5% of the mean B_z response, respectively (Fig. S20). These results establish a predominantly out-of-plane magnetic response, consistent with the out-of-plane moment orientation reported by bulk neutron diffraction (52), while allowing smaller in-plane components.*

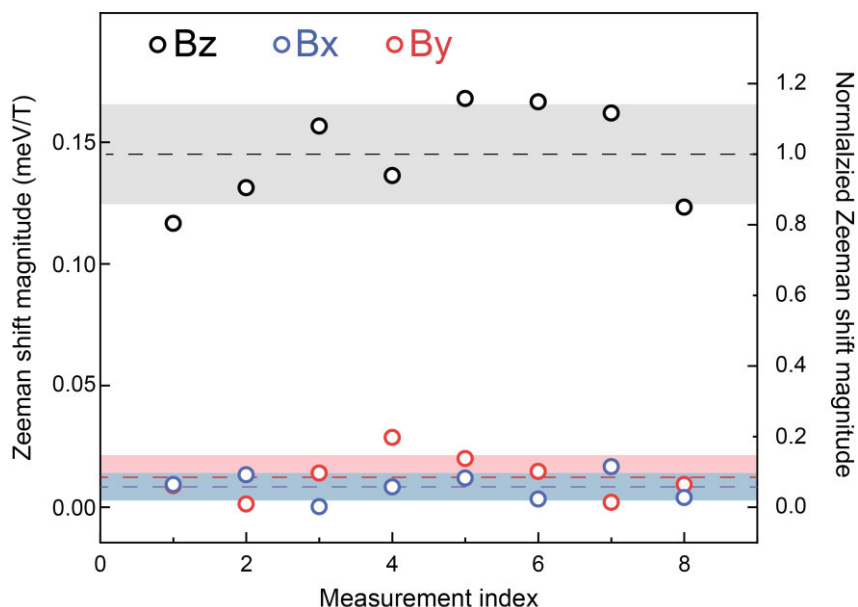


Fig. R1 Quantitative anisotropy of the magnetic-field response of defect-associated excitations in KV_2Se_2O . Field-induced energy shifts per unit field extracted from multiple regions for fields applied along B_z (black), B_x (blue), and B_y (red). For each region, the shift is obtained from fits to 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. The mean B_x and B_y responses are 5.8% and 8.5% of the mean B_z response, respectively; the largest observed B_y response is close to 20%. These data demonstrate a B_z -dominated response while allowing a smaller in-plane component.

When I manually read out the shift, it appears to be 0.2 of the response to the B_z -aligned field. If this approximate ratio of field-induced shifts reflects the corresponding sample spin-polarization components, the stated tip geometry would make their contributions to the tunnelling contrast comparable: 0.2 is similar to the geometric factor $\cos(80^\circ) \sim 0.17$ that reduces the out-of-plane component. The data therefore do not yet establish that an in-plane contribution is negligible in the measured tunnelling contrast. This requires a quantitative bound on the in-plane magnetic response and an explanation of how this bound applies to the states contributing to the q_x and q_y QPI features.

Reply: We agree with the referee that the smaller in-plane magnetic response observed in Fig. R1 cannot be regarded as negligible in the measured QPI contrast, because the Bi_4Br_4 -tip junction is more sensitive to in-plane sample spins. We also agree with the referee that, the measured field-dependent Zeeman shift might not be directly identified as the ratio of in-plane and out-of-plane spin components of the sample. Thus, the following geometric comparison should be regarded only as an estimation, rather than a quantitative decomposition of the QPI contrast.

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. 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 indeed comparable to the out-of-plane projection, as pointed out by the referee.

We also clarify how this comparison relates to the q_x^1 and q_y^1 QPI features. The defect-associated excitations exhibit directional oscillations with a period close to π/k_F and are linked to the same quasi-1D d_{xz} and d_{yz} electronic structure that gives rise to the q_x^1 and q_y^1 scattering features. This provides a qualitative connection between the vector-field spectroscopy and the QPI measurements. Nevertheless, the anisotropic spin-polarization ratio of local defect resonances and the extended QPI channels might not be identical. We therefore do not use the measured Zeeman-shift ratios as a quantitative calibration of the spin components contributing to the QPI intensities.

Accordingly, we totally agree with the referee that because the junction is more sensitive to in-plane sample spins, even the smaller in-plane response allowed by Fig. R1 may contribute appreciably—and even comparably—to the measured QPI contrast. We have revised the manuscript to describe the defect-associated excitations as exhibiting a predominantly out-of-plane magnetic response, while acknowledging that the QPI contrast measured with the Bi_4Br_4 tip can contain contributions from both in-plane and out-of-plane spin components. On page 8, it reads: “*For $\text{KV}_2\text{Se}_2\text{O}$, vector-field spectroscopy establishes a predominantly out-of-plane magnetic response of the defect-associated excitations, while allowing smaller in-plane responses. These excitations exhibit directional oscillations with a period close to π/k_F and are associated with the same quasi-1D V d_{xz} - and d_{yz} -derived bands that generate the orthogonal QPI features. However, their field-response ratios do not directly determine the spin-component weights of the extended states contributing to QPI. A typical tip tilting angle of 10° relative to the surface normal in our experimental setup (Methods) can introduce sensitivity to out-of-plane sample spins while retaining stronger in-plane sensitivity. Consequently, even a smaller in-plane sample polarization could contribute appreciably, and potentially comparably, to the measured QPI contrast (Supplementary Note 7).*”

I am not sure I fully understand their quantitative argument. One reading would be that there is a $\cos(80^\circ) \sim 0.17$ geometric factor with a 0.85 tip Sz polarization and additionally a $\cos(10^\circ) \sim 0.98$ geometric factor with roughly a 0.15 tip in-plane spin polarization. The total is ~ 0.29 , which would match their 20-30%. However, this assumes that the lack of Sz tip polarization translates into an in-plane polarization available to enhance the spin contrast, which is generally not true because the total spin polarization does not need to vectorially add up to a quantized value.

Reply: We thank the referee for identifying this problem and agree that our previous estimate of a 20–30% effective tip polarization was not justified. Our earlier argument incorrectly treated the

reduction of the calculated S_z value as a compensating in-plane spin component. A normalized expectation value of approximately 0.85 for S_z does not imply that the remaining 0.15 constitutes a well-defined in-plane polarization, which is not physically justified as pointed by the referee. *This 20–30% estimate was provided in rebuttal letter; and we have checked that it is not in the revised manuscript.*

The approximately 10° tip tilt is now invoked only to explain qualitatively why the Bi₄Br₄-tip junction can retain finite sensitivity to an out-of-plane sample component; it is not used to determine the magnitude of the tip polarization. We also clarify that the approximately 12% quantity extracted from the QPI data is an effective junction-dependent QPI contrast, rather than a direct quantitative measurement. A microscopic decomposition of this contrast would require a detailed tunnelling model beyond the scope of the present work.

Using Sr₂RuO₄, whose Fermi-surface segments have pronounced orbital character, the authors find a d-wave QPI anisotropy with a magnitude of at most approximately 3%, compared with 10–18% in KV₂Se₂O. The authors provide in figure R2a a suggestive nesting that highlights the similarity to KV₂Se₂O.

Since the warping of Sr₂RuO₄ appears smaller except from the hybridization points, one could also consider a continuum of scattering vectors parallel to qx but connecting along blue Fermi surface instead of red ones. This might imply that the QPI receives contributions from more than one orbital. The authors do not seem to provide a QPI calculation to show its orbital character. Such a calculation would establish the orbital contrast between the compared QPI features and help assess the tip's orbital sensitivity.

To get an impression, what is known on Sr₂RuO₄, one may consider [Fig. 5, Kreisel et al., <https://doi.org/10.1038/s41535-021-00401-x>], which reports on the orbital-textured QPI based on a tight-binding model with surface reconstruction. However, the calculated arcs do not appear to reproduce the pronounced axial intensity maxima seen in the rebuttal. In [Fig. 7, Bhattacharyya et al., <https://doi.org/10.1103/PhysRevB.107.144505>] a central intensity peak at qx in the dxz QPI feature has also a contribution from dyz. Of course, these references are at different modelling conditions and can only give a qualitative idea. So, the orbital contrast used to extract the rebuttal's anisotropy remains unquantified. From my visual inspection of the outer regions of the arcs no clear anisotropy is visible.

Thus, even though the orbital texture of QPI is not quantified and might be not fully comparable to KV₂Se₂O, I agree that Sr₂RuO₄ provides meaningful evidence against a generic dominant orbital sensitivity of Bi₄Br₄ tips.

Reply: We appreciate the referee's careful assessment of our Sr₂RuO₄ control experiment and their recognition that it provides meaningful evidence against a generic, dominant orbital sensitivity of the Bi₄Br₄ tips.

The Fermi surface of Sr₂RuO₄ has been extensively studied by theoretical calculations and ARPES measurements in the past decade (for example, Phys. Rev. B 88, 134519 (2013), Phys. Rev. Lett. 112, 127002 (2014), Phys. Rev. B 107, 144505 (2023), Nat. Commun. 15, 9521 (2024)), 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 Wang et al. [Nature Physics 13, 799 – 805 (2017)]. 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.

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.* [npj Quantum Mater. 6, 100 (2021)] (Fig. R2). In the calculation, 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} . 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. *We have discussed with Dr. Andreas Kreisel and checked these results. Thus, the main conclusion is valid and we have modified our related statements with “main orbital character”.*

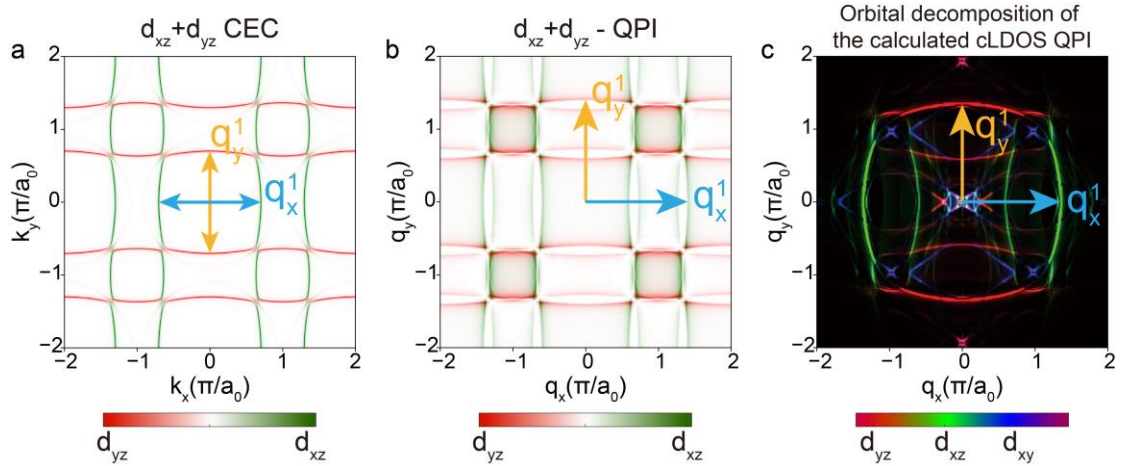


Fig. R2 Main orbital characters of quasiparticle interference in Sr_2RuO_4 . **a**, Orbital-resolved spectral density at the Fermi level for the d_{xz} and d_{yz} states, shown in green and red, respectively. Here, we neglect the contribution from the in-plane d_{xy} orbital, which is typically difficult to detect in STM-based QPI measurements owing to its weak coupling to the STM tip (see, e.g., Nature Physics 13, 799–805 (2017), Phys. Rev. B 107, 144505 (2023), Proc. Natl. Acad. Sci. U.S.A. 117, 5222–5227 (2020)). 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 Nature Physics 13, 799–805 (2017). **c**, Orbital-decomposition QPI pattern with different modeling that is reproduced from Fig. 5 of Kreisel *et al.*, npj Quantum Materials 6, 100 (2021). 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**.

Finally, 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. R3). In all three cases, the anisotropy remains below approximately 3% and does not reverse

sign with bias polarity.

In summary, we totally agree with the referee that different modelling conditions could give rise to different quantitative details in the QPI calculations and a complete quantitative modeling analysis is difficult to achieve, but the main orbital features for the QPI signatures we focused on here is consistently obtained in these models. Thus, these QPI calculations support the referee's conclusion that "Sr₂RuO₄ provides meaningful evidence against a generic dominant orbital sensitivity of Bi₄Br₄ tips".

In the revised version, on page 7, we added a discussion of "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 KV₂Se₂O", and added the calculations and new analysis as Supplementary Note 11.

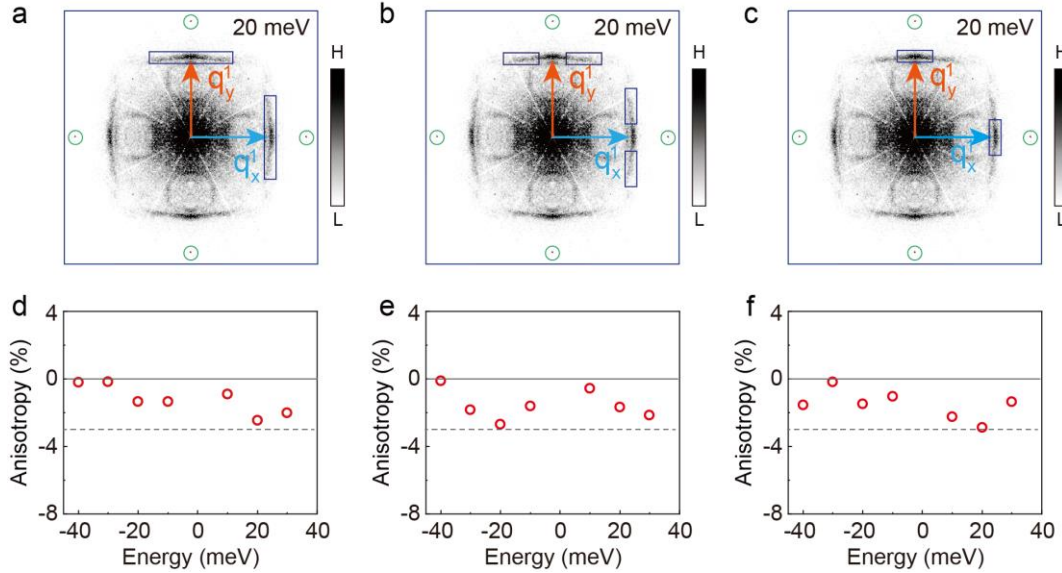


Fig. R3 Robustness of the Sr₂RuO₄ QPI anisotropy against integration-window selection. **a–c**, QPI maps at 20 meV with black rectangles marking the full outer QPI segments (**a**), off-axis portions excluding the axial maxima (**b**), and axial maxima alone (**c**). **d–f**, Corresponding energy-dependent 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.

In conclusion, an in-plane polarization of the QPI signal is potentially picked up with a signal strength comparable to that of the out-of-plane component. As the defect-associated resonances are indeed responsive to Bz and the orbital selectivity has been established as less likely, I would regard some sensitivity to the out-of-plane spin component with a transparent discussion as sufficient. But as of now a "direct experimental evidence" claim applies mostly to the d-wave pattern and not to the proposed out-of-plane spin texture.

Reply: We express our deepest gratitude to the referees for their thorough examination of this work and for providing us with constructive comments.

We agree with the referee for this important clarification. We now reserve the phrase 'direct

experimental evidence' for the observed d-wave, bias-reversing spin-dependent pattern. The vector-field Zeeman spectroscopy is described more specifically as evidence that the defect-associated excitations are mainly out-of-plane spin-polarized, while allowing a smaller in-plane component. Because the Bi₄Br₄ tip is more sensitive to in-plane sample spins, we also state explicitly that this smaller component could contribute appreciably to the QPI contrast and that the present experiment does not quantitatively separate the in-plane and out-of-plane contributions. These revisions preserve the experimentally established d-wave pattern while bringing the claim regarding its spin orientation into line with the quantitative bounds of Fig. R1.

Response to Referee #2

The authors have addressed the major points raised in my previous report. In particular, most of my concerns regarding the experimental observations, data interpretation, and the underlying physical picture have now been adequately mentioned. I note that some aspects of the authors' interpretation may still admit alternative views, and I do not necessarily agree with all of the arguments presented in the response and the revised manuscript. In my opinion, however, these remaining issues mainly concern the interpretation of the results rather than the validity of the experimental observations themselves.

Although there have appeared several similar studies after this work, however, in my view, these subsequent reports do not provide evidence that is as comprehensive and convincing as that presented in the present manuscript. Nevertheless, I consider such differences in interpretation to be scientifically constructive rather than detrimental to the manuscript. The results presented here may stimulate further discussion and experimental and theoretical efforts in this field.

Overall, I believe that the authors have satisfactorily addressed my major concerns, and that the remaining issues do not preclude publication.

Reply: We sincerely thank the referee for the positive assessment and for recognizing the breadth and comprehensiveness of the experimental evidence presented in this work. We appreciate the referee's thoughtful distinction between the robustness of the experimental observations and aspects of their microscopic interpretation that may remain open to alternative views. *Accordingly, we have further revised the manuscript to distinguish the directly established observations from model-dependent interpretations and have moderated our related wording.* We are grateful for the referee's constructive suggestions, which have substantially improved the clarity and rigor of the manuscript, and hope that this work will stimulate further experimental and theoretical investigations.

Response to Referee #3:

In order to come to an appropriate advice how to deal with the manuscript "Atomic-scale spin sensing of a 2D d-wave altermagnet via helical tunneling" by Wang and co-workers I read their detailed rebuttal letter to the three referee reports. I appreciate the enormous amount of data and extra analysis the authors invested to deal with the criticism raised by the referees. Overall, I come

to the conclusion that the amount of data and the insight provided by the data now justifies publication in Nature since it will—to my opinion— have an impact on the scientific debate and very likely trigger future studies in experiment and theory. Even if some of the interpretations offered in the manuscript might be revised based on future results, the breadth and quality of the data justifies their publication in a highly visible journal.

One side remark: To my opinion, large parts of what is written in the paragraph entitled „Functionalization of the tip with helical edge modes“ should be moved to the Methods section, the Extended Data or the Supplementary Material. But I leave the decision to the editorial team which will certainly safeguard Nature’s guidelines.

Reply: We sincerely thank the referee for the positive assessment of our work and for concluding that the breadth and quality of the data justify publication in Nature. We also appreciate the referee’s suggestion concerning the organization of the section entitled “Functionalization of the tip with helical edge modes.” Following this suggestion, *we have tried our best to streamline this section.* The main text now retains only the essential information needed to explain the current-direction-dependent spin-selective tunnelling principle and to summarize the key experimental benchmarks validating the performance of the Bi₄Br₄ tip. We thank the referee for this constructive suggestion, which has substantially improved the clarity and rigor of manuscript.