Evidence of higher-order topology in multilayer WTe₂ from Josephson coupling through anisotropic hinge states

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T³d-WTe₂ (non-centrosymmetric and orthorhombic), a type-II Weyl semimetal, is expected to have higher-order topological phases with topologically protected, helical one-dimensional hinge states when its Weyl points are annihilated. However, the detection of these hinge states is difficult due to the semimetallic behaviour of the bulk. In this study, we have spatially resolved the hinge states by analysing the magnetic field interference of the supercurrent in Nb–WTe₂–Nb proximity Josephson junctions. The Josephson current along the a axis of the WTe₂ crystal, but not along the b axis, showed a sharp enhancement at the edges of the junction, and the amount of enhanced Josephson current was comparable to the upper limits of a single one-dimensional helical channel. Our experimental observations suggest a higher-order topological phase in WTe₂ and its corresponding anisotropic topological hinge states, in agreement with theoretical calculations. Our work paves the way for the study of hinge states in topological transition-metal dichalcogenides and analogous phases.

Recently, it was proposed that bulk MoTe₂ and WTe₂ in their T³d structures are higher-order topological insulators (HOTIs), and that large surface states arising from gapped four-fold Dirac surfaces would be present. Their signatures were actually observed previously but not understood; it was argued that large surface states seen by ARPES and STM in bulk WTe₂ and MoTe₂ were not attributable to the Fermi arcs arising from the Weyl points. In addition, it was proposed that WTe₂, which is the simplest protected conducting helical hinge states, which would be difficult to resolve by transport measurements due to coexisting bulk conducting states. In this work, we coupled a WTe₂ thin film to two superconducting leads formed of niobium and measured the critical superconducting current of this Nb–WTe₂–Nb Josephson junction as a function of vertically applied magnetic field. By analysing the Fraunhofer pattern resulting from the interference of the superconducting currents, we found evidence of conducting channels localized at the edges of the sample, consistent with the presence of topological hinge states due to the higher-order topology of WTe₂. We further found that the hinge states are highly anisotropic; they are localized on edges parallel to the a axis of the crystal, whereas along the b axis they are delocalized and merge into the bulk state. Our claims of the presence of hinge states are supported by theoretical calculations that explain the observed current density profile and Fraunhofer patterns, and by control experiments to distinguish the hinge state from an edge state.

Firstly, we describe the angle-dependent bulk transport in multilayer WTe₂. The high-quality single crystals of WTe₂ used

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in this study were grown by the flux method. There was no intrinsic chemical doping of the WTe₂ crystals, as confirmed by non-saturating magnetoresistance in the bulk crystal and classical Hall measurements of exfoliated multilayer WTe₂ (Supplementary Fig. 1). Unlike many other transition-metal dichalcogenides, such as MoS₂, MoSe₂, and WSe₂, which have hexagonal planar structures, WTe₂ crystallizes in the non-centrosymmetric, orthorhombic, Td structure (space group Pnma, (No. 31)) in which the hexagonal plane is distorted by the formation of tungsten chains running along the a axis (Fig. 1a). This leads to a WSM bulk state and a 2D topological insulating state in the monolayer limit. To investigate the in-plane electrical anisotropy of the crystal, we fabricated a multi-terminal circular WTe₂ device covered with hexagonal boron nitride (Fig. 1b). The angular dependence of the in-plane resistance, \( R(\theta) \), within a single device was measured with a four-probe configuration to avoid contact resistance. Here, we chose the pair of voltage probes nearest to the pair of current electrodes, as shown in Fig. 1b, where \( \theta \) denotes the angle between the direction of the resistance measurement (white dashed line) and the a axis of the WTe₂ crystal (red dashed line). As expected from the crystalline anisotropy, the minimum resistance was observed along the a axis (\( \theta = 0^\circ \)) and the maximum resistance was observed along the b axis (\( \theta = 90^\circ \), Fig. 1c). The ratio of maximum to minimum resistance reached 2.7 at 50 K and dropped to 2.3 at 4.2 K (inset of Fig. 1c), presumably due to the competing temperature dependencies of carrier mobility and carrier density. Other manifestations of such anisotropic transport in WTe₂ have been observed in quantum oscillations and the chiral anomaly. As the anisotropic crystallographic axes are also reflected in the symmetry of the lattice vibrations, polarization-resolved Raman spectroscopy has been used to determine the crystal axis for all other devices in an accurate and non-invasive manner. The A1 Raman mode at around 165 cm⁻¹ shows a characteristic two-fold pattern in the parallel polarization configuration of laser excitation and detection (Fig. 1d). The a axis (b axis) of the crystal can be determined unambiguously on the basis of the angle with maximum (minimum) intensity, as reported previously (see Supplementary Figs. 2 and 3 for more Raman spectroscopic data).

Next, we discuss the proximity Josephson junction created with multilayer WTe₂, in which WTe₂ is placed between two Nb superconducting electrodes as depicted in Fig. 2a. When superconducting contacts are electrically transparent, the superconducting order parameters of the Nb electrodes can extend into WTe₂ and eventually lead to Josephson coupling, mediated by WTe₂. We fabricated eight different Josephson junctions of length \( L \) between 60 and 230 nm, width \( W \) between 1.8 and 5.2 μm and WTe₂ thickness \( t \) between 13 and 27 nm. These devices were divided into two groups with different crystal orientations, the a and b axes, aligned with the current direction (see Methods for details of the fabrication process). We will focus on two representative devices, device A with \((L, W, t) = (190 \text{ nm}, 2.3 \text{ μm}, 13.2 \text{ nm})\) with current flow along the a axis and device B with \((L, W, t) = (100 \text{ nm}, 3.3 \text{ μm}, 25.0 \text{ nm})\) with current flow along the b axis (see Supplementary Table 1 for a summary of the other devices). Figure 2c and its inset show the typical current–voltage (I–V) characteristics of devices A and B, respectively, at various temperatures. On sweeping the bias current of device A from zero to a positive value, the junction voltage switched from zero to a finite value at the Josephson critical current, \( I_c = 0.6 \text{ μA} \) at 145 mK. The retrapping current at which the voltage switched from a finite value back to zero was 0.5 μA, which is slightly smaller than \( I_c \), presumably due to self-heating of the junction. The \( I_c \) decreased monotonically with increasing temperature \( T \) and eventually vanished at around \( T \approx 1.0 \text{ K} \), above which the I–V curve became linear with a slope corresponding to the normal-state resistance \( R_n \). As \( I_c \) scales with cross-sectional area \( A = Wt \) and \( R_n \) scales with \( 1/A \), the \( I_c R_n \) product becomes independent of \( A \) such that it characterizes the strength of Josephson coupling, irrespective of the junction geometry. The measured \( I_c R_n \) product of device A (22.4 μV) was about seven times larger than that of device B (3.0 μV). Anisotropy of the \( I_c R_n \) product along the a and b axes was consistently observed for the other six devices (see Supplementary Table 1). The \( T \) dependence of \( I_c \) shown in Fig. 2d also reveals a more robust Josephson coupling for device A than for device B.

Unlike the insulating bulk states of topological insulators, the bulk states of WSMs such as WTe₂ are conducting, making it a challenge to resolve the proposed topological hinge states. Our approach was to spatially resolve how the Josephson current flows through the Nb–WTe₂–Nb Josephson junction by analysing its magnetic field response. When a magnetic field B is threading the junction area, the Aharonov–Bohm effect leads to a macroscopic quantum phase difference between the superconductors and modulates \( I_c \) according to the d.c. Josephson relationship. If a sinusoidal current–phase relation is assumed, the Josephson current interference pattern is determined by the magnitude of the phase-sensitive integration according to \( I_c(B) = \int_{-\infty}^{\infty} J(x)e^{i\phi_0}dx \), where \( J(x) \) is the Josephson current density, with \( x \) the real-space coordinate along the junction width direction and \( \phi_0 \) is the magnetic field-dependent parameter expressed by \( 2\pi B L \phi_0 / \Phi_0 \), where \( L \phi_0 \) is the effective junction length given by \( L + 2L' \), which takes into account the magnetic flux penetrating into each superconducting electrode by length \( L' \), and \( \Phi_0 \) is the magnetic flux quantum defined as \( h/2e \), where \( h \) is Planck's constant and \( e \) is electron charge. As \( I_c(B) \) can be viewed as the magnitude of the Fourier transform of \( J(x) \), we can extract \( J(x) \) from the inverse Fourier transform (IFT) of the experimentally measured \( I_c(B) \) and visualize the current flow density in the junction. This
In contrast to device A, device B (Fig. 3d) exhibited an interference pattern resembling the conventional single-slit Fraunhofer pattern, in which the oscillation amplitude decays with 1/B and vanishes above ~50 G (Fig. 3c). This is very similar to the results of our control experiment with graphite. The Josephson current density extracted from the IFT shows a uniform current distribution, without any signatures of edge-enhanced transport within the limits of our experimental resolution (Fig. 3f). The six other devices with different crystal axes showed consistent interference patterns and current density distributions (see Supplementary Fig. 6 for a axis and Supplementary Fig. 7 for b axis). The sharp difference in I(x) between devices A and B will be discussed in terms of the distinct shapes of the spatial wave functions of the topological hinge states along the different crystal axes.

We theoretically investigated the hinge states, and their Fraunhofer patterns, by adopting the model Hamiltonian reported previously\(^{25,27}\), which describes the higher-order topological properties of WTe\(_2\) (see Methods for details). Our model showed highly anisotropic band structures (see the insets of Fig. 3h,i), with the helical hinge states showing markedly different localizations along the a and b axes. The wave function of the hinge states along the a axis is localized at the edges of the junction (Fig. 3h), matching the current density profile of device A (Fig. 3c). When the chemical potential was set to introduce a bulk contribution, the calculated interference pattern presented in Fig. 3g, taking into account both the hinge and bulk states, shows a close resemblance to the interference shape recorded for device A (Fig. 3b), especially the non-vanishing behaviour of I\(_c\) even at high B. By contrast, the hinge states along the b axis strongly merge into the bulk states (inset of Fig. 3j) due to a larger spreading of the wave function (stronger hopping) in the orthogonal (a axis) direction (Supplementary Fig. 4). This is consistent with the crystalline and corresponding electronic anisotropy mentioned earlier. Consequently, the wave function is well delocalized over the entire junction in real space, as plotted in Fig. 3j, which matches the uniform current density profile of device B (Fig. 3i). This results in a single-slit Fraunhofer pattern, as shown in Fig. 3i, which is similar to the interference pattern measured for device B (Fig. 3c) and the graphite control experiment (Supplementary Fig. 6). The observed Josephson current mediated by a single edge of the junction, I\(_{\text{edge}}\), for device A is 36.9 ± 6.91 nA for the left edge and 35.5 ± 6.86 nA for the right edge (the shaded areas in Fig. 3c). Here, I\(_{\text{edge}}\) is less than the maximum theoretical value of I\(_{\text{th}}\) = 140 nA for a single hinge state, and three other devices consistently showed I\(_{\text{edge}}\) ≤ I\(_{\text{th}}\) (Supplementary Table 2). The value of I\(_{\text{th}}\) was obtained by considering a short ballisitic junction limit, expressed as c\(_{\text{fl}}\)R\(_{\text{NS}}\) = h\(\Delta c\) where R\(_{\text{NS}}\) = \(\hbar c\) is the normal-state resistance for a single hinge state and \(\Delta c\) = 1.763k\(_B\)T\(_{\text{NS}}\) is the Bardeen–Cooper–Schrieffer (BCS) superconducting gap of the Nb electrode for T\(_{\text{NS}}\) = 7.5 K and k\(_B\) is Boltzmann’s constant. The fact that I\(_{\text{edge}}\) is smaller than I\(_{\text{th}}\) is consistent with the theory that there is only one hinge state on each side of the WTe\(_2\) flake, as depicted in Fig. 3a.d. Other HOTI candidates have exceeded their theoretical maximum I\(_{\text{edge}}\) and the reason for this remains an open question\(^{34,36}\).

We note that there is an alternative, but less likely, explanation for the observation that stems from the quantum spin Hall insulator phase of monolayer WTe\(_2\). When monolayers of WTe\(_2\) with 1D quantum spin Hall edge states are vertically stacked with finite interlayer interactions, 1D topological edge states gradually turn into the 2D Fermi arc surface state of WSMs. In this low-layer limit between the bulk and monolayer, edge states may exist due to quantization of the c-axis momentum in the Fermi arc states. However, this is an unlikely explanation for our observations for three reasons. Firstly, we were closer to the bulk limit in our experiments (18–38 layers) than the monolayer limit. Secondly, many experiments have been carried out on few-layer (2–10 layers) WTe\(_2\), and edge states were observed strictly in the monolayer\(^{15–19}\). Finally, if the Fermi arc states...
were present, they would yield a large number of conducting channels on the side surfaces that would give $I_{J_{ao}}$ values much larger than $I_{J_{ab}}$ (Supplementary Fig. 8); however, this was not the case for all four $a$-axis oriented devices.

Further, to differentiate between the (1D) hinge-state scenario and the (2D) Fermi arc surface-state scenario, we performed a control experiment. We fabricated a Josephson junction device (device AT (Dev. AT)) with $(L, W, t) = (180 \, \text{nm}, 1.35 \, \mu\text{m}, 88 \, \text{nm})$ from the same bulk crystal as used for all the previously discussed $a$- and $b$-axis-oriented devices in which the bottoms of the side surfaces of the WTe$_2$ flake were covered by insulating Al$_2$O$_3$, as depicted in Fig. 4a–c (see Supplementary Fig. 9 for details of the fabrication procedure). Depending on the scenario, one would expect a very different shape of Josephson interference pattern. In the hinge state scenario (Fig. 4a), only the hinge state on top of the flake (filled circle) can contribute to Josephson coupling as the other hinge state at the bottom (empty circle) cannot couple with the superconductor because of the insulating Al$_2$O$_3$. The single hinge state in contact with the superconductor (filled circle) cannot interfere with the other hinge state (empty circle), and results only in a rather standard shape of the Fraunhofer pattern. By sharp contrast, the Fermi arc surface state would be distributed uniformly on both side surfaces of the WTe$_2$ flake that are in direct contact with the superconductors (Fig. 4b and Supplementary Fig. 8). This would lead to a non-vanishing Josephson pattern at high field. As can be seen in Fig. 4d, device AT (orange line) shows a rather standard Josephson pattern with $1/B$ decay, as expected for a single-hinge state and not Fermi arc states on both edges, which is in sharp contrast to the non-vanishing interference pattern of device A (blue line). More quantitatively, the $R^2$ value (0.919) of the fitting to the standard Fraunhofer pattern for device AT is markedly higher than that (0.749) for device A (Supplementary Fig. 10). This same behaviour was consistently observed in two other control devices with the same geometry, strongly showcasing the hinge state scenario.

By investigating the electronic transport in multilayer WTe$_2$ in both normal and superconducting regimes in combination with model Hamiltonian calculations, we can provide evidence for the higher-order topology of non-centrosymmetric Td-WTe$_2$ through proximity-induced superconductivity in its anisotropic hinge states. Several theoretical studies on the effect of superconducting proximity on WSMs have shown that the Andreev reflection in bulk states is suppressed due to the conservation of chirality, whereas the topological boundary state interacts well with superconductivity. Such selective coupling of bulk and boundary states to superconductors makes superconducting heterostructures an ideal platform for investigating topological semimetals that have mixed bulk and topological boundary contributions, particularly HOTI systems. Our study would also provide a foundation for studying superconducting HOTI candidates such as MoTe$_2$ (ref. 40), correspondingly TaIrTe$_4$, and metal-contacted WTe$_2$ at extremely low temperatures or under pressure. Additionally, the anisotropy of the hinge state combined with proximity-induced superconductivity in these materials results in naturally 1D, topologically protected, superconducting wires that may be exploited in superconducting...
Fig. 4 | Fraunhofer pattern of a partially insulated Josephson junction. a, b. Schematic side views of Josephson junctions with Al2O3 on the side surfaces (top panels), the corresponding Josephson current density profiles (middle panels) and the expected interference patterns (bottom panels) for the hinge state case (a) and the Fermi arc surface state case (b). The red filled and empty circles (polygons) in the top schematic of a (b) represent superconductor-coupled and -decoupled hinge states (Fermi arcs surface states), respectively. The black solid lines in the bottom panels represent the standard Fraunhofer pattern with a uniform current density profile. c. Scanning electron micrograph of device AT showing the measurement configuration (left) and the schematic cross-sectional side view (right). d. Interference pattern of the critical current of device AT (orange) fitted with the standard Fraunhofer pattern (black line). For comparison, the interference pattern of device A (blue) is also fitted with the standard Fraunhofer pattern.

electronics. Unlike bismuth, in these transition-metal chalcogenide compounds (which should all host these states), the hinge states can be probed through chemical doping to modify the Fermi energy and the subsequent inter-relation between hinge and bulk states. Also, their exfoliability allows for layer-dependent investigations of the crossover among Weyl semimetals, higher-order topological insulators and quantum spin Hall insulators as well as the interplay of the hinge state with magnetism in artificial heterostructures consisting of WTe2 and a 2D ferromagnet such as Cr3Ge2Te6, Fe3Ge2Te6 or CrI3. Finally, twisted layer stacking will allow study of the hinge state with moiré physics.

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Methods

Device fabrication. A WTe₂ single crystal was mechanically exfoliated to provide WTe₂ thin films with a thickness of 10–20 nm that were placed on silicon substrate covered with a 280-nm-thick insulating layer of SiO₂. The thickness of the WTe₂ flakes was confirmed by atomic force microscopy (using an X-E™ from Park Systems) after electrical measurement. As the bulk single crystal has a needle shape elongated along the a axis, the crystal axis could be tracked during the exfoliation process. Polarized Raman spectroscopy (a home-built set-up based on the confocal microscope) was used to confirm the direction of the crystal axis after electrical measurement. To eliminate possible contamination of the topmost layers of the WTe₂ flakes, in situ argon ion etching was conducted before metal deposition to remove WTe₂ surface layers possibly oxidized in ambient air. A Ti/Nb layer was sputtered on WTe₂ surface at the low rate of 8.33 nm min⁻¹ to minimize the degradation of WTe₂. The Nb layer prevents the adhesion of metal electrodes to WTe₂, and the gold layer protects the Nb superconducting electrode from oxidation. The chamber pressure during titanium evaporation was kept below 8 × 10⁻⁴ torr. The Nb layer was sputtered under argon atmosphere at the low rate of 8.33 nm min⁻¹ to minimize the damage to the WTe₂ flake by argon plasma. During the fabrication process, the top surface of the WTe₂ thin film was exposed to poly(methyl methacrylate) polymer only once to minimize the degradation of WTe₂.

Polarization-resolved Raman spectroscopy. Raman spectra were recorded with a confocal microscope at room temperature. The samples were excited with a HeNe laser (632.8 nm) at normal incidence. Raman signals were collected in the backscattering configuration and analysed using a monochromator equipped with a liquid nitrogen-cooled silicon CCD (charge-coupled device). Two linear polarizers in parallel configuration were placed immediately in front of the laser and before the monochromator to define the polarizations of incident and scattered light, respectively. The crystal orientation relative to the polarization was controlled with a half-wave plate between the beam splitter and WTe₂ flake.

Effective model. The Td-WTe₂ has been identified as a type-II WSM (ref. 1). However, according to both ab initio calculations35 and ARPES measurements47, its Fermi arc states connecting Weyl points is very short. It has been argued that under realistic experimental conditions such as the presence of low strain20,43, the Fermi arc state connecting Weyl points is very short. It has been argued that effective model.

\[ H(k) = H_{\text{Bnd}}(k) + V_c + V_m(k) \]

where \( H_{\text{Bnd}}(k) = (m_1 + \sum_{\alpha=x,y} v_\alpha \cos(k_\alpha) + m_2 \mu^\alpha + m_3 \gamma^\alpha) r^\alpha + \lambda_\beta \sin k_\beta \mu^\alpha \sin k_\gamma \gamma^\alpha, k \) is the crystal momentum, \( V_c = \gamma r^\alpha + \gamma \mu^\alpha \) and \( V_m(k) = \beta_1 \sin k_\beta \mu^\alpha \sigma^\beta \). The Pauli matrices \( \mu \) and \( \sigma \) operate on different orbital space, and \( \sigma \) operates on spin space. The Hamiltonian \( H_{\text{Bnd}}(k) \) gives a bulk monopole node-line (MNL) semimetal20,44,45,47, the narrow separation of Weyl points can be annihilated and the material becomes a \( \sigma \)-WTe₂, which is centrosymmetric with non-symmorphic space group \( P_2_1/m \) and point group \( C_{3h} \). Therefore, to study the topological properties of Td-WTe₂, it is sufficient to use an effective Hamiltonian with the point group \( C_{3h} \) and the same number of double band inversions at time-reversal invariant momenta as Td-WTe₂. The effective Hamiltonian can be written according to equation (1):

\[ H(k) = H_{\text{Bnd}}(k) + V_c + V_m(k) \]

Numerical calculation of the Josephson current. In the calculations, we assumed that the multilayer WTe₂ underwent the Nb electrodes was fully superconducting due to the proximity effect and that their pairing potentials were bound by \( 4\pi \lambda_{\text{ph}}/2 \), respectively. We modelled multilayer WTe₂ with the lattice model discretizing from the Hamiltonian \( H(k) \) in equation (1), and the Josephson current was evaluated using the recursive Green’s function method36 by defining the system with three layers of WTe₂: 25 lattice sites along the current direction, 15 lattice sites perpendicular to the current direction, \( k_0 T = \Delta/200 \) and \( \Delta = 0.5 \).

Data availability

The data that support the findings of this study are available from the corresponding authors on reasonable request.

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K.C.F., M.N.A., K.T.L. and G.-H.L. conceived and supervised the project. Y.-B.C. fabricated the samples. Y.-B.C. and J.P. performed transport experiments. J.Y. and X.-Y. performed Raman experiments. Y.-B.C., Y.X., K.C.F., M.N.A., K.T.L. and G.-H.L. wrote the paper. K.W., J.K., K.C.F., M.N.A., K.T.L. and G.-H.L. conceived and supervised the project. Y.-B.C. fabricated the samples. Y.-B.C. and J.P. performed transport experiments. J.Y. and X.-Y. performed Raman experiments. Y.-B.C., Y.X., K.C.F., M.N.A., K.T.L. and G.-H.L. wrote the paper. K.W., J.K., K.C.F., M.N.A., K.T.L. and G.-H.L. conceived and supervised the project. Y.-B.C. fabricated the samples. Y.-B.C. and J.P. performed transport experiments. J.Y. and X.-Y. performed Raman experiments. Y.-B.C., Y.X., K.C.F., M.N.A., K.T.L. and G.-H.L. wrote the paper.

Competing interests

The authors declare no competing interests.

Additional information

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