RESEARCH ARTICLE

PHYSICS

Edge superconductivity in multilayer WTe₂ Josephson junction

Ce Huang^{1,2,†}, Awadhesh Narayan^{3,†}, Enze Zhang^{1,2}, Xiaoyi Xie^{1,2}, Linfeng Ai^{1,2}, Shanshan Liu^{1,2}, Changjiang Yi⁴, Youguo Shi^{4,5}, Stefano Sanvito⁶ and Faxian Xiu D^{1,2,7,*}

ABSTRACT

WTe₂, as a type-II Weyl semimetal, has 2D Fermi arcs on the (001) surface in the bulk and 1D helical edge states in its monolayer. These features have recently attracted wide attention in condensed matter physics. However, in the intermediate regime between the bulk and monolayer, the edge states have not been resolved owing to its closed band gap which makes the bulk states dominant. Here, we report the signatures of the edge superconductivity by superconducting quantum interference measurements in multilayer WTe₂ Josephson junctions and we directly map the localized supercurrent. In thick WTe₂ (~ 60 nm), the supercurrent is uniformly distributed by bulk states with symmetric Josephson effect ($|I_c^+(B)| = |I_c^-(B)|$). In thin WTe₂ (10 nm), however, the supercurrent becomes confined to the edge and its width reaches up to 1.4 μ m and exhibits non-symmetric behavior $|I_c^+(B)| \neq |I_c^-(B)|$. The ability to tune the edge domination by changing thickness and the edge superconductivity establishes WTe₂ as a promising topological system with exotic quantum phases and a rich physics.

Keywords: WTe2, Josephson junction, Weyl semimetal, edge superconductivity, non-symmetric effect

INTRODUCTION

Layered WTe₂ was suggested as the first material candidate to be a type-II Weyl semimetal, where eight separated Weyl points exist in the bulk and topological Fermi arcs occur on the (001) crystal surfaces owing to the reflection symmetry [1]. An extra set of quantum oscillations arising from Weyl orbits were observed as evidence of Fermi arcs in transport [2]. Intriguingly, when the thickness is reduced to the monolayer, WTe2 turns to be a quantum spin Hall insulator with edge states [3], which have been demonstrated in numerous experiments involving low-temperature transport [4,5], angle-resolved photoelectron spectroscopy [6], scanning tunneling microscopy [7,8], and microwave impedance microscopy [9]. Besides, it has also been predicted that WTe2 has 1D hinge states as a higher-order topological insulator [10].

While the boundary modes of WTe_2 have been well studied in both the 3D and 2D limits [11,12], in multilayers these modes become rather compli-

cated due to the intervening bulk and edge states and thus they remain largely unexplored. Unlike the monolayer WTe2, the nearly-closed bandgap in multilayer WTe₂ results in a large density of bulk states. Therefore, it is difficult to distinguish the edge states through a gating approach. It is then necessary to make them distinct from the coexisting bulk ones. However, separating edge and bulk states in a single electrical conductance measurement may be ambiguous. In contrast, if the charge carriers condense together to form Cooper pairs, the difference can be amplified since the supercurrent properties are largely related to the coherence length [13]. A planar microscopic Josephson junction to realize superconducting TSM is feasible to elucidate the boundary states. For example, in Nb/Bi₂Te₃/Nb Josephson junctions the surface states enable the ballistic Josephson current rather than the diffusive bulk transport [14]. The supercurrent distribution in real space can be also quantitatively extracted from the superconducting quantum interference (SQI)

¹State Key Laboratory of Surface Physics and Department of Physics, Fudan University, Shanghai 200433, China; ²Collaborative Innovation Center of Advanced Microstructures, Nanjing 210093, China; ³SSCU, Indian Institute of Science, Bengaluru 560012, India; ⁴Institute of Physics and Beijing National Laboratory for Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100190, China; ⁵School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100190, China; ⁶School of Physics, AMBER and CRANN Institute, Trinity College, Dublin 2, Ireland and ⁷Institute for Nanoelectronic Devices and Quantum Computing, Fudan University, Shanghai 200433, China

*Corresponding author. E-mail: Faxian@fudan.edu.cn

[†]Equally contributed to this work.

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measurements, where a perpendicular magnetic field induces oscillations in the amplitude of the superconducting current in Josephson junctions. This approach has been widely adopted to reveal the quantum spin Hall edge states in HgTe quantum well [15], topological surface states in TI [16] and quantum Hall edge states in graphene [17], but not yet in TSM.

Here, we report the observation of edge superconductivity in multilayer WTe₂ Josephson junctions. By varying the thickness of WTe₂ in SQI experiments, we have observed the Fraunhofer and the mixture of Fraunhofer and SQUID pattern in thick and thin WTe₂, respectively, which indicates the edge superconductivity in thin WTe₂. The nonuniform supercurrent exists in multilayer WTe₂ up to 16 nm, while the bulk supercurrent density amplitude (J_c) is about 1/3 of the edge in the thinnest sample. $|I_c^+(B)| \neq |I_c^-(B)|$ is also observed in thin WTe₂ due to the inversion symmetry breaking.

WT_{E2} JOSEPHSON JUNCTIONS

We measure several Josephson junctions consisting of WTe2 flakes of different thicknesses contacted by niobium (Nb) leads. The fabrication and characterization details are described in the Methods and SI Section I (Supplementary Figs 1 and 2). A scanning electron microscopy (SEM) image of the actual device and its measurement configuration are displayed in Fig. 1a (device #1, 10 nm-thick WTe₂). The length and width of the superconducting channel are L = 200 nm and $W = 13 \,\mu$ m, respectively. Figure 1b shows the resistance-temperature (*R*-*T*) curve of the junction with two transitions T_{c1} and T_{c2} at zero magnetic field. $T_{c1} \sim 8$ K originates from the Nb superconducting transition, while the resistance continues to drop to 10^{-3} times of the normal resistance below $T_{c2} \sim 0.72$ K which comes from the proximity Josephson coupling. The Josephson effect is highly reproducible across different devices, as shown in Supplementary Fig. 3. Figure 1c and its inset display the I-V characteristics and the differential resistance (dV/dI) of the junction at 45 mK, respectively. From the slope of the *I-V* curve in the high bias region $(I > 10 \,\mu\text{A})$ where the curve is linear, the normal-state resistance $R_N \sim$ 1.7 Ω is extracted. For $|I| < 4.1 \ \mu$ A, the voltage and dV/dI across the junction remain nearly zero, indicating a robust Josephson effect. Ten WTe₂ Josephson junctions with various L and W are studied (see Table S1 for their junction parameters), all exhibiting a finite supercurrent at low temperatures with reproducible behavior. The junction is in the long junction limit [18] (see SI Section IV and

Supplementary Fig. 4 for details). Therefore it follows the 1/L dependence found from the $I_c R_N$ plot versus L in Fig. 1d. In this long junction regime, the critical current is given [18,19] by $I_c \sim \frac{E_{Th}}{eR_N}$, being determined by the Thouless energy E_{Th} , which can be estimated [20] to be $\sim hv_F/L$, yielding $I_c R_N \propto 1/L$.

THE SUPERCONDUCTING QUANTUM INTERFERENCE MEASUREMENTS

Having established the Josephson effect in our Nb/WTe₂/Nb junctions, we then focus on the supercurrent of WTe₂. In our experiments, the spatial distribution of the supercurrent is analyzed by applying a perpendicular magnetic field *B* during the SQI measurements with different thicknesses of WTe₂. The particular shape of the critical current interference pattern depends on the phase-sensitive summation of the supercurrents traversing the junction. In the case of a symmetric supercurrent distribution, this integral takes the simple form [21]:

$$I_{c}^{\max}(B) = \left| \int_{-\infty}^{\infty} J_{c}(x) \cos\left(\frac{2\pi L_{eff} Bx}{\Phi_{0}}\right) dx \right|,$$

where L_{eff} is the effective length of the junction along the direction of the current, accounting for the magnetic flux threading [22] through parts of the superconducting contacts over the London penetration depths. $\Phi_0 = h/2e$ corresponds to the magnetic flux quantum.

In thick WTe₂, the bulk states dominate and along the *y*-axis the supercurrent density has an approximately uniform distribution as shown in Fig. 2a. Thus, the uniform current density yields the singleslit Fraunhofer pattern described by [21]

$$= I_{c0} \left| \sin \left(\frac{\pi L_{eff} B W}{\Phi_0} \right) / \left(\frac{\pi L_{eff} B W}{\Phi_0} \right) \right|.$$

We have measured device #8 with 60 nm-thick WTe₂ as shown in Fig. 2b. The critical current envelope has an oscillation characteristic. We obtain a period of ~0.33 mT, which yields the effective length of $L_{eff} = \Phi_0/\delta B_{lobe} W \sim 1.0 \,\mu$ m. This effective length, larger than the distance between the two Nb electrodes (L = 240 nm), is caused by the London penetration depth and the flux focusing due to the Meissner effect [23,24]. The critical current envelope strongly resembles a single-slit pattern with $2\Phi_0$ central lobe width. The corresponding supercurrent distribution is obtained by transforming the single-slit pattern to the real-space current density,

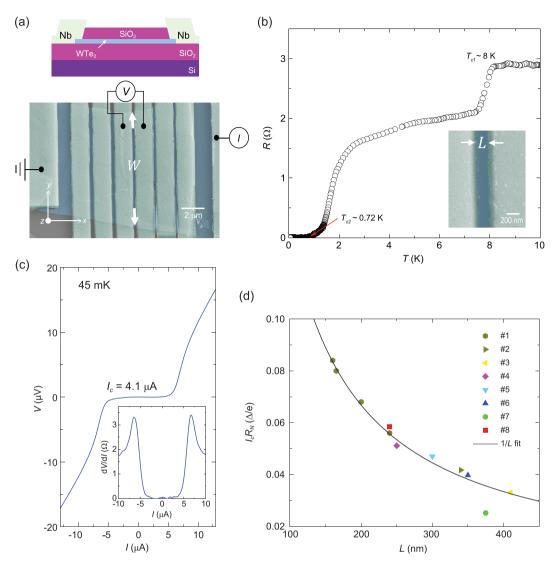


Figure 1. Josephson effect in thin WTe₂. (a) Top: Junctions schematic. Bottom: False-colour SEM image of the device with the measurement configuration. 120 nm-thick Nb is deposited on the top of WTe₂ (device #1). A four-terminal measurement across the interface was performed. The in-plane crystal axis of the WTe₂ flake is unknown. (b) Temperature dependence of WTe₂ Josephson junction resistance. Two transitions are identified: $T_{c1} = 8$ K is from the superconducting Nb, $T_{c2} \sim 0.72$ K is from the proximity Josephson coupling of WTe₂. Inset shows that the junction has a length of L = 200 nm. (c) *I-V* characteristics for Josephson junction in the superconducting states with a critical current of $I_c \sim 4.1 \ \mu$ A under zero magnetic field at 45 mK. Inset: dV/dI characteristics indicate zero resistance below the critical current, the same as the *I-V* curve. (d) Effect of the junction length on supercurrent for eight devices. The product kR_N follows a general trend of $kR_N \propto 1/L$.

 $J_c(z)$, as shown in Fig. 2c. This suggests a nearly uniform supercurrent density throughout the *y* direction. The full details of the extraction procedure can be found in the SI section V and Supplementary Fig. 5. Furthermore, the critical currents overlap each other at different current directions as shown in Fig. 2d which indicates a symmetric Josephson effect with $|I_c^+(B)| = |I_c^-(B)|$, where + and - denote the sweep direction of the bias current and +*B* and -*B* are the magnetic field directions.

When the WTe_2 is thinned down to a few layers, the low bulk density of states coexists with the possible high density at edges as shown in Fig. 2e. The magnetic-field-dependent critical current envelope in a 13 nm-thick WTe₂ device (device #2) demonstrates the periodic SQI with a $1.6\Phi_0$ central lobe width (Fig. 2f). I_c decays slowly which is distinct from the Fraunhofer pattern in Fig. 2b. We use an edge-stepped nonuniform supercurrent model to directly simulate the $I_c - B$ relation as shown by the black line in Fig. 2f (see the model details in SI Section VI and Supplementary Fig. 6). The good fit of both the magnitude and periodicity of I_c indicates the nonuniform supercurrent in

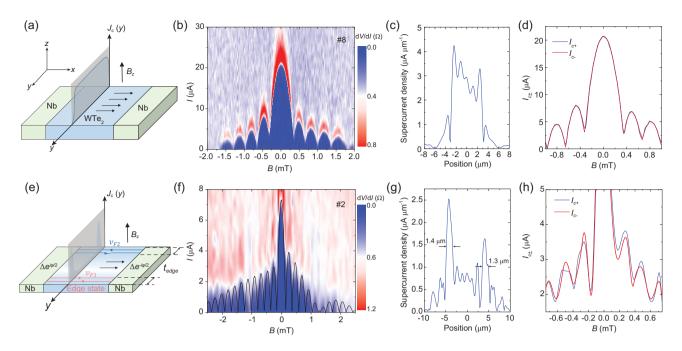


Figure 2. Evolution of edge superconductivity in thin WTe₂. (a) A schematic picture of a lateral Josephson junction with the out-of-plane magnetic field in thick WTe₂. The thick WTe₂ is filled with charge carriers and the supercurrent can flow uniformly across the junction along the *y*-axis, corresponding to a flat supercurrent density $J_c(y)$. (b) The differential resistance at different values of B_z in 60 nm-thick WTe₂ (device #8), showing the single-slit interference characteristics with a uniform supercurrent density. (c) The supercurrent distribution along the *y*-axis, which is calculated by the inverse Fourier transform of the data in (b). The supercurrent density is uniform along the *y*-axis, consistent with trivial bulk charge transport. (d) Critical current l_c as a function of *B* for the two sweep directions (positive as the blue line, negative as the red line). Two curves overlap with each other. (e) A schematic picture of a lateral Josephson junction with the out-of-plane magnetic field where $\Delta e^{\pm i\varphi/2}$ denotes the pairing order parameter of two superconducting Nb electrodes. In thin WTe₂, the bulk domination decreases and the supercurrent is carried by the edge. The edges on two sides of WTe₂ have different Fermi velocities v_{F1} and v_{F2} when the inversion symmetry is broken that gives rise to the asymmetric Josephson effect. (f) Differential resistance across the 13 nm-thick WTe₂ junction (device #2), showing a mixture of Fraunhofer and SQUID-like pattern with a central lobe of width $< 2\Phi_0$ and side lobes of width Φ_0 . The black line shows the fitting results from the edge-stepped supercurrent model. (g) The supercurrent distribution of device #2. The widths of the supercurrent-carrying edge channels are estimated to be $1.3-1.4 \ \mu$ m. (h) Critical current l_c as a function of *B* for the two sweep directions (positive as the blue line, negative as the red line), indicating non-symmetric behavior $l_r^+(B) \neq l_c^-(B)$.

few-layer WTe₂. Furthermore, the mixture of Fraunhofer and SQUID interference pattern corresponds to the development of sharp peaks in the supercurrent density at the mesa edges in Fig. 2g. The widths of the supercurrent-carrying edge are estimated to be in the range 1.3–1.4 μ m. The value is similar to other edge superconductivity systems [15,25] and the additional edge modes or bulk modes coupled weakly to edge states across the junction to carry supercurrent can result in the large edge supercurrent channel. The relation of the critical current with the magnetic field is presented in Fig. 2h and behaves non-symmetric $|I_c^+(B)| \neq |I_c^-(B)|$ in most magnetic fields which is different with thick sample.

We have reproduced the edge superconductivity and the mixture of Fraunhofer and SQUID pattern in a 10 nm-thick WTe_2 device (#1), and the traditional Fraunhofer pattern in a 40 nm-thick WTe_2 device (#5, see SI Section VII and Supplementary Fig. 7 for details). The higher supercurrent density at edges suggests a robust coupling to the superconductor electrodes.

To further distinguish the superconducting proximity Josephson coupling of edge/bulk, we experiment with a 16 nm-thick WTe₂ device (#3) to distinguish the bulk and edge contributions. Two Josephson channels are fabricated as the edgecrossing (R_1) and edge-untouched (R_2) as shown in Fig. 3a. For R_{2} , the junction is easier to be conducted by the bulk because the electrodes are closer in the central region ($L_b \sim 0.4 \,\mu$ m) while far at the edge. The distance on the edge side is $L_s \sim 4 \,\mu m$ that makes it hard to realize the Josephson effect through the edge region. Since the thickness is uniform in this sample, as indicated by the atomic force microscopy (AFM) measurement (Supplementary Fig. 2c and d), we can reasonably assume that the resistance by bulk states is isotropic and inversely proportional to the width. Figure 3b shows the R-T curve at low temperatures. Only edge-crossing R_1 can reach zero to exhibit Josephson effect while R_2 only decreases a little. The differential resistance versus the current measurement in Fig. 3c verifies this property. Since the lengths of two junctions differ slightly, the

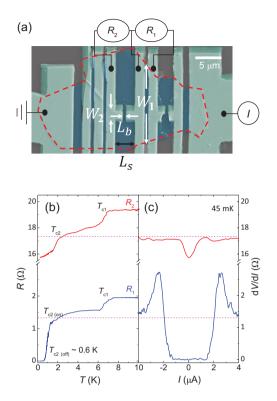


Figure 3. The coupling of superconductor Nb to the bulk and edge WTe₂ channels. (a) False-color SEM image of the device #3 with two measurement configurations. R_1 and R_2 represent the edge-crossing and edge-untouched junctions, respectively. The electrode separation width of R_2 from the edge W_0 is larger than 5 μ m. The length for edge channel L_s is 4 μ m while for the bulk channel $L_b \sim$ 0.4 μ m which makes the edge superconductivity hard to realize. (b) Temperature dependence of resistance in two junctions as shown in (a). T_{c1} is the superconducting transition of Nb while the superconducting WTe₂ emerges at T_{c2} . (c) d V/d/ characteristic at 45 mK.

coherence length of R_1 should be larger than that of R_2 to realize the Josephson effect. A similar mixture of Fraunhofer and SQUID pattern with edgedominated supercurrent is also observed, as shown in Supplementary Fig. 7b, which is consistent with the other two thin devices (#1 and #2). On the contrary, R₂ does not exhibit any oscillation and only the central lobe is observed (see Supplementary Fig. 8 for details). The width W_2 for R_2 is estimated to be 1.9 μ m, which corresponds well to the actual junction width 1.7 μ m as shown in Fig. 3a. The in-complete superconductivity of R_2 is due to the weaker superconducting combining for bulk. If the Josephson channel is further shortened, the bulkonly channel R_2 can also be superconducting in another device #9. However, only the Fraunhofer pattern with uniform supercurrent density is observed and corresponds well to the bulk-dominated

sample #8 in Fig. 2c and d (see Figs S8 and 9 for details).

DISCUSSION

It is necessary to discuss whether the observed edge superconductivity originates from the edge states in WTe2 or other trivial effects. All of the four different devices exhibit the sharp edge superconductivity which can exclude the accidental impurity effect. The exclusion of some trivial effects such as fluctuations and the affection by the SiO₂ substrate and the capping layer in thinner WTe₂ is also discussed in Supplementary Section VIII. However, it is difficult to exclude other trivial effects such as trivial edge states. Moreover, the other trivial mechanisms can also lead to a similar non-uniform supercurrent such as an inhomogeneous interface. A mixture of Fraunhofer and SQUID pattern was also observed in Nb-InGaAs/InP junctions with a step-shaped current density distribution [26]. Therefore, we need to point out that the edge superconductivity we observed is not equivalent to the superconductivity in the edge modes nor any evidence of toplogical superconducting phase. On the contrary, only the superconductivity in the edge region of samples can be concluded in our experiments.

The critical currents following $|I_c^+(B)| \neq |I_c^-(B)|$ in thin WTe₂ are quite interesting. In general, the asymmetric crystal can induce different Fermi velocities at two sides and result in supercurrent asymmetry. Since the supercurrent density is uniform as shown in Fig. 2c with symmetric Fraunhofer pattern (Fig. 2d), the bulk WTe₂ does not contribute to the asymmetry. Consequently, this supercurrent asymmetry may be related to the edge which is consistent with the predicted effect of inversion-symmetry-breaking on Weyl semimetal [27]. The total Josephson current carried by the two edges can be described by [28]

$$I(\Phi,\varphi) \propto I_1 \sin(n\varphi + n\Phi) + I_2 \sin(n\varphi - n\Phi)$$

where I_1 and I_2 represent the Josephson current carried by the two edges, Φ and φ are the phase in WTe₂ (the magnetic-field-related) and Nb regions (the current-related), respectively. The two edges have different energy spectra and $I_1 \neq I_2$ in thin WTe₂ (Fig. 2g) which results from different Fermi velocity of the two edge sides, denoted by the red and blue lines as shown in Fig. 2e. Therefore, the $I(\Phi, \varphi)$ is not symmetric for both φ and Φ anymore. Other possibilities such as vortex trapping, vortex motion during the magnetic field sweep or

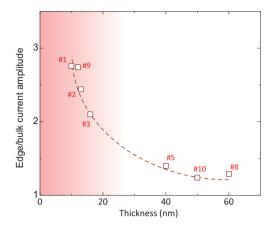


Figure 4. Summary of thickness-dependent edgesupercurrent-density contribution in WTe₂. The edge/bulk current amplitude is estimated by the ratio of edge/bulk $(\frac{Jedge}{Jbulk})$ supercurrent density. j_{edge} and j_{bulk} are estimated by the average value of the left and right peaks and the value of the central region in position-dependent supercurrent density. The dashed line shows the trend.

bulk states asymmetry may contribute. However, the $|I_c^+(B)| = |I_c^-(B)|$ in the thick sample in Fig. 2d helps to largely exclude the other possibility.

We note that two recent preprints [29,30] have also studied WTe₂ Josephson junction and shown evidence of edge states which is explained to be Hinge states [10]. Indeed, it is challenging to unambiguously determine the definite origin of edge superconductivity in our results, and various possibilities exist. However, from the consistent observations of high edge supercurrent density [29], the edge superconductivity is confirmed in the multi-layer WTe₂ system. Compared to the reported data, we further perform thickness-dependence experiments and provide more evidence that edge superconductivity exists in thin WTe₂ but not a thick one.

We summarize the supercurrent density amplitude ratio of the edge and bulk in Fig. 4. The edge superconductivity gradually emerges in thinner ones and the edge/bulk supercurrent amplitude reaches 2.76 in 10 nm-thick WTe₂. The critical thickness for the transition from edge to bulk-dominated superconductivity is estimated to be $t_c = 16-20$ nm. Moreover, various topological semimetals such as the TaAs family [31] (Fermi-arc surface states), layered MoTe₂ [32] (edge states in the 2D limit) and ZrSiS [33] (nodal-line surface states) can be further fabricated into Josephson junctions to detect the surface/edge states.

CONCLUSION

In summary, by studying the Fraunhofer interference, our measurements provide the supercurrent

distribution in type-II Weyl semimetal WTe₂. In thin WTe₂, the existence of edge superconductivity is evidenced. Besides, non-symmetric behavior $I_{c}^{+}(B) \neq I_{c}^{-}(B)$ in WTe₂ through the edge is an intrinsic property of the inversion symmetry breaking, which is distinct from other systems by an external in-plane magnetic field [22]. Furthermore, the Josephson junctions formed from 1D edge states or 2D surface states and s-wave superconducting contacts are expected to emulate spinless p-wave superconductivity [34] and Majorana flat bands [35] via a.c. Josephson effect by Shapiro response measurements. Edge superconductivity establishes WTe₂ as a promising platform for the future realization of topological superconductivity and Majorana bound states.

METHODS WTe₂ crystal growth

High-quality bulk WTe₂ crystals were grown by chemical vapor transport (CVT) method as reported before [36]. Single crystals of WTe₂ were grown by a high-temperature self-flux method. High-purity tungsten powders (99.9%) and Te pieces (99.999%) were inserted into alumina crucibles with a molar ratio of 1:30 in a glove box filled with pure argon then sealed in quartz tubes under high vacuum. The tubes were heated to 1100°C in 20 hours and maintained for 10 hours. Then the furnace was slowly cooled down to 650°C with a rate of 2°C/h followed by separating the Te flux in a centrifuge at 650°C.

Device fabrication

The WTe₂ flakes were mechanically exfoliated onto a Si substrate capped with a 280 nm-thick SiO₂ layer and the thickness of WTe₂ was identified by optical contrast and atomic force microscopy. The WTe₂ Josephson junctions were fabricated by an *e*-beam lithography technique and wet-etched by standard buffered HF solution for 5 s in the electrode regime. We deposited 120 nm-thick Nb electrodes using magnetic sputtering. Then, 40 nm-thick SiO₂ was deposited on top to prevent the WTe₂ oxidization.

Transport measurements

Four-terminal temperature-dependent transport measurements were carried out in a Physical Property Measurement System (PPMS, Quantum Design) with a dilution refrigerator, which achieves a base temperature of 35 mK. The transport properties were acquired using lock-in amplifiers (SR830) and Agilent 2912 meters. We used an excitation current of <50 nA. In differential resistance (dV/dI) measurements, a small *a.c.* current bias (10 nA to 100 nA) is generated by the lock-in amplifier output voltage in combination with a 10 M Ω bias resistor. This small *a.c.* current is added on top of the larger d.*c.* current bias by Agilent 2912, and the induced differential voltage is measured using the lock-in technique with a low frequency (<50 Hz).

DATA AVAILABILITY

The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

SUPPLEMENTARY DATA

Supplementary data are available at NSR online.

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AUTHOR CONTRIBUTIONS

F.X. conceived the ideas and supervised the overall research. Y.S. and C.Y. synthesized high-quality WTe₂ bulk samples. C.H. and E.Z. fabricated the nanodevices. C.H., L.A. and S.L. carried out the PPMS measurements. A.N. and S.S. provided theoretical support. X.X. provided the curve fitting. C.H. and F.X. wrote the paper with assistance from all other authors.

Conflict of interest statement. None declared.

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