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Abstract

We study the electronic structure of WTe₂ thin films with different thicknesses. High-quality thin-film samples are obtained with carrier mobility up to 5000 cm² V⁻¹ s⁻¹, which enables us to resolve the four main Fermi pockets from Shubnikov-de Haas (SdH) oscillations. Angle-resolved SdH oscillations show that the WTe₂ thin films cross from three-dimensional to two-dimensional electronic systems at a thickness of ~ 20 nm. Using the field effect, the nature of the Fermi pockets in thin-film WTe₂ is identified, and the evolution of SdH oscillation frequencies is traced over different sample thicknesses. It is found that the frequencies dramatically decrease at a thickness of approximately 12 nm, which indicates the onset of finite-size effects on the band structure. Our work pins down two critical length scales of the thickness-dependent electronic structure in WTe₂ thin films.

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Thickness-dependent electronic structure in WTe₂ thin films

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I. INTRODUCTION

Tungsten ditelluride, one of the transition-metal dichalcogenides, has many interesting physical properties and has attracted a lot of attention recently. Bulk WTe₂ exhibits a nonsaturated and extremely large magnetoresistance (MR) which could be used to design devices such as magnetic sensors [1]. It is believed that the peculiar magnetoresistance occurs due to the nearly equal electron and hole concentrations with high carrier mobility [1–8], and forbidden backscattering due to strong spin-orbit coupling also plays an important role [9]. Surprisingly, although WTe₂ is a layered material, the anisotropy of the effective mass is small [10], and quantum oscillations can be observed along three different crystal axes [3,11], which makes it essentially a three-dimensional (3D) instead of two-dimensional (2D) electronic system [12].

WTe₂ is also a topological material. Bulk WTe₂ is predicted to be a type-II Weyl semimetal [13], in which Lorenz invariance is absent and the Weyl point appears at the boundary of electron and hole pockets [13,14]. This prediction has triggered renewed interest in this material [15–20]. In the monolayer limit, WTe₂ is predicted to be a topological semimetal in which both topological metallic edge states and 2D metallic bulk states are present [21]. Applying a small tensile strain, however, could lift

the overlap of the conduction and valence bands and turn it into a 2D topological insulator with a band gap of approximately 0.1 eV [21], where only the topological edge state is metallic [22,23]. The topological phase can be further tuned by a vertical electric field [21]. The relatively large band gap and electric field tunability are of great interest for fabrication of topological field-effect transistors with low-energy dissipation [21]. Recent transport measurements indicate that the helical conduction is indeed present on the edge of monolayer WTe₂, but surprisingly, its interior is insulating without applying an external tensile strain [24], which is consistent with a 2D topological insulator. However, even before the monolayer limit is reached, little is known about the evolution of the electronic structure of WTe₂ as the samples are made thinner and cross over to the 2D regime [24,25].

In this paper, we use quantum oscillations to study the electronic structure of WTe₂ thin films with different thicknesses. Angle-dependent Shubnikov–de Haas (SdH) oscillations reveal a crossover from a 3D to 2D electronic system as the WTe₂ sample thickness is reduced. Separate measurements of the field effect enable us to identify the nature of the Fermi pockets and trace their evolution as the thin-film thickness is varied. It is found that the overlap of conduction and valence bands is reduced as the samples are made thinner.

II. EXPERIMENTAL METHOD

WTe₂ thin films with different thickness are cleaved from bulk WTe₂ single crystals by the microexfoliation method onto

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285-nm SiO₂/Si substrates with alignment markers. Electron-beam lithography (EBL) is used to fabricate the alignment makers, electrodes, and bonding pads. Before Ti/Au electrodes with a typical thickness of 10 nm/60 nm are deposited, the contact areas are treated with Ar plasma to remove native oxides. To further reduce the contact resistance, the devices are annealed in a furnace with N₂ gas at 200 °C for 2 h. To minimize the oxidation of the sample surface, after cleaving, the thin films are exposed to air only before the contact area patterning with EBL and during the bonding of the devices to the chip carriers. At other times, the thin films are always covered by poly(methyl methacrylate) (PMMA) and stored in a vacuum desiccator or a N₂ glove box. The thicknesses of the thin films were measured by an atomic force microscope after the magnetotransport measurement. Magnetotransport measurements were performed in Oxford dilution fridges with *in situ* rotators [26] using standard low-frequency lock-in techniques. Unless otherwise stated, magnetotransport measurements were performed at $T = 30$ mK with magnetic fields up to 10 T.

III. RESULTS AND DISCUSSION

We begin by characterizing the film quality to confirm that no obvious degradation happens during the sample fabrication and storage [27–30] and that they are of high enough quality to reveal the intrinsic electronic structure. Figures 1(a)–1(e) show the magnetic field dependence of ΔR_{xx} of five WTe₂ thin films with different thicknesses, which are obtained by subtracting R_{xx} with polynomial backgrounds. Shubnikov–de Haas oscillations can be observed starting from 2 to 5 T depending on the thin-film thickness, which indicates that the carrier mobility is around 5000 to 2000 cm² V^{−1} s^{−1} since SdH oscillations become observable when $\mu B \approx 1$, where μ is the carrier mobility and B is the magnetic field. Beating patterns can be observed in the SdH oscillations of

FIG. 1. Characterization of the thin-film quality. (a)–(e) Shubnikov–de Haas (SdH) oscillations of the thin films with thicknesses of 30 nm (S70), 20 nm (S65), 15 nm (S69), and 10 nm (S68). ΔR_{xx} are obtained by subtracting polynomial backgrounds from magnetic-field-dependent R_{xx} . The insets are optical images for each device. (f) The magnetic-field-dependent magnetoresistance (MR) of corresponding thin films in (a)–(e). (g) Residual resistance ratio (RRR) and mobility of the thin films with different thickness.

all samples shown in Figs. 1(a)–1(e), which suggests that multiple oscillation frequencies are involved. The high carrier mobility and the beating patterns indicate that the quality of the WTe₂ thin films is relatively high. The high quality of the samples can be further confirmed by their magnetoresistance,

FIG. 2. Crossover to two-dimensional electronic systems. (a)–(d) Angle-dependent fast Fourier transform (FFT) frequencies of SdH oscillations of the 10-, 15-, 20-, and 30-nm WTe₂ thin films. The squares, circles, triangles, and diamonds represent the frequencies F_1 , F_2 , F_3 , and F_4 from experiments, respectively. The FFT frequencies in (b) also include the data measured at 200 mK and magnetic field up to 15 T (see Fig. 6 in Appendix A). The solid lines are angle-dependent FFT frequencies F expected from 2D systems where $F = F_0 / \cos(\theta)$ and F_0 are the frequencies at $\theta = 0^\circ$. The insets in (a) and (b) are the schematic diagram of the angle-dependent measurement configuration and a typical FFT spectrum with $\theta = 90^\circ$, respectively. The error bars represent the uncertainty to determine the frequencies in FFT spectra due to FFT resolution. (e) ΔR_{xx} of the 15-nm thin-film sample (S69) at 90° and 0° after the polynomial background subtraction.

FIG. 3. Field effect on a 12-nm WTe_2 film (S67). (a) Back-gate voltage-dependent R_{xx} at 200 mK and 0 T. (b) Plot of SdH oscillations at different V_{bg} . The color scale indicates the amplitude of the SdH oscillations. The measurement is performed at 200 mK and magnetic field up to 15 T. (c) Color map of the FFT spectrum of SdH oscillations at different V_{bg} . The cyan dashed lines are a guide to the eye showing how each frequency changes with V_{bg} . (d) FFT spectra of SdH oscillations at different V_{bg} .

MR = $[R_{xx}(B) - R_{xx}(0)] \times 100\% / R_{xx}(0)$, as shown in Fig. 1(f). The magnitude of the MR in compensated semimetals depends on the mobility of the carriers, $MR = \mu_e \mu_h B^2$, where μ_e and μ_h are electron and hole mobilities, respectively. Therefore, the larger the MR is, the higher the mobility is. The magnetoresistance of our thin films varies between 600% and 80% [see Fig. 1(f)], which is comparable to the largest MR reported in non-h-BN-encapsulated WTe_2 thin-film samples with a similar thickness [28,29,31,32]. We also note that the mobility decreases as the sample thickness is reduced, as shown in Fig. 1(g). The decrease in the mobility can be ascribed to surface scattering, which affects the mobility of thinner samples more than thicker samples [28,31]. This is consistent with the decrease in the resistivity residual ratio R_{300K}/R_{4K} shown in Fig. 1(g) as the sample thickness is reduced, which is another indication of the sample quality [1,3,4].

We now analyze the electronic structure of the five WTe_2 thin films. Angle-resolved quantum oscillations can map the detailed structure of the Fermi surface [33,34]. We measured SdH oscillations of WTe_2 thin films over a wide range of angles using the measurement configuration shown in the inset of Fig. 2(a). θ is the tilt angle between the normal of the sample surface and the magnetic field direction. Since WTe_2 has multiple Fermi surfaces [3–5,11], to illustrate the detailed structure of each Fermi surface, the SdH oscillations have been fast Fourier transformed to resolve corresponding oscillation frequencies. Four main frequencies, F_1 , F_2 , F_3 , and F_4 , can be resolved from thin-film samples, as shown in a typical fast Fourier transform (FFT) spectrum in the inset of Fig. 2(b) (for a detailed description of the method we used to trace the four frequencies at different angles, see Appendix A), and agree

with the four fundamental frequencies in the high-mobility bulk samples [3,4,11].

However, in the thin-film samples with thickness $t \leq 20$ nm, the angle dependence of these four frequencies follows the expected trend for a 2D system, $F = F_0 / \cos(\theta)$, where F_0 are the frequencies at $\theta = 0^\circ$ [see Figs. 2(b)–2(e)]. This is in contrast to the angle dependence of bulk samples, which follows the ellipsoidal model of 3D electronic systems [3,11]. This is also different from the 30-nm sample (S70) in Fig. 2(d), the frequencies of which start to deviate from the 2D model at approximately 20° – 30° . Furthermore, as shown by the black trace in Fig. 2(e) for a typical thin-film sample following the 2D model, no SdH oscillations can be observed at $\theta = 90^\circ$, and the corresponding amplitude of ΔR_{xx} is comparable to the noise level of ΔR_{xx} at low magnetic fields when $\theta = 0^\circ$. However, in bulk samples, quantum oscillations can be observed when magnetic field is parallel to the sample surfaces, i.e., $\theta = 90^\circ$ [3,11]. Therefore, the angle-dependent measurements indicate the WTe_2 thin-film samples with $t \leq 20$ nm are in the 2D regime, and the thin-film sample with a thickness of 30 nm is more electronically 3D-like. Therefore, angle-dependent SdH oscillation measurements show that as the WTe_2 samples are made thinner, the spatial confinement leads to crossover from 3D to 2D electronic systems. This also agrees with the observation of the higher 2D subbands in the 20-nm sample (see Appendix D).

Next, we wish to study the evolution of the Fermi pockets associated with the four main frequencies as the sample thickness is varied. To do this, we begin by identifying the nature (electron or hole) of the associated Fermi pockets. We utilize a back-gate voltage V_{bg} , which has two effects on the transport properties. First, V_{bg} can vary the density of electrons and

holes in opposite ways, which alters the FFT frequency of SdH oscillations. Second, V_{bg} can change the surface scattering (mobility) of electrons and holes in different ways, which alters the FFT amplitude of SdH oscillations [32]. Hence, by studying the V_{bg} dependence of the FFT frequency and amplitude for each of the four frequencies, it is possible to determine whether they correspond to electrons or holes. To that end, we use the 12-nm sample (S67) to study the back-gate dependence. We first show that the field effect indeed works: R_{xx} monotonically increased as the back gate was swept from -60 to 60 V, as shown in Fig. 3(a), and both the amplitude and period of the SdH oscillations, which were measured at 200 mK and magnetic field up to 15 T, are affected by V_{bg} [see Fig. 3(b) and Appendix C].

To identify the nature of the Fermi pockets by the V_{bg} dependence of FFT frequency, SdH oscillations measured at different V_{bg} are fast Fourier transformed and shown in a color map [Fig. 3(c)]. When the back gate is swept from -60 to 60 V, the Fermi level is shifted up. Meanwhile, the size of electron pockets becomes larger, while the size of hole pockets becomes smaller, because in WTe₂ the Fermi level lies in both conduction and valence bands. According to the Onsager-Lifshitz equation, $F = (\hbar/2\pi e) A_F$ where A_F is the area of electron and hole pockets [33], the oscillation frequencies F from electron pockets should become larger, and those from hole pockets should become smaller. From Fig. 3(c), it can be seen that F_1 and F_4 monotonically decrease as the back gate is swept from -60 to 60 V, which is consistent with hole behavior. F_2 and F_3 do not shift for negative V_{bg} but increase for positive V_{bg} , which is consistent with electron behavior. In addition, as shown in Fig. 3(d), the amplitudes of the F_2 and F_3 peaks are monotonically suppressed as the back gate is swept from -60 to 60 V. This also agrees with electron behavior as the negative V_{bg} would push electrons away from the surface and reduce the surface scattering, thus increasing the mobility, while the positive gate voltage has the opposite effect. The nature of the Fermi pockets we have assigned here for the thin films agrees with that in bulk samples [3,11].

In the high-mobility samples of bulk WTe₂, four fundamental frequencies are identified [3,4,11]. In our high-quality samples of thin-film WTe₂, four main frequencies are also observed, as indicated by the dashed lines in Fig. 4(a) [35]. The observation of both electron and hole pockets shows that the thin-film samples down to 10 nm thick are in the semimetal regime. By comparing the oscillation frequencies for different thicknesses as shown in Fig. 4(b), it can be seen that all four frequencies decrease dramatically for the samples thinner than 12 nm. A decrease in the frequencies (Fermi surface area) for both the electron and hole pockets in a 2D system cannot be caused by a shift in the Fermi level E_F because this would make the electron and hole oscillations change in opposite directions [compare to Fig. 3(c), where a change in E_F is caused by the gate bias]. An alternative is that the finite-size effects are affecting the band structure, reducing the overlap between the conduction and valence bands, as shown schematically in Fig. 4(c). (This is consistent with the increases in band gap observed in thin-film 2D semiconductors such as MoS₂ and black phosphorus [36,37]). To illustrate this point, we use a minimal model of WTe₂ to calculate the thickness-dependent oscillation frequencies from the Fermi pocket sizes (see Ap-

FIG. 4. Reduced overlap of conduction and valence bands in WTe₂ thin films. (a) FFT spectra of SdH oscillations of WTe₂ samples with different thicknesses at $\theta = 90^\circ$. The FFT spectrum of the 12-nm sample is measured at 200 mK and magnetic field up to 15 T. The FFT of the 10-nm sample is an average of $F \cos \theta$ over 11 angles from 0° to 25° to improve the signal-to-noise ratio. (b) Thickness dependence of FFT frequencies. The thicknesses of bulk samples from Refs. [3–5,11] are set as infinite compared with the thin-film samples. The error bars represent the uncertainty to determine the frequencies in FFT spectra due to FFT resolution. (c) Schematic diagrams showing the thickness-dependent Fermi pocket size in half of the Brillouin zone (top panels) and the band structure near the Fermi level along the Γ -X direction (bottom panels). Note that the schematic diagram shows only the relative change in the Fermi pocket size and the band overlap; the change in the band curvature and the Fermi pocket position cannot be measured in the current experiment. (d) The thickness-dependent quantum oscillation obtained from a minimal model of WTe₂ (see Appendix F).

pendix F). As shown in Fig. 4(d), this model captures the decrease of the four main frequencies as the thickness is reduced. In particular the frequencies decrease most dramatically in the thin regime. Although this minimal model is too simple to quantitatively fit the experimental data, the behavior of thickness-dependent frequencies in Fig. 4(d) agrees qualitatively with the experimental observation in Fig. 4(b). Therefore, our results suggest that spatial confinement may drive the change in electronic structure, especially for samples thinner than 12 nm.

IV. CONCLUSION

We have performed a systematic study of the electronic structure of high-quality WTe₂ thin films as the sample thickness is varied and found two critical length scales. The angle-dependent quantum oscillations reveal a crossover from 3D to 2D electronic systems at a thickness of approximately 30 nm. The other critical thickness is approximately 12 nm, where the SdH oscillation frequencies show a dramatic decrease, which

FIG. 5. ΔR_{xx} vs B at different angles for (a) 15-, (b) 20-, and (c) 30-nm samples. The data are vertically offset for clarity. (d)–(f) FFT spectra corresponding to the SdH oscillations in (a)–(c), respectively.

indicates the spatial confinement starts to have a significant effect on the band structure.

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FIG. 6. (a) ΔR_{xx} vs B , (b) the corresponding FFT spectra, and (c) color map of FFT spectra at different angles for the 10-nm sample (S68). The dashed lines in (c) are the angle-dependent FFT frequencies expected from 2D systems, where $F = F_0 / \cos(\theta)$ and F_0 are the frequencies at $\theta = 0^\circ$. (d) ΔR_{xx} vs B at different angles measured at 200 mK and a magnetic field up to 15 T. (e) The corresponding FFT spectra; the dashed lines indicate the frequencies F_2 and F_3 . The data in (a), (d), and (e) are vertically offset for clarity.

FIG. 7. Angle-dependent FFT frequencies extracted from (a) bulk reference samples [3] and our (b) 30-nm and (c) 20-nm thin-film samples. The squares, circles, triangles, and diamonds represent the frequencies F_1 , F_2 , F_3 , and F_4 from experiments, respectively. The solid lines are angle-dependent FFT frequencies F expected from 2D systems, where $F = F_0 / \cos(\theta)$ and F_0 are the frequencies at $\theta = 0^\circ$.

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APPENDIX A: ANGLE-DEPENDENT SDH OSCILLATIONS

To resolve the detailed structure of the Fermi surfaces of WTe_2 thin films, we perform angle-dependent SdH oscillations over a wide range of angles. In Figs. 2(a)–2(d) of the main text, we showed the angle-dependent FFT frequencies; here we show how we obtain these frequencies. Figures 5(a)–5(c) show the SdH oscillations ΔR_{xx} at different tilt angles, which

are obtained by subtracting R_{xx} with a polynomial background. Figures 5(d)–5(f) show the corresponding FFT spectra. Thanks to high carrier mobility in the 15-, 20-, and 30-nm samples, SdH oscillations measured at 30 mK with a magnetic field up to 10 T are enough to resolve clear FFT spectra for identifying the four main frequencies over a wide range of angles.

In the 10-nm sample (S68), the mobility is relatively low, which damps the amplitude of SdH oscillations [see Fig. 1(e) in the main text] and decreases the number of SdH oscillations that can be observed below a magnetic field of 10 T [see Fig. 6(a)]. As a result, the signal-to-noise ratio of the FFT spectra as shown in Fig. 6(b) is not as good as those in Figs. 5(d)–5(f). To distinguish the FFT frequencies of SdH oscillations from the noise frequencies, the FFT spectra in Fig. 6(b) are replotted in the form of a color map [see Fig. 6(c)], where the four main FFT frequencies of SdH oscillations can

FIG. 8. (a) R_{xx} vs B at different V_{bg} for the 12-nm sample (S68) and (b) the corresponding ΔR_{xx} after polynomial background subtraction.

FIG. 9. (a) Color map of the FFT spectra of the 10-nm sample (S68) at different angles after converting F to $F \sin \theta$. (b) FFT spectrum after summing $F \cos \theta$ over 11 different tilt angles from 0° to 25° in (a).

be more easily traced than those in Fig. 6(b). The four main frequencies follow the 2D model where $F = F_0 / \cos(\theta)$ and F_0 are the frequencies at $\theta = 0^\circ$, as indicated by the dashed lines. In order to resolve the FFT frequencies for a wider range of angles, we also measured this 10-nm sample in another cryostat at 200 mK with a magnetic field up to 15 T after a thermal cycle, as shown in Fig. 6(d). The relatively high measurement temperature and the thermal cycle damped the SdH oscillations from the low-mobility carriers in the hole pockets; however, the FFT frequency from two electron pockets can still be observed up to 55° , as shown in Fig. 6(e).

To better illustrate the difference between the 3D ellipsoidal and 2D Fermi surfaces, in Fig. 7 we show a comparison between the angle-dependent FFT frequencies of the bulk WTe₂ samples with a 3D ellipsoidal Fermi surface from Ref. [3] and our thin-film samples. The solid lines show the angle-dependent FFT frequencies expected from a 2D electronic system, where $F = F_0 / \cos(\theta)$ and F_0 are the frequencies at $\theta = 0^\circ$. The frequencies from the bulk reference samples in Fig. 7(a) and our 30-nm sample in Fig. 7(b) deviate from the 2D trend at $|\theta| \sim 20^\circ$ – 30° . However, for the frequencies obtained

from our 20-nm sample in Fig. 7(c), no deviations from the 2D model are visible.

APPENDIX B: FIELD EFFECT ON SDH OSCILLATIONS

Using the field effect to identify the nature of the Fermi pockets, we choose a relatively thin sample with a thickness of 12 nm so that the total carrier density can be tuned by a back gate. The magnetotransport measurement was performed in a cryostat at 200 mK with a magnetic field up to 15 T to increase the number of SdH oscillations that can be observed [see Fig. 8(a)]. It can be seen that the back-gate voltage V_{bg} changes the magnetic field dependence of R_{xx} . The SdH oscillations shown in Fig. 8(b) are obtained by subtracting the magnetic-field-dependent R_{xx} with polynomial backgrounds. The amplitude of SdH oscillations is gradually damped as the back gate is swept from -60 to 60 V. The oscillation period is also shifted by V_{bg} , and the shift is more obvious in the low magnetic field where the degeneracy of Landau levels is smaller than that in the high field.

TABLE I. Two-dimensional carrier densities based on the four main frequencies calculated for samples with $t \leq 20$ nm using $n_{2D} = g_s g_v e F / h$, where g_s and g_v are the spin and valley degeneracies and e , F , and h are the electron charge, SdH oscillation frequency, and Planck constant, respectively.

Thickness (nm)	$n_1 (\times 10^{13} \text{ cm}^{-2})$	$n_2 (\times 10^{13} \text{ cm}^{-2})$	$n_3 (\times 10^{13} \text{ cm}^{-2})$	$n_4 (\times 10^{13} \text{ cm}^{-2})$	$n_{\text{total}} (\times 10^{13} \text{ cm}^{-2})$
10	0.66	1.11	1.21	1.29	4.27
12	0.77	1.20	1.31	1.46	4.74
15	0.80	1.22	1.33	1.48	4.82
20	0.78	1.19	1.33	1.52	4.83

FIG. 10. Color map of FFT spectra of the 20-nm-sample (S65) at different angles after converting F to $F \sin \theta$. The dashed lines indicate the four main frequencies, and the dotted lines indicate the frequencies from higher 2D subbands.

APPENDIX C: IDENTIFYING THE FOUR MAIN FREQUENCIES OF THE 10-NM SAMPLE

In Fig. 6 we show how we identify the four main FFT frequencies for the 10-nm sample (S68). In order to compare its FFT frequencies with those from other samples as we discuss in Fig. 4 of the main text, a single FFT spectrum showing clear FFT peaks from SdH oscillations at $\theta = 0^\circ$ is necessary. To that end, we sum up $F \sin \theta$ over 11 different tilt angles from 0° to 25° to improve the signal-to-noise ratio. Because the 10-nm-thick sample is in the 2D regime, as we show in Fig. 6 and also Fig. 2(a) of the main text, $F \cos \theta$ at different θ should be the same, and the sum of $F \cos \theta$ at different θ could increase the amplitude of FFT frequencies from SdH oscillations. But the noise frequencies are random, and their amplitude will be averaged after the sum. So, first, we convert F to $F \cos \theta$ for 11 different tilt angles from 0° to 25° , as shown in Fig. 9(a). We can see that the four frequencies align as horizontal lines,

as expected from a 2D system. Then we sum them up and get the single FFT spectrum shown in Fig. 9(b), which is the FFT spectrum of the 10-nm sample in Fig. 5(a) of the main text.

APPENDIX D: EMERGENCE OF HIGHER 2D SUBBANDS

Figure 10 shows a color map of the FFT spectra of the quantum oscillations of a 20-nm sample (S65) at different angles. We can see that besides the four main frequencies indicated by dashed lines, some oscillation frequencies with a relatively small amplitude can also be observed, as indicated by the dotted lines, which cannot be observed in the FFT spectrum of the WTe_2 bulk samples. Although the amplitudes of these oscillation peaks is relatively small compared to those of the four main frequencies, they are distinguished from the noise background peaks. More importantly, they can be systematically traced over different angles and follow the angle-dependent oscillation frequency of 2D systems. These features are consistent with the spatial-confinement-induced multiple higher 2D subbands because, as the samples are made thinner, the spatial confinement will increase the energy spacing of different subbands, which leads to multiple other frequencies in addition to the four main frequencies in the FFT spectra. If the Fermi surface of WTe_2 is elongated only as the sample is made thinner, it is not possible to observe the frequencies from the higher 2D subbands. This observation agrees with the crossover from a 3D to 2D system observed in Fig. 2

APPENDIX E: TWO-DIMENSIONAL CARRIER DENSITIES CALCULATED FROM THE FOUR MAIN SDH OSCILLATION FREQUENCIES

As we showed in Fig. 2 of the main text, the WTe_2 thin films with $t \leq 20$ nm are in the 2D electronic regime; therefore, we can calculate their 2D carrier density, $n_{2D} = g_s g_v e F / h$, from the four main SdH oscillation frequencies F , with spin degeneracy $g_s = 2$, valley degeneracy $g_v = 2$, oscillation frequency F , elementary charge e , and Planck constant h , as shown in Table I. It should be noted that our samples (down to 10 nm) have not yet reached the 2D quantum limit; that is, only a single subband is occupied in each valley. As we can see in Fig. 10, higher 2D subbands are present. This also agrees with the fact that as the back-gate voltage varies, the total carrier density change calculated from the four main frequencies is smaller than that calculated from the back-gate capacitance.

APPENDIX F: TOY MODEL

A simple $\mathcal{T}$ -symmetric model that can describe a quantum transition from a topological semimetal to a trivial semimetal is

$$\hat{\mathcal{H}}_{\text{toy}} = \begin{pmatrix} \hat{h}(\mathbf{k}) & 0 \\ 0 & \hat{h}^*(-\mathbf{k}) \end{pmatrix}, \quad (\text{F1})$$

with

$$\hat{h}(\mathbf{k}) = \begin{pmatrix} E - \beta[k_x^2 + k_y^2 + (k_z + k_s)^2] & \alpha k_- \\ \alpha k_+ & -E + \beta[k_x^2 + k_y^2 + (k_z - k_s)^2] \end{pmatrix}. \quad (\text{F2})$$

For $k_s = 0$, this 2×2 block reads

$$\hat{h}(k_s = 0) = \begin{pmatrix} E - \beta \mathbf{k}^2 & \alpha k_- \\ \alpha k_+ & -E + \beta \mathbf{k}^2 \end{pmatrix}, \quad (\text{F3})$$

which is nothing but the minimal model of a type-I Weyl semimetal. Thus, the total Hamiltonian describes a Dirac semimetal with two copies of the Weyl semimetal. If $E\beta > 0$, the two bands intersect at $(0, 0, \pm k_w)$ with $k_w \equiv \sqrt{E/\beta}$. As one turns on k_s , the upper and lower bands move towards opposite

directions along k_z . For a nonzero k_s with $k_s < k_w$, the system turns into a tilted Dirac semimetal. If $k_s > k_w$, the system becomes a trivial semimetal with both an electron and a hole pocket on the Fermi surface. And the topological transition point is located at $k_s = k_w$.

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