

RESEARCH ARTICLE | JUNE 20 2023

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Appl. Phys. Lett. 122, 253101 (2023)

<https://doi.org/10.1063/5.0151240>





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Cite as: Appl. Phys. Lett. **122**, 253101 (2023); doi: [10.1063/5.0151240](https://doi.org/10.1063/5.0151240)

Submitted: 20 March 2023 · Accepted: 6 June 2023 ·

Published Online: 20 June 2023



View Online



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ABSTRACT

We report the magneto-transport measurements of thin film devices of the topological Weyl semimetal WTe₂ with the applied current along and vertical to the in-plane directions. The device is composed of a van der Waals thin film of WTe₂ sandwiched between top and bottom Au electrodes. At low temperatures, we observe not only a large unsaturated magnetoresistance and Shubnikov–de Haas oscillations with current in the in-plane direction but also a saturated vertical longitudinal magnetoresistance and quantum oscillations with current in the out-of-plane direction in a thin film of WTe₂. Our work provides insight into the origin of the unsaturated magnetoresistance in WTe₂ and may inspire non-planar engineering to reach higher integration in spintronics.

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WTe₂ is a layered transition-metal dichalcogenide (TMD) consisting of a tungsten layer surrounded by two tellurium layers stacked along the *z* axis. It exhibits an extremely large magnetoresistance (XMR) and shows no sign of saturation in high fields.¹ The XMR in the type-II Weyl semimetal WTe₂ recently stimulates extensive theoretical and experimental magneto-transport investigations.^{2–6} ARPES experiment in WTe₂ suggests that the XMR is attributed to the compensation between the balanced electron and hole populations,⁶ like the behavior expected in a perfectly compensated semimetal.⁷ In contrast to the normal semimetal, WTe₂ is a type-II Weyl semimetal, where Fermi surfaces (FSs) consist of a pair of electron- and hole-pockets contacting at the Weyl node.^{5,8} Shubnikov–de Haas (SdH) oscillations and Seebeck and Nernst measurements support the Fermi surface consisting of two pairs of electron-like and hole-like pockets.^{2,3}

With the reduction of thickness, a metal-to-insulator transition is observed in thin WTe₂ flakes.⁹ Gate-tunable magnetoresistance is found in ultra-thin WTe₂.^{10,11} In atomically thin WTe₂, the magnetoresistance can be tuned from positive to negative.¹² In addition, by electrostatically doping ultra-thin WTe₂, the XMR is turned on and off.¹³ In a monolayer crystal WTe₂, the quantum spin Hall effect¹⁴ and

Landau quantization¹⁵ are observed. In bilayers of WTe₂, the nonlinear Hall effect is observed in spite of its non-magnetic and time-reversal-symmetric conditions.¹⁶ It needs further investigation in the WTe₂ ultra-thin flake, where the electronic structure is modulated by the dimension.¹⁷

In this paper, we fabricate a thin film device of WTe₂ with top and bottom electrode structures. By choosing the current directions, we performed anisotropic magneto-transport measurements in this device. We found a large unsaturated magnetoresistance with current in the in-plane direction and a saturated vertical longitudinal magnetoresistance. The Shubnikov–de Haas oscillations are also observed in the magnetoresistance at both directions at low temperature.

Single crystals of WTe₂ were synthesized using a self-flux method. WTe₂ thin flake was transferred with the polyvinyl alcohol (PVA), based on our previous technique.¹⁸ Figure 1 shows the schematics of the vertical contact fabrication and assembly process comprised of the following steps: (a) the electrodes are patterned when the mask is exposed to UV light. (b) Gold is deposited to the SiO₂/Si substrate and the residual photoresist is removed with a standard liftoff process as depicted. (c) PVA solution was spin-coated on the SiO₂/Si

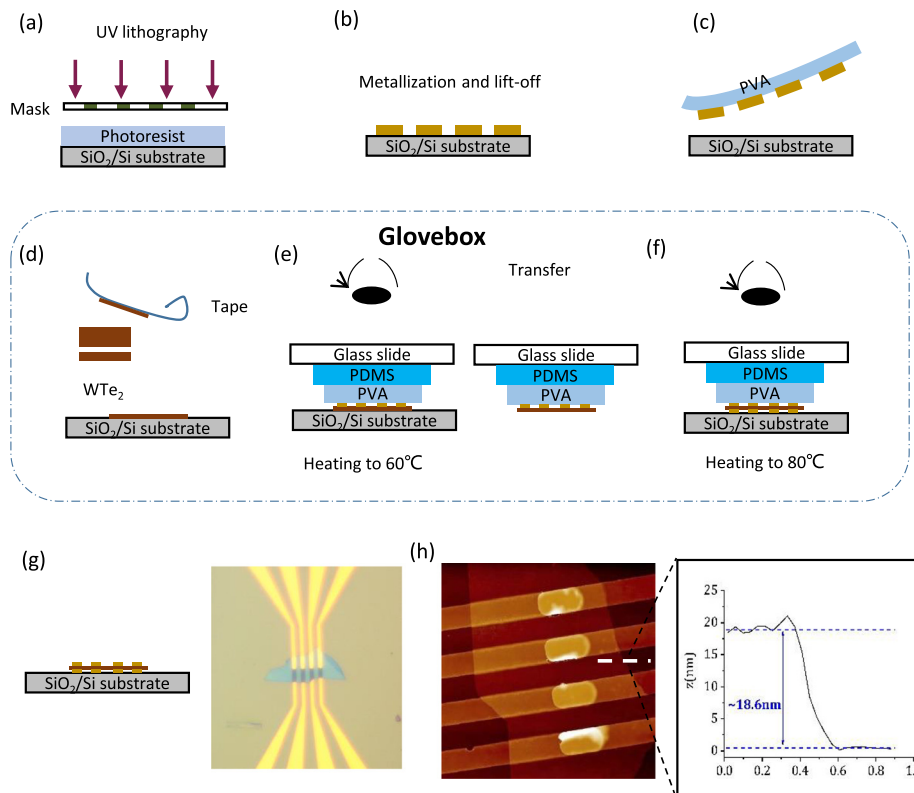


FIG. 1. Schematics of the fabrication process. (a) UV lithography patterning onto the SiO_2/Si substrate. (b) Au is deposited onto the SiO_2/Si substrate, and Au electrodes are left after a standard liftoff process applied to remove the photoresist. (c) The Au electrodes are peeled off by polyvinyl alcohol (PVA) at room temperature from the SiO_2/Si substrate. (d) Preparation of exfoliated WTe_2 flake on the SiO_2/Si substrate in glovebox. (e) The Au/PVA/PDMS prepared on glass slide are in contact with the thin WTe_2 film when heating the stage to 60°C . After heated for 2 min, the $\text{WTe}_2/\text{Au}/\text{PVA}/\text{PDMS}$ structure is obtained after gentle separation of the glass slide from the SiO_2/Si substrate. (f) Adjusting the relative position of Au electrodes on the SiO_2/Si substrate and $\text{WTe}_2/\text{Au}/\text{PVA}/\text{PDMS}$ on glass slide under an optical microscope and make gentle contact. The stage is kept heating to 80°C for 2 min. (g) After gentle separation from the PDMS on the glass slide, the Au-electrodes/ WTe_2 /Au-electrodes device is obtained. Schematic structure and optical image of the device are shown. (h) AFM micrograph of fabricated WTe_2 vertical structures shows the vertical channel region. Height profile (right inset) is extracted along the dotted line.

substrate with Au electrodes at a speed of 4000 rpm. It created a PVA film of about 900 nm. After 12 h in atmosphere, the PVA becomes dry and solid. At room temperature, the Au electrodes are gently peeled off from the SiO_2/Si substrate by PVA.^{18,19} (d) The WTe_2 thin flakes can be exfoliated using scotch tapes and attached to the SiO_2/Si substrate prior to plasma cleaning. (e) A piece of the Au/PVA strip is attached to the Polydimethylsiloxane (PDMS) and glass slide, under the microscope, they are aligned to the thin sample flake. After a short baking of 60°C , the sample can be gently separated from the substrate and grabbed by the PVA side. (f) We align the Au electrodes part on the SiO_2/Si substrate with the $\text{WTe}_2/\text{Au}/\text{PVA}/\text{PDMS}$ part on the glass slide under the microscope. After a short baking of 80°C , the top PDMS is detached. (g) After the PVA is washed away in de-ionized water and the part left is a Au/ WTe_2 /Au vertical structure on the SiO_2/Si substrate. The optical microscope image of the vertical device is shown in the right part. Atomic force microscopy (AFM), Fig. 1(h), reveals the vertical channel region (in bright yellow color) of the device. The width of the vertical channel is about $3\ \mu\text{m}$, and the height profile in the inset shows that the thickness of the WTe_2 flake is 18.6 nm, approximately 26 layers taking account of the monolayer 0.7 nm.

Figure 2(a) shows the scanning electron micrograph (SEM) of the WTe_2 van der Waals device. The thin flake of WTe_2 is sandwiched between Au electrodes. The top and bottom Au electrodes are laterally edge-to-edge separated by $3\ \mu\text{m}$ from each other, respectively. The mismatch among top and bottom electrodes is inspected by the magnified area in the inset. The mismatch length (d_s) is about 0.69 and $0.72\ \mu\text{m}$, with a small inclination of about 4.3%. The device can be

used for measuring the planar and vertical transport properties at the same sample. For the in-plane transport configuration, the electrodes 5 and 8 are used as current probes and inner electrodes 6 and 7 are connected to a voltmeter. While for vertical longitudinal transport configuration, the electrodes 2 and 6 are used as current probes and the electrodes 3 and 7 as voltage probes. The resistance was measured by two sets of lock-in amplifiers Stanford SR830 in a 14 T Oxford instrument Teslatron PT. To avoid the influence of noise from different directions, the vertical longitudinal transport measurements were performed subsequently after the in-plane measurements. In our measurement, the four probe resistances for R_{ab} and R_c measured at 300 K were 45.7 and $0.66\ \Omega$, respectively.

Figures 2(b) and 2(c) show the temperature (T) dependence of the normalized resistances (R_{ab} and R_c) at the same WTe_2 sample under the perpendicular external magnetic fields and the applied current (I) along the in-plane and the out-of-plane directions, respectively. It is found that the R_{ab} at 0 T decreases monotonically from room temperature as the temperature cools down, yielding a residual resistivity ratio (RRR) about 12.5 in Fig. 2(b). The R_{ab} shows a remarkable increase after reaching a minimum at the turn-on temperature, when a magnetic field is applied. The turn-on temperature is found to be shifted to a higher temperature as increasing H at the investigated magnetic field range. On the other hand, the RRR for R_c at 0 T is about 6.4 [Fig. 2(c)], which is less than the one for R_{ab} . Furthermore, the R_c shows a negligible increase at low temperature at the investigated $H \leq 14\ \text{T}$. It, therefore, suggests that the in-plane and out-of-plane transport have distinct electronic properties.

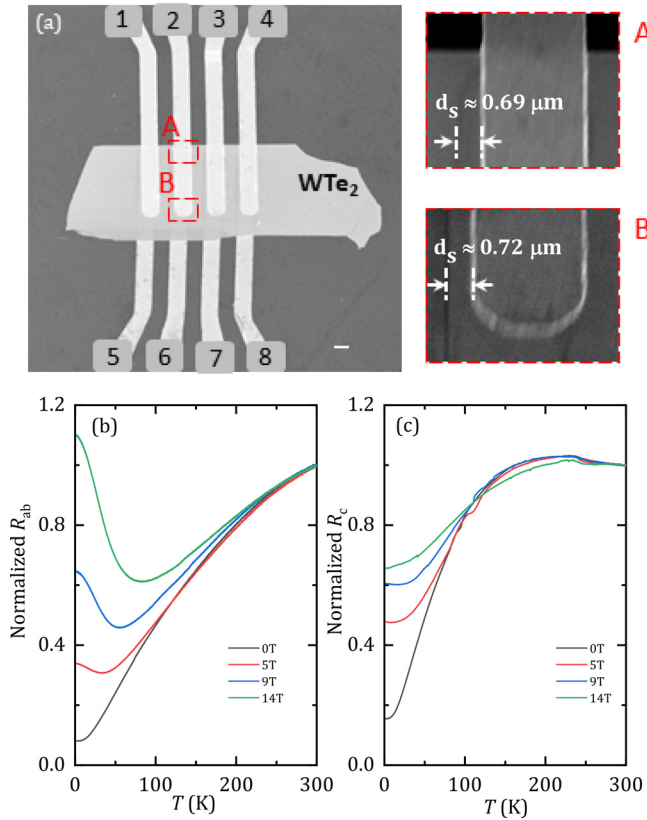


FIG. 2. Structure and transport behavior of the WTe₂ device. (a) Scanning electron micrographs of the WTe₂ device, consisting of the ultrathin WTe₂ film sandwiched between the top (marked with 1, 2, 3, and 4) and bottom (marked with 5, 6, 7, and 8) gold electrodes. The scale bar is 3 μm . The mismatch between the top and bottom electrodes is about 0.69 μm for the region A in the enlarged area while about 0.72 μm for the region B. The normalized resistance $R(T)/R(300\text{ K})$ vs T with the current along ab and c directions (b) and (c), respectively, under various magnetic fields H parallel to the c axis.

Figure 3(a) shows the H dependence of the in-plane magnetoresistance (MR_{ab}) at various temperatures (T). The MR_{ab} increases with H without a sign of saturation, consistent with the literature.^{1,3,20–22} At high H , the Shubnikov–de Haas quantum oscillations are observed in all the temperatures from 1.6 to 10 K. The oscillatory component can be extracted by subtracting a second-order polynomial background over the MR. The $\Delta R_{ab}/R_{ab}(H)$ as a function of $1/H$ is plotted in Fig. 3(b). Fast Fourier transformation (FFT) has been performed, and the amplitude spectra are illustrated in Fig. 3(c). The frequencies of the thin flake show four major peaks at 77.8, 100, 133, and 161 T, originated from two pairs of electrons and hole pockets.^{2,3,5} The decrease and shift of the SdH oscillation frequencies are due to the spatial confinement contributing to the electronic structure in thin samples.^{5,17} In addition to the four distinct frequencies, a fifth frequency ($P_5 = 254\text{ T}$) is detected. It is externally caused by the magnetic breakdown as suggested by a previous study.^{2,3} The peaks correspond to the orthogonal cross-sectional area of the Fermi surface (FS) A_F , as described by the Onsager relation $F = (\Phi_0/2\pi^2)A_F$, where Φ_0 is the flux quantum. The effective cyclotron mass of carriers at the observed Fermi surface

sheets is evaluated from the temperature dependence of normalized FFT amplitude using the Lifshitz–Kosevich (LK) formula,

$$\text{FFTamp} \propto \frac{\alpha m^* T/H}{\sinh(\alpha m^* T/H)},$$

where $\alpha = 2\pi^2 K_B / e\hbar$ and $m^* = m/m_e$ is the effective mass. Normalized FFT amplitudes of the four peaks for the MR_{ab} as a function of T are plotted in Fig. 3(d). Their effective masses are estimated to be $0.266m_e$, $0.206m_e$, $0.278m_e$, and $0.446m_e$ for oscillation peaks P_1 , P_2 , P_3 , and P_4 , respectively.

When the current is applied through the vertical channel, we can probe the vertical longitudinal magnetoresistance transport properties in one single sample. The H dependences of MR_c under different T are shown in Fig. 3(e). The MR_c at 1.6 K increases with H and saturates at large magnetic field, which is different with MR_{ab} of WTe₂. Since in the in-plane current configuration, the magnetoresistance of WTe₂ increases significantly with magnetic field without saturating,^{2,23} the mismatch or lateral component should not lead to a non-monotonic and saturated out-of-plane MR. The MR_c shows oscillations at the temperature range from 1.6 to 15 K but then disappear at higher temperatures. Similar to the MR_{ab} , the oscillatory component of MR_c is obtained after a background subtraction, and $\delta R_c/R_c(H)$ is periodic with $1/H$ as shown in Fig. 3(f). The resistance in the two directions is mainly limited by the geometry if ρ_{ab} and ρ_c are comparable in quantity. Since the current channel length is different, it is 3 μm for in-plane resistance while 18.6 nm for the out-of-plane resistance, the out-of-plane resistance is much smaller than the in-plane resistance in our measurement. Thus, the resolution of quantum oscillation is worse in the out-of-plane direction. We observe a weak peak $Q_1 = 66.7\text{ T}$ and an obvious sharp peak $Q_2 = 106\text{ T}$ for the MR_c in Fig. 3(g). According to the previous reports in the bulk sample, the observed frequencies with out-of-plane current are around those with in-plane current.^{3,24} The frequency Q_2 is similar to the P_2 , in agreement with the literature. After a fitting with the normalized amplitude A_c , the effective masses are estimated to be $0.124m_e$ and $0.143m_e$ [Fig. 3(h)], smaller than those obtained from the in-plane direction transport measurements.

In addition to the in-plane oscillation, we discover the quantum oscillation in the out-of-plane magnetoresistance. One possible interpretation would be that the Fermi arcs on the top and the bottom surface contribute to a novel type of quantum oscillation phenomena.²⁵ However, the underlying origin needs further experimental evidence and theoretical analysis. In bulk WTe₂, the resistivity first increases with magnetic field at low magnetic field before it starts oscillating with a damping out-of-plane magnetoresistance.²⁴ However, no sign of saturation is observed in the bulk. There is no previous report for out-of-plane magnetoresistance in WTe₂ thin films. The quantum oscillation appears in the suppressed magnetoresistance in the ultrathin film, consistent with the bulk, while it is much more suppressed to saturation. A similar magnetoresistance has been observed in a topological Weyl semimetal TaAs, which has a quadratic field dependent in-plane MR but a negative out-of-plane MR.^{26,27} It was interpreted that the electrical current along c direction causes an imbalance of the chemical potentials between the opposite chiral Weyl nodes, which leads to the negative magnetoresistance called as Adler–Bell–Jackiw anomaly.^{26,28–30} Thus, for the topological Weyl semimetal WTe₂, the saturated or negative out-of-plane magnetoresistance might

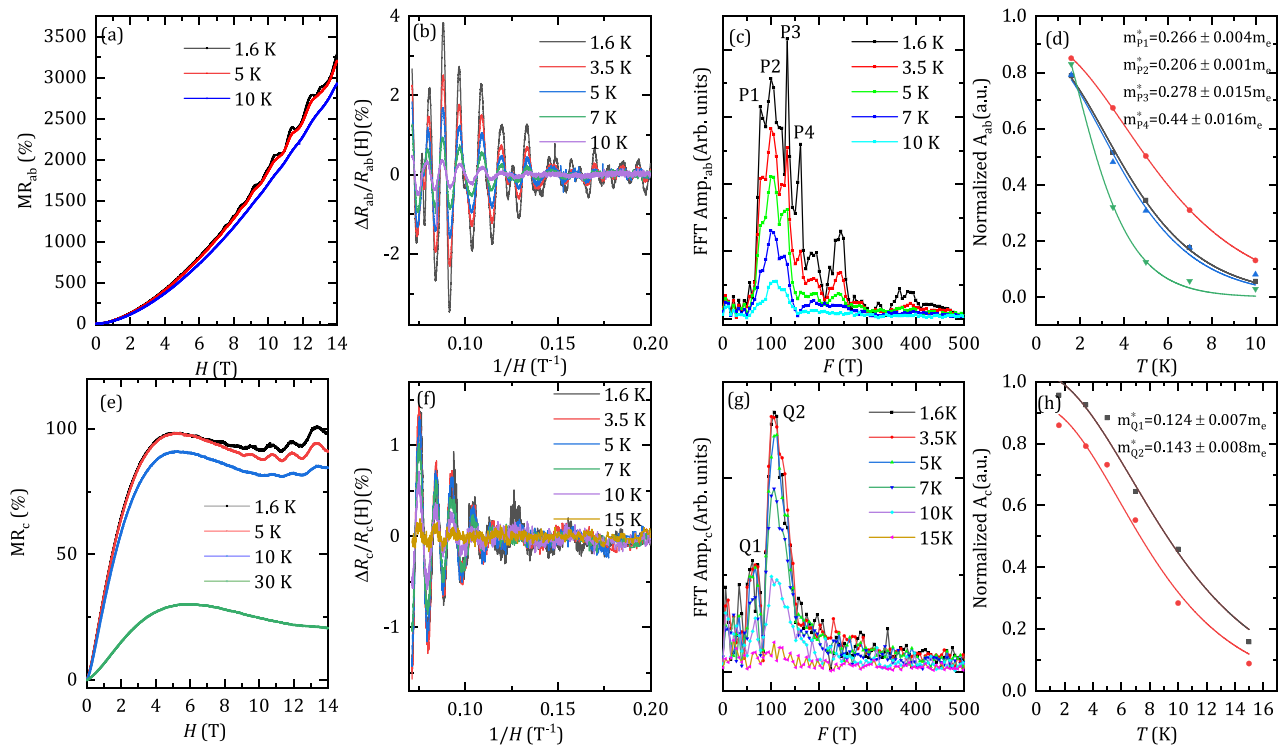


FIG. 3. The magneto-transport behaviors in thin film WTe_2 . The H dependence of the magnetoresistance (MR) with the current along ab (a) and c directions (e) at different T . SdH oscillations in MR_{ab} (b) and MR_c (f). The fast Fourier transform (FFT) spectrums of MR_{ab} (c) and MR_c (g). The T dependence of the normalized FFT amplitudes of MR_{ab} (d) and MR_c (h).

be attributed to the electrical current induced Adler–Bell–Jackiw anomaly.

In summary, we introduced the fabrication of the vertically assembled $\text{Au}/\text{WTe}_2/\text{Au}$ device and characterized its out-of-plane and in-plane transport properties. A saturated out-of-plane magnetoresistance is observed at low temperatures while it remains unsaturated for the in-plane magneto-transport. There are quantum oscillations discovered in the out-of-plane transport in addition to the in-plane transport. Our results suggest that the presence of different behaviors of magnetoresistance could be determined by applied current directions.

T.H. acknowledges the support of NSFC Grant No. 11574338. H.X. acknowledges the support of NSAF Grant No. U1530402. Y. G. Shi acknowledges the support of NSFC Grant No. U2032204 and the Informatization Plan of Chinese Academy of Sciences (No. CAS-WX2021SF-0102).

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Yinshang Liu: Conceptualization (equal); Data curation (equal); Formal analysis (equal); Writing – original draft (equal). **Hong Xiao:** Formal

analysis (equal). **Chi Zhang:** Data curation (equal); Formal analysis (equal). **Cuiwei Zhang:** Data curation (equal). **Yonguo Shi:** Data curation (equal). **Tao Hu:** Data curation (equal); Formal analysis (equal); Writing – review & editing (equal); Supervision (equal). **Claus M. Schneider:** Writing – review & editing (equal); Supervision (equal).

DATA AVAILABILITY

The data that support the findings of this study are available within the article.

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