Anisotropic Landau level splitting and Lifshitz transition induced magnetoresistance enhancement in ZrTe$_5$ crystals

I. Zhou$^1$, A. Ramiere$^1$, P. B. Chen$^1$, J. Y. Tang$^1$, Y. H. Wu$^1$, X. Lei$^1$, G. P. Guo$^1$, J. Q. He$^{1,3}$, and H. T. He$^{1,3}$

$^1$Department of Physics, Southern University of Science and Technology, Shenzhen 518055, People’s Republic of China
$^2$Key Laboratory of Quantum Information, CAS University of Science and Technology of China, Hefei 230026, People’s Republic of China
$^3$Authors to whom any correspondence should be addressed.

E-mail: hejtu@sustec.edu.cn and ht@ustc.edu.cn

Keywords: anisotropic landau level splitting, lifshitz transition, two-carrier transport

Abstract

Magneto-transport study has been performed in ZrTe$_5$ single crystals. The observed Shubnikov–de Hass quantum oscillation at low temperature clearly demonstrates the existence of a nontrivial band with small effective mass in ZrTe$_5$. Furthermore, we also revealed the highly anisotropic nature of high-field Landau level splitting in ZrTe$_5$, suggesting the dominant role of orbital contribution to the splitting. Besides these, an abnormal large enhancement of magnetoresistance appears at high temperatures, which is believed to arise from the Lifshitz transition induced two-carrier transport in ZrTe$_5$. Our study provides more understanding of the physical properties of ZrTe$_5$ and sheds light on potential application of ZrTe$_5$ in spintronics.

Dirac semimetals (DSMs) are three dimensional analogs of graphene, featuring gapless Dirac cones in the bulk states [1]. The well-studied DSMs, such as Na$_3$Bi and Cd$_3$As$_2$, have been shown to exhibit ultra-high mobility, extremely large positive magnetoresistance (MR), surface Fermi arc states, and a novel negative MR related with the chiral anomaly in high energy physics [2–9]. Therefore, DSMs have triggered extensive studies exploring the potential applications in spintronics, such as magnetic sensors, as well as the intriguing Dirac fermion physics [1].

Among various DSM candidates, ZrTe$_5$ is of particular interest. It has a layered crystal structure, which means that thickness-dependent physics can be easily accessed, even down to the ultrathin limit, i.e. one monolayer of ZrTe$_5$ [10, 11]. Besides this, ZrTe$_5$ has been shown to exhibit many intriguing phenomena, such as the chiral anomaly induced negative MR [12], the nontrivial Berry phase revealed in quantum oscillation experiment [13], the anomalous Hall effect even with the magnetic field in–plane [14], and possible emergence of density wave states in the quantum limit [15, 16]. What makes ZrTe$_5$ more special is that it lies at the boundary between weak and strong topological insulators [17]. Whether ZrTe$_5$ is a real DSM or not is still under active investigation in the literature [18–20].

In this work, we have performed systematic magneto-transport study of high-quality ZrTe$_5$ single crystals. Clear quantum MR oscillations were observed at low temperatures, revealing the existence of a nontrivial band with tiny effective mass in ZrTe$_5$. In the high field region, we also observed well-resolved Landau level splitting. It shows a very strong dependence on the tilting angle of the magnetic field, indicating the highly anisotropic nature of it, very different from previous transport studies [15]. Besides these, we also revealed an abnormal large enhancement of MR up to 820% at high temperatures, which was interpreted in terms of the Lifshitz transition induced two-carrier transport in ZrTe$_5$ and might be important for potential application of ZrTe$_5$ in spintronics.

The ZrTe$_5$ single crystals we studied in this work were grown by the chemical vapor transport method. Stoichiometric proportions of high purity Zr (GRINM 99.995%) and Te (Alfa Aesar 99.999%) were loaded in an evacuated quartz tube at a pressure below $3 \times 10^{-5}$ Pa and hold at 480°C for 12 d. The solid-state reaction created polycrystalline ZrTe$_5$ that was hand-milled, then loaded in a new evacuated quartz tube with 5 mg cm$^{-3}$ of iodine, and finally placed in a horizontal dual zone furnace with a temperature gradient 430 °C–520 °C for two weeks to grow the single crystals. ZrTe$_5$ has a layered orthorhombic crystal structure, with the 2D layers stacked...
in the \( b \)-axis. Each 2D layer consists of prismatic \( \text{ZrTe}_5 \) chains along the \( a \)-axis linked by zigzag chains of Te atoms in the \( c \)-axis. Therefore, the obtained \( \text{ZrTe}_5 \) crystals are usually needle-like and preferably grown along the \( a \)-axis. To investigate the magneto-transport properties of these \( \text{ZrTe}_5 \) crystals, six-terminal Hall devices were fabricated with gold wires attached by silver paint, as schematically shown in the inset of figure 1. The devices were then measured in an Oxford TeslatronPT system with the base temperature and highest magnetic field of 1.6 K and 14 T, respectively. A lock-in technique was implemented to improve the measurement accuracy. The ac amplitude and frequency of the measuring current is 1 mA and 17 Hz, respectively. Since similar results were obtained on different devices, we only present the data of a Hall device which has the thickness, width, and length of 33, 119, and 849 \( \mu \text{m} \), respectively.

Figure 1 shows the temperature \( (T) \) dependence of resistance \( (R) \) of the bulk \( \text{ZrTe}_5 \) sample. The resistance firstly increases with \( T \) decreasing from room temperature. But below 138 K, it shows a metallic behavior. A resistance peak thus appears at \( T_p = 138 \text{ K} \), similar to previous studies of \( \text{ZrTe}_5 \) [13, 15]. Note that the values of \( T_p \) can vary in a wide temperature range, depending on the thickness or carrier doping of \( \text{ZrTe}_5 \) [10, 21]. The emergence of such a peak anomaly is recently ascribed to the Lifshitz transition in \( \text{ZrTe}_5 \) [19–21].

We then investigated the magnetoresistance (MR) with the magnetic field \( (B) \) in the \( b \) direction, as shown in figure 2(a). At \( T = 1.6 \text{ K} \), clear Shubnikov–de Hass (SdH) oscillations are observed in low fields, indicating the formation of quantized Landau levels (LLs) in our sample. Also noticed is the huge MR peak appearing around \( 8 \text{ T} \), accompanied by a dramatic drop of MR around 12 T. Although previous study has explained this phenomenon in terms of the spin density wave in the quantum limit [13], direct evidence is still lacking. To examine the quantum oscillations more clearly, we subtract the smooth MR background from each \( R(B) \) curve in figure 2(a). The obtained \( \Delta R \) has been plotted as a function of \( 1 / B \) at each temperature, as shown in figure 2(b). For clarity, the \( \Delta R (1 / B) \) curves have been offset vertically. As expected for the SdH oscillation [13, 15], the MR oscillations in figure 2(b) are periodic in \( 1 / B \) and gradually fade out with increasing temperatures.

The Landau quantization condition is given by the Onsager relation
\[
\frac{\Delta R}{\Delta B} = 2\pi \left( n + \frac{1}{2} - \frac{\phi_b}{2\pi} - \delta \right) = 2\pi (n + \gamma),
\]
where \( \Delta R \) is the extremal cross-sectional area of Fermi surface perpendicular to the field, \( \hbar \) is the Plank’s constant, \( e \) is the electronic charge, \( n \) is the Landau level index, \( \phi_b \) is a geometrical phase, also known as the Berry phase, and \( \delta \) is a phase shift depending on the dimensionality of the system [21]. The value of \( \gamma \) provides valuable information about the topology of energy bands. For example, in 3D DSMs with linear energy dispersion, \( |\gamma| \) is expected to lie in the range between 0 and \( 1/8 \), indicating the presence of a nontrivial \( \pi \) Berry phase \( \phi_B \) [22, 23]. According to the Onsager relation, the Landau level index \( n \) should be a linear function of \( 1 / B \), the slope and \( n \)-intercept of which yield the values of \( \frac{\Delta R h}{2\pi e} \) and \( \gamma \), respectively. In figure 2(c), the Landau fan diagram \( n (\frac{B}{B_p}) \) at 1.6 K is plotted, with the peaks (or valleys) of the SdH oscillations indexed by integers (or half-integers). Indeed, the fan diagram can be well fitted linearly. The best linear fit of the fan diagram reveals a non-trivial Berry phase with \( \gamma = 0.09 \). From the slope of the fitting curve, \( A_F = 5.13 \times 10^{-4} \text{ A}^{-2} \) is also obtained, similar to previous studies of \( \text{ZrTe}_5 \) [13]. The carrier concentration is thus estimated to be around \( 7.1 \times 10^{16} \text{ cm}^{-3} \). It should be noted that the value is underestimated since the spherical geometry of the Fermi surface of \( \text{ZeTe}_5 \) is assumed in the calculation for simplicity, instead of the cylindrical geometry.
According to the Lifshitz-Kosevich (LK) formula\cite{21,23}, the SdH oscillation amplitude should decrease with increasing temperatures, which is described by the thermal damping factor $R_T = [(aTm^*/B) / \sinh (aTm^*/B)]$, where $a = 2\pi k_B / (e\hbar)$, $m^*$ is the effective mass, and $k_B$ is the Boltzmann constant. Therefore, from the analysis of the temperature dependence of the oscillation amplitude, one can derive the effective mass of the carriers. Figure 2(d) shows the fits of the normalized oscillation amplitude $(\Delta R_{xx}/\Delta R_{ax}(1.6 \text{ K}))$ of the 5th landau level to the damping factor $R_T$. The fitting yields $m^* = 0.033 m_0$, where $m_0$ is the free electron mass.

Therefore, the quantum oscillation results shown in figure 2 collectively reveal the presence of a band with a nontrivial Berry phase and a small effective mass in ZrTe$_5$. As discussed in previous transport studies, it is the conduction band at the $\Gamma$ point giving rise to the observed SdH oscillations\cite{13,15}. But recent ARPES and infrared spectroscopy studies of ZrTe$_5$ suggest that ZrTe$_5$ is more like a weak or strong topological insulator with a small gap at the $\Gamma$ point at low temperatures, rather than the DSM\cite{18–20}. This discrepancy suggests that the transport study alone cannot conclusively determine the nature of ZrTe$_5$, even a nontrivial Berry phase is extracted from the analysis of SdH data.

Figure 2(b) also shows a clear splitting of the 2nd LL at $T = 1.6$ K, which was previously ascribed to the splitting of the Dirac node to two energetically separated Weyl nodes in high magnetic fields\cite{13,15}. As the temperature increases, the splitting-induced double-peak structure gradually evolves into a single peak at $T = 14.6$ K, as seen in figure 2(b). This reflects the smearing out of the splitting by the increased thermal energy at high temperatures. According to previous studies\cite{24,25}, there are both Zeeman and orbital contributions to the exchange splitting induced by a magnetic field. The former mainly depends on the magnitude of the field and is isotropic with regard to the field direction, but the latter is highly anisotropic.

In order to discriminate the Zeeman and orbital contribution, we further studied the LL splitting in tilted magnetic fields at 1.6 K, as shown in figure 3(a). $\theta$ is the angle of the field with respect to the $b$-axis in the $ab$ plane (see the inset of figure 1). At small angles, the splitting of the 2nd LL is clearly resolved. But as $\theta$ increases, the two split peaks gradually emerge into a single one. In figure 3(b), we plot the angle dependence of the spacing between the two peaks, i.e. $\Delta_2 = 1/[(B_{z+} - B_{z-}) \cos \theta]$, where $B_{z,\pm}$ is the field position for the two split peaks.
of the 2nd LL, respectively. It can be seen that $\Delta_2$ almost keeps unchanged with increasing $\theta$, but beyond about 70°, a sharp drop of it to zero is observed. Such a strong angle dependence of $\Delta_2$ indicates the dominant anisotropic orbital splitting, rather than the isotropic Zeeman splitting. Similar orbital-dominant LL splitting has been also observed in some Dirac semimetals, such as Cd$_3$As$_2$.[25, 26]. Furthermore, in a previous study of ZrTe$_5$, [15], due to the limited range of $\theta$ ($\theta \leq 60^\circ$), the LL splitting was found to scale with the normal component of field $B_z$. But by studying the LL splitting in an extended range of $\theta$ as shown in figure 3, the splitting does not scale with $B_z$, but vanishes at large tilting angles. These observations thus provide new insight into the LL splitting in ZrTe$_5$ with strong spin–orbit interaction, i.e. it is the anisotropic orbital term that dominates the LL splitting in ZrTe$_5$, instead of the isotropic Zeeman term [24, 25]. Since the orbital splitting is much more sensitive to the tilting angles than the Landau quantization [25], this also explains why the splitting of the 2nd LL disappears at large tilting angles, but only a small shift of the 3rd LL is observed in figure 3(a). Note that the observed splitting is also not likely due to the exchange enhanced splitting observed in quantum Hall systems, as it predicts a square root $B$ dependence of the splitting [27]. This is not consistent with the experimental observation that the splitting $\Delta_2$ vanishes at large tilting angles, although the $B_z$, at which the splitting is observed, remains the same for different tilting angles.

We also studied the MR of ZrTe$_5$ at higher temperatures. As shown in figure 4(a), ZrTe$_5$ exhibits typical classical MR behaviors around 100 K, i.e. the MR firstly increases with increasing fields and then tends to saturate in high magnetic fields. But as the temperature increases above 140 K, a non-saturating MR up to 14 T is observed. In order to demonstrate the change of MR with $T$, the MR ratio, defined by $R(14 T)/R(0 T)$, is plotted as a function of $T$ in figure 4(b). Although the MR ratio is only about 230% at 100 K, it shows a rapid increase above 140 K. After a maximum value of about 820% is obtained at 170–180 K, the ratio decreases with increasing temperatures. Therefore, a large enhancement of MR is clearly observed around 170 K. Such an enhancement should be associated with the non-saturating behavior of MR shown in figure 4(a).

It has been shown that when a gapless semiconductor with linear energy dispersion enters the quantum limit, i.e. all carriers occupy the lowest LL in high fields, a linear and non-saturating MR is expected [28]. But this is apparently not applicable to our results, since the non-saturating MR in figure 4(a) is observed at quite high temperatures (> 140 K). Besides this quantum mechanism, inhomogeneity or mobility disorder can also give rise to a non-saturating MR in high fields [29]. But the observation of clear SdH oscillations in relatively low fields as shown in figure 2 indicates the high quality of our ZrTe$_5$ single crystals. After ruling out the above two mechanisms, we intend to understand what we observed in terms of the classical two-carrier transport model, which has been shown to result in extreme magnetoresistance (XMR) in topological semimetals with perfect compensation between hole and electron carriers, such as WTe$_2$ and TaAs$_2$.[30, 31].

To provide more evidence for this two-carrier transport model, we have also measured the Hall effect of our sample at high temperatures, as shown in figure 5(a). At 100 K, the measured Hall resistivity curve $R_{xy}(B)$ reveals the dominance of $n$-type carriers in ZrTe$_5$. But with temperatures increasing to about 140 K, the slope of the $R_{xy}(B)$ curve in low fields changes from negative to positive, implying the participation of holes in the transport in addition to electrons. One can also see from figure 5(a) that the hole contribution becomes more important with increasing temperatures and finally dominates the Hall effect up to 14 T at high temperatures. This Hall measurement clearly demonstrates the coexistence of electrons and holes in ZrTe$_5$, especially around 170 K, which coincides well with the temperature where the enhancement of MR is observed, as shown in figure 4(b).
Such a coincidence leads us to believe that it is the two-carrier transport mechanism that yields the non-saturating MR in figure 4(a). We have also used the following two-band model (equation (1)) to fit the nonlinear $R_{yx}(B)$ curves in figure 5(b) [30, 31]

$$\rho_{yx} = \frac{(n_p \mu_p - n_e \mu_e)}{e(n_p \mu_p + n_e \mu_e)^2} B^2 (n_p - n_e)$$

(1)

$$\rho_{xx} = \frac{(n_p \mu_p + n_e \mu_e) + (n_p \mu_p \mu_e + n_p \mu_p \mu_e \mu_p \mu_e)}{e(n_p \mu_p + n_e \mu_e)^2 B^2 (n_p - n_e)^2} B^2$$

(2)

with the constraint condition $n_e = (\sigma_0 - e n_p \mu_p)/(e \mu_e)$. $n_p$ ($n_e$) and $\mu_p$ ($\mu_e$) are the density and mobility of holes (electrons), respectively. $\sigma_0$ is the longitudinal conductivity measured at $B = 0$. Therefore, there are in fact only three independent fitting parameters $n_p$, $\mu_p$, and $\mu_e$. The obtained fitting parameters at 170 K are $n_p = 4.3 \times 10^{17}$ cm$^{-3}$, $\mu_p = 21900$ cm$^2$ V$^{-1}$ s$^{-1}$, $n_e = 3.7 \times 10^{20}$ cm$^{-3}$, and $\mu_e = 24.5$ cm$^2$ V$^{-1}$ s$^{-1}$, respectively. Compared with the high-density electrons, the low-density holes have much higher mobility. This enormous difference in the mobility also indicate the different nature of the carriers. Due to the tiny effective mass of the Dirac-like hole and its possible topological protection from back scattering [5], it is expected to have a much larger mobility than the trivial electrons. One can also see that, even in the two-carrier transport regime, our sample is away from the perfect electron–hole compensation. That is the reason why relatively small MR is obtained in our ZrTe$_5$ samples, compared with the XMR in WTe$_2$ and TaAs$_2$ [30, 31].

In figure 5(b), the MR at 170 K can be also well described by equation (2) of the two-band model. Due to the more complex nature of MR than the Hall effect, the fitting of $R_{xx}(B)$ was done with different parameters. The best fitting yields the following parameters: $n_p = 6.0 \times 10^{17}$ cm$^{-3}$, $\mu_p = 15220$ cm$^2$ V$^{-1}$ s$^{-1}$, $n_e = 3.6 \times 10^{20}$ cm$^{-3}$, and $\mu_e = 32.1$ cm$^2$ V$^{-1}$ s$^{-1}$, close to the parameters extracted from the fitting of $R_{yx}(B)$, thus suggesting the validity of the two-band model in interpreting our results.

Note that the coexistence of holes and electrons in ZrTe$_5$ has been also revealed in previous ARPES or transport studies of ZrTe$_5$ [18–21]. Briefly speaking, there is a Fermi pocket at the $\Gamma$ point and another trivial electron pocket along the YM edge in the first Brillouin zone of ZrTe$_5$ [18]. At low temperatures, only electrons...
from both two pockets contribute to the transport in ZrTe$_5$. But as the temperature increases above $T_p$ ($=138$ K), a Lifshitz transition occurs, leading to the change of the Fermi pocket at the $\Gamma$ point from electrons to holes $[18–21]$. This scenario explains the emergence of high-mobility holes above 140 K ($\sim T_p$), as discussed in figure 5. Therefore, our MR and Hall results in figures 4 and 5 can be regarded as a direct consequence of the Lifshitz transition in ZrTe$_5$. But it is worth pointing out that besides the Lifshitz transition, the electron pocket along the YM edge is also essential to the occurrence of the abnormal enhancement of MR shown in figure 4. Without this pocket, the two-carrier transport mechanism cannot be realized in ZrTe$_5$.

Although the two-carrier transport under perfect electron–hole compensation can give rise to XMR in some materials, such as WTe$_2$ and TaAs$_2$, this is usually achieved only at very low temperatures $[30, 31]$. For example, the MR of TaAs$_2$ at 9 T can be as high as 1200 000% at 2 K, but it will dramatically drop to only about 12% at 300 K $[31]$. For practical application of XMR in spintronics, one must find a way to realize the perfect electron–hole compensation condition at room temperature. As enlightened by our results shown in figure 4, if we can use a gate to tune the Fermi level (or the electron and hole densities) of our sample, we may possibly shift the temperature region where the large enhancement of MR is observed towards higher temperatures, which would be important for potential application of ZrTe$_5$ in magnetic sensors.

We also like to point out that although data similar to figures 4(a) and 5(a) have been also reported in the supplementary information of $[15]$, the purpose of that paper is to extract the carrier concentrations and mobilities at different temperatures from the two-band model analysis. But in this work, we especially focus on the abnormal enhancement of MR at high temperatures due to the Lifshitz transition and discuss its implication for potential spintronics application. Therefore, different from the previous study $[15]$, a new perspective on the MR and Hall data of ZrTe$_5$ is provided.

In conclusion, the measured SdH oscillations clearly demonstrate the existence of a non-trivial band with tiny effective mass in ZrTe$_5$. The vanishing of the LL splitting at large tilting angles further reveals the dominant role of anisotropic orbital splitting, instead of the isotropic Zeeman term. Besides these, the Lifshitz transition induced two-carrier transport, as indicated by the nonlinear Hall effect, gives rise to an abnormal enhancement of MR at high temperatures, which might be enlightening in potential application of ZrTe$_5$ in spintronics.
Acknowledgments

This work was supported by the National Natural Science Foundation of China (No. 11574129 and 11874194), the National Key Research and Development Program of China (No. 2016YFA0301703), and Technology and Innovation Commission of Shenzhen Municipality (No. KQJSCX20170727090712763 and KQTD2016022619565991). L Z and R A contributed equally to this paper.

ORCID iDs

G P Guo @ https://orcid.org/0000-0002-2179-9507

References

[1] Armitage N P, Mele E J and Vishwanath A 2018 Rev. Mod. Phys. 90 015001
[2] Liu Z K et al 2014 Science 343 864
[3] Xiong J, Kushwaha S K, Liang T, Krizan J W, Hirschberger M, Wang W D, Cava R J and Ong N P 2015 Science 350 413
[4] Neupane M et al 2014 Nat. Commun. 5 3786
[5] Liang T, Gibson Q, Ali M N, Liu M H, Cava R J and Ong N P 2015 Nat. Mater. 14 280
[6] Li C Z, Wang L, Li H W, Wang J, Liao Z M and Yu D P 2015 Nat. Commun. 6 10137
[7] Li H, He H T, Lu H Z, Zhang H C, Liu H C, Ma R, Fan Z Y, Shen S Q and Wang J N 2016 Nat. Commun. 7 10301
[8] Vasconcelos T C, Ramos J G G S and Barbosa A L R 2016 Phys. Rev. B 93 115120
[9] Ramos J G G S, Vasconcelos T C and Barbosa A L R 2018 J. Appl. Phys. 123 034304
[10] Niu J, Wang J Y, He Z J, Zhang C L, Li X Q, Cai T C, Ma X M, Jia S, Yu D P and Wu X S 2017 Phys. Rev. B 95 035420
[11] Lu J W et al 2017 Phys. Rev. B 95 125135
[12] Li Q, Kharzeev D E, Zhang C, Huang Y, Pletikosic I, Fedorov A V, Schneeloch J A, Gu G D and Valla T 2016 Nat. Phys. 12 550
[13] Zheng G L et al 2016 Phys. Rev. B 93 115414
[14] Liang T et al 2018 Nat. Phys. 14 451
[15] Liu Y W et al 2016 Nat. Commun. 7 12516
[16] Tang F D et al 2019 Nature 569 537
[17] Weng H M, Dai X and Fang Z 2014 Phys. Rev. X 4 011002
[18] Zhang Y J et al 2017 Nat. Commun. 8 15512
[19] Xu B, Zhao I X, Marsik P, Sheveleva E, Lyzwa F, Dai Y M, Chen G F, Qiu X G and Bernhard C 2018 Phys. Rev. Lett. 121 187401
[20] Xiong H et al 2017 Phys. Rev. B 95 195119
[21] Chi H, Zhang C, Gu G D, Kharzeev D E, Dai X and Li Q 2017 New J. Phys. 19 015005
[22] Murakawa H, Bahramy M S, Tokunaga M, Kohama Y, Bell C, Kaneko Y, Nagaosa N, Hwang H Y and Tokura Y 2013 Science 342 1490
[23] Wang C, Lu H Z and Shen S Q 2016 Phys. Rev. Lett. 117 077201
[24] Wang Z J, Sun Y, Chen Q, Franchini C, Xu G, Weng H M, Dai X and Fang Z 2012 Phys. Rev. B 85 195320
[25] Jeon S, Zhou B B, Gynis A, Feldman B E, Kimchi I, Potter A C, Gibson Q D, Cava R J, Vishwanath A and Yazdani A 2014 Nat. Mater. 13 851
[26] Cao J Z et al 2015 Nat. Commun. 6 7779
[27] Wu X J, Li T X, Zhang C and Du R R 2015 Appl. Phys. Lett. 106 012106
[28] Abriskov A A 1998 Phys. Rev. B 58 2788
[29] Parish M M and Littlewood P B 2003 Nature 426 162
[30] Ali M N et al 2014 Nature 514 205
[31] Wu D S et al 2016 Appl. Phys. Lett. 108 042105