Spin zero and large Landé g-factor in WTe₂

Ran Bi¹, Zili Feng², Xinqi Li³, Jingjing Niu¹, Jingyue Wang¹, Youguo Shi², Dapeng Yu³,⁴,⁵ and Xiaosong Wu⁴,⁵

¹ State Key Laboratory for Artificial Microstructure and Mesoscopic Physics, Peking University, Beijing 100871, People’s Republic of China
² Institute of Physics and Beijing National Laboratory for Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100190, People’s Republic of China
³ State Key Laboratory for Artificial Microstructure and Mesoscopic Physics, Beijing Key Laboratory of Quantum Devices, Peking University, Beijing 100871, People’s Republic of China
⁴ Collaborative Innovation Center of Quantum Matter, Beijing 100871, People’s Republic of China
⁵ Department of Physics, Southern University of Science and Technology of China, Shenzhen 518055, People’s Republic of China

E-mail: ygshi@iphy.ac.cn and xswu@pku.edu.cn

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Abstract

We study the c axis transport of WTe₂. An enhancement of quantum oscillations allows for observation of an astonishing spin zero effect at certain field orientations for both hole bands and electron bands, providing transport evidence for their spin–orbit splitting origin. Based on the requirements for the spin zero effect, a lower limit for the Landé g-factor, as large as 44.7, has been obtained at a field angle of 63.7° with respect to the c axis towards the b axis for the hole band. The field dependence of the band splitting is experimentally determined.

1. Introduction

Observation of an extremely large and unsaturated magnetoresistance (MR) in WTe₂ has stimulated considerable theoretical and experimental activities [1–15]. Though the carrier compensation has been mostly accepted as the main mechanism for the MR behavior [1–5, 15], the role of the spin–orbit coupling (SOC) has been postulated [8, 9, 13, 14]. Angle resolved photoemission spectroscopy (ARPES) has revealed an anisotropic spin texture due to SOC, which, combined with anisotropic MR, suggests the importance of SOC in MR [9–11]. Moreover, recent spin–resolved ARPES works reveal the spin–polarized Fermi pockets, which provides vital information to understand the large and nonsaturating MR in WTe₂ [12, 13]. However, the detailed mechanism underlying the MR has yet to be determined. Recently, theories have predicted WTe₂ to be a type-II Weyl semimetal in three-dimensional bulk form and a quantum spin Hall insulator in monolayer of 1T’ structure, which has further fueled the interest in this material [16–23].

As SOC not only plays an important role in MR, but also is one of the key ingredients for the above topological phases, information on this coupling and its effect can provide a critical insight into these phenomena [24, 25]. Since the calculated electronic structure is extremely sensitive to the atomic coordinates and electronic correlations in WTe₂, experiments are particularly valuable in this regards [9, 17, 26]. Extensive ARPES and quantum oscillation studies have been carried out and four pairs of energy bands along the X–Γ–X direction have been identified, namely two pairs of hole bands and two pairs of electron bands, in agreement with first-principles calculations [3, 6, 26, 27]. According to calculations, the twofold degeneracy of the bands is lifted by SOC. Although the resultant spin texture has been detected by spin–resolved ARPES, its transport signature has not been reported [9–11]. Moreover, the field dependence of the electronic structure, especially relevant in MR, is less studied. The Landé g-factor is unknown.

Previous quantum oscillation studies have focused on the angular dependence of the oscillation frequencies, by which the topography of the Fermi surfaces has been inferred [6, 8]. No information on the spin could be extracted. In this work, we report the first c axis transport study of WTe₂. A strong enhancement of quantum oscillations in this configuration enables us to observe the angular dependence of the oscillation amplitude,
which exhibits an astonishing spin zero effect. The fact that all electron/hole bands display a spin zero at the same angle, offers transport evidence that they are non-degenerated spin–orbit split bands, confirming the ARPES experiments \[9, 12, 13\]. For particular field directions, a sizable splitting of 12.3 meV estimated from data places a lower limit on the Landé g-factor, 44.7. The magnetic field effect on the band splitting is manifested by the field dependence of the oscillation frequencies.

2. Method

Single crystals of WTe$_2$ were grown by a solid state reaction method. The details on the growth and the in-plane transport properties of our samples can be found elsewhere \[6, 7\]. Crystals with a relatively large dimension in the $\hat{c}$ axis were selected for this study. To achieve a good electrical contact, 300 nm Au was deposited on freshly cleaved (001) surfaces of the crystal by magnetron sputtering. Then, the ribbonlike crystal was cut into one with typical dimensions of $0.6 \times 0.13 \times 0.1$ mm$^3$. Electrical contacts were made by silver paste on two Au coated surfaces. A four-probe method was used for the transport measurement. Low temperature transport measurements were carried out using a standard lock-in method in an OXFORD variable temperature cryostat.

3. Results and discussion

When the current is in-plane (along either $\hat{a}$ or $\hat{b}$ axis) and the magnetic field is parallel to the $\hat{c}$ axis, WTe$_2$ displays extremely large MR, seen in figure 1(a). MR, defined as $[\rho(B)-\rho(0)]/\rho(0)$, reaches $\approx 2.1 \times 10^4\%$ at 10 T, consistent with previous studies \[1, 10\], which indicates the high quality of our sample \[6\]. Shubnikov de Hass oscillations (SdHOs) are evident above 5 T. When the magnetic field is parallel to the $\hat{b}$ axis, seen in the upper panel of figure 1(c), the magnitude of SdHOs is strongly suppressed and the onset for SdHOs is shifted to a higher field. Similar trend can also be seen in other studies \[6, 10\]. The suppression is even more severe when the field is parallel to the $\hat{a}$ axis, shown in the lower panel of figure 1(c). The oscillations are hardly discernible when the current $I \parallel \hat{a}$. Such a suppression can be understood by the presumed conductivity anisotropy, i.e., $\sigma_{\hat{a}} > \sigma_{\hat{b}} > \sigma_{\hat{c}}$, inferred by the layer structure and the in-plane W chain of WTe$_2$ \[1\]. The formation of Landau levels will be hindered when the cyclotron orbit involves motion along the low conductivity axis. Consequently, a higher field is required to observe SdHOs in these field directions. Note that a high filed can modify the energy band of WTe$_2$, as indicated by an earlier study and discussed later in this work \[8\]. When obtaining the topography of the Fermi surface by SdHOs, one should keep in mind that it may be different from the topography in zero field. Thus, it would be helpful to have the oscillations starting at a low field so that the field dependence of the topography can be mapped.

MR measurements have also been carried out when the current is out-of-plane. The signal-to-noise ratio is improved due to a larger resistivity. The test structure for the $\hat{c}$ axis transport is sketched in figure 1(b). To ensure a current uniformly distributed in the sample, a thick Au layer is used to spread the current. For the same reason, a pseudo four-point measurement is employed, although the configuration compromises the advantage of a true four-point measurement in excluding the contact resistance between Au and the sample. Fortunately, it only leads to an underestimation of the relative SdHOs amplitude. Moreover, a true four-point setup has been employed to measure other samples and no statistically significant difference in resistivity has been found between two setups, suggesting a negligible contact resistance (see the supplementary materials available online at stacks.iop.org/NJP/20/063026/mmedia). The short separation between the electrodes also gives rise to a geometric MR, which, similar to the effect of the contact resistance, increases the background MR, hence reduces the relative oscillation amplitude. The reduction is monotonically enhanced when the field is tilted towards to the $\hat{a}$ or $\hat{b}$ axis (perpendicular to the current). It will not introduce a non-monotonic change of the oscillation amplitude, one of the results in this study, which will be discussed later. The SdHOs in the $\hat{c}$ axis transport are plotted for two in-plane field directions in figure 1(c). Intriguingly, MR in both cases exhibits much stronger SdHOs compared with the above in-plane transport. The onset field is as low as 5 T, similar to that with the field parallel to the $\hat{c}$ axis (figure 1(a)). This result is rather surprising if one realizes that the formation of the Landau levels should not depend on the direction of the probe current, but the direction of the field, which is normal to the cyclotron orbit plane. The origin of the effect is not clear. Nevertheless, the strong SdHOs in the $\hat{c}$ axis transport offer a substantially better resolution and a lower field range in study of the Fermi surface topography. From now on, we focus on the $\hat{c}$ axis transport, unless otherwise specified.

Figure 2(a) shows the temperature dependence of the $\hat{c}$ axis resistivity at 0 and 9 T with the field parallel to the $\hat{b}$ axis. The zero field resistivity at room temperature and 1.5 K is $\rho(300 \text{ K}) = 94 \mu\Omega\text{m}$ and $\rho(1.5 \text{ K}) = 12 \mu\Omega\text{m}$, respectively. It yields a residual resistivity ratio (RRR) $\rho(300 \text{ K})/\rho(1.5 \text{ K}) = 7.83$, which is significantly smaller than that of the in-plane resistivity, $\text{RRR} = 5.91 \mu\Omega\text{m}/0.024 \mu\Omega\text{m} \approx 246.3$. The
transverse MR (current perpendicular to field) displays the same turn-on behavior as in the in-plane transport [1, 11]. However, its size at low temperature is only about 1000%, almost two orders of magnitude smaller. The difference is in agreement with the positive correlation found between the transverse MR and RRR [6].

We now turn to the angular dependence of MR. Figure 2(b) shows MR up to 14 T for different tilt angles of the magnetic field. The field is within the $b$–$c$ plane and the angle $\theta$ is with respect to the $c$ axis. As the angle increases, MR is enhanced, which we attribute to the orbital contribution due to the Lorentz force. The most interesting feature is the amplitude of SdHOs. It changes with the angle and almost vanishes at $\theta = 63.7^\circ$. When the field rotates in the $a$–$c$ plane, similar damping of SdHOs has also been observed, except the angle is different. Observation of the strong suppression of amplitude at certain angles benefits from the enhanced oscillations in our out-of-plane transport. We have noticed that a similar feature has appeared in the in-plane transport of an earlier study, too, in which the oscillation signal is enhanced by measuring the thermoelectric effect (see figure 3 of the supplementary materials in reference 6).

To see this feature more clearly, the oscillatory part of MR is obtained by subtracting a polynomial background and plotted in figures 3(a) and (b), for $B$ within the $b$–$c$ plane and the $a$–$c$ plane, respectively. There is obviously a minimum of oscillations. Fast Fourier transformation (FFT) has been performed and the amplitude spectra are illustrated in figures 3(c) and (d). The spectra are consistent with other reports [4, 6, 28]. Four peaks in the low frequency range, denoted as $P^1$–$P^4$ from low to high frequency, are ascribed to four fundamental frequencies corresponding to two pairs of electron ($P^2$ and $P^3$) and hole ($P^1$ and $P^4$) pockets. These high frequencies are results of magnetic breakdown. For field along the $\hat{c}$ axis, $P^1 = 93.4$ T, $P^2 = 126.4$ T, $P^3 = 143.9$ T, $P^4 = 163.4$ T. The frequencies of the four peaks gradually increase as the magnetic field is tilted away from $\hat{c}$ toward the $b$ or the $\hat{a}$ axis, which is in agreement with the observations of previous reports [6, 8]. The detail of the angular dependence of the frequency is shown in supplementary materials.
A close-up look at the evolution of the four frequencies with the angle confirms the damping of SdHOs for certain field orientations. It also shows that they damp differently. In particular, for field in the \( b-c \) plane, \( P^1 \) and \( P^4 \) (holes) disappear at \( \theta = 63.7^\circ \) and \( 90^\circ \), while for field in the \( a-c \) plane, \( P^2 \) and \( P^3 \) (electrons) disappear at a slightly different angle, between \( 64.4^\circ \) and \( 75.1^\circ \). This phenomenon is better illustrated in figures 3(e) and (f), in which the amplitudes of \( P^1-P^4 \) are plotted against the angle. For field in the \( b-c \) plane, \( P^1 \) and \( P^4 \) fall to near zero at \( 63.7^\circ \) and \( 90^\circ \), while \( P^2 \) and \( P^3 \) remain finite. In contrast, for field in the \( a-c \) plane, \( P^2 \) and \( P^3 \) reach zero between \( 64.4^\circ \) and \( 75.1^\circ \), while \( P^1 \) and \( P^4 \) show a finite minimum at around \( 75.1^\circ \). The angular dependence of \( P^1 \) and \( P^4 \) displays a striking similarity, which suggests that two hole bands are closely related. The same conclusion can be drawn for \( P^2 \) and \( P^3 \), too.

Disappearance of SdHOs can occur in layered materials. In these quasi-two-dimensional systems, carriers are largely confined in each layer. Consequently, the Fermi surface is a warped cylinder along the interlayer direction [29–31]. When the field is in-plane, Landau levels cannot be formed due to incoherent interlayer transport. But, this is obviously not the case here, because the Fermi surface of \( \text{WTe}_2 \) consists of ellipsoids with mild anisotropy, which result from coherent electrons moving from layer to layer, as shown in [6, 26] and our data. The transport is essentially three-dimensional [11, 26]. In addition, the fact that SdHOs remain when the field is in-plane, \( \theta = 90^\circ \), rules out this scenario.

According to the Lifschitz–Kosevich theory for SdHOs, the fundamental of oscillation can be expressed as

\[
\frac{\Delta \rho}{\rho_0} = AR_f R_T R_s \cos \left( 2\pi \frac{B_i}{B} - \gamma \right),
\]

where \( A \) is the overall amplitude, \( B_i \) is the oscillation frequency, \( \gamma \) is a phase factor, \( R_f, R_T, R_s \) are amplitude reduction factors due to temperature, scattering and spin splitting [32]. The higher harmonics are negligible in our study (see the supplementary materials). At low temperature, \( R_f \approx 1 \). \( R_T = \exp(-2\pi^2 k_B T_D m^*/\hbar B) \) depends on the effective mass \( m^* \), where \( T_D \) is the Dingle temperature. So, a large \( m^* \) can suppress the oscillations. But, no significant enhancement of \( m^* \) has been observed around the angles at which the oscillations vanish (see figure S4 in the supplementary materials). In addition, the Fermi surfaces around this angle obtained
by the oscillation frequencies are also quite smooth, inconsistent with a severe deformation expected by the enhancement of $m^*$. Other experiments have also shown that the deviation of the Fermi surface from an ellipsoid is small [6]. Finally, it is most likely that $m^*$ in the $c$ axis is, or at least close to, the largest due to the layer structure of WTe$_2$. The fact that the oscillation amplitudes of $P_2$ and $P_3$ peak when $B \parallel \hat{b}$ strongly suggests that $R_D$ is not the dominant factor determining the angular dependence.

We believe that the explanation lies in the spin factor. Spin splitting of Landau levels produces two oscillation frequencies, which interfere with each other. The interference modulates the oscillation amplitude, described by

$$R_s = \cos \left( \frac{\Delta E}{h \omega_c} \pi \right)$$

where $\Delta E$ is the size of the splitting, $h$ is the reduced Plank constant and $\omega_c$ is the angular frequency of the cyclotron, which is linear in $B$. If $\Delta E$ is not linear in $B$, $R_s$ will be field dependent, leading to a beating pattern in oscillations. Analysis of such beating pattern has been widely used to extract the spin–orbit splitting [33–35]. On the other hand, for the Zeeman splitting, $\Delta E_z = g_{\mu_B} B_z$, where $g$ is the Landé factor and $\mu_B$ is the Bohr magneton. Thus, $R_s = \cos \left( \frac{\pi gm^*}{2m_e} \right)$ becomes independent of $B$. When $gm^*/m_e = 2n + 1$, where $n$ is an integer, the oscillations disappear at arbitrary field, called spin zero [32]. Although the condition can be met in two-dimensional systems by tuning the ratio between the out-of-plane field responsible for cyclotron and the total field responsible for the Zeeman effect, this knob is not applicable in three dimensions. Therefore, the spin zero is not commonly seen in three dimensions. Still, we have observed it in WTe$_2$. This can be understood by considering a large $g$-factor, which helps to meet the condition. We shall discuss the $g$-factor shortly.

Since the stringent condition for the spin zero depends only on the band parameters, $g$ and $m^*$, and is essentially accidental, the fact that $P^2$ and $P^4$ annihilate concomitantly, as well as $P^2$ and $P^3$, can hardly be a coincidence. The only plausible explanation is that $P^2$ and $P^4$ are a pair of non-degenerated spin–orbit split bands, so are $P^3$ and $P^1$. The same angular dependence of the amplitude shown in figures 3(e) and (f) further confirms it. Our results provide solid transport evidence for the spin–orbit split nature of the bulk bands.

The size of the splitting has been predicted to be around 15 meV for holes on average [9]. To estimate the size from our data, we obtain the Fermi wave vectors along the $a$ axis, $k_{14}^i = 0.46, 0.49, 0.51, 0.57$ nm$^{-1}$, from the oscillation frequencies considering ellipsoidal Fermi surfaces. The effective masses for the electron and hole bands can be calculated from the temperature reduction factor $R_T$, $m_{lh}^* = 0.44, 0.36, 0.40, 0.49 m_e$ (see the

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**Figure 3.** Evolution of quantum oscillations with the field orientation. (a), (b) Oscillation component as a function of $1/B$ as $B$ is tilted from $c$ to $\hat{b}$ and from $c$ to $a$, respectively. (c), (d) FFT spectra of (a), (b). (e), (f) The angular dependence of the FFT amplitudes obtained from (c), (d) for four fundamental frequencies, $P^2$–$P^4$, from top to bottom.
supplementary materials). Assuming simple parabolic bands, the Fermi energy along the \( \hat{a} \) axis for four bands can be estimated, \( E_{\text{F}} \approx 17.2, 24.0, 26.5, 26.4 \) meV. This yields 9.21 and 2.48 meV for the hole and electron band splittings, respectively. These values are consistent with first principle calculations and ARPES results \([6, 9]\).

It is worth noting from equation (2) that a field independent spin zero occurs only if the size of the splitting is linearly proportional to \( B \). In a magnetic field, the splitting is affected by an interplay of the spin–orbit splitting and Zeeman effect and depends on the detail of the band structure \([36–39]\). For example, in 2D electron gas with Rashba SOC, \( \Delta E = \sqrt{(\hbar \omega - g^* \mu_B B)^2 + \Delta_{\text{Ze}}^2 - \hbar \omega}, \) where \( \Delta_{\text{Ze}} \) is the spin splitting in zero field \([33, 34, 40]\). Also in Dirac semi-metal Cd$_3$As$_2$, the spin-degenerate conduction bands are lifted by the combination of orbital and Zeeman splitting \([38, 39]\). Given that the zero field spin splitting is already over 10 meV for the hole bands in WTe$_2$, a very large \( g \)-factor is required so that the Zeeman splitting, linear in \( B \), can be dominant. Taking a splitting \( \Delta E = 12.3 \) meV estimated from the oscillation frequencies for the holes and assuming \( \Delta E_{\text{h}} > 2 \Delta E \) in order to be dominant in the range \( 5 < B < 14 \) T, we determine a lower limit for the \( g \)-factor in the direction when the spin zero takes place, \( g > 44.7 \). A similar calculation yields \( g > 10.1 \) for the electron bands when the spin zero occurs in the \( \hat{c} \)-\( \hat{a} \) plane.

This large \( g \)-factor leads to a marked magnetic field dependence of the splitting, which can be seen in the FFT spectra. Thanks to the enhanced SdHOs in the \( \hat{c} \) axis transport, such observation can be made down to a much lower field. As seen in the upper panel of figure 1(c), when \( \vec{B} \parallel \hat{b} \), the splitting induced beating pattern is clear. The number of periods between adjacent beating nodes increases from 10 to 12 with increasing field, indicating a decrease of the splitting. To quantitatively illustrate the effect, a moving FFT has been performed on the oscillations from low to high fields. The results are shown in figure 4. With increasing field, \( P^2 \) and \( P^3 \) get closer. From their frequencies, the splitting energy is obtained and plotted as a function of the field. It starts from 5.1 meV at 7.1 T and drops to 3.0 meV at 10.3 T. The large change of the splitting is consistent with a large \( g \)-factor. Although it has been proposed that the change of the spin texture with magnetic field plays an important role in the extremely large MR of WTe$_2$, there have been few experimental investigations on such a field dependence. Our study has experimentally determined the field dependence of the spin splitting and the \( g \)-factor, which provide basic information for understanding the field dependence of spin-related properties of this material.

4. Conclusions

In summary, we performed a study of angle-resolved quantum oscillations of the \( \hat{c} \) axis transport in WTe$_2$. The angular dependence of the oscillation amplitude exhibits a rare spin zero effect. The effect takes place at the same angle for two pairs of electron/hole bands, confirming their spin–orbit splitting origin. By analysis of the
requirements for the spin zero effect, we have determined a sizable Zeeman energy, yielding a large Landé g-factor no less than 44.7 in certain directions. The field dependence of the band splitting has been determined by the oscillation frequency as a function of the field. Our results may shed light on MR and other spin-related phenomena in this material.

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ORCID iDs

Xiaosong Wu @ https://orcid.org/0000-0001-9224-9871

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