# Quantum oscillations in type-II Dirac semimetal PtTe<sub>2</sub>

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Type-II Dirac semimetal (DSM) phase, in which Lorentz invariance is broken, has been experimentally realized in  $PtSe_2$  class. We study the transport properties and Fermi surface (FS) morphology of  $PtTe_2$  by de Haas–van Alphen (dHvA) quantum oscillations. Quantum oscillations analyses for out-of-plane and in-plane magnetizations revealed multiple frequencies with low effective mass and high quantum mobility. First-principles calculations are performed to confirm the existence of type-II Dirac fermions in  $PtTe_2$  and distinguish the contributing pockets for each oscillation mode. Among all the observed frequencies, one around 4510 T is identified to be derived from the electron pocket that participates in the formation of the type-II Dirac cone.

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### I. INTRODUCTION

Though Dirac and Weyl fermions were initially proposed in high-energy physics [1], Weyl fermions have never been discovered experimentally as elementary particles. In recent years, their counterpart (low-energy excitations) in condensed matter physics have been implemented, which has ignited explosive research on three-dimensional (3D) topological semimetals [2-8]. 3D DSMs were first realized in Na<sub>3</sub>Bi [9,10]and  $Cd_3$ -As<sub>2</sub> [11,12] with quadruple degenerate linear band crossings (Dirac points) appearing near the Fermi level. If the inversion symmetry or time-reversal symmetry is broken, a spin-degenerate Dirac fermion will evolve into two Weyl fermions with opposite chirality [2,3,13]. 3D Weyl semimetals (WSMs) were realized in the TaAs family [2,3] and feature open Fermi arcs as topological surface states. Due to the linear dispersion near the crossing, Dirac and Weyl fermions can be described by massless Dirac and Weyl equations, respectively. In 3D DSMs or WSMs, the existence of Dirac or Weyl fermion in the vicinity of the Fermi level gives rise to a variety of exotic physical properties, such as negative magnetoresistance induced by chiral anomaly [14,15] and quantum oscillations arising from Fermi arcs [16,17].

Lorentz invariance is a strict restriction in high-energy physics, however it is not necessary in condensed matter physics. The type-II WSMs that break the Lorentz invariance are predicted and experimentally confirmed in WTe<sub>2</sub> [18,19], MoTe<sub>2</sub> [20,21], Mo<sub>x</sub>W<sub>1-x</sub>Te<sub>2</sub> [22,23], and LaAlGe [24]. Different from the type-I WSMs with pointlike FS, type-II WSMs have finite FS, forming heavily tilted Weyl cones at the points where electron and hole pockets contact each other [18]. Because of its unique band structure, type-II WSMs have many unique properties that differ from the type-I WSMs: field-selective chiral anomaly [25], intrinsic anomalous Hall effect [26], and momentum space Klein tunneling [27]. Recently, another kind of topological material that violates the Lorentz invariance, the type-II DSMs, has attracted a great deal of research [28–31]. The PtSe<sub>2</sub> [28] and VA1<sub>3</sub> [32] family are predicted to be type-II DSMs by theoretical calculation. Tilted Dirac cones along the  $k_z$  direction were verified by ARPES experiments in most of the members of the PtSe<sub>2</sub> class [29,30,33]. In addition, a topological nontrivial quantum oscillation mode is observed in PdTe<sub>2</sub> [31], which may be related to the Dirac fermions. However, the transport properties of PtTe<sub>2</sub> are not reported yet, and the relevant research urgently needs to be expanded.

Here we study the magnetic transport of  $PtTe_2$  by dHvA quantum oscillations. Multiple frequencies are extracted from the analysis of isothermal magnetization in low temperature and high field. We obtained all important transport parameters such as Dingle temperature and quantum mobility by multiband LK fit. First-principles calculations were conducted to identify the contributing pockets of oscillations frequencies and assist the understanding of band structure of  $PtTe_2$ .

#### **II. EXPERIMENTAL DETAILS**

A high quality single crystal of  $PtTe_2$  was grown via self-flux methods. The mixture of Pt and Te with the molar ratio 1:49 was sealed in a quartz tube, which was sealed under vacuum. The quartz tube was heated in a furnace at 800 °C for 48 h to melt and homogenize the starting materials. The furnace was then slowly cooled to 495 °C after 2 weeks to crystallize  $PtTe_2$  in Te flux. A single crystal with regular shapes of  $PtTe_2$  was obtained via centrifugation after 3 days. The isothermal magnetization measurements were performed with a vibrating sample magnetometer (VSM) oven option in 14 T DynaCool (Quantum Design). The magnetotransport

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FIG. 1. Crystal structure and characterization of  $PtTe_2$ . (a) and (b) Side and top views of the crystal structure of  $PtTe_2$ . Green balls are Te atoms, and red balls are Pt atoms. (c) X-ray diffraction pattern of  $PtTe_2$  measured at room temperature. A image of the few-millimeter-size single crystal is shown in the inset, and the directions of *a* and *b* are labeled on the sample. (d) Raman spectrum of  $PtTe_2$  measured at room temperature. (e) Temperature dependent resistivity of  $PtTe_2$ . (f) The magnetic-field dependent resistivity up to 14 T at 2 K, with a magnetic field applied along the *c*-axis direction. The inset shows magnetic field dependence of the Hall resistivity.

measurements, including resistance and Hall effect measurements, were performed using a standard Hall-bar geometry in DynaCool.

#### **III. RESULTS AND DISCUSSIONS**

PtTe<sub>2</sub> has a centrosymmetric CdI<sub>2</sub>-type structure that satisfies the space group P3m1 and point group  $D_{3d}(-3m)$ . The adjacent Te atomic layers are connected by weak van der Waals interactions, which make PtTe<sub>2</sub> a quasi-two-dimensional layered material. The side and top views of the PtTe<sub>2</sub> crystal structure are shown in Figs. 1(a) and 1(b). Figure 1(c) shows the x-ray diffraction pattern of the PtTe<sub>2</sub> single crystal. Only the sharp (00n) peaks are observed and no impurity peaks appear, which imply the high quality of the single crystal. The inset of Fig. 1(c) is a typical PtTe<sub>2</sub> single crystal with several-millimeter size. The Raman spectrum in Fig. 1(d) shows the  $E_g$  and  $A_{1g}$  modes at 110 and 157 cm<sup>-1</sup>, respectively. The  $E_g$  and  $A_{1g}$  modes correspond to the in-plane oscillations and out-of-plane vibration of the Te atoms, respectively, which are typical features of the 1 T structure [34]. Figure 1(e) shows the temperature-dependent longitudinal resistivity of PtTe<sub>2</sub>, and the resistivity decreases with decreasing temperature, exhibiting a metallic behavior with a residual resistance ratio about 28. Figure 1(f) is the field dependent resistivity of the sample measured at 2 K, which shows a classic parabolic behavior. The inset is the Hall resistivity of the sample at 2 K and its nonlinearity implies the multiband transport with contributions from different pockets near the FS.

In order to understand the electronic structure of  $PtTe_2$ and to explore the origin of its quantum oscillation modes, we have performed first-principles band structure calculations. Bulk Brillouin zone (BZ) is shown in Fig. 2(a) with the highsymmetry points, lines, and Dirac points (*D*) being indicated. Figures 2(d) and 2(e) is the band dispersion with SOC along the high-symmetry lines (the in-plane *S*-*D*-*T* and out-of-plane *A*-*D*- $\Gamma$  directions). There are remarkable band crossings in the vicinity of *D* along both the *S*-*D*-*T* and *A*-*D*- $\Gamma$  lines below the Fermi level. Because of belonging to different irreducible representations (2D  $\Delta_4$  and 1D  $\Delta_{5+6}$ , respectively), the two valence bands intersect each other without opening a gap [28]. It is noteworthy that the Dirac cone inclines strongly along  $k_z$ direction ( $\Gamma$ -*A*) but keeps upright along  $k_x$ - $k_y$  plane (*S*-*D*-*T*), which is the typical characteristic of a type-II Dirac cone. According to our calculations, the Dirac points reside at 0.82 eV below the Fermi energy at the *D* point (0, 0, ±0.37)  $2\pi/c$ , where *c* is the lattice constant along *z* direction.

We have calculated the 3D FS of bulk PtTe<sub>2</sub> to compare the calculations and quantum oscillation modes. Figures 2(b) and 2(c) are the top view and side view of the 3D FS, respectively. The FS consists of one apple-core-like hole pocket  $\alpha$  in the center of BZ, one flowerlike electron pocket  $\beta_1$  encompassing the hole pocket, six double-triangular-pyramid-like electron pockets  $\gamma$  located around the K points of the first BZ, and a dozen of fingerlike electron pockets  $\beta_2$ . The electron pockets  $\gamma$  are wrapped by the flowerlike electron pocket, so they cannot be observed in Fig. 2(b). Both the hole and electron pockets have threefold rotation and inversion symmetry, which is consistent with the symmetry of the crystal structure. The apple-core-like hole pocket  $\alpha$  and flowerlike electron pocket  $\beta_1$  have been observed by ARPES experiments [29]. Since the pockets are sensitive to the position of Fermi level, with the Fermi level shifting from the Dirac point to the current location as indicated in Figs. 2(d) and 2(e), the initial electron pocket participating in the forming of the type-II Dirac cone evolves into the current hole pocket  $\alpha$  and the initial hole pocket



FIG. 2. (a) BZ of bulk with the high-symmetry points being indicated. 3D Dirac point is highlighted by D. (b) the top and (c) side views of the FS in BZ. The aqua green and purple one correspond to electron pockets, and the blue one is hole pocket. (d) The band structure along high symmetry lines in-plane S-D-T direction. (e) The band structure along high symmetry lines out-of-plane A-D- $\Gamma$  direction.

disappears. Meanwhile, the other electron pockets in Figs. 2(b) and 2(c) arise. Therefore, in Figs. 2(b) and 2(c) only the current hole pocket contributes to the forming of a type-II Dirac cone, while the other electron pockets do not.

Quantum oscillation is an important experimental tool to study the electronic properties of materials. As shown in Figs. 3(a) and 3(d), we measured the isothermal magnetization of PtTe<sub>2</sub> up to 14 T in B//c and B//ab orientations at various temperatures. Both the out-of-plane (B//c) and in-plane (B//ab) isothermal magnetizations curves exhibit remarkable dHvA quantum oscillations at low temperature starting from 2.1 and 3.5 T, respectively, indicating a threedimensional character of the FS. The different background for out-of-plane and in-plane magnetization can be ascribed to the difference of sample holders used for measurements. After subtracting a smooth paramagnetic (B//c) or diamagnetic (B//ab) background from the measured curves, the periodic oscillations become more clear in 1/B up to 25 K as shown in Figs. 3(b) and 3(e). Strong oscillations with the amplitudes of  $\sim$ 15 emu/mol at T = 2 K are clearly observed for B//c which is comparable to that of ZrSiH (H = Se and Te) [35]. In the geometry of B/(ab), PtTe<sub>2</sub> show weaker oscillations amplitude as compared to B//c. The oscillations pattern for B//c are likely to involve two close frequencies or one dominant frequency, while the quantum oscillations pattern for B//abseems to involve multiple frequencies, which confirms the conspicuous anisotropy of the FS.

The number and magnitude of all the frequencies involved in quantum oscillation can be derived from its fast Fourier transform (FFT) analysis as shown in Figs. 3(c) and 3(f). For the case of B//c, we get two close and strong frequencies F1 = 102.7 T, F2 = 107.8 T, and a weak frequency F3 = 243.2 T from the FFT spectra. The observation of beat (F1 and F2) means this section is not centered at a high symmetry point in momentum space [36]. F1 and F2 can also be observed in another sample (see the Supplemental Material [37]). The frequency around 207.2 T should be a second harmonic peak of F1. These frequencies observed in FFT spectra are closely related with the orthogonal extremal cross-sectional area ( $A_F$ ) of the Fermi pockets by the Onsager relation  $F = (\Phi_0/2\pi^2)A_F$ , where  $\Phi_0 = h/2e$  is the magnetic flux quantum. According to the formula,  $A_F$  corresponding to F1, F2, and F3 are  $A_{F1} = 98.0 \times 10^{-4} \text{ Å}^{-2}$ ,  $A_{F2} = 102.9 \times 10^{-4} \text{ Å}^{-2}$ , and  $A_{F3} = 232.2 \times 10^{-4} \text{ Å}^{-2}$ , occupying 0.35%, 0.37%, and 0.82% of the cross-sectional area of the first BZ in the *ab* plane, respectively. For the case of B//ab, we get F4 = 148.9 T, F5 = 206.2 T, F6 = 225.3 T, and  $F7 \approx 4510 \text{ T}$  from the FFT analysis. A slightly different background subtraction will not affect the frequency and amplitude of ultrahigh frequency F7 (see the Supplemental Material [37]).  $A_F$  corresponding to F4, F5, F6, and F7 are  $A_{F4} = 142.1 \times 10^{-4} \text{ Å}^{-2}$ ,  $A_{F5} = 196.8 \times 10^{-4} \text{ Å}^{-2}$ ,  $A_{F6} = 215.1 \times 10^{-4} \text{ Å}^{-2}$ , and  $A_{F7} = 4305 \times 10^{-4} \text{ Å}^{-2}$ , occupying 0.57%, 0.78%, 0.86%, and 17% of the cross-sectional area of the first BZ in the *bc* plane, respectively. The observation of multiple frequencies in the in-plane quantum oscillations proves the complexity of the band structure of PtTe<sub>2</sub>, consistent with the first-principles calculations shown above.

To determine the contributing pocket of each quantum oscillation mode, we calculate possible extremal cross-sectional area of electron and hole pockets perpendicular to the direction of the magnetic field. According to our calculation, the frequencies F1 = 102.7 T, F2 = 107.8 T for B//c correspond to the maximum cross section of fingerlike electron pockets  $\beta_2$ , while F3 = 243.2 T comes from the electron pocket  $\gamma$  located around the *K* point. For the case of B//ab, F4 = 148.9 T can be ascribed to the electron pocket  $\gamma$ , while F5 = 206.2 T and F6 = 225.3 T come from the branches of the flowerlike electron pocket  $\beta_1$ . What is more, the highest frequency  $F7 \approx$ 4510 T originates from the hole pocket  $\alpha$ , which is related to the formation of the type-II Dirac cone.

The dHvA quantum oscillation of 3D Dirac system can be described by Lifshitz-Kosevich (LK) formula [38,39], which takes the Berry phase into account [40]:

$$\Delta M \propto -B^{0.5} R_T R_D R_S \sin\left[2\pi \left(\frac{F}{B} - \gamma - \delta\right)\right], \quad (1)$$

where  $R_T = (\alpha T m^*/B)/[\sinh(\alpha T m^*/B)]$ ,  $R_D = \exp(-\alpha T_D m^*/B)$ , and  $R_S = \cos(\pi g m^*/2m_e)$ .  $m^*$  and  $m_e$  is the effective cyclotron mass and free electron mass, respectively.  $T_D$  is the Dingle temperature and  $\alpha = 2\pi^2 k_B/e\hbar$  is a constant.  $R_T$  and  $R_D$  derive from the broadening in Landau levels induced by temperature effect in Fermi-Dirac



FIG. 3. (a) and (d) Isothermal out-of-plane magnetization and in-plane  $(B//[10\bar{1}0])$  magnetization for PtTe<sub>2</sub> at various temperatures. (b) and (e) The oscillatory components of the (a) out-of-plane and (d) in-plane magnetization for PtTe<sub>2</sub>. The inset of (e) shows the enlarged high field data of in-plane oscillatory components. (c) and (f) The FFT spectra of the (b) out-of-plane and (e) in-plane oscillatory components at various temperatures. For (c), the FFT spectra are acquired under field interval  $\Delta H = 1-14$  T. The inset shows the FFT spectra of the same oscillatory components under field interval  $\Delta H = 2.22-14$  T. For (f), The FFT spectra are acquired under field interval  $\Delta H = 3-14$  T. The inset shows the FFT spectra of the same oscillatory components under field interval  $\Delta H = 10-14$  T. An ultrahigh frequency around 4500 T is clearly observed.

distribution and electron scattering, respectively.  $R_T$  describes the temperature dependence of the oscillation amplitude, while  $R_D$  captures the field dependent damping of the oscillation amplitude. What is more, the periodic oscillation of  $\Delta M$  is depicted by the sine function term with a phase factor  $-\gamma - \delta$ , where  $\gamma = \frac{1}{2} - \frac{\phi_B}{2\pi}$  and  $\phi_B$  is the Berry phase. The dimensionality of FS determines the phase shift  $\delta$ , specifically  $\delta = 0$  and  $\pm 1/8$ , for the 2D and 3D cases, respectively.

Through the fit of the temperature dependence of the FFT amplitude to the thermal damping factor  $R_T$ , we obtain the effective mass of different frequencies as shown in Figs. 4(a) and 4(c). The effective mass for all the observed frequencies are in the range of 0.151 to 0.907  $m_e$ , most of which (except  $m^*$  for F7) are comparable to that of another kind of type-II DSM PdTe<sub>2</sub> [31]. The average  $m^*$  for B//c is lower than that for B//ab, which originates from the anisotropy of the band structure. For quantum oscillation involving multiple frequencies, it is difficult to get the Dingle temperature of different frequencies by drawing the Dingle plot. However, the Dingle temperature and quantum mobility for every frequency except F7 can be extracted by fitting the oscillation patterns at a given temperature (T = 2 K) to the multiband LK formula for the 3D case [35]. Multiband LK formula can be considered as

the linear superposition of LK formula for a single band, and this approach has been proved to be valid for extracting the transport parameters from quantum oscillations of multiband topological semimetals in previous work [31,35]. As shown in Figs. 4(b) and 4(d), the  $\Delta M$  for both B//c and B//ab can be precisely reproduced by multiband LK formula and all the parameters determined form the fit are summarized in Table I,

TABLE I. The derived parameters from dHvA oscillations for PtTe<sub>2</sub>. *F* is the oscillation frequency,  $A_F$  is the extremal cross-sectional area of the pockets,  $T_D$  is the Dingle temperature, m\* (in terms of  $m_e$ ) is the effective mass,  $\tau$  is quantum relaxation time, and  $\mu_q$  is quantum mobility.

	$F(\mathbf{T})$	$A_F \; (10^{-4} \text{\AA}^{-2})$	$T_D(\mathbf{K})$	$m^*/m_e$	$\tau$ (ps)	$\mu_q \ (\mathrm{cm}^2/\mathrm{Vs})$
<i>B</i> // <i>c</i>	102.7	98.0	4.3	0.196	0.28	9642
	107.8	102.9	9.5	0.151	0.13	6007
	243.2	232.2	4.2	0.289	0.29	3754
B//ab	148.9	142.1	6.8	0.255	0.18	1238
	206.2	196.8	2.8	0.318	0.44	2443
	225.3	215.1	4.2	0.337	0.29	1523
	4510	4305		0.907		



FIG. 4. (a) and (c) Temperature dependence of out-of-plane (B//c) and in-plane (B//ab) FFT amplitude; the solid lines represent the LK fit for effective mass. The results of the fit are shown in (a) and (c). (b) and (d) The multiband LK fit (red line) of the out-of-plane and in-plane oscillation patterns (black line) at T = 2 K.

in which quantum relaxation time and quantum mobility are obtained by  $\tau = \hbar/2\pi k_{\rm B}T_D$  and  $\mu = e\tau/m^*$ , respectively. The Dingle temperature is determined to be from 2.8 to 9.5 K, and corresponding quantum relaxation time is from 0.13 to 0.44 ps. Furthermore, the quantum mobility for B//c is estimated to be from 3754 to 9642 cm<sup>2</sup>/V s, while that for B//ab is calculated to be from 1238 to  $2443 \text{ cm}^2/\text{V}$  s. Obviously the quantum mobility for in-plane cyclotron motions (B//c) is much larger than that for out-of-plane cyclotron motions (B//ab). The layered structure of PtTe<sub>2</sub> may explain such anisotropic quantum mobility. The higher scattering rate along the interlayer direction is likely responsible for the suppression of quantum mobility for B//ab. The difference in the amplitude of the dHvA oscillations between the in-plane and out-of-plane directions may be induced by the difference of the quantum mobility and the effective mass between the two directions. Furthermore, it is worth mentioning that the in-plane cyclotron motion quantum mobility for PtTe<sub>2</sub> is significantly higher than that for PdTe<sub>2</sub>. The low effective mass and high quantum mobility hint at the superior transport properties of type-II Dirac semimetal PtTe<sub>2</sub>.

The angle dependence of dHvA provides further insight into the shape of the FS. The FFT spectra of the dHvA oscillations with field rotated in the *ab* plane are presented in Fig. 5(a). The FFT peaks can be generally divided into two groups, one group around 130 T, and another group around 210 T. Split peaks and small peaks around F5 might be due to strong spin-orbit coupling or complicated FS of flowerlike electron pocket  $\beta_1$  [41]. The angles dependent positions of two main FFT peaks are summarized in Fig. 5(b). Both F4 and F5 evolve with the direction of magnetic field with a period of  $60^{\circ}$  in the *ab* plane, which is consistent with threefold rotation symmetry of the FS. When the angle  $\theta$  deviates from 0°, F4 increase gradually, and reach maximum value around 148.9 T at  $\theta = 30^{\circ}$ . When  $\theta$ changes from 30° to 60°, F4 will approach minimum value again at  $\theta = 60^{\circ}$  around 124 T. This can be explained by the double-triangular-pyramid-like electron pocket  $\gamma$ . We present the FFT spectra of the same oscillatory components under field interval  $\Delta H = 10-14$  T in Fig. 5(c). The highest frequency appears only when the magnetic field is along or near  $\Gamma$ -M direction in the BZ. The strange property of the highest frequency may arise from the angular dependence of the minimum orthogonal cross-sectional area for the hole pocket  $\alpha$ , which take the lowest value when the magnetic field is along  $\Gamma$ -M direction. If the magnetic field deviates from  $\Gamma$ -M direction, the minimum orthogonal cross-sectional area will increase, which make it more difficult to detect the ultrahigh frequency above 4000 T. Therefore, the hole pocket  $\alpha$  in the center of BZ shows strong anisotropy. The FS measured by dHvA is consistent with the theoretical calculation, providing indirect evidence for the existence of tilted Dirac cones in PtTe<sub>2</sub>.

## **IV. CONCLUSIONS**

In summary, we study the magnetic transport properties and band structure of type-II DSM PtTe<sub>2</sub> by dHvA quantum



FIG. 5. (a) FFT spectra of the dHvA quantum oscillations with field rotated in the *ab* plane. Inset in (a) shows the definition of field orientation angle  $\theta$ . The data collected at different  $\theta$  have been shifted vertically for clarity. (b) The angular dependencies of the main oscillation frequencies obtained from (a). (c) The FFT spectra of the same oscillatory components under field interval  $\Delta H = 10-14$  T.

oscillations. Strong oscillations are observed for both the outof-plane and in-plane isothermal magnetization in low temperature. Multiple frequencies are extracted from the oscillations and analyzed based on the Lifshitz-Kosevich (LK) formula. The low effective mass for most frequencies is comparable to that of another kind of type-II Dirac semimetal PdTe<sub>2</sub>, while the average mobility for most frequencies is significantly higher than that of PdTe<sub>2</sub> [31]. Angle dependent in-plane dHvA oscillations provide further insights into FS properties. In addition, the first-principles calculations are conducted to help us verify the topological nature and identify the contributing pocket of each frequency. Especially the ultrahigh frequency around 4510 T is considered to result from the electron pocket that contributes to the formation of the type-II Dirac cone.

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