Normal-state negative longitudinal magnetoresistance and Dirac-cone-like dispersion in PtPb$_4$ single crystals: a potential Weyl-semimetal superconductor candidate

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Abstract
Magnetotransport measurements of PtPb$_4$ single crystals with the $T_c$ of $\sim 2.80$ K reveal pronounced exotic topological nature. An anomalous negative longitudinal magnetoresistance (MR) related to the chiral anomaly and a linear-like transverse MR examined within the framework of quantum MR theory were studied. Results indicate the presence of chiral-anomaly-induced quantum transport associated with the Weyl states and show a Dirac-cone-like band dispersion in PtPb$_4$. This work reveals the drastic impact of the concept that the surface electrons in a Weyl fermion state may dominate the normal-state magnetotransport in PtPb$_4$, leading to the conclusion that PtPb$_4$ can be a Weyl-semimetal superconductor candidate.

1. Introduction
Topological superconductors [1–4], newly emerging topological materials, have attracted much attention in recent years due to their potential applications in quantum computations [4] and the realization of novel excitations such as Majorana fermions in condensed-matter systems, whereas they are the condensed-matter analogues of low-energy quasiparticles in relativistic high-energy physics [5]. So far, topological materials have attracted significant attention due to their unconventional properties, such as the presence of linear-band-dispersion surface/edge states, absence of backscattering, topological Fermi arc surface states, and many other novel quantum behaviors [1–3]. They reveal different characteristics of band structure, and thus can be categorized into several groups: the topological insulators, Weyl semimetals, and Dirac semimetals, etc. By introducing a superconducting gap in topological materials, being the so-called topological superconductors, the superconducting state will inherit the nontrivial topology of their surface electronic structure, giving rise to unconventional properties such as nodal phases and two-dimensional superconductivity observed [1]. Among the topological materials, the Weyl semimetal is strikingly different from other classes of materials with nontrivial band topology because of the presence of surface Fermi arcs [2, 3]. It is known that a Dirac point may split into Weyl points when inversion or time reversal symmetry is broken, and the Dirac semimetal becomes a Weyl semimetal, where the Weyl points (nodes) appear in pairs with the opposite chirality. A pair of Weyl points is protected by Chern numbers $\pm 1$ of the constant energy surfaces enclosing either Weyl node, leading to a surface Fermi arc, which may not exist in the Dirac semimetal. Therefore by introducing a superconducting gap in Weyl semimetals, being the so-called Weyl superconductor, it is expected that Weyl superconductors will host exotic phenomena, such as nonzero-momentum pairing due to their chiral node structure, or zero-energy Majorana arcs at the surface [6, 7]. Weyl superconductors could serve as a platform for realizing Majorana zero modes and topological quantum computation [4, 8]. Possible superconducting states in Weyl semimetals due to the doping or proximity to a superconductor have also been studied theoretically [9–11]. Recently, the conventional A15-structure superconductors were shown have nontrivial band topology in the bulk electronic band.
structures, leading to the formation of topological surface states near the Fermi energy [12]. Therefore, the realization of topological superconductivity in a single material of conventional superconductors is advantageous considering the possible complexity at the interfaces of heterostructures, in which the interface superconductivity should be induced by the superconducting proximity effect [2, 13]. A recent study, presented by Lee et al [14] showed that another conventional superconductor PtPb4, which is closely related to the topological PtSn4 semimetal, reveals significant Rashba splitting as well as a linear-like surface band dispersion into their angle-resolved photoemission spectroscopy (ARPES) data. D1c-type structure of PtPb4, revealing superconductivity with $T_c$ of 2.8 K, as reported in the 1960s [15, 16], is isostructural with the Dirac nodal arc semimetals PtSn4 and PdSn4 [16]. However, only few studies on the transport properties of PtPb4 have so far been made regarding the exotic topological properties and superconductivity of PtPb4. Novel state in these traditional low-$T_c$ superconductors seems to have stimulated new light on the study of nontrivial topological superconductivity, in addition to a simple realization of topological superconductors.

On the other hand, the recent discovery of Weyl fermions in three-dimensional condensed matter systems, such as TaAs [17, 18], Cd3As2 [19, 20], Na3Bi [21], ZrTe5 [22, 23], TaP [24], CdPtBi [25, 26], $\beta$-Ag2Se [27], NbP and NbAs [28], has attracted considerable interest on the quantum dynamics of relativistic field theory in condensed matter physics. Another unique characteristic feature is the so-called chiral anomaly associated with Weyl fermions. This phenomenon is referred to as the Adler–Bell–Jackiw anomaly, indicating that the chiral current is not conserved [29]. This effect has been suggested to present as an anomalous ‘negative’ longitudinal magnetoresistance (NLMR) in topological materials when the applied electric field is parallel to the magnetic field [17–28]. So far, these topological materials also show a common magnetotransport characteristic in which a weak antilocalization (WAL) effect is observed at low applied electric field parallel to the magnetic field [17–28].

2. Experiments

PtPb4 single crystals were grown from Pb flux with a starting composition of Pt:Pb = 0.08:0.92. The mixtures of Pt powder and Pb powder were sealed under vacuum in a quartz tube. The quartz ampoule was heated to 600 °C for 24 h, cooled to 330 °C for 6 h and then slowly cooled down to 290 °C at a rate of 0.5 °C h−1. The remaining Pb flux on the surface of crystals was washed by 5% HNO3 aqueous solution and distilled water. Several platelet-like crystals with a typical size of 1.5 × 1.0 × 0.1 mm3 were obtained. The phase purity and the crystal structure of obtained crystals were characterized by powder x-ray diffraction (Bruker D2 phaser) measurements with Cu Kα radiation on single crystals. For electrical transport measurements, the cleaved shiny crystals were cut into dimensions of ~1.5 × 0.5 × 0.05 mm3. Five leads were soldered with indium, and a Hall-measurement geometry was formed to allow simultaneous measurements of both longitudinal ($\rho_{xx}$) and transverse (Hall) resistivities ($\rho_{xy}$) using the standard dc four-probe technique. In materials with high mobility, negative NLMR can arise from current jetting [28, 35]. Thus, for a homogeneous current injection, current contacts were soldered, covering the complete cross-section of the samples. Hall voltages were taken in opposing fields parallel to the c-axis up to 6 T and at a dc current density of ~50 A cm−2. The magnetization was measured in a superconducting quantum interference device system (MPMS from quantum design).

3. Results and discussion

Figure 1(a) is a typical picture of the PtPb4 cleaved single crystals. The crystals are ductile and exhibit a silvery luster, but they cannot be mechanically exfoliated to be a few-layered thick like graphene. The PtPb4 crystals are stable in the air or diluted HNO3 aqueous solution. The crystal structure in real space is schematically shown in figure 1(b). The crystallographic c axis is anticipated to be perpendicular to the
indicating that the [001] direction is perpendicular to the plane of the crystals. The lattice constant of the ρ shows the temperature dependence of resistivity for S1 in zero fields at temperatures near smaller FC signal, compared with the ZFC signal, is caused by vortex pinning. The inset of figure 2(b) crystalline specimen, in which only the (00\( n \)) \(( n = 1, 2, 3, \) and 4) diffraction peaks were observed, indicating that the [001] direction is perpendicular to the plane of the crystals. The lattice constant of the c axis was determined precisely to be 5.971 Å and is very close to 5.966 Å as reported earlier [36].

Figure 2(a) shows the entire temperature range of resistivity \( \rho_{xx} \) for a typical sample S1. The normal-state resistivity for PtPb\(_4\) reveals a metallic-like character \(( \partial \rho_{xx} / \partial T > 0 )\), with a residual resistivity \( \rho_0 \) of \( \sim 0.709 \mu \Omega\) cm and a residual resistivity ratio (RRR) of 113, where the RRR value is determined by \( \rho_{xx}(300 \text{ K})/\rho_{xx}(3 \text{ K}) \). The low residual resistivity and large RRR indicate the exceptional quality of samples. The upper inset of figure 2(a) shows the temperature dependence of resistivity \( \rho_{xx}(T) \) in zero fields at low temperatures. As seen in the lower inset of figure 2(a), the low-temperature resistivity data can fit in the temperature range of 3.0 K to 40 K by using the formula \( \rho_{xx}(T) = \rho_0 + AT^2 \), and the electron-electron scattering coefficient \( A = 5.6 \times 10^{-3} \mu \Omega\) cm K\(^{-2} \) was obtained. The obtained \( A \) for PtPb\(_4\) is much larger than those of 2–7 \( \times 10^{-4} \mu \Omega\) cm K\(^{-2} \) reported for PtSn\(_4\) [37] and PdSn\(_4\) [38, 39], respectively. The larger electron–electron scattering coefficient indicates that PtPb\(_4\) should behave more characteristic of correlated semimetals. Figure 2(b) shows the zero-field-cooling (ZFC) and field-cooling (FC) magnetizations in field \( H = 5 \text{ Oe} \) parallel to the crystal c axis for a PtPb\(_4\) single crystal, demonstrating the superconducting transition temperature \( T_c \approx 2.80 \text{ K} \), which is consistent with a previous report of 2.8 K [15, 16]. The smaller FC signal, compared with the ZFC signal, is caused by vortex pinning. The inset of figure 2(b) shows the temperature dependence of resistivity for S1 in zero fields at temperatures near \( T_c \); a sharp resistive transition occurs at 2.80 K, which agrees with magnetization measurement. Basic electrical transports of zero-field resistivity \( \rho_{xx} \), temperature-dependent MR, Hall coefficient \( R_{HH} \), and Hall mobility characteristics of PtPb\(_4\) single crystals from different batches are presented and briefly discussed in the supplementary material, as seen in the supplementary figures S1–S4.

Figure 3(a) shows a typical main result of transverse magnetoresistance (TMR) behavior for sample S1 with the application of B fields perpendicular to the crystalline ab plane. Here, the MR is calculated by \( \text{MR}\% = | R(B) - R(0) | / R(0) \times 100\% \), where \( R(B) \) is the resistance at the applied magnetic field \( B \), and \( R(0) \) is the measured resistance at \( B = 0 \). As shown in figure 3(a), the MR increases with the increased applied fields, and a clear non-saturating MR can be seen for the entire range of temperature and field of measurement. A sharp resistance dip is clearly observed at a temperature of 5 K, and as the temperature increases, the MR dip at a low B is broadened indicating the reduction of the surface state. Furthermore, the MR becomes linear for the intermediate temperature range, and at higher temperatures, it shows a quadratic-like behavior in the low-field region. We shall return to this subject later. A close analysis of the TMR in the low-field region, as shown in figure 3(b), shows a striking anomaly of the negative TMR at the low-field region and temperatures below 4.5 K. Actually, above 10 K, the TMR is positive and can be adequately described by a quadratic field dependence in low fields. Below 6 K, there exists a critical B-field for each curve, below which a pronounced negative MR region (corresponding to a negative slope in a resistance against magnetic field plot) accompanied with a MR dip can be observed in the low fields of \( B < 0.07–0.2 \text{ T} \), deepening upon the temperature of cooling down. At \( T = 4 \text{ K} \) and \( B = 0.08 \text{ T} \), the highest negative MR of 21% can be obtained. Therefore, the curves in the negative MR region show a cusp-like feature in the region close to the zero-field point. It is also noted in figure 3(b) that besides the apparent negative MR, a small resistance dip appears close to the zero field \( B = 0 \) as \( T < 3.5 \text{ K} \). Such a dip is believed
Figure 2. (a) The entire temperature range of resistivity for sample S1. The upper inset shows $\rho_{xx}(T)$ at low temperatures. The lower inset shows $\rho_{xx}(T)$ as a function of $T^2$, and the dashed line is the fitting of $\rho_{xx}(T) = \rho_0 + AT^2$. (b) ZFC and FC magnetizations in $H = 5$ Oe parallel to the crystal $c$ axis for a PtPb$_4$ single crystal are shown. The inset shows $\rho_{xx}(T)$ of sample S1 (https://stacks.iop.org/NJP/23/093030/mmedia) in zero field at temperatures near $T_c$.

to arise from the weak anti-localization effect in Dirac semimetals [29]. The low-field negative TMR is unexpected in the non-ferromagnetic PtPb$_4$. So far, the negative TMR has been observed on the magnetic semimetal GdPtBi [25, 26] and nonmagnetic Bi nanowires [40] and has never been examined in detail. One of the main mechanisms for the negative TMR is explained by the spin-disorder scattering process in Kondo systems [41], in which the resistance is predicted to decrease as the amplitude of the spin fluctuation is suppressed by the induced magnetic moment $M$ in magnetic fields with $MR \propto -M^2$, and commonly has been observed on the ferromagnetic materials such as manganites at high fields [42]. Here the observed low-field negative TMR for non-ferromagnetic PtPb$_4$ seems to be inconsistent with this origin. Here we focused on the negative NLMR which is correlated to the so-called chiral anomaly associated with Weyl fermions as mentioned previously. Figure 3(c) shows the longitudinal MR (LMR) ($B//I$) for S1 at different temperatures, showing a bell-shaped negative LMR at low temperatures. At $T \approx 0.25$ T and 3 K, the LMR decreases to a negative minimum value of $\approx 30\%$ below the zero-field value and then increases to be positive at higher fields. The NLMR weakens and gradually narrows upon warming up and ultimately changes to positive with an MR dip in the low-field range at temperatures above 5 K, as seen in figures 3(c) and (d). Upon warming to $T > 10$ K, the low-field LMR behaves according to the classical quadratic field dependence. The observed NLMR is an important point and will be discussed in the later sections. Similar types of NLMR behavior have been reported for some topological Weyl semimetals [17–28].

In addition to the negative TMR, which needs further investigation, we attempted to explain the origin of the NLMR in PtPb$_4$ with the chiral anomaly in the semi-classical regime [29]. NLMR related to the chiral anomaly can be traced to the four-component massless Dirac equation. The above equation describes particles, known as Weyl fermions, with a definite chirality, in which the number of fermions with plus or minus chirality is conserved separately. However, the relativistic theory of charged chiral fermions in three spatial dimensions presumes that a non-conservation of chiral charge can be induced by external gauge fields with non-trivial topology, known as the chiral anomaly or the so-called Adler–Bell–Jackiw (ABJ) anomaly [43, 44]. In 1980s, a physical picture of the chiral anomaly in the context of condensed matter
Figure 3. (a) A typical result of the field-dependent TMR \((B \perp I)\) behavior for sample S1. (b) The corresponding TMR in the low-field region. (c) LMR \((B//I)\) for S1 at different temperatures, and (d) The corresponding LMR in the low-field region.

physics was provided \[45\], which predicted an enhanced longitudinal magneto-conductivity (MC) due to the pumping of charge from one node to another in presence of an electric field \(E\) parallel to \(B\). A result, the enhanced MC due to the chiral anomaly, \(\sigma_{CA}\), is described by a quadratic \(B\) dependence in the form,

\[
\sigma_{CA} = C_W(T) \cdot B^2,
\]

where the field independent constant, \(C_W(T)\), has the inverse \(T^2\) dependence,

\[
C_W(T) = \frac{e^2}{\pi \hbar} \frac{3}{8} \frac{\nu_F}{\pi} \frac{\tau_c}{T^2} + \frac{\mu_c}{e^2}.
\]

(1)

Here \(\nu_F\), \(\tau_c\), and \(\mu_c\) are the Fermi velocity, chirality changing scattering time and chemical potential, respectively \[22, 23\]. In addition to the negative quadratic MR associated with a chiral anomaly, Kim et al have also shown that the coexistence between the WAL and NLMR for topological Bi\(_1-x\)Sb\(_x\) can be attributed to the ABJ anomaly in the presence of WAL corrections \[29\], where the conductivity contribution due to WAL can be described by \(\sigma_{WAL} = a(T) \cdot \sqrt{B}\) with a negative value of \(a(T)\) for WAL transport (note that: weak localization transport has a positive \(a(T)\) value). Thus, for a complete quantitative analysis, the fitting of conductivity \(\sigma\) for LMR data can be carried out with the formula,

\[
\sigma = \sigma_0 + \sigma_{CA} + \sigma_{WAL} + \sigma_N = \sigma_0 + C_W(T) B^2 + a(T) \sqrt{B} + (\rho_0 + A_N B^2)^{-1},
\]

(2)

where \(\sigma_0\) is the residual conductivity, and \(\sigma_N\) is the small positive MR due to the conventional nonlinear band contribution around the Fermi level, which is commonly considered and has the field dependence of \(\sigma_N = (\rho_0 + A_NB^2)^{-1}\) with a temperature-dependent coefficient \(A_N\). However, in our data analyses, the contribution of the fourth term \(\sigma_N\) was very small and negligible. Thus we tried to fit our LMR data using an expression for the change of longitudinal MC,

\[
\Delta MC = \sigma - \sigma_0 = C_W(T)B^2 + a(T)\sqrt{B}.
\]

(3)

Figure 4(a) shows the field-dependent longitudinal \(\Delta MC\) for PtPb\(_4\) at low temperatures as well as the fitting curves using equation (3). As seen, the \(\Delta MC\) in the intermediate field range of \(\sim 0.2\)–\(1.0\) T can be
well described by the chiral anomaly associated with the WAL transport formula. However, in the low-field region of $B < 0.1$ T, an increase in MC, corresponding to the NLMR, is observed for PtPb$_4$ at temperatures below 4.5 K. As shown in figure 4(b) for the low-field data, the zero-field $\Delta$MC exhibits a linear dependence upon the magnetic field, $\Delta$MC $\propto$ B, implying that the MC of chiral anomaly outcompetes the WAL conductivity in the low-field region. The shape of low-field NLMR for Cd$_3$As$_2$ [20] nanowires or ZrTe$_5$ [22] was analyzed and revealed a chiral-anomaly-dominated conductivity, i.e. $\Delta$MC $\propto$ $B^2$. However, the obtained NLMR data for sample S1 in the low-field region look similar to those for GdPtBi semimetals, but regrettably the analyses of $\Delta$MC for their LMR were lacking. The phenomenon of $\Delta$MC $\propto$ B has been proposed by considering the low-dimension chiral dynamics in an ultra-quantum limit, where it is predicted that the correction term of the longitudinal MC is linearly proportional to $B$ [29]. A linear-B MC arising from the Landau degeneracy has also been obtained in some theoretical works before [46–49], based on the assumption that the potential range in an intrinsic topological Weyl semimetal is finite and the interplay of the Landau degeneracy and impurity scattering in the transport is considered. However, both the theoretical derivations are inconsistent with our data because predicted linear B-dependent MC requires the system to lie in the ultra-quantum limit. That is, one should have $\omega_T \gg 1$ ($\omega$ is the cyclotron frequency and $\tau$ is the transport life time) with a sufficiently strong field and only the lowest Landau band crosses the Fermi level, which is clearly not the case for our linear B-dependent MC observed in the small-field region of $B < 0.05$ T. Actually, the minimum magnetic field for the PtPb$_4$ S1 sample needed to enter the quantum limit is $\sim 0.35$ T as we shall see later. Another recent theoretical work has predicted a linear B-dependent MC in weak non-quantizing magnetic fields by considering the intervalley scattering in a lattice model of tilted Weyl fermions within the Boltzmann approximation [50]. It has shown that a finite tilt of Weyl cones will introduce a linear-in-$B$ term in the longitudinal MC and thus also introduce a corresponding asymmetric MC around zero magnetic field, however, being clearly not the case for the symmetric MC as presented here. The origin of $\Delta$MC $\propto$ B in low fields for PtPb$_4$ needs further investigation. Figures 4(c) and (d) respectively show the temperature dependences of the obtained $C_W$ and $a$ values, where $C_W$ and $a$ were determined from the lines of best fit shown in figure 4(a). As seen, both the $C_W$ and $-a$ values decreased with an increase in temperature. In the inset of figure 4(c), the $C_W^{-1}$ vs $T^2$ are plotted and the $C_W^{-1}$ is observed to be almost linear in $T^2$, as predicted theoretically by equation (1). Thus, the NLMR implies the chiral-anomaly-induced quantum transport associated with the existing Weyl states in PtPb$_4$. In addition, as seen in figure 4(d), parameter $a$ is negative due to the WAL effect and at higher temperatures; the decrease in $-a$ values with an increase in temperature indicates a gradual absence of the WAL effect on the field-dependent longitudinal MC. Furthermore, we observed that the $-a(T)$ values exhibit a power-law dependence of $\sim T^{-1.77}$ for sample S1 as shown in the inset of figure 4(d). The temperature dependence of coefficient $a(T)$ has never been examined in detail [17, 51]. It may safely be assumed that the power-law dependent $a(T)$ is correlated with the coherence length $\xi_0$, which describes the characteristic magnetic field in the Hikami–Larkin–Nagaoka model for the WAL effect [52, 53]. The coherence length $\xi_0$ has been theoretically predicted to vary with $T^{-0.75}$, being also power-law dependent, for three-dimension systems [52].

Focusing on the subject of the linearly field-dependent TMR as seen in figure 3(a), we attempted to explain this phenomenon within the framework of Abrikosov’s ‘quantum linear MR’ mechanism [33, 34], as introduced in the supplementary material. It has been pointed out that the Landau level (LL) splitting $\Delta_{LL}$ in a Dirac cone state scales with the square root of $B$, as described by $\Delta_{LL} = \pm v_F \sqrt{2eB}\phi$, can lead to a much larger LL splitting than that in a conventional parabolic band under the same $B$ strength, where $v_F$ is the Fermi velocity. The linear MR has been identified in many materials hosting Dirac fermions with linear energy dispersion, such as some layered compounds with two-dimensional (2D) Fermi surface like SrMnBi$_2$ [54], quasi-2D semimetal NbTe$_2$ [31], topological Fe-doped Bi$_2$Te$_3$ [32], and the iron-based family of Ba(FeAs)$_2$, FeSe and FeTe$_{0.6}$Se$_{0.4}$ single crystals [55–57]. High-field Abrikosov’s quantum MR most likely apply to the TMR behavior of our PtPb$_4$ single crystals. A crossover of TMR values from a semi-classical weak field $B^0$ dependence to a nearly linear $B$ dependence can be observed when the magnetic field is beyond critical field $B^*$. The critical field $B^*$ can be determined by the plot of differential TMR versus the field, $dTMR/dB$, as shown in figure 5(a) for the sample S1. The $dTMR/dB$ was observed to be linearly proportional to $B$ with a large positive slope in low fields but reaches a nearly constant value when the field is higher than $B^*$, which is determined by an intersection of two linear fitting lines, as shown by the guideline in figure 5(a). Here, the derivative anomalies of $dTMR/dB$ due to the negative TMR occurring at the near-zero fields and at low temperatures, as mentioned previously, are not presented. The derived $B^*$ values varying with temperatures are shown in figure 5(b), showing that $B^*$ shifts to a higher field with the increase in temperature. According to Abrikosov’s model of the quantum linear MR in layered semimetals [34], the LL splitting $\Delta_{LL}$ can be expressed with $\Delta_{LL} = E_F + k_B T$ at critical field $B^*$, leading to satisfy the regime of the quantum limit. Therefore, the $B$-linear MR originates from the Dirac cone states, and the
observed $B'(T)$ corresponds to the limit of $B'(T) = (1/2e)(E_F + k_B T)^2$. The critical field $B^*(T)$ was examined via the plot of $\sqrt{B}$ versus $T$, as shown in the inset of figure 5(b). As shown, the temperature dependence of critical field $B^*$ fitted well with the $\sqrt{B}$-T plotting at temperatures below 25 K. The fitting results yield Fermi level of the Dirac cone state $E_F \approx 3.05$ meV and Fermi velocity $v_F \approx 1.45 \times 10^5$ ms$^{-1}$ for sample S1. The obtained value of $E_F$, corresponding to the energy of temperature 38 K, is indeed larger than the temperatures for $B^*(T)$ analyzed, being accordant with the condition of quantum limit. The yielded $E_F$ and $v_F$ values are close to the previous reports in iron-based family of Ba(FeAs)$_2$ ($E_F \sim 2.48$ meV, $v_F \sim 1.88 \times 10^5$ m s$^{-1}$) [55] and FeTe$_{0.6}$Se$_{0.4}$ ($E_F \sim 5.5$ meV, $v_F \sim 1.1 \times 10^5$ m s$^{-1}$) [57]. A classic Parish and Littlewood’s model (PL model) [58] was also proposed to explain the mechanisms of the linear MR behavior. However, the observed linear TMR behavior for PtPb$_4$ may not be close to the mechanism of the PL model, as proved in the supplementary material. The main transport characteristics of our high-quality PtPb$_4$ samples are similar and have been consistently described by the presented formulas. The MR and MC characteristics of PtPb$_4$ single crystals from different batches are presented and discussed in the supplementary material. The obtained basic electrical transport parameters are also summarized therein.

As mentioned, the electronic structure of PtPb$_4$ by using ARPES and density functional theory (DFT) calculations has been reported by Lee et al [14]. They have shown a linear-like surface band dispersion close to the zone boundaries. Therefore, it seems reasonable that the observed chiral-anomaly transport is possibly associated with the WAL effect, as well as the Abrikosov’s quantum TMR of PtPb$_4$ is derived from the surface carriers in hole pockets around zone boundaries. Taking into account $\Delta_{\perp LL} = E_F + k_B T$ with $E_F = 3.05$ meV and $T = 10$ K, we have the LL splitting $\Delta_{\perp LL}(10 K) = 3.91$ meV, which corresponds to the splitting between the lowest and the first LL, $\Delta_{-1}$, at $T = 10$ K. Thus, referring to the hole-pocket surface band shape determined by ARPES [14], we demonstrate a sketch of the linear surface state dispersion for PtPb$_4$ at 10 K as seen in figure 5(c), where the LL splitting $\Delta_{-1}$ and the 2D surface $E_F$ below the top of the surface valence band is illustrated. Furthermore, we estimated the surface Dirac carrier density $n_D$ to be $\sim 3.9 \times 10^{11}$ cm$^{-2}$ based on the 2D surface $E_F = 3.05$ meV and the 2D formula $E_F = \frac{\hbar^2}{2m_e} \cdot n_D$, where $m_e$ is
the electron mass used for an approximate estimation. Looking more carefully into the ARPES and related DFT results around the X point, however, one can see that the Dirac point has to be more than 50 meV above the Fermi level (refer to figures 3(j)–(o) and figures 4(d)–(f) of reference [14] and assume the linear-like band below the Fermi level is going to generate a Dirac point), while a much smaller value of 3.05 meV was estimated from our transport results. This discrepancy can be understood with recalling the fact that ARPES probes the zero-field band structure while the value 3.05 meV was obtained for the quantum limit under field. In this case with the presence of magnetic fields, there would possibly be no topological features below Fermi level observable by ARPES in the related band structure. Besides, it should be noted that the linear dispersion deduced from the quantum linear MR mechanism may also be related to bulk Dirac cones and thus the possibility of quantum linear MR arising from the bulk Dirac cones cannot be ruled out. Further study on high-quality PtPb4 thin films will be worthwhile examining this subject more closely. On the other hand, we noted that no Shubnikov–de Haas (ShdH) oscillation was observed in the field range of linear TMR. The linear TMR without ShdH oscillation has also been presented for the bulk samples of Ba(FeAs)2, FeSe and FeTe0.6Se0.4 single crystals [55–57]. We may attribute the linear MR observed in our experiments to a smaller Dirac cone state with a small value of oscillation frequency around the zone boundaries according to the ARPES data [14], leading to the difficulty in observing the quantum oscillation. Recalling the fact that PtPb4 reveals multi-band transport as seen in the supplementary material, the ShdH oscillations can arise from the parabolic Fermi surface with larger energy scale.

Figure 5. (a) The plot of differential TMR, dTMR/dB versus field for sample S1 at T < 50 K. The critical field $B^*$ is determined by an intersection of two linear fitting lines, as shown by the guidelines. (b) The temperature dependence of obtained critical field $B^*$ for S1. The $B^*$–T plot is divided by $B^*(T)$, showing the crossover from a semi-classical weak field $B^2$ dependence (shaded in pale green) to a linear and non-saturating MR transport (shaded in pale yellow). The inset shows the plots of $\sqrt{B^*}$ versus T, where the dashed line represents the linear fitting of $\sqrt{B^*} \propto T$. (c) A sketch of the surface state dispersion $E$–$k$ for sample S1 at 10 K, where the solid line represents the suggested surface Fermi level $E_F$ below the top of the surface valence band, and the dashed line indicates the splitting between the lowest and the first LL, $\Delta_{-1}$, at $T = 10$ K. The sketch is plotted by referring to the hole-pocket surface band shape determined by ARPES [14].
ΔLL = ±vF \sqrt{2\hbar eB}. This can be the next attractive topic for studying quantum behaviors of the Dirac cone states in PtPb4, especially in thin-film samples.

4. Conclusion

In conclusion, the normal-state magnetotransport properties of PtPb4 single crystals with the Tc of ~2.80 K were investigated to explore the exotic topological nature of the PtPb4 superconductor. An unexpected negative TMR, as well as an anomalous NLMR related to the chiral anomaly in low fields, were observed for PtPb4 at temperatures below 5 K. The corresponding longitudinal MC in the intermediate fields could be described by the chiral anomaly associated with the WAL transport formula, implying the association of chiral-anomaly-induced quantum transport with the existing Weyl states in PtPb4. Here both the chiral anomaly and WAL effect showed a gradual absence with an increase in temperatures. Moreover, a linear-like TMR in high fields was examined within the framework of Abrikosov’s quantum MR theory and show a Dirac-cone-like dispersion in PtPb4 at temperatures below 25 K. The critical field B∗(T) could satisfy the regime of quantum limit and be examined via the plotting of \sqrt{B} versus T, giving the Fermi level of Dirac cone state E_F ≈ 3.05 meV and Fermi velocity vF ≈ 1.45 × 10^5 m s^-1 for sample S1. Thus, according to the ARPES data presented by Lee et al, a schematic band structure with a possible surface Dirac-cone state for PtPb4 at 10 K is illustrated with the LL splitting and Fermi level in the valence band. The results strongly support the concept that the surface electrons in a Weyl fermion state dominate the normal-state magnetotransport in PtPb4 single crystals, suggesting that PtPb4 can be a Weyl-semimetal superconductor candidate. Besides, the possibility of quantum linear MR arising from the bulk Dirac cones cannot be ruled out. Further study on high-quality PtPb4 thin films will be worthwhile examining this subject more closely, being an attractive topic for studying quantum behaviors of the Dirac cone states in PtPb4. Considering the convenient realization of topological superconductivity in a single material, the transport properties of PtPb4 have interesting consequences not only due to its fundamental importance but also because of the possibility of application in the formation of Majorana fermions, paving the way for developing quantum computations.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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