Field-induced Lifshitz transition in the magnetic Weyl semimetal candidate PrAlSi

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Lifshitz transition (LT) refers to an abrupt change in the electronic structure and Fermi surface and is associated to a variety of emergent quantum phenomena. Amongst the LTs observed in known materials, the field-induced LT has been rare and its origin remains elusive. To understand the origin of field-induced LT, it is important to extend the material basis beyond the usual setting of heavy fermion metals. Here, we report on a field-induced LT in PrAlSi, a magnetic Weyl semimetal candidate with localized 4f electrons, through a study of magnetotransport up to 55 T. The quantum oscillation analysis reveals that across a threshold field $B^* \approx 14.5$ T the oscillation frequency ($F_1 = 43$ T) is replaced by two new frequencies ($F_2 = 62$ T and $F_3 = 103$ T). Strikingly, the LT occurs well below the quantum limit, with obvious temperature-dependent oscillation frequency and field-dependent cyclotron mass. Our work not only enriches the rare examples of field-induced LTs but also paves the way for further investigation of the interplay among topology, magnetism, and electronic correlation.

**INTRODUCTION**

The Lifshitz transition (LT) has received renewed attention in condensed matter physics. An LT in an electronic topological transition of the Fermi surface (FS) driven by the variation of the band structure and/or the Fermi energy. Since such a transition does not necessarily require simultaneous symmetry breaking, and meanwhile, it can occur at $T = 0$, and be tuned by parameters other than temperature (such as pressure, strain, doping, magnetic field, etc.)1,2, it, therefore, can be deemed as a topological quantum phase transition. In the vicinity of Lifshitz transitions, many peculiar emergent phenomena may appear, such as van-Hove singularity, non-Fermi-liquid behavior, unconventional superconductivity, and so on (e.g. refs. 3,4).

Compared with a number of cases tuned by doping or pressure that have been widely seen in topological systems6,14,16, cuprate superconductors17,18, iron pnictides superconductors5,10, and other strongly correlated materials5,11, the examples of LT driven by a magnetic field are rare. This is because the energy scale of a laboratory magnetic field, in the order of $1–10$ meV, is much smaller than the characteristic energy scale of most metals ($\approx 10^2 – 10^3$ meV). Only in a few cases, mostly limited in heavy-fermion (HF) metals12–20, the hybridization between conduction electrons and localized f electrons leads to narrow renormalized bands with a small Fermi energy and thus the Zeeman term can be sufficient strong to shift the spin-split FS15. Recently, field-induced LTs were also observed in some low Fermi energy non-magnetic semimetals, such as bismuth31, TaP32, and TaAs33, where magnetic field beyond the quantum limit can empty a Dirac or Weyl pocket with small Fermi energy. However, in these cases, no additional Fermi pocket emerges and the carriers of the empty pocket were transferred to other pockets (previously existing).

Here we present a new example of field-induced LT beyond heavy-fermion systems in the magnetic Weyl semimetal candidate PrAlSi, by a systematic study of quantum oscillation (QO) effect with the magnetic field extending up to 55 T. We observe a single frequency ($F_1 = 43$ T) below a critical field of $B^* = 14.5$ T, in agreement with what was previously reported24. Above $B^*$, we see clearly the emergence of two new frequencies ($F_2 = 62$ T and $F_3 = 103$ T) and the disappearance of the original $F_1$. We exclude the possibility of magnetic breakdown and identify $B^*$ as a critical point where the field-induced LT occurs. By comparing the reported Fermi surface of NdAlSi and the theoretical calculation of PrAlSi, we conclude that the LT occurs in the hole-like Weyl pockets along the direction of $\Gamma–X$ of the Brillouin zone (BZ, hereafter). Our work not only enriches the rare examples of field-induced LTs but also paves the way for further investigation on the interplay among topology, magnetism, and electronic correlation.

#### RESULTS

**Experimental results and analysis**

High-quality single crystals of PrAlSi were synthesized using the flux method. The inset of Fig. 1a shows the crystal structure of

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from the measured measurements, the magnetic field was along the c-axis. At low temperatures, oscillations are visible above the Dingle mobility of 1/\(c\)PrAlSi with the magnetic moments of Pr easily orientated along the c-axis. Quantum oscillations in the magnetoresistance of PrAlSi. Fig. 1. The field-dependent of MR measured under a static field at 2 K and a pulsed-field at 1.8 K. The inset is the crystal structure(I41/md) of PrAlSi as the magnetic field easily polarizes the moments of Pr along the c-axis. b Field dependence of the oscillatory part of magnetoresistance \(\Delta \rho(B)\) in low magnetic field measured under static field, showing only a single QO frequency. The experiments were carried out with a Leiden dilution refrigerator (14 T) at 80 mK and with a refrigerator of Oxford Instrument (16 T) at \(T = 10, 17, \) and 25 K. c \(\Delta \rho(B)\) in high magnetic field measured by the pulsed magnetic field, showing more QO frequencies with a complex pattern.

PrAlSi with the magnetic moments of Pr easily orientated along the c-axis after the application of a small field\textsuperscript{24}. In our transport measurements, the magnetic field was along the c-axis and the electrical current was along the b-axis (more details are presented in the “Methods” section and Supplemental Material). Figure 1a shows magnetoresistance measured in the static field at 2 K (red curve) and pulsed field at 1.8 K (black curve). Normalized magnetoresistance \((\rho(B)−\rho(0))/\rho(0)\) reaches 115 at 55 T and remains non-saturating. The purple dashed line corresponds to \(B^2\). Figure 1b presents the oscillatory part of the longitudinal resistivity \(\Delta \rho(B)\), obtained by subtracting a smooth background from the measured \(\rho(B)\) (see Supplementary Fig. 2 for more raw data). At low temperatures, oscillations are visible above the field as low as 3 T, indicating the good quality of the sample. The rough Dingle mobility of \(1/B_1 = 0.33\, \text{T}^{-1}\) is close to the average mobility of 0.26 m\(^2\)/Vs yielded from the amplitude of the quadratic low-field magnetoresistance (see the inset of Supplementary Fig. 2a).

The single-frequency QO for static field measurements retains until temperature down to 80 mK. A more complex pattern emerges when the larger magnetic field is applied, as seen in Fig. 1c.

Figure 2a and b show the results of the fast Fourier transformation (FFT) of the oscillatory part of the magnetoresistance \(\Delta \rho\) as a function of \(1/B\). The SdH frequencies extracted from the low-field \((B < 14.5\, \text{T})\) and high-field \((B > 14.5\, \text{T})\) data show a dramatic difference. Note that, since this compound is ferromagnetic, we took into the demagnetization factor to correct the applied field in all the analyses of SdH effect (see Section 9 of the Supplemental Material). We identified \(B = 14.5\, \text{T}\) as a critical field, after checking several fields close to \(B^\ast\). As shown in Supplementary Fig. 3, we have investigated a field with different values of 10, 14, 15, and 20 T for the segmented FFT analyses. We can see the crossover from the lower QO frequency to the two higher QO frequencies as the segment changes between the 5.7 T and the selected field. Two higher QO peaks emerge when we segment the field at 20 T and we note that the low QO peaks persist because of the inclusion of the low-field QOs below 14.5 T. From the comparison of the segmented FFT analyses, 14.5 T was determined as the field of the LT. There is only one fundamental frequency in the FFT spectrum until down to 80 mK below \(B^\ast\) (see Fig. 2a). It should be noted that \(F_1\) gradually decreases with temperature, changing from 43 T at 80 mK to 32 T at 35 K, as shown in the inset. Similar temperature dependence in \(F_1\) was also reported in earlier work on PrAlSi\textsuperscript{24}, whereas the values of \(F_1\) are relatively smaller than ours. We attribute this discrepancy to the difference in stoichiometry\textsuperscript{19}. Figure 2b shows that above \(B^\ast\) there are two new QO frequencies \((F_2 = 62\, \text{T}, F_3 = 103\, \text{T})\) and their higher harmonics. Such a change in the Fermi surface is also manifested in the effective cyclotron mass \(m^\ast\). The value of \(m^\ast\) for each frequency can be deduced from the fitting of FFT amplitude according to the temperature damping factor, and this yields the small \(m^\ast = 0.08m_e \) for \(F_1\) (with field range 5.7–14.5 T), and 0.23\(m_e\) and 0.28\(m_e\) for \(F_2\) and \(F_3\) respectively, where \(m_e\) is the mass of a free electron. Interestingly, a careful look into the temperature dependence of the amplitude of the oscillatory peak leads to the
fact that the effective mass is enhanced 2-fold between 2.8 T and B*, as shown in Fig. 2c. This feature is reminiscent of HF systems displaying an LT and will be discussed more later on. The effective masses of F_2 and F_3 as the function of a field higher than B* are absent here, mainly because it is rather difficult to extract the actual amplitude of entangled peaks in the oscillatory part from two frequencies and their harmonic terms. More explanation about the effective mass mentioned above is exhibited in Supplementary Fig. 5. The Fermi energy of the band corresponding to F_1 is then estimated ε_F ≈ 125 meV.

To further demonstrate the field-induced Fermi surface change near B* ≈ 14.5 T, we performed Lifshitz–Kosevich (LK) fitting on Δρ(B) measured under the pulsed field. We notice that all the analyzed FFT data are from the pulsed fields. More details can be found in Section 4 of the Supplementary Material. As is shown in Fig. 3a, the Δρ for the field below B* can be well reproduced by the LK fitting with a single F_1 (cf. the red dot line). However, such a fitting collapse when the field exceeds B*. This problem can be fixed in an alternate fitting by employing both F_2 and F_3, seeing the blue dot line in Fig. 3b. Noteworthy that this 2-frequency LK fitting fails in the low-field window, implying that F_2 and F_3 appear only in the high-field range.

Thus, the variation of QO frequencies with the disappearance of 43 T and the emergence of 62 and 103 T clearly point to the change in Fermi surface topology, viz an LT. Firstly, we can rule out magnetic breakdown as the origin, because there is no extra QO frequency (19 T and 60 T) in the low-field range even for temperature as low as 80 mK. Moreover, the peak with the frequency of 43 T does not persist under a high magnetic field, either. Secondly, we can also exclude a metamagnetic transition as the driver of this process. Figure 4a shows the field dependence of magnetization at 2 K with the magnetic field applied along the c-axis. One clearly finds that the magnetization saturates to ~3.1μ_B/Pr at a small field 0.48 T, and no additional transition can be resolved nearby 14.5 T except for some traces of de Haas–van Alphen oscillations (inset of Fig. 4a). This is different from the case of NdAlSi, where a magnetic transition to the final Weyl-mediated helical magnetism leads to a change in QO frequency.

In order to further clarify this field-induced LT, it is helpful to estimate the density of carriers of each Fermi pocket. According to the Lifshitz–Onsager relation, F_{2} = \binom{h}{2πne}A_k, where h is Planck’s constant and A_k is an extremal cross-sectional area of the Fermi surface perpendicular to the field with Fermi wave vector k. The bands become non-degenerate due to spin–orbit coupling. Assuming these Fermi pockets are spheres, we find that the LT wipes out n_2 = 3.2 × 10^{10} cm^{-3} and produces n_3 = 5.5 × 10^{10} cm^{-3} and n_4 = 1.2 × 10^{10} cm^{-3} per four pockets. Note that the total number of pockets would be a multiple of 4, due to the symmetric requirement (see below).

The total carrier density of the hole and electron can also be extracted by fitting the Hall resistivity with the two-band model,

\[ \rho_{xy}(B) = \frac{B}{3\pi^2} \left( \frac{n_2 \mu_h^2 - n_3 \mu_e^2}{\mu_h^2 + \mu_e^2 (n_2 - n_3)} \right) + \frac{2\pi^2}{\beta} \left( \frac{n_2 \mu_h^2}{\mu_h^2 + \mu_e^2 (n_2 - n_3)} \right), \quad (1) \]

Here, n and μ represent carrier density and mobility, and the subscripts h and e denote hole and electron, respectively. We obtain n_2 = 4.3 × 10^{10} cm^{-3}, n_3 = 5.3 × 10^{10} cm^{-3}, μ_h = 0.18 m^2/V s and μ_e = 0.22 m^2/V s. These values fit both Hall resistivity and magnetic effects within the Fermi pockets.
magneto-resistivity reasonably well up to $B^*$ (see Fig. 4c and Supplementary Fig. 7). The deduced mobilities are also close to the value obtained from quantum oscillations and magnetoresistance. The average zero-field mobility ($\mu$) extracted from the residual resistivity $\rho_0 = 15 \, \mu\Omega \, \text{cm}$ is about 0.5 m$^2$/V s, slightly larger than the finite-field mobility. Such a discrepancy has been observed in other semimetals$^{37,38}$ and attributed to the field-induced mobility reduction. The carrier densities of hole and electron are within 10% of the compensation $2[(n_e n_h)/(n_e + n_h)]$, compared to ~4% in bismuth$^{39}$ and WTe$_2$.$^{40}$ This near compensation would explain the observed unsaturated magnetoresistance.

This fit, which properly works up to $B^*$ (shown by a black arrow in Fig. 4c), falls above $B^*$. The change occurring at $B^*$ is evident in Fig. 4b, which shows the first derivative of the longitudinal and Hall resistivities. The violet dotted lines demonstrate the apparent change of slope in MR and Hall resistivities at 14.5 T. We let $|n_e - n_h|$ to stay constant across $B^*$, in order to respect the Luttinger theorem$^{41}$. We infer that $F_2$ and $F_3$ should correspond to carriers of opposite signs with a density difference of $|n_e - n_h| = 6.5 \times 10^{18}$ cm$^{-3}$. Assuming that there are 8 pockets for $F_1$ ($2n_e = 6.4 \times 10^{18}$ cm$^{-3}$), would be compatible with the Luttinger theorem. Both types of carriers increase by about $0.6 \times 10^{19}$ cm$^{-3}$ ($n_e - 2n_{F_1}$ and $n_{F_2}$, respectively). By fitting the Hall resistivity curve with $n_e = 4.9 \times 10^{19}$ cm$^{-3}$ and $n_h = 5.9 \times 10^{19}$ cm$^{-3}$, we obtain $\mu_e = 0.05(1)$ m$^2$/V s and $\mu_h = 0.16(1)$ m$^2$/V s. This is shown in Fig. 4c with a blue line. The mobility ($\mu = e^\ast/\hbar$) of holes drop (~72%) more than that of electron (~27%), implying the sign of $F_1$ with a small mass should be hole-like, since its mass increases by 2.5 times by assuming the same scattering time $\tau$. We conclude that $F_2$ has an electron-like sign and $F_3$ is a hole-like one. Thus, each hole pocket ($F_1$) evolves into a larger hole pocket ($F_2$) and an additional electron pocket ($F_3$). This indicates the existence of a van-Hove singularity (saddle point) in this system.

To get more information about the Fermi surface of PrAISi, we performed the measurements of angular-dependent MR with a pulsed magnetic field at $T = 1.8$ K. Figure 5a shows the oscillatory component extracted by subtracting the smooth background from the MR measured at different $\theta$; which is defined as the angle between the c-axis and the magnetic field, and the current was along b-axis as shown in the inset (see the Supplementary Fig. 10 for the raw MR data). The SdH oscillations evolve systematically and can be observed in all angles as the magnetic field is rotated from $\theta = 0^\circ$ to $\theta = 87^\circ$. Figure 5b and c present the segmented FFT spectra for different angles $\theta$. The inset of Fig. 5b shows the critical fields at various angles, which indicates that the LT also evolves with $\theta$. We determined the $B^*$ as the same method mentioned above and the detail is shown in Supplementary Fig. 4. The oscillation frequency $F_1$ shown in Fig. 5b is weakly dependent on...
the angle, indeed indicating a nearly spherical Fermi surface of this band and the evolution of angle-dependent oscillation frequencies $F_2$, $F_3$, and their harmonic terms shown in Fig. 5c. According to the experimental results, Fig. 5d presents the angle dependence of the quantum oscillation $F_1$ which is shown as black symbols as well as $F_0$, $F_2$, $F_3$, and $F_4$ are the calculated frequencies obtained from the SKEAF program$^{42}$. The Fermi level was shifted with 3 meV in the calculation because of the uncertain doping. The $F_2$ is quite close to the observed $F_1$, whereas the higher frequencies obtained from the calculation are not observed by experiment in our case, may be due to the low mobility of these bands.

Next, we tried to locate these pockets in the BZ, and our similar calculation results (see Supplementary Fig. 11). The electronic structure also resembles that of NdAlSi$^{36}$. Since the center of the BZ is not occupied, symmetry imposes four-fold degeneracy of each pocket. According to the calculation, the schematic of the positions and the quantum limits have been reached to empty a Dirac or a reasonable assumption that they locate on the symmetry $C_E F$ splits into two non-Kramers doublets and five singlets$^{49}$. A recent analysis based on specific heat measurements revealed that the ground state is probably a doublet, while the magnetic entropy gain reaches $R \ln 3$ at about 20 K, $R \ln 4$ at about 30 K, and saturates to $R \ln 9$ at a temperature as low as $-95 K^{45}$. This suggests that at least one excited state sitting not far above the ground doublet, potentially in the order of 10 K. It is reasonable to speculate that a magnetic field of $\sim 10T$ might be sufficient to modify the CEF energy levels and the orbital characters, which possibly changes the Fermi surface topology. In addition, this field-induced evolution of CEF levels is also qualitatively consistent with the temperature-dependent QO frequency and field-dependent $m^*$ as observed experimentally. Actually, this scenario was also proposed recently for the field-induced Fermi surface reconstruction in CeRhIn$_5$.$^{50}$ To further address this possibility, more experiments like inelastic neutron scattering are needed to figure out the diagram of the CEF splitting.

In summary, we grew high-quality single crystals and observed pronounced SdH oscillations in PrAlSi with a magnetic field up to 55 T. A LT transition occurs around 14.5 T. The change in carrier densities and Fermi pockets revealed by QO and the Hall effect are consistent with each other. By comparison with theoretical calculations, we propose that LT occurs along the $\Gamma-X$ orientation and involves the Weyl pockets. One hole pocket becomes an electron pocket and a hole pocket, which indicates the existence of a van-Hove singularity. PrAlSi, therefore, represents a unique case of field-induced LT beyond the HF systems.

**METHODS**

**Sample growth and characterizations**

Single crystals of PrAlSi used in our studies were synthesized using the flux method. The starting materials are high-purity chunks of praseodymium, silicon, and aluminum, mixed into an alumina crucible. Then, the alumina crucible and quartz wool were placed in a resistance furnace and the crucible was removed by centrifuging. Powder and single crystal x-ray diffraction (XRD) have been used to obtain the XRD pattern and confirm the structure and the orientation. The atomic proportion was determined by energy-dispersive x-ray spectroscopy (EDS).

**Measurements**

The low-field magnetic transport measurements were performed on an Integra AC (Oxford Instruments) with a 16 T superconducting magnet and a Leiden dilution refrigerator with a 14 T superconducting magnet. Temperature- and field-dependent resistivity measurements were made in the standard four-probe method with a pair of the current source (Keithley 6221) and DC-Nanovoltmeter (Keithley 2182A). The high-field magnetic transport measurements were carried out under a pulsed magnetic field at Wuhan National High Magnetic Field Center (WHMFC).

Golden wires were attached using silver paste on the rectangular sample and every contact resistance was maintained to be $<2 \Omega$ in the measurements.

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AUTHOR CONTRIBUTIONS

Z.Z. and L.Z. conceived and oversaw this work. L.W., H.Z., and L.Z. performed the experiments. Z.Z., G.X., L.W., and S.C. performed band structure calculations. L.W., L.Z., Y.L., and Z.Z. wrote the manuscript with inputs from co-authors.

COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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