Shubnikov–de Haas oscillations and nontrivial topological states in Weyl semimetal candidate SmAlSi

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Abstract
The RAlX (R = Light rare earth; X = Ge, Si) compounds, as a family of magnetic Weyl semimetal, have recently attracted growing attention due to the tunability of Weyl nodes and its interactions with diverse magnetism by rare-earth atoms. Here, we report the magnetotransport evidence and electronic structure calculations on nontrivial band topology of SmAlSi, a new member of this family. At low temperatures, SmAlSi exhibits large non-saturated magnetoresistance (MR) (as large as $\sim 5500\%$ at 2 K and 48 T) and distinct Shubnikov–de Haas (SdH) oscillations. The field dependent MRs at 2 K deviate from the semiclassical ($\mu_0 H$)$^2$ variation but follow the power-law relation $\text{MR} \propto (\mu_0 H)^m$ with a crossover from $m \sim 1.52$ at low fields ($\mu_0 H < 15$ T) to $m \sim 1$ under high fields ($\mu_0 H > 18$ T), which is attributed to the existence of Weyl points and electron–hole compensated characteristics with high mobility. From the analysis of SdH oscillations, two fundamental frequencies originating from the Fermi surface pockets with non-trivial $\pi$ Berry phases and small cyclotron mass can be identified, this feature is supported by the calculated electronic band structures with two Weyl pockets near the Fermi level. Our study establishes SmAlSi as a paradigm for researching the novel topological states of RAIX family.

Keywords: topological semimetal, electrical transport, Shubnikov–de Haas oscillations, Berry phase, Hall effect

(Some figures may appear in colour only in the online journal)
1. Introduction

Weyl semimetal (WSM) as a new type of quantum state of matter hosting low energy relativistic quasiparticles, has attracted significant attention for both scientific community and potential quantum device applications [1–4]. The typical feature related to the exotic quasiparticles is the topologically protected linear crossings called Weyl nodes near Fermi surface (FS) [5, 6], which come in pairs with a definite chirality. Generally, there are two approaches to generate WSM, either by breaking space-inversion or time-reversal symmetry. The former WSM states have been well studied in various types of nonmagnetic materials with specific crystalline symmetry since its initial discovery in TaAs structural family [7, 8]. Comparatively, the latter named as magnetic WSMs have been experimentally verified only in a handful of materials, such as Mn3X (X = Sn, Ge) [9, 10], Co3Sn2S2 [11, 12], Fe3Sn2 [13] and Co2MnGa [14]. Magnetic WSMs can exhibit unique quantum transport phenomena despite the extremely large magnetic susceptibility (MR) [15] and chiral-anomaly-induced negative MR in nonmagnetic WSM [16], indeed large anomalous Hall conductivity and topological Hall effect can appear even at zero field originating from the interplay between diverse magnetism and nontrivial Weyl band topology [12, 17]. In experiment, quantum transport study is an important approach to identify the WSM state, which can extract the information associated with topological characteristics of WSM, such as nontrivial \( \pi \) Berry phase in Shubnikov–de Haas (SdH) oscillations [7, 8], large linear unsaturated MRs [15, 18], small cyclotron mass [7, 19] and high carrier mobility [3, 5]. On the other side, the above experimental evidences provide the scenario on identifying new topological WSM materials.

Recently, rare-earth based RAIX compounds (R is light rare-earth element; X is Si or Ge) with noncentrosymmetric space group \( \text{I}4_1\text{md} \) and magnetic Weyl fermions by choice of R ions [20, 21], and topological characteristics of Weyl fermions have been detected by angle-resolved photomission spectroscopy. More importantly, this family provides a rare system for comparative study on nonmagnetic WSM (\( R = \text{La} \)) and magnetic WSM (\( R = \text{Pr–Sm} \)) while keeping the same crystal structure, it also enables the tunability of conduction electron of Weyl nodes with different magnetic ground states and anisotropic magnetic behaviors by varying rare-earth ions, such as Ising-like anisotropic ferromagnetic (FM) PrAlIX [22, 23] and ferrimagnetic NdAlSi [24, 25], easy-plane anisotropy of antiferromagnetic (AFM) CeAlIX [26, 27], etc. Even quantum SdH oscillations were studied in RAIX (\( R = \text{Pr, Nd} \)) [23, 24], experimental identifications on existence of Weyl fermions based on magnetotransport study are still insufficient and under debate due to its sensitivity to R ions. The investigation on new RAIX members exhibiting distinct SdH oscillations will help to disclose the underlying Weyl physics. Among this family, SmAlSi has the smaller unit-cell compared to the previously studied light rare-earth based RAIX (\( R = \text{La–Nd} \)) series. Magnetic measurements reveal that it experiences successive magnetic transitions at low temperatures (see appendix, figure 8), providing a suitable compound for studying the topological properties in RAIX family.

In this work, we present a systematical magnetotransport study combined with the first-principle calculations on SmAlSi single crystal. High field magnetotransport reveals SmAlSi show a large unsaturated MRs (reaching \( \sim 5200\% \) at \( 2 \text{ K} \) and \( 48 \text{ T} \)) at low temperatures, accompanied by a field induced metal–insulator transitions. The field dependent MRs follow the power-law relation MR \( \propto (\mu_0 H)^m \) (\( m \leq 1.52 \)) but violate the parabolic dependence observed in compensated semimetal originating from the existence of Weyl points and electron–hole compensated characteristics. By analyzing the quantum SdH oscillations, two small FS pockets are unveiled with nontrivial Berry phase and small cyclotron mass, which is corroborated by the electronic band structure calculations.

2. Experiment

Single crystals of SmAlSi were grown by a self-flux method [22], high-purity SM (99.9%, Alfa Aesar), Si (99.99%, Alfa Aesar), and Al (99.9%, Alfa Aesar) pieces were used as starting materials. The ingredient ratio with Sm:Si:Al = 1:1:10 were weighted and sealed inside a crucible under vacuum in quartz tube. After heating at 1150 °C for 12 h, the loaded quartz tube was cooled slowly down to 800 °C, taken out from the furnace, and decanted by a centrifuge to remove excess Al flux. Large plates of SmAlSi single crystals were obtained with typical dimensions 3 mm × 2 mm × 1 mm (see the inset of figure 1(c)). To identify the crystal structure of SmAlSi, powder and single-crystal x-ray diffraction (XRD) at room temperature were characterized by using a Rigaku x-ray diffractometer with Cu Kα radiation and analyzed by Rietveld method. Magnetic properties were measured using a commercial physical property measurement system (PPMS, Quantum Design) equipped with a vibrating sample magnetometer option. Electrical transport measurements were conducted in PPMS-16 T by a standard four-probe technique using rectangle shaped samples. To eliminate the influence of longitudinal MR on Hall resistivity due to the misalignment of electrodes, the raw Hall results were measured in both field directions and anti-symmetrized by \( \rho_{xy}(H) = [\rho_{xy}(+H) - \rho_{xy}(-H)]/2 \). In addition, high field magnetotransport measurements were carried out in pulsed magnetic field up to 48 T with the pulse duration of 50 ms in Wuhan National High Magnetic Field Center.

The electronic structures were calculated by density functional theory (DFT) by including the spin–orbital coupling using the projector-augmented wave method [28] as implemented in Vienna Ab-initio Simulation Package [29]. The exchange-correlation were included using the Perdew–Burke–Ernzerhof [29, 30]. After convergence test, a 300 eV energy cut-off was used, and self-consistent cycles were proceeded on a \( 12 \times 12 \times 12 \) Monkhorst-Pack k-point mesh. The electronic structures of SmAlSi in magnetic-ordered state was adopted, in which Sm f electrons are put in the valence and a Hubbard energy U of 6.4 eV is used. The maximally localized Wannier functions for Sm f, Al s, p; and Si s, p orbitals were determined
using the Wannier90 code [31]. The electronic band structures near Fermi level (FL) and Weyl node positions were calculated using the WannierTools package [32].

3. Results and discussions

3.1. Structural analysis

SmAlSi crystallizes a noncentrosymmetric tetrahedral structure with space group $I4_1$md (No.109), as presented in figure 1(a). The refined profiles match well the experimental powder XRD data (powders were obtained by crushing single crystals) with good reliability parameters $R_p = 3.78\%$, $R_{wp} = 5.46\%$, and $\chi^2 = 1.26$ (see figure 1(b)), the structural lattice parameters and atomic coordinates are summarized in table 2 (see appendix). Notably, the refined unit-cell lattice parameters $a = b = 4.1586$ Å and $c = 14.4332$ Å of SmAlSi is the smallest among the RAlSi(Ge) family members with $R = \text{La-Sm}$ [20–26]. Single crystal XRD pattern of SmAlSi is presented in figure 1(c), which indicates that the surface of crystal is $ab$ plane and $c$-axis is perpendicular to the plate.

3.2. Transverse and longitudinal MR

Temperature ($T$) dependence of resistivity $\rho_{xx}(T)$ under current ($I$) along $a$-axis ($I // a$) and field ($\mu_0 H$) parallel to $c$-axis ($\mu_0 H // c$) of SmAlSi are shown in figure 2(a). Zero field $\rho_{xx}(T)$ shows typical metallic behavior, two resistivity anomalies are observed near the magnetic transition temperatures $T_{N1} \sim 11.5$ K and $T_{N2} \sim 5$ K (see appendix, figure 8). Compared to its sister RAIX ($R = \text{La–Nd}$) compounds [20–26], SmAlSi has relatively larger residual resistivity ratio $\rho_{xx}(300 \text{ K})/\rho_{xx}(2 \text{ K}) = 5.5$ indicative of high quality crystals. Under magnetic field, low temperature $\rho_{xx}(T)$ changes to an upturn behavior for $\mu_0 H \geq 4$ T, and high temperature $\rho_{xx}(T)$ is enhanced together with the occurrence of broad humps at $T = T_1$ and valleys at $T = T_2$ in figure 2(a). For $\mu_0 H \geq 6$ T, the humps and valleys shift to low and high temperatures, respectively. By combining the above $\rho_{xx}(T)$ and magnetic results, we plot a temperature–field ($T–\mu_0 H$) phase diagram as shown in figure 2(b). To highlight the evolution of conductivity under $\mu_0 H$, a contour plot is constructed using the field derivative of resistivity $d\rho/dT$ data. At high temperatures, a crossover from metallic ($d\rho/dT > 0$) to insulating ($d\rho/dT < 0$) behavior happens separated by $T_1$ or $T_2$. This kind metal–insulator transition can be attributed to the multiband effect generally observed in semimetals with electron–hole compensation [33–35] or to the induced excitonic gap by magnetic field [36]. Below $T_{N1}$, SmAlSi changes from metallic ($d\rho/dT > 0$) to insulating ($d\rho/dT < 0$) with a more dramatic change of resistivity compared to its paramagnetic (PM) state, signifying the effect of magnetic ordering on electron conductivity.

In figure 2(c), we present the magnetoresistance MR = $[\rho_{xx}(B) - \rho_{xx}(0)]/\rho_{xx}(0) \times 100\%$ of SmAlSi with field up to 48 T for $\mu_0 H // c$ axis. It exhibits the large non-saturated MR behaviors at low temperatures, reaching $\sim 5200\%$ under $\mu_0 H = 48$ T at 2 K. To clarify this field dependent MR behavior, a double-logarithmic plot of MR versus $\mu_0 H$ is shown in figure 2(d). At 2 K, the field dependent MR follows a power-law dependence MR $\propto (\mu_0 H)^m$ with a crossover from $m \sim 1.52$ at low field regime ($\mu_0 H < 15$ T) to linear dependence in high fields ($\mu_0 H > 18$ T). As $T$ increases to 300 K, isothermal MR follows the $(\mu_0 H)^{1.65}$ dependence. The above relationship obviously violates from the quadratic $(\mu_0 H)^2$ behavior in compensated semimetals with balanced electrons and holes, such as the report in WTe$_2$ [34] and rare-earth monopnictides [35, 37]. On the other side, linear field dependent MR can be derived from the linear dispersive band structures in Dirac/Weyl materials [15, 38], in that case, the crossover from $m \sim 1.52$ to $m = 1$ as increased fields indicate...
the dominant influence of topological state in high field regimes.

To get insight into this unsaturated MR behavior, we measure the Hall resistivity \( \rho_{xy}(\mu_0H) \) under \( I//a \) and \( \mu_0H // c \) configurations, the results are presented in figure 3(a). The isothermal \( \rho_{xy}(\mu_0H) \) curves at different temperatures show obvious nonlinear behaviors, indicative of SmAlSi being a multiband system. Additionally, the slope of \( \rho_{xy}(\mu_0H) \) changes its sign from positive to negative as increased \( \mu_0H \), signifying the variable dominant carriers at different fields. To extract the carrier mobility and density from this nonlinear profile, we fit Hall conductivity \( \sigma_{xy}(\mu_0H) \) by the two carrier model [39–41],

\[
\sigma_{xy}(\mu_0H) = \frac{\rho_{xy}(\mu_0H)}{\rho_{xx}(\mu_0H) + \rho_{xx}^2(\mu_0H)} = \left[ \frac{n_e \mu_e^2}{1 + (\mu_e\mu_0H)^2} - \frac{n_h \mu_h^2}{1 + (\mu_h\mu_0H)^2} \right] \mu_0H
\]

where \( \mu_e (\mu_h) \) and \( n_e (n_h) \) correspond to the mobility and density of the electron (hole). Within this model, zero-field resistivity \( \rho_{xx}(0) \) is related to carrier concentration and mobility through the following equation \( \rho_{xx}(0) = \frac{1}{\xi n_{\mu_e} + n_{\mu_h}} \) [40]. Combined this limitation, the fitted \( \sigma_{xy}(\mu_0H) \) curves at 2 K as examples are shown in the inset of figure 3(b), the extracted carrier density are \( n_e = 1.5 \times 10^{19} \text{ cm}^{-3} \) and \( n_h = 3.63 \times 10^{19} \text{ cm}^{-3} \) with carrier mobility \( \mu_e = 9735 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1} \) and \( \mu_h = 2195 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1} \). With increased temperatures, both carrier density \( n_e, n_h \) and mobility \( \mu_e, \mu_h \) exhibit a nonmonotonic temperature dependence, as illustrated in figures 3(b) and (c). In all temperatures, low carrier density of \( 10^{19} \text{ cm}^{-3} \) and high carrier mobility support SmAlSi being a semimetal, but the large ratio \( \eta = n_h/\mu_h > 2 \) deviated from \( \eta = 1 \) reveals the absence of perfect electron–hole compensation.

3.3. SdH oscillations and FS properties

As another typical feature, SdH oscillations are observed in both isothermal MRs and Hall resistivities, letting us analyze the characteristics of electronic FS of SmAlSi. Figures 4(a) and (b) present the out-plane \( \rho_{xx}(\mu_0H //a, \mu_0H // c) \) and in-plane \( \rho_{xx}(\mu_0H //a, \mu_0H // b) \) resistivity curves at different temperatures. For \( \mu_0H // c \), SdH oscillations can be clearly observed down to low field \( \sim 3 \text{ T} \) in contrast to the LaAlSi(Ge) with oscillations detected only above 10 T [23, 42], signifying the high quality crystals of SmAlSi. After subtracting a smooth background, the oscillatory components of \( \Delta \rho_{xx} \) and \( \Delta \rho_{yx} \) versus \( 1/\mu_0H \) are presented in figures 4(c) and (d). From the fast Fourier transform (FFT) analysis, two independent frequencies \( F_\beta = 14.9 \text{ T}, F_\alpha = 46.1 \text{ T} \) and \( F_\beta = 17.7 \text{ T}, F_\alpha = 52.4 \text{ T} \) at 2 K) and their high order harmonics are derived for \( \mu_0H // c \) and \( \mu_0H // b \) (see figures 4(e) and (f)). Given that the oscillation frequency \( F \) is linked to the FS cross-sectional area \( A_F \) by the Onsager relation \( F = (h/2e\alpha)A_F \), the Fermi wave vector \( (k_F) \) can be evaluated via \( k_F = (A_F/\pi)^{1/2} \), the calculated values at 2 K are to \( 0.20 \text{ nm}^{-1} \), \( 0.36 \text{ nm}^{-1} \) and \( 0.22 \text{ nm}^{-1} \), \( 0.39 \text{ nm}^{-1} \) frequencies \( F_\beta \) and \( F_\alpha \) respectively. The small \( A_F \) less than 0.7% of cross-sectional area of Brillouin zone (BZ) indicates the existence of small Fermi pockets (see table 1). As increased temperature from 2 K to 20 K, \( F_\alpha \) and \( F_\beta \) shrink from 46.1 and 52.4 T to 40.3 and 42.7 T, showing strong temperature dependence. This demonstrates the change of FS as temperature is cooled down from PM to magnetic-ordered states. Similar phenomena are observed in isostuctural magnetic RAISi (R = Pr, Nd) [23, 24] but not in magnetic AlSi (X = Si, Ge) [19, 42], which suggests the significant influence of electronic FSs from the local 4f–electron magnetism of Sm moments. In combination with the abnormal variation of \( \mu_e (\mu_h) \) and \( n_e (n_h) \) near the transition temperatures (see figures 3(b) and (c)), the tunability of FS is linked to the development of magnetic ordering or magnetic structures driven by \( \mu_0H \).

The analysis on amplitude of SdH oscillations are performed by Lifshitz–Kosevich (LK) formula [43–46]:

\[
\frac{\Delta \rho}{\rho_{xx}(0)} \propto \frac{\lambda T}{\sinh \lambda T} \exp(-\lambda T_D), \lambda = \frac{2\pi^2 k_m^*}{\hbar e\mu_0H}.
\]

Here, \( m^* \) denotes the cyclotron mass of the carrier, \( T_D \) is the Dingle temperature defined by \( T_D = h/2\pi \k \tau_Q \) with \( \tau_Q \) being the quantum scattering lifetime. By fitting the temperature dependence of relative FFT amplitude (see the inset of figures 4(e) and (f)), cyclotron mass \( m^* \) can be obtained with \( m_e^* = 0.09 \; m_e, m_h^* = 0.07 \; m_e (\mu_0H // c) \) and \( m_e^* = 0.07 \; m_e, m_h^* = 0.06 \; m_e (\mu_0H // b) \), where \( m_e \) is the free-electron mass. These oscillation masses are very small and comparable to the topological semimetal Cd₃As₂ [47], TaP [48],SrAs₃ [49], etc. The estimated Fermi wave vectors \( (V_F = h \k F / m^*) \) and Fermi energy \( (E_F = h^2 k_F^2 / 2m^*) \) are summarized in table 1. The total oscillations \( \Delta \rho_{xx} \) are fitted based on the existence of two Fermi pockets by fixing \( m^* \) obtained from FFT amplitude, as shown in figures 4(g) and (h). Using this fit, Dingle temperatures for \( \alpha \) and \( \beta \) pockets are extracted to 1.8 K,
14.9 K (\(\mu_0H \parallel c\)) and 1.5 K, 13.1 K (\(\mu_0H \parallel b\)), respectively. Subsequently, quantum mobility \(\mu_q = e\tau_Q/m^*\) can be estimated to 13 000 cm² V⁻¹ s⁻¹ (\(\alpha\) pocket) and 2000 cm² V⁻¹ s⁻¹ (\(\beta\) pocket) for \(\mu_0H \parallel c\), which are in the same order of mobility obtained from Hall effect. These results support SmAlSi has small cyclotron mass and high mobility as typical features of WSM.

Nontrivial Berry phase is considered to be a key evidence of Weyl fermions, which can be extracted from SdH oscillations. According to the LK equation, the oscillation components can be described by \(\Delta \rho \propto \cos 2\pi \left(\frac{F}{\mu_0H} + \gamma + \delta\right)\) where \(\gamma = -\frac{\phi_b}{\pi}\) and \(\phi_b\) is the Berry phase, \(\delta\) represents the FS dimension-dependent correction to the phase shift, which takes 0 or \(\pm 1/8\) for the two dimensional and three dimensional (3D) systems, respectively. As known, \(\phi_b\) is zero for non-relativistic system and finite value \(\pi\) for topological materials with linear energy dispersion. To extract \(\phi_b\), the LK formula is directly used to fit the multi-frequency SdH oscillation by the fixing parameters of frequencies and cyclotron masses. As shown in figures 4(g) and (h), two-band LK fitting reproduces the resistivity oscillations well at 2 K with phase factor \(\phi = \gamma + \delta\). Then, Berry phases \(\phi_b\) are yielded to 1.22 \(\pi\), 0.74 \(\pi\) and 1.06 \(\pi\), 0.75 \(\pi\) for \(F_{\alpha}\) and \(F_{\beta}\), respectively. These values close to \(\pi\) demonstrate the nontrivial Berry phase of both \(\alpha\) and \(\beta\) Fermi pockets. To obtain the accurate Berry phase, we also mapped the Landau level (LL) fan diagram of SdH oscillation, from that \(\phi_b\) can be extracted from linear extrapolation of LL index (\(N\)) versus \(1/\mu_0H\) by the Lifshitz-Onsager quantization relation \(\phi = N + 1/2 - \frac{\phi}{\pi} + \delta\) [44, 50-52]. Here, the peak positions of oscillation components \(\Delta \rho_{Q}\) are assigned to the integer indices \(N\) as a measure of the LLs (see figure 5(a)). Considering that SdH oscillations exhibit complex behaviors for \(\mu_0H > 25\) T due to the Zeeman splitting effects or other undetected frequency, the data for \(\mu_0H < 20\) T are used to extract \(\phi_b\) and herein small index \(N = 3\) let the extrapolation reliable. The intercept for \(F_{\beta}\) pocket results in \(\phi \sim 0.131\) corresponding to \(\phi_b \sim 0.988\) \(\pi\) indicative of nontrivial topological state. Also, the extrapolated frequency 46.9 T is consistent with 46.1 T of \(F_{\beta}\) from the FFT analysis.

To capture the FS anisotropy of SmAlSi, angle-dependent MRs were measured under field rotating within the bc-plane and ac-plane, the schematic diagrams of experimental setup are shown in the insets of figure 6(c). From the FFT spectra of SdH oscillations at different angles (\(\theta, \varphi\)) (see figures 6(a) and (b)), the presence of SdH oscillations in whole angles from 0° to 90° points out the 3D FS feature of SmAlSi. As changed angles, \(F_{\beta}\) displays a nonmonotonic angle dependence reaching maximum at 60°, above that \(F_2\) and \(F_{2\beta}\) become indistinguishable as field rotated within ac plane. Generally, for the elongated ellipsoidal FS pocket with anisotropic 3D

![Figure 4](image-url)
Figure 5. (a) The oscillatory components \( \Delta R_{xx} \) versus \( 1/\mu_0 H \) under \( \mu_0 H \parallel c \). The inset shows magnetic-field dependence of original \( R_{xx} \) (red line) and smooth background (dashed black line) with field up to 48 T. (b) The Landau-level fan diagram for the \( F_c \) pocket at 2 K.

Figure 6. (a) and (b) Angular dependence of FFT spectra obtained from SdH oscillations for field rotating within the \( ac \)-plane and \( bc \)-plane, respectively. The dashed lines are guides to the eyes. (c) Angular dependence of frequencies for \( \alpha \) and \( \beta \) pockets. Inset shows the geometric configurations of measurement, where \( \theta \) or \( \phi \) is defined as the angle between field and \( c \) axis.

3.4. Electronic band structures

The electronic band structures of SmAlSi are calculated. The bulk BZ is shown in figure 7(a). The calculations reveal SmAlSi has magnetic moment of 1.03 \( \mu_B/\text{Sm} \) in close to the experimental value \( \sim 0.76(2) \mu_B/\text{Sm} \) (see appendix, figure 8). As shown in figure 7(b), electron and valence band cross each other close to the FL at several points indicative of its semimetal states, and band crossings are mainly along the \( \Sigma - \Gamma - \Sigma_1 - N \) lines similar to its isostructural \( R \)-AlSi \((R = \text{La, Nd}) \) [20, 24, 25]. As shown in figure 7(c), three electron Weyl pockets can be identified: one small hole pockets located around \( \Sigma \) points in the \( \Gamma - \Sigma \) path, one small electron pocket in the \( N - \Sigma_1 \) path and one large hole pockets along \( \Gamma - Z - \Sigma_1 \) path. The two experimental observed frequencies of SdH oscillations \( F_{\alpha} \) and \( F_{\beta} \) may be ascribed to the small and large Weyl pockets. From the mapped FS, there are totally 56 Weyl points in the whole BZ (see appendix, figure 9), and the electron and hole pockets are nearly compensated, which can explain the observed large unsaturated MRs. Further focusing on the energy window of \(-0.1 \text{ eV} < E_F \), the characteristics of energy dispersions can be more clearly determined as presented in figures 7(d)–(g), which show two examples of these Weyl nodes. From that, the small pockets near \( \Sigma \) points crossing near \( E_F \) belong to type I Weyl fermions and the other pockets near \( \Sigma_1 \) points belong to type II Weyl states. Near the \( \Sigma_1 \) points, the calculated band
top of the hole band to the FL is \(\sim 45\) meV, which is close to \(E^c_F = 50.5\) meV obtained from SdH oscillations. In combination with the light cyclotron mass and nontrivial Berry phase, SmAlSi is proposed to a promising magnetic WSM system for exploiting the interplay between magnetism and Weyl states.

4. Conclusions

In summary, we carried out the systematic magnetotransport studies on SmAlSi single crystals, which exhibit large non-saturated MRs up to \(\sim 48\) T and field induced metal–insulator transitions at low temperatures. The field dependent MRs follow linear dependences at high field regimes (\(\mu_B H > 18\) T) deviating from the semiclassical \((\mu_B H)^2\) relation in compensated semimetal. From the analysis of SdH oscillations, we can identify two-periodic quantum oscillations exhibiting nontrivial \(\pi\) Berry phase and light cyclotron mass, in consistent with the electronic band structure calculations with Weyl nodes located near \(E_F\). Our results indicate SmAlSi as an interesting material on understanding WSM physics in RAX family.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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Appendix

Crystal Structure and Magnetic Properties of SmAlSi

SmAlSi crystallizes into tetragonal LaPtSi type structure with space group \(I4_1md\) (No. 109), where the Sm, Al, Si are located at 4a (0, 0, 0.573 60), 4a (0, 0, 0.166 80) and 4a (0, 0, 0) Wyckoff sites. Room temperature x-ray diffraction of SmAlSi was measured on powders from crushed single crystals, the diffraction patterns were refined by Rietveld method using the GSAS software, and the obtained results are shown in table 2.

Figure 8(a) presents temperature dependence of magnetic susceptibility \(\chi(T)\) under \(\mu_B H = 1\) T along \(b\)-axis and \(c\)-axis, respectively. Two magnetic transitions can be identified at \(T_{N1} \sim 11.5\) K and \(T_{N2} \sim 5\) K as marked by the green and violet dashed lines. The above anomaly indicates the presence of successive antiferromagnetic transitions with incommensurate spin structures [53, 54], at which \(\rho_{\alpha\beta}(T)\) also show anomaly near transition temperatures (see the inset of figure 8(a)). Since Sm ions can exhibit weak temperature-dependent Van-Vleck paramagnetic behavior [55–57], we fitted the magnetic susceptibilities by a modified Curie–Weiss law: \(\chi = \chi_0 + C/(T - \theta_{CW})\), where \(C\) is the Curie constant, \(\theta_{CW}\) is the Curie-Weiss temperature and \(\chi_0\) is temperature independent Van Vleck paramagnetic component (\(\chi_0\)) of Sm\(^{3+}\) ions. The contribution of \(\chi_0\) can be from the excited multiplets as observed in many Sm-containing intermetallic compounds such as in SmPd\(_2\)Al\(_3\) [55] and SmBo [56, 57], etc. As shown in figure 8(b), taking \(\chi_0 = 0.00029\) emu mol\(^{-1}\) (\(\mu_B H \parallel b\)) and \(\chi_0 = 0.00052\) emu mol\(^{-1}\) (\(\mu_B H \parallel c\)), high temperature (50 K < \(T < 150\) K) CW fitting yield \(\theta_{CW}\) and effective magnetic moments \((\mu_{eff})\) \(\theta_{CW} = -31.5\) K (\(\mu_B H \parallel b\)), \(\mu_{eff} = 0.74\) \(\mu_B/\text{Sm}\) (\(\mu_B H \parallel b\)) and \(\theta_{CW} = -19.6\) K (\(\mu_B H \parallel c\)), \(\mu_{eff} = 0.76\) \(\mu_B/\text{Sm}\) (\(\mu_B H \parallel c\)), respectively. The field-dependent magnetization \(M(\mu_B H)\) curves at 2 K are presented in figure 8(c). For \(\mu_B H \parallel c\), the
Table 2. Structural parameters and atomic coordinates of SmAlSi by Rietveld refinement.

| Atom | Wyck. | x   | y   | z   | occ |
|------|-------|-----|-----|-----|-----|
| Sm   | 4a    | 0   | 0   | 0.5736 | 0.98 |
| Al   | 4a    | 0   | 0   | 0.1668 | 1    |
| Si   | 4a    | 0   | 0   | 0    | 0.95 |

Final R indexes \(R_p = 0.0378; \quad R_{wp} = 0.0546\)

Goodness-of-fit on \(F^2\) 1.26

Figure 8. (a) Temperature dependence of susceptibility \(\chi(T)\) for \(\mu_0 H \parallel b\) and \(\mu_0 H \parallel c\), the inset show the enlarged \(\chi(T)\) and resistivity \(\rho_{xx}(T)\) at low temperatures. The green and violet dashed lines mark the positions of \(T_{N1}\) and \(T_{N2}\), respectively. (b) The inverse susceptibility and its Curie–Weiss fitting. (c) Isothermal magnetization curves for field along \(b\) axis (red) and \(c\) axis (blue) at 2 K.

\(M(\mu_0 H)\) curve exhibits nearly linear field dependence, while it shows a spin-flop transition for \(\mu_0 H \parallel b\) with a distinct hysteresis behavior. At \(\mu_0 H = 7\) T, SmAlSi exhibits small magnetic moment \(\sim 0.03\ \mu_B/\text{Sm}\) analogous to the observation in SmB$_6$ [56, 57].

The first-principle calculations on electronic band structures of SmAlSi

To obtain the electronic band structure, the first-principle calculations are performed by including the spin–orbital coupling using the projector-augmented wave method as implemented in Vienna Ab-initio Simulation Package. The calculation is started from the nonmagnetic state, in that case, the Sm \(f\) electrons are kept in the core. The calculated Fermi surface at energy \(E_F\) and \(E = \pm 50\) meV are illustrated in figure 9(a), from that, we can find the conduction and valence bands touch at several points close to the \(E_F\), indicative of its semimetal state. Totally, we can identify 56 Weyl points in the whole Brillouin zone as denoted in figure 9(b). Further checking the calculated band structures (see figure 9(c)), the band crossings are mainly along the \(\Gamma-\Sigma-N-\Sigma_1-Z\) lines, several band crossings with linear dispersion characteristics around \(E_F\). Weyl nodes emerge in the vicinity. Additionally, the electronic structures of SmAlSi in magnetic-ordered state are calculated, in which Sm \(f\) electrons are put in the valence and a Hubbard energy \(U\) of 6.4 eV was used in the calculation. As displayed in figure 9(d), the Sm \(f\) orbital is partially occupied, giving rise to the local magnetism of SmAlSi. In case of ferromagnetic state, the calculations reveal SmAlSi has magnetic moment of 1.03 \(\mu_B/\text{Sm}\). Compared to the nonmagnetic case, the spin-up and spin-down sub-bands split in magnetic ordered state (see figure 9(e)), the energy dispersions and positions near the crossing points in the band structure can be slightly tuned, signifying the modulation of Weyl nodes by magnetism. As an example, the Weyl cone near N-\(\Sigma_1\) and \(\Sigma_1-Z\) points are tilted indicative of possible type II Weyl states. These calculated results share some similarities with its isostructural RAlX (\(R = \text{La, Ce, Pr; X = Ge, Al}\)) compounds identified as WSM materials [20, 24, 27], where the Weyl nodes stem from the broken inversion symmetry and magnetism in this family.
Figure 9. (a) The Fermi surfaces of SmAlSi at $E_F$ and $E = \pm 50 \text{ meV}$ in the whole BZ. (b) The Weyl points in the whole BZ. (c) Band structure for SmAlSi in nonmagnetic state with SOC. (d) Calculated DOS of SmAlSi in FM state. The spin-up and spin-down partial DOS are plotted in red and purple colors, respectively. The 4f states of Sm are represented by blue shaded area. The inset shows the Sm f states below $E_F$. (e) The band structure of SmAlSi in the FM state with SOC.

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