Anomalous quantum oscillations and a non-trivial Berry phase in SmSb

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Abstract

We provide evidence for anomalous behavior in the quantum oscillations of the antiferromagnetic semimetal SmSb. Magnetotransport measurements in high magnetic fields reveal that SmSb has a non-trivial $\pi$ Berry phase inferred from the Landau index analysis of Shubnikov-de Haas (SdH) oscillations. Furthermore, striking differences are found between the temperature dependence of the amplitudes of de Haas-van Alphen effect oscillations, which are well fitted by the Lifshitz-Kosevich (LK) formula across the measured temperature range, and those from SdH measurements which show a significant disagreement with LK behavior at low temperatures. Our findings of unusual quantum oscillations in an antiferromagnetic, mixed valence semimetal with a non-trivial Berry phase can provide an opportunity for studying the interplay between topology, electronic correlations and magnetism.

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INTRODUCTION

The discovery of topological insulators with bulk band gaps but robust topological surface states protected by time reversal symmetry triggered much research into topological phases of matter [1,2]. Topological insulators are bulk insulators yet have conducting surface states as a consequence of the topologically non-trivial band structure, where a Dirac cone lies within the bulk gap [3,4]. Subsequently, a number of semimetals with non-trivial topologies have also been discovered. In some cases such as Dirac and Weyl semimetals, the electronic structure is gapless and contains Dirac or Weyl points at the positions where the bands cross with linear dispersions and the excitations are well described as Dirac or Weyl fermions [5–11]. Meanwhile in other topological semimetals, the band structure contains similar band inversion features to topological insulators, but the Fermi level does not lie within the band gap [12,13]. The role of electronic correlations in topological systems has also become of particular interest after the proposal that SmB$_6$ is a topological Kondo insulator, where the surface state lies within the gap opened due to the Kondo interaction [14–16]. One striking feature is the presence of quantum oscillations in SmB$_6$ from de Haas-van Alphen
effect (dHvA) measurements [17, 18], despite the material being a bulk insulator at low
temperatures, stimulating debate over whether these arise from the surface [17, 19], or if
this is a bulk property [18, 20]. Furthermore, Tan et al. found that the dHvA amplitude
shows an anomalous increase at low temperatures below 1 K, and the origin of this highly
unusual deviation is currently not resolved [18].

Recently, the $X$(Sb,Bi)($X$ = lanthanide) series of semimetals with a cubic rocksalt struc-
ture have attracted considerable interest. These materials are commonly found to have an
extremely large magnetoresistance (XMR), generally attributed to the nearly perfect comp-
pensation of electron and hole carriers [21–27]. Meanwhile, non-trivial band topologies have
been theoretically predicted [28], and evidence for topological states is found experimentally
for some compounds from ARPES and transport measurements [25, 27, 29, 33], while other
members appear to be topologically trivial [24, 34, 35]. In LaSb, it was suggested that the
low-temperature plateau in the temperature dependence of the resistivity in applied mag-
etic fields was due to a topologically protected surface state, similar to that observed in
SmB$_6$ [21]. However, ARPES measurements show conflicting results over whether the band
topology is trivial [33, 34], while the resistivity plateau can be explained by a three band
model with nearly perfect electron-hole compensation [34]. Furthermore, by substituting for
lanthanide elements with partially filled 4f electron shells, the effect of tuning the spin-orbit
coupling, magnetism and electronic correlations can be studied. For example, CeSb shows
evidence for Weyl fermions in the field-induced ferromagnetic state from both theoretical
calculations and the measurement of a negative longitudinal magnetoresistance [25]. NdSb
has also been proposed to be a topological semimetal from the observation of a Dirac-like
semi-metal phase using ARPES, as well as from band structure calculations and analysis of
the Landau indices [30, 36].

In this work, we report magnetoresistance and quantum oscillation measurements of the
antiferromagnet SmSb [37]. We find clear evidence for a non-trivial Berry phase in SmSb,
which is derived from an analysis of the Landau indices obtained from the results of high
field magnetotransport experiments. Moreover, striking differences are found between the
temperature dependences of the quantum oscillation amplitudes from Shubnikov-de Haas
(SdH) and dHvA measurements. The amplitudes of the SdH oscillations show anomalous
behavior at low temperatures, which may arise due to the presence of multiple Fermi surface
sheets, or correspond to a conduction channel related to the topological state.
RESULTS

The temperature dependence of the electrical resistivity $[\rho(T)]$ of SmSb is displayed in Fig. 1a, where the data up to 300 K is in the inset. The $\rho(T)$ data show a small hump at around 65 K, which is explained as resulting from the splitting of the crystalline electric fields \cite{38}, as well as a sharp drop of $\rho(T)$ at the antiferromagnetic transition at $T_N = 2.2$ K \cite{39}. While $T_N$ changes very little in applied magnetic fields \cite{40}, an upturn appears in $\rho(T)$ at low temperatures, which becomes more pronounced as the field is increased. As shown in Figs. 1a and 1b, there is a strong increase of $\rho(T)$ in-field below $T_N$, before reaching a nearly constant value below around 1 K. Figure 1c shows the field dependence of the magnetoresistance $[\rho(H)/\rho(0)]$ of SmSb up to 15 T at various temperatures down to 0.3 K. At 0.3 K $\rho(H)/\rho(0) = 74000$, which decreases rapidly as the temperature is increased. Figure 1d displays measurements performed up to higher fields at 1.8 K, where the magnetoresistance increases quadratically to about 5558 at 60 T. The magnetoresistance of SmSb is one of the largest observed among the XSb family of compounds \cite{25, 34-36, 41}, and the quadratic field dependence suggests that the XMR most likely results from electron-hole carrier compensation \cite{42}. In this case, the increase of mobility below $T_N$ may lead to both the small low temperature values of $\rho(T)$ in zero-field, as well as the very large magnetoresistance and low-temperature plateau of $\rho(T)$ in high fields. While this low temperature plateau has also been observed in other XSb materials \cite{21, 25, 27}, the appearance of the plateau at a lower temperature in SmSb is likely due to the comparatively low $T_N$.

To examine the topology of the electronic structure of SmSb, we also measured the SdH effect in the resistivity, so as to determine the residual Landau index. In particular, to reliably extrapolate to the zeroth level, high field quantum oscillation experiments are necessary, and therefore the magnetoresistance was measured in pulsed magnetic fields up to 60 T. Figure 2a displays the $1/B$ dependence of the quantum oscillations for two samples S2 and S6 in applied fields up to 32 T and 60 T respectively along the [001] direction, after subtracting the background contribution. Landau indices were assigned to the positions of the valleys, as displayed in Fig. 2b. The data were fitted with a linear temperature dependence and the extrapolated residual Landau indices are $n_0 = 0.53(5)$ for sample S2 and $n_0 = 0.52(3)$ for sample S6. This corresponds to a $\pi$ Berry phase, which is evidence
for a non-trivial band topology \cite{43,45}. Importantly, since the lowest observed Landau index is 6 in the 60 T measurements, the uncertainty associated with the fitted intercept is small, indicating that the determination of the Berry phase is reliable. Therefore our results strongly suggest that the electronic structure of SmSb is topologically non-trivial. The origin of this Berry phase is currently not resolved, and further studies are necessary to determine if it originates from a gapless topological state such as in Dirac or Weyl semimetals \cite{2,3,11}. It is also noted that evidence for a $\pi$ Berry phase was also found in LaBi \cite{46}, where both an odd number of band inversions and surface states with an odd number of massless Dirac cones were observed in ARPES measurements \cite{31,33}.

In addition, we also performed field-dependent ac susceptibility measurements down to low temperatures where the presence of dHvA oscillations are clearly observed. For comparison, SdH and dHvA oscillations are shown for various temperatures down to 0.3 K in Figs. 3h and 3i, respectively, after subtracting the background contribution. The respective fast Fourier transformations (FFT) are shown in Figs. 3j and 3k, where two principal frequencies were observed, which remain unchanged below $T_N$, similar to previous reports \cite{40}. The observed values are $F_\alpha \approx 334$ T and 328 T, and $F_\beta \approx 633$ T and $\approx 590$ T for SdH and dHvA respectively, where the differences may be due to a small misalignment. It can be seen that the amplitudes of the SdH oscillations in $\Delta \rho$ corresponding to both the $\alpha$ and $\beta$ bands increase significantly with decreasing temperature below 3 K, while in dHvA measurements the change is more gradual. Furthermore, the amplitude of the $\alpha$-band oscillations is maximum at around 0.8 K, and upon further decreasing the temperature, the amplitude decreases. Meanwhile, the angular dependence of $F_\alpha$ is consistent with a two-dimensional Fermi surface (see Supplementary Information), although in general it is difficult to distinguish this scenario from that of three-dimensional ellipsoidal pockets \cite{47}.

Figures 4a and 4b present the temperature dependence of oscillation amplitudes from both SdH and dHvA measurements. Since there is a rapid change of the resistivity both with changing temperature and field, the SdH oscillation amplitudes are displayed as $\Delta \rho/\rho_0$, so as to normalize by the background values. It can be seen that below around 3 K, there is a distinct deviation between the SdH and dHvA results. The dHvA amplitudes are well fitted across the whole temperature range (0.3 to 10 K) for both the $\alpha$ and $\beta$ bands using the canonical Lifshitz-Kosevich (LK) formula \cite{45}, with effective cyclotron masses of 0.26 $m_e$ and 0.28 $m_e$ respectively. The results are consistent for measurements of both the ac and
dc susceptibility (samples S3 and S5 respectively). However, although the SdH and dHvA data coincide well at higher temperatures, below around 2 K there is a significant deviation which cannot be accounted for by the LK formula. Moreover, for the α-band, the amplitude reaches a maximum at around 1.6 K, before beginning to decrease. We note that these results are highly repeatable, as shown by the measurements of the two samples displayed in Fig. 4 and when the SdH amplitudes are plotted as $\Delta \rho$, the deviation from LK behavior is even more pronounced (see Supplementary Information).

One possible explanation for the deviation between dHvA and SdH measurements is that the unusual behavior of SdH oscillations is related to the topologically non-trivial electronic structure which manifests a $\pi$ Berry phase. We note that dHvA measurements of floating zone grown crystals of the proposed Kondo insulator SmB$_6$ reveal a steep increase of the oscillation amplitude below 1 K which cannot be explained by LK theory [18, 48], while there is also evidence for a non-trivial Berry phase [17]. Currently, there is considerable debate over whether the quantum oscillations in SmB$_6$ are from the insulating bulk, or the surface [17, 18]. Proposals such as the magnetic breakdown scenario give rise to quantum oscillations in the bulk of an insulator which deviate from LK behavior [20], while it has also been suggested that the anomalous increase of dHvA quantum oscillation amplitudes in SmB$_6$ arises due to surface quantum criticality [19]. In contrast to SmB$_6$, the amplitudes of our dHvA measurements of SmSb can be well fitted using the LK expression and the anomalous behavior is only seen in SdH measurements. This may suggest that the anomalous behavior of SmSb arises only from certain conduction channels with high mobility, which do not significantly contribute to dHvA measurements. Metallic edge states were proposed to explain the anomalous quantum oscillations of some charge transfer salts, where the deviation of SdH amplitudes from conventional behavior is not observed in the dHvA [49, 50]. However, these systems have highly conducting quasi-two-dimensional planes where the edge states are suggested to arise in the quantum Hall regime, and this is therefore quite a different scenario to that presented by SmSb. Meanwhile, a comparison between SdH and dHvA cannot be made for SmB$_6$, since SdH oscillations in magnetotransport measurements have not been reported.

Another possibility is that the deviation between SdH and dHvA measurements arises due to the difficulty in disentangling the contributions to the conductivity from the different FS sheets. Although it is often assumed that $\Delta \rho/\rho_0$ is proportional to the density of states
and that the temperature dependence is well described by the LK formula, for multiband systems the situation can be more complicated due to the different contributions to the conductivity from each band \[45\]. This effect will not influence the dHvA results, which can be well described by the LK formula across the whole temperature range. We note that below around $T_N$, the normalized oscillation amplitude of the $\alpha$-band is anomalously low relative to the dHvA results, while for the $\beta$-band there is an increase. This suggests that with decreasing temperature, there may be changes of the relative contributions to the conductivity from the two bands. Therefore, for systems with multiple FS sheets and large magnetoresistance, analysis of the cyclotron masses using SdH measurements may be difficult. As a result, the origin of the dramatic departure from conventional behavior in the SdH amplitudes is an open question, and in particular since this deviation onsets near $T_N$, the possible relationship between the antiferromagnetic state and the anomalous behavior also needs to be explored.

To summarize, we find evidence for a $\pi$ Berry phase in SmSb from the analysis of the Landau indices at high fields. Meanwhile the presence of an extremely large magnetoresistance which increases quadratically with increasing applied magnetic field without saturation suggests the compensation of two types of carriers with high mobilities. Furthermore, our quantum oscillation measurements show that the amplitudes of dHvA oscillations are well described by the canonical LK formula, while those from the SdH effect show anomalous behavior at low temperatures. This unusual behavior in SdH oscillations may be a consequence of the non-trivial band topology, or could arise due to different contributions to the total conductivity from multiple FS sheets. As a result further studies such as ARPES are required to understand the origin of the non-trivial Berry phase, as well as any relationship with the anomalous SdH oscillations.

**METHODS**

Single crystals of SmSb were synthesized using an Sn flux-method \[51\]. The elements were combined in a molar ratio Sm:Sb:Sn of 1:1:20 in an $\text{Al}_2\text{O}_3$ crucible before being sealed in evacuated quartz ampoules. These were slowly cooled from 1100°C down to 800°C, before centrifuging to remove the Sn flux. The resulting crystals are cubic with typical dimensions of around 3 mm. The residual resistivity ratio of $RRR = \rho(300\text{K})/\rho(0.3\text{K}) \approx 4000$, indicates
a high sample quality.

Resistivity and magnetoresistance measurements were performed using a Quantum Design Physical Property Measurement System (PPMS) from 300 K to 2 K with a maximum field of 9 T. Four Pt wires were attached to the sample by spot welding so that the current was applied along the [100] direction. The low temperature resistivity and ac susceptibility were measured in a $^3$He system with a base temperature of 0.27 K and a maximum applied magnetic field of 15 T. The high field magnetoresistance measurements were performed up to 60 T using a pulsed field magnet at the Los Alamos National Laboratory, USA, while measurements up to 32 T were carried out using the Cell 5 Water-Cooling Magnet at the High Magnetic Field Laboratory of the Chinese Academy of Sciences.

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ADDITIONAL INFORMATION

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AUTHOR CONTRIBUTIONS

The project was conceived by F.W, C.Y.G. and H.Q.Y.. The crystals were grown by F.W.. F.W., C.Y.G., J.L.Z, Y.C. and J.S. performed the measurements, which were analyzed by
F.W, C.Y.G., M.S, and H.Q.Y. The manuscript were written by F.W, C.Y.G., M.S. and H.Q.Y. All authors participated in discussions.

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FIG. 1. Temperature dependence of the electrical resistivity of SmSb sample S1 at a zero and low applied fields, and b, at high fields. The inset of a, displays the resistivity over the full temperature range from 0.3 to 300 K. c, Field dependence of the transverse magnetoresistance of SmSb sample S1 at various temperatures up to applied fields of 15 T. d, Transverse magnetoresistance of SmSb samples S2 and S6, up to applied fields of 32 T and 60 T respectively. The solid line shows the $B^2$ dependence.
FIG. 2.  a, SdH oscillations of samples S2 and S6 of SmSb plotted as a function of $1/B$. The oscillations were obtained from the field dependence of the magnetoresistance up to 32 T and 60 T, respectively after subtracting the background contribution. b, The $1/B$ dependence of the valley positions in the SdH measurements. The solid lines show linear fits to the data, which both extrapolate to a non-zero residual value close to $n_0 = 0.5$. 

$n_0(S2) = 0.53\pm0.05$

$n_0(S6) = 0.52\pm0.03$
FIG. 3.  a, SdH oscillations obtained from the field dependence of the magnetoresistance of sample S1 of SmSb at various temperatures. b, dHvA oscillations at several different temperatures from field dependent ac susceptibility measurements of sample S3 of SmSb. The measurements were performed with the magnetic field applied along the principal crystallographic axis. The corresponding FFT spectra are displayed for oscillations in c, SdH, and d, dHvA.
FIG. 4. Temperature dependence of the SdH (filled symbols) and dHvA (empty symbols) amplitudes of the fundamental a, α-band and b, β-band frequencies, for several different samples, where the SdH amplitudes are displayed as $\Delta \rho/\rho_0$. The field ranges over which the data were analyzed are displayed, while the vertical dashed line marks the position of $T_N$. The solid lines display fits to the dHvA amplitudes using the Lifshitz-Kosevich formula, with effective masses of 0.26 $m_e$ for the α-band and 0.28 $m_e$ for the β-band.