Understanding the Quantum Oscillation Spectrum of Heavy-fermion Compound SmB$_6$

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SmB$_6$ is a mysterious compound that is electrically insulating but yet it exhibits quantum oscillations, which are a telltale signature of the metallic state. Adding to the enigma is the possibility that SmB$_6$ is a topological Kondo insulator. Here, we report first-principles, parameter-free all-electron electronic-structure calculations on SmB$_6$, which yield the band structure and crystal-field splittings within the $f$-electron complex in accord with experiments. Predicted energies of several magnetic phases where charge, spin and lattice degrees of freedom are treated on an equal footing are found to be extremely close, indicating the key role of spin fluctuations in SmB$_6$. Our results show that the topological Kondo state of SmB$_6$ is robust regardless of its magnetic configuration. The Fermi surfaces derived from our predicted ground state explain the experimentally observed bulk quantum oscillations, and our large calculated effective mass of electrons at the Fermi surface explains how the material is essentially insulating, with a measured specific heat that is in excellent agreement with our calculations.

Introduction

Heavy fermion physics—a rapidly growing field of study usually involving open $f$ shells or rare-earth elements—is home to many anomalous and fascinating phenomena¹, including unconventional superconductivity, non-Fermi liquid behavior, and the Kondo effect. SmB$_6$ has been of particular interest as a possible strongly correlated topological insulator (TI). Indeed, its possible surface state was reported over 50 years ago, long before TIs were known². Quantum oscillations (QOs) were initially thought to be the evidence for this surface state but later suggested to appear because of an anomalous bulk effect³,⁴. The recent theoretical classification of SmB$_6$ as a new class of strongly correlated electron system—a topological Kondo insulator (TKI)⁵—has
attracted considerable theoretical and experimental attention. However, searches for the expected topological surface states have been ambiguous. Some angle-resolved photoemission spectroscopy (ARPES)\textsuperscript{12–14,16}, scanning tunneling spectroscopy\textsuperscript{17,18}, and de Haas-van Alphen (dHvA) oscillations studies\textsuperscript{9,19} claim to have found the topological surface states. On the contrary, other dHvA experiments\textsuperscript{3,4} found that the quantum oscillations (QOs) of SmB\textsubscript{6} are bulk like. Moreover, the experimentally measured low-temperature linear specific heat of SmB\textsubscript{6} cannot be attributed to a surface state\textsuperscript{4,20,21}. These puzzling and seemingly incompatible properties must be understood before a robust understanding of the strange electronic and topological properties of SmB\textsubscript{6} can be reached.

First-principles theoretical investigations that are usually used to extract the microscopic mechanisms behind materials properties have not been able to solve the aforementioned puzzles satisfactorily. For example, various widely used flavors of the density-functional theory (DFT)\textsuperscript{7,8,22,23} have not been able to provide accurate valence f-band splittings and Fermi surface topology of SmB\textsubscript{6} in accord with experimental results. These fundamental failures are usually ascribed to the difficulty of describing strongly localized f electrons due to the self-interaction errors in DFT.\textsuperscript{24} Other complementary approaches including dynamical mean-field theory (DMFT)\textsuperscript{25} and DFT+Gutzwiller\textsuperscript{6} have been applied to SmB\textsubscript{6}. However, one still does not obtain the correct valence f-band splittings observed in experiments\textsuperscript{12,14,16}. Besides, the aforementioned theoretical studies assume a non-magnetic ground state although recent experiments have demonstrated that there are localized magnetic moments in SmB\textsubscript{6} at low temperatures. Notably, magnetic fluctuations and paramagnetic ordering were observed in muon-spin-rotation (μSR) experiments\textsuperscript{26}. The more recent studies show that all degrees of freedom, including lattice, charge, and spin, should be considered together to properly describe strongly-correlated materials\textsuperscript{27–29}.

In this work, we revisit SmB\textsubscript{6} and report the electronic and magnetic structures of this puzzling system using the strongly-constrained-appropriately-normed (SCAN)\textsuperscript{30} density functional that is capable of treating all degrees of freedom on equal footing without invoking any free parameters such as the Hubbard U parameter. SCAN has proven its ability to handle similar strongly correlated systems such as cuprates\textsuperscript{27,31,32}, binary 3d oxides\textsuperscript{29}, and ABO\textsubscript{3} materials\textsuperscript{28}. We consider several magnetic configurations for SmB\textsubscript{6} and discuss their electronic structures. Our calculations not only capture the correct Sm f state splittings and bulk fermi surface but also reconcile the seemingly incompatible experimental observations for the existence of topological surface states.

Results

SmB\textsubscript{6} is known to crystallize in the CsCl-type structure with the Sm atoms located at the corner and B\textsubscript{6} octahedral cluster lying at the body center of the cubic lattice. To better understand the electronic and structural ground state of this material, we consider non-magnetic (NM),
paramagnetic (PM), and several different magnetic states, including the ferromagnetic (FM) as well as classical $A$, $C$, and $G$-type antiferromagnetic (AFM) configurations. The PM phase is modeled by the special-quasi-random structure (SQS)$^{33,34}$ with a 56-atom supercell. These models adopt a $2 \times 2 \times 2$ supercell with different magnetic configurations as shown in Fig. 1. As reported in Table 1, the relaxed lattice constants of all magnetic configurations agree well with the experimental value of 4.133 Å$^{16}$. The calculated magnetic moments are $\sim 5.4 \ \mu_B$ whereas the average Sm 4$f$ occupation number is around 5.5. As seen from Table 1, the total energies of different magnetic configurations are very close with the $A$-type AFM ($A$-AFM) phase being the most stable one. This is in strict contrast to the NM model considered in earlier studies which is 717.12 meV/atom higher than the $A$-AFM energy. These results further indicate the dominance of quantum fluctuations and highly competing orders in SmB$_6$. Interestingly, the energy and local magnetic moments in the SQS-PM model are almost same as that of AFM and FM phases whereas these are much lower than the NM model. These findings are consistent with those on other strongly correlated materials with many competing orders$^{27-29,31,32}$.

![Fig. 1. The crystal structure of SmB$_6$ with different magnetic configurations.](image)

(a) Non-magnetic (NM), (b) ferromagnetic (FM), (c) special quasirandom structure-paramagnetic (SQS-PM), (d) $A$-type antiferromagnetic ($A$-AFM), (e) $C$-type antiferromagnetic ($C$-AFM), and (f) $G$-type antiferromagnetic ($G$-AFM) phase. The blue and aqua balls represent Sm and B atoms, respectively. The orange and purple arrows denote different magnetic moment directions.
Table 1. Energies and lattice constants for different magnetic configurations of SmB$_6$ as calculated by the SCAN density functional.

| Mag. Configurations | Lattice Constant (Å) | Energy (meV/atom) | $M_{\text{Sm}}$ (μ$_{\text{B}}$) | Topological character |
|---------------------|----------------------|-------------------|-----------------------------|-----------------------|
|                     | a       | b       | c       |                        |                      |
| A-AFM               | 4.126   | 4.126   | 4.129   | 0                      | 5.447                | Non-trivial         |
| SQS-PM              | 4.128   | 4.128   | 4.128   | +2.29                  | 5.446                | Non-trivial         |
| FM                  | 4.129   | 4.129   | 4.129   | +3.42                  | 5.458                | Non-trivial         |
| C-AFM               | 4.126   | 4.126   | 4.122   | +5.18                  | 5.433                | Non-trivial         |
| G-AFM               | 4.129   | 4.129   | 4.129   | +5.88                  | 5.425                | Non-trivial         |
| NM                  | 4.119   | 4.119   | 4.119   | +717.12                | 0                    | Non-trivial         |

The preceding structural analysis suggests that the SQS-PM or magnetic configurations are essential to describe the ground state properties of SmB$_6$. We, therefore, present the band structure of $A$-AFM and SQS-PM magnetic configurations in Fig. 2. Interestingly, our calculated band structures of both the AFM and SQS-PM configurations correctly reproduce the crystal field split $f$ states in agreement with the ARPES experiments,$^{12,14,16}$ which are schematically illustrated in Fig. 2(a). Notably, the $2 \times 2 \times 2$ supercell band structure of $A$-AFM and its unfolded representation on the original $1 \times 1 \times 1$ Brillouin zone (BZ) (Figs. 2(b)-(d)) show that the three crystal-field split $f$ states are located at -0.18, -0.38, and -1.0 eV. Similar $f$ band splitting is also seen in the SQS-PM band structure (Fig. 2(e)) as well as in the band structure of other magnetic configurations (see supplementary materials (SM) for details). However, as expected, the degeneracy of $f$ bands in the SQS-PM model is lowered as compared to other magnetic configurations because of the local spin-disorder on Sm sites. We find that the states near the Fermi level ($E_F$) are dominated by the flat Sm $f$ orbitals along with a small $d$-$f$ hybridized bandgap. These results are in substantial accord with the ARPES measurements and, to the best of our knowledge, such level of agreement has never been achieved by any earlier theoretical studies including DFT and DFT+DMFT calculations.$^{6,7,22,23,25}$

Our results in Fig. 2 show that the low energy states in SmB$_6$ are mainly governed by the dispersive Sm 5$d$ and flat 4$f$ states irrespective of the magnetic configuration. There is a hybridization gap due to quantum mechanical mixing of $d$ and $f$ bands away from the high-symmetry points at $E_F$. Moreover, we find that the Sm $f$ bands are flat, having a high density of states (DOS) of 61.80 states/eV at $E_F$ (see Fig. 2(b)). Such high bulk DOS leads to a calculated specific heat of $\gamma = 18.21$ mJ K$^{-2}$mol$^{-1}$ in close agreement with the experimentally measured value of 10–50 mJ K$^{-2}$mol$^{-1}$.$^{4,20,21}$ This strongly indicates that the unusual specific heat capacity of SmB$_6$ observed in experiments is a bulk effect.
Figure 2. Bulk band structure of SmB$_6$ with different magnetic configurations. (a) Schematic illustration of the observed SmB$_6$ band structure in ARPES experiments.$^{11,12,14}$ The red, blue and green lines represent $f$, $d$- and $p$-states, respectively. (b) The orbital resolved band structure and density of states (DOS) of $A$-AFM in the $2 \times 2 \times 2$ supercell Brillouin zone (BZ). (c) The corresponding unfolded band structure in the primitive $1 \times 1 \times 1$ BZ. (d) Closeup of the area highlighted by a red dashed rectangle in (c). The flat $f$ band and $d$-$f$ hybridized bandgap are resolved near $E_F$. (e) The unfolded band structure calculated using the SQS-PM configuration. The $f$ state degeneracy is further reduced due to the disordered spins at the Sm sites.

Although there is a $d$-$f$ band hybridization gap in SmB$_6$, our results show that the predicted ground state $A$-AFM band structure may be metallic with extremely flat, heavy-fermion-like bands near $E_F$ (Fig. 2(c)). Such a band structure can lead to QOs similar to those found in experiments. We, therefore, use this structure to analyze the experimentally observed QOs.$^4$ In this connection, we plot the evolution of the Fermi surface with energy using a $1 \times 1 \times 2$ supercell in Fig. S2. The Fermi surface consists of two distinct surfaces with their symmetry-related replicas in $1 \times 1 \times 2$ superlattice BZ. The larger surface centered at the $X$ point near the $k_z = 0$ plane is labeled as $\epsilon$ whereas the surfaces lying on the $k_z = \pm \pi/c$ plane are identified as $\rho$ in Fig. S2 and Fig. 3. We find that $\epsilon$ surface is very sensitive to changes in $E_F$. It changes shape drastically with 1 meV change of $E_F$ (see Fig. S2). Importantly, we find that the results obtained by raising $E_F$ by 1 meV from the calculated value produce results in reasonable agreement with experiments, and thus we discuss the calculated results at $E = E_F + 1$ meV in Fig. 3.
We present the calculated QOs with different magnetic field orientations associated with ρ Fermi pockets in Fig. 3(d). The area of this surface varies from 540-718T which is somewhat narrower than the experimentally observed value of 309-750T for ρ_{exp} in Ref. 4 but has the same range. Notably, there are eight symmetry-related ρ surfaces which lie on the k_z = ±π/c planes (Figs. 3(a) and (c)). This is in contrast to the experiments which find 24 such surfaces lying on the k_x = ±π/a, k_y = ±π/a, and k_z = ±π/c planes. These experimental observations can be explained if one considers three types of A-AFM domains with short axis along x, y, and z directions. However, for the sake of simplicity, we have discussed results only with the z-oriented A-AFM domains.

The calculated QO frequencies for ϵ pockets of the Fermi surface are shown in Fig. 3(e). Interestingly, the ϵ pockets, illustrated in Figs. 3 (a) and (b) have an anomalous ‘flatfish’ shape. Their flat portion comes from the Sm f states whereas the large cross-sections consist of the Sm d states (see Figs. 2 (b)-(d)). We find that the oscillation frequency determined from this anomalous ‘flatfish’ is quite low everywhere, except for fields near [100] or [010] where they approach the experimental α_{exp}-frequency. We understand the experimental QO spectrum as follows. A very flat band ϵ_{exp} seen in Ref. 4 agrees well with our ϵ flatfish band along [001]-[111]-[110] directions. The α_{exp} band seen in the experiments may appear due to magnetic breakdown with a purely d-like Fermi surface decoupled from the f states. The experimental band labeled α’_{exp}, near 7000T, maybe another breakdown feature, possibly related to mixing the top of the theoretical ϵ frequencies and experimental α_{exp}-band frequencies. Thus, our results can account for all the main features observed in the experiments.

It should also be noted that the anomalous ‘flatfish’ ϵ pockets can explain the possible insulating character of SmB_{6} observed in experiments. The large experimental α_{exp} band seen by dHvA makes no contribution to the transport, being a product of high-field magnetic breakdown. In contrast, the very flat ϵ-band with large effective masses at the order of 10 m_{e} (see Fig.3(e)) leads to nearly localized heavy-fermion carriers which can act as strong scatterers of the experimental ρ_{exp}-electrons. Indeed, we find evidence for an effective mass divergence lying close to the Fermi level (see SM), which will further enhance fluctuations. Fluctuations associated with the many low-lying magnetic phases can also contribute to strong scattering. Furthermore, the flatness of the ϵ band will make it difficult to observe true topological surface states.
Figure 3. The calculated Fermi surface and the quantum oscillation (QOs) frequencies of $A$-AFM. (a) The calculated Fermi surface of $A$-AFM in the $1 \times 1 \times 2$ superlattice BZ. The $\epsilon$ and $\rho$ Fermi pockets are marked explicitly. The color code indicates the Fermi velocity. (b,c) The projection of (b) $\epsilon$ Fermi pockets on the $k_z = 0$ plane and (c) $\rho$ Fermi pockets on the $k_z = \pi/c$ plane. (d) The calculated quantum oscillation frequencies of $\rho$ Fermi pockets with field rotating from the [001] to [010] directions. The effective masses ($m^*/m_e$) in [001], [111], [110] and [010] directions are marked besides the aqua lines. (e) Same as (d) but for $\epsilon$ Fermi pockets. Comparatively higher effective masses can be seen in the [001], [111], [110] and [010] directions.

Having resolved the structural and electronic properties of bulk SmB$_6$, we now discuss its topological electronic state. As noted above, the low energy states in SmB$_6$ are dominated by Sm 5$d$ and 4$f$ states. Importantly, Sm 5$d$ states show an exceptionally large degree of itinerancy with a large bandwidth of $\sim$3.0 eV irrespective of the magnetic structure. Notably, the Sm 5$d$ states of an Sm$^{2+}$ ion are expected to be empty. However, due to their large bandwidth in SmB$_6$, they span across $E_F$ and open an inverted hybridization gap at $E_F$. The nontrivial topological state thus stays robust and is immune to the magnetic ordering. Interestingly, we find that the band dispersions of the magnetic states are adiabatically connected to the NM band structure despite their distinct Fermiology of bands around the $E_F$. We thus analyze the band topology using the SCAN calculated NM band structure in Figs. 4(a) and (b). There is a clear $d$-$f$ hybridization gap at $E_F$ with an inverted band ordering. To access the topological state, we compute the topological invariants $Z_2 = (v_0,v_1,v_2,v_3)$ defined for systems with time-reversal and inversion symmetries. Since the quantum mechanical mixing of opposite parity states vanishes at the time-reversal invariant momentum...
(TRIM) points ($\Gamma$, $3 \times X$, $3 \times M$, and $R$), the parity for each band is well defined at these TRIM points. The calculated parity eigenvalues of each occupied band are marked explicitly in Fig. 4(a) and (b) whereas the parity of the occupied manifold at each TRIM point ($\delta_i$ with $i = X, M,$ and $R$) is shown in Fig. 4(c). The parity is inverted at the three X points, leading to a TKI state with $Z_2 = (1;111)$ in agreement with the results reported in previous works $^6,7$. Furthermore, we find that the inverted parity of the occupied manifold at X points is preserved in all the magnetic configurations (Fig. 4(d)). The TKI state is thus robust in SmB$_6$. It should be however noted that the classification of the topological state and associated nontrivial surface states would be sensitive to a particular magnetic configuration$^{36,37}$.

![Figure 4. The topological state of SmB$_6$.](image)

(a) The calculated band structure of the NM phase in the primitive ($1 \times 1 \times 1$) unit cell. The calculated parity eigenvalues of occupied bands at the TRIM points are marked. (b) The closeup of the bands along the $\Gamma$-$X$-$M$ line in (a). This band structure is adiabatically similar to the magnetic configurations. (c) Parity ($\delta_i$) of the occupied bands at the eight TRIM points and related associated $Z_2$ invariants. (d) The schematic band diagram of the calculated band structure of magnetic configurations and associated parity eigenvalues. The anticipated topological surface states are also shown as dashed lines.
In conclusion, we have systematically studied SmB$_6$ using the SCAN functional within the framework of the DFT. We find that many magnetic phases are extremely close in energy which suggests that SmB$_6$ exhibits competing magnetic orders and spin fluctuations. Importantly, we obtain accurate crystal-field split $f$ bands, which are in substantial agreement with the available ARPES dispersions. Our analysis gives insight into the surprisingly large bandwidth of Sm 5$d$ states and their hybridization with 4$f$ states, and support the mixed-valence SmB$_6$ ground state with pronounced Kondo physics. Additionally, we find that band inversion happens at the X points irrespective of the magnetic configuration, indicating the robustness of the TKI state. We also report detailed Fermi surface topology of SmB$_6$ and show that our predicted magnetic ground state explains the bulk QOs in substantial accord with the experimental results. Also, the calculated effective mass of electrons at the Fermi surface indicates why the material is essentially insulating, with the measured specific heat being in excellent agreement with our theoretical predictions.

**Computational Details**

All calculations were performed by using the pseudopotential projector-augmented wave method as implemented in the Vienna *ab-initio* simulation package (VASP). A high energy cutoff of 520 eV was used to truncate the plane-wave basis set. The exchange-correlation effects were treated using the strongly-constrained-and appropriately-normed (SCAN) meta-GGA scheme. For the energies and unfolded band structure calculations, we adopted a $2 \times 2 \times 2$ supercell for all the magnetic structure with a $6 \times 6 \times 6 \Gamma$-centered $k$ mesh to sample the bulk BZs. Spin-orbit coupling effects were included self-consistently. The crystal structures and ionic positions are fully optimized with a force convergence criterion of 0.01 eV/Å for each atom and a total energy tolerance of $10^{-5}$ eV. For the PM phase, the polymorphous representation of magnetic moments at different Sm sites is implemented with SQS model. We used the stochastic generation algorithm implemented in the alloy theoretic automated toolkit (ATAT) code to search for the best SQS for the 56 atom supercell. To explore the Fermi surface topology, we adopted a $1 \times 1 \times 2$ supercell for A-AFM model with 4225 $k$ points in order to get a high-quality plot. The predicted quantum oscillation frequencies were calculated using the SKEAF program. We used BandUp to obtain the unfolded band structure. The Fermi surface was obtained with FermiSurfer code.
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Supplementary Materials (SM) for
“Understanding the Quantum Oscillation Spectrum of Heavy-fermion Compound SmB₆”

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Figure S1. Unfolded 2 × 2 × 2 superlattice band structure of SmB₆ with different magnetic configurations. (a) Ferromagnetic (FM), (b) C-type Antiferromagnetic (C-AFM), and (d) G-type Antiferromagnetic (G-AFM) phases. Although the local band structure is sensitive to magnetic configurations, an effective d-f band hybridization remains preserved. The various magnetic configurations are illustrated in Fig.1 of the main text.
Figure S2. Evolution of the Fermi surface of $1 \times 1 \times 2$. $A$-AFM configuration as a function of Fermi energy $E_F$. The Fermi surface at (a)-(c) $E_F$, (d)-(f) $E_F + 1$ meV, (g)-(i) $E_F + 5$ meV, and (g)-(i) $E_F + 10$ meV. The left column shows the Fermi surface whereas the middle and right columns show $\epsilon$ and $\rho$ Fermi pocket projections, respectively. The Fermi surface changes geometric shape drastically with a small change in energy w.r.t $E_F$.

In Fig. S2, the left-hand column shows the evolution of the $\epsilon$ Fermi surface with $E_F$. For larger $E_F$, the surface is convex. For a 1meV shift of $E_F$ (frame d), a concave dimple develops on the lateral surface, which collapses at lower energy, causing the Fermi surface to break up at the original $E_F$ (frame a). The change in curvature implies that the corresponding effective mass changes sign from positive (convex) to negative (concave). This is clearly seen from the cross sections at $E_F + 5$meV (frame h) and $E_F + 1$meV (frame e). At some intermediate energy the band is flat, corresponding to an effective mass divergence. This result could be relevant to cuprate physics, where the QO effective mass of YBCO is found to diverge at both ends of the pseudogap phase.$^1$

![Figure S2](image1)

Figure S3. The calculated quantum oscillation (QO) frequencies with applied magnetic fields for $A$-AFM using a $1 \times 1 \times 2$ supercell at different Fermi levels. (a)-(b) The calculated QO frequencies for $\rho$ Fermi pockets at (a) $E_F$...
(c-d) Same as (a)-(b) but for $\epsilon$ Fermi pockets. The applied magnetic field is rotated from [001] to [010] directions.

References:

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