

Original citation:

Liu, Hsu, Hartstein, Máté, Wallace, Gregory J, Davies, Alexander J, Ciomaga Hatnean, Monica, Johannes, Michelle D, Shitsevalova, Natalya, Balakrishnan, Geetha and Sebastian, Suchitra E (2018) Fermi surfaces in Kondo insulators. Journal of Physics: Condensed Matter, 30 (16). 16LT01. doi:10.1088/1361-648X/aaa522

Permanent WRAP URL:

http://wrap.warwick.ac.uk/101666

Copyright and reuse:

The Warwick Research Archive Portal (WRAP) makes this work by researchers of the University of Warwick available open access under the following conditions. Copyright © and all moral rights to the version of the paper presented here belong to the individual author(s) and/or other copyright owners. To the extent reasonable and practicable the material made available in WRAP has been checked for eligibility before being made available.

Copies of full items can be used for personal research or study, educational, or not-for-profit purposes without prior permission or charge. Provided that the authors, title and full bibliographic details are credited, a hyperlink and/or URL is given for the original metadata page and the content is not changed in any way.

Publisher's statement:

"This is an author-created, un-copyedited version of an article accepted for publication in: Journal of Physics: Condensed Matter. The publisher is not responsible for any errors or omissions in this version of the manuscript or any version derived from it. The Version of Record is available online at http://dx.doi.org/10.1088/1361-648X/aaa522"

A note on versions:

The version presented here may differ from the published version or, version of record, if you wish to cite this item you are advised to consult the publisher's version. Please see the 'permanent WRAP URL' above for details on accessing the published version and note that access may require a subscription.

For more information, please contact the WRAP Team at: wrap@warwick.ac.uk

Fermi surfaces in Kondo insulators

Hsu Liu,^{1*} Máté Hartstein,^{1*} Gregory J. Wallace,¹ Alexander J. Davies,¹ Monica Ciomaga Hatnean², Michelle D. Johannes³, Natalya Shitsevalova⁴, Geetha Balakrishnan² and Suchitra E. Sebastian¹

E-mail: suchitra@phy.cam.ac.uk

5 March 2018

Abstract. We report magnetic quantum oscillations measured using torque magnetisation in the Kondo insulator YbB_{12} and discuss the potential origin of the underlying Fermi surface. Observed quantum oscillations as well as complementary quantities such as a finite linear specific heat capacity in YbB_{12} exhibit similarities with the Kondo insulator SmB_6 , yet also crucial differences. Small heavy Fermi sections are observed in YbB_{12} with similarities to the neighbouring heavy fermion semimetallic Fermi surface, in contrast to large light Fermi surface sections in SmB_6 which are more similar to the conduction electron Fermi surface. A rich spectrum of theoretical models is suggested to explain the origin across different Kondo insulating families of a bulk Fermi surface potentially from novel itinerant quasiparticles that couple to magnetic fields, yet do not couple to weak DC electric fields.

¹Cavendish Laboratory, Cambridge University, JJ Thomson Avenue, Cambridge CB3 0HE, UK,

²Department of Physics, University of Warwick, Coventry CV4 7AL, UK,

³Center for Computational Materials Science, Naval Research Laboratory, Washington, DC 20375, USA,

⁴The National Academy of Sciences of Ukraine, Kiev 03680, Ukraine.

^{*}These authors contributed equally to this work.

Introduction

Evidence for a bulk Fermi surface in the Kondo insulator SmB_6 has been observed from a variety of experimental techniques, spanning quantum oscillations in the torque magnetisation [1, 2, 3], finite linear specific heat [1, 2, 4, 5], oscillatory magnetic entropy [2], and magnetic field enhanced thermal conductivity [2, 6]. A remarkable possibility is that the existence of a Fermi surface is more universal to correlated insulators. The observation of a Fermi surface in multiple families of correlated insulators would support a new paradigm that is distinct from the traditional idea where Fermi surfaces are the preserve of Fermi liquids. Here, we experimentally explore the possibility of a bulk Fermi surface in the Kondo insulator YbB_{12} , a material that is closely related to SmB_6 [7].

Kondo insulators are characterised by an energy gap arising from collective f-electron-conduction electron hybridisation, as schematically shown in figure 1(a) [8]. In the case of the Kondo insulator YbB₁₂, collective hybridisation occurs between the f-electron band and two conduction electron bands (shown in figure 4(a)), leading to a complex hierarchy of gaps. A small indirect gap of size ≈ 5 meV in YbB₁₂, similar in size to that in SmB₆, is determined from electrical transport measurements (figures 1(a) and (b), [9, 10]), accompanied by a larger direct hybridisation gap of ≈ 200 meV (figure 1(a)) accessed by complementary measurements such as optical spectroscopy [11] and tunneling spectroscopy [12]. Early observations of a finite linear term in the specific heat capacity (figure 1(d), [13]) and a finite density of states in tunneling experiments [10] led to broad debate about the Kondo insulating mechanism in these materials. In view of the itinerant low energy excitations revealed in the Kondo insulator SmB₆ by quantum oscillation measurements [1, 2, 3], we revisit this question by searching for quantum oscillations in the magnetic torque measured on single crystals of YbB₁₂.

Methods

The growth of single phase YbB_{12} is challenging given its peritectic phase diagram and decomposition into YbB_{66} beyond a very narrow temperature range, necessitating careful control of temperature and composition during the growth process [14, 15]. Source YbB_{12} powder was prepared in polycrystalline form by borothermal reduction of a mixture of Yb_2O_3 (99.998 mass % purity) and amorphous B (99.9 mass % purity) at 1700°C under vacuum [16]. The material was then isostatically pressed into a cylindrical rod and sintered at 1600°C in argon gas flow for several hours. Single crystal growth of YbB_{12} was carried out at the University of Warwick by the traveling solvent floating zone technique under conditions similar to those previously reported in [14] using a four-mirror Xenon arc lamp (3 kW) optical image furnace (Crystal Systems Incorporated, Japan). The growths were performed in a reducing atmosphere

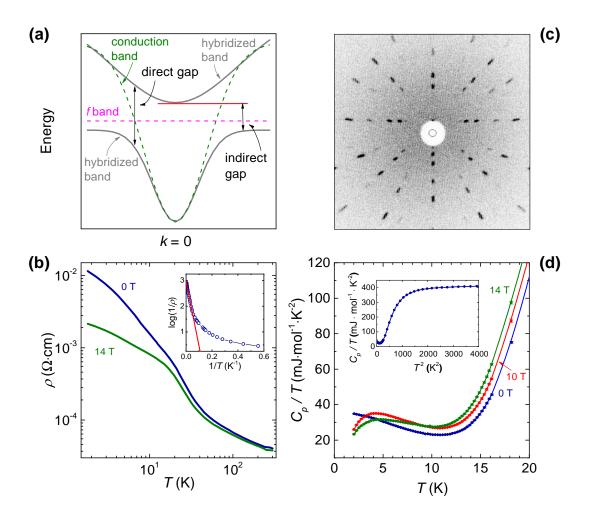


Figure 1. (a) Schematic depiction of the gap formation in a Kondo insulator by collective hybridisation of the f-electron and conduction electron bands. (b) Measured electrical resistivity as a function of temperature in magnetic fields of 0 T and 14 T. The inset shows the exponential fit $1/\rho \propto \exp\left(-2\Delta/k_{\rm B}T\right)$ to the electrical resistivity ρ in YbB $_{12}$ at 0 T, where Δ is the activation gap. The indirect hybridisation bandgap is found to be \approx 5 meV. (c) X-ray Laue back reflection photograph of a crystal of YbB $_{12}$ (on which measured quantum oscillations are shown in figure 2) along the [001] direction, showing single crystallinity. (d) Measured specific heat capacity divided by temperature of YbB $_{12}$ at 0 T, 10 T, and 14 T. A finite linear specific heat coefficient at low temperatures is seen, similar to that observed in SmB $_6$ [1, 2, 4, 5]. The measured linear specific heat coefficient is similar to that reported in [13]. While the finite value of γ above the upturn is enhanced by a magnetic field, as would be expected for a reduction in the activation gap, the low temperature upturn in specific heat capacity is suppressed by a magnetic field, suggesting its correspondence to a secondary low energy scale such as that associated with magnetic impurities or magnetic excitons, which is suppressed by a magnetic field. The inset shows the zero-field specific heat capacity divided by temperature versus T^2 up to 63 K.

(Ar + 3% H₂) at a growth rate of 18 mm/hr with the feed and seed rods counter-rotating at 20–30 rpm. LuB₁₂ single crystals were prepared at the National Academy of Sciences of Ukraine, Kiev by the inductive floating zone method as described in [16]. Laue x-ray imaging with a Multiwire Laue system was used to determine the quality of the grown crystal boules and to select and orient single crystal samples cut from the as-grown boule. Single crystals were selected that yielded high inverse electrical resistivity residual ratios (figure 1(b)) and well-defined spots in the Laue diffraction pattern, evidencing high single crystal quality (figure 1(c)). Elemental composition analysis was performed on selected single crystals using an FEI Philips XL30 sFEG scanning electron microscope (SEM) to reveal an atomic ratio between Yb and B closely comparable to the stoichiometric ratio of 1 : 12 and distinct from the ratios for YbB₆ and for YbB₆₆. Energy dispersive x-ray (EDX) microanalysis on multiple samples provided comparable results to SEM. Rietveld refinement performed using the Bruker TOPAS software on powder x-ray diffraction data yielded a lattice constant of 7.4686(1) Å, agreeing well with published data for YbB₁₂ [16].

Torque magnetisation measurements were made on YbB₁₂ and LuB₁₂ in DC magnetic fields at the University of Cambridge (up to 14 T) and at the National High Magnetic Field Laboratory (NHMFL) in Tallahassee (up to 45 T) using flexible T-shaped BeCu cantilevers of 20 or 50 μ m thickness, with the narrow end anchored and the wide end floating above a fixed Cu film. Single crystals of dimensions approximately $1 \times 1 \times 0.5 \text{ mm}^3$ were mounted on the wide end of the cantilever. The cantilever and the Cu film form the two plates of a capacitor whose capacitance is measured using a General Radio analogue capacitance bridge in conjunction with a Stanford Research Systems lock-in amplifier. The measured change in capacitance ΔC is proportional to the change in magnetic torque. Specific heat measurements were made using the standard heat capacity option for the Physical Property Measurement System (PPMS) from Quantum Design Inc (figure 1(d)).

Density functional theory Fermi surfaces were calculated with the Wien2k augmented plane wave plus local orbital (APW+lo) code [17]. The modified Becke-Johnson (mBJ) potential was used, which is a semi-local approximation to the exact exchange plus a screening term [18] and which improves the band gap in many semiconductor materials. Application of mBJ resulted in a non-magnetic ground state with an indirect band gap of 21 meV and a direct gap of 80 meV, whereas the standard Perdew Burke Ernzerhof (PBE) potential produced a semimetal with overlapping valence and conduction bands. Spin-orbit coupling was included via the second variational method and resulted in a strong reordering of the bands. Self-consistent calculations were converged using a k-mesh of $15 \times 15 \times 15$ followed by a non-self-consistent calculation with a $30 \times 30 \times 30$ mesh for calculation of Fermi surfaces. Extremal cross-sectional areas of the Fermi surfaces were calculated for the magnetic field in the [001], [110] and [111] cubic crystal directions using the open source visualization software, OpenDX.

Effective masses were obtained by shifting the Fermi energy up/down by 0.7 meV from its original value, obtaining the new cross-sections and then calculating the cyclotron effective mass using the resulting finite differences.

Quantum oscillations in YbB₁₂

Figure 2(a) shows the magnetic torque measured on a single crystal of YbB₁₂ as a function of DC magnetic fields up to 45 T, from which a monotonic smooth background has been subtracted. Quantum oscillations periodic in inverse magnetic field are observed for magnetic field tilt angles θ spanning [001], [111] to [110] crystal directions, and extending down to magnetic fields at least as low as 22 T. An example Fourier transform of quantum oscillations in inverse magnetic field shown in figure 2(c) reveals multiple quantum oscillation frequencies between 300 - 1500 T. The quantum oscillation frequency >1 kT is seen most clearly in the high magnetic field range (figure 2(c) inset). While this higher quantum oscillation frequency is close to a harmonic of lower frequency quantum oscillations, there is no obvious observation of frequencies corresponding to harmonics of dominant amplitude low frequency quantum oscillations. The temperature dependence of the measured quantum oscillation amplitude follows a Lifshitz-Kosevich (LK) form, yielding cyclotron effective masses of m^*/m_e between 3-10 for the various measured quantum oscillation frequencies (figure 2(d), [19, 20]). Figure 2(b) shows the angular dependence of the measured quantum oscillation frequencies for magnetic field tilt angles spanning [001], [111] to [110] crystal directions, which reveals only a subtle variation in quantum oscillation frequency as a function of angle, consistent with a three-dimensional Fermi surface geometry.

We calibrate the absolute amplitude of quantum oscillations we observe in YbB₁₂ in units of $\mu_{\rm B}$ per Yb unit cell by using the spring constant of the cantilever used to make torque magnetisation measurements. The method to convert the measured capacitance to absolute units of magnetic torque is the same as that detailed in [2]. Keeping the notation consistent with [2], we have cantilever length L=3.1 mm, distance between cantilever and fixed Cu plate $d_0=0.1$ mm, spring constant $k=190~{\rm N\cdot m^{-1}}$, lattice constant $a_{\rm u.c.}=0.747~{\rm nm}$, and crystal volume $s^3=0.9\cdot 0.5\cdot 0.3~{\rm mm^3}$. We thus convert the measured torque magnetisation in terms of capacitance (C) to an absolute magnetic moment p_s in units of Bohr magneton per unit cell by the expression:

$$\Delta p_s = \frac{20}{\mu_0 H \sin \theta_M} \cdot \Delta C \quad \text{T} \cdot \text{pF}^{-1} \,\mu_{\text{B}} \text{ per unit cell.}$$
 (1)

Here, $20 \text{ T} \cdot \text{pF}^{-1}$ is calculated with the parameters above according to [2], and θ_M is the angle between the magnetic field $\mu_0 H$ and the total magnetic moment.

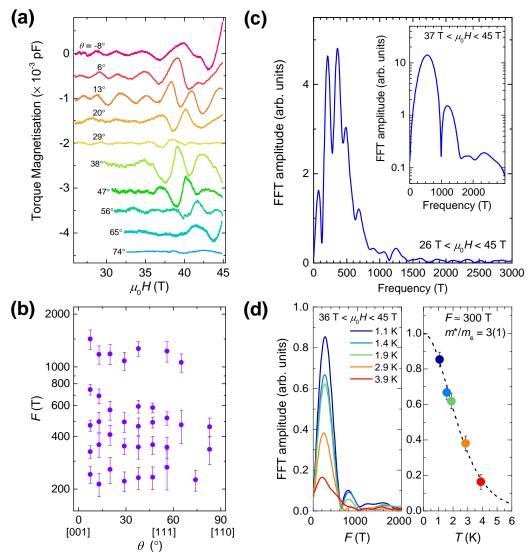


Figure 2. (a) Solid lines show de Haas-van Alphen oscillations measured on a single crystal of YbB₁₂ using torque magnetisation at T=0.4 K for μ_0H oriented at different tilt angles (θ) away from the [001] crystalline direction, passing through the [111] crystalline direction, and approaching the [110] crystalline direction. (b) Angular dependence of measured quantum oscillation frequencies for values of magnetic field tilt angle spanning the [001] crystalline direction, through the [111] crystalline direction, and approaching the [110] crystalline direction. (c) Example Fourier transform of the magnetic field sweep at $\theta\approx13^\circ$ showing quantum oscillation peaks for a magnetic field window between 26 T and 45 T. The inset shows the Fourier transform of the magnetic field sweep at $\theta\approx13^\circ$ for a high magnetic field window between 37 T and 45 T, more clearly showing the quantum oscillation peak at 1.2 kT. (d) Left panel shows an example Fourier transform at $\theta\approx6^\circ$ measured at different temperatures. Right panel shows the quantum oscillation amplitude obtained from the peak height of the Fourier transforms shown in the left panel, plotted as a function of temperature T. Performing a Lifshitz-Kosevich fit (shown by a dashed line) yields an effective mass of $m^*/m_e=3(1)$.

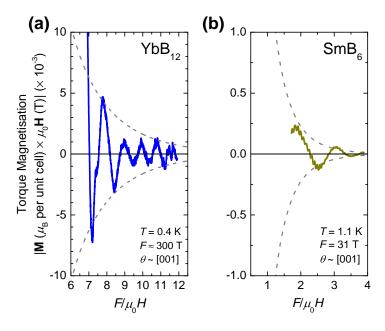


Figure 3. Quantum oscillatory magnetic moment (in μ_B per unit cell) corresponding to the measured oscillations versus inverse magnetic field in YbB₁₂ in (a), and SmB₆ in (b) (reproduced from [2]). Dashed lines represent the calculated magnetic field dependence of the quantum oscillation amplitude from exponential Dingle damping, for a damping factor of ≈ 100 T in the case of YbB₁₂, and 30 T in the case of SmB₆. The theoretical Lifshitz-Kosevich estimate for the quantum oscillatory magnetic moment taking into account the angular anisotropy term, Dingle and spin-splitting damping factors is found to be $1-4 \times 10^{-4}$ μ_B per unit cell at $F/\mu_0H = 6.9$ in the case of YbB₁₂, and $0.2-1 \times 10^{-4}$ μ_B per unit cell at $F/\mu_0H = 1.9$ in the case of SmB₆.

We compare the measured size of the quantum oscillations to the theoretical Lifshitz-Kosevich estimate for a quantum oscillatory magnetic moment of bulk origin, including the angular anisotropy term, Dingle and spin-splitting damping factors as detailed in [2]. For the quantum oscillations observed in YbB₁₂, we use the experimentally measured values inferred from figure 2, and a Dingle damping factor $R_{\rm D}=\exp(-100~{\rm T}/\mu_0 H)$. We find the expected theoretical amplitude of the magnetic moment for the $F\approx 300~{\rm T}$ frequency of YbB₁₂ to be of the order of $1\text{--}4\times 10^{-4}~\mu_{\rm B}$ per unit cell at $\mu_0 H=45~{\rm T}$. This theoretical estimate is comparable in order of magnitude to the experimentally measured amplitude of quantum oscillations shown in figure 3(a).

We consider a breadth of experimental observations to discern whether the observed quantum oscillations correspond to the bulk volume of the sample. Firstly, we find that the experimentally measured quantum oscillation amplitude (shown in figure 3) is in agreement with the theoretical estimate from the Lifshitz-Kosevich theory for quantum oscillations arising from the bulk volume of the sample, as calculated above. Secondly, we compare two

samples with differing impurity concentrations, and examine whether the amplitude of quantum oscillations scales with the Dingle impurity term as would be expected for quantum oscillations arising from the bulk volume of the sample. We measure quantum oscillations in a single crystal of YbB₁₂ with more domains in the Laue pattern signaling grain boundaries or inclusions, and a lower value of inverse electrical residual resistivity ratio than the sample on which quantum oscillations are shown in figures 2 and 3. Quantum oscillations are observed on this sample, with a substantially higher Dingle impurity damping factor of $R_D = \exp(-300 \text{ T}/\mu_0 H)$. We find the amplitude of quantum oscillations observed in this single crystal with increased impurity levels to be more than an order of magnitude lower than the higher quality single crystal shown in figures 2 and 3, as expected for quantum oscillations originating from the bulk of the single crystal rather than from small secondary phase inclusions. Thirdly, we study the neighbouring semimetal YbB₆ in the peritectic phase diagram, to examine the likelihood of the observed quantum oscillations arising from inclusions of this phase. We find that torque magnetisation measurements on pure single crystals of YbB₆ yielded no discernible quantum oscillations up to an applied magnetic field of 14 T under similar experimental conditions of low temperature and high measurement sensitivity, making this an unlikely source of the quantum oscillations observed in single crystals of YbB₁₂. Fourthly, we note the quantum oscillation amplitude at the highest magnetic fields potentially shows a growth in amplitude beyond that expected solely from Dingle impurity damping for magnetic field tilt angles close to the [001] crystalline direction (figures 2(a) and 3(a)). Especially given that the magnetic field at which an insulatormetal transition occurs in YbB₁₂ is lowest (≈ 50 T) along the [001] crystalline direction [21], a rapid increase in quantum oscillation amplitude at high magnetic fields would be a natural consequence of the approach to an insulator-metal phase transition in bulk YbB12, unlike in the case of a secondary phase inclusion.

Fermi surface origin in YbB₁₂

We explore band structure calculations to shed light on the origin of the Fermi surface observed in YbB₁₂ yielding quantum oscillations with low frequency and high effective mass, in contrast to the quantum oscillations of both low and high frequency and low effective mass observed in SmB₆. Band structure calculations of YbB₁₂ using the mBJ potential are shown in figure 4. States at both the valence band maximum and conduction band minimum are mainly derived from Yb f-states, but are heavily hybridised with the dispersive boron s and p states in the vicinity. We decompose the eigenvectors at each k-point both orbitally and atomically. Integrating over all k-points, we find, for the 14 states of the Yb f-complex, 13.2 below the Fermi energy and 0.8 above (figures 4(a), 4(b)). Counting the empty states indicates 0.8 holes in the f-complex, consistent with a nominally Yb³⁺ semiconducting state that corresponds well

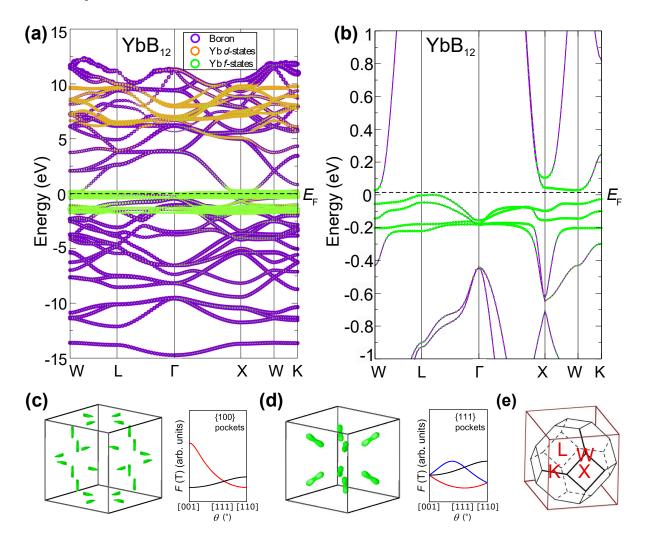


Figure 4. Calculated band structure of YbB_{12} shown over a wide energy range in (a) with an expanded view around the Fermi energy $E_{\rm F}$ in (b). Several characters are projected out of the eigenvectors at each k-point and the resulting weight is indicated by a circle of proportional size. Green circles are Yb f-states, orange circles are Yb d-states, and violet circles are boron states. (c) Small needle-shaped Fermi surfaces of YbB_{12} obtained using the modified Becke-Johnson potential for a small positive energy shift. Expected angular dependence of the quantum oscillation frequencies in the [001]-[111]-[110] rotation plane are shown by approximating the shown Fermi surfaces as prolate ellipsoids. (d) Small peanut-shaped Fermi surfaces of YbB_{12} obtained using the modified Becke-Johnson potential for a small negative energy shift. Expected angular dependence of the quantum oscillation frequencies in the [001]-[111]-[110] rotation plane are shown by approximating the shown Fermi surfaces as prolate ellipsoids. (e) A schematic of the conventional face-centred cubic Brillouin zone used for the band structures within the cubic Brillouin zone used for the Fermi surfaces.

with experiment [7] and band structure calculations that use a GW potential [22]. Interestingly, application of DFT+U, the most commonly used technique for dealing with metallic f-states, instead results in all 14 states being filled and corresponds to Yb²⁺, which is inconsistent with experiment, as also found by previous band structure calculations [23]. A crucial distinction is therefore seen between the band structure of SmB₆ and YbB₁₂. In the case of SmB₆, the boron bands are filled and consequently a single half-filled unhybridised conduction d-electron band crosses the Fermi energy. Hybridisation of this conduction band with the f-electron band yields the Kondo gap. In contrast, in the case of YbB₁₂, two partially filled unhybridised s-p conduction electron bands that are cumulatively half-filled cross the Fermi energy with electron-like character, and are gapped by hybridisation with the f-electron band as shown in figures 4(a) and 4(b).

Given the absence of a finite electronic density of states at the Fermi energy, constituent neutral quasiparticles have been proposed to explain the observation of a bulk Fermi surface in Kondo insulators. Neutral quasiparticles invoked by various theoretical models include spinons in the case of single band Mott insulating organic spin liquids [24, 25, 26, 27], and in the case of single band Kondo insulators such as SmB₆, magnetic excitons [28], composite excitons [29, 30], Majorana fermions [31, 32, 33], and others [34, 35]. A natural way to think of a Fermi surface of such neutral quasiparticles is in terms of slow fluctuations in space and time between the insulating ground state where a Fermi surface is absent due to filling of the Brillouin zone, and the neighbouring metallised ground state in phase space, which is characterised by a Fermi surface [1, 2]. The character of such a neutral Fermi surface may thus be expected to be akin to the Fermi surface of the neighbouring metallised ground state. Metallisation in the case of SmB₆ requires a decoupling of the f-electron and conduction electron bands, resulting in a solely conduction electron Fermi surface occupying half the Brillouin zone. Accordingly, comparison of the observed Fermi surface in insulating SmB₆ with a conduction electron Fermi surface similar to that in metallic LaB₆ was found to yield good agreement both in frequency and effective mass [1, 2]. In contrast, metallisation of YbB_{12} can arise through two routes: either (i) a decoupling of the f-electron and conduction electron bands yielding a Fermi surface corresponding to the conduction electron band, or (ii) an effective relative shift of each of the two hybridised conduction electron bands, yielding a Fermi surface corresponding to a heavy fermion semimetal [29, 30].

We consider the first scenario, and make a comparison of the small heavy Fermi surface sections observed in insulating YbB_{12} with a conduction electron Fermi surface similar to that in metallic LuB_{12} . In LuB_{12} , all 14 f-states are well below the Fermi energy in the calculated band structure, resulting in a metallic ground state with highly dispersive boron bands and light carriers (figure 5(a), [36, 37]). Quantum oscillations we measure using torque magnetisation in LuB_{12} (shown in figures 5(e)–(g)) yield good agreement with the calculated

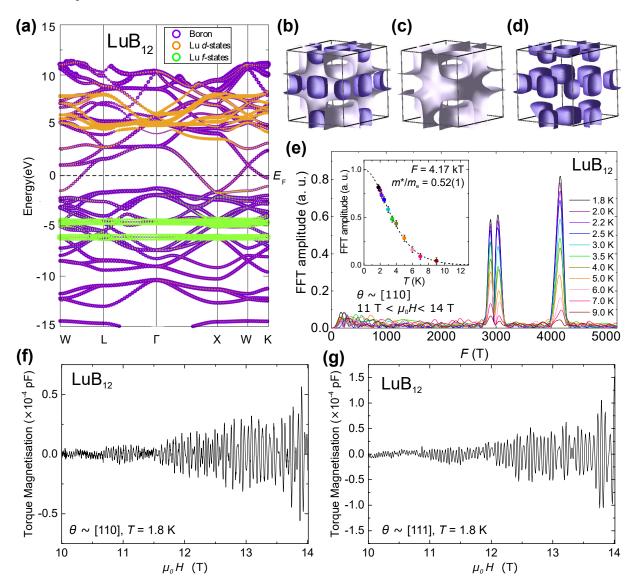


Figure 5. (a) Calculated band structure of LuB₁₂. Several characters are projected out of the eigenvectors at each k-point and the resulting weight is indicated by a circle of proportional size. Green circles are Lu f-states, orange circles are Lu d-states, and violet circles are boron states. (b) - (d) the Fermi surfaces of LuB₁₂ shown together in (b) and separately for clearer viewing in (c) and (d). (f) - (g) De Haas-van Alphen oscillations measured using torque magnetisation at 1.8 K in LuB₁₂. (e) Fourier transform of the magnetic field sweeps taken at different temperatures. The inset shows the Lifshitz-Kosevich fit (dashed line) to the frequency F=4.17 kT. The effective masses m^*/m_e are found to be 0.40(1), 0.53(2), and 0.52(1) for the frequencies 2.90 kT, 3.03 kT, and 4.17 kT for $\mu_0 H$ oriented a few degrees away from [110], corresponding well to calculated frequencies 2.92 kT and 3.93 kT for $\mu_0 H$ || [110] with effective masses m^*/m_e of 0.36 and 0.43 respectively. Frequencies 2.89 kT, 3.51 kT, 3.75 kT, and 5.64 kT are measured for $\mu_0 H$ oriented a few degrees away from [111], comparable to calculated frequencies of 2.82 kT and 5.80 kT for $\mu_0 H$ || [111] with effective masses m^*/m_e of 0.35 and 0.67 respectively.

band structure (figures 5(a)–(d)), and with previous quantum oscillation measurements [36, 38]. A correspondence is not immediately obvious between the observed quantum oscillations with a relatively high effective mass in YbB₁₂ (figure 2(d)) and the large and light conduction electron Fermi surface expected from band structure and observed in LuB₁₂ (figure 5). It is possible that owing to subtle materials differences between YbB₁₂ and LuB₁₂, a band shift could yield small Fermi surface pockets from the conduction electron band similar to those observed in YbB₁₂. Meanwhile quantum oscillations corresponding to large Fermi surface pockets might not be observed due to even higher effective masses than for the small Fermi surfaces observed. Alternatively, in the second scenario, metallisation in YbB₁₂ can be achieved by a small effective relative shift of the two conduction bands while retaining hybridisation or potentially reduced hybridisation. In this case, small heavy Fermi surface sections characteristic of a heavy fermion semimetal [8] would be expected, as can be seen from performing small energy shifts to the band structure (figures 4(c) and (d)). Such heavy Fermi surface sections would also yield a sizable linear heat capacity as observed in YbB₁₂ (figure 1(d), [13]). This scenario of a neutral Fermi surface in Kondo insulating YbB₁₂ with properties similar to the Fermi surface of a heavy fermion semimetal, arising from a small relative shift to the two hybridised conduction bands, is intriguing to pursue theoretically. The realm of Fermi surfaces in Kondo insulators may be even richer than previously thought. Just as materials differences between metals yield differences in band structure and consequently Fermi surface character, we find that differences between the nature of f-electron and conduction electron hybridisation in SmB₆ and YbB₁₂ yield potentially important differences in Fermi surface character between the two Kondo insulators.

Magnetic field tuning in YbB_{12}

Another salient difference between SmB₆ and YbB₁₂ is the effectiveness of magnetic field in tuning these materials toward an insulator-metal transition. While applied magnetic fields as high as $\mu_0 H = 93$ T only result in a negative magnetoresistance of ≈ 7 % in SmB₆ [39], applied magnetic fields of $\mu_0 H \approx 45$ T are found to reduce the electrical resistivity by an order of magnitude in YbB₁₂ (figure 1(b), [21]). The enhanced response of YbB₁₂ to applied magnetic fields compared to SmB₆ may reflect small band shifts due to Zeeman splitting or the involvement of magnetic degrees of freedom in the ground state of YbB₁₂. The close proximity of YbB₁₂ to the insulator-metal transition may be expected to lead to an increased propensity for quantum oscillations originating from neutral quasiparticles [24, 25, 26, 27, 29, 30, 31, 32, 33]. Such an effect would yield larger amplitude quantum oscillations at higher magnetic fields, potentially yielding the increase in quantum oscillation amplitude beyond that expected from Dingle damping suggested from the experimental data for magnetic field tilt angles near

the [001] crystalline direction in YbB $_{12}$ (figures 2(a) and 3(a)). Alternatively, the reduced charge gap at high magnetic fields could also provide an explanation in terms of conventional quasiparticles tunneling through a narrow energy gap that are more likely to yield quantum oscillations [40, 41, 42, 43]. Taken in conjunction with the observation of a finite linear specific heat capacity (figure 1(d), [13]) even in zero magnetic field, an explanation for the observed quantum oscillations in terms of a Fermi surface originating from novel quasiparticles that couple to a magnetic field but not to a weak DC electric field appears more likely than an origin from conventional tunneling through a magnetic field reduced charge gap.

Summary

The observation of magnetic quantum oscillations in at least two families of Kondo insulators, SmB₆ and YbB₁₂, suggests a more universal phenomenon across correlated insulators. Differences we uncover between the character of the underlying bulk Fermi surface in the two systems further add to the richness of potential Fermi surface models relevant to the broad panorama of Kondo insulators. While the Fermi surface observed in SmB₆ corresponds in geometry and effective mass to the conduction electron Fermi surface, the Fermi surface observed in YbB₁₂ corresponds more closely to that of a heavy fermion semimetal, suggesting important differences between theoretical models of relevance to each of these systems. The magnetic field tuning we find to influence the quantum oscillations observed in YbB₁₂, while having little effect on the quantum oscillations in SmB₆, further informs our understanding of the itinerant low energy excitations involved in each of these materials and their approach to the neighbouring insulator-metal quantum critical point. Looking ahead, magnetic field dependent quantum oscillation measurements in other quantities, as well as complementary measurements such as thermal conductivity and nuclear magnetic resonance at low temperatures, are important in the quest to examine the character of the novel itinerant low energy excitations in YbB₁₂. Our results provide further impetus to the search underway for new theoretical paradigms to explain the unexpected discovery of a bulk Fermi surface in Kondo insulating materials [24, 25, 26, 27, 29, 30, 31, 32, 33, 40, 41, 42, 43].

Acknowledgments

We thank the team at the National Academy of Sciences of Ukraine, Kiev for assistance in the preparation of polycrystalline YbB_{12} as well as single crystals of LuB_{12} . We are grateful to Y. Matsuda for discussions of their unpublished measurements on YbB_{12} . We acknowledge valuable discussions with G. G. Lonzarich, T. Senthil, D. Chowdhury, I. Sodemann, and G. Baskaran. We are grateful for the experimental support provided by the NHMFL, Tallahassee,

including J. Billings, J. T. Camacho, R. Carrier, E. S. Choi, W. A. Coniglio, B. L. Dalton, D. Freeman, L. J. Gordon, M. Hicks, S. A. Maier, T. P. Murphy, J.-H. Park, J. N. Piotrowski, J. A. Powell, E. Stiers.

H.L, M.H., G.W., A.J.D, and S.E.S. acknowledge support from the Royal Society, the Leverhulme Trust through the award of a Philip Leverhulme Prize, the Winton Programme for the Physics of Sustainability, EPSRC UK (grant number EP/M000524/1) and the European Research Council under the European Unions Seventh Framework Programme (grant number FP/2007-2013)/ERC Grant Agreement number 337425. M.C.H. and G.B. acknowledge financial support from the EPSRC UK (grant number EP/M028771/1). M.D.J. acknowledges support for this project by the Office of Naval Research (ONR) through the Naval Research Laboratory's Basic Research Program. A portion of this work was performed at the National High Magnetic Field Laboratory, which is supported by National Science Foundation Cooperative Agreement No. DMR-1157490, the State of Florida, and the Department of Energy (DOE).

References

- [1] Tan B S, Hsu Y T, Zeng B, Ciomaga Hatnean M, Harrison N, Zhu Z, Hartstein M, Kiourlappou M, Srivastava A, Johannes M D, Murphy T P, Park J H, Balicas L, Lonzarich G G, Balakrishnan G and Sebastian S E 2015 *Science* **349** 287–290
- [2] Hartstein M, Toews W H, Hsu Y T, Zeng B, Chen X, Ciomaga Hatnean M, Zhang Q R, Nakamura S, Padgett A S, Rodway-Gant G, Berk J, Kingston M K, Zhang G H, Chan M K, Yamashita S, Sakakibara T, Takano Y, Park J H, Balicas L, Harrison N, Shitsevalova N, Balakrishnan G, Lonzarich G G, Hill R W, Sutherland M and Sebastian S E 2018 Nat. Phys. 14 166–172
- [3] Li G, Xiang Z, Yu F, Asaba T, Lawson B, Cai P, Tinsman C, Berkley A, Wolgast S, Eo Y S, Kim D J, Kurdak C, Allen J W, Sun K, Chen X H, Wang Y Y, Fisk Z and Li L 2014 *Science* **346** 1208–1212
- [4] Flachbart K, Gabáni S, Neumaier K, Paderno Y, Pavlík V, Schuberth E and Shitsevalova N 2006 *Physica B* 378-380 610–611
- [5] Gabáni S, Flachbart K, Konovalova E, Orendáč M, Paderno Y, Pavlík V and Šebek J 2001 *Solid State Commun.* **117** 641–644
- [6] Boulanger M E, Laliberté F, Badoux S, Doiron-Leyraud N, Phelan W A, Koohpayeh S M, McQueen T M, Wang X, Nakajima Y, Metz T, Paglione J and Taillefer L 2017 *arXiv preprint* arXiv:1709.10456
- [7] Kasaya M, Iga F, Takigawa M and Kasuya T 1985 J. Magn. Magn. Mater. 47-48 429-435
- [8] Hewson A C 1997 The Kondo Problem to Heavy Fermions (Cambridge University Press)
- [9] Sugiyama K, Iga F, Kasaya M, Kasuya T and Date M 1988 J. Phys. Soc. Jpn. 57 3946–3953
- [10] Bat'ková M, Bat'ko I, Konovalova E, Shitsevalova N and Paderno Y 2006 Physica B 378 618-619
- [11] Okamura H, Michizawa T, Nanba T, Kimura S i, Iga F and Takabatake T 2005 J. Phys. Soc. Jpn. 74 1954–1957
- [12] Ekino T, Umeda H, Iga F, Shimizu N, Takabatake T and Fujii H 1999 Physica B 259 315-316
- [13] Iga F, Kasaya M and Kasuya T 1988 J. Magn. Magn. Mater. 76-77 156-158
- [14] Iga F, Shimizu N and Takabatake T 1998 J. Magn. Magn. Mater. 177-181 337-338
- [15] Schlesinger M E 1998 J. Phase Equilib. Diffusion 19 49–55

- [16] Werheit H, Filipov V, Shirai K, Dekura H, Shitsevalova N, Schwarz U and Armbrüster M 2011 *J. Phys. Condens. Matter* **23** 065403
- [17] Blaha P, Schwarz K, Madsen G, Kvasnicka D and Luitz J 2001 wien2k: An augmented plane wave+ local orbitals program for calculating crystal properties Vienna University of Technology Institute of Materials Chemistry
- [18] Tran F and Blaha P 2009 Phys. Rev. Lett. 102 226401
- [19] Shoenberg D 1984 Magnetic Oscillations in Metals (Cambridge, UK: Cambridge University Press)
- [20] Landau L 1957 Sov. Phys. JETP 3 920–925
- [21] Iga F, Suga K, Takeda K, Michimura S, Murakami K, Takabatake T and Kindo K 2010 *J. Phys. Conf. Ser.* **200** 012064
- [22] Weng H, Zhao J, Wang Z, Fang Z and Dai X 2014 Phys. Rev. Lett. 112 016403
- [23] Antonov V N, Harmon B N and Yaresko A N 2002 Phys. Rev. B 66 165209
- [24] Anderson P W 1992 Phys. Scr. T42 11-16
- [25] Grover T, Trivedi N, Senthil T and Lee P A 2010 Phys. Rev. B 81 245121
- [26] Motrunich O I 2006 Phys. Rev. B 73 155115
- [27] Katsura H, Nagaosa N and Lee P A 2010 Phys. Rev. Lett. 104 066403
- [28] Knolle J and Cooper N R 2017 Phys. Rev. Lett. 118 096604
- [29] Chowdhury D, Sodemann I and Senthil T 2017 arXiv preprint arXiv:1706.00418
- [30] Sodemann I, Chowdhury D and Senthil T 2018 Physical Review B 97 045152
- [31] Coleman P, Miranda E and Tsvelik A 1993 Physica B 186-188 362–364
- [32] Baskaran G 2015 arXiv preprint arXiv:1507.03477
- [33] Erten O, Chang P Y, Coleman P and Tsvelik A M 2017 Phys. Rev. Lett. 119 057603
- [34] Pixley J, Yu R, Paschen S and Si Q 2015 arXiv preprint arXiv:1509.02907
- [35] Kagan Y, Kikion K and Prokof'ev N 1992 Physica B 182 201–208
- [36] Heinecke M, Winzer K N J K H, Grieb H, Flachbart K and Paderno Y B 1995 Z. Physik B 98 231-237
- [37] Harima H, Yanase A and Kasuya T 1985 J. Magn. Magn. Mater. 47–48 567–569
- [38] Okuda N, Suzuki T, Ishii I, Sayaka H, Iga F, Takabatake T, Fujita T, Kadomatsu H and Harima H 2000 *Physica B* **281–282** 756–757
- [39] Wolgast S, Eo Y S, Sun K, Kurdak, Balakirev F F, Jaime M, Kim D J and Fisk Z 2017 Phys. Rev. B 95 245112
- [40] Knolle J and Cooper N R 2015 Phys. Rev. Lett. 115 146401
- [41] Erten O, Ghaemi P and Coleman P 2016 Phys. Rev. Lett. 116 046403
- [42] Xu Y, Cui S, Dong J K, Zhao D, Wu T, Chen X H, Sun K, Yao H and Li S Y 2016 Phys. Rev. Lett. 116 246403
- [43] Zhang L, Song X Y and Wang F 2016 Phys. Rev. Lett. 116 046404