Layered Kondo lattice model for quantum critical $\beta$-YbAlB$_4$

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We analyze the magnetic and electronic properties of the quantum critical heavy fermion superconductor $\beta$-YbAlB$_4$, calculating the Fermi surface and the angular dependence of the extremal orbits relevant to the de Haas–van Alphen measurements. Using a combination of the realistic materials modeling and single-ion crystal field analysis, we are led to propose a layered Kondo lattice model for this system, in which two dimensional boron layers are Kondo coupled via interlayer Yb moments in a $J_z = \pm 5/2$ state. This model fits the measured single ion magnetic susceptibility and predicts a substantial change in the electronic anisotropy as the system is pressure-tuned through the quantum critical point.

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Heavy electron materials have played a long-standing role in strongly correlated electron physics, providing key insights into emergent quantum mechanical behavior that many hope can be scaled up in energy and temperature in actinide and transition metal compounds. One of the remarkable aspects of heavy fermion materials is that they can be tuned, by applying pressure or magnetic field at low temperatures, through a quantum critical point $\frac{\beta}{c}$, where they exhibit non-Fermi liquid behavior $[1, 2, 3, 4]$ and a marked predisposition towards superconductivity $[5, 6]$. The recent discovery of a layered Yb based heavy fermion material $\beta$-YbAlB$_4$ $[7]$ which is both stoichiometrically quantum critical and superconducting at $T_c = 80$ mK has attracted a great deal of interest. There are many examples of heavy electron superconductivity, often in close proximity to a quantum critical point $[1]$. However, although heavy electron behavior and quantum critical behavior have been observed in Ce, U and Yb materials $[5, 8, 9, 10, 11, 12, 13]$, $\beta$-YbAlB$_4$ is the first ytterbium based heavy fermion superconductor. This material raises many fascinating questions. Why, for example, is this system intrinsically quantum critical, with a specific heat coefficient that is finite in a magnetic field, but which diverges as the field is removed?

In this paper, we present an analysis of the magnetic and electronic properties of $\beta$-YbAlB$_4$. We use a combination of electronic structure calculations and crystal field analysis to develop a simple model for the low energy physics. Our results are consistent with the almost localized nature of the Yb $f$-electrons. In particular, we find that the electron effective mass, as measured by the low temperature specific heat coefficient $\gamma = C_v/T$, exceeds the band-structure value by more than a factor of 30. Moreover, in an LDA+U band-structure calculation, the system is found to develop an antiferromagnetically ordered ground-state, underlining the close vicinity to magnetic instability.

We have calculated the Fermi surface and the angular dependence of the extremal orbits relevant to the de Haas–van Alphen measurements. Despite the layered crystal structure, hybridization with the $f$-electrons gives rise to a three-dimensional Fermi surface. Combining the band-structure calculations and crystal field analysis, we are led to propose an anisotropic Kondo lattice description of this material, in which Yb magnetic moments at a site of seven-fold symmetry hybridize with neighboring boron planes, to give rise to the observed 3-dimensional band structure. We show that the crystal field configuration with maximum interlayer overlap, $|J = 7/2, m_J = \pm 5/2 \rangle$ (Fig. 1), accounts well for the anisotropy in the high temperature magnetic susceptibility.

The compound $\beta$-YbAlB$_4$ crystallizes in an orthorhombic structure (group Cmmm) $[14]$, with ytterbium and aluminium atoms sandwiched between boron layers. In this unusual structure the Yb atoms are centered between seven-member boron rings, as illustrated in Fig. 1, while the Al atoms are centered between pentagonal rings of boron. A central question raised by the structure, is whether the electronic structure will be quasi-two dimensional as in the case of intercalated graphite compounds. Our calculations indicate instead that the two-dimensional layers of boron are “short-circuited” by electrons of Yb and Al. The role of the Kondo effect in this setting is particularly intriguing and will be discussed below.

For the band structure calculations, a full potential linear

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**FIG. 1:** (Color online) Local coordination of Yb atom (large spheres, blue), suspended between two planes of boron heptagons (small green spheres) and surrounded in-plane by a distorted rectangular of Al atoms (red spheres). The proposed $|m_J = \pm 5/2\rangle$ ground state wavefunction is also shown, see Eq. 4. Boron conduction electrons $c^\dagger(\mathbf{r}_G)|0\rangle$ interact with Yb spins $S_j$ via layered Kondo lattice model Eq. 4.
The calculated LDA Fermi surface and the underlying Fermi surface topology [19, 20, 21]. The calculated LDA Fermi surface $\beta$-YbAlB$_4$ (see Fig. 3) contains two sheets (heavy bands 89 and 91) that are $f$-like in character, and a very small pocket (light band 93, not shown) associated with boron $p$-electrons. The topology of the heavy sheets is non-trivial, featuring a combination of parallel quasi-1D sheets that are connected by a cylindrical quasi-2D tube. For comparison with future de Haas-van Alphen (dHvA) experiments we have determined the angular dependence of the extremal orbits [22], shown in Fig. 3(b) for the the lightest bands with LDA-calculated $m^* < 10m_e$. The heavier bands, with larger frequencies, are unlikely to be observed in dHvA measurements due to the quasi-particle damping.

Experimentally, the low temperature thermodynamics of $\beta$-YbAlB$_4$ is dominated by the spin entropy of the Yb ions [7], indicating that this system is a Kondo lattice compound. We now determine the symmetry of the low lying magnetic dou-
blets and construct the corresponding Kondo lattice model. In $\beta$-YbAlB$_4$, the Yb ions are suspended between two seven-member rings of boron atoms (Fig. 1) giving rise to an approximate 7-fold symmetry. In this low symmetry environment, we expect the eight states of the $J = 7/2$ multiplet to split into four Kramers doublets. The approximate local symmetry group of the Yb ions is $C_7 \times C_{2v}$. The only crystal field opera-

FIG. 2: (Color online) Projected density of states (DOS), showing relative contribution of different atoms. The DOS at the Fermi level corresponds to the specific heat coefficient $\gamma \equiv C_v/T = 6.7$ mJ/(mol Yb$\times$K$^2$) per Yb atom.

augmented plane wave method (LAPW) was carried out using the WIEN2k code [15], using a Local Density Approximation (LDA) with a generalized gradient correction [16]. The strong spin-orbit coupling of the Yb atoms was taken into account using a scalar-relativistic approach [13, 17]. The calculated electron density of states (DOS) of $\beta$-YbAlB$_4$ is shown in Fig. 2. The Yb $f$-bands are split by the spin-orbit (SO) interaction into $J = 5/2$ and $J = 7/2$ multiplets separated by 1.4 eV, and are heavily hybridized with the lighter conduction electrons of B, and (to a lesser extent) Al atoms. The DOS at the Fermi level is dominated by the Yb $f$-bands, with a band mass around 5–6 $m_e$ in the (ab)-plane and about $m_e$ along the c-axis. The specific heat coefficient $\gamma_{\text{Yb}} = 6.7$ mJ/(mol$\times$K$^2$), extracted from the value of the DOS at $E_F$ contrasts with a measured value $\gamma_{\text{exp}} = \lim_{T \to 0} C_M/T \gtrsim 170$ mJ/(mol Yb$\times$K$^2$) at a field of 0.50 Tesla [7]. Experimentally, the zero temperature specific heat coefficient is field dependent, diverging as $B^{-1/4}$. On the basis of this comparison,

$$m^*(B) \approx \frac{25}{(B - B_c)^{1/4}},$$

where the critical field $B_c = 0$ within current experimental resolution. This divergent mass enhancement indicates that the Yb $f$-bands become localized in the zero field limit.

The LDA calculation predicts that the Yb ions are in a mixed valence state, with $n_f \approx 13.4$. This is clearly inconsistent with the susceptibility measurements, which reveal a Curie-Weiss susceptibility at high temperatures, with the full moment of the $4f^{13}$ state: a signature of local moment behavior. We attempted to cure this failure using the LDA+U method [18] where the Anderson $U$ is treated at the mean-field level for the localized Yb $4f$-orbitals. These calculations predict a magnetic ground state with an in-plane AFM arrangement of Yb spins and the effective moment of 0.6 $\mu_B$ per Yb ion. While no magnetic ordering is observed in the material, we may regard the LDA+U result as a mean-field description of strong underlying magnetic correlations.

Although LDA cannot capture the large mass renormalizations of strongly correlated systems, it generally provides a good guide to the underlying Fermi surface topology [19, 20, 21]. The calculated LDA Fermi surface $\beta$-YbAlB$_4$ (see Fig. 3) contains two sheets (heavy bands 89 and 91) that are $f$-like in character, and a very small pocket (light band 93, not shown) associated with boron $p$-electrons. The topology of the heavy sheets is non-trivial, featuring a combination of parallel quasi-1D sheets that are connected by a cylindrical quasi-2D tube. For comparison with future de Haas-van Alphen (dHvA) experiments we have determined the angular dependence of the extremal orbits [22], shown in Fig. 3(b) for the the lightest bands with LDA-calculated $m^* < 10m_e$. The heavier bands, with larger frequencies, are unlikely to be seen in dHvA measurements due to the quasi-particle damping.

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tors with both $2\pi/7$ rotational and time reversal symmetry are polynomial functions $P[J^2_\alpha]$ of $J^2_\alpha$ [23], so to good approximation, the magnetic doublets are eigenstates of definite $m_J$. Empirically, the observation of a Schottky peak in the specific heat at $T_{\Delta} \approx 30$–$40$ K, indicates that the lowest lying crystal field excitation has energy $\Delta \approx 2k_B T_{\Delta} \sim 60$–$80$ K.

To determine the symmetry of the lowest lying Yb doublet, we appeal to geometric considerations. The lowest energy magnetic doublet will correspond to the $4f$ wavefunction with maximum overlap with the boron rings. This rules out the states with $|m_J| = 1/2$ which are aligned along the c-axis, and the states with $|m_J| = 7/2$ which are planar, lying in the ab plane. The remaining contenders with $m_J = 3/2$ or 5/2 are “conical dumbbells” aligned along the c-axis. The seven-fold boron rings subtend an angle of 50° about the c-axis, closely corresponding to the aperture angle ($52°$) of the conical wavefunction of the $|\pm 5/2\rangle$ multiplet. We expect this state to have the maximum hybridization with the boron rings. The most likely crystal field excitation state is the mixture of $|\pm 1/2\rangle$ and $|\pm 3/2\rangle$ configuration, with the second-largest overlap. This simple model of the Yb ion provides a good fit to the experimentally measured magnetic susceptibility at high temperatures (Fig. 4), provided one fits the Curie–Weiss temperature $\theta_{CW} \sim 230$ K due to antiferromagnetic RKKY interaction between Yb moments. The best fit is obtained with the excited state being mainly $|\pm 1/2\rangle$, with a small admixture of the $m_J = \pm 3/2$ as indicated in the inset of Fig. 4 with $\gamma \sim 0.28$. This is likely due to crystal fields of Al atoms that break the seven-fold symmetry of the boron rings.

Given this single-ion picture, the simplest effective model for the low-energy degrees of freedom is the one in which low lying $|J = 7/2, m_J = \pm 5/2\rangle$ 4f doublet interacts with the neighboring layers of two-dimensional boron $p$-electrons via a Kondo exchange interaction. Based on our calculations, we expect the spins on neighboring Yb ions to interact via an in-plane antiferromagnetic RKKY interaction $J_{ij}$. This leads us to propose a “layered Kondo lattice model” for $\beta$-YbAlB$_4$,

$$H = H_0 + H_K + H_H, \quad \text{where}$$

$$H_0 = -\sum_{p=\pm\frac{\pi}{2},(m,n)\in(ab)} t_{m,n} c^\dagger_p(r_{mp})c_p(r_{np}),$$

$$H_K = J_K \sum_{\langle i,j \rangle \in(ab)} \left( \sum_{J,p=\pm} c^\dagger_{Jop} \sigma_{\alpha\beta} c_{Jp} + \sum_{J,p\neq p'} c^\dagger_{Jop} \sigma_{\alpha\beta} c_{Jp'} \right),$$

$$H_H = \sum_{\langle i,j \rangle \in(ab)} J_{ij} S_i \cdot S_j \quad (2)$$

where the $c^\dagger_p(r_{mp})$ operators create conduction electrons with spin $\sigma = \pm 1/2$ in the two dimensional boron planes at site $r_{m,\pm} = r_m \pm c/2$. The $S_i$ are the Yb spin operators, interacting with boron layers via the Kondo coupling $J_K$. The $\{c_{Jop}\}$ represent boron Wannier states in the $p$th layer projected onto Yb site $R_j$ in the $|\alpha = \pm \frac{5}{2}\rangle$ state, given by

$$c_{Jop} = \sum_{\sigma} \sum_{m=1}^7 \gamma^{5/2}_{\alpha\sigma}(r_{mp} - R_j) c_{\sigma}(r_{mp}), \quad (3)$$

where summation is over atomic positions of the seven-member boron ring above or below the Yb site. Here the form-factors $\gamma^{5/2}_{\alpha\sigma}(\hat{r}) = C_{\alpha\sigma}(r_{mp}) Y_{\alpha\sigma}(\hat{r})$, arise due to spin-orbit coupling [24], where $C_{\alpha\sigma}(r_{mp}) \equiv \langle \alpha - \sigma | J \frac{5}{2} \rangle$ are the Clebsch-Gordan coefficients for the $J = 7/2, l = 3$ state and $Y_{\alpha\sigma}(\hat{r})$ are spherical harmonics. Written explicitly

$$Y^{5/2}_{\alpha\sigma}(\hat{r}) = \frac{1}{\sqrt{7}} \begin{pmatrix} \sqrt{6} Y_{3,2}(\hat{r}) & Y_{3,3}(\hat{r}) \\ Y_{3,-2}(\hat{r}) & \sqrt{6} Y_{3,-3}(\hat{r}) \end{pmatrix}. \quad (4)$$

The angular probability distribution function of the $|\pm \frac{5}{2}\rangle$ doublet is given by $\sum_{\alpha\sigma} |\gamma^{5/2}_{\alpha\sigma}(\hat{r})|^2$ as shown in Fig. 1.

The above model (2) provides a starting point for future theoretical studies of $\beta$-YbAlB$_4$. Here we focus on the qualitative physics of the model. At high temperatures, local moments form on the Yb ions, which interact via RKKY interaction, whose scale is however too small to allow magnetic ordering. The high temperature electronic behavior is then governed mainly by the “small” Fermi surface formed out of the quasi-two-dimensional boron bands, coupled weakly along the c-axis via the interplanar aluminum atoms. As the temperature is lowered below the Kondo temperature $T_K \sim D e^{-D/2} J_K$ (here $D$ is the conduction electron band width), the quenched local moments will give rise to resonant bound-states that link neighboring boron layers, forming a three-dimensional Fermi surface of heavy quasiparticles.

The observation of field-tuned quantum criticality in this material indicates that the RKKY interactions and the Kondo temperature are of comparable size, estimated [2] to be of order of 100 K, a value substantially larger than other quantum critical systems, such as YbRh$_2$Si$_2$ [25] ($T_K \sim 25$ K) or CeCu$_6-y$Au$_y$ [2] ($T_K \sim 6$ K). The enhanced Kondo temperature suggests that this system balances a more mixed valent Yb ion with stronger RKKY interactions. Set against these observations, the tiny size of the superconducting transition temperature is quite mysterious, for a large magnetic interaction might be expected to induce stronger pairing [5].

A striking feature of this compound is the highly
anisotropic, low-symmetry structure. The layered Kondo model Eq. (2) is akin to a quantum dot symmetrically coupled to two leads [26], where at high temperatures or in an applied field the electrons are localized and the system is effectively Coulomb-blockaded. As the Kondo effect develops at low temperature, the interlayer channels opened up by the Yb Kondo resonances will lead to co-tunneling processes described by the second term in $H_K$, enhancing the c-axis conductivity. In $\beta$-YbAlB$_4$, neighboring boron planes are weakly coupled by the s,p electrons of the Al and Yb atoms, leading to a finite c-axis conductivity even in the absence of the Kondo coupling. We expect however that the interlayer Kondo effect will resonantly “short-circuit” the boron planes at low temperatures. In the light of the above discussion, the conventional scenario of the quantum critical point (1) would suggest an immediate drop in $c$-axis conductivity as the system is tuned into the magnetic phase by applied pressure.

The most puzzling feature of this material is the quantum critical behavior observed under ambient conditions. It is conceivable that the system is fine-tuned to a quantum critical point. In this case, pressure tuning will immediately drive $\beta$-YbAlB$_4$ into an antiferromagnetic phase, with a mass divergence following the form Eq. (1), but where $B_c(P)$ is now finite. In this scenario, the fact that $B_c = 0$ at $P = 0$ would be purely accidental. A more intriguing possibility is that the field-tuned properties of $\beta$-YbAlB$_4$ are signature of a robust quantum critical phase. In this case, the critical field would be expected to remain zero over a finite range of pressure, while the exponent $\alpha$ may vary with pressure, according to

$$m^*(B)/m_e \propto B^{-\alpha(P)}, \quad (5)$$

The possibility of critical metallic phases in the Kondo lattice has recently been put forward by Anderson [27]. The existence of a paramagnet over a range of pressures between the fully developed Fermi liquid and antiferromagnet has also been proposed in the context of “Kondo breakdown” models of quantum criticality [28,29], however in that case the intermediate phase is a non-critical spin liquid phase.

The existence of quantum critical point at zero field is actually well known in the context of the two impurity Kondo model [30,31], where a competition between the Kondo effect and valence bond formation between the local moments leads to non-Fermi liquid behavior. Could this phase be stabilized by the formation of a spin liquid in the Yb layers? This work was funded by NSF grant DMR 0605935. The authors are grateful to Zachary Fisk for bringing Ref. [14] to our attention. A.H.N. thanks P.M.C. Rourke and S. R. Julian for kindly providing the authors with computer code for Fermi surface analysis. Note added in proof: after this Letter was submitted, a report on de Haas–van Alphen measurements by O’Farrell et al. [32] has become available, in good agreement with our prediction of two heavy sheets of the Fermi surface.

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