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HEAVY-ELECTRON SUPERCONDUCTOR UPt_3

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Abstract

Local-density-approximation (LDA) calculations for the Fermi-surface extremal cross-sectional areas of UPt₃ are presented and compared to deHaas-van Alphen experiments of Taillefer et al. The topology of the calculated surfaces is in excellent agreement with experiment and allows a determination of the directional dependence of the anisotropic mass-renormalization factor. The source of this renormalization is briefly discussed.

Heavy-electron compounds form a remarkable class of materials with many unusual properties that often seem to defy conventional solid-state experience.¹ While an enormous effort, both experimentally and theoretically, has therefore been devoted to these systems, they are nonetheless still mysterious and poorly understood.² For example, no good criteria exists to predict which materials will become heavy or how heavy they will become. There is also no satisfactory explanation for why some heavy-electron compounds are superconducting, some are magnetic, and some are neither. There is no general agreement on the basic mechanism of the anomalous superconducting properties of these systems (see below, however) or the nature of the superconducting pairing (singlet or triplet). Indeed, it is not even known whether one general theory can be developed to explain all of these compounds or whether different physical mechanisms are necessary to explain the properties of different heavy-electron materials.

Despite all of these difficulties, there has been a general consensus that the large low-temperature specific-heat coefficients that characterize these systems as well as all of the other anomalous properties are a consequence of strong electron-electron correlations. This focus on the electronic structure of these materials has, however, not been without its own set of problems, especially since naive conventional interpretations immediately lead to other difficulties. For example, if the large specific heats were to be attributed to a large static (i.e., ground state) band-structure density of states (DOS), such a system would almost certainly distort by either a structural or magnetic phase transition in order to remove such an energetically costly configuration. Also, the presence of a high ground-state DOS would normally imply a very narrow band, which should lead to local-moment formation by electron-electron correlations. None of these expectations occur: the heavy effective masses remain to quite low temperatures and the electronic states appear to remain itinerant. We believe that the resolution to these problems is that

the electronic band structure gives a correct description of the zero-temperature ground state. Most experiments, however, cannot probe this state directly and instead mainly see large excitations about this ground state. Because the excitations are dynamical, there is no need for the system to structurally distort.

The most important part of the basic electronic structure is believed to consist of a narrow set of f states (either localized or itinerant) that are at or near the Fermi energy and are coupled to a wide conduction band. Attempts to experimentally probe the details of the electronic structure have met with difficulty, largely because many experiments are so physically complex that interpretation of the data is often prejudiced by the necessity for detailed theoretical models. Often several conflicting models are in equal agreement with the data from a given type of experiment or no attempt is made to interpret the data by removing obscuring experimental aspects of the measurement such as the presence of surfaces and other geometrical effects, energy-dependent cross-sections, final-state effects, and most importantly of all excited state contributions. Because these materials have such extraordinary low-temperature properties, many believe that electron-electron correlations are so important for these systems that ordinary electronic band-structure (EBS) or local-density approximation (LDA) calculations can give no useful information about their electronic structure. Such beliefs are almost always difficult to resolve because no experiment ever produces a pure unadulterated DOS to compare with band calculations. The problem with heavy electron compounds is that dynamical effects so dominate their low temperature properties that it is difficult to extract the static properties that the LDA is designed to calculate.

One experimental technique, x-ray photoelectron spectroscopy (XPS), in our opinion is sufficiently simple to allow meaningful extraction of information about the intrinsic electronic structure, since in this case the only corrections required are for the energy-dependent cross sections, which are reasonably easy to accurately

calculate. The XPS data for an important compounds in this class of materials, viz. UPt_3 , has been obtained and is in excellent agreement³ with band-structure results. This suggest that the uranium f electrons are genuinely itinerant in UPt_3 . Nonetheless, because the heavy-electron state seems to form at relatively low temperatures and the experiments were done at higher temperatures than this as well as the poor spectrometer resolution in XPS experiments, such agreement is hardly a definitive test.

With the deHaas van Alphen (dHvA) data of Ref. 4a and the preceding paper [4b], we finally have an extremely sensitive and definitive comparison between experimental electronic structure and band-structure results for UPt_3 . This, of course, is in addition to the importance of observing dHvA signals at all, since such signals are a crucial demonstration that a well-defined Fermi surface exists, even despite the presence of the strong electron-electron correlations that are believed to cause the large specific-heat enhancements. While the density-functional Fermi surface has never been shown to be equivalent to the true quasiparticle surface, the difference is known to be small in free-electron-like metals.⁵ Unless the LDA is not valid for the system in question, one therefore expects the Fermi surface to be reproduced to a good approximation.

Besides the Fermi surface topology, one can also extract the effective masses of the Fermi surface quasiparticles from dHvA data. These can differ substantially from the LDA band masses if large dynamical correlations are present, since "excited state" effects are not included in an LDA theory. Thus, if one finds agreement of the LDA surface with experiment, comparison of the experimental masses with the LDA masses gives an estimate for the orbit-averaged dynamical mass renormalizations and their variation over different directions in the Brillouin zone.

In Ref. 6 it was shown that many different types of EBS calculations were all in agreement with each other and furthermore were in agreement with the limited dHvA data then available. However, dHvA experiments only provide topological Fermi surface information about the magnitude of extremal orbital areas perpendicular to the magnetic field. For only one measured direction, which was all that was available before the data of the preceding paper, it was possible that the agreement in Ref. 6 was purely fortuitous and also that the correspondence between the theoretical orbits and measured areas were incorrectly assigned. With the availability of data for the additional directions provided in the preceding paper, it is now unambiguously possible to follow extremal orbits in different directions and accurately test the theoretical Fermi-surface predictions. The excellent agreement with our calculations indicates that our calculated bands are correct to within a few millirydbergs.

The LDA band structure for UPt₃ that we use in our analysis is obtained from a fully relativistic (Dirac equation) linearized muffin-tin orbital calculation with combined correction terms (LMTO-CC). The latter terms correct for the non-spherical shape of the unit cell, and represent an improvement over the commonly employed atomic-sphere approximation (ASA). The details of this calculation are essentially identical to those of an earlier ASA calculation that are given in Ref. 7. The a priori self-consistent calculations require only the atomic nuclear charges of the atoms (Z-numbers) as input. After achieving self-consistency, eigenvalues were generated at 126 k points in the irreducible wedge of the Brillouin zone. the bands were then fit with a Fourier spline series utilizing 330 star functions.⁸

In Figure 1, we show the Fermi-surface cross sections in various symmetry planes. Five bands form the surface. Band 1 forms a disk centered about the A point. Band 2 also forms A-centered disks, but with interconnecting arms in the MKLH planes. Bands 3, 4, and 5 form closed surfaces about the Γ point; Band 3 also has an

additional closed surface about the K point. If the Fermi energy is raised a few mRy, then the band 3 Γ and K pieces become connected. This topology is the same as that found from a linearized augmented plane wave (LAPW) calculation,⁶ and differs only slightly from that of an earlier LMTO-ASA calculation which had rings instead of disks.^{6,9} Our computed areas for bands 1 and 2 are shown in Figures 2, and for bands 3, 4, and 5 in Figure 3.

When compared to the experimental data of the previous paper,^{4b} the same two topologies are found. The orbit α is around the arm of the band 2 disk, the orbit β is an L centered orbit exterior to the band 2 arms, the orbit λ is around the band 2 ~~disk~~ ^{disk} ~~surface~~, the orbit δ is around the band 1 disk, the orbit ξ is around the band 3 K centered surface, the orbit ϵ is around the band 4 surface, the orbit ω is around the band 3 Γ centered surface, and the orbit γ is around the band 5 ~~disk~~ ^{surface}. The orbits θ , ϕ , and ψ represent magnetic breakdown orbits between the band 1 and band 2 disks (not shown). One notes, though, that although some orbits like α end at angles predicted by theory, other orbits exist over shorter angular ranges than theory predicts. For the δ orbit, the signal loss is due to the rapid variation of area with angle, whereas for the β , λ , and ϵ orbits, the source of the signal loss is unknown. With current resolution, it is difficult to tell whether the short angle range of the ξ and ω orbits are due to signal loss or to the formation of a neck between the band 3 Γ and K surfaces.

The unambiguous assigning of orbits allows us to estimate the relative uncertainty in the bands. To do this, we adjust the Fermi energy until the orbit area agrees with experiment. These results are shown in Table I. From this table, one can see that the bands responsible for the large area orbits are accurate to within 1 mRy. The band 2 arms, though, are too large, and the band 1 disk too small. Also listed are the band masses compared to their experimental analogs. The resultant mass renormalizations are large, and have a significant momentum component, with

$m^*/m_b^{\wedge}$ varying from 12 to 23 across the zone. There is some theoretical uncertainty in the band-structure masses since they are derivative quantities and hence sensitive to the spline fit. For example, the LAPW calculation analyzed in Ref. 6, which has a Fermi surface in good agreement with our LMTO-CC results, had different band masses for some of the orbits. Nonetheless, the accuracy is sufficient to demonstrate the large mass-renormalization anisotropy over different directions in the Brillouin zone.

In rare-earth systems where the f electrons form local moments, a correct Fermi-surface model requires that the f electrons be treated as core (or localized) states. To test this for UPt_3 , two calculations were performed which forced the f electrons into the core with occupation numbers of 2 and 3 (with the f states removed from the basis for the band states). A Fermi-surface plot for the calculation with the occupation of 2 is shown in Figure 4. One can easily see that this surface is in gross disagreement with experiment. In particular, it has tubes open along the c axis which are not observed.

The agreement of the LDA Fermi surface with experiment has several important implications: First, the ground-state f electrons are itinerant and hybridize with other conduction-band electrons. this can be contrasted with the related compound UPd_3 , which seems to be a local-moment compound and hence agrees with dHvA data only when the f electrons are treated as localized states.¹⁰ Second, the LDA is probably a reasonable approximation to the true density functional for this system. In fact, we believe that the high accuracy for our UPt_3 Fermi surface relative to that typically achieved for normal transition metals is because the large direct Coulomb interaction on the Uranium sites tends to "lock" the f-bands to the Fermi energy such that there is an occupation number of 2-3 f electrons per uranium atom; thus there appears to be little sensitivity to the choice of exchange-correlation potential.

Additional corroboration for our view of the electronic structure of UPt_3 is provided by a recent quantitative moment-fluctuation theory that seems to explain the size of the mass renormalizations as well as the observed superconducting transition temperature.¹¹ Experimental evidence that moment fluctuations are the most important low-temperature excitation process is provided by the presence of an additional $T^3 \ln T$ term in the specific heat¹ as well as by the moment-fluctuation spectrum observed in neutron scattering experiments.¹² Because the Fermi surface geometry is a crucial input to the moment-fluctuation theory, the elucidation of the Fermi surface via dHvA and band structure calculations is leading to a quantitative understanding of heavy-electron electronic structure.

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FIGURE CAPTIONS

- Figure 1. Fermi surface plots in the symmetry planes from the LMTO-CC calculation. (All figures are for the calculated Fermi energy.)
- Figure 2. Extremal areas versus applied field direction for band 1 (dashed line) and band 2 (solid line).
- Figure 3. Extremal areas versus applied field direction for band 3 (solid line), band 4 (dashed line), and band 5 (dotted line).
- Figure 4. Fermi surface plots in the symmetry planes from the f core calculation.

Table I. Comparison of experimental to band masses for UPt_3 . The first column is the orbit identification, the next two columns the experimental areas (MG) and masses, the next two columns the theoretical areas and masses, and the final column the mass renormalization. The numbers in () are the shifts of E_F in mRy necessary to get the best fit between theoretical and experimental areas.

Orbit	Area	Mass	Area	Mass	m^*/m_b
a axis					
ML2	5.1	25	5.3(+2.0)	1.6	16
Γ_5	7.3	40	7.1(-0.5)	1.8	22
K3	14.2	50	13.6(+0.0)	3.7	14
Γ_4	21.2	60	21.3(-0.5)	4.8	13
Γ_3	59.2	90	59.0(+1.0)	5.3	17
b axis					
ML2	4.1	15	3.9(+2.5)	1.3	12
A1	12.3	30	12.0(-3.0)	1.3	23
A2	25.1	50	26.2(+0.0)	4.2	12