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Fermi surface of UPt_3 within the local-density approximation

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The Fermi surface of UPt_3 is constructed from three different local-density band calculations, one utilizing a linearized muffin-tin orbital approach in the atomic-sphere approximation (LMTO-ASA), the second an LMTO with additional combined correction terms (LMTO-CC), and the third a general potential linearized augmented plane-wave (LAPW) approach. The topology of the three surfaces are similar, except for the sheets coming from the lowest two bands which are slightly different between the cases. These surfaces are then compared to the experimental de Haas–van Alphen (dHvA) results of Taillefer *et al.* This comparison indicates that the Fermi surface of the LAPW and LMTO-CC calculations can be put in quantitative agreement with experiment with adjustments of the bands of only ~ 2 mRy. More adjustments are needed in the LMTO-ASA case. There are still some possible ambiguities in assigning certain theoretical extremal orbits to experimental data which call for further angle-dependent dHvA experiments. Nevertheless, these results clearly indicate that the local-density energy bands give an accurate description of the Fermi surface and are an important ingredient in understanding the anomalous properties of UPt_3.

The electronic nature of the heavy-fermion superconductor UPt_3 has generated much controversy recently. In particular, it has been questioned whether anything can be learned from performing "one-electron" calculations on such a highly correlated system. It is our contention that there are two questions to be addressed here. The first question is the nature of the superconductivity in this system, and whether band calculations can reflect on this point. We will not address this question in this Rapid Communication, but will refer the reader to two different conclusions obtained from band calculations on this matter.[1,2] The second question, though, is actually of a more fundamental nature. Can band calculations be useful as a first-principles basis for the description of heavy-fermion properties in the normal state? This is a pertinent question, because density functional theory (DFT) is designed to obtain correct ground-state properties, and we wish to know to what extent the local-density approximation is adequate for heavy-fermion materials. This is the question we address here.

Because of the apparent large renormalizations and anomalous temperature dependences of thermodynamic and transport properties in heavy-fermion systems, it is difficult to find experimental results to directly compare with band calculations. It must be remembered that these ground-state calculations do not include any renormalization effects involving frequency derivatives of self-energy terms, and thus the validity of such calculations cannot be judged on the basis of specific-heat data alone. In fact, one would expect that the density of states from a band calculation would be modest. The reason is that all heavy-fermion systems are either paramagnets, weak magnets, or low-temperature superconductors. This implies that the Stoner criteria is either not exceeded or just weakly exceeded in these systems. Hence, the band density of states must be of moderate value. In practice, this is realized from the local-density calculations.

Recently, though, de Haas–van Alphen (dHvA) data have been obtained on both UPt_3 (Ref. 3) and $CeCu_6$.[4] This is of extreme importance since such experiments measure the Fermi surface topology and quasiparticle effective masses directly. Although the Fermi surface is not a ground-state property, it is very close to one.[5] Obtaining a correct Fermi surface topology is a good indication of the validity of the band calculation. This in turn allows one to characterize the mass enhancements in the system by a comparison of band to experimental dHvA masses. In this paper, local-density band calculations per-

formed using three different methods are described for UPt_3, and details for the extraction of a Fermi surface are presented. We also compare the topology of the three calculated surfaces with the recent dHvA data. In addition, some speculations and conclusions are offered regarding heavy-fermion UPt_3.

The two theorems by Hohenberg and Kohn[6] allow us to find, in principle, the ground state of an interacting system of electrons by minimization of an energy functional involving the single-particle density. In practice, such a functional is difficult to construct, so Kohn and Sham[7] introduced an approximate functional known as the local-density approximation (LDA). Such an approximation assumes that locally the system behaves like an electron gas (the appropriate functional is then based on many-body calculations for the electron gas). One would naively assume that such an approximation is too drastic to have any meaning, but in practice it works extremely well. It has thus provided the foundation for most electronic structure calculations done on extended systems. For systems involving electrons that are close to being localized, though, one might expect such an approximation to fail because an electron gas functional operating on extended Bloch functions may not work in this case. Although the f electrons in heavy-fermion metals are itinerant at low temperatures, their wave functions are spatially localized. Hence, it is important to perform local-density calculations to test the adequacy of the LDA for these systems.

UPt_3 is a key system, since although it has an enormous specific heat coefficient (420 mJ/mole K^2), it shows similarities to less enhanced systems like UAl_2 and $TiBe_2$, particularly the presence of a $T^3 \ln T$ term in the specific heat which has been attributed to itinerant spin fluctuations.[7] It also shows a resistivity whose temperature dependence is also more typical of normal metals, rather than the Kondo-like behavior seen in heavier compounds such as UBe_{13}.[8] In fact, one might (naively) regard UPt_3 as an enhanced version of Pd, which has given rise to all the discussion on possible triplet superconductivity. Because of these reasons, much of the attention of band theorists has been focused on UPt_3.

Some of the first calculations to be performed on this metal[1,9] were done using the linearized muffin-tin orbital in the atomic-sphere approximation (LMTO-ASA) method.[10] The advantage of this method is its speed as compared to more accurate methods such as the linearized augmented plane wave (LAPW) one. This is an important factor since UPt_3 has eight atoms per unit cell. The LMTO-ASA calculation reported here is that of Ref. 9 which is very similar to that of Ref. 1. The calculation is for the observed $SnNi_3$ structure with $a=10.892$ a.u. and $c=9.258$ a.u. (equal radii Wigner-Seitz spheres were employed). A fully relativistic code was employed and the electron gas functional used was that of von Barth and Hedin.[11] After convergence of the self-consistent calculation utilizing 84 k points in the irreducible wedge, a final iteration was done at 126 k points. These bands were then fitted using a Fourier series spline technique[12] involving 330 star functions, and a density of states determined by the linear tetrahedron method.[13] The Fermi surface was then traced using this spline fit;[14] extremal orbits on the surface were identified whenever the line integral of dA/dk is zero, where A is the area enclosed by the orbit, and k is along the magnetic field direction (perpendicular to the orbit). The mass is then determined by the line integral of dA/dE. After this, a new calculation was performed including the so-called combined correction (CC) terms.[10] These are correct for the nonspherical shape of the atomic polyhedra, and for the truncation of the partial-wave expansion at finite maximum angular momentum. The analysis for this calculation (LMTO-CC) was the same as for the LMTO-ASA case.

The LAPW method involves a more accurate calculation at a more substantial cost. The LMTO-ASA method approximates the system by overlapping atomic spheres, with the potential being treated as spherical inside each muffin tin. The LMTO-CC method corrects for the overlaps, but still uses a spherical potential. In the LAPW method described here though, one uses nonoverlapping spheres, with nonspherical terms being kept both outside and inside the spheres. In this calculation,[2] lattice constants of $a=10.870$ and $c=9.239$ a.u. were used. Self-consistency was obtained using a special 21-point mesh in the irreducible wedge including a temperature broadening factor of 2 mRy. The functional employed was that of Hedin and Lundqvist,[15] and also included transverse exchange corrections.[16] Much speed was gained by treating the spin-orbit correction as a second variational step within the self-consistency cycle.[17] This approximation works well for uranium systems, although it does introduce some error for the platinum $6p$ states. On the final iteration, bands were generated at 84 k points. The fit procedure to the bands and the Fermi surface tracing are then done as described above. As one can see, the details of the three calculations are quite different, and should thus give us a feel for how sensitive the results are to different types of approximations.

In Fig. 1, we show the six bands in the vicinity of the Fermi energy (E_F) from the three calculations. As a guide to the reader, we include Fermi surface plane plots for the LAPW case in Fig. 2. The bands look very similar, except for one point. At A, bands 1 and 2 are above E_F in the LAPW and LMTO-CC calculations, but below E_F in the LMTO-ASA one. This means that the Fermi surface topology for these two bands is slightly different between the cases. In the LMTO-ASA case, band 1 forms a ring centered about A. For band 2, this ring has large arms attached to it along the A-L directions, the arms interconnecting the rings in the MKLH planes. (This surface is essentially equivalent to that of Ref. 1.) In the other cases, band 1 forms a saucer with center at A and bulged about A with small holes poked in it along the A-H directions. The band-2 surface is similar to that in the LMTO-ASA case, except that the hole in the ring along the Γ to A direction has been replaced by an inward bulge. For band 2 in all cases open orbits exist. For bands 3, 4, and 5, the topologies are all the same. These three bands form closed electron surfaces around Γ, and band 3 also forms a closed surface around K.

In Table I, we show the areas and masses of the extremal orbits for the theoretical calculations for field directions along the $\langle 010 \rangle$ axis in reciprocal space (this

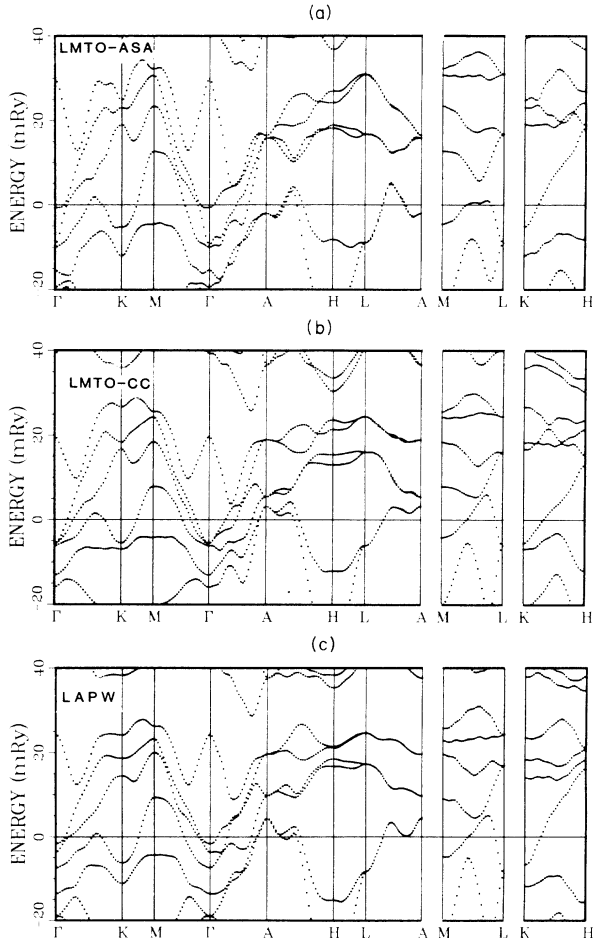

FIG. 1. Energy dispersions of the six bands in the vicinity of the Fermi energy for (a) the LMTO-ASA, (b) the LMTO-CC, and (c) the LAPW calculations. The Fermi energy is marked by a straight line.

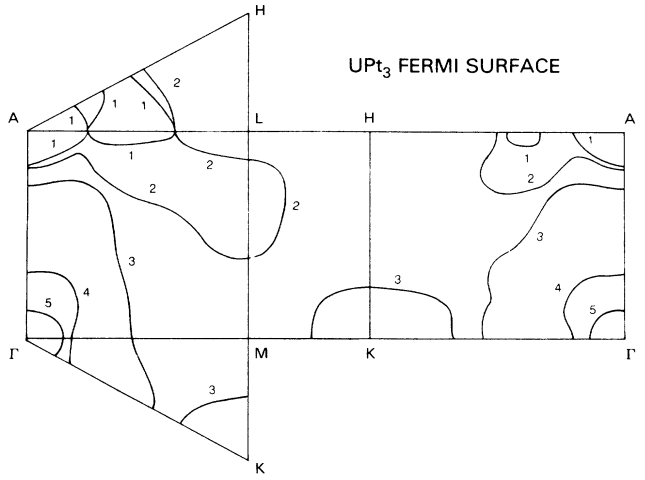

FIG. 2. Fermi surface plots for the LAPW calculation in the high-symmetry planes.

corresponds to the ⟨100⟩ axis in real space). The orbits are labeled by the position of the center of the orbit and the band (note that the band-3 electron surface around K does not have its orbit centered at K). Comparison to the experimental dHvA data (given in Table II) shows that there is fairly good agreement with the experimental data, especially in the LMTO-CC and LAPW cases. To obtain better results, one can try moving the Fermi energy and finding the resultant new extremal areas (this is equivalent to moving the bands). It should be noted that the orbital assignments chosen are tentative because of the lack of experimental data along other field directions. In Table II, we show the experimental data in the first column. In the next two columns, the corresponding theoretical data are shown, with the number in parentheses being the direction the Fermi energy has to be shifted to obtain that value. As can be seen, the orbital assignments of the calculations are the same, except for the three small area orbits which are rather close in size, and thus hard to distinguish. It is interesting to note that all the calculations show only seven closed orbits in agreement with experiment (the LMTO-ASA shows an eighth orbit which is open in the other two calculations; its high mass though, would have made it difficult to observe). As one can see from this table, some shifting around has to be done to get the LMTO-ASA results to agree with experiment, but not much for the other two cases. For the LAPW case, it can be concluded that if the electron surfaces are moved downwards ~2 mRy, quantitative agreement can be achieved with experiment. In the LMTO-CC

TABLE I. Extremal areas and masses for the three calculations evaluated at the calculated Fermi energy. The first column gives the center for the orbit and identifying band, the second the area in megagauss, the third the electron mass (a negative number indicates a hole orbit). The field is along the ⟨010⟩ axis in reciprocal space (⟨100⟩ in real space).

LMTO-ASA			LAPW			LMTO-CC		
Band	Area	Mass	Band	Area	Mass	Band	Area	Mass
ML-2	3.35	−1.09	*ML*-2	6.92	3.36	*ML*-2	10.42	2.15
AL-1	2.88	−0.80	*A*-1	9.01	−1.62	*A*-1	9.07	−1.90
L-2	7.54	1.18	*L*-2	5.72	0.79	*L*-2	5.23	1.01
L-2	49.66	−8.05	*L*-2	open		*L*-2	open	
Γ-3	41.62	3.74	Γ-3	53.26	4.46	Γ-3	52.78	5.34
MK-3	10.89	2.42	*MK*-3	12.98	3.52	*MK*-3	13.58	3.74
Γ-4	15.76	2.98	Γ-4	11.85	3.34	Γ-4	24.03	4.63
Γ-5	1.58	1.41	Γ-5	3.12	1.29	Γ-5	8.19	1.97

TABLE II. Extremal areas and masses for the three calculations compared to the experimental dHvA data. The number in parentheses indicates the adjustment to the Fermi energy necessary to obtain the quoted value (in mRy).

Expt.		LMTO-ASA			LAPW			LMTO-CC		
Area	Mass	Band	Area	Mass	Band	Area	Mass	Band	Area	Mass
5.1	25	*ML*-2	3.35(0)	−1.1	*ML*-2	6.92(0)	3.4	*ML*-2	5.27(+2)	1.6
6.0	· · ·	*AL*-1	5.05(−2)	1.1	*L*-2	5.72(0)	0.8	*L*-2	5.23(0)	1.0
7.3	40	Γ-5	4.58(+2)	1.4	Γ-5	6.54(+2)	2.0	Γ-5	8.19(0)	2.0
13.7	· · ·	*L*-2	7.54(0)	1.2	*A*-1	12.41(−1)	2.7	*A*-1	11.19(−1)	1.7
14.2	50	*MK*-3	14.12(+1)	3.1	*MK*-3	12.98(0)	3.5	*MK*-3	13.58(0)	3.7
21.2	60	Γ-4	19.60(+1)	3.6	Γ-4	19.60(+2)	3.5	Γ-4	24.03(0)	4.6
59.2	90	Γ-3	50.66(+2)	4.0	Γ-3	58.27(+1)	4.1	Γ-3	59.01(+1)	5.3

case, adjustments again of only 2 mRy are needed.

Also shown in Table II are the dHvA masses and the calculated band masses. The dHvA mass is related to the band mass by the factor $(1+\lambda)$, where λ is the energy derivative of the (unknown) heavy-fermion self-energy. Note that this factor is not only very large, but it is also not a constant throughout the Brillouin zone, just as found for mixed valent $CeSn_3$.[18]

In conclusion, it appears that there is encouragingly good agreement with experiment, especially in the LAPW and LMTO-CC cases. It is rather striking that such agreement can be attained with adjustments of the local density bands of only ~2 mRy. Much larger adjustments than this are needed for transition metals like niobium, for instance.[19] It is also pleasing that both the LMTO-CC and LAPW calculations predict essentially the same Fermi surface topology. Even the LMTO-ASA surface differs in only one aspect from the other two surfaces. Angle dependent dHvA data, though, are needed to allow a complete and unambiguous mapping of the Fermi surface.

The most important result of this study is that good quantitative agreement can be achieved with the experimental data with only minor adjustments of the local-density Fermi surface. This agreement was also previously found for mixed valent $CeSn_3$.[18] In the case of UPd_3 though, agreement was found with the experimental data only when the *f* electrons were artificially localized into the core.[20] This indicates that there is a sharp breakdown in the local-density Bloch description of the *f* electrons in that case, but that UPt_3 is still in the itinerant regime. The clear implication is that local-density studies are a useful basis to start from in unraveling the anomalous properties of heavy-fermion metals.

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