Origin of heavy quasiparticles in UPt$_3$

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We propose a microscopic description of heavy fermions in UPt$_3$. It is based on the assumption that two of the three 5f electrons of the U ions are localized. Band-structure calculations based on this supposition reproduce the observed de Haas-van Alphen frequencies very well. The observed enhancement of the quasiparticle mass as compared with the LDA band mass results from the local Coulomb interaction of the delocalized f electrons with the localized ones. Diagonalization of the Coulomb interaction matrix yields the level scheme of the localized $f^2$ states. Assuming a splitting of the ground-state doublet by the crystal field of similar size as in UPd$_2$Al$_3$ results in a mass enhancement factor of order 10 in agreement with experiments.

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The intermetallic compound UPt$_3$ is a well known heavy fermion system [1]. The Sommerfeld $\gamma$ coefficient of the linear low-temperature specific heat is strongly enhanced, i.e., $\gamma = 420$ mJ/(mole $\cdot$ K$^2$) and so is the Pauli-like spin susceptibility $\chi_S$. Both findings can be explained by attaching a large effective mass $m^*$ to the quasiparticles, i.e., one which is by a factor of order 20 bigger than the band mass $m_0$ obtained from local density approximation (LDA) calculations for the electronic structure [2,3]. Indeed, heavy quasiparticles have been observed in de Haas-van Alphen (dH-vA) experiments [1,4]. These experiments unambiguously confirm that UPt$_3$ has to be regarded as a strongly correlated Fermi liquid. Although a complete picture of the low temperature phase of UPt$_3$ has emerged, a comprehensive theoretical picture of the heavy quasiparticles is still missing. It is generally accepted that the latter are derived from the U 5f states. The number of itinerant U 5f electrons as well as the microscopic mechanism yielding the high effective masses are still controversial. It has been considered a success of the LDA that the dH-vA frequencies could be related to extremal orbits on the Fermi surface obtained by band-structure calculation which treats the U 5f states as itinerant. From these findings, however, one should not conclude that the U 5f states are ordinary band states which can be described by conventional electronic structure calculation. As pointed out above, the calculated energy bands are too broad for explaining the effective masses. Moreover, they are too small in order to fit the photoemission data [5]. The latter shows a broad peak just below the Fermi energy $E_F$. It is quite different from the data of heavy fermion systems involving Ce$^{3+}$ instead of U ions, such as CePt$_3$. Here a small Kondo resonance is observed near $E_F$ together with a broad 4f peak approximately 2 eV below that energy [6,7]. We add that the photoemission data near $E_F$ of UPt$_3$ resembles that of UBe$_{13}$ and UPd$_2$Al$_3$, two other heavy-fermion systems. It is also similar to that of UPd$_3$, which has localized 5f electrons except that in this case there is no f weight left at $E_F$. From the above it follows that a more microscopic understanding of the heavy-quasiparticle excitations is highly desirable. It seems clear that the Kondo effect as well as more exotic versions of it [8] can be excluded for UPt$_3$, for various reasons.

There are indications that U ions may have two types of f orbitals, i.e., with localized and with delocalized electrons, depending on the degree of hybridization. For example, quantum chemical calculations for uranocene U(C$_8$H$_8$)$_2$ have demonstrated this [9]. They have shown that the system has low-lying excitations which are due to local rearrangements of the 5f electrons. Also neutron inelastic scattering experiments on UPd$_2$Al$_3$ point towards the existence of localized as well as delocalized 5f electrons in that material [10]. Note that the susceptibilities of UPt$_3$ and UPd$_2$Al$_3$ look very much alike. These findings are the basis of our theory.

The aim of the present letter is to suggest that in UPt$_3$ we deal with localized as well as delocalized 5f electrons and that the coupling of the two subsystems results in heavy fermions. The mass enhancements of the quasiparticles are shown to follow from the local exchange interaction of the delocalized 5f electrons with localized $f^2$ configurations. The situation resembles that in Pr metal where a mass enhancement of the conduction electrons by a factor of 5 results from virtual crystal field (CEF)
excitations of localized $4f^2$ electrons [14]. As shown below, the theory suggested here allows for an explanation of the observed dH-vA frequencies. The attained accuracy is similar, if not better than that of previous LDA band structure calculations, although the Fermi surface shows distinct differences in the two cases.

In the following we consider two of the $5f$ electrons as localized, in agreement with the absence of any Kramers doublets in cases where a CEF splitting of U states has been observed. We put the localized $f$ electrons into $5f \ j = \frac{5}{2}$ orbitals, with $j_z = \pm \frac{5}{2}$ and $\pm \frac{3}{2}$. This choice is also consistent with the observed ground state, e.g., of UPd$_2$Al$_3$ [15,16]. The $5f \ j = \frac{3}{2}, j_z = \pm \frac{3}{2}$ states, on the other hand, are treated as itinerant band electrons. This yields the observed dH-vA frequencies when subsequently a band structure calculation is done.

A plot of the calculated dH-vA frequencies and of the observed ones is shown in Figure 1. We should like to emphasize that the theoretical data were obtained without any adjustments of the bands.

The Fermi surface from which the data are derived is displayed in Figure 2. It is formed by two bands which are doubly degenerate and which are derived from the $5f \ j = \frac{5}{2}, j_z = \pm \frac{3}{2}$ states.

![Figure 1](image1.png)

**FIG. 1.** DeHaas-vanAlphen cross sections as calculated within the present theory and from experiment (Kimura et al). Green and blue triangles correspond to the first and second band, respectively. The labelling indicates the origin of the orbit. Branches without labels are derived from those extremal areas of the lower band which are not centered on symmetry points.

![Figure 2](image2.png)

**FIG. 2.** Fermi surfaces calculated assuming that one of the uranium $5f$-electrons ($j=5/2, j_z=\pm 3/2$) is included in the Fermi surface volume while the remaining two are treated as localized. Two bands are contributing to the the Fermi surface. Calculated Fermi surface (a) from band 1, (b) from band 2. The small $\Gamma$-centered hole surface derived from band 2 is not displayed here. The symmetry points of the Brillouin zone are indicated in (a).

The first band gives rise to a pancake-like hole surface which is centered at $A$. The surfaces in neighboring cells are connected by arms along $AL$. These features imply the existence of open orbits for magnetic fields pointing along the $a$-direction as suggested by magnetoresistance data [17]. The second band intersects the Fermi surface several times giving rise to a $\Gamma$-centered hole ellipsoid which is enclosed by a large closed $\Gamma$-centered electron surface. The latter is strongly anisotropic in the basal plane. Finally, there are closed ellipsoids centered at $H$. The assignment of the observed quantum oscillation frequencies to extremal orbits on this Fermi surface is as follows: The thermodynamically most important orbit $\omega$ is assigned to the $\Gamma$-centered strongly anisotropic electron surface. The variation with magnetic field direction of the dH-vA frequencies is quantitatively reproduced. Of particular interest is the spitting obtained in the $a$-$c$ plane. This feature which is also found in LDA calculations has been confirmed by recent experiments [17].

The observed effective masses and calculated band masses on this sheet are listed in Table I for selected directions of the magnetic field.

| Magnetic Field | $m_b/m_0$ (LDA) [7] | $m_b/m_0$ (present theory) | $m_b^\ast/m_0$ (exp) [17] |
|----------------|-------------------|---------------------------|---------------------------|
| $H \parallel c$ axis | 5.09 | 17 | 110 |
| $H \parallel a$ axis | 5.79 | 6 and 9.5 | 82 |
| $H \parallel b$ axis | 6.84 | 14 | 94 |
The enhancement of the observed effective masses \( m^* \) over the ones calculated within the present theory \( m_b \) is of the order of 10. It can be quantitatively explained by the interaction of the itinerant 5f-states with the localized f\(^2\) configuration. The ratio \( \frac{m^*}{m_b} \) is obtained in analogy to Ref. [4] resulting in the equation

\[
\frac{m^*}{m_b} = 1 + 4a^2 N(0) \frac{2|M|^2}{\delta} .
\]  

(1)

Here \( N(0) \) is the density of states at \( E_F \) as obtained from our band structure calculation. The prefactor \( a \) is the 5f-weight per spin and U atom of the conduction electron states near \( E_F \).

The band structure calculations have been performed with appropriate numbers put into Eq. (1) we find \( m^*/m_b \approx 10 \). Here \( N(0) \) is the density of states at \( E_F \) as obtained from our band structure calculation. The prefactor \( a \) is the 5f-weight per spin and U atom of the conduction electron states near \( E_F \). The matrix element M describes the transition between the ground-state singlet and the low-lying excited singlet state at energy \( \delta \) of the localized f\(^2\) subshell in the presence of a CEF. With appropriate numbers put into Eq. (1) we find \( m^*/m_b \approx 10 \).

In the following, we present the details of the calculations. Band-structure calculations have been performed starting from the self-consistent LDA potentials but excluding the U 5f \( j = \frac{5}{2} \), \( j_z = \pm \frac{5}{2} \) and \( j_z = \pm \frac{3}{2} \) states from forming bands. The localized 5f\(^2\) orbitals are accounted for in the self-consistent density and, concomitantly, in the potential seen by the conduction electrons. The 5f-bands are calculated by solving the Dirac equation. The intrinsic bandwidth of the itinerant U 5f \( j = \frac{5}{2} \), \( j_z = \pm \frac{3}{2} \) is taken from the LDA calculation while the position of the corresponding band-center C is chosen such that the density distribution of the conduction states as obtained within LDA remains unchanged. The position of the f band relative to the calculated Pt d states is consistent with photoemission data. The band centers of the remaining U 5f states are shifted to higher energies. As a consequence the corresponding orbitals cannot form bands in the energy range of interest. They affect the bands in the vicinity of the Fermi level via the hybridization tails only. It was found that the U 5f bands with \( j_z = \pm \frac{3}{2} \) hybridize strongly near the Fermi level so that the f occupancy per U atom for the delocalized 5f electrons amounts to \( n_f = 0.65 \) indicating that we are dealing with a mixed valent situation.

We next turn to the discussion of the localized U 5f states. The multiplet structure of the localized f\(^2\) states is calculated by diagonalizing a 6 x 6 Coulomb matrix. The spin-orbit splitting is rather large and therefore a jj-coupling scheme is used. This simplification gives six 2-particle states build from \( |j = \frac{5}{2}, j_z = \pm \frac{5}{2} \rangle \) and \( |j = \frac{3}{2}, j_z = \pm \frac{3}{2} \rangle \). The resulting eigenstates are generally no longer eigenstates of the total angular momentum \( J^2 \), but remain eigenstates of \( J_z \). The Coulomb matrix elements are calculated following Condon and Shortley [3]. Inputs are the Slater-Condon parameters \( F^{\text{KR}} \) (Coulomb integrals) and \( G^K \) (exchange integrals) which are evaluated with the radial function \( R_{1s}^{\text{Kr}}(r) \) for U which is determined from a self-consistent band structure potential. The radial function is calculated for an energy given by the center of gravity of the 5f bands. The required integrations are done within the atomic sphere surrounding the U ion. Diagonalization of the matrix yields a doubly degenerate ground state \( J_z = \pm 3 \) which must be an eigenstate of \( J = 4 \). Note that the Pauli principle permits even values of \( J \) only, i.e., \( J = 0, 2, 4 \) in our case. The states \( |j = \frac{5}{2}, j_z = \pm \frac{3}{2}; J = 4, J_z = \pm 3 \rangle \) have an overlap of 0.865 with the Hund’s rule ground state \( \text{J}^2 \text{H}_4 \) derived from the LS-coupling scheme. Therefore the choice of \( j \) vs. LS coupling should only weakly affect the results obtained for the ground-state multiplet. The two-fold degeneracy of the ground-state is lifted by a CEF yielding the two states

\[
|\Gamma_3\rangle = \frac{1}{\sqrt{2}}(|J = 4; J_z = 3\rangle + |J = 4; J_z = -3\rangle)
\]

\[
|\Gamma_4\rangle = \frac{1}{\sqrt{2}}(|J = 4; J_z = 3\rangle - |J = 4; J_z = -3\rangle) .
\]  

(2)

Note that \( |\Gamma_4\rangle \) has been suggested as ground state of UPd\(_2\)Al\(_3\) [14,15]. We assume that the splitting energy \( \delta \) between \( |\Gamma_3\rangle \) and \( |\Gamma_4\rangle \) is of order 20 meV as in UPd\(_2\)Al\(_3\) [10]. The next-higher eigenstates of the Coulomb matrix is a doublet with \( J_z = \pm 2 \). The excitation energy separating it from the ground state is rather large (0.4 eV). We therefore neglect all higher levels. For the evaluation of the effective mass enhancement we need to determine the coupling between the localized and delocalized f electrons. It is directly obtained from the expectation values of the Coulomb interaction \( U_{\text{Coul}} \) in the 5f\(^3\) states. For the latter we use the product states \( |f^2; J = 4, J_z = \pm 3\rangle \otimes |f^1; j = \frac{5}{2}, j_z = \pm \frac{5}{2}\rangle \). The difference \( \langle f^1; \frac{5}{2}, \frac{3}{2}\rangle \otimes \langle f^2; 4, \pm 3 |U_{\text{Coul}}| f^1; 4, \pm 3 \rangle \otimes |f^1; \frac{5}{2}, \frac{3}{2}\rangle - \langle f^1; \frac{5}{2}, \frac{3}{2}\rangle \otimes \langle f^2; 4, -3 |U_{\text{Coul}}| f^1; 4, -3 \rangle \otimes |f^1; \frac{5}{2}, \frac{3}{2}\rangle \) amounts to -0.38 eV from which we deduce the transition matrix element

\[
M = \langle f^1; \frac{5}{2}, \frac{3}{2}\rangle \otimes (\text{Coul} | f^1; \frac{5}{2}, \frac{3}{2}\rangle - \langle f^1; \frac{5}{2}, \frac{3}{2}\rangle \otimes \langle f^2; 4, -3 |U_{\text{Coul}}| f^1; 4, -3 \rangle \otimes \langle f^1; \frac{5}{2}, \frac{3}{2}\rangle ).
\]  

(3)

We may also rewrite M in form of an exchange coupling between the itinerant and localized f states. In that case the Landé g factor for the localized f\(^2\) states must be determined. It turns out that this \( g \) factor is \( g_{\text{eff}} = 0.63 \) and that the exchange integral is \( I \approx 1 eV \), which is of the correct size for f\(^3\) electrons.

Let us now turn to the mass enhancement Eq. (1). The density of states \( N(0) \) corresponding to the Fermi surface in Fig. 3 equals \( \approx 15.5 \) states/ (eV cell), the other parameters are \( |M|^2 = 0.036 \text{ eV}^2 \), \( 4a^2 = 0.13 \) and \( \delta \approx 20 \text{ meV} \). When these parameters are put into Eq. [4] a mass enhancement \( \frac{m^*}{m_b} \) of approximately 8.3 is obtained. This is in excellent agreement with experiment.

The above scenario provides for a satisfactory explanation of the heavy quasiparticles observed in UPt\(_3\). The
different hybridization behavior of the various U 5f orbitals requires further investigations. In a separate study we shall investigate the effect of strong local electron correlations on the different LDA hybridization matrix elements. In concluding we want to point out that the way of treating the $f$ electrons in UPt$_3$ may be also applicable to other uranium compounds.

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