Metamagnetic Transition and Magnetic Properties of UPt$_3$

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We report fully relativistic band structure and total energy studies of UPt$_3$ in the different magnetic states. We find that the ground state of UPt$_3$ is nonmagnetic, even though stable ferromagnetic and antiferromagnetic solutions are also found. We find that the total energy calculations predict the direction of the ordered moment correctly and for the first time the "metamagnetic transition" is found from a first principles total energy calculation for heavy fermion compounds.

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UPt$_3$ has emerged as one of the most thoroughly studied heavy fermion compounds [1]. The neutron scattering experiments show the presence of antiferromagnetic correlations in this compound persisting on long and short time scales and a static moment of $0.02\mu_B$ is ordered along the basal plane at $\mathbf{Q} = (\frac{1}{2},0,0)$ below 5 K [2]. This weak antiferromagnetic order has unusual features which are characteristic of a heavy fermion compound: the moment is weak, the correlation length is finite, and in UPt$_3$ it does not show up in any bulk property [1].

Another feature of UPt$_3$ is the metamagnetic transition, which is typical of a heavy fermion compound. The susceptibility shows a peak which develops into a first order jump in the magnetization in the limit of very low temperatures. In UPt$_3$ this transition occurs in a magnetic field of about 22 T with the field direction in the basal plane, the manifestation of which is shown in a sudden jump in the magnetization at the U site of $0.3\mu_B$ [3]. The quantum oscillatory magnetoresistance measurements of Julian *et al.* [4] provides evidence for nonlinear magnetic splitting of the quasiparticle bands and possible rearrangement of the Fermi surface at the transition field.

In this Letter we shall be concerned with local (spin) density ground state energy of UPt$_3$ in (a) the observed antiferromagnetic (AF) phase at $H = 0$ and (b) the nonmagnetic (NM) and ferromagnetic (F) phases at $H = 0$ and at several values of external magnetic field. The key issue we shall address here is the band theoretical investigation of the metamagnetic transition in UPt$_3$. The total energy calculations in the NM and in the F phases in an external magnetic field predict a first order transition with a jump in the magnetic moment at the U site in accordance with experiment [3].

There exist many first-principles calculations of UPt$_3$ within local density approximation (LDA) or local spin density approximation (LSDA) [5–7]. The LDA calculations were successful in predicting the Fermi surface which are observed in the de Haas–van Alphen experiments [8]. The f electrons were treated as valence electrons in band structure calculations and the Fermi surface agreement with experiment clearly established the itinerant nature of f electrons. Norman *et al.* [6] have performed several spin-polarized band structure calculations both in the simple antiferromagnetic structure ($\mathbf{Q} = 0,0,1$) and in the observed antiferromagnetic phase ($\mathbf{Q} = 0.5,0,0$) with different orientations of magnetic moment. The largest moment of $0.8\mu_B$ was found in the orthorhombic phase with the moments lying in the basal plane; however, the total energy for various magnetic states were not calculated. Sticht and Kubler [7] have calculated the magnetic moment and the heat of formation of UPt$_3$ in the nonmagnetic, the F and AF phases; in their calculations spin-orbit coupling is included only at the end. We compare our results later.

Compared with other heavy fermion compounds, UPt$_3$ has a fairly simple structure (two formula units in a hexagonal unit cell) and good quality crystals can be grown for experiments. We do not consider the weak structural modulation which has been observed recently [9]. All calculations are performed at the experimental lattice spacing [5]. We use the fully relativistic spin-polarized linear muffin tin orbital (SPRLMTO) method [10], where the Dirac equation is solved properly within the relativistic local spin density functional theory. The contribution of the orbital magnetic moment to the effective single particle potential is ignored. The usual nonrelativistic local exchange correlation potential of von Barth and Hedin [11] is used; the relativistic corrections to the local potential were found to have little effect on the structural and magnetic properties [12]. While fully relativistic spin-polarized calculations have been performed in the study of the magnetic anisotropy of transition metals and their surfaces and interfaces [13,14], the method is applied here for the first time to a heavy fermion system. Finally, the combined corrections terms were not included in the LMTO.

In these calculations, the basis functions include s, p, d, and f states on both U and Pt sites. The valence states are taken as $6p^6$, $7s^2$, $6d^1$, and $5f^3$ of U and $5d^9$, $6s^1$ of Pt with p electrons of U being treated in the lower panel. The core charge density is frozen.

In the antiferromagnetic state at $\mathbf{Q} = (0.5,0,0)$, with moments lying in the basal plane, the unit cell doubles up. Consequently, the computations in this orthorhombic structure are extremely time consuming. With 90 k

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points in the irreducible wedge (IRW) of the Brillouin zone and the Hamiltonian size of 512×512 and two panels, the calculations required nearly 4 h of Cray-YMP8 processor per iteration.

At $H = 0$, we find stable nonmagnetic, ferromagnetic, as well as antiferromagnetic solutions. For comparison, all calculations were carried out in the same orthorhombic unit cell. We took particular care to obtain well converged total energies (which takes about 30 iterations in the AF case) and we also made sure the k integrations in all cases were done over the same IRW with the same number of k points. All these solutions lie extremely close in energy within 1 mRyd/cell of each other and moreover the nonmagnetic solution has the lowest energy. Neither the F nor AF solution is the ground state. This is unique in our experience: In all other cases if a polarized state is stable, it is also the ground state.

For all further calculations in the NM and the F phases we use the smaller hcp unit cell, computationally less demanding than those calculations in the orthorhombic unit cell. In the F phase, calculations were also done for different orientations of the magnetic field, along the $[11\bar{2}0]$, $[10\bar{1}0]$ axes in the basal plane and along [0001] direction. For the moments lying along the "c" axis the hexagonal symmetry survives and the IRW is 1/24th of the Brillouin zone. For the moments lying in the basal plane, along the "a" or the "b" axis, the IRW is 1/8th of the Brillouin zone. 196 k points were used in the IRW in the former case and 588 k points in the latter.

First at $H = 0$, we calculated the total energy in NM and F phases with moments along the a axis. We find that the NM solution is the ground state which confirms our earlier results in the orthorhombic structure. The total energy results obtained in both the structures are shown in Table I. Small differences between these values are due to the numerical inaccuracies resulting from integrations in different Brillouin zones. However, the difference between the F and NM states for the same structure is in better agreement.

All the calculations were performed for the observed lattice constant. Earlier calculations [7] showed that the LDA predicts a lattice constant to within 0.5% of the experimental values and we have not repeated these calculations with our fully relativistic method. UPt$_3$ shows a small magneto volume expansion of $\epsilon = 10^{-4}$ [15] at the metamagnetic transition. We have estimated the error in the ground state energy of the magnetic configuration by assuming a "frozen lattice" using the observed value of the bulk modulus B [16]. We find an elastic energy of 6.4×10^{-5} mRyd which is negligible.

TABLE I. Total energy in the different phases of UPt$_3$ in the orthorhombic and hexagonal structures per hcp unit cell in Ry.

	Ortho	hcp
NM	−474.283 063	−474.281 676
AF	−474.282 696	
F	−474.282 224	−474.280 858
ΔE = F−NM	0.000 839	0.000 818

For more insight, we compared the different contributions to the total energy in the NM and magnetic (F) states with that of the ferromagnetic material, bcc Fe. The total energy E can be decomposed as

$$E = E_{\mathrm{K.E.}} + E_{\mathrm{ext}} + E_{\mathrm{Hart}} + E_{\mathrm{xc}} \ , \tag{1}$$

where $E_{\mathrm{K.E.}}$ is the kinetic energy, E_{ext} is due to the external potential including nuclear Coulombic potential, E_{Hart} is the Hartree part, and E_{xc} is the exchange correlation energy. In Table II we give the energy difference of these various contributions to the total energy between the magnetic state (F) and the nonmagnetic state for both UPt$_3$ (in hcp unit cell) and bcc Fe. The striking difference between the two systems as we can see from this table is that in Fe the numbers are considerably bigger. In contrast with Fe, the gain in the exchange correlation energy in the magnetic phase of UPt$_3$ is small compared with the loss due to the kinetic energy. While in Fe, the magnetic solution lies lower than the nonmagnetic one by more than 30 mRyd, in UPt$_3$ the magnetic solution is about 1 mRyd higher.

We also looked at the radial dependence of the exchange correlation energy in the magnetic phase of Fe and U in UPt$_3$. In Fig. 1, we have plotted the radial dependence of the polarization x $[\rho \uparrow (r)/\rho(r)]$, measure of the electron density, r_S $(\frac{3}{4\pi\rho(r)})^{\frac{1}{3}}$, and part of exchange correlation energy which depends on the polarization x.

The polarization x is small in U, because of the large number of electrons and it rises slowly for $r > 0.5$. In Fe, x is bigger and moreover it is finite even at lower values of r, where r_S [Fig. 1(b)] is small (hence at higher densities). This gives rise to larger exchange correlation energy gain in the magnetic phase of Fe and smaller in UPt$_3$. The spin magnetic moment at the U site is considerable: −1.08 (F) and −1.07 (AF). These spin moments are compensated by large orbital moments of 1.49 (F) and 1.53 (AF) giving a net moment of 0.41 (F) and 0.46 (AF).

Comparing the energy of the F solutions with different orientations of the magnetic moment, we find the solution with the spin moment along the a axis is more stable than

TABLE II. Decomposed energy differences between the NM and F states for bcc Fe and hcp UPt$_3$.

Energy (mRyd)	Fe	UPt3 U	 Pt
K.E.	+475.90	+159.54	+43.28
E_{ext}	−458.70	−155.33	−44.93
E_{Hart}	+76.24	+20.29	+5.56
E_{xc}	−125.11	−22.60	−4.38
Net/atom	−31.67	+ 1.90	−0.47
Net/u.cell	−31.67	+0.49	

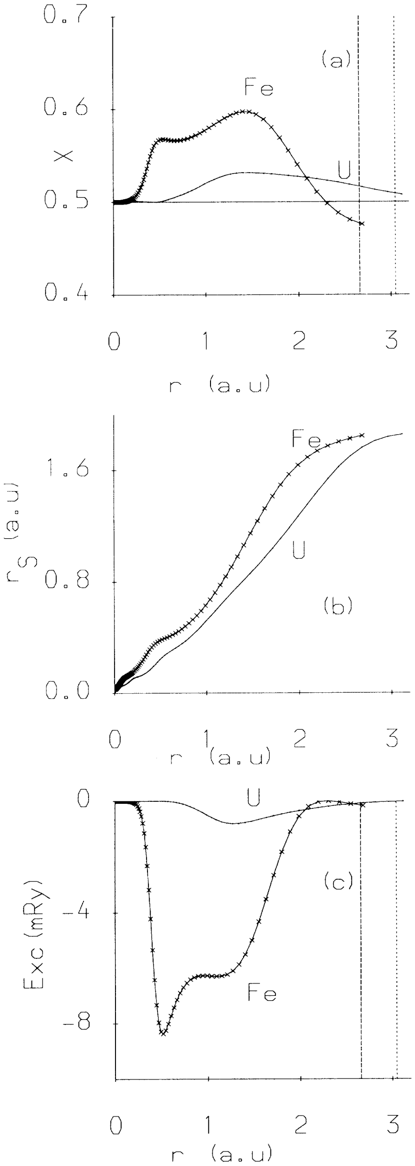

FIG. 1. The radial dependence of (a) polarization x, (b) r_S, measure of electronic density, and (c) part of E_{xc} which depends on x. In (a), $x = 0.5$ refers to the paramagnetic state. The dotted lines at $r = 2.662$ a.u. and $r = 3.049$ a.u. represent the Wigner-Seitz sphere radius of Fe and UPt$_3$, respectively.

the solution with the spin moment lying along the c axis by an energy difference of 0.36 mRyd. Small changes occur to the orbital moments: 1.48 ($\hat{\mathbf{M}}\|\hat{\mathbf{a}}$) and 1.34 ($\hat{\mathbf{M}}\|\hat{\mathbf{c}}$). The spin moments are −1.07 ($\hat{\mathbf{M}}\|\hat{\mathbf{a}}$) and −1.04 ($\hat{\mathbf{M}}\|\hat{\mathbf{c}}$). Studying this magnetic anisotropy further we also find that the in-plane anisotropy is very small, namely 0.1 mRyd.

We now report the results in a finite magnetic field. The electronic structure and the total energy in the F and NM phases were determined in an external magnetic field. Here the magnetic field couples only to the spin of the electrons and it is straightforward to add a term $-2\boldsymbol{\sigma} \cdot \mathbf{B}$ to the one electron Hamiltonian in our band structure calculations. As a function of the magnetic field we find the changes in the electronic structure (Fermi surface) are not significant while significant changes occur as a function of the direction of the field [17].

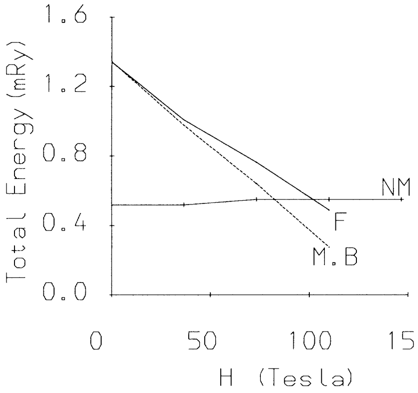

FIG. 2. Total energy (with respect to −474.2822 Ry) as a function of the magnetic field B in the F phase and NM phase of UPt$_3$. The dotted line in the F phase is calculated using Eq. (3).

From thermodynamics the change in the ground state energy as a function of the magnetic field is given by

$$\Delta\xi = -\mathbf{M} \cdot \mathbf{B} - \frac{1}{2}\chi B^2 - O(B^3). \qquad (2)$$

In a ferromagnet the first term is expected to dominate (the second term only becomes important when the applied field induces an additional moment which is comparable to the spontaneous moment). Thus we expect that the energy of the ferromagnet should vary approximately linearly with the field and that of the paramagnetic phase should be essentially independent of B. This is confirmed by the calculation as is shown in Fig. 2. The total energy calculation for the paramagnetic phase is almost independent of B—but it does show a slight positive slope. This is an artifact of the calculation.

We can also calculate the energy of the ferromagnetic state from

$$\xi(B) = \xi_0 - \mathbf{M}_S \cdot \mathbf{B} \qquad (3)$$

using the calculated value for the total spin moment M_S. This is shown as the dotted line in Fig. 2. The difference between this line and the solid line of our first-principles total energy calculations is due to the spin-orbit effects being included—the first principles calculation is fully relativistic. The total moment is aligned along the field and hence the effective g value for the spin component is reduced below 2. We have performed a similar calculation for iron—in this case the two slopes are identical.

However, the important thing to note from this figure is the crossing at 102 T. Around 102 T, our calculated F solution with the moments lying along the *a* axis crosses the nonmagnetic solution and becomes the ground state. Thus a phase transition from a nonmagnetic to a ferromagnetic state occurs with a jump in the net magnetic moment of about $0.4\mu_B$. Experimentally, the metamagnetic transition is obtained at 22 T at which the magnetic moment jumps by $0.3\mu_B$.

The value of the critical field obtained by this method is too high. This is a consequence of the fact that the total energy calculation has *overestimated* the energy difference between the paramagnetic and ferromagnetic states. This is because the LDA fails to take full account of the correlations associated with the quasilocalized *f* electrons. Inclusion of the magnetism associated with the orbital motion will further increase the value of the predicted critical field.

The importance of this result is that it is the first time that the metamagnetic transition in UPt_3 or any other heavy fermion compound has been predicted by total energy calculation. This is a consequence of the fact that in this material the total energy calculations predict a metastable ferromagnetic phase. The calculations also predict the *a* axis to be the easy axis which is in agreement with the experiment.

We note that for a first order transition there may be three relevant fields: (i) the field at which the energies of the phases are equal, H_0; (ii) the field at which the low moment state is unstable, H_u; and (iii) the field at which the high moment state is unstable, H_l, one always has $H_l < H_0 < H_u$. In this work we find H_l does not exist since the high moment state is stable down to $H = 0$ and we do not calculate H_u. In their study of Pd, Jarlborg and Freeman [18] calculated H_l and H_u but not H_0.

Our results not only reveal the nature of the metamagnetic transition but also provide the explanation for the nonlinear spin splitting of the quasiparticle bands observed by Julian *et al.* [4]. At the transition field, a transition from the NM to F state with a jump in the magnetization results in the nonlinear spin splitting of the bands. Moreover, the calculations also show that the induced magnetization is described completely by a difference between the up and down Fermi surface volumes. Finally, from the topology of the bands in the NM and F states [17] there is clear evidence of Fermi surface rearrangements at the transition.

The two special features of this calculation are the fact that it is fully relativistic and the *k* mesh is much denser than the previous calculations [6,7]. Norman *et al.*, finds a large net moment ($0.81\mu_B$) in the antiferromagnetic phase. While Sticht and Kubler find a net moment similar to ours, the ordering of ground state energies are different [7]: NM−F=28 meV against the present result of −5.6 meV per UPt_3.

The purpose of this Letter is to show that a first principles calculation carried out fully relativistically and to a high accuracy is able to explain the metamagnetic transition in a heavy fermion compound as well as to predict correctly the size of the moment in the high field state and the easy direction of the magnetization. This is an important result because it is well known that LDA methods are successful in explaining some properties, for example, Fermi surface and magnetic moments but not others, for example, effective mass and photoemission linewidths. This calculation shows that the extent to which the crude LDA approximation accounts for the correlations appears to be sufficient for a qualitative understanding of the metamagnetic transition provided the relativistic effects are treated correctly.

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