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## Magnetization density in heavy-fermion UPd<sub>2</sub>Al<sub>3</sub>

L Paolasini††, J A Paixão†¶, G H Lander†, A Delapalme§, N Sato|| and T Komatsubara||

† Commission of the European Communities, Joint Research Institute, Institute for Transuranium Elements, Postfach 2340, 76125 Karlsruhe, Federal Republic of Germany

‡ Département de la Recherche Fondamentale sur la Matière Condensée (CEA), SPSMS, MDM, 85X, 38041 Grenoble Cédex, France

§ Laboratoire Léon Brillouin (CEA-CNRS) Centre d'Etudes Saclay, 91191 Gif-sur-Yvette, France

|| Department of Physics, Faculty of Science, Tohoku University, Sendai 980, Japan

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**Abstract.** The induced magnetization density has been measured at  $T = 36$  K (above the antiferromagnetic ordering temperature of 14 K) in a single crystal of the heavy-fermion material UPd<sub>2</sub>Al<sub>3</sub>. A magnetic field of 5 T was applied along the easy axis of magnetization. The magnetization density as measured by the polarized-neutron technique is associated totally with the uranium atom; no spin transfer is found on the Pd atoms. The induced moment, as measured by neutrons, is some 12% below that measured by magnetization measurements on the same crystal. This indicates the presence of a significant positive (with respect to the uranium moment) conduction electron polarization. The ratio of the orbital to the spin moment of uranium found experimentally is  $-2.01(25)$ , compared with the free-ion values of  $-2.56$  (for U<sup>3+</sup>) and  $-3.29$  (for U<sup>4+</sup>), and a recent theoretical prediction of  $-1.35$ .

### 1. Introduction

The large variety of behaviours exhibited by uranium atoms in compounds has led to an extensive research effort on their magnetic and electronic properties. Certainly the most celebrated of these are the so-called heavy-fermion materials (Ott and Fisk 1987, Grewe and Steglich 1991, Sigrist and Ueda 1991) which exhibit large Sommerfeld coefficients ( $\gamma$ , the low-temperature electronic coefficient of the specific heat). A major question in this research is the nature of the electron wavefunctions, especially at low temperatures. Furthermore, in systems involving d transition metals there is the important question of the role of the d electrons, especially in view of the possibility of f–d hybridization.

Such complications involving f–d electron hybridization may be well illustrated in our recent research on another set of U compounds, those with the ZrNiAl (hexagonal) structure, and with the formula URhAl and URuAl, for example. The  $\gamma$ -values of these materials are modest, being in the range 40–150 mJ mol<sup>-1</sup> K<sup>-2</sup>, but they exhibit enormous anisotropy and always have their easy axis along the unique hexagonal  $c$  axis. None (yet) has been found to become superconducting. In our studies of URhAl (Paixão *et al* 1992) and URuAl (Paixão *et al* 1993) we have found that a large polarization of the d electrons is found at the transition-metal site. This amounts to about 35% of that of the U 5f electrons. We have proposed that this large polarization of the transition-metal electrons is a consequence of

¶ On leave from Departamento de Física, Universidade de Coimbra, Portugal.

the 5f–4d hybridization and is the cause of the strong uniaxial anisotropy in these materials (Paixão *et al* 1993).

Recently, a new heavy-fermion compound UPd<sub>2</sub>Al<sub>3</sub> has been discovered (Geibel *et al* 1991a, b) that orders antiferromagnetically (Krimmel *et al* 1992) at  $T_N = 14$  K with a moment of  $0.85(3)\mu_B$  per U atom and becomes superconducting at about 2 K. It has a hexagonal crystal structure and the easy axis of magnetization is in the (001) plane rather than along the unique *c* axis (Sato *et al* 1992). In this respect the anisotropy is similar to that found in the hexagonal heavy-fermion UPt<sub>3</sub> and different from the situation discussed in the preceding paragraph for the hexagonal uranium 1:1:1 compounds.

Our experiments to examine the magnetization density of UPd<sub>2</sub>Al<sub>3</sub> were motivated principally by the question of whether there is any induced moment on the 4d electrons at the Pd site; a second question was whether the tetravalent ground state proposed for the uranium ion by Grauel *et al* (1992) could be verified by observing the angular anisotropy of the magnetization density. Sticht and Kübler (1992), moreover, have predicted values for the orbital and spin moments in this compound, a subject that is addressed in our experiments.

## 2. Experimental details and results

The measurements reported in this paper have been performed at the 5Cl polarized-neutron diffractometer at the Orphée Reactor of the Laboratoire Léon Brillouin (Saclay) on a small single crystal of composition UPd<sub>2</sub>Al<sub>3.06</sub>, prepared by Czochralski pulling using a tri-arc furnace at Sendai. The crystal is a parallelepiped with dimensions of 2.3 mm × 1.5 mm × 6.3 mm with the long dimension parallel to a twofold *b\** reciprocal axis. Here *b\** is a crystallographic direction bisecting two equivalent (100)\* directions in reciprocal space. The sample was placed in a cryomagnet with a 5 T magnetic field applied along the long dimension. Within the scattering plane (perpendicular to the field direction), reflections obeying the relationship  $H + 2K = 0$  were observed, and the use of the normal-beam technique allowed access up to higher levels with  $H + 2K = 3$ . *L*-values of up to 4 were accessible. The neutron wavelength used was 0.829 Å obtained from a Heusler crystal with an erbium filter to reduce second-order contamination. In a secondary experiment the positions and integrated intensities of a small number of reflections were measured on the same apparatus to determine the lattice parameters and to test for extinction. A refinement of the lattice parameters gave  $a = b = 5.28$  Å and  $c = 4.114$  Å at 36 K, which are consistent with values in the literature for samples made with excess aluminium. The integrated intensities were consistent with those calculated with the hexagonal PrNi<sub>2</sub>Al<sub>3</sub> structure (space group, *6/mmm* (No. 191)), so that we do not expect extinction to be a serious problem. Further details of the experimental method can be found, for example, in the work of Paixão *et al* (1992).

The polarized-beam method relies on the fact that there is interference between the magnetic and nuclear signal, and through this interference the method is sensitive to small magnetic signals. A consequence of the requirement of this interference is that in the usual configuration the method is sensitive to *ferromagnetic* (i.e.  $q = 0$ ) components only. Since UPd<sub>2</sub>Al<sub>3</sub> is *antiferromagnetic* below  $T_N = 14$  K we have performed our experiments at  $T = 36$  K, at which temperature the bulk susceptibility has a maximum. Thus, our experiment is not aimed at investigating the superconducting state ( $T < 2$  K), but we determine the wavefunction in an applied field in the paramagnetic state. We do not expect the nature of the wavefunctions to change on entering the antiferromagnetic state.

In figure 1 we show the susceptibility measured on our sample (*H* is parallel to the long axis of the crystal) with a field of 5 T, i.e. exactly the experimental conditions of the neutron

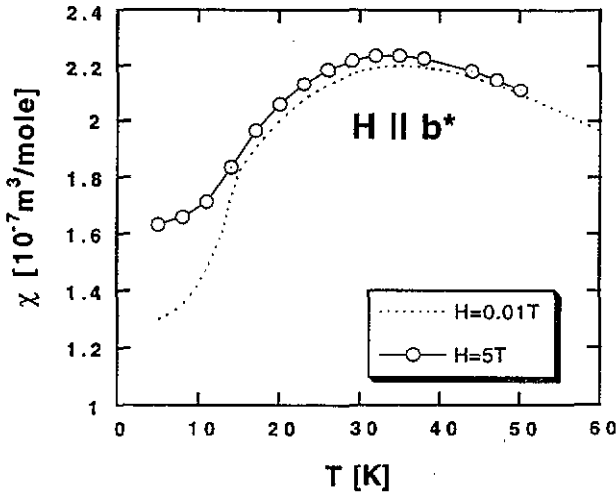


Figure 1. DC magnetic susceptibility versus temperature as measured on the same crystal as used for the neutron studies measured in applied fields of 5 T (○) and 0.01 T (---).

experiment. These susceptibility measurements were performed on the SQUID magnetometer at the Centre d'Etudes Nucléaires, Grenoble collecting 15 different temperatures from 5 to 50 K. Measurements of the susceptibility at  $H = 0.01$  T (performed at Sendai) are shown also for this direction in figure 1. The susceptibility is slightly non-linear with field; for example  $H = 0.01$  T the maximum value is  $2.2 \times 10^{-7} \text{ m}^3 \text{ mol}^{-1}$ , whereas at 5 T we find this has increased to  $2.26 \times 10^{-7} \text{ m}^3 \text{ mol}^{-1}$ .

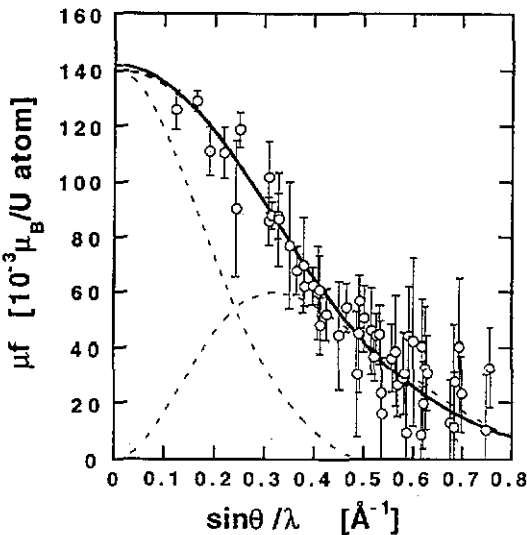


Figure 2. Magnetic scattering amplitudes as measured at 36 K in an applied field of 5 T and plotted versus  $(\sin \theta)/\lambda$  assuming that the total response is located at the uranium site. The best fits to the dipole approximation for  $U^{4+}$  and  $U^{3+}$  are shown as full and bold broken curves respectively. They are almost indistinguishable. The broken curves correspond to the functions  $\langle j_0 \rangle$  and  $\langle j_2 \rangle$  for  $U^{3+}$  used in the fit (see Freeman *et al.* (1976)).

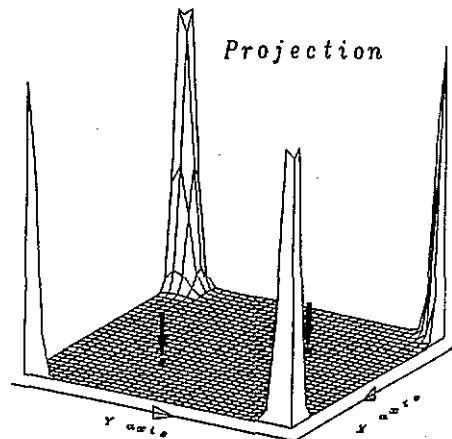


Figure 3. Maximum-entropy reconstruction of the magnetization density projected on the basal plane of the hexagonal  $UPd_2Al_3$  structure. The high values are associated with the density at the uranium sites; the arrows indicate the positions of the palladium sites. The  $x$  axis is parallel to the hexagonal  $a$  axis, and the  $y$  axis is perpendicular to it.

The results of measuring 55 reflections are shown as a form factor in figure 2. This method of presentation assumes that all the moment is associated with the uranium atom. Four reflections have been omitted from this plot. For these four reflections the nuclear scattering is weak, and there is a high probability that the small intensities are contaminated by multiple-scattering effects. The first qualitative point to be made with respect to this figure is that it appears to be much like a conventional uranium form factor, and our initial guess is that there is almost no moment on the Pd sites.

### 3. Analysis and discussion

The question of whether any induced moment resides on the Pd sites may be examined either by the Fourier technique or, with greater certainty, by applying the method of maximum entropy (Papoular and Gillon 1990) to the Fourier transform. The result is shown in figure 3. The induced moment on the U site is  $0.140(4)\mu_B$  and that on the Pd atom is zero to within a precision of  $0.004\mu_B$ , i.e. less than 3% of the moment induced on the U site. We have assumed that any moment on the Pd site would have a form factor similar to that for elemental Pd (Cable *et al* 1975). The absence of any induced moment on the Pd site is *different* from the situation that we have found in the hexagonal uranium 1:1:1 compounds containing 4d transition-metal atoms. Some of these differences may be related to the structural coordination, although in both cases the structures may be considered as a succession of weakly interacting layers perpendicular to the *c* axis. In the 1:1:1-type compounds with the ZrNiAl structure there is a relatively short *single* U-T<sub>1</sub> bond of 2.94 Å. Strong hybridization is found to occur between electrons on the uranium and T<sub>1</sub> sites (Paixão *et al* 1993). However, in the UPd<sub>2</sub>Al<sub>3</sub> structure there are *six* equidistant Pd atoms around the U atom at 3.12 Å. We assume that the absence of an induced moment on the Pd atoms is evidence for the absence of any strong hybridization between the U 5f and Pd 4d electrons. Since the model developed for the 1:1:1 compounds associates the strong uniaxial anisotropy with hybridization, the absence of *both* these effects in the present compound is consistent with the model. The easy axis of magnetization is then determined by other factors, such as exchange or crystal-field interactions.

The susceptibility value of  $2.26 \times 10^{-7} \text{ m}^3 \text{ mol}^{-1}$  corresponds to a total moment induced at 5 T of  $0.160\mu_B \text{ mol}^{-1}$ . The above analysis of the form factor shows that the moment at the uranium atom is  $0.140(4)$  and that there is nothing at either the Pd or Al sites, so that the difference of  $0.02\mu_B \text{ mol}^{-1}$  must be ascribed to the plane-wave states, i.e. the conduction electron polarization. Such states, with an extended spatial extent, possess a form factor that falls rapidly to zero by about  $0.1 \text{ \AA}^{-1}$  in  $(\sin \theta)/\lambda$  and thus are not observed in the neutron experiments. The conduction electron states are, of course, always present in metallic systems and have been studied extensively in uranium-based ferromagnets. Normally, in ferromagnetic materials such as US (Wedgwood 1972) or UFe<sub>2</sub> (Lebech *et al* 1989) they attain values of about 10% of the total moment but are directed *opposite* to the localized moments. We have less experience of the conduction electron moments induced in *paramagnetic* systems. In URuAl (Paixão *et al* 1993), for example, no significant difference could be detected between the value of the moments as deduced from neutron and magnetization measurements, suggesting a negligible conduction electron polarization. Certainly the value of about 12% of the total moment in UPd<sub>2</sub>Al<sub>3</sub> is significant, and the fact that it is parallel to the uranium moment may suggest a large contribution from the outer Pd electrons. The interest in this aspect is that the quasi-particle states that are generally believed to be responsible for the high value of  $\gamma$  arise from hybridization between the uranium 5f and conduction states.

The U form factor in figure 2 is essentially a smooth curve as a function of the momentum transfer  $Q(= (4\pi \sin \theta)/\lambda)$ . The relatively poor statistics, together with the absence of any appreciable anisotropy in the form factor, mean that we are unable to make any statement about whether the crystal-field ground state of the 5f electrons proposed by Grauel *et al* (1992) is correct or not. The method of analysis of the form factor in terms of crystal-field wavefunctions is reliable only when appreciable anisotropy is observed in the form factor (Lander *et al* 1976, Boucherle and Schweizer 1985). Instead, we can use the dipole approximation (Lander *et al* 1991, Lebech *et al* 1991) to estimate the ratio of the orbital to spin moment at the uranium site. The dipole analysis is normally not valid for  $(\sin \theta)/\lambda > 0.6 \text{ \AA}^{-1}$ , but the poor statistics for these data mean that they are given little weight in the least-squares analysis. The results for such an analysis are presented in table 1 for the  $U^{4+} 5f^2$  and  $U^{3+} 5f^3$  configurations. Also given is the ratio predicted by the calculations of Sticht and Kübler (1992). Sticht and Kübler have performed calculations with a ferromagnetic ground state, in which they find  $\mu_L/\mu_S = -1.29$ , and for an antiferromagnetic ground state they find the ratio  $-1.41$ . We have tabulated the mean of these two. Clearly, the amount of reduction in the orbital moment predicted by Sticht and Kübler is not observed experimentally, although the experimental values of the  $|\mu_L/\mu_S|$ -ratio are significantly lower than the free-ion ratios, especially for the  $U^{4+}$  state. Sticht and Kübler find a configuration of  $5f^{2.7}$  for the U atom in the solid, which is close to  $U^{3+}$ . We cannot distinguish between the  $U^{3+}$  and  $U^{4+}$  by the fits to the form factor; both are equally good. As is the case for the effective moments  $\mu_{\text{eff}} = 3.58\mu_B$  for  $U^{4+}$  and  $\mu_{\text{eff}} = 3.62\mu_B$  for  $U^{3+}$ , the form factors for these two ionic states are indistinguishable (Freeman *et al* 1976).

**Table 1.** Results of the least-squares refinements of the 55 measured reflections using the single-electron wavefunctions of Freeman *et al* (1976) and the dipole approximation. For all measurements the sample is at 36 K and in an applied magnetic field of 5 T. The total moment is from the bulk magnetization measurement. The conduction electron polarization is derived simply from the difference between the second and third columns. The last column refers to the calculations of Sticht and Kübler (1992) who obtained an f count of 2.7 5f electrons, i.e. close to  $5f^3$ .

	$\mu$ ( $m\mu_B$ )			$-\mu_L/\mu_S$		
	Total	Uranium	Conduction electrons	Observed value	Free-ion value	Sticht and Kübler
$U^{4+} 5f^2$	161(1)	142(4)	19(4)	2.28(32)	3.29	
$U^{3+} 5f^3$	161(1)	140(4)	21(4)	2.01(25)	2.56	$\approx 1.35$

#### 4. Summary

Our experiments have determined that the magnetization density in  $UPd_2Al_3$  is associated only with the U atom. No significant polarization of the Pd 4d electrons occurs, in agreement with the calculations of Sticht and Kübler (1992) and presumably because the Pd 4d states are too far below  $E_F$ . Our measurements are unable to address the question of the crystal-field ground state proposed for the U site by Grauel *et al* (1992), nor can we say whether the ground state is  $5f^2$  or  $5f^3$ . There is a significant orbital moment present at the U site. The ratio of the orbital to the spin moment is reduced from that expected from a free-ion

state, presumably as a result of correlation effects (Eriksson *et al* 1991), but this reduction is not as great as proposed by the local-density calculations of Sticht and Kübler (1992).

A simple model is that the Pd 4d band is filled and there is little overlap in the energy of the U 5f and Pd 4d bands; this model is consistent with that proposed for UPd<sub>3</sub> (Eriksson *et al* 1989), which is known to be one of the few uranium systems where the free-ion crystal-field picture is applicable (McEwen *et al* 1993). It seems possible, therefore, that a crystal-field picture may also apply to UPd<sub>2</sub>Al<sub>3</sub>, although no direct spectroscopic measurements of such levels by neutron inelastic scattering has yet been reported.

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