# High field dilatation experiments on $U(Pt_{0.95}Pd_{0.05})_3$; Magnetic ordering versus heavy fermion behaviour

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## Abstract

A still open question is whether long-range magnetic ordering as observed in some heavy fermion (HF) systems is intimately related to the HF state or is an extra phenomenon interacting with it. This problem might be addressed by studying the compound $U(Pt_{0.95}Pd_{0.05})_3$. Doping $UPt_3$ with 5% Pd induces long range antiferromagnetic order (LRAFO) which coexists with the heavy-fermion (HF) behavior. We present thermal expansion and magnetostriction data on $U(Pt_{0.95}Pd_{0.05})_3$ along all three crystallographic axes in magnetic fields ($B \parallel a$-axis) up to 17 T and the temperature range 1.5–12 K covering the critical fields and temperatures of both the LRAFO ($T_N = 5.8$ K, $B_{AF} = 13$ T) and the HF state ($T^* \approx 8.2$ K, $B^* \approx 12$ T). We observe features related to the HF-behaviour and to the LRAFO separately, but also features which we interpret as originating from the interaction between LRAFO and HF-behaviour.

Doping $UPt_3$ with 5% Pd ($U(Pt_{0.95}Pd_{0.05})_3$) induces long range antiferromagnetic order (LRAFO) which coexists with the heavy-fermion (HF) behaviour [1]. $UPt_3$ itself can be considered as a prototype system for the U-based heavy-fermion (HF) systems. Therefore $U(Pt_{0.95}Pd_{0.05})_3$ is an ideal system to address the still open question whether long range magnetic ordering (LRAFO) as observed in some heavy fermion (HF) systems is intimately related to the HF state itself or is an extra phenomenon interacting with it.

Because of the importance of the ligand hybridization for the HF-state and because of the strong dependence of the LRAFO on the intersite exchange parameter both magnetostriction (MS) and thermal expansion (TE) are sensitive tools to study simultaneously both HF-behaviour as well as LRAFO. In addition, length change measurements are a thermodynamical technique sensitive to crystallographic directions. Field and length changes can be applied c.q. measured along well-specified directions.

Neuron diffraction experiments on $U(Pt_{0.95}Pd_{0.05})_3$ reveal the presence of antiferromagnetic ordering with a ordered moment equal to $0.6 \pm 0.2\mu_B$/U-atom [2] pointing along the $b$-axis. The magnetic structure consists out of a doubling of the crystallographic unit cell along the $b$-axis. The $\lambda$-like anomaly in the specific heat data [3] and the Cr-type anomaly in the resistivity data [4] confirm the presence of long range antiferromagnetic order (LRAFO) of the spin density wave type. The Néel-temperature and the critical field for $B \parallel a$-axis are $T_N = 5.8$ K, and $B_{AF} = 13$ T respectively. The characteristic field and temperature as e.g. deduced from specific heat, resistivity or susceptibility for the HF state itself are $T^* \approx 8.2$ K, $B^* \approx 12$ T ([5]).

The experiments have been performed on the same sample as is used in previous specific-heat measurements ([3]). Length changes were measured using a parallel-plate capacitance method. The field was always applied along the $a$-axis of the hexagonal close packed structure, while the length changes were measured along the $a$-, $b$- and $c$-axis. For all three crystallographic directions both TE as forced MS-data

were taken in fields up to 17 T and in the temperature range 1.5–12 K, covering critical field and temperature of both the LRAFO and the HF state.

The forced MS-data along the a-, b- and c-axis have similar features; a broad shoulder related to the metamagnetic transition of the HF-state (having its maximum at $B^* \approx 12$ T) with on top of it a sharp feature related to the critical phenomena at $B_{AF}$ of the LRAFO (see e.g. Fig. 1). The broadness of the metamagnetic transition as compared to MS-data taken on pure UPt$_3$ could be explained by the fact that U(Pt$_{0.95}$Pd$_{0.05}$)$_3$ is not a single crystal in the strictest sense of the word. Pd is replacing Pt and is homogeneously distributed on the scale of several unit cells but it is not always the same Pt position in a unit cell which is replaced by Pd. The locations of the anomalies in the $B–T$ plane correspond nicely with the results obtained by magnetoresistance experiments [7]. For the a- and b-axis an extra feature is observed at approximately 1 T, related to the presence of antiferromagnetic domains. By comparing field up and down sweeps [6], it appeared that a minimum field of 8 T is needed to create a single domain sample.

The thermal expansion, $\alpha_i = L_i^{-1}\, \Delta L_i/\Delta T$, along all three crystallographic directions, indicated by the subscript $i = a, b$ or $c$, at different fixed field values are shown in Fig. 2. The lowest accessible regulated field is 0.5 T. The data taken on the sample cooled in zero field and measured at respectively 0.5 T and 0 T were identical. But they differ significantly for the a- and b-axis from the 0.5 T data measured after the sample
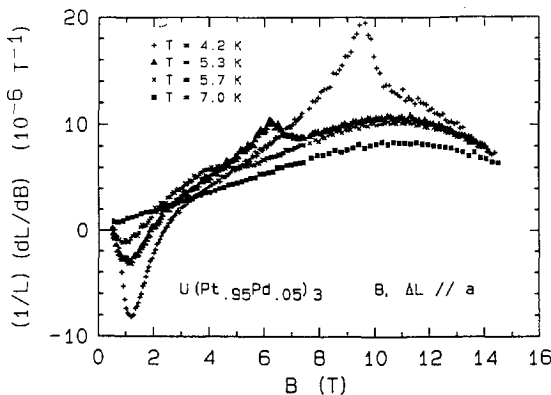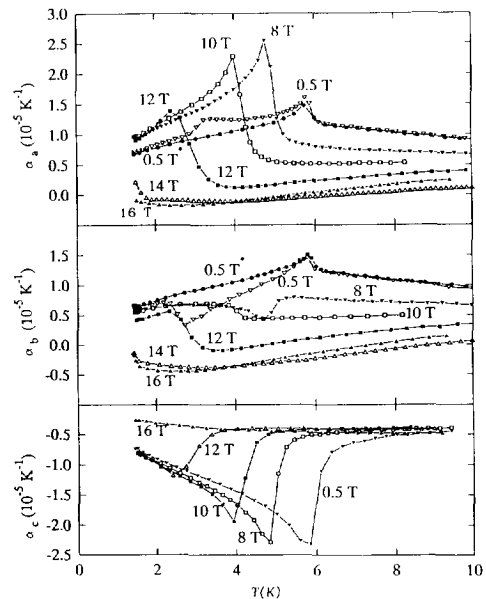


Fig. 2. Thermal expansion data along the crystallographic a-, b- and c-axis for $B \parallel a$-axis. Field and temperature covering both the critical fields and temperatures of the long range magnetic ordering and the heavy-fermion state. The * indicates the curves which are not cooled in a field of 10 T.



Fig. 1. Some typical magnetostriction curves $B \parallel a$-axis in temperatures above and below $T_{Néel}$ along the crystallographic a-axis.

was cooled in a field higher than 10 T (see the different curves of $\alpha_a$ and $\alpha_b$ for 0.5 T in Fig. 1). Cooling the sample in a field exceeding 10 T will cause the sample to be single domain and we therefore attribute this difference to the sample being either multi (denoted by ★ in Fig. 1) or single domain. Along the c-direction a negative sharp anomaly at $T_N(B)$ is observed. No domain effects were found. The effect of the field is to decrease the height of the feature and the temperature $T_N(B)$ at which edge occurs. For fields $B > 12$ T no features are observed in $\alpha_c$. The observed features in the $\alpha_a$- and $\alpha_b$-curves for different fields cannot easily be described in terms of simple trends. A sharp extra feature is observed at $T_N(B)$ being either an extra negative, in the case of the b-axis, or an extra positive, in the case of the a-axis contribution. For $T > T_N(B)$ and $T < 9$ K, the similarity with the TE-data on pure UPt$_3$ [6], the field dependence of the TE-curves (phonons are hardly field dependent), and the still visible metamagnetic transition in MS-data suggest the presence of a HF-state for $T$ exceeding $T_N(B)$. At least as long as $B$

does not exceed 12 T. Therefore the LRAFO and the HF-behaviour should be considered as two separate phenomena interacting with each other.

We plotted $\alpha_0 \equiv (1/2)(\alpha_a + \alpha_b)$, which is proportional to the 'surface' TE of the $a$, $b$-plane, and $\alpha_a - \alpha_b$ (see Fig. 3). The $\alpha_0$-curves for fields smaller than 12 T look similar to the HF-feature as found in $UPt_3$ [6] with on top of it a feature related to the LRAFO. Plotting these curves as a function of $(T/T_N(B))$ (Fig. 3), reveals that the $T/T_N(B) < 1$ parts of the curves are almost identical to each other, except for the extra little feature observed at $T_A \approx 2.5$ K $(T_A/T_N(B) \approx 0.4)$ in the $\alpha_0(B = 0.5$ T)-curve. This suggests that the important energy scale in this region is $T_N(B)$, and that the influence of the LRAFO on the HF-state is

much stronger than the direct influence of the field on the HF-state.

We think the extra feature at $T_A$ in the $\alpha_0$ $(B = 0.5$ T)-curve is related to the anisotropy field of the LRAFO. Some thermal energy is needed to overcome the energy related to the anisotropy field, after which the moments can rotate minimizing the total free energy. (At temperatures below $T \approx 2.5$ K $(T_A)$ the shape of $\alpha_0(B = 12$ T)-curve (see Fig. 2) still suggests the dominance of a HF-state, while at higher temperatures it changes to a curve similar to those found for $UPt_3$, corresponding to a field suppressed HF-state [8] $(B > 20$ T, which is the characteristic field of the HF state in $UPt_3$). Reaching $B_{AF}$, the LRAFO correlation length will tend to zero but still some fluctuating antiferromagnetic ordering is present.) As is discussed in Ref. [9] the HF-state can be pictured as an extended state, and one of the influences determining the magnetic topology of this HF-state is the magnetic structure already present of another origin as e.g. LRAFO in our case. For $B = 12$ T, the balance of influences on the HF-state between LRAFO, which tries to restore the HF-state, and the field, which in this field region $(B = B^*)$ partly suppresses the HF-state, becomes critical. Increasing the temperature causes the LRAFO to disappear and the balance will tip over in favour of the field. For even higher fields the $\alpha_0$- and $\alpha_{a,b}$-curves are smooth. Its physics is being dominated by the localized moments on the uranium sites.

In Fig. 3 also $\alpha_a - \alpha_b$-curves are plotted for several fixed fields. A peak at $T_N(B)$ is observed.

From both MS- and TE-data the length itself in the studied temperature and field regime can be deduced. It was found that decreasing the temperature into the LRAFO-state (passing $T_N(B)$) causes the surface in the $a$, $b$-plane to shrink. The $a$-axis shrinks considerably more than the $b$-axis. This is consistent with the peak-like feature observed in $\alpha_a - \alpha_b$. A possible explanation of the observed feature in the $\alpha_c$-curves (see Fig. 2) could also be given in terms of this strong deformation of the $a$, $b$-plane.

Finally we also calculated the volume TE $(\alpha_V = (1/3)(\alpha_a + \alpha_c))$ for several fixed fields. We find that $\alpha_V$ in the LRAFO-state is almost a constant independent of field and temperature.

In conclusion, from analyzing our data we find that the effect of LRAFO, on the HF-state is substantial and much stronger that the direct effect of the applied field. For $T < T_N(B)$ the important energy scale is $T_N(B)$ (see Fig. 4). It is therefore not by accident that
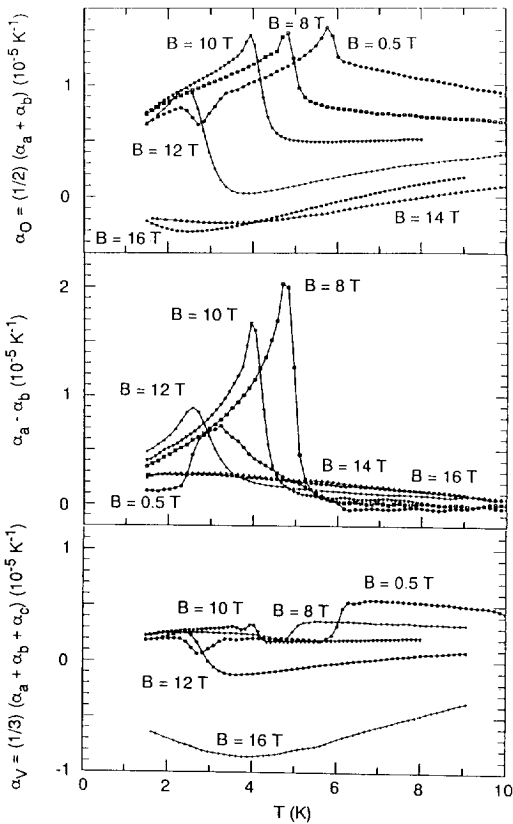


Fig. 3. Plot of $\alpha_0 = (1/2)$ $(\alpha_a + \alpha_b)$ (upper part figure) and $\alpha_a - \alpha_b$ (middle part of the figure) as determined from the measured thermal expansion curves (see Fig. 2). The lower part of the figure contains the volume thermal expansion.
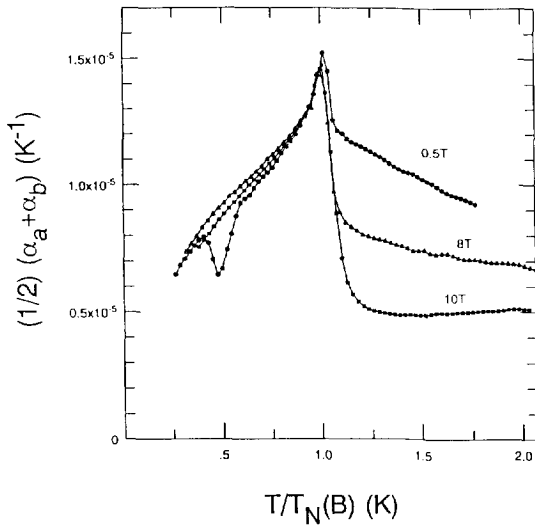
Fig. 4. A plot of the $\alpha_0 = (1/2)(a_a + \alpha_b)$ of several fixed fields as a function of the temperature normalized with respect to the Néel-temperature of the specific field the $\alpha_0$-curve is measured in.

for $T$ approaching 0 K, $B_{AF} = B^*$. Also consistent with this is the fact that $\alpha_v$ in the LRAFO-state is a constant almost independent of field and temperature.

## References

[1] A. de Visser, J.C.P. Klaasse, M. van Sprang, J.J.M. Franse, A. Menovsky, T.T.M. Palstra and A.J. Dirkmaat, Phys. Lett. A 113 (1986) 489.
[2] P.H. Frings, B. Renker and C. Vettier, J. Magn. Magn. Mater 63 & 64 (1987) 202.
[3] H.P. van der Meulen, J.J.M. Franse, A. de Visser, J.A.A.J. Perenboom and H. van Kempen, Physica B 165 & 166 (1990) 441.
[4] R. Verhoef, A. de Visser, A. Menovsky, A.J. Riemersma and J.J.M. Franse, Physica B 142 (1986) 11.
[5] J.J.M. Franse, H.P. van der Meulen and A. de Visser, Physica B 165 & 166 (1990) 383.
[6] U. Wyder et al., to be published.
[7] K. Bakker, A. de Visser, A.A. Menovsky and J.J.M. Franse, Physica B 186–188 (1993) 687.
[8] U. Wyder, H.P. van der Meulen, A. de Visser, P. van der Linden, J.A.A.J. Perenboom, A. Menovsky and J.J.M. Franse, Physica B 199 & 200 (1994) 178.
[9] U. Wyder, Physica B 204 (1995) 255.