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HIGH-FIELD MAGNETISATION OF SINGLE-CRYSTALLINE U(Pt, Pd)$_3$

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High-field magnetisation measurements on single-crystalline samples of the heavy fermion compounds U(Pt, Pd)$_3$ at 4.2 K reveal metamagnetic types of transitions for field directions perpendicular to the hexagonal axis. The transition is shifted to lower field values for increasing Pd content and approaches zero field for the 10% Pd alloy.

1. Introduction

Anomalous high-field properties have been observed in a number of heavy fermion systems like UPt_3, $CeRu_2Si_2$, URu_2Si_2 and $CeCu_2Si_2$. These anomalies have been studied in magnetisation, magneto-transport, specific heat, magnetostriction, thermal expansion and magneto-acoustic experiments (for a review, see ref. [1] and references therein). In the systems mentioned, large magnetic anisotropies are involved. This anisotropy stresses the need for work on single-crystalline samples in particular. In the present contribution, experiments on three $U(Pt_{1-x}Pd_x)_3$ samples with x values of 0.00, 0.05 and 0.10 are compared.

The system U(Pt, Pd)$_3$ has been discussed before on several occasions, in particular in relation to the occurrence of superconductivity and long-range antiferromagnetic order [2]. Superconductivity in UPt_3 is easily destroyed by substituting a few tenths of a percent of Pt by Pd. Long-range antiferromagnetic order below 6 K is found for a Pd content around 5 at% with values for the ordered moment of about $0.6\mu_B$ per uranium atom [3]. In pure UPt_3, long-range antiferromagnetic order below 6 K has been claimed as well, in this case with values for the ordered moment as small as $0.02\mu_B$ per uranium atom [4]. This order has not been observed in all samples studied so far [5] and might be related to special features of the superconducting state.

Within the composition range up to 10 at% Pd, the pseudo-binary U(Pt, Pd)$_3$ compounds have been found to be single phase. At higher Pd concentrations, additional lines in the X-ray spectrum reveal some superlattice structure or the presence of a second phase. Superlattice structures are easily formed in the UPt_3 phase as non-stoichiometric UPt_3 samples reveal. An excess of 1 at% Pt already induces these additional lines in the X-ray spectrum [6].

The antiferromagnetic long-range order that is found around 5 at% Pd in the U(Pt, Pd)$_3$ system, is visible in the specific heat by a sharp peak near 6 K [2]. For lower and higher Pd concentrations, the specific-heat peak broadens and shifts to lower temperatures. At 10 at% Pd there is hardly any peak structure left in the specific heat and this compound is thought to be at the border between antiferromagnetic order and paramagnetic behaviour. Irrespective of the presence of long-range antiferromagnetic order, all U(Pt, Pd)$_3$ compounds with a Pd content less than 10 at% Pd, belong to the class of heavy fermion systems. Values for the electronic coefficient of the linear specific heat term reach their maximum value near 10 at% Pd. Apparently, the formation of stable uranium moments of $0.6\mu_B$ does not suppress the heavy fermion state at all. This conclusion is compatible with high-field specific heat results for UPt_3 that indicate large values for the electronic specific heat coefficient even in magnetic fields well above the metamagnetic transition field of 20 T [7]. From these experiments we conclude that the short-range antiferromagnetic correlations that are suppressed in magnetic fields above 20 T, are not the only origin for the enhancement of the electronic specific heat coefficient. In this respect it is tempting to compare the low-temperature and high-field behaviour of UPt_3 with that of $CeRu_2Si_2$. In this latter compound the metamagnetic transition occurs for a field of about 8 T applied along the c-axis [8]. The electronic mass enhancement is considered in this case to be partly a single-ion Kondo phenomenon, partly to arise from short-range antiferromagnetic correlations that could be observed in neutron scattering experiments.

In the U(Pt, Pd)$_3$ system we have the possibility to shift the metamagnetic transition field to lower field values by increasing the Pd content. This shift has been studied before in polycrystalline samples. In the present contribution we report on high-field magneti-

sation measurements on three $U(Pt_{1-x}Pd_x)_3$ compounds with x-values of 0.00, 0.05 and 0.10.

2. Experimental

High-field magnetisation measurements were performed on spherical samples (diameter 3 mm) in the Amsterdam High Field Facility. The single-crystalline spheres were cut out of cylindrical batches grown by the Czochralski method in a tri-arc furnace. The single-crystalline batches were investigated by microprobe, X-ray and mass density analyses. No change in composition was found along the cylindrical axis. The lattice parameters, as resulting from the X-ray analyses, hardly show any change with increasing Pd content for the single-crystalline samples. Previous experiments on a more extended series of polycrystalline $U(Pt, Pd)_3$ samples yield a constant a-parameter within the experimental accuracy, $a = 5.752 \pm 3$ Å, and a c-parameter linearly decreasing with Pd content from 4.897 ± 3 Å down to 4.892 ± 3 Å for 15 at% Pd [2]. X-ray measurements over a wider range of diffraction angles on a series of polycrystalline $U(Pt, Pd)_3$ samples reveal extra lines in the high-angle range indicating lattice mismatch for compounds with Pd contents as low as 7 at%. These extra lines most likely reflect anomalies in the stacking sequence along the c-axis and may point to quasi-cubic positions for the uranium atoms.

3. Experimental results

Magnetisation measurements on the $U(Pt, Pd)_3$ system have been performed by De Visser et al. [2] and by Van Sprang [9] on polycrystalline samples at 4.2 K and reveal metamagnetic-like transitions. The field where the metamagnetic transition takes place, gradually changes from 20 T to zero field at increasing the Pd content up to 10 at% Pd. The occurrence of long-range antiferromagnetic order below 6 K around 5 at% Pd apparently does not effect the trends in the metamagnetic transition very much. Low-field (field range 0.6–1.3 T) susceptibility measurements on polycrystalline samples [2, 9] confirm the trend of the high-field magnetisation measurements. The temperature at which the susceptibility has its maximum gradually shifts from 18 K to zero temperature at increasing the Pd content up to 10 at% Pd. Single-crystalline susceptibility data for the 0 and 5 at% Pd compounds are shown in fig. 1.

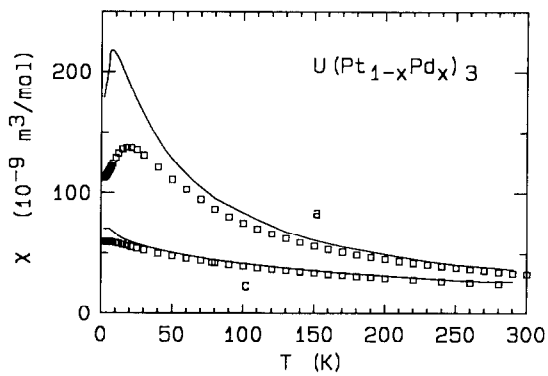

Fig. 1. Low-field susceptibility of single-crystalline UPt_3 (□) and $U(Pt_{0.95}Pd_{0.05})_3$ (full line) along the c-axis and a-axis.

High-field magnetisation measurements on the single-crystalline samples at 4.2 K are presented in fig. 2 for field directions parallel and perpendicular to the hexagonal axis. The magnetisation curves along the c-axis coincide for the three compounds and show a linear behaviour up to a field of 35 T. Perpendicular to the c-axis, the different compounds reach almost the same magnetisation value at 35 T of 5 Am^2/mol, equivalent with about $0.9\mu_B$ per uranium atom. The field dependence of the differential susceptibility as deduced from the magnetisation data of fig. 2, exhibits the same maximum value at 4.2 K of about 400×10^{-9} m^3/mol for all three compounds, although the field where the maximum is reached changes from 20 T for UPt_3 to zero field for the 10 at% Pd alloy.

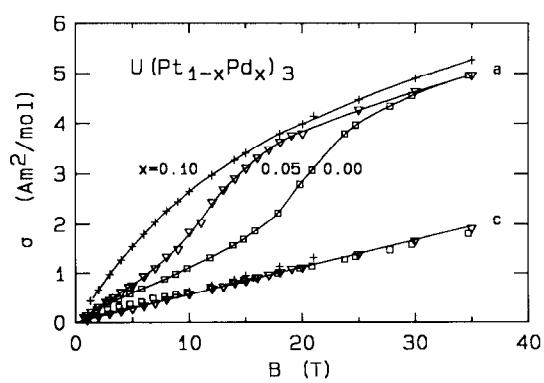

Fig. 2. Magnetisation curves of single-crystalline $U(Pt_{1-x}Pd_x)_3$ compounds at 4.2 K for field directions along the c-axis and the a-axis; $x = 0.00$ (□), $x = 0.05$ (▽) and $x = 0.10$ (+).

4. Discussion

The high-field data of fig. 2 represent magnetisation curves of samples in different magnetic states: no macroscopic long-range order is present in UPt_3 and in the 10 at% Pd alloy, whereas the 5 at% Pd compound is, at least at lower field values, in a state of long-range antiferromagnetic order with well-developed uranium moments of $0.6\mu_B$ approximately. Careful studies of the temperature dependence of the magnetisation in fields up to 18 T on this compound [10] reveal that the metamagnetic transition is still present at temperatures well above the ordering temperature of 5.8 K. Nevertheless, some anomalies have been found in the field dependence of the differential susceptibility at 4.2 K that could point to a double transition in field: one related to the breaking of the long-range order, the second to short-range antiferromagnetic correlations. The same experiments indicate magnetic anisotropy in the hexagonal plane and slightly different values for the transition field along the a- and b-axes. This anisotropy in the hexagonal plane has been confirmed in magnetic torque experiments on the 5 at% Pd compound at 4.2 K in a field of 4 T, see fig. 3. Analysing the 6θ term we calculate an anisotropy in the susceptibility between the a-axis and b-axis of 4.7×10^{-9} m^3/mol, in satisfying agreement with the magnetisation data of De Visser et al. [10]. The torque curves, however, reveal a large asymmetry and point to a complex magnetisation process. In this respect, magnetoresistance measurements reveal some more details. The low-temperature resistivity changes drastically with Pd content from a quadratic temperature dependence for UPt_3 to Kondo behaviour for the 10 at% Pd compound. In the resistivity curve of the 5 at% Pd alloy the transition to the magnetically ordered state is clearly visible by a minimum in the temperature derivative. Applying a magnetic field along the a-axis, this minimum shifts from 5.8 K in zero field to 4.3 K in 8 T. These magnetoresistance measurements reveal large differences below 11 T for field directions along the a- and b-axes, see fig. 4, and are compatible with the neutron results that give the b-axis as the preference direction for the uranium moment.

The present magnetisation measurements on single-crystalline $U(Pt, Pd)_3$ compounds confirm in a quantitative way the qualitative picture deduced before from experiments on polycrystalline samples: the temperature at which the low-field susceptibility takes its maximum (T^*) is intimately connected to the field (B^*) where at low temperatures the metamagnetic transition is found. The energy $k_B T^* \approx \mu_B B^*$ gradually drops from 1.6 meV for pure UPt_3 to zero for the 10 at% Pd compound. This energy is considered to be associated with the reduction of the antiferromagnetic (inter-site) correlations. While the antiferromagnetic correlations can be suppressed by either alloying with 10 at% Pd or by applying a large magnetic field, we emphasize that the heavy-fermion state is preserved as evidenced by the appropriate specific-heat experiments (ref. [2] and references therein). This indicates that the formation of the heavy-fermion state is closely connected to the presence of a second type of interaction, probably Kondo-like (on-site) fluctuations in a similar way as in $CeRu_2Si_2$ [11]. In order to finally

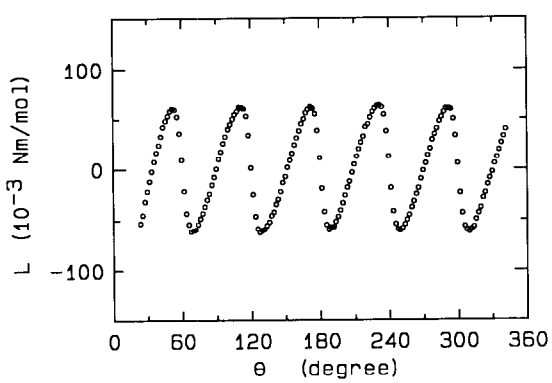

Fig. 3. Magnetic torque curve of single-crystalline $U(Pt_{0.95}Pd_{0.05})_3$ in the hexagonal plane as a function of the magnetic field angle; experiments have been performed at 4.2 K in a field of 4 T.

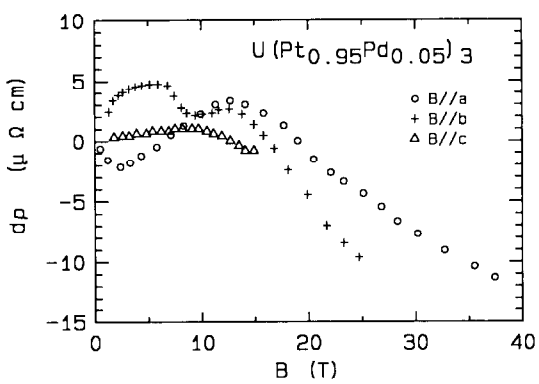

Fig. 4. The change in resistivity as a function of applied field for single-crystalline $U(Pt_{0.95}Pd_{0.05})_3$; experiments have been performed at 4.2 K for field directions along the a-axis, b-axis and c-axis; the current direction is always along the a-axis.

suppress this second type of interaction, Pd concentrations above 15 at% Pd are needed, or magnetic fields far above B^*. The details of the magnetisation process of the ordered 5 at% Pd compound still have to be clarified. Specific-heat experiments on single-crystalline samples of this compound in magnetic fields up to 20 T are now in progress.

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