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RESISTIVITY OF THE HEAVY-FERMION $U(Pt,Pd)_3$ ALLOYS

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Electrical resistivity measurements have been performed on a series of mono- and polycrystalline $U(Pt_{1-x}Pd_x)_3$ compounds ($x \leq 0.15$) in the temperature range 1.4–300 K. On alloying with Pd the spin-fluctuation-like resistivity of pure UPt_3 transforms into a more complex curve for $x = 0.02$ and $x = 0.05$ revealing a transition at 3.3 K and 5.6 K, respectively. At higher Pd concentrations ($x = 0.10$, 0.15) the resistivity curves change to a Kondo-type of behaviour. In the same concentration range a second phase appears in the X-ray patterns.

The effect of substituting Pt by isoelectronic Pd in the heavy-fermion compound UPt_3 is quite intriguing [1]. The low-temperature normal-state properties of pure UPt_3 can be described in terms of a spin-fluctuation model. The main conclusion derived from specific heat [1–3], susceptibility [1] and high-field magnetisation [1] experiments, performed so far on a series of pseudo-binary $U(Pt_{1-x}Pd_x)_3$ alloys, can be briefly sketched as follows: an enhancement, with respect to pure UPt_3, of the spin-fluctuation phenomena for small Pd concentration ($x \leq 0.10$) and a suppression of these phenomena for higher Pd concentration. In conjunction herewith we observe a shift, on alloying, towards lower temperatures and fields, of the anomalies present for pure UPt_3 in the susceptibility at 17 K and in the high-field magnetisation at 21 T (at 4.2 K). Besides these effects new low-temperature anomalies have been observed in the specific heat data of the $U(Pt_{0.98}Pd_{0.02})_3$ and $U(Pt_{0.95}Pd_{0.05})_3$ compounds, at 3.6 K and 5.8 K, respectively. The origin of these anomalies is not clear, but possibly is associated with an antiferromagnetic type of order. Superconductivity ($T_c = 0.5$ K for pure UPt_3 [4,5]) has not been found in these pseudo-binary compounds, not even for very small amounts of Pd ($x = 0.005$).

In order to study the transport properties of these pseudo-binary compounds we have performed electrical resistivity measurements, in the temperature range 1.4–300 K. The resistivity of pure UPt_3 [6], as a function of temperature, reveals a strong increase at low temperatures and a tendency to saturate in the high-temperature regime. The normal-state low-temperature resistivity follows a T^2-law up to approximately 2 K [6–8]. The resistivity of hexagonal UPt_3 is strongly anisotropic, which is reflected in the coefficients of the T^2-term, i.e. 1.6 $\mu\Omega$ cm/K^2 for a current in the basal plane, and 0.7 $\mu\Omega$ cm/K^2 for a current along the hexagonal axis, and in the room-temperature values of 238 $\mu\Omega$ cm (basal plane) and 132 $\mu\Omega$ cm (hexagonal axis), as well. As we will see below, the low-temperature resistivity of pure UPt_3 is strongly modified by alloying with Pd.

Electrical resistivity measurements (a standard four-point low-frequency ac or dc method) have been performed on a number of polycrystalline samples, as well as on small monocrystalline whiskers. The method of preparation and the characterization of the polycrystalline compounds have been described elsewhere [1]. Experiments have been performed on cylindrical samples (diameter = 1.5 mm and length = 4 mm), with a current density of 0.6 A/cm^2. The absolute accuracy in the resistivity values is mainly determined by the uncertainty in the geometrical factor and amounts up to 5%.

The small needle-like whiskers used for this experiment jump spontaneously out of the homogeneous melt under controlled solidification [9]. Since the formation of the whiskers

takes place in less than 10 ms and the diffusion velocities in the melt will have an upperbound of about 0.01 cm/s, we conclude that the composition of the whiskers equals that of the melt. The dimensions of a whisker are roughly 15 (a-axis) × 60 (b-axis) × 1000 (c-axis) (μm)^3. In order to measure the resistivity of such a small sample it was pressed onto an anodized aluminium sample holder by means of four phosphorus-bronze springs, that served as current and voltage contacts. The tiny springs, with sharp edges to ensure well-determined contacts, were shaped by means of a miniature spark erosion technique. The current density ($I \| c$-axis) amounts to 13 A/cm^2. The uncertainty in the geometrical factor has, in this case, an upperbound of 30%.

The experimental results are shown in figs. 1–3. In order to illustrate the relative temperature dependence of the resistivity, the data are plotted as $\rho(T)/\rho(300\,\mathrm{K})$. Absolute values for ρ_0 and $\rho(300\,\mathrm{K})$ are listed in table I. From figs. 1 and 2 it follows that the experimental data on the whiskers roughly coincide with the results obtained on the polycrystalline samples. This illus-

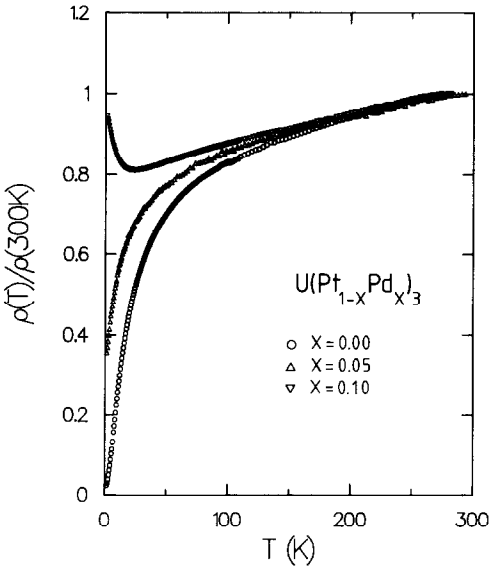

Fig. 1. Reduced resistivity, $\rho(T)/\rho(300\,\mathrm{K})$, of monocrystalline $U(Pt_{1-x}Pd_x)_3$ whiskers, for $x = 0.00$ (○), 0.05 (△) and 0.10 (▽). The current is applied along the c-axis.

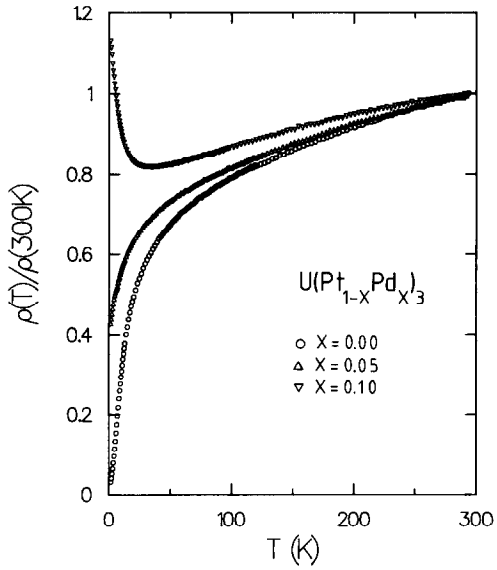

Fig. 2. Reduced resistivity, $\rho(T)/\rho(300\,\mathrm{K})$, of polycrystalline $U(Pt_{1-x}Pd_x)_3$ samples, for $x = 0.00$ (○), 0.05 (△) and 0.10 (▽).

Table I.
Residual resistivity, ρ_0, and room-temperature values, ρ (300 K), in μΩ cm for mono- and polycrystalline $U(Pt_{1-x}Pd_x)_3$ compounds

Compound	Whiskers ($I \| c$-axis)		Polycrystalline samples	
x	ρ_0	ρ (300 K)	ρ_0	ρ (300 K)
0.00	2.2	147	6.1	240
0.01	4.5	126	18.6	264
0.02	6.4	81	36.4	238
0.05	24.6	75	101	251
0.10	46.6	48	633[a),b)]	539
0.15	—	—	355[b)]	334

[a)]Irregular texture.
[b)]Kondo-type compound.

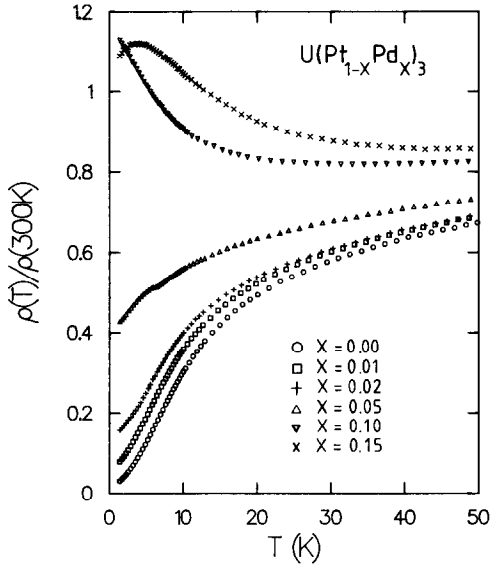

Fig. 3. Reduced resistivity, $\rho(T)/\rho(300\,\mathrm{K})$, of polycrystalline $U(Pt_{1-x}Pd_x)_3$ samples, for $x = 0.00$ (○), 0.01 (□), 0.02 (+), 0.05 (△), 0.10 (▽) and 0.15 (X).

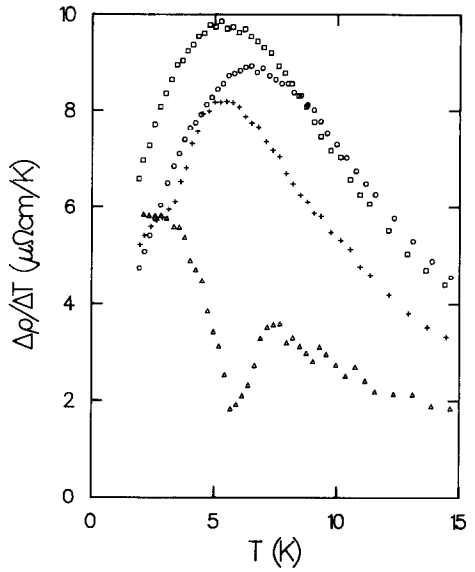

Fig. 4. Temperature derivative of the resistivity, $\Delta\rho/\Delta T$, of polycrystalline $U(Pt_{1-x}Pd_x)_3$ samples, for $x = 0.00$ (○), 0.01 (□), 0.02 (+) and 0.05 (△).

trates that the whiskers indeed may be considered to represent the bulk material. The difference in the absolute values between the two types of samples partly has its origin in anisotropy. Due to preferential orientations a polycrystalline sample has a resistivity almost equal to the basal plane value (238 μΩ cm for pure UPt_3), whereas a whisker represents the c-axis value (132 μΩ cm). Furthermore, cracks and an irregular texture enlarge the resistivity of the polycrystalline samples. This is in particular the case for the high Pd concentrations ($x = 0.10$, 0.15).

An interesting result of our measurements is the gradual change from the spin-fluctuation-like behaviour for pure UPt_3 to a Kondo-like resistivity for high Pd concentration. In combination herewith we observe a decrease in $\rho(300\,\mathrm{K}) - \rho_0$ with increasing Pd concentration. Temperature derivatives, $\Delta\rho/\Delta T$, are plotted in fig. 4 for some selected compounds. The maximum in $\Delta\rho/\Delta T$, at 6.5 K for UPt_3, shifts towards 5.5 K for the 1% Pd sample. For the 2% and 5% Pd samples, $\Delta\rho/\Delta T$ shows an anomaly near the temperature at which a maximum in the specific heat is observed [1]. In particular a sharp minimum occurs at 5.6 K for the 5% Pd sample. The resistivity of the Kondo-like compounds ($x =$ 0.10, 0.15) is shown in a plot of $\rho(T)/\rho(300\,\mathrm{K})$ versus $\ln T$ in fig. 5. A low-temperature maximum in the resistivity, as we observe for $x =$ 0.15 around 5 K, has also been observed by Stewart and Giorgi [10] for an $U(Pt_{0.80}Pd_{0.20})_3$ sample near 20 K. A T^2-temperature dependence, as observed for UPt_3 up to 2 K is not observable in the samples with low Pd contents, in the temperature range investigated.

In searching an explanation for the Kondo-type of resistivity curves of the samples with x equal to 0.10 and 0.15, we compare UPt_3 with UPd_3. Although the distances between neighbouring uranium atoms are very much alike in both compounds, their crystal structures are different. UPt_3 has a hexagonal closed-packed structure ($d_{U-U} = 4.13$ Å), and UPd_3 a double hexagonal one ($d_{U-U} = 4.11$ Å). In UPd_3 two uranium positions exist, hexagonal and quasi-cubic ones, whereas in UPt_3 only hexagonal positions are present. From neutron experiments it has been concluded that UPd_3 has well-localised

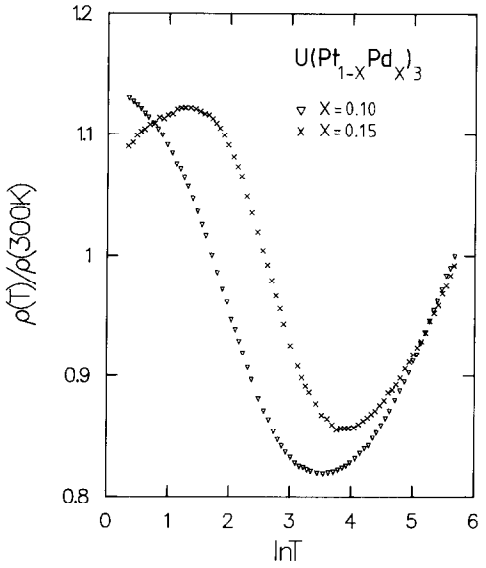

Fig. 5. Reduced resistivity, $\rho(T)/\rho(300\,\mathrm{K})$, versus ln T of polycrystalline $U(Pt_{1-x}Pd_x)_3$ samples, for $x = 0.10$ (▽) and 0.15 (X).

f-electrons, with a $5f^2$ configuration in an L–S ground state [11]. Both uranium states have been claimed to have a singlet ground state and a first excited doublet level, at 24 K (quasi-cubic sites) and at 164 (hexagonal sites), due to crystal-field splitting. The effect of alloying UPt_3 with Pd on the lattice parameter is rather small: the a-parameter remains constant, the c-parameter decreases linearly with Pd concentration: 0.1% for $x = 0.15$. A concentration of 10 percent or more of Pd on the Pt sublattice in UPt_3 could, in principle, be sufficient to destroy, at least partially, the hcp structure, possibly leading to quasi-cubic uranium sites which could act as Kondo impurities. It turned out to be difficult to resolve the dhcp structure from the hcp structure by the X-ray patterns of the polycrystalline samples: any sign of a long-range dhcp structure was lacking and no substantial broadening of the diffraction lines was observable for $x \leqslant 0.15$. On the contrary, analysing the X-ray patterns for higher Pd concentrations ($x = 0.15, 0.20, 0.30$) additional lines are found indicating the presence of a second phase which is not the UPd_3 phase. Whether the Kondo-type of resistivity curves are related to this second phase remains a question yet to be answered.

The most interesting features of our resistivity measurements are the anomalies that are observed in the polycrystalline as well as monocrystalline samples with x values of 0.02 and 0.05. The minimum in the $\Delta\rho/\Delta T$ vs. T curve for the $x = 0.05$ alloy at 5.6 K almost coincides with a peak in the specific heat curve that has been observed for a $x = 0.05$ sample of a different melt at 5.8 K. Experiments on $(U_{1-x}Th_x)Pt_3$ compounds reveal similar anomalies [12,13]. These observations suggest that the compound UPt_3 is close to an electronic instability. Substitutions in the range of a few percent on the uranium or the platinum sublattice cause a transition around 6 K which is rather sensitive to the application of a magnetic field as specific-heat data on the $U(Pt_{0.95}Pd_{0.05})_3$ sample show [1]. The precise nature of the transition, however, is not yet clear. Whereas a second phase starts in the $U(Pt_{1-x}Pd_x)_3$ compounds around $x = 0.15$, the UPt_3 phase seems to be stable in the $(U_{1-x}Th_x)Pt_3$ compounds for x-values up to 0.20 to 0.30. In the thorium-substituted compounds, the anomaly around 6 K has been observed in a $x = 0.20$ sample as well [13], whereas for the $U(Pt_{1-x}Pd_x)_3$ series the anomaly is already absent for the $x = 0.10$ alloy, although the c/T-value at 1.4 K is largest for this alloy.

Why small amounts of Pd in UPt_3 destroy the superconductivity remains puzzling. From our resistivity data we conclude that the absence of superconductivity in a 0.5% Pd compound [1] cannot simply be due to an impurity effect. From table I it follows that the ρ_0-value of a 0.5% Pd sample is between 3 and 10 μΩ cm. An annealed monocrystalline sample of UPt_3 with a ρ_0-value of 6.2 μΩ cm still showed a superconducting transition near 0.5 K [5].

In summary, we have performed electrical resistivity measurements on a series of $U(Pt_{1-x}Pd_x)_3$ compounds for $x \leqslant 0.15$. The spin-fluctuation-like resistivity curve for pure UPt_3 transforms into a more complex curve for the $x = 0.02$ and 0.05 alloys. These curves reflect the anomalies that previously have been observed in the specific heat of samples with the same nomi-

nal composition. At higher Pd concentrations the resistivity curves change to a Kondo-type of behaviour. In the same concentration range a second phase appears in the X-ray patterns.

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Note added in proof

Polarised neutron experiments have unambigiously proven that the transition at 5.8 K in $U(Pt_{0.95}Pd_{0.05})_3$ is of antiferromagnetic nature. The magnetic moments that are involved are of the order of $(0.6 \pm 0.2)\mu_B$ and are directed perpendicular to the c-axis (Frings et al., to be published).

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