

MAGNETIC AND ELECTRONIC PROPERTIES OF PSEUDO-BINARY $U(Pt_{1-x}Pd_x)_3$

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A number of $U(Pt_{1-x}Pd_x)_3$ compounds has been investigated in specific heat, susceptibility and resistivity measurements. Besides for UPt_3 , superconductivity is also found for $x = 0.001$ and $x = 0.002$ alloys. Between $x = 0.02$ and $x = 0.10$ a spin-density-wave-like anomaly is observed in the electrical resistivity at temperatures below 7 K. Measurements on a single-crystalline $x = 0.05$ sample reveal that the anomaly only appears in the basal plane.

The intermetallic compound UPt_3 , being a heavy-fermion superconductor and a spin fluctuation system, has been the subject of a variety of experimental investigations. Part of these aimed to clarify the superconducting state while others concerned the normal state properties. The latter appeared to be strongly anisotropic as was found, for instance, in resistivity, thermal expansion and susceptibility measurements on single-crystalline UPt_3 [1–3]. Both superconducting and normal state properties were found to be extremely sensitive to a substitution of Pt by iso-electronic Pd. No superconductivity has been observed for $U(Pt_{1-x}Pd_x)_3$ compounds with $x \geq 0.005$ down to 40 mK [4]. Higher Pd concentrations ($0.01 \leq x \leq 0.10$) caused an enhancement of the spinfluctuation properties followed by a suppression for $x \geq 0.15$ [4,5]. Furthermore, anomalies were observed in specific heat and resistivity for the $x = 0.02$ and 0.05 compounds at 3.6 and 5.8 K, respectively [4]. In the present paper we report on specific heat, resistivity, susceptibility and high-field magnetisation for a number of $U(Pt_{1-x}Pd_x)_3$ compounds, mainly concentrating on experimental results obtained on single-crystalline $x = 0.05$ samples.

From the starting materials (U with a RRR value of 20, Pt and Pd MRC-MARZ grade) two master alloys UPt_3 and UPd_3 were prepared by arc-melting in a titanium-gettered argon atmosphere. First, the $x = 0.10$ compound was constituted and by enriching with UPt_3 successively polycrystalline compounds with $x = 0.07$, 0.05 , 0.002 and 0.001 were prepared, thereby avoiding

the handling of very small amounts of Pd. The polycrystalline samples were annealed in evacuated sealed silica tubes at 900°C for 7 d. X-ray diffraction measurements on powdered samples confirmed the hexagonal $MgCd_3$ -type of structure. Lattice parameter studies showed a decrease of the c -parameter of $(5 \pm 2) \times 10^{-4} \text{ \AA}$ per at% Pd whereas the a -parameter hardly changes. These values are close to earlier obtained results [4]. From all the compounds single-crystalline whiskers could be obtained [6] indicative for a low contamination content. A large piece (20 gram) of the $x = 0.05$ compound served as the starting material for the growth of a single crystal, which was prepared in a tri-arc melting apparatus by the Czochralski-method [7]. From this large piece two cylinders with diameter of 1.3 mm were spark eroded, one with its cylindrical axis along the crystallographic a -direction (length 5 mm) and the other along the c -direction (length 11 mm).

To investigate the suppression of the superconducting ground state by alloying with Pd, compounds with $x = 0.001$ and 0.002 were checked for superconductivity by a low-frequency ac-susceptibility technique. The compounds have a superconducting transition temperature of 0.460 and 0.357 K, respectively, as determined by the 50% value of the susceptibility step.

Specific heat data on large pieces of polycrystalline samples (mass about 3 g) with Pd concentrations $x = 0.05$, 0.07 and 0.10 were taken by means of an adiabatic method. The results are presented in fig. 1 in which also the data for the compounds with $x = 0.00$ and 0.02 from ref. [4]

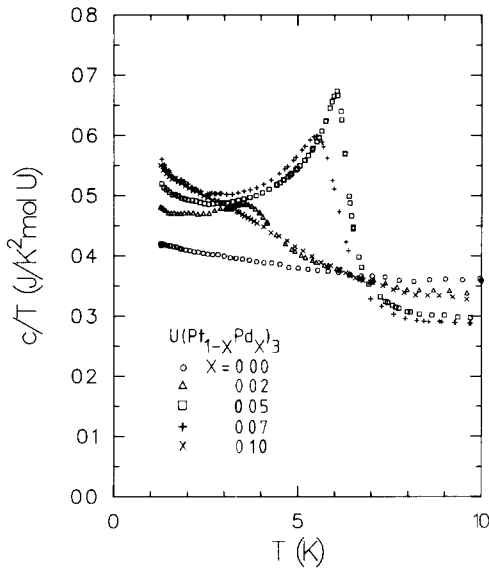


Fig 1 Specific heat data for $U(Pt_{1-x}Pd_x)_3$ compounds in a plot of c/T versus T

are shown. The results for the compounds with $x = 0.05$ and 0.10 closely resemble the earlier results from ref [4]. The presence of the anomaly in the compounds with $x = 0.02$, 0.05 and 0.07 at $T = 3.6$, 6.1 and 5.5 K, respectively and its absence for $x = 0.00$ and 0.10 shows that the corresponding ordering phenomena only exist in a limited concentration range. In the $(U_{1-x}Th_x)Pt_3$ compounds a similar anomaly develops as was measured by Ramirez et al [8]. The anomaly was identified with a spin density wave.

The results of susceptibility experiments on single-crystalline $U(Pt_{0.95}Pd_{0.05})_3$ for two different crystallographic directions are shown in fig 2. These (pendulum magnetometer) data were derived from the linear magnetisation curves in fields lower than 1.3 T. In the same figure the susceptibility of pure UPt_3 [3] is shown. The large anisotropy in the $x = 0.05$ compound is qualitatively the same as for pure UPt_3 . Absolute values are, however, larger in the former case. The susceptibility along the a -direction is much larger than along the c -direction and shows a broad maximum around 8 K, to be compared with the maximum at 17 K in UPt_3 . The inset in fig 2 shows the peak of the a -axis susceptibility for the

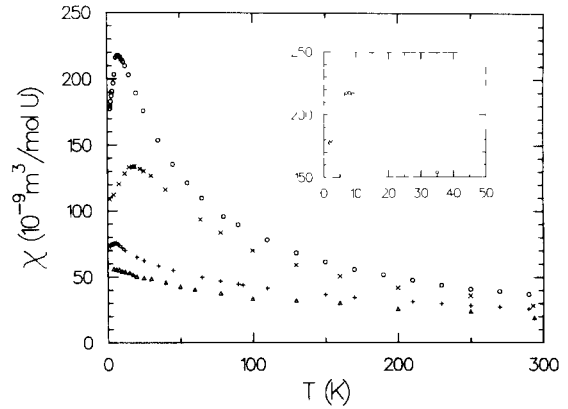


Fig 2 Magnetic susceptibility versus temperature of single-crystalline $U(Pt_{1-x}Pd_x)_3$, along two crystallographic directions ($x = 0.00$ a -axis \times , c -axis Δ , $x = 0.05$ a -axis \circ , c -axis $+$). The inset shows in more detail the susceptibility peak of the $x = 0.05$ compound for the a -axis, the accuracy of the data points is indicated by the size of the markers

$x = 0.05$ compound in more detail. A small irregularity can be observed at 6 K which can be identified with the anomaly in the specific heat at the same temperature. For the c -direction no anomaly can be observed. The small decrease in the susceptibility for the c -direction below 8 K is probably not intrinsic and due to a misalignment of about two degrees of the sample in this particular case. Fits of the high-temperature data to a Curie-Weiss behaviour result in a paramagnetic Curie temperature of -44 (-190) K and an effective magnetic moment of 2.77 (2.81) μ_B per U-atom for the a (c)-direction.

In fig 3 high-field magnetisation data at 4.2 K are shown for the two single-crystalline samples of the $x = 0.05$ compound. The data for single-crystalline UPt_3 [3] are also shown for comparison. Again, the anisotropy is seen not to be much affected by the 5% Pd substitution. The magnetic moment at 35 T is the same for the two compounds whereas the transition observed in the a -direction magnetisation curves is shifted from 21 T for UPt_3 to 11 T for the 5 at% Pd alloy, thereby enhancing the low-field susceptibility. The initial slope of the magnetisation curve amounts to 188 (63) $10^{-9} \text{ m}^3/\text{mol U}$ for the a (c)-direction for this latter alloy.

The next property we investigated is the elec-

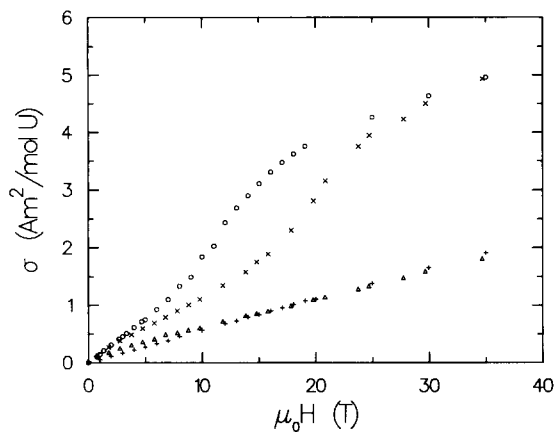


Fig 3 Magnetisation versus magnetic field of single-crystalline $U(Pt_{1-x}Pd_x)_3$ at 4.2 K along two crystallographic directions ($x = 0.00$ a -axis \times , c -axis \circ , $x = 0.05$ a -axis Δ , c -axis $+$)

trical resistivity of $U(Pt_{0.95}Pd_{0.05})_3$ from 1.5 K up to room temperature by a standard 4-point ac method. The resistivity appeared to be as anisotropic as in pure UPt_3 [1]. At room temperature, with the current in the a -direction, the resistivity amounts to $287 \mu\Omega \text{ cm}$, whereas for the c -direction $140 \mu\Omega \text{ cm}$ is found. The resulting residual resistance ratios are 2.4 and 3.7 for the a - and c -direction, respectively. In the low-temperature interval an anomaly appears around 6 K, better visible in a plot of the temperature derivative of the resistivity versus temperature, see fig. 4. In this figure a large dip is located at $T = 5.8 \text{ K}$ for the a -direction. For the c -direction the anomaly is less pronounced. The same anomaly has previously been observed by resistivity measurements on polycrystalline bulksamples and single-crystalline whiskers with $x = 0.02$ and 0.05 [9]. Recent neutron experiments by Frings et al. reveal that the anomaly coincides with the onset of antiferromagnetic order [10].

The close similarity between the low temperature properties of the Pd and Th doped compounds suggests the same origin of the anomalies. Resistivity measurements under pressure on the $x = 0.05$ (Pd) compound show a suppression of the ordering temperature with $\partial \ln T_N / \partial P = -55 \text{ Mbar}^{-1}$ [11]. The pressure

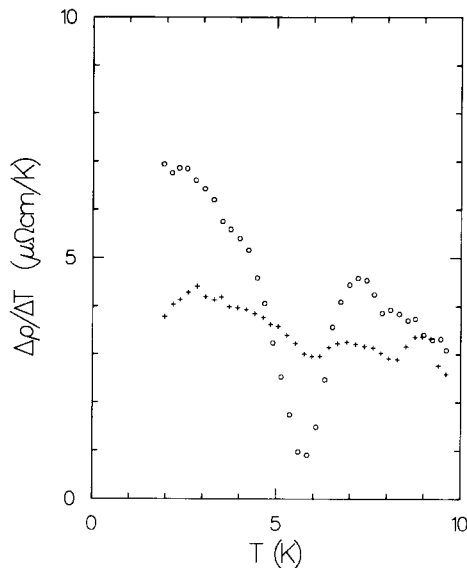


Fig 4 Temperature derivative of the resistivity of single-crystalline $U(Pt_{0.95}Pd_{0.05})_3$, with the current applied along the a -axis (\circ) and along the c -axis ($+$)

effect on the Néel temperature is comparable with that in Cr, a well known spin density wave system [12].

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