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LOW-TEMPERATURE RESISTIVITY MEASUREMENTS OF HEAVY-FERMION $U(Pt,Pd)_3$ ALLOYS

A. DE VISSER, P. HAEN, T. VORENKAMP *, M. VAN SPRANG *, A.A. MENOVSKY * and J.J.M. FRANSE *

Centre de Recherches sur les Très Basses Températures, CNRS, BP 166X, F-38042 Grenoble Cédex, France
** Natuurkundig Laboratorium UvA, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands*

The magnetic and superconducting phase diagram of the heavy-fermion $U(Pt_{1-x}Pd_x)_3$ alloys ($x \leq 0.05$) has been studied in detail by resistivity measurements in the temperature range 0.016 K $< T <$ 10 K. It is confirmed that near 1% Pd neither antiferromagnetism nor superconductivity is observed. Heavy-quasiparticles are still present in the low-temperature limit, as evidenced by the large coefficients of the T^2-term in the resistivity.

The magnetic and superconducting phase diagram [1] of the $U(Pt_{1-x}Pd_x)_3$ alloys has recently been determined by means of specific-heat [2] and resistivity [3] measurements. These studies showed that on alloying the heavy-fermion superconductor UPt_3 ($T_c \cong 0.5$ K, $\gamma = 430$ mJ/molK^2 [4]) with Pd, antiferromagnetism occurs for Pd contents between 2 and 10%, whereas superconductivity is rapidly depressed. The Néel temperature (T_N) is observed as a sharp peak in the specific heat (c), and has a maximum value of ~ 6 K near 5% Pd. However, in the magnetic state c/T does not drop below the values for pure UPt_3, but, instead, starts to rise again at $\sim T_N/2$, leading to considerably larger γ-values than for pure UPt_3. This indicates that the heavy quasiparticles (spin fluctuations), giving rise to a Fermi-liquid behaviour, still exist below T_N. Neutron-diffraction experiments on a 5% Pd sample [5] have shown an antiferromagnetic propagation vector along the b-axis (hexagonal structure). The ordered moment, $(0.6 \pm 0.2)\mu_B$/U-atom, points along the b-axis as well.

Surprisingly, very similar ordering phenomena have been observed in a series of $(U,Th)Pt_3$ alloys [6]. An explanation for the antiferromagnetic ground state in both series of compounds might be found in Fermi-surface instabilities, as first has been suggested by Ramirez et al. [6]. Due to alloying induced small hybridisation changes, a spin-density wave transition takes place, accompanied by the opening of a gap over a part of the Fermi surface. A substantial support for this explanation comes from the type of anomaly present in the resistivity, $\rho(T)$: an upswing below T_N resulting in an additional contribution to ρ.

In the case of the $U(Pt,Pd)_3$ series, $\rho(T)$ measurements (1.4 K $< T <$ 300 K) were presented for polycrystalline samples, as well as for monocrystalline whiskers, for $x = 0.00$, 0.01, 0.02, 0.05, 0.10 and 0.15 [3]. It was concluded that (i) at low Pd contents the spin-fluctuation effects are enhanced (the temperature at which the maximum in $d\rho/dT$ occurs, $T_{max} \propto T_{sf}$, drops with Pd content, implying a lower excitation energy), and (ii) for $x \geq 0.10$ a Kondo-type of curve is present, possibly related with the appearance of a second phase in the same concentration range. The T_N's were observed as sharp minima in $d\rho/dT$: $T_N = 3.6$ and 5.8 K for 2% and 5% Pd, respectively. Due to the limited temperature range in our previous experiments, the $\rho = AT^2$ term, characteristic for the Fermi liquid and present in UPt_3 below ~ 2 K [4], was not observed in the alloyed compounds. The purpose of the present paper is to investigate this term by extending the $\rho(T)$ data down to 16 mK. We also studied the $\rho(T)$ of 0.1 and 0.2% Pd samples, that previously had been characterized by χ_{ac} experiments only.

We report on resistivity measurements on $U(Pt_{1-x}Pd_x)_3$ compounds for $x = 0.00$, 0.001, 0.002, 0.01, 0.02 and 0.05 in the T-interval 0.016 K $< T <$ 10 K. Polycrystalline samples were prepared by arc-melting, single-crystalline samples ($x = 0.00$ and 0.05) by the Czochralski technique. All samples were carefully annealed in a way described elsewhere [4]. Electrical resistivity measurements have been performed with a four-

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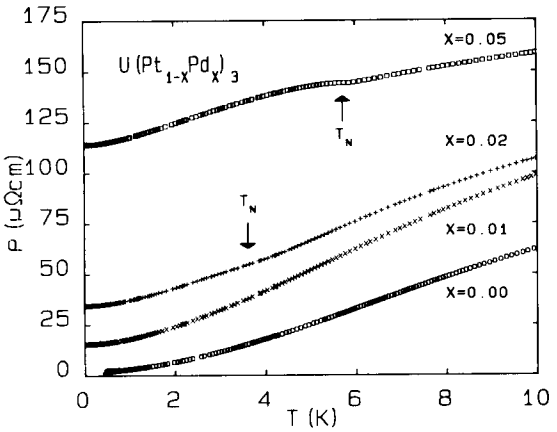

Fig. 1. Electrical resistivity of $U(Pt_{1-x}Pd_x)_3$ compounds for $x = 0.00$ (○) and $x = 0.05$ (□) (single-crystalline samples, $I \| a$), and for $x = 0.01$ (×) and 0.02 (+) (polycrystalline samples).

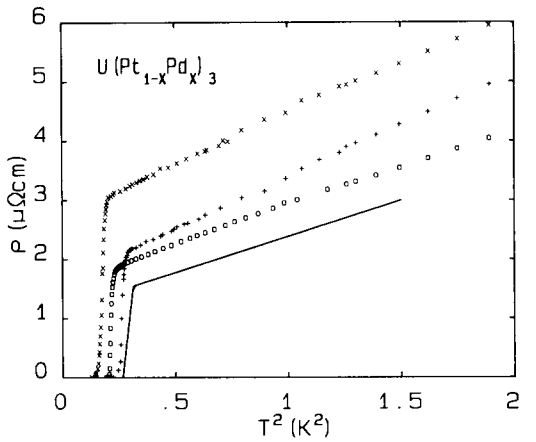

Fig. 2. Electrical resistivity vs. T^2 for single- (○) and polycrystalline (solid line) UPt_3, and for 0.1% (+) and 0.2% Pd (×).

point ac-technique (32 Hz). For temperatures below 1.2 K, the samples were mounted in the mixing chamber of a dilution refrigerator, whereas for $T > 1.2$ K a bath cryostat was used (with the sample immersed in the liquid below 4.2 K). The absolute accuracy of the $\rho(T)$ data amounts to 5%, due to the uncertainty in the geometrical factor.

The experimental results for $x = 0.00$, 0.01, 0.02 and 0.05 are shown in fig. 1. The agreement with previous data (see ref. [1] for 5% Pd and [3] for 1% and 2% Pd) is quite satisfactory. In fig. 2 we have plotted ρ vs T^2 for some selected compounds (UPt_3, 0.1% and 0.2% Pd). An overall collection of resistivity parameters can be found in table 1.

As follows from table 1, the temperature range of the T^2-term decreases on alloying ($T < 0.5$ K for 5% Pd) whereas the coefficient A increases. Since the $\rho(T)$ of hexagonal UPt_3 was found to be strongly anisotropic ($\rho_{ab} \cong 2\rho_c$, see ref. [7]) we calculate average A-values for the single-crystalline samples according to $A_p = (2A_{ab} + A_c)/3 \cong 5A_{ab}/6$. This value for A_p may be compared to the listed values for the polycrystalline samples. Also the value of T_{max} is anisotropic: T_{max} equals 6.2 K and 7.5 K for $I \| b$ and $I \| c$, respectively

Table 1
Room-temperature resistivity (ρ(300 K)), residual resistivity (ρ(0 K)), residual resistance ratio (ρ(300 K)/ρ(0 K)), coefficient (A) and T-interval of the T^2-term, T_{order}, maximum temperature for $d\rho/dT$ (T_{max}), extrapolated γ-values, and the ratio A/γ^2 (in units 10^{-5} μΩcm(molK/mJ)^2), for $U(Pt_{1-x}Pd_x)_3$ compounds

Compound	ρ(300 K) (μΩcm)	ρ(0 K) (μΩcm)	RRR	A (μΩcm/K^2)	T-int	T_{order} (K)	T_{max} (K)	γ (mJ/molK^2)	A/γ^2
UPt_3 [a]	196	1.54	127	1.42(1.18 [d])	$T < 1$ K	$T_c = 0.462$	6.14(6.6 [d])	430	0.77(0.64 [d])
UPt_3 [b]	225	1.16	194	1.22	$T < 1.5$ K	$T_c = 0.53$	n.m.	430	0.66
0.1%Pd [b]	296	1.60	185	1.80	$T < 1.4$ K	$T_c = 0.512$	6.30	435 [c]	0.95
0.2%Pd [b]	256	2.70	94.8	1.78	$T < 1$ K	$T_c = 0.420$	6.20	440 [c]	0.92
1%Pd [b]	272	15.1	18.0	2.96	$T < 0.6$ K	–	5.2	480	1.28
2%Pd [b]	257	34.2	7.5	3.26	$T < 0.5$ K	$T_N = 3.6$	–	527	1.17
5%Pd [a]	270	113.8	2.4	4.40(3.67 [d])	$T < 0.5$ K	$T_N = 5.7$	–	615	1.16(0.97 [d])

[a] Single-crystalline sample ($I \| a$).
[b] Polycrystalline sample.
[c] Estimated value.
[d] Averaged value.
n.m. = not measured

[7]. The averaged value for T_{max} amounts to ~ 6.6 K. By alloying with 1% Pd, T_{max} (and thus T_{sf}) drops by 21%. Assuming $A \propto 1/T_{sf}^2$, the calculated depression of T_{sf} is even larger: 38% for 1% Pd. From both estimates of T_{sf} we conclude a strong enhancement of the spin-fluctuation phenomena. For the pure UPt_3 sample the ratio $A/\gamma^2 \propto (T_F/T_{sf})^2$ (T_F is the Fermi temperature) amounts to 0.65×10^{-5} μΩcm (molK/mJ)^2, which differs somewhat from the universal value for heavy-fermion compounds (1×10^{-5}) noticed by Kadowaki and Woods [8]. Using extrapolated γ-values we calculate that the ratio A/γ^2 passes through a maximum on alloying with Pd (see table 1). This would imply that the mass enhancement is not linear proportional to the effective interaction. However, accurate measurements of the γ-values below 1.4 K are necessary to verify this hypothesis.

The T_c's of UPt_3 (polycrystalline), 0.1% Pd and 0.2% Pd (all samples made with the same type of pure uranium) equal 0.53, 0.51 and 0.42 K (midpoint of transitions). Note that for the monocrystalline UPt_3 sample the T_c is somewhat lower (0.46 K), which is ascribed to the lower purity of the uranium used in this case. Although the resistive T_c's are slightly higher than the inductively observed values (0.46 and 0.36 K for 0.1% and 0.2% Pd, respectively [1]), a strong depression of T_c with increasing Pd content follows. Furthermore, for $x = 0.005$ superconductivity has not been observed above 40 mK [1], and for $x = 0.01$ not above 16 mK (fig. 1). On the other hand, no sign of antiferromagnetic order has been observed in the resistivity curve of the 1% compound. This confirms that in a small concentration range near 1% Pd, neither superconductivity, nor antiferromagnetic order is present. As follows from table 1 the T_c depression is not linear with Pd contents, which is likely related to the non-linear ρ_0-values at these low Pd concentrations. However, as has been pointed out in ref. [4], the depression of T_c originates from several mechanisms, among which the dirt effects probably plays a significant rôle. Recently, it was claimed that UPt_3 is an antiferromagnet below 5 K with an extremely small ordered moment ($0.02\mu_B$) [9]. Antiferromagnetic order at 5 K in UPt_3 is unlikely according to the present findings. However, one cannot exclude that antiferromagnetism in UPt_3 is of a different nature (Fermi surface nesting) and is rapidly destroyed on alloying with Pd [10].

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