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Physica B 223&224 (1996) 178–180

PHYSICA B

The magnetic phase diagram of UPt_3 alloyed with Pd

F.S. Tautz[1], A. de Visser*, M. Mihalik, A.A. Menovsky, J.J.M. Franse

Van der Waals–Zeeman Institute, University of Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands

Abstract

In the course of an investigation of pseudobinary compounds based on heavy-fermion UPt_3, we have prepared $U(Pt_{0.98}Pd_{0.02})_3$ in single-crystalline form. The location in the B–T plane of the antiferromagnetic phase boundary and of the metamagnetic-like transition field has been measured by means of high-field magnetoresistance experiments ($B < 25$ T, $B \| b$). The new results are discussed with reference to the magnetic phase diagrams reported for other compounds in the $U(Pt, Pd)_3$ series.

1. Introduction

The pseudo-binary system $U(Pt, Pd)_3$ is considered as an exemplary heavy-fermion series [1], because it features most of the electronic instabilities that are at the center of the heavy-fermion problem: small-moment and large-moment antiferromagnetism, pseudo-metamagnetism and unconventional superconductivity. As is well known, unconventional superconductivity ($T_c = 0.5$ K) and antiferromagnetic order ($T_N = 5$ K), with extremely small moments ($|\mu| = 0.02\mu_B$/U-atom [2]) coexist in pure UPt_3. On alloying with Pd, superconductivity disappears beyond 0.3 at% Pd, while for larger concentrations antiferromagnetic order (re-)appears in the phase diagram [1]. Investigations of $U(Pt_{0.95}Pd_{0.05})_3$ single crystals by neutron diffraction [3] have revealed this antiferromagnetism ($T_N = 5.8$ K) to be of the conventional type, in that the ordered moment reaches a value of $0.6\mu_B$/ U-atom. However, the relationship between the magnetic order parameter in UPt_3 and the doped compounds is not well understood. For example, we do not know in which fashion the order parameter evolves between the extremes UPt_3 and $U(Pt_{0.95}Pd_{0.05})_3$, and particularly whether there exists any order between 0.3% and ~1% Pd doping.

A second (and related) question pertaining to the pseudo-binary series $U(Pt_{1-x}Pd_x)_3$ is the relationship between the long-range magnetic order, whose emergence can be observed at distinct ordering temperatures $T_N(B)$ and distinct ordering fields $B_N(T)$, and short-range correlations, the suppression of which is connected to a metamagnetic-like anomaly at some characteristic field B^*. UPt_3 has a well-known metamagnetic anomaly at $B^* = 21$ T [4]. In $U(Pt_{0.95}Pd_{0.05})_3$, where $B_N(T)$ has been measured accurately, one finds $B^* = B_N(0) = 12$ T ($B \| b$) [5]. The question then arises whether this equality is purely coincidental or not. Hydrostatic pressure experiments [6] have shown the possibility of splitting B^* and $B_N(0)$ apart. However, the influence of doping concentration on this equality has not been investigated so far. In order to make some progress in this area, we have studied the phase diagram of single-crystalline $U(Pt_{0.98}Pd_{0.02})_3$ in the B–T plane, using the anomaly in the magnetoresistivity at the ordering transition.

2. Experimental

A single-crystalline sample with composition $U(Pt_{0.98}Pd_{0.02})_3$ has been prepared by the Czochralski method in a tri-arc furnace under argon atmosphere. As starting materials we used 5N platinum, 5N palladium and 4N uranium. A single crystal of approximately 5 mm diameter and 25 mm length was pulled from the melt. Electron probe microanalysis ascertained a uniform distribution of the dopant Pd over the whole crystal. The crystal was characterized by elastic neutron scattering

* Corresponding author.

[1] Present address: Technische Universität, D-98693 Ilmenau, Germany.

PII S0921-4526(96)00070-1

experiments at IRI (Delft). We found a mosaicity of 3.5° (with respect to the hexagonal axis) over the whole crystal. The sample used in the present investigation was cut from a part of the crystal with a mosaicity of less then 2°. The electrical resistivity experiments were performed on a bar-shaped sample with the long axis parallel to the a-axis. Measurements in the temperature range 0.4 to 4.2 K in fields up to 8 T were performed in a ^{3}He cryostat equipped with a superconducting magnet. Magnetoresistance experiments at 1.5 and 4.2 K in fields up to 25 T were performed in the pulsed field facility of the University of Amsterdam. In both cases, magnetic fields were oriented along the crystallographic b-axis, with currents running parallel to the a-axis (we define b orthogonal to the a- and c-axis).

3. Results

In Fig. 1 we show the resistivity versus temperature in zero field and applied fields of 2, 4, 6 and 8 T ($B\|b$). The curves have been vertically offset for better clarity. Extrapolating the 0 T curve to absolute zero, we obtain a residual resistivity ($I\|a$) of ~ 27 μΩcm. This compares to 114 μΩcm for the 5 at% Pd compound, and to a value of 1.5 μΩcm for UPt_3 [7]. All five curves in Fig. 1 show an anomaly, which is shifted to lower temperatures by the application of the magnetic field. Differentiating the data clearly reveals a sharp minimum in dR/dT which we

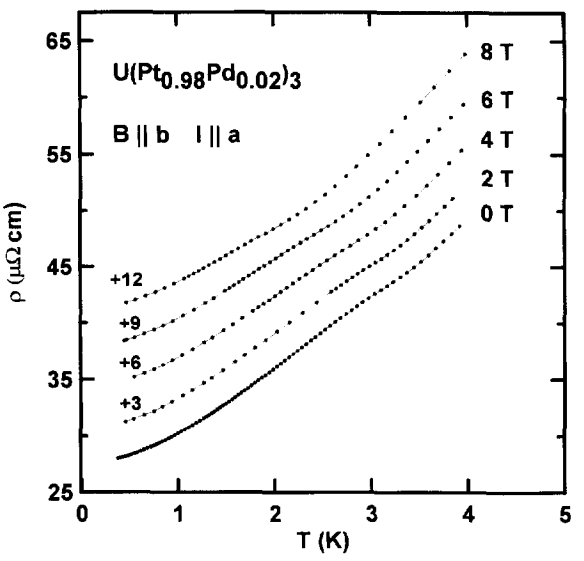

Fig. 1. Electrical resistivity of $U(Pt_{0.98}Pd_{0.02})_3$ ($I\|a$) in zero field and magnetic fields as indicated ($B\|b$). For clarity, the curves in field are offset by 3 μΩcm each. The solid lines are to guide the eye.

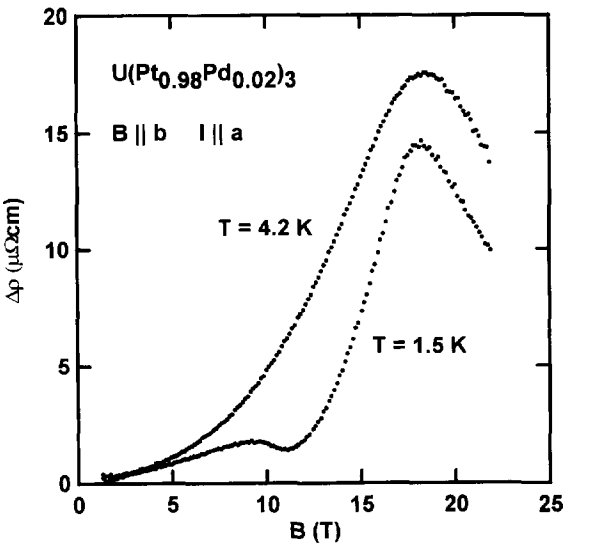

Fig. 2. Magnetoresistance of $U(Pt_{0.98}Pd_{0.02})_3$ ($I\|a$, $B\|b$) at temperatures of 1.5 and 4.2 K.

took as defining $T_N(B)$. Thus T_N decreases from 3.2 K at 0 T to 1.9 K at 8 T. The value $T_N(0) = 3.2$ K should be compared to a value of 3.5 K as determined from the maximum in the specific-heat data taken on a polycrystalline sample [1].

In Fig. 2, we have plotted the magnetoresistance of $U(Pt_{0.98}Pd_{0.02})_3$ at temperatures of 1.5 and 4.2 K and fields up to 22 T. The data are taken from continuous pulses with a total pulse duration of about 1 s. The continuous data have been checked against data points derived from step pulses, where the resistivity is measured under stationary conditions. Both types of pulses give good agreement. From Fig. 2, one sees that at 1.5 K a local maximum appears, which can be assigned to the antiferromagnetic ordering transition. The local maximum at 9 T corresponds well to the suppression field B_N. The absolute maximum at higher fields, present in both the 1.5 and 4.2 K traces, signifies the suppression of short-range antiferromagnetic correlations ($B^* = 18.5$ T). This is to be compared with a value of 16 T as deduced from high-field magnetization measurements on a polycrystalline sample [1].

4. Discussion

Fig. 3 collates the results of the present experiments with data points from earlier investigations on $U(Pt, Pd)_3$ single crystals into a B–T phase diagram ($B\perp c$). For the 5 at% Pd compound, data for $B\|a$ and

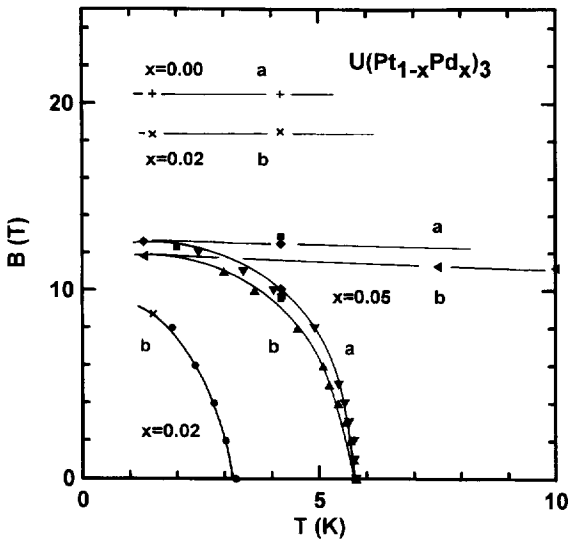

Fig. 3. Magnetic phase diagram for $U(Pt_{1-x}Pd_x)_3$. Symbols have the following meaning. For $x = 0.00$ ($B\|a$): (+)B^* from magnetization [5] and magnetoresistivity [1] experiments. For $x = 0.02$ ($B\|b$): (×)B^* and $B_N(T)$ from magnetoresistivity (this work); (●) $B_N(T)$ from resistivity in field (this work). For $x = 0.05$ ($B\|b$): (▲) $B_N(T)$ from specific heat in field [5]; (◀) B^* from magnetization [8]. For $x = 0.05$ ($B\|a$): (▼) $B_N(T)$ from specific heat in field [5]; (◆) $B_N(T)$ from magnetization [8]; (■) $B_N(T)$ from magnetoresistance [6]; (◆) B^* from magnetization [8]; (■) B^* from magnetoresistance [6]. The solid lines are guides to the eye. Notice that the antiferromagnetic order ($T_N = 5$ K) for pure UPt_3 is not indicated.

$B\|b$ have been reported. The latter give $T_N(B)$ and $B_N(T)$ slightly smaller than the former, which is naturally explained by the fact that the magnetic moment is oriented along the b-axis. For pure UPt_3, the Néel temperature could, so far, not be followed as function of the magnetic field in the basal plane. The pseudo-metamagnetic transition field drops with increasing Pd concentration and is no longer observed for $U(Pt_{0.90}Pd_{0.10})_3$ [1].

Compared to $U(Pt_{0.95}Pd_{0.05})_3$, both T_N and B_N have decreased in $U(Pt_{0.98}Pd_{0.02})_3$. If the phase diagram is plotted with reduced axes T/T_N and B/B_N, the two phase lines for the 2% and 5% Pd compounds fall on top of each other. This is obviously not the case for B^*, which has a value of 12 T in the 5% compound and 18.5 T in the 2% compound ($B\|b$). The equality $B_N(0) = B^*$, which holds in the 5% compound is shown to be accidental by our new results for the 2% compound. While the reduction of the doping level destabilizes long-range order, it stabilises the relevant short-range correlations.

Acknowledgements

We would like to thank the Interfacultair Reactor Instituut in Delft for the possibility to perform neutron-scattering experiments. FST acknowledges support from the European Community.

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