Superconductivity and antiferromagnetic order in the U(Pt,Pd)$_3$ system

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A phase diagram is presented for the superconducting and antiferromagnetic states in the heavy-fermion compounds U(Pt$_{1-x}$Pd$_x$)$_3$, $x \leqslant 0.10$. Superconductivity is depressed for x values larger than 0.005, whereas antiferromagnetism is observed in the composition range $0.01 < x < 0.1$. Specific heat experiments show a sharp transition at the Néel temperature of 6.1 K for $x = 0.05$, in contrast to the alloys with $x = 0.02$ and 0.07 in which rather broad anomalies are observed around 3.6 and 5.5 K, respectively. Experimental observations indicate an optimal condition for antiferromagnetism around $x = 0.05$. The effect of pressure on the phase diagram is discussed.

The intermetallic compound UPt$_3$ is regarded as one of the most interesting heavy-fermion systems because of its unexpected superconducting ground state that develops at $T_c \simeq 0.48$ K.[1] The low-temperature normal state of UPt$_3$ is characterized by pronounced spin-fluctuation effects as indicated by the existence of a $T^3 \ln(T/T^*)$ term in the specific heat[2,3] and in the volume thermal expansion.[3,4] Recent neutron scattering experiments[5,6] further support the concept of spin fluctuations. An intriguing question regarding UPt$_3$ is whether the same many-body interactions cause the superconductivity as well as the spin fluctuations. Although several theoretical approaches[7–9] suggest unconventional odd-parity superconductivity in this system, there is no clear experimental evidence available yet. Note that a conventional type of superconductivity cannot be excluded.[10]

So far, theoretical approaches to UPt$_3$ do not take into account anisotropy, whereas the experiments on UPt$_3$ revealed strongly anisotropic properties. We mention magnetization,[11,12] magnetic susceptibility,[11,12] electrical resistivity,[13] thermal expansion,[4] magnetoresistance,[14] and magnetostriction.[15] The low-temperature behavior turns out to be sensitive to alloying or changes of external parameters such as magnetic field and pressure. A depression of superconductivity by stresses or impurities is a typical example.[1,16] When Pd is substituted for Pt in UPt$_3$ the superconductivity is rapidly lost in the resulting pseudobinary compound U(Pt$_{1-x}$Pd$_x$)$_3$, i.e., below 40 mK for $x = 0.005$. Samples with $x = 0.001$ and $x = 0.002$ had a superconducting transition temperature of 0.460 and 0.357 K, respectively.[17]

The effect of alloying by Pd on the normal state properties is large. Specific heat experiments[16] show that for $x \leqslant 0.10$, the γ value, obtained from an extrapolation to zero temperature, increases with increasing Pd concentration, reaching a maximum value of ~ 600 mJ/K^2 mol U for $x = 0.10$. For higher Pd concentrations the γ value rapidly drops ($\gamma = 30$ mJ/K^2 mol U for $x = 0.30$). Since the large γ value is attributed to spin fluctuations, this increasing behavior indicates further enhancement of the spin-fluctuation effects. An interesting feature is the peak that is found for $x = 0.02, 0.05$, and 0.07 at 3.6, 6.1, and 5.5 K, respectively (see Fig. 1). This peak represents antiferromagnetic order-

ing, as was shown recently. Neutron experiments[18] on the $x = 0.05$ compound gave clear evidence for a long-range antiferromagnetically ordered state with an ordered moment of $0.6 \pm 0.2 \mu_B$ on the uranium sites. It is surprising that in the magnetically ordered state, the c/T value does not drop at all, still keeping its increasing behavior with decreasing temperature. This suggests that the antiferromagnetic state is formed in the spin-fluctuation state without suppressing the spin fluctuations.

The peak at the transition temperature T_N is sharp for the $x = 0.05$ compound, whereas it is less pronounced at both sides of this Pd concentration. The disappearance of the antiferromagnetic transition for $x \geqslant 0.10$ may be caused by a second crystallographic phase. The existence of a second phase has been confirmed by x-ray investigations which we performed on polycrystalline samples with $x \leqslant 0.30$. This second phase has not been identified yet. By applying a magnet-
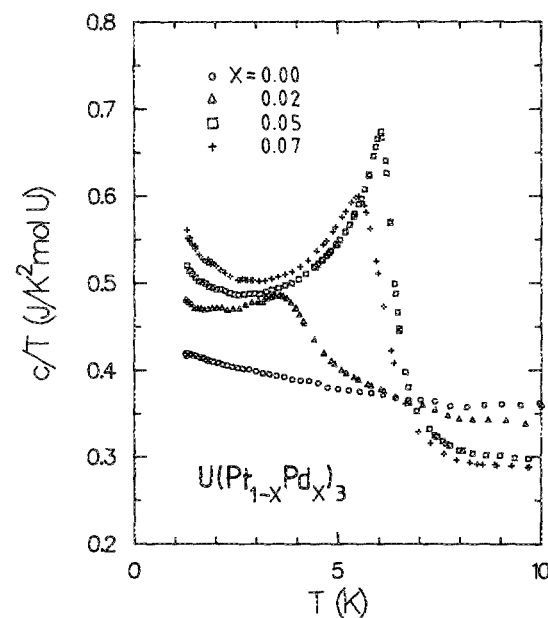

FIG. 1. Temperature dependence of the specific heat of polycrystalline U(Pt$_{1-x}$Pd$_x$)$_3$ compounds.

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ic field of 5 T for $x = 0.05$, the transition temperature is shifted downwards with 0.3 K.[16]

The effect of Pd substitution has also been studied by the electrical resistivity[19] of $U(Pt_{1-x}Pd_x)_3$. In the normal state UPt_3 has a spin-fluctuation-like resistivity curve: a T^2 law is obeyed up to 2 K, followed by a steep increase and a tendency to saturate in the room temperature region. Up to $x = 0.10$ alloying results in a large increase of ρ_0 and a less steep resistivity curve. For $x = 0.10$ and $x = 0.15$ a Kondo-like increase of the resistivity at low temperatures has been found. The antiferromagnetic ordering is for $x = 0.05$ reflected by an anomaly in the resistivity around T_N. For $x = 0.02$ and $x = 0.07$ the anomalies are less pronounced.

Following the experimental results for the $U(Pt_{1-x}Pd_x)_3$ series that have been obtained so far, a phase diagram up to 10 at. % Pd is presented in Fig. 2. From the phase boundaries we expect that in a narrow concentration range $0.005 < x < 0.01$, neither superconductivity nor antiferromagnetism is found. The arrows in Fig. 2 indicate the sign of the pressure effect for pure UPt_3 on the superconducting transition temperature: $\partial \ln T_c/\partial P = -26$ Mbar^{-1} and on the spin-fluctuation temperature T_{SF}: $\partial \ln T_{SF}/\partial P = 30$ Mbar^{-1}.[20] In order to complement these data, resistivity experiments were performed to determine the effect of pressure on the Néel temperature. A standard four-point ac technique was used and helium served as the pressure transmitting medium. It was performed on a $x = 0.05$ single-crystalline sample with the current along the a axis. The data are shown in Fig. 3. The anomaly at $T_N = 5.8$ K shifts to lower temperature with pressure at the rate of -0.3 K/kbar. The relative pressure dependence of T_N amounts to -55 Mbar^{-1}. It is concluded that the superconducting as well as the antiferromagnetic phase are depressed by pressure. However, the relative pressure dependences of T_c and T_N differ by a factor of 2. Another interesting feature is the depression of the resistivity curve with increasing pressure. At 1.4 K the relative pressure dependence of the resistivity along the a axis is -50 Mbar^{-1}, whereas at room

FIG. 3. Temperature dependence of the electrical resistivity of single-crystalline $U(Pt_{0.95}Pd_{0.05})_3$ along the a axis. The sample was spark eroded from a large single-crystalline sample that has been used in a specific heat experiment, yielding $T_N = 5.8$ K (as indicated by the arrow).

temperature it is -6 Mbar^{-1}. This decrease of the resistivity over the whole temperature range can be explained by the pressure effect on the spin-fluctuation phenomena as seen in UPt_3.[13,20,21] Assuming a linear depression of $\rho(1.4$ K$)$ with increasing pressures, it is expected that a pressure of 18 kbar for the $x = 0.05$ compound is nearly sufficient to reach the resistivity values, which are characteristic for UPt_3. Apart from this, the low-temperature resistivity is decreased in such a way that a pressure-dependent ρ_0 cannot be excluded. Since ρ_0 is usually attributed to the impurity concentration in metals, this strong pressure dependence is not easy to account for. It would be necessary to look for new mechanisms, which might be related to pressure-induced changes of the electronic states near the Fermi level.

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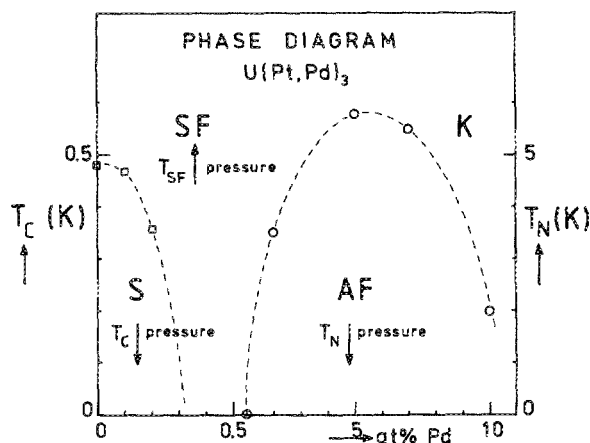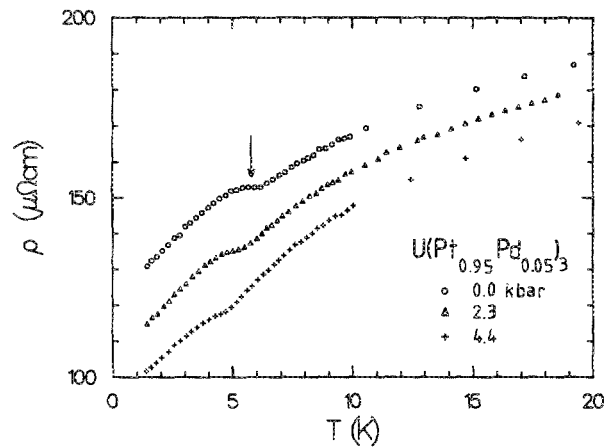
FIG. 2. T_c and T_N as function of Pd concentration for the $U(Pt_{1-x}Pd_x)_3$ system up to 10 at. % Pd. S indicates the superconducting phase, AF the antiferromagnetic phase, K the Kondo regime, and SF the spin-fluctuation region which persists into the AF region. The arrows indicate the sign of the pressure effect on T_c, T_{SF}, and T_N. Note the expanded scale for Pd concentrations below 0.5 at. %.

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