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Specific heat of $U(Pt,Pd)_3$ under pressure

J. Tang[a,*], A. de Visser[b], A. Matsushita[a], T. Matsumoto[a]

[a] *National Research Institute for Metals, 1-2-1 Sengen, Tsukuba, Ibaraki 305, Japan*
[b] *Van der Waals-Zeeman Institute, University of Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, Netherlands*

Abstract

We report on a high-pressure specific-heat study of the heavy-fermion antiferromagnetic $U(Pt_{0.95}Pd_{0.05})_3$. Experiments were carried out on a polycrystalline sample at pressures up to 1.6 GPa in the temperature range 0.7–10 K. The Néel temperature ($T_N = 6.1$ K at ambient pressure) decreases with increasing pressure and vanishes at about the maximum pressure.

Keywords: Heavy-fermion compounds; Antiferromagnetism; Pressure effect; Specific heat

The heavy-fermion pseudobinaries $U(Pt_{1-x}Pd_x)_3$ with $x \leqslant 0.10$ form an appealing system to study the antiferromagnetic instability of the strongly renormalized Fermi liquid [1]. Pure UPt_3 orders antiferromagnetically at a Néel temperature (T_N) of about 6 K, albeit with an extremely small ordered moment ($\mu = 0.02\ \mu_B$/U-atom) [2]. By progressively replacing Pt by isoelectronic Pd, long-range antiferromagnetic order with substantially larger ordered moments is induced for Pd concentrations between ~ 1 and ~ 7 at%. For optimal doping ($x = 0.05$), T_N attains a maximum value of 5.8 K [1, 3] and the ordered moment equals $0.62 \pm 0.05\ \mu_B$/U-atom [4, 5]. The antiferromagnetic ordering for compounds with $x > 0.01$ is of the spin-density wave type, as revealed by the activation-law behaviour in the specific heat and the chromium-like anomaly in the electrical resistivity. Resistivity data under pressure ($P < 0.5$ GPa) [6], taken on a single-crystalline sample, showed that the antiferromagnetic state is rather sensitive to pressure (T_N decreases at a rate of -3 K/GPa), confirming the itinerant nature of the ordering. In this paper we present specific-heat experiments under pressure, which show that the antiferromagnetic state is fully suppressed at a critical pressure $P_c \sim 1.6$ GPa.

A polycrystalline $U(Pt_{0.95}Pd_{0.05})_3$ sample was prepared by arc-melting the constituents in a water-cooled copper crucible in a continuously titanium gettered argon atmosphere. The sample was remelted three times to ensure homogeneity and then cast into the form of a cylinder ($l = 6$ mm, $\varnothing = 3$ mm). The as-cast sample was annealed for one week at a temperature of 900°C. Its Néel temperature ($T_N = 6.1$ K) measured by the specific heat was found to be slightly higher than that of previous samples (5.8 K). The sample was mounted in a copper–beryllium pressure vessel with silver chloride as the pressure transmitting medium. The heat capacity of the assembly of sample and cell was measured using a conventional semi-adiabatic heat-pulse technique. The heat capacity of the sample was then determined by subtracting the contribution of the pressure cell, determined in a separate run, from the total heat capacity.

The specific heat under pressure of the polycrystalline $U(Pt_{0.95}Pd_{0.05})_3$ sample is shown in a plot of c/T versus T in Fig. 1. The data taken at zero pressure are in good agreement with previous results [1, 3]. The contribution of the antiferromagnetic ordering, $\Delta_{af}(c/T)$, appears as

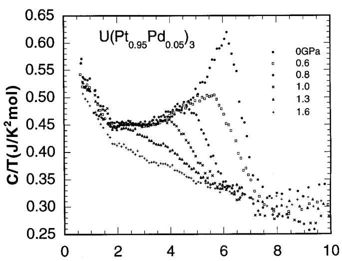

Fig. 1. c/T versus T for polycrystalline $U(Pt_{0.95}Pd_{0.05})_3$ at pressures as indicated.

* Corresponding author. Tel.: + 81 298 531171; fax: + 81 298 531093; e-mail: tang@nrim.go.jp.

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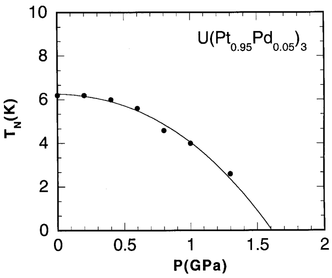

Fig. 2. Pressure variation of T_N for polycrystalline $U(Pt_{0.95}Pd_{0.05})_3$. The solid line is to guide the eye.

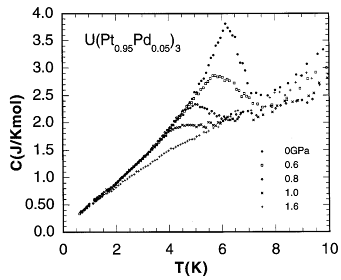

Fig. 3. Temperature dependence of the specific heat of $U(Pt_{0.95}Pd_{0.05})_3$ at pressures as indicated.

a pronounced anomaly superposed on a large heavy-fermion background. In order to determine $T_N(P)$, defined by the temperature of the maximum in $\Delta_{af}(c/T)$, the heavy-fermion background, represented by the data taken at $P = 1.6$ GPa, has been subtracted from the data. $T_N(P)$ is shown in Fig. 2. At low pressures ($P \leqslant 0.4$ GPa) the suppression of the anomaly at T_N is small, but at further raising the pressure the suppression takes place at a much faster rate. At the maximum pressure (1.6 GPa), the anomaly at T_N can no longer be discerned. Besides the considerable broadening of the transition, which finds its explanation in pressure gradients in the polycrystalline sample, it is clear that the transition also weakens with increasing pressure. Fig. 3 shows the relationship between c and T at various pressures. The $c(T)$ data for $P \leqslant 1.3$ GPa overlap each other for $T < 2$ K, within the experimental uncertainty of about 10% (the 'kink' in the plot c/T versus T at 2 K is an artifact of the experiment which could be attributed to the thermometer). This indicates that the heavy-fermion background is rather insensitive to pressure.

A most striking feature of the data shown in Figs. 1 and 2 is that for all pressures the curves overlap for $T < T_N$, as if the curve at $P = 0$ yields a limiting behaviour. Under pressure the ordered moment reduces. The entropy related to the magnetic ordering at $P = 0$ is small, $\sim 0.13R \ln 2$, and is further reduced by applying pressure. The removed entropy should be recovered at $T > T_N$, but due to the large uncertainty for $T > 7$ K and the limited temperature range this is not observable in the data. The low-temperature c/T-values are almost pressure independent, which is surprising as the coefficient of the linear term in the specific heat of UPt_3 ($\gamma = 420$ mJ/mol K^2) is reduced at a rate of -120 (mJ/molK^2)/GPa [7]. This strongly suggests that the reduction of the itinerant ordered moment under pressure coincides with an enhancement of the electronic correlations, which counterbalances the normal suppression of the γ-value.

An interesting observation is that pressure suppresses the magnetic ordering induced by Pd doping, in such a way, that the situation of UPt_3 is recovered at $P_c \sim 1.6$ GPa. Thus, pressure has the same effect as reducing the Pd contents. For instance, the data at $P = 1.0$ GPa for $U(Pt_{0.95}Pd_{0.05})_3$ are very similar to the data for $x = 0.02$ at zero pressure ($T_N = 3.5$ K and $\mu = 0.35 \pm 0.05$ μ_B/U-atom [5]). For $U(Pt_{0.90}Pd_{0.10})_3$, the compound at the verge of vanishing magnetic ordering [1, 3], magnetic order can be induced by pressure [8]. Roughly speaking, the effect of doping 1 at% Pd corresponds to an external pressure of about -0.33 GPa [9]. The strong effects on the exchange parameter and the appearance of antiferromagnetic order in UPt_3 by doping with Pd, can be understood, to a certain extent, by a reduction of the c/a ratio upon alloying (and not by a volume effect as the volume decreases). The effect of pressure is to increase the c/a-ratio again, because of the anisotropic compressibility ($\kappa_c < \kappa_a$) [9]. These effects are, however, subtle and a satisfactory quantitative analysis is hampered by the limited accuracy in the values of the lattice constants of the doped samples and the compressibility.

In summary, we have investigated the effect of pressure on the heavy-fermion antiferromagnet $U(Pt_{0.95}Pd_{0.05})_3$. At ~ 1.6 GPa the magnetic state is fully suppressed, underlining the itinerant nature of the ordering, while the heavy-fermion state is surprisingly robust to pressure.

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