ENHANCEMENT OF SPIN FLUCTUATIONS BY ALLOYING HEAVY-FERMION UPt_3 WITH Pd

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In the course of our investigation of the low-temperature properties of heavy-fermion UPt_3, a number of pseudobinary $U(Pt_{1-x}Pd_x)_3$ compounds has been prepared and their specific heat measured in the temperature range 1.2–30 K. For low Pd concentrations the linear term in the specific heat increases, pointing to an enhancement of the spin-fluctuation effects at low temperatures. Superconductivity has not been observed down to 40 mK in the $U(Pt_{0.99}Pd_{0.01})_3$ sample.

The intermetallic compound UPt_3 has attracted a great deal of interest since its classification in the small group of heavy-fermion superconductors [1,2]. At present a large variety of studies has been carried out over a wide temperature range (40 mK–1000 K), among which are specific heat, thermal expansion, sound velocity, electrical and thermal conductivity, thermopower, susceptibility, magnetoresistance, magnetostriction, reflectivity, photo-emission and neutron experiments. These investigations include the application of high-magnetic fields (35 T) and high pressures (200 kbar). Yet, no satisfactory description in terms of spin-fluctuation phenomena, crystal-field effects, antiferromagnetism and singlet or triplet superconductivity can be offered, as has been pointed out by Franse et al. in a recent review paper [3]. The large value of the linear term in the normal-state specific heat, $\gamma = 422$ mJ/(K^{2}mol U), is characteristic for a highly correlated electron system, thus causing a high effective electron mass, $m \simeq 200 m_0$. We are inclined to believe that the same many-body effects are responsible for remarkable anomalies in the magnetic and transport properties [3].

In our further investigation of the low-temperature properties of UPt_3 we decided to dilute UPt_3 by substituting Pt by isoelectronic Pd. On substituting, a large effect on the coefficient of the linear term in the specific heat is expected since the γ-value for UPd_3 is not larger than 5 or 10 mJ/(K^{2}mol U) (refs. [4] and [5], respectively). Recently some information on alloying UPt_3 became available [6]: from specific heat data taken on a $U(Pt_{0.80}Pd_{0.20})_3$ sample Stewart and Giorgi concluded a substantial suppression of the spin-fluctuation effects compared to pure UPt_3. In our case a number of pseudobinary compounds $U(Pt_{1-x}Pd_x)_3$ has been prepared ($x = 0.01, 0.02, 0.05, 0.10, 0.15, 0.20, 0.30$) and their specific heat has been measured in the temperature range 1.2–30 K. Parts of these results have been published elsewhere [3].

Polycrystalline compounds were prepared by arc melting the appropriate amounts of the pure elements, U (Koch Light, purity 99.8%), Pt and Pd (MRC-Marz grade), in a titanium gettered argon atmosphere. All samples were annealed, in evacuated sealed silica tubes, at 1000°C for a period of 10 days. X-ray diffraction patterns taken on powdered samples at room temperature confirmed the hexagonal $MgCd_3$-type of structure. Samples with $x \geq 0.15$ showed additional unresolved diffraction lines, pointing to at least one second phase. Small needle-like single-crystalline whiskers were obtained, for all pseudobinary compounds, from the arc melted buttons, just as for pure UPt_3 [7]. Lattice parameter determinations from the X-ray diffraction patterns on the powdered samples and on the whiskers ($x = 0.10, 0.20$ and 0.30) show that the a-parameter remains constant within the experimental accuracy, $a = 5.752(3)$ Å, on diluting. The c-parameter decreases linearly with Pd concentration, from 4.897(3) Å for pure UPt_3 down to 4.886(3) Å for $x = 0.30$.

An adiabatic method served to obtain specific heat data on the polycrystalline samples (mass 3–4 g). Data were taken in zero and in a 5 T applied field. The zero field data are presented in fig. 1 in a plot of c/T versus T. On diluting UPt_3 two remarkable features can be observed: (1) for $x \leq 0.10$ the γ-value increases with respect to pure UPt_3, and (2) an anomaly develops at low temperatures for the 2 and 5% buttons. The former observation points to an enhancement of the many-body effects at low temperatures. Although the extrapolation of the linear term in the specific heat to zero K is not unambigious, γ might easily amount to 600 or 700 mJ/(K^{2}mol U) for the 5 and 10% compounds. This signifies a surprisingly large increase of the γ-value with respect to pure UPt_3 with almost 50%. In a magnetic field of 5 T the γ-values are only slightly modified, as indicated by the c/T-values at 1.4 K in fig. 2. The entropy difference, in the temperature interval 1.2–20 K, between the curve for pure UPt_3 and the curve for $U(Pt_{0.80}Pd_{0.20})_3$ equals 2.4 J/(Kmol U). On diluting by

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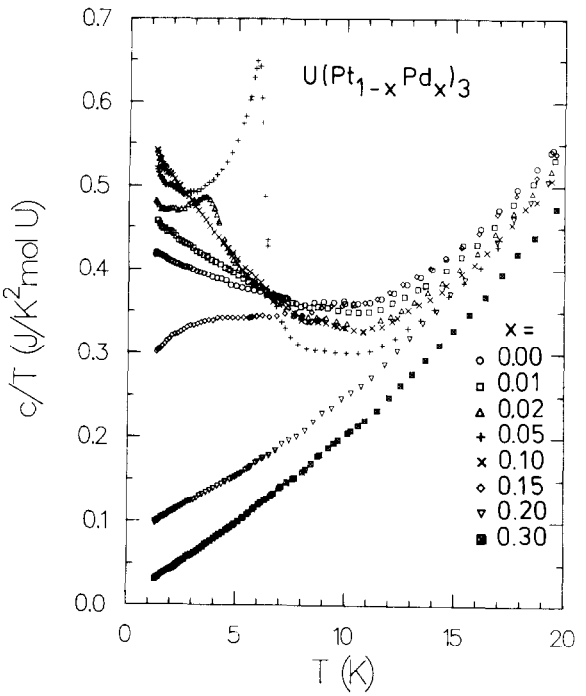

Fig. 1. Specific heat data for $U(Pt_{1-x}Pd_x)_3$ compounds in a plot of c/T versus T, for $x \leq 0.30$.

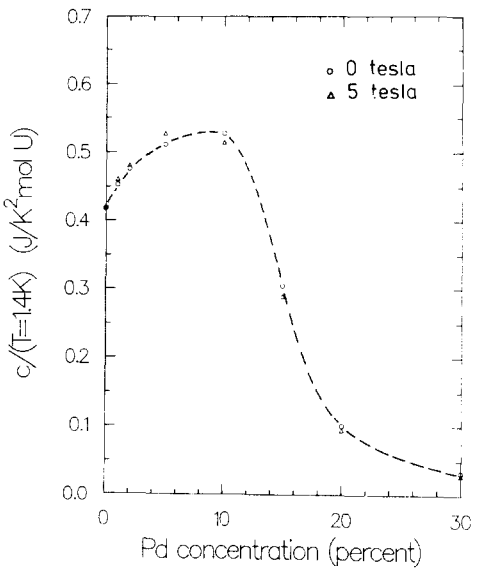

Fig. 2. c/T values at $T = 1.4$ K versus Pd concentration for $U(Pt_{1-x}Pd_x)_3$ compounds ($x \leq 0.30$) in a magnetic field of 5 T (○) and at zero field (△). The broken line serves as a guide to the eye.

Pd, the corresponding entropy differences with the 20% compound remain 2.4 J/(Kmol U), for $x \leq 0.05$. The entropy difference between the curves for $x = 0.20$ and $x = 0.30$ amounts to 1.0 J/(Kmol U) below 20 K.

The specific heat data of UPt_3 have been analysed with a $T^3\ln(T/T^*)$ contribution, characteristic for spin-fluctuation effects [1,2]. A computer fit to the data, in the temperature interval 1.2–10 K, including such a term, reveals a reduction of the characteristic temperature, T^*, from 29 K (pure UPt_3) to 22 K (1% Pd) and 19 K (10% Pd). For $x \geq 0.15$ the spin-fluctuation properties are rapidly lost.

The nature of the anomalies in the specific heat data for the 5% and 2% compounds is not clear, but possibly indicates an antiferromagnetic type of order. In a magnetic field of 5 T the temperature at which the maximum in c/T is observed shifts from 5.8 K to 5.4 K (5% Pd), and from 3.6 K to 3.3 K (2% Pd), but the shape of both peaks remains essentially unchanged. These anomalies remind one on the other hand to the phase transitions in UPd_3 at 5 and 7 K, both crystallographic in nature [4,8]. From entropy considerations it follows that the anomalies in these pseudobinary compounds cannot be due to a second phase of UPd_3 (that might be overlooked in the X-ray patterns). For UPd_3 the excess entropy up to 15 K equals 3 J/(Kmol U) [4], whereas the entropy involved in the peaks of the 5 and 2% samples amounts to 0.8 and 0.2 J/(Kmol U), respectively.

Superconductivity has not been observed in a $U(Pt_{0.99}Pd_{0.01})_3$ sample down to 40 mK. Hence, a destructive influence on the superconducting properties must been concluded from alloying UPt_3 with Pd.

It is a pleasure to thank A.C. Moleman and P.H.M. van Berge Henegouwen for the X-ray analysis of the samples. This work was part of the research program of the Dutch Stichting FOM (Foundation for Fundamental Research of Matter).

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