ENHANCEMENT OF SPIN FLUCTUATIONS BY ALLOYING HEAVY-FERMION UPt₃ WITH Pd

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In the course of our investigation of the low-temperature properties of heavy-fermion UPt₃, a number of pseudobinary U(Ptₓ₋ₓPdₓ)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(Pt₀.₉₆Pd₀.₀₄)₃ sample.

The intermetallic compound UPt₃ 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, γ = 422 mJ/(K²mol U), is characteristic for a highly correlated electron system, thus causing a high effective electron mass, mₑ = 200m₀. 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 UPt₃ we decided to dilute UPt₃ 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 U(Pd₃) is not larger than 5 or 10 mJ/(K²mol U) (refs. [4] and [5], respectively). Recently some information on alloying UPt₃, became available [6]: from specific heat data taken on a U(Pt₀.₈₀Pd₀.₂₀)₃ sample Stewart and Giorgi concluded that the γ-parameter remains constant within the experimental accuracy, γ = 3.752(3) A, on diluting. The c-parameter decreases linearly with Pd concentration, from 4.897(3) A for pure UPt₃ down to 4.886(3) A 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 3 T applied field. The zero field data are presented in fig. 1 in a plot of c/T versus T. On diluting UPt₃ two remarkable features can be observed: (1) for x ≤ 0.10 the γ-value increases with respect to pure UPt₃, 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 unambiguous, γ might easily amount to 600 or 700 mJ/(K²mol U) for the 5 and 10% compounds. This signifies a surprisingly large increase of the γ-value with respect to pure UPt₃ 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 UPt₃ and the curve for U(Pt₀.₈₀Pd₀.₂₀)₃ equals 2.4 J/(Kmol U). On diluting by
Pd, the corresponding entropy differences with the 20% compound remain 2.4 J/(Kmol U), for $x < 0.05$. The entropy difference between the curves for $x = 0.20$ and $x = 0.30$ amounts to 1.0 J/(Kmol U) below 2 K.

The specific heat data of UPt$_3$ have been analysed with a $T \ln(T/T^*)$ 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 (15% 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$_{0.85}$Pd$_{0.15}$)$_3$ sample down to 40 mK. Hence, a destructive influence on the superconducting properties must be concluded from alloying UPt$_3$ with Pd.

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