Specific heat of UPd_2Au_3: Evidence for an unusual heavy-fermion state

B. Andraka
Department of Physics, University of Florida, Gainesville, Florida 32611
(Received 11 August 1995; revised manuscript received 12 February 1996)

Low-temperature specific heat and magnetic susceptibility are reported for $UPd_{5-x}Au_x$ alloys, for $2.5<x<3.2$ and $x=1$. These low-temperature properties reflect a competition between ferromagnetic and antiferromagnetic interactions between U moments. UPd_2Au_3 orders antiferromagnetically at 3 K. Between 5 and at least 14 K, its C/T is proportional to T^2 with a large value of the C/T intercept γ_H of about 670 mJ/K^2 and a slope 3 mJ/K^4 mol. Below T_N, its specific heat has a magnon-derived T^3 term and a linear term ($\gamma_L T$), $\gamma_L=500$ mJ/K^2 mol. Both values of γ_L and γ_H place this alloy into a heavy-fermion category. [S0163-1829(96)06826-9]

According to a recent study by Tran *et al.*,[1] $UPd_{4-x}Au_x$ alloys crystallize into two different cubic crystal structures. UPd_4Au forms in a $AuCu_3$-type structure, while alloys corresponding to x between 2 and 3 form in a $AuBe_5$-type structure. The reported results of the magnetic susceptibility and electrical resistivity indicated the possibility of observing a heavy-fermion state[2] for alloys belonging to this latter structure. In particular, the room-temperature electrical resistivity for the $x=3$ concentration exhibited a Kondo-like increase of the electrical resistivity with a decrease of temperature, while the 4.2 K magnetic susceptibility was strongly enhanced with respect to ordinary metals.

$UPd_{5-x}Au_x$ samples, corresponding to $x=1$, 2, 2.5, 2.8, 3, 3.2, and 3.5, were prepared by a standard inert atmosphere arc-melting technique. UPd_2Au_3 was investigated as cast and after annealing at 800 °C for 10 days. This annealing did not affect either the low-temperature specific heat (above 1.1 K) or the magnetic susceptibility, and therefore all other samples have only been studied as cast. Our $x=2$ sample was two phase. In addition to strong lines corresponding to the $AuBe_5$-type structure, we observed much weaker lines indexable to the $AuCu_3$-type structure. All other investigated alloys with $x\geqslant 2.5$ were single phase, $AuBe_5$-type, within the resolution (about 5%) of our x-ray diffraction analysis. However, the diffraction lines corresponding to the $x=3.5$ alloy were significantly broader and less intense than those corresponding to $x=3$ or 3.2, suggesting an upper x limit of the stability of the $AuBe_5$-type phase. Also, the lattice constant, which increases approximately linearly with x between 2.5 and 3.2, shows a tendency to saturate beyond $x=3.2$. Therefore, we concentrated our study on $UPd_{5-x}Au_x$ alloys with x between 2.5 and 3.2.

Our magnetic susceptibility and magnetization results were in very good agreement with those reported in Ref. 1 for overlapping ranges of temperatures and fields. In general, we have performed measurements to lower temperatures (to 1.8 K), which allowed us to further clarify the magnetic nature of ground states. The complete set of results on these measurements will be published elsewhere. Here we briefly discuss only our low-temperature magnetic data, which complement those presented in Ref. 1 and which will serve as a foundation for a discussion of other thermodynamical and transport properties.

The magnetic measurements reveal a broad range of different magnetic phases in $UPd_{5-x}Au_x$ alloys. Our susceptibility data confirmed ferromagnetic order[1] taking place at about 3.4 K in $UPd_{2.5}Au_{2.5}$. However, this sample showed also thermal hysteretic effects below about 2.5 K. A discrepancy between the zero-field-cooled (ZFC) and field-cooled (FC) magnetizations at 100 G is shown in Fig. 1(a). Thus $UPd_{2.5}Au_{2.5}$ can be viewed as a reentrant spin glass.[3] The lack of a saturation plateau in the 1.8 K magnetization versus magnetic field as high as 5.5 T (not shown) corroborates

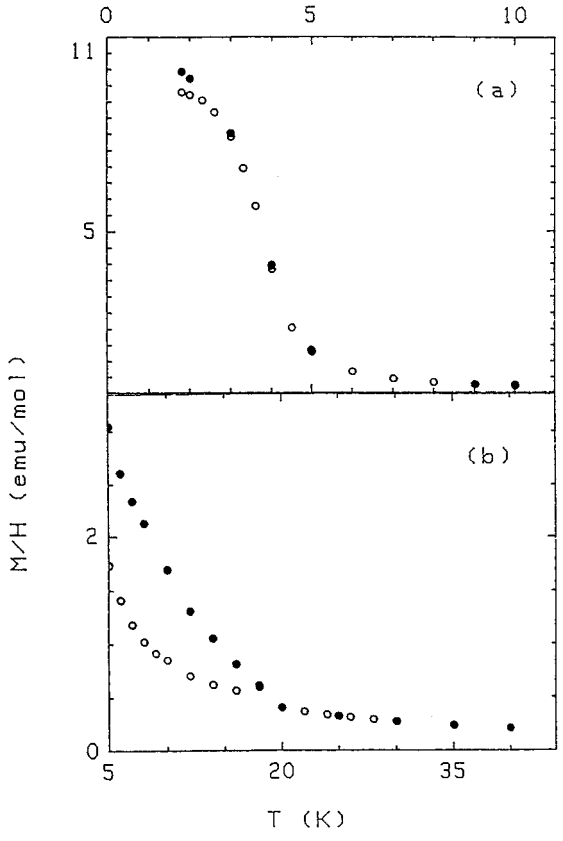

FIG. 1. Magnetization divided by field (100 G) vs temperature for $x=2.5$ (a), $x=2.8$ (b); open dots correspond to the zero-field-cooled, solid dots to the field-cooled data.

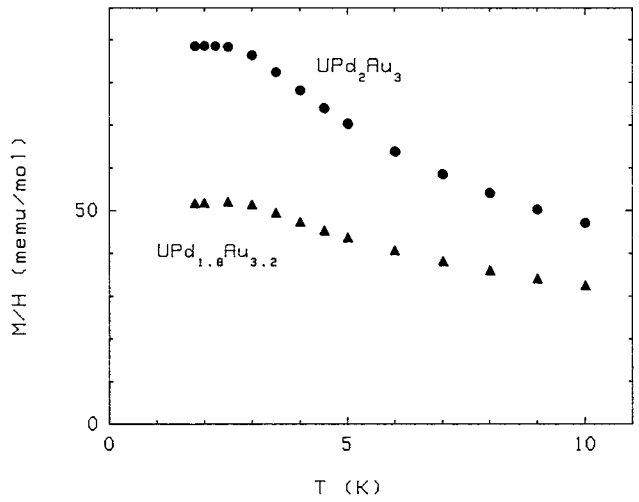

FIG. 2. Magnetization divided by field (5 kG) for $x=3$ and $x=3.2$.

such an interpretation. The discrepancy between ZFC and FC susceptibilities persists to much higher temperatures, about 18 K, in $UPd_{2.2}Au_{2.8}$ [Fig. 1(b)]. At this temperature, the ZFC data for fields of the order 100 G have a small local maximum below which the susceptibility still increases with a decrease of temperature.

On the other hand, we could not detect any thermal hysteretic effects for $x=3$ and 3.2 compositions at accessible temperatures (down to 1.8 K) and fields. The susceptibility data for both these alloys (Fig. 2) exhibit weak maxima near 3 K, suggesting an antiferromagnetic phase transition occurring at this temperature. This interpretation is corroborated by the specific heat data discussed further below.

Additional information on the low-temperature states of the investigated alloys can be obtained from the temperature dependence of the magnetic susceptibility in paramagnetic states when analyzed in terms of a Curie-Weiss law, $\chi^{-1}=3k_B(T-\theta)/\mu_{\rm eff}^2$. θ, obtained from the low-temperature data (below 20 K) for which the Curie-Weiss law approximately holds is a monotonic function of x. It is positive for $x\leqslant 2.8$ and negative for $x\geqslant 3$. Based on this observation and a variety of detected magnetic phase transitions, we can argue for the presence of competing magnetic interactions in these alloys. Alloys with $x\geqslant 3$ are dominated by antiferromagnetic-type interactions, while those corresponding to $x\leqslant 2.8$ by ferromagnetic-type interactions. The spin-glass characteristics detected for some of the samples imply also the coexistence of these interactions. Some magnetic frustration can be anticipated considering a disorder on the Be sites, occupied by Pd and Au, in the $AuBe_5$-type structure. There are two inequivalent Be sites with an occupancy ratio of 4:1. Thus Pd and Au cannot be separated on inequivalent crystallographic sites in the investigated alloys.

Figure 3 shows the low-temperature specific heat data for $x=1$, 2.5, and 3, in the form of C/T versus T^2, for temperatures between 1 and 10 K. A maximum in C/T for UPd_2Au_3 at 3 K is consistent with a bulk phase transition implied by the magnetic susceptibility. At temperatures much lower than 3 K, the specific heat for this alloy has a temperature variation of antiferromagnetic spin waves; i.e.,

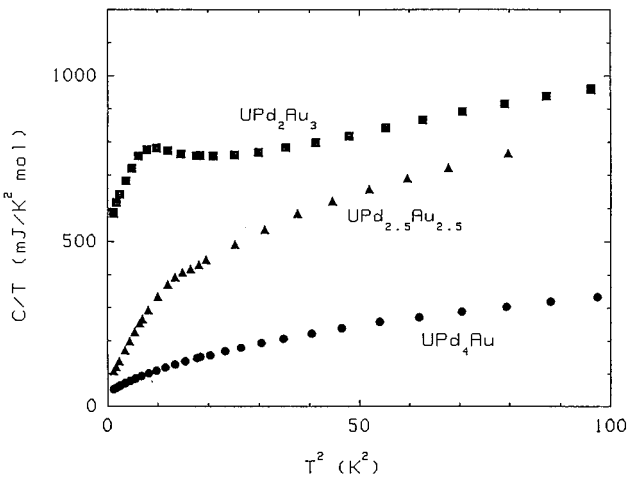

FIG. 3. C/T vs T^2 for $x=1$ ($AuCu_3$-type crystal structure), 2.5, and 3 for temperature lower than 10 K.

C/T is proportional to T^2 (Fig. 4).

The normal-state specific heat of UPd_2Au_3 is anomalous. Between 7 and at least 14 K (data above 10 K are not shown in Fig. 3), C/T changes with temperature as in ordinary metal (C/T is proportional to T^2) with, however, the linear temperature coefficient (γ_H) exceeding that for ordinary metals by three orders of magnitude. γ_H for UPd_2Au_3 is about 670 mJ/K^2 mol. This linear temperature coefficient is reduced by an antiferromagnetic order to about 500 mJ/K^2 mol (γ_L). The slope of C/T versus T^2 above 7 K, or the β coefficient, is about 3 mJ/K^4 mol. In ordinary metals, β is related to the Debye temperature, which measures the stiffness of the lattice. By making a similar assumption for UPd_2Au_3, we arrive at the Debye temperature of 160 K, a value somewhat small, but not unreasonable. The specific heat of $UPd_{1.8}Au_{3.2}$ (not shown, measured between 1 and 10 K) is similar to that of UPd_2Au_3, but shows a broader anomaly associated with the phase transition, which makes the determination of γ_H and especially γ_L less reliable. In any case, its γ_H and γ_L lie in within 10% of the corresponding values found for the $x=3$ composition.

A similar temperature dependence of the specific heat to

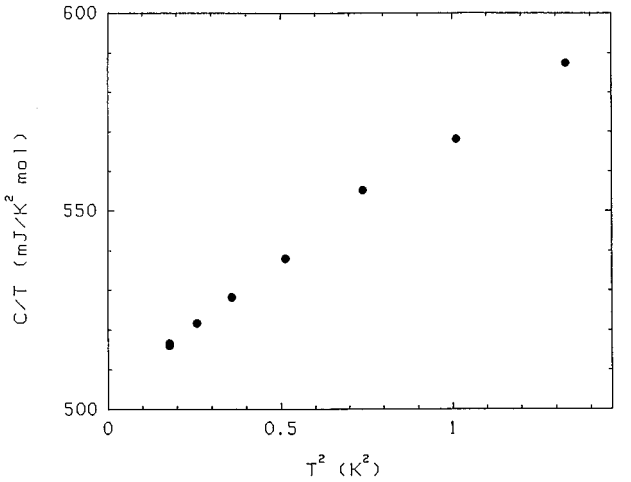

FIG. 4. C/T vs T^2 for UPd_2Au_3 below 1.5 K.

that for UPd_2Au_3 has been previously observed for two other U-based heavy-fermion antiferromagnets U_2Zn_{17} (Ref. 4) and UCd_{11} (Ref. 5). The similarities between UPd_2Au_3 and UCd_{11} are quite remarkable. This latter compound, forming also in a cubic-type crystal structure, orders antiferromagnetically at 5 K. Besides the similar temperature dependence of their specific heats below and above T_N, their γ_L, γ_H, and β values are also alike. The consequence of such a temperature dependence of the specific heat is large entropy at relatively low temperatures. In both UPd_2Au_3 and UCd_{11}, the entropy associated with the linear term of the specific heat exceeds $R\ln 2$ already at temperatures lower than 9 K and $R\ln 3$ at 13 K. Moreover, C/T is still linear in T^2 at 13 K for both systems. Thus either the degeneracy of the crystal field ground state is larger than 3 or crystal electric field states are not well separated in energy. This second scenario is more plausible considering all other more thoroughly investigated U-based heavy-fermion systems. To date,[6] there has been no single, direct evidence reported (like inelastic neutron scattering) of crystal electric field levels in any U system with strongly enhanced C/T at $T=0$.

The lack of any theoretical understanding of such a temperature dependence of the specific heat in UPd_2Au_3, the lack of any information on crystal fields, and the large measured entropy values makes the interpretation of γ_H and γ_L difficult. By no means can we directly relate these values to the electronic density of states at the Fermi level as it is in ordinary metals. At the same time, the order of magnitude of γ_H and γ_L is a clear indication of the heavy-fermion state in UPd_2Au_3.

Other investigated $UPd_{5-x}Au_x$ alloys corresponding to $x=1$ and 2.5 (Fig. 3) have much smaller, but still enhanced, C/T values for $T\rightarrow 0$. (Note that UPd_4Au belongs to the $AuCu_3$-type crystal structure.) The temperature dependence of their specific heats is also interesting and will be further investigated in the future. For both alloys, C/T above 5 K increases faster than linearly, but slower than quadratically, with temperature.

Although an antiferromagnetic order clearly manifests itself in the specific heat and magnetic susceptibility for UPd_2Au_3, there is no indication of any phase transition in the electrical resistivity.[1] This observation can be understood in terms of two noninteracting or only very weakly interacting electronic subsystems, e.g., $5f$ electrons of U, responsible for the susceptibility and the specific heat, and conduction electrons determining the low-temperature electronic transport. However, further inspection of the temperature variation of the resistivity bears witness to rather unusual interactions between conduction electrons and U moments. Tran *et al.*[1] have found that the resistivity for UPd_2Au_3 below 5 K and down to at least 1.1 K changes with temperature as $T^{1.5}$. Our measurements in the same temperature regime confirmed such a temperature variation. Moreover, our measurements in the ^{3}He range, to about 0.35 K, are also consistent with this temperature dependence.

The $T^{1.5}$ temperature variation has been theoretically predicted for nearly antiferromagnetic metals[7] and experimentally observed[8] in some antiferromagnetic materials just above the critical pressure separating magnetic and paramagnetic phases. Although UPd_2Au_3 order antiferromagnetically according to its magnetic susceptibility and the specific heat, it can also be viewed as a limiting case of the antiferromagnetism since it lies near the borderline in the alloy parameter space separating ferromagnetic and antiferromagnetic phases. To the best of our knowledge, there have not been other reports of such an exotic (non-Fermi-liquid-like) temperature dependence of the resistivity for systems with competing antiferromagnetic and ferromagnetic interactions.

In summary, we report a heavy-fermion system UPd_2Au_3 with an unusual temperature dependence of the specific heat and electrical resistivity. The magnetization results for $UPd_{5-x}Au_x$ indicate the presence of competing antiferromagnetic and ferromagnetic exchange interactions. Alloys dominated by ferromagnetic interactions have moderate values of C/T for $T\rightarrow 0$; alloys dominated by antiferromagnetic interactions have large C/T at $T=0$ K.

The author acknowledges fruitful discussions with G. R. Stewart and P. Kumar. This work was supported by the National Science Foundation, Grant No. DMR-9400755.

[1] V. H. Tran, Z. Zolnierek, and R. Troc, J. Alloys Compounds **196**, 219 (1993).

[2] For reviews of the field, see G. R. Stewart, Rev. Mod. Phys. **56**, 755 (1984); N. Grewe and F. Steglich, in *Handbook on the Physics and Chemistry of Rare Earths*, edited by K. A. Gschneinder, Jr. and L. Eyring (Elsevier, Amsterdam, 1990), Vol. 14, Chap. 97.

[3] M. J. P. Gingras, in *Magnetic Systems with Competing Interactions (Frustrated Spin Systems)*, edited by H. T. Diep (World Scientific, Singapore, 1994), p. 238.

[4] H. R. Ott, H. Rudigier, P. Delsing, and Z. Fisk, Phys. Rev. Lett. **52**, 1551 (1984).

[5] Z. Fisk, G. R. Stewart, J. O. Willis, H. R. Ott, and F. Hulliger, Phys. Rev. B **30**, 6360 (1984).

[6] F. Steglich, C. Geibel, R. Modler, M. Lang, P. Hellmann, and P. Gegenwart, J. Low Temp. Phys. **99**, 267 (1995).

[7] K. Ueda, J. Phys. Soc. Jpn. **43**, 1497 (1977).

[8] M. Kamada, N. Mori, and T. Mitsui, J. Phys. C **10**, L643 (1977).