RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 34, NUMBER 1 1 JULY 1986

Heavy-fermion behavior in a pseudobinary system: $U(In_{1-x}Sn_x)_3$

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(Received 24 April 1986)

The temperature dependence of the magnetic susceptibility, the low-temperature specific heat, and the field dependence of the high-field magnetization were measured for the pseudobinary system $U(In_{1-x}Sn_x)_3$ for $0 \leq x \leq 1$. The results show a clear evolution from long-range antiferromagnetism for $x < 0.45$ (In rich) to a heavy-fermion region for $0.45 \leq x \leq 0.80$ and to highly enhanced paramagnetism for $x > 0.8$ (Sn rich). These results represent the first systematic study of the onset of the salient features associated with heavy-fermion behavior in a pseudobinary system.

Heavy-fermion (HF) metallic systems are characterized by an extremely large low-temperature electronic coefficient of specific heat ($\gamma > 400$ mJ/mol K^2) and several other low-temperature properties which reflect the presence of a contribution at the Fermi energy of electrons with an anomalously large effective mass ($\sim$200 times the free-electron mass).[1] Superconductivity, long-range magnetic order, and enhanced but nonordering paramagnetism have been reported for HF systems. To date all the identifiable HF systems have involved Ce and light actinide-based stoichiometric intermetallic compounds, e.g., $CeCu_2Si_2$, UBe_{13}, UPt_3, and $NpBe_{13}$.[1] The loosely bound f electrons of the Ce and lighter actinides and their hybridization with the *spd* electrons are responsible for the rather remarkable properties reported for the HF systems. Theoretical attempts to describe these systems have included one-electron, Landau-Fermi liquid, and many-body models.[2]

In this Rapid Communication we report the first observation of HF behavior in a pseudobinary system. An important feature of $U(In_{1-x}Sn_x)_3$ is that the HF regime ($0.5 < x < 0.8$) is well removed from the antiferromagnetic regime ($x < 0.45$) as well as the enhanced paramagnetic Fermi-liquid regime ($x > 0.80$). Thus HF behavior can be fine tuned by varying the Sn to In ratio. Experiments performed were low-temperature heat capacity C, for 1.3 K $< T <$ 15 K, electrical resistivity ρ, and magnetic susceptibility χ, over the temperature range 1.2 K $\leq T <$ 300 K. For $x = 0.6$, C was measured down to 0.4 K and in magnetic fields H, to 10 T.[3] In addition, room temperature lattice constant measurements and high-field (to 20 T) magnetization M, measurements at $T = 4.2$ K and 1.3 K were made.

In the light actinide (Ac) and Ce-based intermetallic compounds, the Ac-Ac (Ce-Ce) distance is often cited as an important determining factor for the existence of localized moments on the Ac (Ce) ion. For $d_{\text{Ac-Ac}} < 3.4$–3.6 Å, the f-f overlap between neighboring U ions is sufficiently large to quench magnetic moment formation.[4] For $d_{\text{Ac-Ac}} > 3.4$–3.6 Å, magnetic moment formation can occur but is mediated by f-*spd* hybridization. For the stoichiometric intermetallic compounds UX_3 with $X =$ Si, Ge, Sn, In, and Pb, $d_{\text{U-U}}$ is larger than 4 Å. Thus f-*spd* hybridization is the controlling interaction which influences the rich variety of magnetic properties found for these systems.[5,6] For example, USn_3 is a highly enhanced paramagnetic system which does not order magnetically, and previous studies of pseudobinary alloys of USn_3 and the antiferromagnetic UPb_3 ($T_N \simeq 32$ K) reveal the appearance of antiferromagnetism at only 8 at.% Pb substituted for Sn.[7] In the present study of $U(In_{1-x}Sn_x)_3$ antiferromagnetism does not occur until more than 55 at.% In is introduced into the Sn sublattice. This study shows HF behavior for lower In concentration, well removed from the critical concentration for the onset of long-range antiferromagnetism. Such behavior shows the role of f-*spd* hybridization on the evolution of magnetic behavior from the highly enhanced paramagnetic regime to HF behavior and then to the long-range magnetically ordered regime. It is noteworthy that In and Sn are nearly identical in size, so presumably any electron concentration and subtle Fermi surface effects mediate the f-*spd* hybridization needed for HF behavior.

The polycrystalline samples used in this study were prepared in an inert atmosphere arc furnace and annealed in vacuum at 600 °C for about four days. A small weight loss primarily due to the evaporation of the more volatile constituents (In and Sn) occurred during arc melting. Additional In and Sn were added in proportion to their relative vapor pressures and consistent with the weight loss obtained in melting the terminal compounds. Assuming these estimates were correct, the final weight of the pseudobinary alloy was within 0.2% of the expected weight for a stoichiometric ratio of atoms. The concentrations given are the nominal concentrations corrected for the small weight losses. The $U(In_{1-x}Sn_x)_3$ system crystallizes with

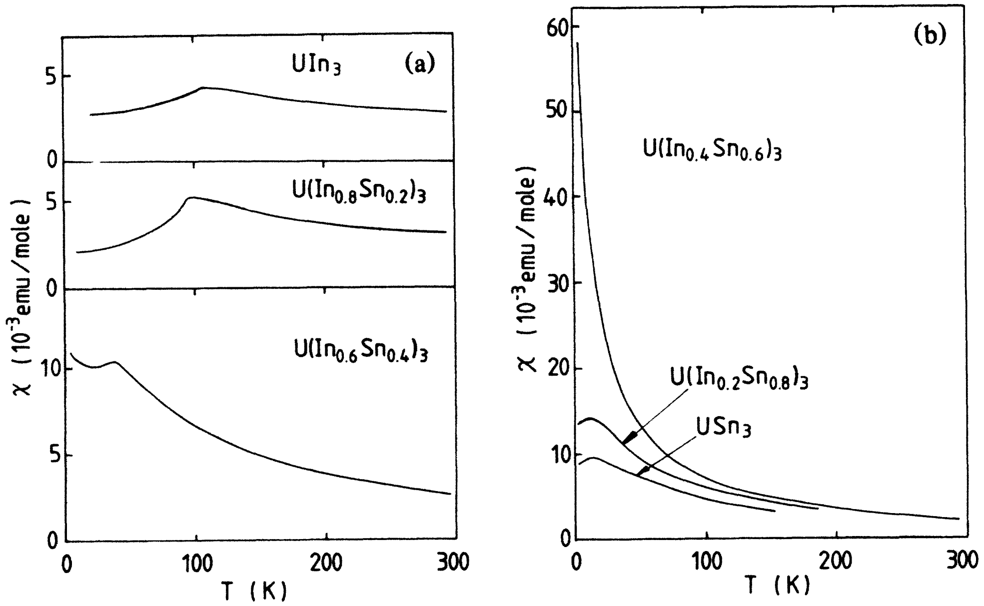

FIG. 1. Low-field magnetic susceptibility vs temperature for several $U(In_{1-x}Sn_x)_3$ samples.

the ordered Cu_3Au structure and room temperature x-ray measurements indicate the samples were all single phase. The lattice constant, a_0, vs x followed Vegard's law with $a_0=4.601$ Å for UIn_3 increasing linearly to $a_0=4.610$ Å for USn_3. Heat capacity for $1.3\ K < T < 15\ K$ was performed on samples of about 3–5 g using a conventional heat pulse technique. For $x=0.6$, a small sample relaxation method was used down to 0.4 K and in magnetic fields H to 10 T.[3] The electrical resistivity was measured on rectangular pressure cast bars ($10\times 1\ mm^2$) using a conventional four-probe dc technique. Vibrating sample magnetometers were used for all magnetic measurements.

The widely varying magnetic properties of $U(In,Sn)_3$ are shown in Figs. 1(a) and 1(b) which are representative χ vs T data on six of the 12 samples investigated. The measuring field was typically 1 T. However, for those samples where $M(H)$ was nonlinear for $H < 1$ T, lower fields were used, so that $\chi(T)$ data are the low-field limits. For $x=0$, 0.2, and 0.4, well-defined cusps in the $\chi(T)$ indicate the onset of antiferromagnetic order at the Néel temperature, T_N. The value of T_N for UIn_3 ($T_N=108$ K) agrees well with previous work.[8] In Fig. 1(b), for decreasing x starting with $x=1.0$, $\chi(T)$ shows a weak Curie-like behavior at high temperatures with $\chi(T)$ nearly temperature independent as $T\rightarrow 0$ K. For $x=1.0$ and $x=0.8$, weak maxima in $\chi(T)$ are seen at $T=12$ K and 6 K, respectively. Note, the strong enhancement of $\chi(0)$ as x decreases; $\chi(0)$ reaches its largest value in the vicinity of $x=0.6$ which is well removed from the critical concentration ($x<0.45$) for the onset of antiferromagnetism.

Figure 2 shows heat capacity data for four representative samples, $x=0.5$, 0.6, 0.8, and 1.0. The data for USn_3 agree well with previous work.[9] The data in Fig. 2 are displayed as C/T vs T^2 for the low-temperature region $T<15$ K. The distinguishing feature is the pronounced enhancement of C/T as x approaches 0.6 followed by a decrease in the enhancement with further In substitution. For $x=0.6$, C/T reaches 530 $mJ/mol\,K^2$ at 1.3 K. Subsequent measurements to lower T revealed a weak maximum in C/T vs T at about 0.7 K. This maximum is strongly suppressed by a magnetic field. This broad feature is reminiscent of the coherence anomaly seen in $CeCu_2Si_2$ and $CeAl_3$ by Bredl *et al.*[10] rather than magnetic ordering. Details of these measurements will be reported elsewhere.[3]

Figure 3 shows the electronic specific heat coefficient $\gamma(\gamma=C/T)$ at 1.35 K for $x\geq 0.45$ and the antiferromagnetic-paramagnetic phase boundary for $x\leq 0.4$. As mentioned above, the maxima in γ and χ at $T=1.3$ K occur at $x=0.6$, which is well removed from the antiferromagnetic region. Thus, the enhancement in γ and χ cannot be attributed to short- or long-range magnetic ordering. A further indication that this region represents HF or at least narrow f-band behavior is illustrated in Fig. 4,

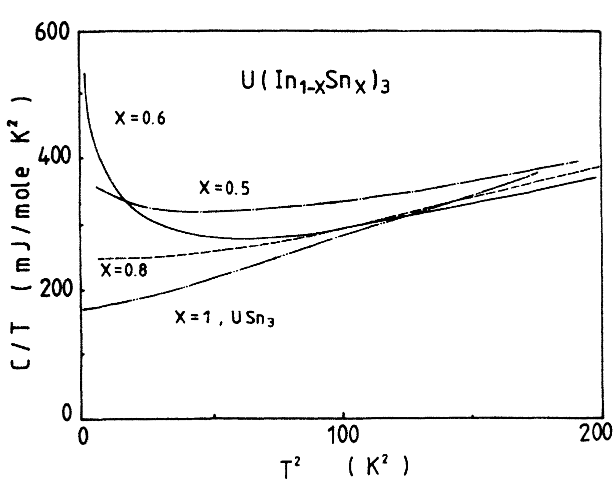

FIG. 2. Specific heat, C, divided by temperature, C/T, vs T^2 for several $U(In_{1-x}Sn_x)_3$ samples.

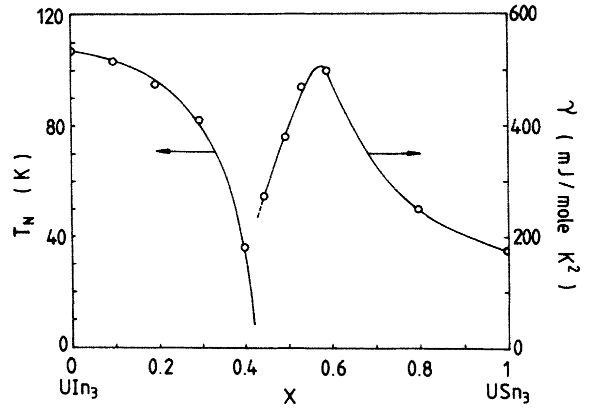

FIG. 3. Néel temperature, T_N, and low-temperature electronic specific-heat coefficient, $\gamma = C/T$, at $T = 1.3$ K vs x for $\mathrm{U(In_{1-x}Sn_x)_3}$.

where M vs H at 4.2 K is shown for four samples. The degree of nonlinearity in M vs H correlates with the enhancement in γ and $\chi(0)$. As seen in Fig. 4, M vs H is linear up to 9 T for $\mathrm{USn_3}$, which is in the enhanced paramagnetic regime. It is also linear to 9 T for all antiferromagnetic samples. The nonlinearity increases as x approaches 0.6, then decreases again. This parallels the variation of γ and $\chi(0)$ vs x. We believe the strong nonlinearity in M vs H can be attributed to a narrow f band, rather than conventional localized moment behavior. If the latter were the case, one might expect a magnetically ordered state to exist at finite temperatures, whereas none was detected down to 1.3 K for any of the sample in Fig. 4. For $x = 0.6$ sample, no magnetic order was observed down to 0.4 K.[3]

In Fig. 5, we show $\gamma(0)$ and $\chi(0)$ for several cubic isomorphic $\mathrm{U}X_3$ compounds and $\mathrm{U(In_{1-x}Sn_x)_3}$. For the $\mathrm{U(In_{1-x}Sn_x)_3}$ system and for $x < 1$, $\gamma(1.3\ \mathrm{K})$ and $\chi(1.3\ \mathrm{K})$ are shown rather than $\gamma(0)$ and $\chi(0)$ because of the uncertainity in the extrapolation to $T = 0$ K for both γ and χ. The χ values shown are the measured susceptibilities and have not been corrected for spd-band contributions, which are expected to be small. The solid line in Fig. 5 is derived from a free-electron model. Assuming the deviations from free-electron behavior can be attributed to spin fluctuation enhancement and neglecting spd contributions to χ, the $\mathrm{USi_3}$, $\mathrm{UGe_3}$, and $\mathrm{UAl_3}$ are only slightly enhanced. However, $\mathrm{U(In_{0.4}Sn_{0.6})_3}$ becomes strongly enhanced with a Stoner enhancement factor more than 10. The Stoner factor has a maximum at $x = 0.6$ and is much larger than those reported for other HF systems.[1]

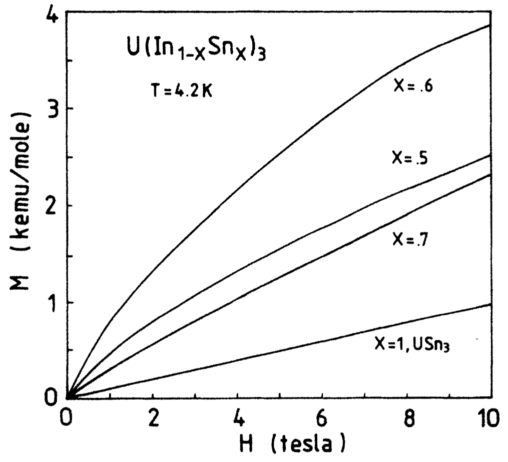

FIG. 4. Magnetization at $T = 4.2$ K vs applied magnetic field for several $\mathrm{U(In_{1-x}Sn_x)_3}$ samples in the HF regime and for $\mathrm{USn_3}$.

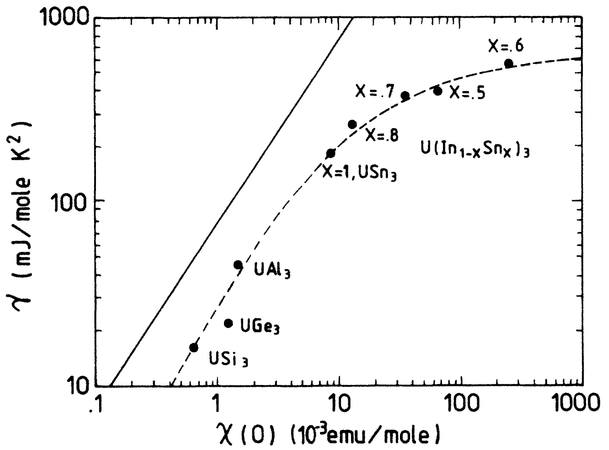

FIG. 5. Electronic specific-heat coefficient γ vs low-temperature limit of the magnetic susceptibility, χ, for four $\mathrm{U}X_3$ compounds. The solid line is the free electron γ vs χ. For $\mathrm{U(In_{1-x}Sn_x)_3}$ samples in the HF regime, γ and χ are taken at $T = 1.35$ K.

In summary, we have reported for the first time a systematic measurement of several thermodynamic properties of a system which evolves from the enhanced paramagnetic regime, through a strongly enhanced HF regime, and finally to an antiferromagnetically ordered regime. Qualitatively, the T and x dependences of the specific heat and magnetic susceptibility, and the H and x dependences of the high-field magnetization, reflect the behavior expected for a narrow f band near the Fermi energy. An interesting feature presented elsewhere[3] is that the resistivity does not show any anomalous enhancement in the HF regime. Such behavior seems to be consistent with the predictions based on many-body phenomenological theories which predict enhancements in χ and γ but no enhancement for the transport properties.[2] Work is continuing to more clearly establish the features associated with HF behavior in this anomalous U-based pseudobinary system and to compare these features to predictions of emerging theories. A search is in progress for other U-based pseudobinary and pseudoternary systems which show HF behavior near magnetic instabilities.

The research at Temple University and at Tufts University was supported by the National Science Foundation through Grants No. DMR 82-19782 and No. DMR 82-03690, respectively. The Francis Bitter National Magnetic Laboratory, Massachusetts Institute of Technology, Cambridge, Massachusetts, is supported by the National Science Foundation.

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