Magnetic-to-nonmagnetic transition in the pseudobinary system $U(Ga_{1-x}Sn_x)_3$

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The temperature dependences of both the electrical resistivity and magnetic susceptibility were measured for the pseudobinary alloy $U(Ga_{1-x}Sn_x)_3$. Moreover, the temperature variation of the specific heat was determined for UGa_3. The results show a clear evolution in magnetic behavior of the system studied from a weakly temperature-dependent paramagnetism with a long-range antiferromagnetic ordering at 67 K in UGa_3 to a strongly temperature-dependent paramagnetism, but without any magnetic order, in USn_3. The properties of UGa_3 are discussed in terms of itinerant $5f$-electron magnetism. In contrast, some arguments are given for local $5f$-electrons in USn_3, which behaves as a nonmagnetic "Kondo-lattice" system with $T_K \cong 60$ K.

INTRODUCTION

The uranium intermetallic phases UX_3, where X is a group-IIIB or IVB element of the periodic table, form a very interesting family of compounds spanning the whole range of magnetic behavior from a temperature-independent, strongly enhanced, Pauli-like paramagnetism (USi_3, UGe_3) to a local-moment ordering (UPb_3). All these phases crystallize in the cubic $AuCu_3$-type structure with the U-U distances being much larger than the Hill limit for uranium compounds. Therefore, a great variety of magnetic properties observed in the UX_3 series is commonly attributed to a hybridization effect between the $5f$-electron states of uranium and the s-, p-, d-electronic states of neighboring atoms.[1] Depending on the strength of these interactions a nonmagnetic, strongly exchange-enhanced paramagnetic to a local-moment magnetic behavior may be realized.

The two terminal compounds in the present study, namely UGa_3 and USn_3, seem to possess a special position among the UX_3 phases. The gallide, placed between the weakly temperature-dependent paramagnet UAl_3 and the antiferromagnetically ordered UIn_3, exhibits an unusual magnetic behavior. Although the magnetic susceptibility of UGa_3 was found previously[2,3] to be weakly temperature dependent with a minor deflection between 60 and 70 K, and the electrical resistivity measured on polycrystalline samples[3] did not show any anomaly on its temperature dependence, an antiferromagnetic ordering of the uranium moments at 70 K has been established by neutron-diffraction investigations.[4,5] More recently, the antiferromagnetic transition in UGa_3 has also been evidenced by electrical-resistivity studies on a single crystal.[6] As found in alloying experiments on the pseudobinary $U(Ga_{1-x}Ge_x)_3$ system,[7] the antiferromagnetic state in this compound is very sensitive to any disorder in the metalloid sublattice. Upon substitution of Ge for Ga, a rapid decrease in the Néel temperature was observed, and for $x \geq 0.18$ the magnetic order was found to disappear. Similar properties were also found for another isomorphous alloy system, $U(Ga_{1-x}Al_x)_3$, where antiferromagnetic order disappears at $x \cong 0.2$.[8] From these results a relatively strong interaction of the uranium $5f$-electron states with a broad conduction band resulting in a considerable delocalization of the $5f$ states has been anticipated for UGa_3.[7]

USn_3, the other terminal compound in the present investigation, occupying in the UX_3 series the position between the weakly temperature-dependent paramagnet UGe_3 and the well moment-localized antiferromagnetic UPb_3, appears to be a strongly temperature-dependent paramagnetic system, close to a magnetic instability. USn_3 exhibits a fairly high value of the electronic specific-heat coefficient, $\gamma(0) = 169$ mJ/mole K^2,[9] and the magnetic susceptibility χ for $T \to 0$ saturates and reaches a value as large as 9500×10^{-6} emu/mole.[2] Further-

more, a T^2-type variation of the electrical resistivity at low temperatures and an upturn of the C/T vs T^2 curve observed below 3.5 K in the specific heat of this stannide have been commonly interpreted as being characteristic of a spin-fluctuation behavior.[3,9–11] As the alloying experiments in the pseudobinary system $U(Sn_{1-x}Pb_x)_3$ (Refs. 12 and 13) have shown, a small substitution of Pb for Sn, resulting in a small expansion of the lattice, is sufficient to drive the alloy into a long-range magnetically ordered state. A similar transition, but with an almost negligible change in the lattice volume, has also been reported for the $U(Sn_{1-x}In_x)_3$ system.[14,15]

The above-mentioned results on transitions from the strongly exchange-enhanced paramagnet USn_3 to both the *well moment-localized* antiferromagnet UPb_3 and the *less moment-localized* antiferromagnet UIn_3 motivated us to investigate a transition from USn_3 to the *itinerant-electron* antiferromagnetic UGa_3. In this paper we address the magnetic and electrical-resistivity behavior of the highly hybridized pseudobinary system $U(Ga_{1-x}Sn_x)_3$. Moreover, further evidence of an itinerant-electron nature of magnetic ordering in UGa_3 is given from our heat-capacity studies.

EXPERIMENT

Polycrystalline samples of $U(Ga_{1-x}Sn_x)_3$ were prepared by arc-melting the constituent elements in an argon atmosphere and subsequent annealing in vacuum at 600 °C for one week. Some small weight losses due to evaporation of tin and gallium were carefully compensated by correcting the starting compositions in accord with the relative vapor pressures of these elements. The sample purity was checked by x-ray-powder diffractometry. All buttons were found to be single phase, with the expected cubic $AuCu_3$-type crystal structure. The corresponding lattice parameters are shown in Fig. 1 as a function of the Sn concentration x. As is apparent from this figure, the terminal compounds UGa_3 and USn_3 exhibit mutual solubility and the lattice parameter a follows Vegard's law in the whole concentration range.

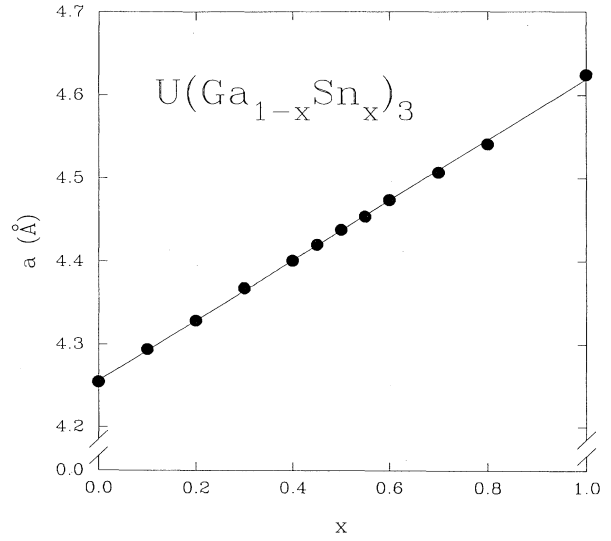

FIG. 1. Lattice parameter a vs Sn concentration x for the $U(Ga_{1-x}Sn_x)_3$ system. The solid line corresponds to Vegard's law.

Magnetic-susceptibility measurements were performed in the temperature range 4.2–300 K using a R. H. Cahn electrobalance. The electrical resistivity was measured over the range 4.2–300 K, using a conventional four-point dc technique. Specific-heat studies on UGa_3 were carried out within the temperature interval 4.2–300 K, using a quasiadiabatic method described elsewhere.[16]

RESULTS

The temperature dependence of the magnetic susceptibility of the samples studied is displayed in Figs. 2 and 3. The $\chi(T)$ variation for UGa_3 agrees well with those reported previously,[2,3,6,7] but our absolute values of χ are higher than the literature data. As seen in Fig. 2, the susceptibility is only weakly temperature dependent, and in such a situation the Néel temperature of 67 K manifests itself on an inflection point in the $\chi(T)$ curve [a maximum in $d\chi(T)/dT$]. As found previously,[2,3,6,7] we observe an upturn in the susceptibility at low temperatures. At present we cannot distinguish if this feature is an intrinsic effect or due to a small amount of paramagnetic impurities present in our UGa_3 sample. However, it is certainly remarkable that a nearly universal $\chi(T)$ of UGa_3 at low temperatures is observed in all these investi-

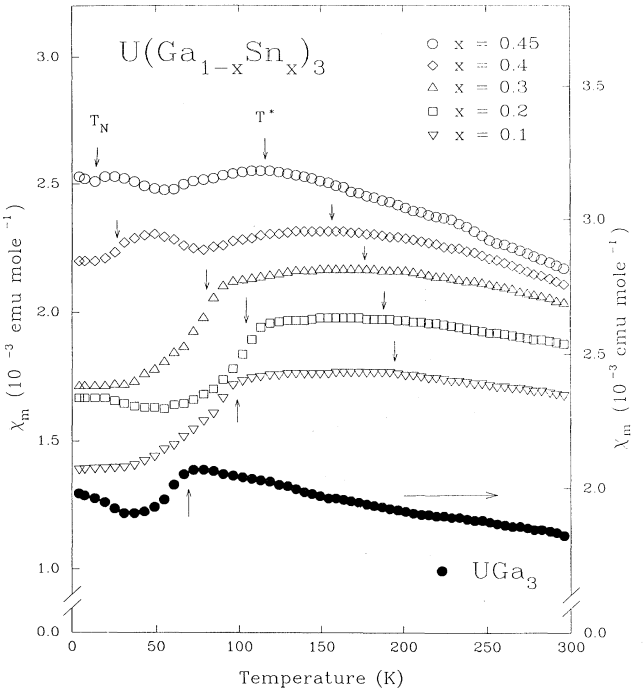

FIG. 2. Magnetic susceptibility vs temperature for UGa_3 (right-hand scale) and $U(Ga_{1-x}Sn_x)_3$, where $0<x<0.5$ (left-hand scale). For the sake of clarity the $\chi(T)$ curves are shifted by 0.1×10^{-3} emu/mole downwards and upwards for $x=0.1$ and $x=0.3$, respectively. The arrows indicate the characteristic temperatures T_N's and T^*'s (see the text).

gations. Upon substitution of Sn for Ga the characteristic deflection in $\chi(T)$, corresponding to the magnetic phase transition, initially shifts to a higher temperature, reaching a maximum value of 102 K at $x=0.2$, but then falls to lower temperatures and finally disappears for the $x=0.5$ sample. Simultaneously, the low-temperature upturn of χ disappears as well. Interestingly, all alloys with $x<0.5$ show a non-Curie-Weiss behavior of the susceptibility above T_N. Instead, a broad maximum in their $\chi(T)$ dependence gradually develops, starting from $x=0.1$. This maximum is most strongly pronounced for the $x=0.4$ and 0.45 compositions (see Fig. 2). It should be noted that a similar temperature variation of the susceptibility was found for a new heavy-fermion superconductor UNi_2Al_3.[17] For this compound, a broad maximum in $\chi(T)$, centered at $T^*=100$ K, has been interpreted, within the Kondo-lattice concept, as a continuous transition from the localized to the delocalized $5f$-electron behavior on going from higher to lower temperatures. The same situation has recently been considered for UAl_2 on the basis of ^{27}Al nuclear magnetic resonance NMR results[18] (see also Ref. 19). It seems likely that a similar interpretation may also be applied to the $U(Ga_{1-x}Sn_x)_3$ system associating the characteristic temperature T^* with the spin-fluctuation temperature T_{sf} (Ref. 19). As is clear from Fig. 2, this temperature decreases from about 200 K at $x=0.1$ to about 110 K at $x=0.45$. For the $x=0.5$ sample, the maximum in $\chi(T)$ transforms to a distinct plateau around 50 K [see Fig. 3(a)] and no indication of magnetic ordering at low temperatures is observed. Instead, the latter $\chi(T)$ behavior well resembles the one characteristic of the spin-fluctuation system UAl_2, showing a T^2 dependence of the magnetic susceptibility at low temperatures.[20] For the samples with higher Sn concentration, the susceptibility follows a Curie-Weiss law, and the temperature range in which this behavior is observed extends substantially to lower temperatures with increasing x [see Fig. 3(b)]. The effective magnetic moment decreases from $\mu_{eff}=3.68\ \mu_B$/U atom at $x=0.5$ to 3.12 μ_B/U atom for the $x=0.8$ sample, with a simultaneous drop in the negative paramagnetic Curie temperature from $\theta_p=-443$ to -159 K, respectively. Interestingly, our μ_{eff} values are much larger than those reported previously for USn_3 [$\mu_{eff}=2.5\ \mu_B$/U atom (Ref. 12)]. The large decrease in the negative values of θ_p indicates, presumably, a rapid development of the local-moment character of the $5f$ electrons on going from $U(Ga_{0.5}Sn_{0.5})_3$ to pure USn_3. Furthermore, the value of χ at $T=4.2$ K continuously increases with increasing x, also reflecting an increasing local-moment behavior in these alloys. Whereas, the χ(4.2 K) values for the $x=0.5$ and 0.6 samples can be compared with χ(4.2 K)$=3.8\times10^{-3}$ emu/mole found for UAl_2 (Ref. 20), this value for $U(Ga_{0.2}Sn_{0.8})_3$ becomes as large as about 6×10^{-3} emu/mole, thus approaching that reported for pure USn_3 [i.e., χ(4.2 K)$=9.5\times10^{-3}$ emu/mole (Ref. 2)].

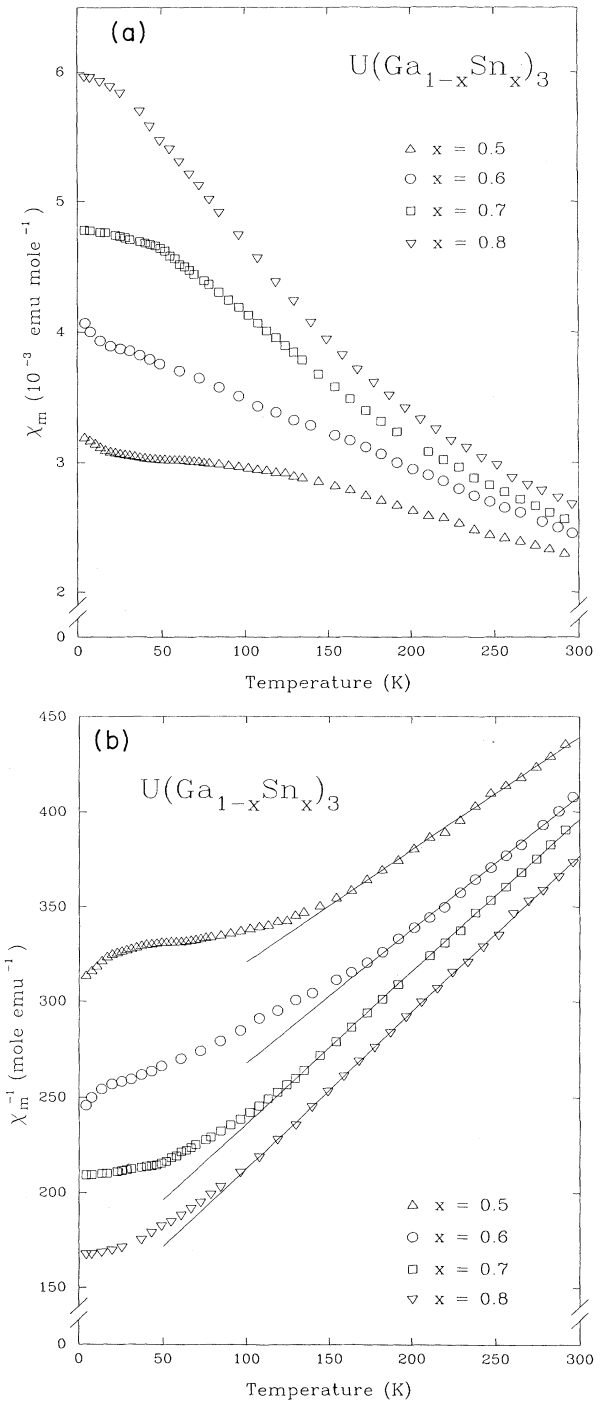

FIG. 3. (a) Magnetic susceptibility vs temperature for $U(Ga_{1-x}Sn_x)_3$ where $x\geq0.5$. (b) Reciprocal magnetic susceptibility vs temperature for $U(Ga_{1-x}Sn_x)_3$ where $x\geq0.5$. Solid lines are fits of $\chi^{-1}(T)$ to the Curie-Weiss law.

The results of our electrical-resistivity studies on the $U(Ga_{1-x}Sn_x)_3$ system are presented in Figs. 4–6. Shown in Fig. 4 is $\rho(T)$ for UGa_3. It is worthwhile noting that the resistivity value at room temperature of about 100 $\mu\Omega$ cm agrees well with that reported previously for a single-crystalline sample.[6] However, in contrast to the previous data,[6] our residual resistivity ratio (RRR$=\rho_{300\ K}/\rho_{4.2\ K}$) of about 100 is considerably larger than that reported[6] for the single crystal (~1.7). As shown in Fig. 4, a T^2 dependence of the resistivity is observed below 20 K.

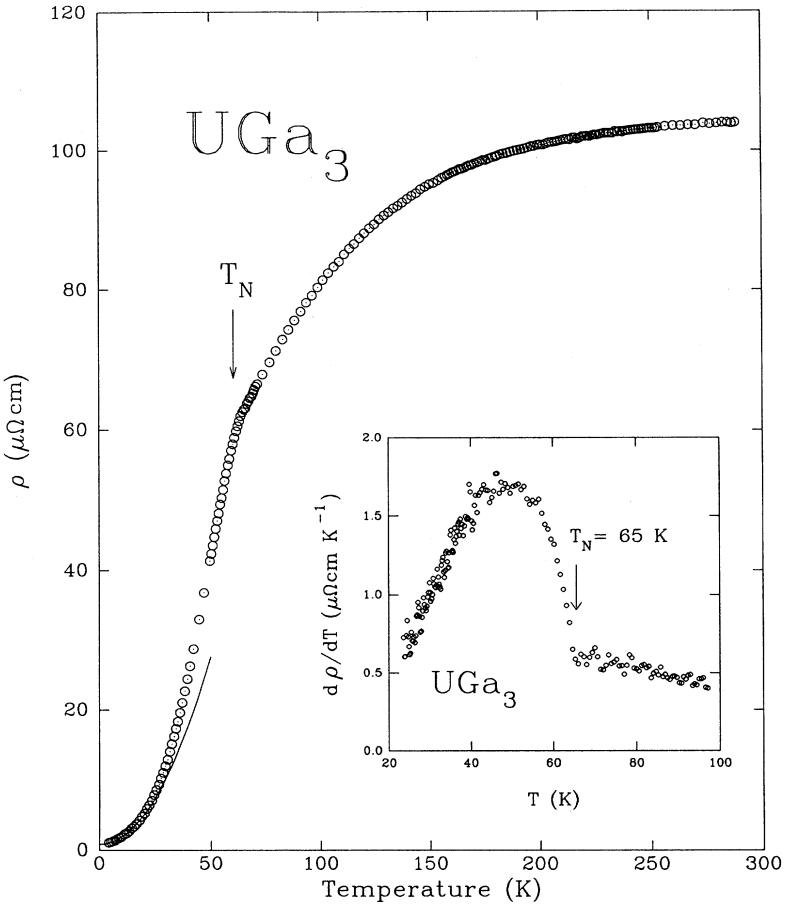

FIG. 4. Temperature dependence of the electrical resistivity of UGa_3. The thin solid line marks a T^2 variation of the resistivity at low temperatures. The inset shows the temperature derivative of the resistivity in the vicinity of T_N.

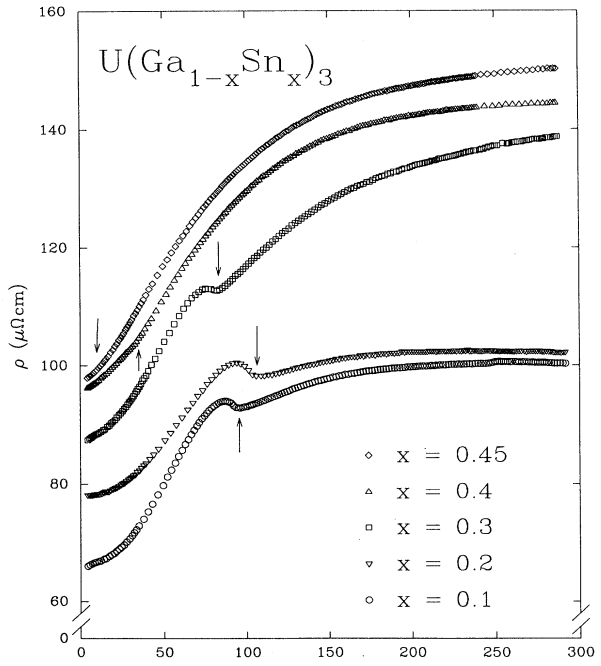

FIG. 5. Electrical resistivity vs temperature for $U(Ga_{1-x}Sn_x)_3$ where $0<x<0.5$. For the sake of clarity, the $\rho(T)$ curves are shifted upwards by 10 $\mu\Omega$ cm for $x=0.2$, 20 $\mu\Omega$ cm for $x=0.3$, 30 $\mu\Omega$ cm for $x=0.4$, and 60 $\mu\Omega$ cm for $x=0.45$. The arrows indicate T_N's.

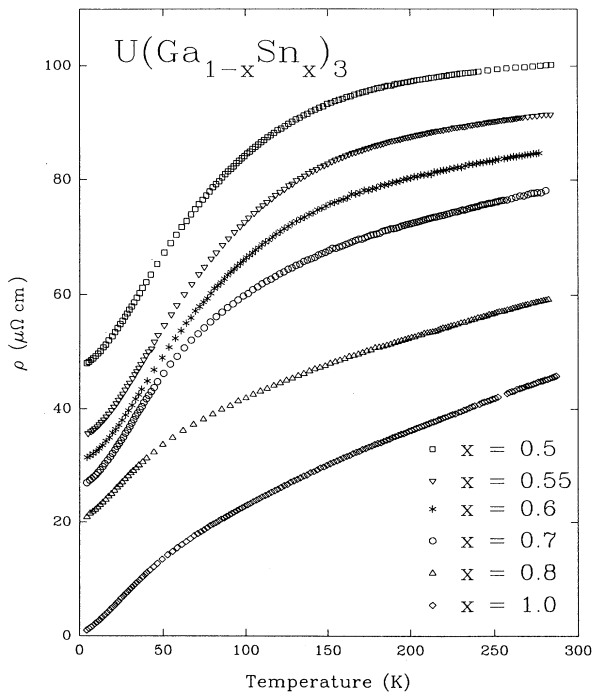

FIG. 6. Electrical resistivity vs temperature for $U(Ga_{1-x}Sn_x)_3$ where $0.5 \le x \le 1$. For the sake of clarity the $\rho(T)$ curve for $x=0.5$ is shifted upwards by 10 $\mu\Omega$ cm.

The Néel temperature found for our sample of UGa_3 manifests itself as a kink in $\rho(T)$ and a distinct anomaly in the $d\rho/dT$ vs T variation (see Fig. 4). This is at variance from the single-crystal results which showed a pronounced maximum in $\rho(T)$ just below T_N.[6] As seen in Fig. 5, such pronounced maxima in $\rho(T)$ were found, however, for the $U(Ga_{1-x}Sn_x)_3$ samples with $x=0.1$, 0.2, and 0.3. These anomalies correspond unambiguously to antiferromagnetic phase transitions, and the ordering temperatures derived from the $d\rho/dT$ vs T curves (not shown) agree well with those determined from the susceptibility data. For the $x=0.40$ and 0.45 compositions, the corresponding singularities in $\rho(T)$ at T_N are hardly visible. However, in this case the values of the Néel temperature taken from the temperature derivative of the resistivity for these samples agree well with the magnetic-susceptibility results. Above $x=0.5$, no indication of magnetic ordering is observed in both the $\rho(T)$ and $d\rho/dT$ vs T dependences. The residual resistivity ρ_0 strongly rises upon substitution of Sn for Ga in $U(Ga_{1-x}Sn_x)_3$ from $\rho_0=1$ $\mu\Omega$ cm for $x=0$ to about $\rho_0=70$ $\mu\Omega$ cm for $x=0.1$ and remains almost constant for the composition range $0.1\leq x\leq 0.4$ [note the shifts upward of $\rho(T)$ in Fig. 5]. Then, ρ_0 starts to fall down with a further increase in the Sn content and finally drops again to as small a value as $\rho_0=1$ $\mu\Omega$ cm for the fully crystallographically ordered compound USn_3 (RRR=46).

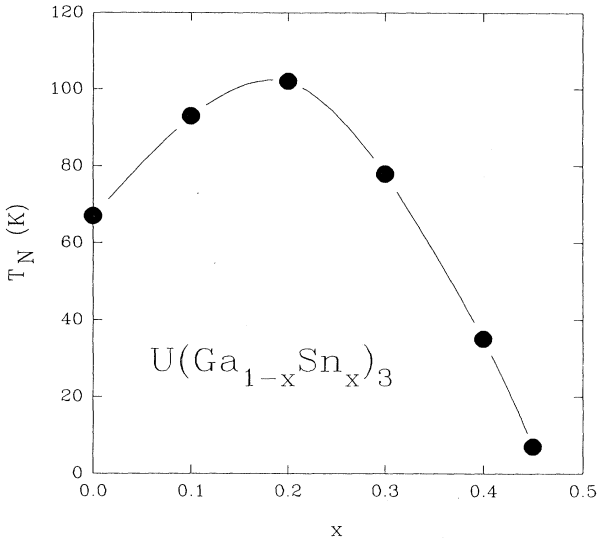

FIG. 7. Néel temperature T_N vs Sn concentration x for $U(Ga_{1-x}Sr_x)_3$. The solid curve is a guide for the eye.

DISCUSSION

Summarizing the behavior of the $U(Ga_{1-x}Sn_x)_3$ system, one sees that the increase of the Sn concentration in the samples causes a gradual transformation of the magnetic susceptibility from the form characteristic of an itinerant-electron antiferromagnet to that typical for a spin-fluctuation system and finally to a more local-moment behavior for $x\cong 1$ (see discussion below). A similar evolution is also inferred from the electrical-resistivity results. Furthermore, the Néel temperature initially strongly rises with increasing x, reaches a maximum value of 102 K at $x=0.2$, and finally decreases to zero at $x\cong 0.5$ (see Fig. 7). This behavior resembles very much the one observed previously for the $U(Pb_{1-x}Sn_x)_3$ system,[13] but with some quantitative differences. Whereas, for the latter system, T_N increases only by $\sim 23\%$ upon substitution of Sn for Pb and the magnetic order extends up to about 93% exchange of Pb by Sn, for the $U(Ga_{1-x}Sn_x)_3$ system, the corresponding numbers are 52 and 50 %, respectively.

An itinerant-electron nature of antiferromagnetic order in UGa_3 has already been suggested by several authors,[1,3,9] mainly on the basis of the peculiar temperature variation of its magnetic susceptibility (see Fig. 2). Indeed, a rise in the χ value at the Néel temperature and the lack of a Curie-Weiss behavior in the temperature range up to 1000 K (Ref. 2), where the susceptibility is dominated by the Pauli-like contribution, both these features are in keeping with the mean-field theory of itinerant antiferromagnetism, evaluated by Fedders and Martin.[21] Moreover, if one also considers the reduced value of the ordered magnetic moment of uranium [$0.82(6)\mu_B$ (Ref. 4) or $0.95(7)\mu_B$ (Ref. 5)] and the enhanced electronic-specific-heat coefficient $\gamma(0\text{ K})=52$ mJ/mole K^2,[9] it seems that these criteria for the itinerant-electron magnetism of UGa_3 are similar to those given previously for UN.[22,23]

Another criterium concerns the reduced magnetic entropy which, for an itinerant-electron system, should be much lower than R ln2. In order to test this latter prediction for UGa_3, we have measured the temperature dependence of its specific heat. The results are shown in Fig. 8. As is apparent from this figure, $C_p(T)$ is dominated by the phonon contribution, superimposed by a mean-field-type anomaly at $T_N=67$ K. The $\gamma(0)$ value determined from the $C_p(T)$ data below 10 K agrees well with that reported previously,[9] being of about 50 mJ/mole K^2. To provide an estimate of the magnetic entropy, the phonon part was fit to the Debye model. We approximated the lattice specific heat by the Debye function with a temperature-dependent Debye temperature $\theta(T)$, assuming that the magnetic contribution to the measured specific heat is negligible except for the temperature range in the vicinity of the phase transition. This way, a magnetic entropy ΔS_m of about 0.8 J/mole K, i.e., $0.14R$ ln2, was estimated.

Moreover, in the inset to Fig. 8 we present the electronic specific heat C_{el} of UGa_3 plotted as C_{el}/T vs T. According to the theoretical predictions of Fedders and Martin,[21] $C_{el}(T)$ of an itinerant-electron antiferromagnet in the vicinity of the magnetic phase transition can be described by a gap equation very similar to that for a BCS superconductor. In particular, the jump in the C_{el} value at the Néel temperature, expressed as the ratio $\Delta C_{el}/\gamma T_N$, should be close to 1.43. Indeed, for UGa_3 the above ratio is found to be 1.3, in satisfactory agreement with that obtained within mean-field theory (BCS).

Hence, the basic characteristics of itinerant-electron

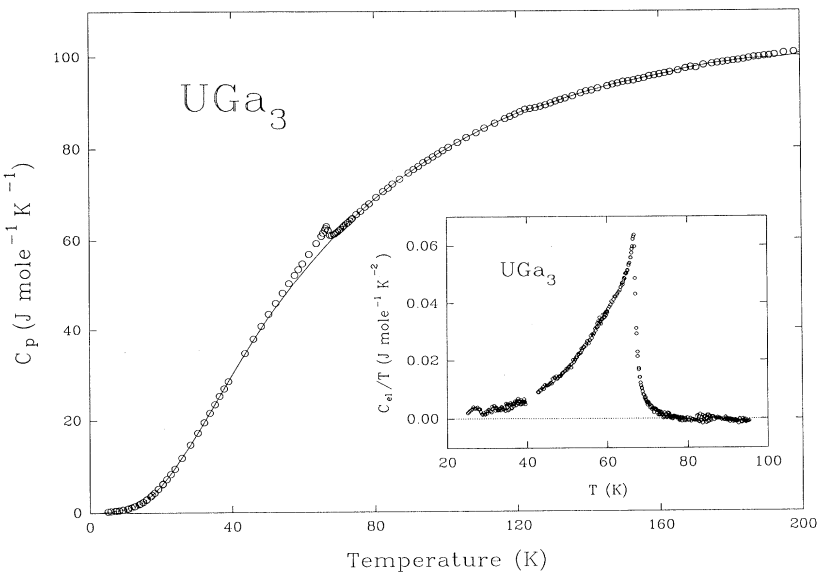

FIG. 8. Specific heat vs temperature for UGa_3. The solid curve is a Debye approximation of the phonon contribution to the specific heat. The inset shows the magnetic contribution to the specific heat in the vicinity of the magnetic phase transition.

antiferromagntism appear to be met in UGa_3. Further support for this interpretation can be expected from future high-pressure investigations.

As to USn_3, a larger distance between the uranium and ligand atoms ($d_{U\text{-}Sn} \cong 3.3$ Å) prevents the strong hybridization effects seen in UGa_3 ($d_{U\text{-}Ga} \cong 3.0$ Å). Despite the arguments provided previously for the spin-fluctuation behavior in USn_3,[3,9–11] it seems that the strongly temperature-dependent susceptibility of the Sn-rich alloys ($x \geq 0.7$) and the tendency to the saturation of their low-temperature susceptibility into large values when $T \rightarrow 0$, all these findings indicate rather a local-moment behavior of the uranium ions in these alloys. In what follows, we will try to give arguments on this point of view.

A detailed analysis of heat-capacity data for USn_3 by taking into account a realistic phonon-heat-capacity contribution, the authors of Ref. 11 were able to remove a paramagnon-derived $T^3 \ln T$ term and to conclude that the spin fluctuations in this compound are local in nature. In this interpretation, the enhancement of the electronic-specific-heat coefficient [$\gamma(0) = 169$ mJ/mole K^2 (Ref. 9)] is not directly connected to the local one-particle conduction-band density of states at the Fermi level; it rather reflects the strong f-electron correlations of a Kondo-lattice system being very close to the onset of the magnetic order.[24,25] The same dramatic enhancement in $\gamma(0)$ is also found in UPb_3 [$\gamma(0) = 173$ mJ/mole K^2 (Ref. 13)], considered up to now as a highly moment-localized system. This latter idea is supported by the positive pressure dependence of T_N in UPb_3.[12] In addition, the authors of Ref. 12 also stated that no dramatic change in the degree of localization of the $5f$ electrons is apparent in the $U(Pb_{1-x}Sn_x)_3$ system.

Furthermore, the temperature dependence of the U-derived electrical resistivity $\Delta\rho(T)$, for $La_{1-x}U_xSn_3$ (Ref. 25) shows a slope change at temperatures $T > 50$ K from a slightly negative ($x < 0.8$) to a positive value ($x \rightarrow 1$). For very small U concentrations ($x \leq 0.05$), the variation $\Delta\rho(T)/x$ vs T strongly resembles that expected for a dilute Kondo alloy with a large Kondo temperature ($T_K \cong 310$ K). Neither the $\rho(T)$ curves nor the lattice parameters indicate any significant change in the magnetic character of the $5f$ electrons upon dilution of the uranium sublattice.[26]

However, there is no clear evidence of well-defined inelastic peaks due to crystal-field (CF) excitations either in USn_3 (Ref. 27) or UPb_3 (Ref. 28). The lack of CF excitations in these, as in many other U-based compounds, probably arises from the presence of strong $5f$-s,p exchange interactions which tend to damp out the transitions within the crystal-field levels. Also, the observed large residual quasielastic linewidth, half width at half maximum (HWHM), $\Gamma(0) \cong 60$ K, in USn_3 (Ref. 27) suggests that the energy of the many-body f-s,p interactions is significantly larger than the thermal energy k_BT already at moderate temperatures. Note that $\Gamma(0)$ is a very good empirical measure for the Kondo temperature.[25]

In our opinion, the magnetism in USn_3 should, from the onset, be discussed in terms of a combined action of CF and Kondo-type effects. The picture of a conventional CF effect in USn_3 was postulated many years ago by Mulak and Misiuk.[29] The CF-level schemes of the split multiplet $^3H_4$ ($5f^2$ configuration) for both negative and positive ligand charges in the UX_3 compounds were given. Assuming negative charges, either a nonmagnetic doublet (Γ_3) or a magnetic triplet (Γ_5) can be the ground state for the U^{4+} ion, depending on the ratio of the fourth- and sixth-order potential terms. The $T = 0$ K value of the magnetic (van Vleck-type) susceptibility for USn_3, $\chi(0) = 9.5 \times 10^{-3}$ emu/mole, corresponds to a Γ_3-Γ_5 splitting $E_{\Gamma_3,\Gamma_5} \cong 100$ K, and the interval in which the susceptibility is temperature independent amounts to $\frac{1}{3}E_{\Gamma_3,\Gamma_5} \cong 35$ K (Ref. 2), in rough agreement with the experiment.[2] For positive ligand charges, the ground term is always the singlet Γ_1. However, this solution yields

considerably lower values of $\chi(0)$.[2] Within the U^{3+} ionic configuration one always gets a magnetic CF ground state.

In the case of Kondo systems (strong f-s,p exchange interactions) a new approach has recently been proposed,[30] based upon the Anderson model for local moments in metals. The CF splitting here is dominated by the anisotropy of the f-s,p mixing interactions, where the mixing matrix elements $V_{k\sigma,m}$ depend on the orbital state m of the f electron. Taking into account this orbitally driven anisotropic hybridization, Kalvius *et al.*[31] have recently shown that the $5f$ electrons in $NpSn_3$ [$\gamma(0)=88$ mJ/mole K^2 (Ref. 32)] appear to be localized. One should remember that until recently this compound has been regarded as the prototypical itinerant-electron antiferromagnet. By comparison, we consider USn_3 as containing localized $5f$ electrons, too. In contrast to $NpSn_3$, however, it assumes a nonmagnetic (moderately heavy Fermi liquid) ground state.

Finally, we wish to state that the change in the strength of the correlation effects caused by the aforementioned anisotropic f-s,p hybridization does not affect the lattice parameters of the $U(Ga_{1-x}Sn_x)_3$ system on going from UGa_3 to USn_3 because Vegard's law is always very well followed. This has also been observed for related systems[7,8,12–15,26] and rules out interconfigurational (valence) fluctuations as a potential source for the dramatic changes in magnetic behavior when the composition is varied in these systems.

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