MAGNETIC AND TRANSPORT PROPERTIES OF THE PSEUDOBINARY SYSTEM $UGa_{3-x}Ge_x$*

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Magnetic, electrical and thermodynamic properties of the pseudobinary alloy system $UGa_{3-x}Ge_x$, $0 \leq x \leq 3$, have been studied by means of magnetic susceptibility, electrical resistivity, thermoelectric power and specific heat measurements. The results indicate a gradual transformation from itinerant antiferromagnetism in Ga-rich alloys to Pauli paramagnetism in Ge-rich materials. Total suppression of the magnetic order occurs near the composition $x \approx 0.6$. For $UGa_{2.4}Ge_{0.6}$ curious behavior of the low-temperature bulk properties has been found that develops presumably because of the closeness of this alloy to a magnetic quantum critical point.

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The uranium compound UGa_3 is an archetypal itinerant antiferromagnet with large orbital moments [1,2]. Upon applying hydrostatic pressure its Néel temperature $T_N = 67$ K sharply decreases and becomes zero at about 2.6 GPa [3]. Around the quantum critical point the electrical resistivity shows a non-Fermi liquid behavior with some hints of a transition to superconducting state [3]. This recent finding motivated us to undertake a re-investigation of the pseudobinary alloy system $UGa_{3-x}Ge_x$, $0 \leq x \leq 3$, in which the substitution for Ga of a smaller atom Ge is believed to simulate a positive chemical pressure [4,5]

Polycrystalline samples of $UGa_{3-x}Ge_x$ were synthesized by arc-melting the constituents in a titanium-gettered argon atmosphere and subsequent annealing in vacuum at 850° C for a week. All the alloys were proved by X-ray powder diffraction to be single phases with the expected $AuCu_3$-type crystal structure. With increasing Ge-content the cubic lattice parameter gradually decreases, approximately in a Vegard's manner.

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The physical properties of $UGa_{3-x}Ge_x$ were studied by means of magnetic susceptibility, electrical resistivity, thermoelectric power and specific heat measurements, performed in the temperature range 1.7–300 K. Upon the substitution of Ge for Ga the magnetic susceptibility, which is almost temperature independent for all the compositions [4], systematically decreases. Simultaneously, the magnetic ordering, manifested by a small maximum in $\chi(T)$, gradually shifts to lower temperatures and vanishes at $x \approx 0.6$. In general, the magnetic properties of Ga-rich alloys are reminiscent of those of UGa_3 [6] (in Fig. 1(a) there is shown as an example the $\chi(T)$ curve for $UGa_{2.9}Ge_{0.1}$). Most surprisingly, the susceptibility of $UGa_{2.4}$ $Ge_{0.6}$, measured in weak magnetic fields, drops abruptly at $T_c = 6.4$ K and at lower temperatures it becomes diamagnetic (see Fig. 1(b)). Moreover, the field dependence of the magnetization taken at $T = 1.7$ K is characteristic of type-II superconductors.

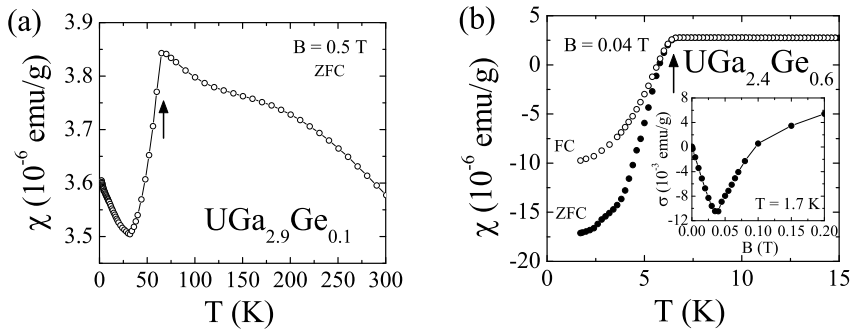

Fig. 1. Temperature variation of the magnetic susceptibility of $UGa_{2.9}$ $Ge_{0.1}$ (a) and $UGa_{2.4}Ge_{0.6}$ (b). ZFC-zero field cooled; FC-field cooled. The arrow in panel (a) indicates the Néel point $T_N = 64$ K. The arrow in panel (b) marks the onset of diamagnetism at $T_c = 6.4$ K. Inset: field dependence of the low-temperature magnetization taken with increasing magnetic field.

Nevertheless, any supposition that the sample becomes superconducting should be rather excluded because its low-temperature resistivity remains final. In fact, as displayed in Fig. 2(a), the $\rho(T)$ curve of $UGa_{2.4}Ge_{0.6}$ is featureless in the whole temperature range studied. Below about 70 K it is a linear function of temperature, being in a striking contrast to the Fermi-liquid-like behavior of the resistivity for all the alloys with $x < 0.6$ (see as an example in Fig. 2(a) $\rho(T)$ for $UGa_{2.9}Ge_{0.1}$). Actually, fitting the resistivity data of $UGa_{2.9}Ge_{0.1}$ to the formula $\rho(T) = \rho_0 + A\left(\frac{T}{T_0}\right)^n$ [7] gives for $T < 50$ K the parameters: $\rho_0 = 44$ $\mu\Omega$cm, $A = 0.09$ $\mu\Omega$cm, $T_0 = 3.5$ K and

$n = 2.04$, whereas the same procedure performed for $UGa_{2.4}Ge_{0.6}$ yields: $\rho_0 = 42$ $\mu\Omega$cm, $A = 0.45$ $\mu\Omega$cm, $T_0 = 1.9$ K and $n = 1.05$. Interestingly, a similar change in $\rho(T)$ was recently reported for a UGa_3 single crystal subjected to hydrostatic pressure [3]: whereas below 2 GPa a $\rho \sim T^2$ dependence was observed, a non-Fermi liquid behavior was found in the vicinity of the magnetic-nonmagnetic instability occurring at $p_c = 2.6$ GPa. However, unlike the $UGa_{2.9}Ge_{0.1}$ case ($T < 70$ K), the NFL-like features in UGa_3 were limited to temperatures below 2 K only.

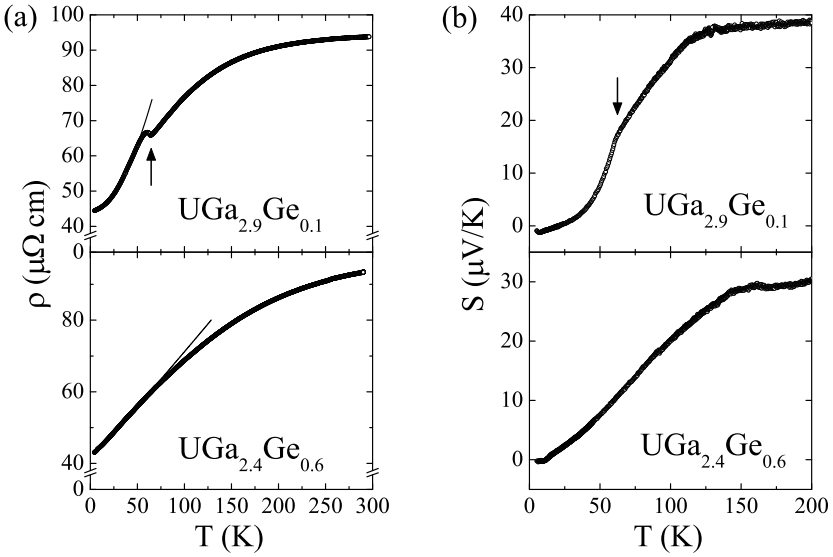

Fig. 2. Temperature dependence of the electrical resistivity (a) and the thermoelectric power (b) of $UGa_{2.9}Ge_{0.1}$ (upper panels) and $UGa_{2.4}Ge_{0.6}$ (lower panels). The arrows mark the antiferromagnetic phase transition. The solid lines are least-squares fits of the experimental data to the function $\rho(T) = \rho_0 + A\left(\frac{T}{T_0}\right)^n$, which yield $n = 2.04$ and 1.05 for $UGa_{2.9}Ge_{0.1}$ and $UGa_{2.4}Ge_{0.6}$, respectively.

In Fig. 2(b) there are shown the temperature variations of the thermoelectric power of the two alloys discussed. These $S(T)$ results are consistent with the data reported for UGa_3 [8] in respect of both positive sign and overall shape. Large absolute values of the thermopower at 200 K are of the order of those characteristic of heavy fermion compounds.

The low-temperature specific heat of UGa_3 exhibits a regular dependence $C_p = \gamma T + \beta T^3$ with moderately enhanced γ value of about 50 mJ/K^{2}mole [1, 8]. For $UGa_{2.4}Ge_{0.6}$ the Sommerfeld coefficient extrapolated from the region 10–20 K amounts to 96 mJ/K^{2}mole, and $C_p(T)$ shows an anomaly

near $T_c = 6.4$ K that results in a steep upturn in C_p/T when $T \to 0$. The details on this finding will be given elsewhere, together with the specific heat data for other $UGa_{3-x}Ge_x$ alloys.

In summary, the substitution in $UGa_{3-x}Ge_x$ of Ge atoms for Ga atoms brings about a gradual suppression of the long range magnetic order. Such a behavior is consistent with an itinerant-electron nature of the magnetism in this system that is mainly driven by the hybridization of uranium $5f$ electronic states with p-orbitals of gallium and germanium. For the critical composition $x \approx 0.6$, at which T_N goes to zero, some unusual features have been found, like a linear T-dependence of the resistivity, a strongly diamagnetic susceptibility and the formation of a low-temperature tail in the C_p/T ratio. These findings can be compared with the non-Fermi liquid behavior of the electrical resistivity of single-crystalline UGa_3, driven into the magnetic quantum critical point by applying hydrostatic pressure [3]. Furthermore, the diamagnetism in $UGa_{2.4}Ge_{0.6}$ strongly recalls the superconductor-like AC susceptibility response of UGa_3 single crystals [6]. These and other extraordinary low-temperature properties of the alloys $UGa_{3-x}Ge_x$ as well as of the terminal compound UGa_3 will be addressed in our forthcoming paper.

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