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Magnetic and electrical properties of the $UCu_{4-x}Mn_xAl_8$ alloys ☆

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Abstract

The $UCu_{4-x}Mn_xAl_8$ system exists in a single-phase form for the whole concentration range. The magnetic and electrical properties have been examined in the temperature ranges 1.7–300 K and 4.2–300 K, respectively. The lattice parameters increase with increasing x, however with $da/dx > dc/dx$. The magnetic susceptibility follows the modified Curie–Weiss law at $T > 20$ K. The increase of the Mn concentration, x, is reflected in a higher value of the temperature independent part of the susceptibility which can be related to the increase of the Pauli-like susceptibility of the band electrons. The electrical resistivity is typical for metallic compounds but for lower x there are anomalies in $\rho(T)$ plots corresponding roughly to the Néel point of UCu_4Al_8

Keywords: $UCu_{4-x}Mn_xAl_8$; Magnetic properties; Electrical properties

1. Introduction

The discovery of heavy fermion-like behavior in the UCu_xAl_{12-x} system [1,2] has aroused the interest in the properties of its derivatives. These alloys crystallize in the $ThMn_{12}$-type of structure (*I4/mmm*) in which uranium atoms occupy *2(a)* positions whereas Cu and Al atoms are located in the remaining *8(f)*, *8(i)* and *8(j)* positions. The examination of UCu_xAl_{12-x} compounds has shown an alloying induced transition from an antiferromagnetic ground state for low Cu concentration ($x \leqslant 5.25$) to pure heavy-fermion behavior for $x \geqslant 5.5$ [2]. Recently, Nishioka et al. [3] have presented a phase diagram of UCu_xAl_{12-x}, derived from the specific heat and magnetic susceptibility measurements and have established that in this system there can exist three states: antiferromagnetic (AF), heavy fermion (HF) and non-Fermi liquid (nFL) depending on the value of x. Moreover, Krimmel et al. [4] claim that for $x = 4.75$ an indication of spin fluctuations (SF) is observed.

In turn, UMn_xAl_{12-x} alloys exhibit also the $ThMn_{12}$-type structure and exist in a single phase form for $3 \leqslant x \leqslant 7$ [5]. These materials are paramagnetic and their magnetic susceptibility follows a Curie–Weiss law at $T > 100$ K with an effective magnetic moment and the Weiss constant varying linearly with composition. UMn_4Al_8 is paramagnetic down to 1.5 K [6] and its electrical resistivity, ρ, is weakly temperature dependent, however, at $T \approx 200$ K there is a diffuse minimum in a $\rho(T)$ plot [7].

The present research has been undertaken to check whether substitution of Mn for Cu would enhance the magnetic order and induce the nFL state in the $UCu_{4-x}Mn_xAl_8$ system.

2. Experimental

The alloys were prepared by melting the elements in stoichiometric quantities in an arc furnace under a protective argon atmosphere. For measurements the as-cast samples were used. An annealing process resulted in the appearance of parasitic lines in the X-ray pattern. The magnetic measurements were carried out in the temperature range 1.7–300 K and in a magnetic field of 0.5 T using a SQUID magnetometer. The standard equipment in our laboratory was used for electrical investigations at $T = 4.2$–300 K.

3. Results and discussion

The $UCu_{4-x}Mn_xAl_8$ system exists for all concentrations in principle in single phase. Very weak additional lines in the X-ray pattern can be identified as belonging to the $CuAl_2$ phase which is nonmagnetic in the temperature

☆Dedicated to Professor Andrzej Szytuła on the occasion of his 60th birthday.

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Table 1
Lattice parameters *a* and *c*, and magnetic data for the $UCu_{4-x}Mn_xAl$ system

x	Lattice parameters (nm) *a*	*c*	$\chi_0 \cdot 10^{-3}$ (cm^3/mol)	θ (K)	p_{eff} (μ_B)
0.5	0.8744	0.5096	1.15	−19.8	1.29
1.0	0.8799	0.51211	1.41	−44.6	2.61
2.0	0.88122	0.51024	4.18	−40.7	2.86
3.0	0.88229	0.51061	6.55	−84	2.82

range of the present investigation. The alloys exhibit a tetragonal $ThMn_{12}$-type structure (space group *I4/mmm*). In analogy with the neutron diffraction study of the UCu_xAl_{12-x} system [4] one can assume that both Cu and Mn atoms are located in the *8(f)* site. The lattice parameters *a* and *c* are listed in Table 1. Substitution of larger Mn atoms for Cu atoms increases both lattice parameters, however, this increase is more pronounced for the *a* parameter. The reciprocal magnetic susceptibility, χ^{-1}, versus temperature for the $UCu_{4-x}Mn_xAl_8$ system is presented in Fig. 1. The $\chi(T)$ plot follows a modified Curie–Weiss law above 20 K shown by lines in Fig. 1. Magnetic data, χ_0, temperature independent susceptibility, p_{eff}, effective magnetic moment and θ, Weiss constant, are also listed in Table 1. The deviation in $\chi(T)$ at low temperature could be caused by antiferromagnetic correlations. The negative values of θ seem to support this concept. The value of the effective magnetic moment perhaps can be attributed exclusively to the 5f electrons of the uranium because the paramagnetism of YMn_4Al_8, $LaMn_4Al_8$, $ThMn_4Al_8$, $LaCu_4Al_8$ and $ThCu_4Al_8$ is apparently very weak (for a review see [8]). The data reported in [9] for YMn_4Al_8 are unrealistic. However, such comparisons should be made with extreme caution as it has been proven by the examination of specific heat of YMn_4Al_8, and $LaMn_4Al_8$ [10]. The determined effective magnetic moments are difficult to interpret because their values are a result of the interplay of many interactions as crystal field, hybridization or polarisation by conduction electrons. The increase of the Mn concentration is reflected in the increase of paramagnetism (see Fig. 1) which in turn could result from the increase of the temperature independent susceptibility, which for the sample with $x=3$ is close to the value for $ThMn_4Al_8$ [11].

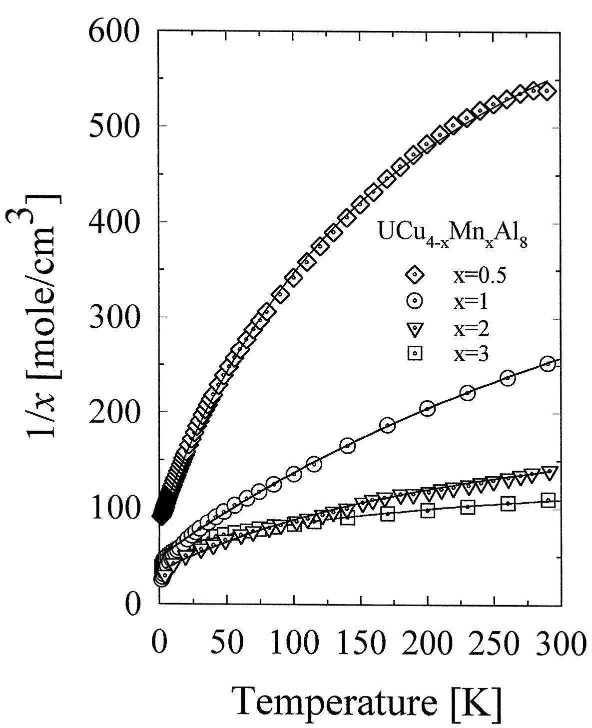

Fig. 1. Reciprocal magnetic susceptibility, χ^{-1}, of the $UCu_{4-x}Mn_xAl_8$ system versus temperature for various Mn concentration *x*.

Fig. 2 shows the magnetic susceptibility, χ, of the $UCu_{4-x}Mn_xAl_8$ system at low temperature ($T<50$ K). One can see that χ in this temperature range is weakly temperature dependent, with a slight increase when the temperature decreases below $T=5$–10 K. It is an indication that the substitution of Mn for Cu does not enhance the magnetic order in UCu_4Al_8 but on the contrary, the magnetic order is suppressed by Mn. It is strange at the first sight because there is magnetic order observed in the lanthanide compounds [8]. Considering the much higher hybridization in the uranium compounds than in the corresponding lanthanide compounds and a lack of magnetic ordering in UMn_4Al_8 [6], this behavior is justified. Recently, we have made other substitutions, namely we have obtained another type of solid solution, $UCu_x(Al_{1-y}Ga_y)_{12}$ in which only a very weak influence of Ga on the magnetic ordering is observed [12]. This can be explained by inspecting the crystal structure in which Mn is most probably located in the *8(f)* sites whereas Ga is substituted in the *8(i)* and *8(j)* positions. Therefore, Mn atoms due to closer location (nearest neighbors) have a much stronger influence on the uranium atom.

The electrical resistivity of the $UCu_{4-x}Mn_xAl_8$ system versus temperature for various Mn concentrations *x* is shown in Fig. 3 in the normalized form ρ/ρ(290 K). The measurements were performed on polycrystalline samples, therefore, the conclusions from Fig. 3 have only qualitative character. The general shape of ρ/ρ(290 K) vs. T is metallic. The plots for samples with lower Mn concentration exhibit some anomalies which most probably are related to antiferromagnetic correlations. For $x=0.5$ the $\rho(T)$ plot can be approximated by a $\rho \propto T$ dependence at low temperature which could be considered as an indication of the nFl state. Attempts to approximate the temperature dependence of the electrical resistivity for $x=0.5$ and 1.0 by $\rho \propto AT^2$ (proposed for the HF) turned out to be nonphysical ($A \approx 10^{-5}$).

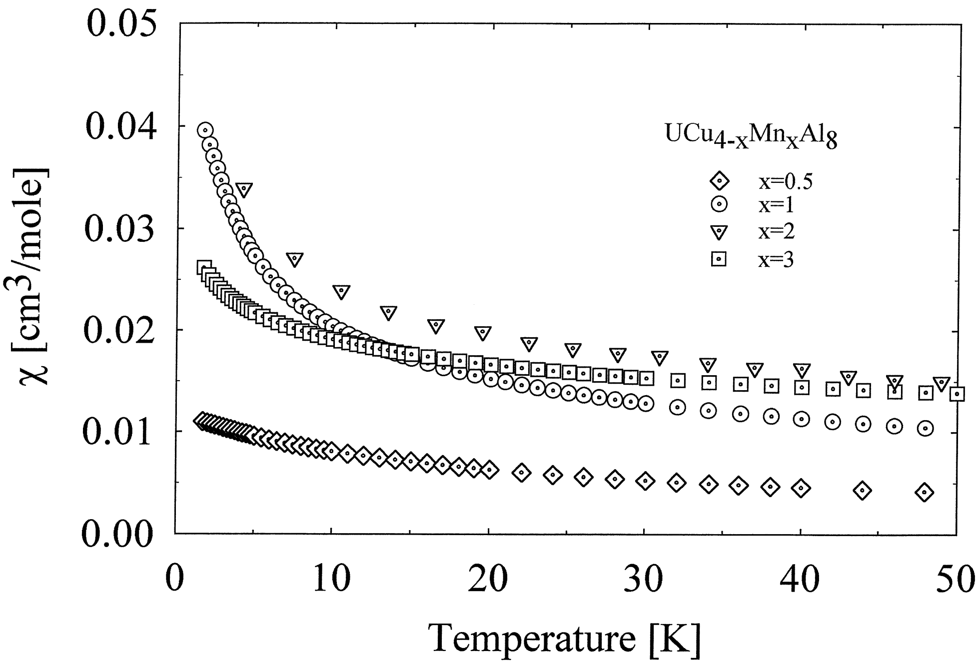

Fig. 2. Magnetic susceptibility, χ, of the $UCu_{4-x}Mn_xAl_8$ system versus temperature $T<50$ K, for various Mn concentration x.

4. Conclusions

Two goals of this research were formulated at the beginning of this paper. The first one, the indication of the enhancement of magnetism and of the nFl state, produced negative results. As far as the second goal is concerned, we have obtained the $\rho \propto T$ dependence for the $x=0.5$ sample which could be a hint of the nFl state, but this conclusion needs further confirmation.

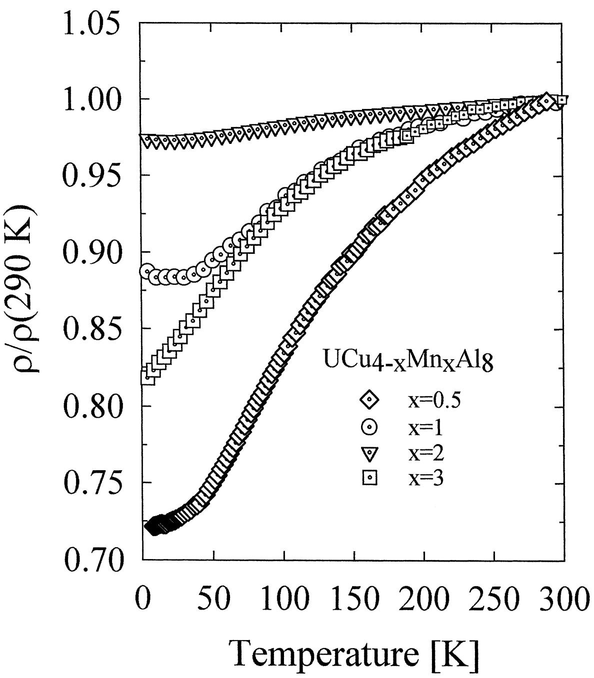

Fig. 3. Electrical resistivity, ρ, normalized to room temperature value, ρ/ρ (290 K) of the $UCu_{4-x}Mn_xAl_8$ system versus temperature for various Mn concentration x.

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