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# Magnetic, Electrical and Thermodynamic Properties of UCuT$_x$Al$_{11-x}$ Alloys Where T = Mn, Fe and $x = 4$ and 5

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**Abstract** The structure, magnetic, electrical, and thermodynamic properties of UCuT$_x$Al$_{11-x}$ alloys, where T = Mn or Fe and $x = 4$ or 5 are presented. The behavior of the Fe alloys is ferromagnetic-like with the Curie points amounting to 180 and 230 K, and the saturation magnetic moments under magnetic field of 5 T equal to 4.75 and 6.02 μ$_B$/f.u., respectively, whereas under a magnetic field of about 34 T the magnetic moments amount to 6.9 and 9.0 μ$_B$/f.u. for the alloys with $x = 4$ and 5, respectively. The Curie points are reflected in the temperature dependence of the specific heat in which the anomalies are found at 180–200 and 230 K for alloys with $x = 4$ and 5, respectively, however, it shows no reflection in the temperature dependence of the electrical resistivity. The field dependence of the magnetization at $T = 1.9$ K for both compounds exhibits considerable hysteresis. There is a pronounced difference between ZFC and FC magnetization in its temperature dependence below the Curie point for materials with $x = 4$ and 5. The Mn alloys exhibit ferrimagnetic-like character for which, supposedly, the interplay of the uranium and manganize sublattices is responsible. Magnetic transitions are determined at $T_N = 300$ ($x = 4$) and 380 K ($x = 5$). However, those anomalies do not find confirmation in the temperature dependence of the specific heat and the electrical resistivity. Magnetic moments determined at $T = 1.9$ K and in a magnetic field of 5 T are very low and in both cases amount to about 0.35 μ$_B$/f.u. and

these values are slightly higher in a magnetic field of 34 T reaching a value of about 1.5 μ$_B$/f.u. Also for the Mn alloys the clear difference between ZFC and FC magnetization in its temperature dependence below the Curie point is observed.

**Keywords** Uranium intermetallics · Magnetic properties · Electrical properties · Specific heat

## 1 Introduction

Prolonged investigation of the UCu$_x$Al$_{12-x}$ derivatives have shown the possibility of the substitution of Cu by other 3$d$ elements. Particularly, the substitution by Mn and Fe instead of Cu as the transition elements results in many pseudoternary alloys showing magnetic anomalies having ferrimagnetic character (see e.g. Refs. [1] and [2]). However, the anomalies observed in the temperature dependence of the magnetic susceptibility (magnetization) did not find a confirmation in measurements of the temperature dependence of the electrical resistivity and specific heat examined both in magnetic field and in zero magnetic field [2]. Moreover, the low temperature coefficient of electronic specific heat, $\gamma$, for all alloys exhibits strongly enhanced value. Unfortunately, we cannot decide if the reason of the enhanced $\gamma$ is heavy fermion state or crystallographic disorder. However, it is clear from our former experiments that $\gamma$ is roughly independent on composition and particularly on Cu content [3].

At present we report on magnetic, electrical, and thermodynamic properties of UCuT$_x$Al$_{11-x}$ alloys, where T = Mn or Fe and $x = 4$ or 5. The attempts of obtaining the alloys with T = Cr, Co, and Ni proved to be unsuccessful.

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**Table 1** Lattice parameters and magnetic data for the $UCuT_xAl_{11-x}$ alloys

| T | $x$ | $a$ [nm] | $c$ [nm] | $T_{C,N}$ | $p_s$ [$\mu_B$/f.u.][*] | $\chi_0$ [cm$^3$/mol] | $\theta_p$ [K] | $p_{eff}$ [$\mu_B$/f.u.] |
|---|---|---|---|---|---|---|---|---|
| Fe | 5 | 0.8632 | 0.4982 | 230 | 9.0 | −0.015 | 253 | 11.7 |
| Fe | 4 | 0.8761 | 0.4991 | 180 | 6.9 | −0.007 | 201 | 9.3 |
| Mn | 5 | 0.8779 | 0.5075 | 380 | 1.5 | | | |
| Mn | 4 | 0.8783 | 0.5079 | 300 | 1.5 | | | |

[*]In pulsed magnetic field of 34 T at $T = 4.2$ K

## 2 Experimental

The investigated compounds were obtained as described previously [1, 2]. The magnetization was examined at 1.9 K in magnetic field up to 5 T, with a SQUID magnetometer, and at 4.2 and 77 K in the steady magnetic field to up 14 T with a string magnetometer, and in pulsed field up to 34 T with a pulse duration of 10 ms [4]. The temperature dependence of magnetization was investigated using a SQUID magnetometer technique in the temperature range $T = 1.9$–400 K in various magnetic fields in zero field (ZFC) and field cooled (FC) modes. The standard equipment in our laboratory was used for electrical measurements at 4.2–300 K. The determination of the specific heat in the temperature range 2–400 K was carried out using Quantum Design PPMS machine.

## 3 Results and Discussion

### 3.1 X-ray Diffraction

X-ray diagrams were single phases, and all reflections belong to the corresponding ThMn$_{12}$-type structure, at least in the range of accuracy of the measurements equals 5 %. Unfortunately, from present experiments we cannot propose the distribution of the T atoms to be in the $8f$, $8i$, and $8j$ positions as has been done for the $UCu_xFe_{5-x}Al_7$ with ($x = 2$, 3.5) alloys by the $^{57}$Fe Mössbauer effect examination [5]. The lattice parameters are collected in Table 1 and one can see that the change of composition or substituting atom T is not reflected in a serious change of lattice parameters.

### 3.2 Magnetic Properties

In Fig. 1 the magnetization of the alloys with $x = 4$ in magnetic fields up to 5 T at 1.9 K is presented. The behavior of the Fe alloy is like for typical ferromagnet with considerable high hysteresis (70 %). Saturation is attained in relatively low magnetic field and the saturation magnetic moment M$_s$ amounts to 4.75 $\mu_B$/f.u. For the Mn alloy the magnetization, after initial sharp increase with increase of magnetic field
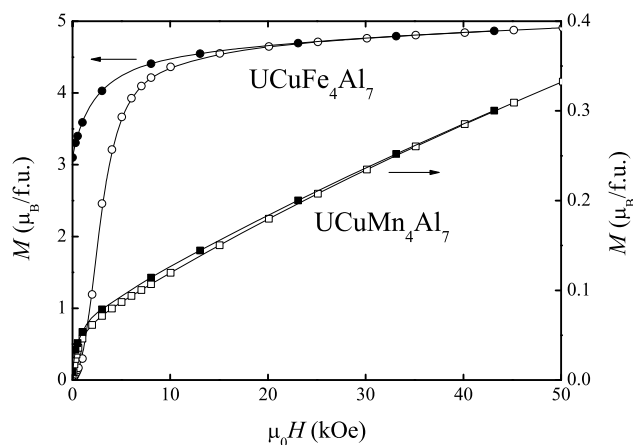


**Fig. 1** Magnetization versus magnetic fields for the $UCuT_xAl_{11-x}$ alloy with $x = 4$ at 1.9 K

then exhibits moderate but linear increase and in the magnetic field of 5 T its value amounts to 0.33 $\mu_B$/f.u. Such a field dependence can be indication of the ferrimagnetic ordering, which is possible for the material with two potentially magnetic atoms, however, the magnitude of magnetic moment can suggest also other magnetic phenomena, e.g. spin fluctuation or spin glass state.

The temperature dependence of magnetization of the Mn compound with $x = 4$ is presented in Fig. 2. The magnetization is measured in the field of 50 Oe in ZFC and FC mode. The plot is ferromagnetic-like but the numerical values are very low. Below $T$"$_N$" $= 300$ K the magnetization values for the FC run are apparently higher. This anomaly (at 300 K), though, did not find confirmation in the temperature dependence of specific heat or electrical resistivity. Such a behavior at relatively high temperature (see below) does not exclude the existence of magnetic ordering.

The temperature dependence of magnetization of the Fe compound with $x = 4$, measured in magnetic fields of 50 and 100 Oe in ZFC and FC mode, is shown in Fig. 3. The character and numerical values of magnetization strongly suggest ferromagnetic character of the material below Curie point amounting to $T_C = 180$ K. The clear difference between ZFC and FC runs below $T_C$ most probably is a result from the domain structure effects.
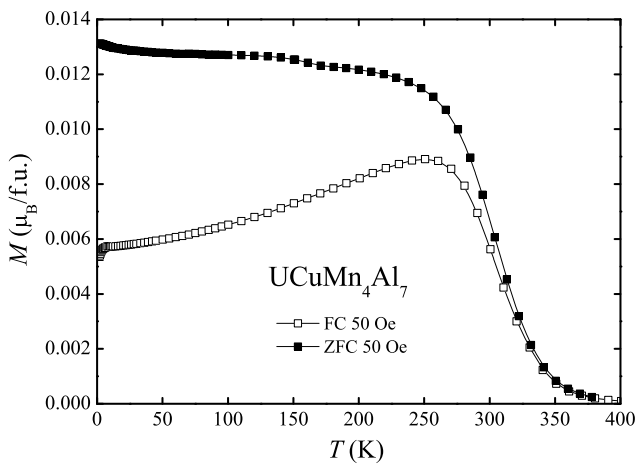
**Fig. 2** Magnetization versus temperature for UCuMn$_4$Al$_7$ in ZFC and FC mode in magnetic field of 50 Oe
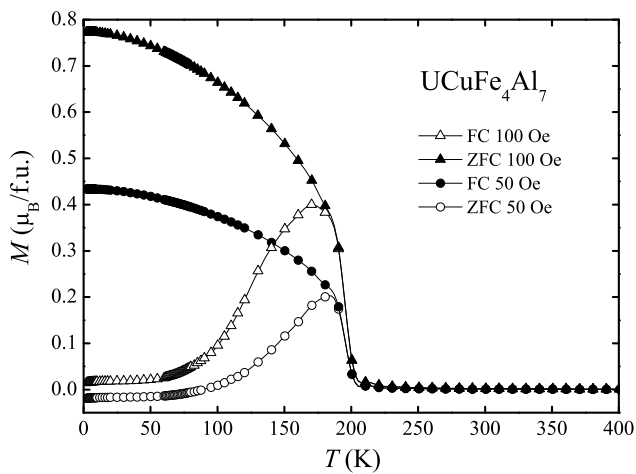


**Fig. 3** Magnetization versus temperature for UCuFe$_4$Al$_7$ in ZFC and FC mode in various magnetic fields



**Fig. 4** Magnetization versus magnetic fields for the UCuT$_x$Al$_{11-x}$ alloys with $x = 5$ at 1.9 K
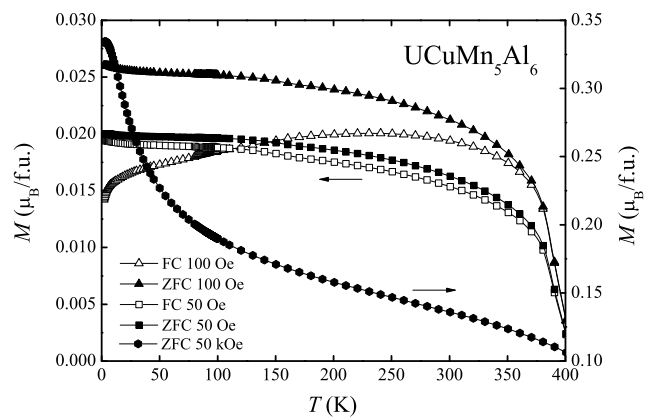


**Fig. 5** Magnetization versus temperature for UCuMn$_5$Al$_6$ in ZFC and FC mode in various magnetic fields. Note that ZFC the run in 50 kOe corresponds to right scale

As presented in Fig. 4, the magnetization versus magnetic field plot for the compounds with transition element concentration $x = 5$ at $T = 1.9$ K is qualitatively similar to that with lower concentration of Fe and Mn (Fig. 1). However, even for the former alloy full saturation is not observed in applied magnetic field range and "saturation" magnetic moment amounts to $M_s = 6.02$ μ$_B$/f.u. Hysteresis is also substantial and equals ∼ 70 %. The value of the saturation magnetic moment is an indication that only Fe is responsible for magnetic ordering in this compound. Moreover, the absence of additional anomalies in $M(T)$ plots (see also Fig. 6) can be an indication that only the Fe sublattices order below $T_C$. It does not exclude the possibility that the Fe and U sublattices order at the same temperature. The $M(T)$ plot for the Mn alloy does not attain saturation and the highest measured value of magnetic moment in 5 T equals also 0.35 μ$_B$/f.u.

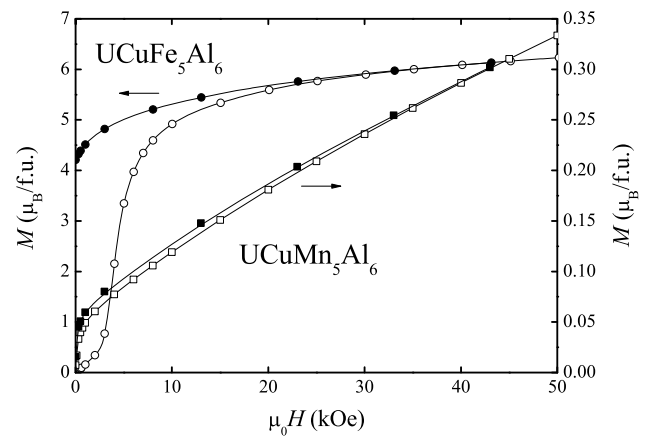Shown in Fig. 5 is the temperature dependence of magnetization for Mn alloy with $x = 5$; it is rather complex. Mag-

netization is very low, with transition temperature 380 K; it is, however, determined for low fields only (for 100 Oe and below). For a magnetic field of 50 kOe (5 T) the $M(T)$ plot has different shape and at present we do not understand this behavior. Only one conclusion can be drawn: that the transition temperature is very sensitive to the magnetic field.

Magnetization versus temperature plot for the Fe compound with concentration $x = 5$ (Fig. 6) is very similar to that for $x = 4$ except that the Curie temperature amounts to 230 K. Moreover, ZFC magnetization measured at 50 Oe in the low temperature region exceeds that obtained at 100 Oe. This last run attains negative values at very low temperature, a phenomenon which was observed in many multicomponent $f$-electron intermetallics and discussed in an extensive review by one of the present authors [6].

In Fig. 7 the magnetization at $T = 4.2$ K under pulsed magnetic field with a pulse duration of 10 ms is presented for all investigated alloys. The iron compounds exhibit markedly higher magnetization than the Mn compounds. The Fe alloy with $x = 5$ even in these intense fields does not
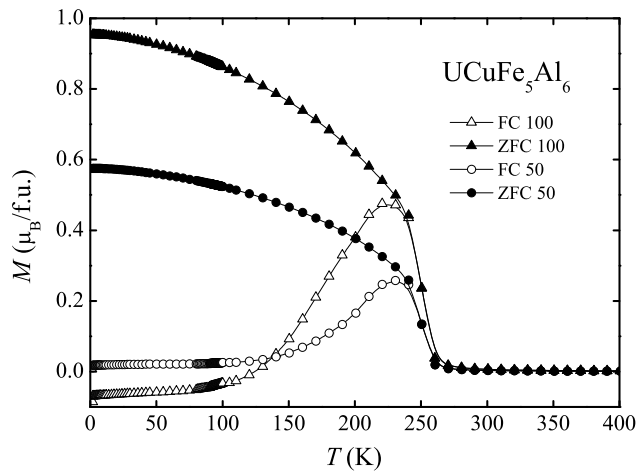
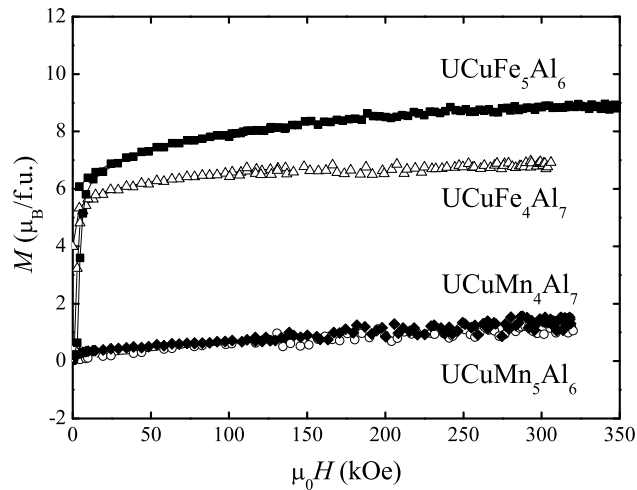**Fig. 6** Magnetization versus temperature for $UCuFe_5Al_6$ in ZFC and FC mode in various magnetic fields



**Fig. 8** Reciprocal magnetic susceptibility versus temperature of the $UCuFe_xAl_{11-x}$ alloys under magnetic field of 100 Oe. *Solid lines* correspond to the fitted plots



**Fig. 7** Magnetization versus pulsed magnetic field for the $UCuT_xAl_{11-x}$ alloys at $T = 4.2$ K with pulse duration 10 ms



**Fig. 9** Electrical resistivity versus temperature for the $UCuT_xAl_{11-x}$ alloys

show the saturation. We do not understand at present this lack of saturation which can result from a paraprocess but also from other reasons. The "saturation" magnetic moment obtained in the highest magnetic field is equal to 9.0 $\mu_B$/f.u. and this value indicates that not only five Fe atoms participate in magnetic ordering. The Fe alloy with $x = 4$ exhibits the saturation of magnetization with magnetic moment amounting to 6.9 $\mu_B$/f.u. and this value seems to suggest also the contribution of both magnetic elements (Fe and U) in total magnetic moment. The magnetization of the Mn compounds is rather weakly dependent on magnetic field, however, in principle in a linear way. The "saturation" magnetic moment for both materials reaches a value of about 1.5 $\mu_B$/f.u.

The temperature dependence of the reciprocal magnetic susceptibility $\chi^{-1}$ of the Fe alloys obtained in a magnetic field of 100 Oe is presented in Fig. 8. One can see that
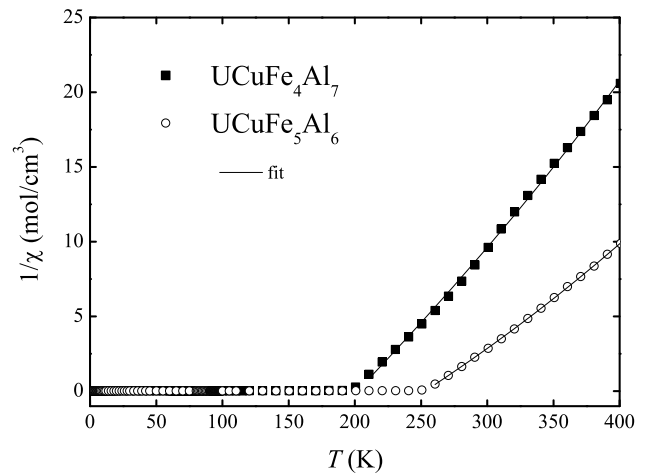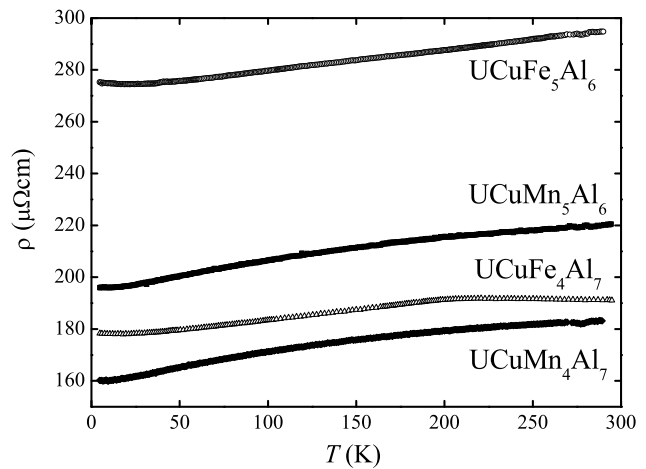
the approximately linear temperature dependence of $\chi^{-1}$ is observed above $T = 250$ K and 210 K for $x = 5$ and 4, respectively. The linear part can be described by modified Curie–Weiss law with the numerical values of the temperature independent susceptibility $\chi_0$, the Weiss constant $\theta_p$ and the effective magnetic moment $p_{eff}$ collected in Table 1. The temperature independent part of magnetic susceptibility is small, whereas the effective magnetic moments and the Weiss constant are higher for the alloy with $x = 5$. This observation is a simple consequence of the higher iron concentration. However, these data do not provide firm information about the uranium contribution to the total magnetism likely as in the case of the saturation moment.

We did not try to determine effective magnetic moments for the Mn alloys because the paramagnetic range is too narrow.
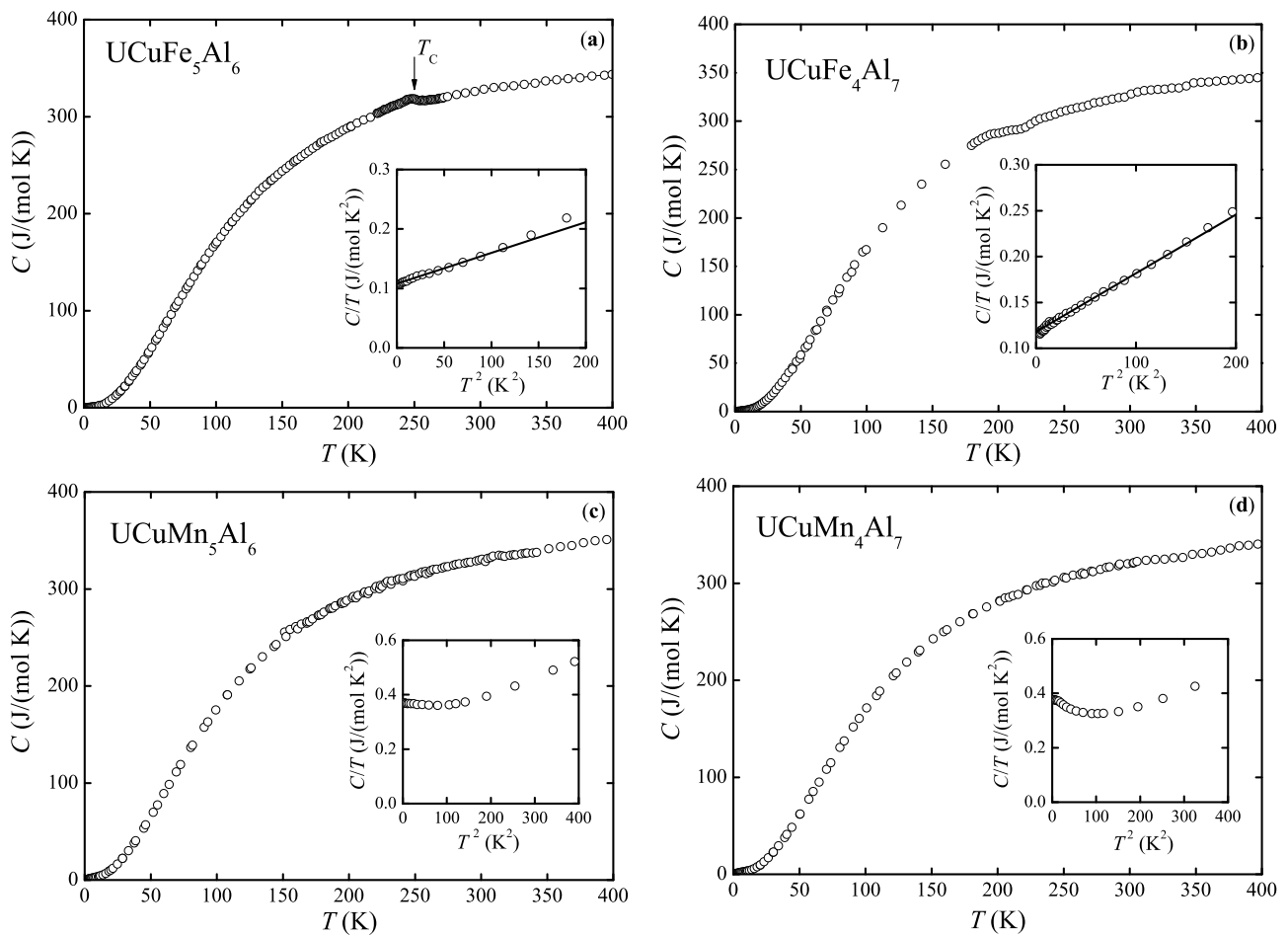
**Fig. 10** Specific heat versus temperature for the $UCuT_xAl_{11-x}$ alloys. *Insets* present $C/T$ vs. $T^2$ at low temperature: (**a**) for T = Fe, $x = 5$; (**b**) for T = Fe, $x = 4$; (**c**) for T = Mn, $x = 5$; (**d**) for T = Mn, $x = 4$

**Table 2** Thermodynamic data for the $UCuT_xAl_{11-x}$ alloys

| T | $x$ | $\gamma$ [mJ/mol K$^2$] | $\Theta_D$ [K] | $T_C$ [K] |
|---|---|---|---|---|
| Fe | 5 | 108(1) | 365(2) | 230 |
| Fe | 4 | 118(1) | 341(1) | 180–200 |
| Mn | 5 | 380 | | |
| Mn | 4 | 380 | | |

The electrical resistivity versus temperature of all investigated materials is presented in Fig. 9. It is clear that resistivity is weakly temperature dependent with low RRR. We do not know if this feature is innate for these materials or only results from imperfections of the samples. All plots are smooth without the traces of anomalies observed in the temperature dependencies of magnetization and specific heat (for Fe alloys).

The thermodynamic properties of the $UCuT_xAl_{11-x}$ alloys are presented in Figs. 10a–10d and in Table 2. The specific heat versus temperature in each case presents in principle smooth curves and for the Fe alloys weak anomalies corresponding to the Curie points (arrows in Figs.) are seen. The anomalies observed in magnetometric measurements of the Mn alloys have no corresponding anomalies in $C(T)$. The insets present the $C/T$ vs. $T^2$ dependencies at low temperature which allows to determine the coefficient of electronic specific heat, $\gamma$, and the Debye temperature, $\Theta_D$. For the Fe alloys $\gamma$ is moderately enhanced, possible due to the magnetic ordering, however, for the Mn compounds this enhancement is substantial. Unfortunately, we cannot present any firm conclusion about its reason. Moreover, these data are not very precise because we have no results for non-magnetic references. The Debye temperatures demonstrate

a magnitude which is common for the $f$-electron intermetallics (see e.g. Ref. [2]). Some time ago Gschneidner et al. [7] have presented the concept of the false heavy fermions for compounds which exhibit a variable composition range, which permits non-magnetic atom disorder (Cu or Al). That behavior gives rise to the spin glass behavior and accounts for the enhanced C/T value. But to clarify this behavior further experiments like neutron diffraction or a.c. susceptibility are indispensable.

## 4 Conclusions

The observed properties of the $UCuT_xAl_{11-x}$ alloys allow us to conclude that the materials with T = Fe exhibit clear ferromagnetic properties which, however, do not provide a clear-cut answer on the contribution of the uranium atom to the total magnetic ordering. For T = Mn alloys the magnetic properties are complicated and we cannot learn about the magnetic ordering in them if any. All alloys exhibit metallic character, however, with relative high electrical resistivity both at low and high temperature. The determination of the thermodynamic characteristics is only approximate because there are no data for reference non-magnetic alloys. The Sommerfeld coefficient $\gamma$ is enhanced

for all alloys, although for the Fe compounds only moderately and the reason for that could be ferromagnetic ordering. Strong enhancement for the materials with Mn could be related to crystallographic disorder or to the spin-glass state. Further improvement of these results could be expected from measurements on the single crystal materials, from neutron diffraction or a.c. magnetic susceptibility.

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