SPECIFIC HEAT OF THE $UCu_{4+x}Al_{8-x}$ DERIVATIVES*

W. Suski[a,b], A. Czopnik[a], K. Wochowski[a] and M. Sołyga[a]

[a]Institute of Low Temperature and Structure Research
Polish Academy of Sciences, P.O. Box 1410, 50–950 Wrocław, Poland

[b]International Laboratory of High Magnetic Fields and Low Temperatures
Gajowicka 95, 53–421 Wrocław, Poland

(Received July 10, 2002)

The low temperature ($T < 70$ K) specific heat obtained for the $UCu_{4+x}Al_{8-x}$ derivatives in which the $3d$ elements (Cr, Mn and Ni) have been substituted for Cu, and Ga for Al is presented. The parent alloys have exhibited an alloying induced transition from simple antiferromagnetic ordering for low Cu–concentration (for $x \leq 1.25$) to heavy-fermion-like properties (for $x \geq 1.5$). The crystallographic disorder introduced by a change of stoichiometry could be the reason for enhanced γ. These components slightly change both the crystallographic relations and electronic structure, and the determined thermodynamic data strongly suggest other reasons for an enhanced γ than crystallographic disorder. The occurrence of the $T^3 \ln T$ term in specific heat for all alloys except that of Ni provides the conclusion about spin fluctuation state. The Ni compound is antiferromagnetic below $T_N = 27.5$ K.

PACS numbers: 71.20.Lp, 71.27.+a, 75.40.Cx

1. Introduction

The discovery of heavy fermion-like behaviour in $UCu_{4+x}Al_{8-x}$ ternaries [1,2] has aroused interest in the properties of their derivatives. The investigation of parent alloys has shown an alloying induced transition from simple antiferromagnetic ordering for low Cu concentration ($x \leq 1.25$) to heavy fermion (HF)-like behaviour ($x \geq 1.5$) [2]. Recently, Nishioka *et al.* [3] have presented a phase diagram of this system and have established that there probably exist three states: antiferromagnetic (AF), heavy fermion (HF) and non-Fermi liquid (nFL) ones depending on x. As the most pronounced indication of the HF state, enhanced coefficient of the electronic

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

specific heat (γ) is suggested. These alloys exhibit the tetragonal $ThMn_{12}$-type structure ($I4/mmm$ space group) with four nonequivalent crystallographic sites: $2a$ occupied by uranium atoms, and $8f$, $8i$ and $8j$ positions in which the Cu and Al atoms are located. Such a structure can produce a crystallographic disorder which in turn can be the reason for the high value of γ. To explain this problem, we have started the research on the derivatives of $UCu_{4+x}Al_{8-x}$ alloys in which Fe [4], Ni [5], Mn [6] and Cr [7] being substituted for Cu, and Ga for Al [8], have been examined. These components slightly change both crystallochemical relations (the U–U separation and possibly the crystallographic sites occupation) and electronic structure. The magnetic and electrical properties of these systems investigated so far did not provide any clear-cut indication of the influence of these added components on the behaviour of pseudo-ternaries. Currently, we report on specific heat investigation of the selected representatives of these pseudo-ternary alloys: UCu_3CrAl_8, $UCu_{3.5}Mn_{0.5}Al_8$, UCu_3NiAl_8 and $UCu_4Al_{7.5}Ga_{0.5}$ at temperature $T = 2$–70 K. Preliminary results for the Cr compound have been published in [7].

2. Experimental

The compounds have been prepared as it was described previously [5–8] by melting the elements in stoichiometric quantities in an arc furnace under protective argon atmosphere. For measurements the as-cast samples were used. An annealing process resulted in the appearance of weak parasitic lines in the X-ray pattern. The home made, fully automatic calorimeter was used for measurements of specific heat at $T = 4.2$–70 K.

3. Results and discussion

The investigated samples are single phases with the lattice parameters given in [5–8]. The results of present investigation are shown in Figs. 1, 2 and in Table I. In figures there are C/T *versus* T^2 plots for the Mn, Cr and Ga alloys presented, whereas for the Ni compound the same plot is shown as inset and in the main figure C/T as a linear function of T is plotted. The reason for that will be explained further. The thermodynamic data collected in the table have been obtained from the figures via the equations:

$$C/T = \gamma_{\mathrm{HT}} + \beta T^2 , \tag{1}$$

$$\beta = 12/5\pi^4 Nk/\Theta_{\mathrm{D}}^3 , \tag{2}$$

where Θ_{D} is Debye temperature, and N and k have the usual meaning, whereas γ_{HT} was evaluated extrapolating the linear part of C/T *versus* T^2 plot to $T = 0$. γ_{LT} was obtained by extrapolation of the low temperature part of the same plot to $T = 0$. It is seen from Figs. 1(a), 1(b) and 2(a)

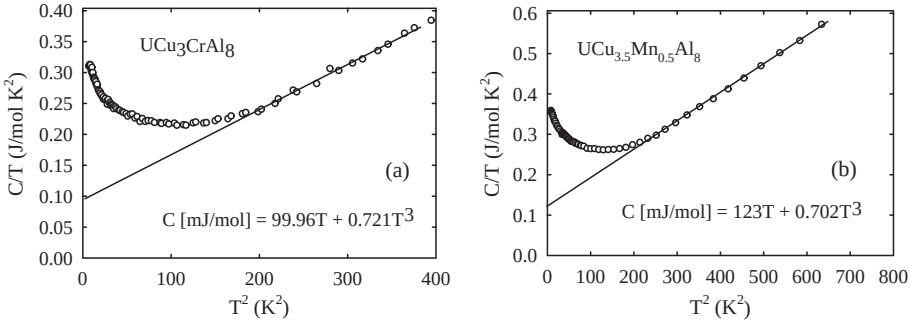

Fig. 1. Low temperature specific heat C/T versus squared temperature for UCu_3CrAl_8 (a) and $UCu_{3.5}Mn_{0.5}Al_8$ (b).

that independently of the alloy composition, all thermodynamic data are fairly close to each other. It is an indication that crystallographic disorder is not the reason for the enhanced γ. Therefore, the uranium atoms seem to be responsible for the γ values. The Figs. 1(a), 1(b) and 2(a) do not reveal any phase transition and hence a large low temperature γ corresponds to a strong correlation of the uranium electrons. The shape of the C/T *versus* T^2 plots and the temperature dependence of magnetic susceptibility at low temperature [6–8] resemble the type 5 spin-fluctuating materials (*e.g.* $CeSi_{2-x}$) as shown by Ikeda *et al.*, [9]. A contribution of the term $T^3 \ln T$ occurs for all three alloys (not shown) and the upturn is quite strong in these materials. However, the weakness of the $T^3 \ln T$ term is probably due to the low T_{sf}. As it is seen from Fig. 2(b) the Ni compound exhibits a different behaviour related to an antiferromagnetic ordering below $T_{\mathrm{N}} = 27.5$ K confirmed by the neutron diffraction experiment [10]. However, γ_{HT} value is very close to those for other compounds and at present we cannot explain this observation.

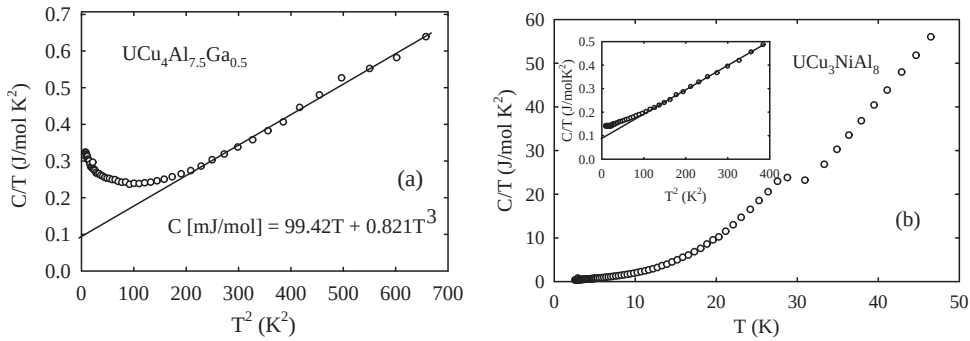

Fig. 2. Low temperature specific heat C/T *versus* squared temperature for $UCu_4Al_{7.5}Ga_{0.5}$ (a) and *versus* temperature for UCu_3NiAl_8 (b). Inset shows C/T versus T^2.

TABLE I

Thermodynamic data for the in $UCu_{4+x}Al_{8-x}$ derivatives.

Compound	γ_{LT} [mJ/mol K^2]	γ_{HT} [mJ/mol K^2]	β [mJ/mol K^4]	Θ_D [K]
UCu_3CrAl_8	330	100	0.721	165
$UCu_{3.5}Mn_{0.5}Al_8$	370	123	0.702	166
UCu_3NiAl_8	150	99.8	0.991	$T_N = 27.5$ K
$UCu_4Al_7.5Ga_{0.5}$	350	99.4	0.821	158

4. Conclusions

The investigated alloys exhibit fairly close thermodynamic data, except for the Ni compound. Probably, they exist in the spin-fluctuation state at low temperature which is suppressed by Ni doping, even though electron correlations, as indicated by γ_{HT}, are similar for all measured compounds. Further experiments are necessary to elucidate the properties of these intriguing materials.

REFERENCES

[1] M. Drulis, A. Baran, B. Staliński, W. Suski, R. Felten, F. Steglich, L. Pawlak, *Thermochim. Acta* **139**, 219 (1989).

[2] C. Geibel, U. Ahlheim, A.L. Giorgi, G. Sparn, H. Spile, F. Steglich, W. Suski, *Physica B* **163**, 194 (1990).

[3] T. Nishioka, Y. Kurakahashi, R. Fukuda, M. Kotani, *Czech. J. Phys.* **46**, 2065 (1996).

[4] W. Suski, A. Baran, L. Folcik, K. Wochowski, T. Mydlarz, *J. Alloy. Compd.* **181**, 249 (1992)

[5] W. Suski, K. Wochowski, D. Badurski, *Bull. Pol. Acad. Sci. Chem.* **42**, 286 (1994)

[6] W. Suski, K. Wochowski, *J. Alloy. Compd.* **317–318**, 395 (2001).

[7] W. Suski, A. Czopnik, K. Wochowski, *J. Nuclear Science Techn.* in press.

[8] W. Suski, K. Wochowski, D. Badurski, *Physica B* **312–313**, 286 (2002).

[9] K. Ikeda, S.K. Dhar, M. Yoshizawa, K.A. Gschneidner Jr., *J. Magn. Magn. Mater.* **100**, 293 (1991).

[10] G. André, F. Bourée, K. Wochowski, W. Suski, *Solid State Commun.* **104**, 189 (1997).