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ANOMALOUS TRANSPORT AND THERMAL PROPERTIES OF THE SOLID SOLUTIONS $U_{1-x}Th_xCu_5M$ (M = Al, Ga, In AND Sn)

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UCu_5M-type compounds are moderate heavy fermion systems with an antiferromagnetic order below T_N = 18, 16–25, and 25 K for M = Al, (Ga, In), and In, respectively, and a ferrimagnetic order with T_C = 54 K for M = Sn. We present the results of magnetoresistance measurements as a function of temperature and applied magnetic field for the pure compounds and for their solid solutions containing Th, i.e. $U_{1-x}Th_xCu_5M$. Only in the case of UCu_5In a substantial positive contribution to the magnetoresistance was observed at low temperatures, as for normal antiferromagnets. For the Al- and Sn-based alloys the magnetoresistance at low temperatures is negative, and its field variation proves the existence of Kondo-type interactions. On Th-substitution the magnetic phase transition is shifted to lower temperature and simultaneously the magnetoresistance becomes less negative. The field dependence of the magnetoresistance of these alloys can be well described by the Coqblin–Schrieffer model. Interestingly, the heat capacity measurements have revealed a general tendency to increase the linear coefficient γ with magnetic dilution of a given UCu_5M compound by Th-substitution. This feature reflects an enlargement of the effective mass of conduction electrons with the increase in both the unit cell volume and magnetic dilution. Such a tendency is also observed for the system $UCu_5(In_{1-x}Ga_x)$, though the unit cell is here suppressed with increasing x. Thus, a similar physical picture is reached in all these systems but involving probably different mechanisms.

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1. Introduction

Within the past two decades the U-based intermetallics have been of great interest because they show a variety of physical phenomena. Some of these intermetallics belong to the family of heavy fermion (HF) compounds in which the Kondo interaction scaled by k_BT_K (T_K is the Kondo temperature) competes with the exchange interaction scaled by k_BT_m (T_m is the temperature of a magnetic order). Whereas the former interaction tends to form a nonmagnetic ground state,

the latter one, which in a simple case can be that of indirect Ruderman–Kittel–Kasuya–Yosida (RKKY)-type, tends to order f-derived local magnetic moments. Closeness in energies of these two interactions leads to a large variety of magnetic and transport properties. Some satisfactory explanation of these properties has been achieved in recent years by the Anderson-type approach to conduction electron – localized electron mixing-type interactions [1]. In dense systems, labeled Kondo lattices (KL), a many-body resonance occurs at low temperatures due to the interference effect of coherent scattering. As a result the electronic specific heat coefficient, $\gamma(0)$, enormously increases and the proportionality is held $\gamma(T \to 0\ \mathrm{K}) \sim T_k^{-1}$. There occurs a strong renormalisation of the conduction band states at the Fermi energy E_F [2] and the following proportionality is also held $\gamma(0) \sim (m_\mathrm{eff})$, where m_eff is the effective carrier mass, being usually of the order of several hundreds of the free electron mass, m_e.

In the last few years one is a witness of a rapidly increasing interest in studying dilute uranium-based alloys. This interest comes from the discovery in some of such systems of the diversity in $\chi(T)$ and the specific heat C over T ratio, $C(T)/T$, as $T \longrightarrow 0$ K. This clear deviation from the Fermi liquid (FL) theory suggests the presence of unconventional Kondo effects in such alloys. To explain this non Fermi liquid (NFL) behaviour at low temperatures, which was encountered in a set of dilute cerium and uranium alloys, many theoretical debates have recently been undertaken in the literature (see for example Ref. [3]), without however finding a final consent.

The central point of the HF compounds is a double role of their f-electrons, which, on the one hand, exhibit at higher temperatures the properties expected for well-localized moments and can be described, e.g. in the context of crystal-field effects, but on the other hand, they behave at low temperatures as itinerant carriers with extremely large effective masses, attributed to the heavy quasiparticles [4, 5]. Since it takes only a very small energy range $k_\mathrm{B}T^*$ ($T^* \sim T_\mathrm{K}$, being usually several tens of K), the linear specific heat contribution becomes enormously enhanced below T^*.

To this kind of HF systems belong also the newly discovered by us UCu_5M-type intermetallics, where M = Al, In, and Sn [6]. These compounds though display at low temperatures moderately enhanced $\gamma(0)$-values (below about 400 mJ/(K^2 mol)), at the same time exhibit magnetic order of the Kondo-reduced magnetic moments.

In this paper we review some so far recognized basic physical properties of UCu_5M-intermetallics, and in addition we report some new results of our X-ray, magnetic, electrical transport, and thermal studies of the $U_{1-x}Th_xCu_5M$-type solid solutions. In such solid solutions, the incorporation of Th into the parent crystal structure of a given UCu_5M compound brings about not only a magnetic dilution and in consequence the expected vanishing of the magnetic order, but also some negative chemical pressure, due to increase in the unit cell volume.

2. Experimental details

All the samples were prepared by arc melting required quantities of the constituent metals under an argon atmosphere. The resulting buttons were turned

over and remelted several times. The samples wrapped in tantalum foil were then annealed under vacuum in quartz tube at 850°C for a week. On the basis of X-ray diffraction analysis of the alloys, the ranges of mutual solubility limits for a given solid solution were roughly established. The susceptibility and magnetization were measured by means of a SQUID magnetometer in the temperature range of 1.7–300 K. The electrical resistivity down to 1.5 K was performed using a standard four-probe DC method. Longitudinal magnetoresistivity data $\Delta\rho/\rho = \{\rho(B,T) - \rho(0,T)\}/\rho(0,T)$ were recorded at selected temperatures in magnetic field $B = 8$ T on zero-field cooled samples. The samples had typical dimensions $0.5 \times 0.5 \times 5.0$ m^3 and the uncertainty in its geometrical factor was of the order of 10%.

3. Results

3.1. (U,Th)Cu$_5$Al system

Since the physical properties of $(U_{1-x}Th_x)Cu_5Al$ have been previously discussed in Refs. [7] and [8], here we will confine ourselves to their basic characteristics only. UCu_5Al crystallizes in its own tetragonal crystal structure type (s.g. $I4/mmm$), which remains unchanged in the $U_{1-x}Th_xCu_5Al$ solid solutions up to $x = 0.35$. Although the variations in the unit cell dimensions versus Th-composition is nonlinear, the c/a ratio for all the samples up to $x \approx 0.35$ keeps a constant value of 0.772. Above this composition other solid solutions with the general chemical formula (U,Th)(Cu,Al)$_5$ are formed, having a different type of structure, namely the hexagonal $CaCu_5$-type. Crystallographic data for UCu_5Al obtained from X-ray and neutron diffraction analysis at room temperature [8] are close to each other and are as follows: $a = 6.411(1)$ Å, $c = 4.951(1)$ Å and $V = 203.5$ Å^3. There is one U-atom position at the 2b sites and two unequal positions for copper atoms, Cu_I at 8h and Cu_{II} at 4d. The latter sites are shared also by the Al atoms with the ratio 1:1 with respect to Cu_{II}. The mean distance between uranium atoms in UCu_5Al is as large as about 5.12 Å. This leads in principle to a conclusion that this ternary uranium compound should be treated as a well-localized $5f$-electron system.

Below $T = 18$ K, UCu_5Al orders antiferromagnetically with a sine-modulated incommensurate magnetic structure described by a wave vector $\boldsymbol{k} = (0, 0, 0.55)$ and the moment aligned along the c-axis with an amplitude $M = 1.45(5)\mu_B$ at 1.4 K [8]. The magnetic susceptibility for $x = 0.0, 0.1$, and 0.2 goes through a distinct maximum at their T_N, which in turn decreases linearly to almost 0 K for x being close to 0.3 [7]. For the latter composition, the low-temperature susceptibility decreases suddenly by a factor of 2.5, which may indicate the change in the crystal-field (CF) ground state occurring near this composition. Thus, one can argue that the relatively short composition range of the stability of both the crystal structure and magnetic ordering under Th^{4+} doping may be attributed to the change in the oxidation state of uranium ion from 3+ to 4+, just near $x = 0.3$. The inverse magnetic susceptibility χ^{-1} plotted against temperature for the above solid solutions was found to be strongly curvilinear indicating an anisotropic behaviour of these alloys in the paramagnetic state. For this reason we have approximated this behaviour by

a modified Curie–Weiss (MCW)-law at temperatures above *ca.* 50 K. The effective magnetic moment μ_{eff} only slightly changes with increasing Th-content, namely between 2.2–2.4μ_{B}. At the same time a large increase in negative paramagnetic Curie temperature, Θ_{p}, is observed for $x = 0.30$ (−120 K) as compared to the remaining compositions (around −30 K). The independent of temperature part of the susceptibility χ_0 varies rather smoothly from 6.5 to 8.0 × 10^{-4} $\text{cm}^3/(\text{mol U})$.

In order to get some insight into the anisotropic behaviour of UCu_5Al, susceptibility measurements were performed on the sample having crystallites "oriented" in the external field. The estimated temperature dependences of $\chi_{\parallel}(T)$ and $\chi_{\perp}(T)$, i.e. those for crystallites oriented along and perpendicular to the *c*-axis with respect to the external magnetic field, respectively, are indeed very anisotropic and have been presented in Fig. 1 of Ref. [9]. The calculated average values, $\chi_{\text{av}}(T)$, were in good agreement with the relevant bulk data. The observed susceptibility anisotropy as well as the low-temperature deviation from the straight-line behaviour of the Knight shift versus susceptibility [9], clearly confirm the presence of CF splitting in UCu_5Al.

The temperature dependence of the electrical resistivity, $\rho(T)$, in the temperature range of 4.2–300 K, for the $U_{1-x}Th_xCu_5Al$ alloys, has been presented in Ref. [7]. For all studied samples, i.e. with $x = 0.0$, 0.1, 0.2, and 0.3, $\rho(T)$ increases logarithmically with decreasing temperature down to about 100 K according to the equation

$$\rho(T) = \rho_0 + \rho_{\text{ph}}(T) - c \ln T,$$

where ρ_0 is the residual resistivity, ρ_{ph} is a phonon contribution to the resistivity and c is the coefficient of the logarithmic term, characteristic of Kondo systems. Below about 100 K, ρ for the $x = 0.0$ and 0.1 goes first through a broad maximum near T_{N}, probably caused by crystal-field effects, and then, just below T_{N}, grows rapidly to saturate finally at the lowest temperatures. As an example, in Fig. 1a we display ρ vs. $\log T$ for UCu_5Al. The sharp increase in $\rho(T)$ just below T_{N} might be at the first sight attributed to the presence of a narrow energy gap in the electronic spectrum of $U_{1-x}Th_xCu_5Al$ alloys. For such an explanation argues the sharpness of the transition of T_{N}, shown in the inset to Fig. 1a. An alternative explanation is the formation of a new Brillouin zone in the antiferromagnetic state, which competes with both the spin-disorder scattering (tendency to decrease the resistivity) and with a single Kondo-type scattering on the ground state (tendency to achieving ρ_{max} at $T = 0$ K). Further additional studies of these alloys on single crystalline samples under pressure, planned at the near future, are thought to throw more light on this problem. In turn, a characteristic feature of $\rho(T)$ for the $x = 0.2$ and 0.3 samples is a distinct diminishing of the jump in $\rho(T)$, and an occurrence of a broad maximum in $\rho(T)$, with only a small upturn at the lowest temperature measured, i.e. at 4.2 K. Thus, we can conclude that both crystal and magnetic instabilities arise around $x \approx 0.35$, i.e. at the limit of the existence of the $U_{1-x}Th_xCu_5Al$ solid solutions.

The results of magnetoresistance (MR) measurements of UCu_5Al sample, taken up to 13 T, have already been reported [10]. Among others it was found that $\Delta\rho/\rho(B)$ at this field and corresponding T_{N}, which shifts down to 10 K,

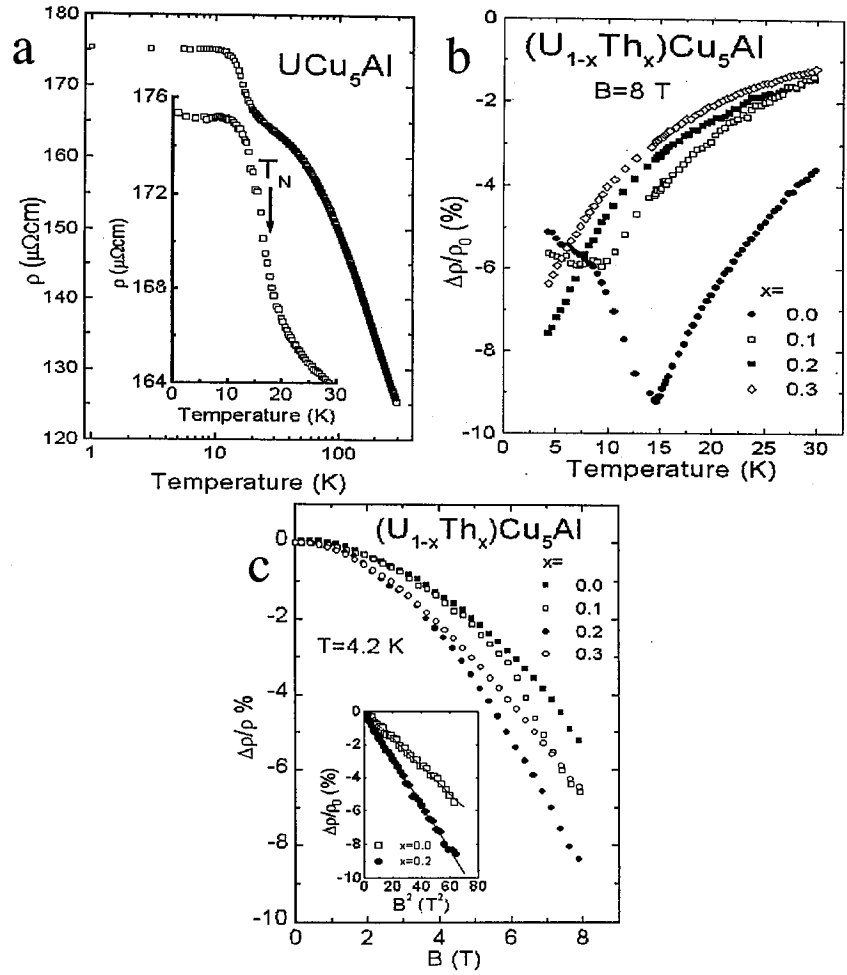

Fig. 1. (a) Electrical resistivity for UCu_5Al, the inset shows $\rho(T)$ at LT. (b) Magnetoresistance vs. temperature for $U_{1-x}Th_xCu_5Al$ alloys. (c) Magnetoresistance vs. field for $U_{1-x}Th_xCu_5Al$ alloys at 4.2 K, the inset: $\Delta\rho/\rho_0$ vs. B^2 for $x = 0$ and 0.2.

reach as a high value as -28%. Also the observed decrease in $\Delta\rho/\rho$ as the field rises was interpreted as a consequence of a field-induced suppression of the Kondo effect. Such a dependence for all studied compositions x but taken up to 5 T is in turn shown in Fig. 1b. As seen, MR for all these alloys is negative and its absolute magnitude increases as the temperature is decreased. As it has already been observed at higher fields [10], the $x = 0.0$ sample exhibits a sharp maximum at T_N being shifted to a lower temperature of 15 K. A less sharp anomaly with reduced $\Delta\rho/\rho$-value is detected for $x = 0.1$. For more dilute alloys, however, no sign of such a maximum is noted down to 4.2 K. In general, the occurrence of the aforementioned anomaly in $\Delta\rho/\rho(T)$ is attributed to increasing positive contribution to the total MR under cooling, caused by the onset of an antiferromagnetic order at T_N. Since the magnetic field as large as 8 T shifts simultaneously T_N to

lower temperatures, the effect of the competition between the Kondo and magnetic exchange interactions could not be seen for more dilute alloys even down to 4.2 K (see Fig. 1b). In Fig. 1c we present the $\Delta\rho/\rho(B)$ dependence for all x. As illustrated in the inset of this figure, the negative MR of $x = 0.0$ and $x = 0.2$ follows at 4.2 K a B^2-dependence, up to the maximum field strength applied. Such a behaviour is characteristic of independent Kondo impurities, a role which uranium atoms play in all the alloys studied, despite their respective large concentration in the compound. However, no signature of the coherent state at lower temperatures was found in any of these materials.

The specific data obtained down to 0.35 K have already been published in Ref. [7]. The C/T versus T^2 curves taken up to 20 K show below the broad phase transition peaks a shallow minimum and a small upturn at the lowest temperatures. An extrapolation of $C(T)/T$ to $T = 0$ K yields $\gamma(0) \approx 180$, 250, 310, and 370 mJ/(K^2 mol U) for $x = 0.0$, 0.1, 0.2, and 0.3, respectively. In conclusion, it is clear that the substitution of Th for U up to 30 at.% in UCu_5Al causes a linear increase in the $\gamma(0)$ value together with a tendency to the disappearance of the antiferromagnetic order, also vanishing in linear manner with x. Thus, the heavy fermion character of these alloys grows distinctly with x up to the dilution limit.

3.2. $(U_{1-x}Th_x)Cu_5In$ system

UCu_5In crystallizes in the orthorhombic $CeCu_5In$-type structure being a ternary ordered variant of the $CeCu_6$-type (space group *Pnma*) [11]. Anomalies observed in the temperature dependences of the susceptibility, transport, and thermal properties indicate that an antiferromagnetic transition occurs at 25 K [6]. As neutron-diffraction experiment has established, the magnetic moments of $1.43\mu_B$ at 1.4 K are tilted in the unit cell to the c-axis by 58°. The X-ray studies have shown that the solid solutions $(U_{1-x}Th_x)Cu_5In$ are formed in the whole range of concentrations x without change in the crystal structure. All three lattice parameters increase with increasing Th-content, showing a positive departure from Vegard's law. The antiferromagnetic order vanishes with a steep drop in T_N for a composition near $x = 0.5$. The observed convex nature of the inverse magnetic susceptibility $\chi^{-1}(T)$, measured for all the alloys, indicates the presence of a strong magnetocrystalline anisotropy. The MCW law applied for the $0 \leq x \leq 0.7$ samples gives a rather small change in μ_{eff} (2.2–2.4μ_B), Θ_p (50–70 K), and χ_0 (6–8 $\times$ 10^{-4} emu/(mol U)). A distinct increase in these values is detected only under further dilution.

The electrical resistivity variation of UCu_5In with temperature has already been presented in Ref. [6]. With lowering temperature it goes through a very broad bump with a maximum at 180 K, probably caused by a strong CF effect. Subsequently, at temperatures close to $T_N = 25$ K, $\rho(T)$ shows first a minimum and then a sharp maximum at about 10 K, as illustrated in the inset of Fig. 2a. In this figure, the temperature dependences of the reduced resistivity of the alloys with x: (a) 0.0, 0.2, and 0.4 and (b) 0.6, 0.7, 0.8, and 1.0 are presented. The most interesting feature of these functions is a sudden change in the character of $\rho(T)$ when passing from $x = 0.0$ to the composition range of $0.2 \leq x \leq 0.4$. Namely,

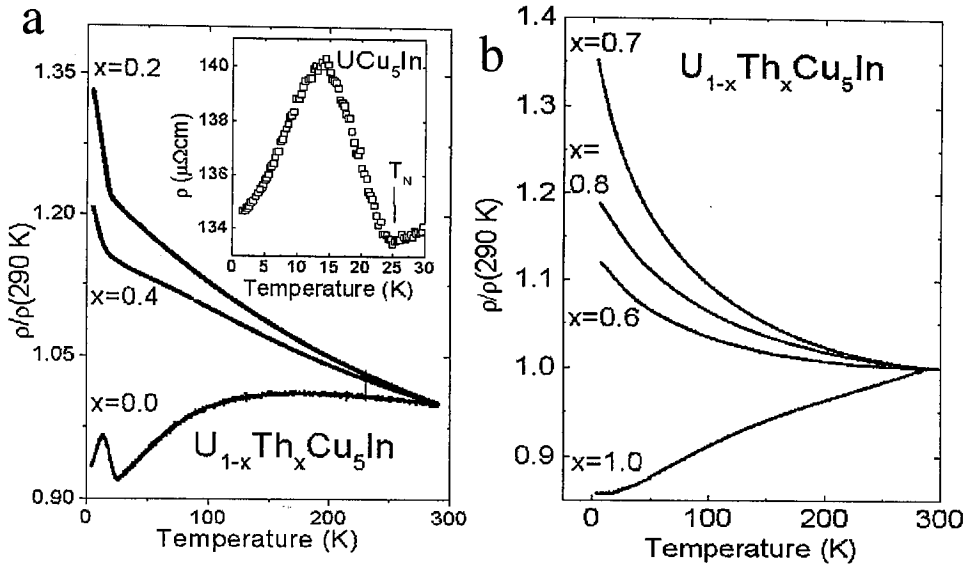

Fig. 2. Reduced resistivity for $U_{1-x}Th_xCu_5In$ alloys with (a) $x < 0.5$ and (b) $x > 0.5$. Inset: $\rho(T)$ of UCu_5In at LT.

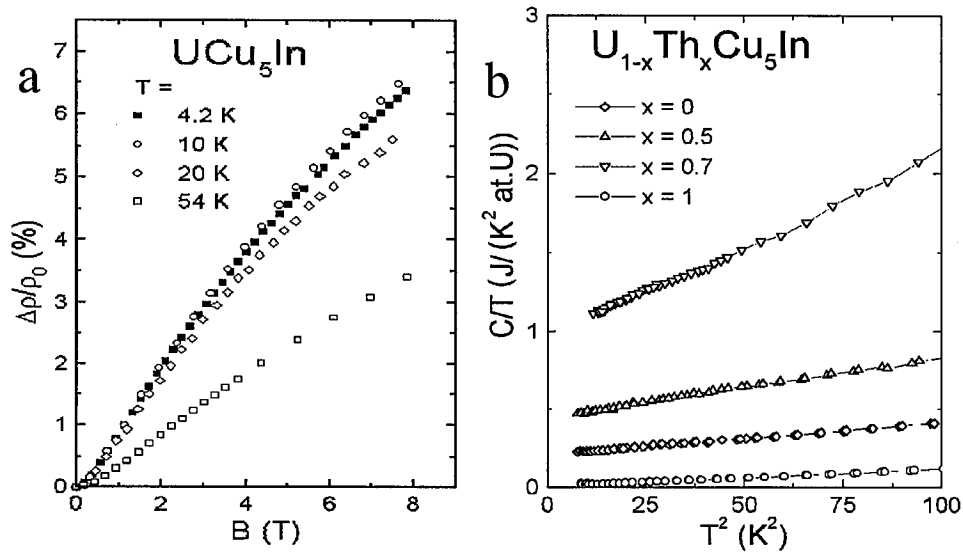

Fig. 3. (a) Magnetoresistance vs. field of UCu_5In at various temperatures; (b) C/T vs. T^2 for $U_{1-x}Th_xCu_5In$ alloys.

for the latter compositions one deals with the Kondo-like behaviour at higher temperatures and with a resistivity jump at T_N, alike for the $U_{1-x}Th_xCu_5Al$ alloys.

The C/T vs. T^2 plots for $x = 0.0$, 0.5, 0.7, and 1.0 are illustrated in Fig. 3b. As seen, the extrapolated C/T values to $T = 0$ K increases from about 220 ($x = 0$)

to 1000 mJ/(K^2 mol U) ($x = 0.7$). Thus, these data give strong evidence for the occurrence of some important change in the electronic structure of UCu_5In with progressive magnetic dilution by Th-content.

3.3. $UCu_5(In_{1-x}Ga_x)$ system

In view of the fact that ternary compound "UCu_5Ga" does not exist we have instead studied the solid solutions $UCu_5(In_{1-x}Ga_x)$, which can be formed up to $x = 0.75$. All of them are antiferromagnetic and their T_N decreases from 25 to 16 K with increasing Ga-content. At the same time the electronic specific-heat coefficient increases by about 25%. The overall behaviour of the electrical resistivity as a function of temperature is reminiscent of that described above for $U_{1-x}Th_xCu_5In$. Like in the latter system, a small Ga/In substitution suppresses the sharp maximum in $\rho(T)$ of UCu_5In and instead a jump in the resistivity occurs just below the respective T_N, with some tendency to saturation at low temperatures. For the alloys with $0.375 \leq x \leq 0.5$ a clear $\ln T$-dependence of the resistivity is observed in the paramagnetic region. For larger Ga-content, the amplitude of the resistivity jump below T_N diminishes and a convex curvature in $\rho(T)$ again arises. The temporary data for this system has been published in Ref. [12].

3.4. $(U_{1-x}Th_x)Cu_5Sn$ system

UCu_5Sn crystallizes with the hexagonal $CeNi_5Sn$-type of structure (space group $P6_3/mmc$), which is related to the orthorhombic structure of UCu_5In [13]. In this structure the uranium atoms occupy two twofold positions, U_I — 2d and U_{II} — 2a with coordination numbers 20 (18Cu+2Sn) and 18 (12Cu+6Sn), respectively. The lattice parameters are $a = 4.998(1)$ Å, $c = 20.271(4)$ Å and $V = 438.5(2)$ Å^3. This crystal structure was found for all compositions within the solid solutions $U_{1-x}Th_xCu_5Sn$.

In contrast to the above described UCu_5M antiferromagnets, UCu_5Sn shows a sharp rise of the magnetization below 54 K indicating a ferromagnetic-like phase transition [14]. From recently performed neutron scattering measurements we have found a ferrimagnetic structure with a propagation vector $\boldsymbol{k} = (0,0,0)$. In this structure the alignment of U magnetic moments is along the hexagonal c-axis. The most striking feature of this structure is that the U moments arranged in the (0,0,0) plane, formed by the sites 2a, are 10 times larger in magnitude than those in the adjacent (0,0,0.25) plane, formed by the sites 2d [15]. The inverse magnetic susceptibility at temperatures 200–1000 K follows a Curie–Weiss law with $\mu_{eff} = 3.11\mu_B$ and $\Theta_p = -63$ K. The negative sign of the paramagnetic Curie temperature implies predominance of negative, antiferromagnetic exchange interactions. The inverse susceptibility plotted against temperature below 300 K for the solid solutions $U_{1-x}Th_xCu_5Sn$ shows for all the compositions studied $0.0 < x < 1.0$ a large convex curvature. This behaviour is caused not only by a large magnetocrystalline anisotropy, as found for the antiferromagnets described above, but also by the ferrimagnetic behaviour of these alloys. As one can conclude

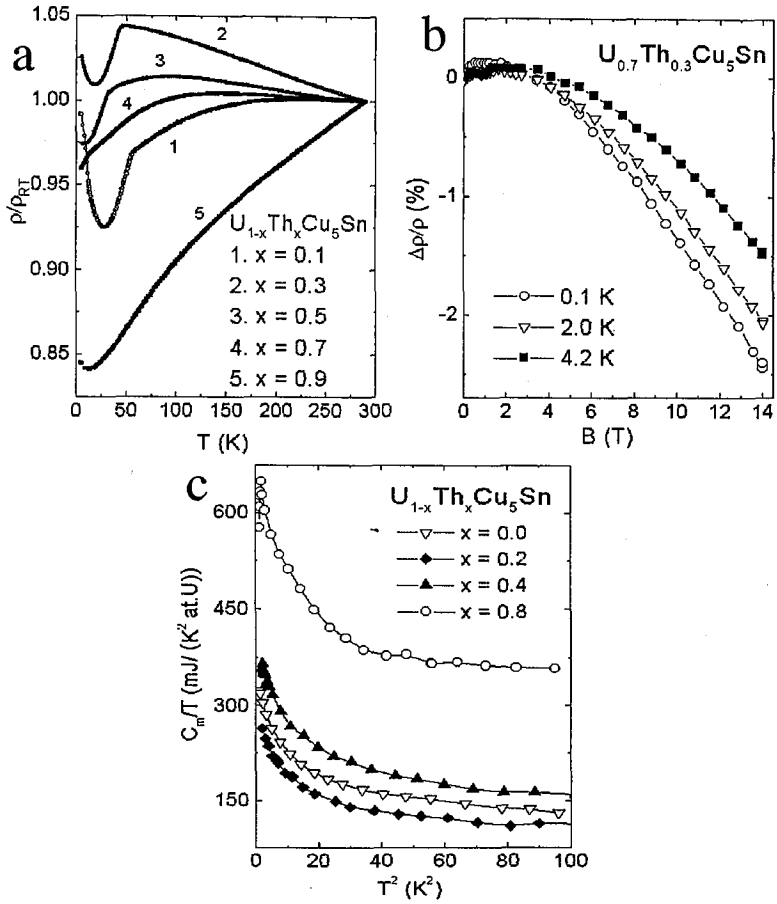

Fig. 4. (a) Reduced resistivity for $U_{1-x}Th_xCu_5Sn$ alloys. (b) Magnetoresistance vs. field of $U_{0.7}Th_{0.3}Cu_5Sn$ at various temperatures. (c) C/T vs. T^2 for $U_{1-x}Th_xCu_5Sn$ alloys.

from the thermomagnetic curves, the Curie temperature T_C varies with x almost linearly and on this basis the ferrimagnetic order has been detected up to $x \approx 0.9$.

The electrical resistivity vs. temperature for $U_{1-x}Th_xCu_5Sn$ is displayed in Fig. 4a. Under cooling from room temperature, the resistivity of UCu_5Sn initially decreases and then near T_C it drops due to the onset of the magnetic order, but at $\sim T_C/2$, $\rho(T)$ goes through a deep minimum and then rises steeply when the temperature is lowered. This steep increase at $T < T_C/2$ we attributed to the Kondo-type scattering (see for details Ref. [15]). As Fig. 4b indicates, already the 10 at.% - substitution U/Th makes the slope of $\rho(T)$ negative down to T_C, and below this temperature a similar behaviour to that found for $x = 0.0$ is observed. With further dilutions the initial deep depression becomes shallow and finally disappears and then there is a progressive change towards the metallic-type behaviour observed for $x \geq 0.9$.

The $\rho(T)$, measured for UCu_5Sn down to 40 mK and corrected for the phonon contribution by substracting $\rho(T)$ of $ThCu_5Sn$, shows saturation and from the inflection point of the ρ_{mag} vs. $\ln T$ curve obtained one would roughly establish $T_K = 15$ K [15]. We have searched for additional signatures of the Kondo character of UCu_5Sn and of the uranium-rich compositions in the solid solutions with Th. The MR for the pure stannide was previously measured at 50 mK, 0.25 K, and 3 K in fields up to 14 T [14]. In these low temperatures and high fields MR shows a negative curvature, depending nearly quadratically on B, which indicates the dominant Kondo-type behaviour of UCu_5Sn. This behaviour could be satisfactorily analyzed in terms of the $J = 1/2$ Coqblin–Schrieffer model (see details in Refs. [14] and [15]). Similar low-temperature dependences of MR for the solid solutions $U_{1-x}Th_xCu_5Sn$ have also been observed up to $x \approx 0.7$. As an example, in Fig. 4b we show the $\Delta\rho/\rho$ vs. B curves for $x = 0.3$ taken at temperatures below 4.2 K.

Finally, in Fig. 4c we present the specific-heat data for several compositions x of the solid solutions with Th, obtained by Andraka at the University of Florida. From this figure it is clear that the values of C_m/T, corrected on the phonon contribution and expressed per mol U for all studied x, increase with lowering temperature. An extrapolation to $T = 0$ K yields for UCu_5Sn a $\gamma(0)$-value as high as 330 mJ/(K^2 mol U) [15]. With medium dilution by the Th-content, $\gamma(0)$ are slightly lower than that of $x = 0.0$, but for the dilution as large as $x = 0.8$, C_m/T increases even to a value of 700 mJ/(K^2 mol) at 1.4 K, but below this temperature its steep decrease is apparent. Nevertheless, all the above data clearly point to the heavy fermion character of all those alloys studied. This is even more striking as all these alloys exhibit the ferrimagnetic properties. To our knowledge, this is the first such an example in all known so far uranium HF compounds.

4. Conclusions

We have studied a new family of moderate heavy fermion alloys with the general chemical formula UCu_5M, where M = Al, In or Sn. Though such a compound with M = Ga does not exist, there is a possibility to substitute In by Ga in UCu_5In up to 75 at.%, holding the same crystal structure.

For all these alloys we have determined crystal structures, magnetic, transport, and thermal properties. Similar studies we have performed on the solid solutions of the $U_{1-x}Th_xCu_5M$-type. We have found that for the antiferromagnets UCu_5Al and UCu_5In the magnetic order vanishes where x approaches 0.35 and 0.5, respectively. All the UCu_5(In,Ga) alloys are also antiferromagnetic and their T_N slightly decreases with In/Ga substitution. On the other hand UCu_5Sn is a ferrimagnet and its T_C linearly decreases with the U/Th substitution up to $x \approx 0.9$. The characteristic feature of all these solid solutions is the presence of the Kondo interactions at low temperatures competing with the exchange interactions. As a result we have observed a general trend to increase the electron correlations with the magnetic dilution.

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