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Magnetic phase diagram of the $URh_{1-x}Ru_xGe$ system

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Magnetic phase diagram of the $URh_xRu_{1-x}Ge$ solid solutions has been reinvestigated by means of dc-magnetization, ac-susceptibility and electrical resistivity. We confirm ferromagnetic order only in alloys with $x \leq 0.2$. For a limited range of concentrations x between 0.3 and 0.35, we found features characteristic of short-range magnetic interactions. For compositions close to $x = 0.4$, non-Fermi liquid signature is observed in the low temperature resistivity. Thus, we report on the evolution of non Fermi-liquid behaviour at a magnetic–nonmagnetic border, neighbouring with the region of the short-range interactions.

Key words: *non-Fermi liquid; uranium ternary compounds, $URh_{1-x}Ru_xGe$; dc-magnetization, ac-susceptibility; electrical resistivity*

1. Introduction

The uranium intermetallic ternaries UT(Si, Ge) with the orthorhombic TiNiSi-type structure form one of the largest isostructural series of the uranium compounds [1]. The physical properties of these intermetallics have been extensively investigated for more than twenty years [2]. It turns out that within this series of compounds, a variety of magnetic properties can be observed by changing T – 3d, 4d or 5d electron transition metals involved. It is widely accepted that the development of magnetic behaviour is intimately associated with the strength of the 5f-ligand hybridization. Bearing in mind this mechanism, one can understand, for instance, the nonmagnetic ground state in URuGe and magnetic order in URhGe (T_C = 9.5 K) [2]. Since Ru possesses the electron configuration of $4d^75s^1$ whereas the configuration in Rh is $4d^85s^1$, the increasing number of conduction electrons in the sequence URuGe–URhGe weakens the 5f–4d hybridization. In consequence, it gives rise to formation of U 5f magnetic moments in the latter compound. Recent study of solid solutions $URh_{1-x}Ru_xGe$ made

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by Sakarya et al. [3] indicated a significant role of the 5f–4d hybridization. Moreover, the authors have shown an interesting magnetic phase diagram. The substitution of Rh by Ru first increases the Curie temperature up to about 11 K at $x = 0.1$, but with more Ru substitution T_C decreases and the ferromagnetic ordering disappears at $x \sim 0.38$. Because of possible experimental realization of a non-Fermi liquid ground state in a strongly correlated electron system at the magnetic-nonmagnetic border [4], we undertook to reinvestigate the magnetic phase diagram of $URh_{1-x}Ru_xGe$ [5]. We have carried out a detailed investigation of the low-temperature ac-magnetic susceptibility, dc-magnetization and electrical resistivity of samples between $0 \leq x \leq 1$. In this contribution, we present these new results, which partly confirm the reported diagram. One of the most remarkable findings to emerge from our study is that the non-Fermi liquid behaviour appears around the critical concentration $x_{cr} = 0.4$. We will also argue that the magnetic order vanishes actually near $x = 0.3$ and short-range correlations take place in alloys with x between 0.3 and 0.35.

2. Experimental details

The alloys with nominal compositions of the $URh_{1-x}Ru_xGe$ system, namely with x equal to 0, 0.1, 0.2, 0.3, 0.35, 0.375, 0.38, 0.4, 0.6, 1.0, were synthesized by arc melting in a Ti-gettered pure argon atmosphere. The samples were then wrapped in a Ta foil and annealed at 800 °C in evacuated silica tube for 60 h. The powder X-ray diffraction diffractometry (DRON and STOE, $Cu_{K\alpha}$ radiation) and EDAX microprobe analysis were used to examine the crystal structure, homogeneity and purity of the prepared materials. The Rietveld refinements were performed using the FULLPROF software. dc-Magnetization of powdered samples was measured using a Quantum Design SQUID magnetometer in the temperature range from 2 to 400 K and in magnetic fields up to 5.5 T. The measurements of ac susceptibility were performed using an Oxford Instrument susceptometer; ac field with the amplitude of 10 Oe and frequency 1 kHz were applied. Electrical resistivity measurements were performed on bar-shaped samples using the standard four-probe DC technique in the temperature range 2–290 K.

3. Results and analysis

3.1. Crystallographic data

The crystallographic data of URuGe and URhGe have been investigated by several groups [1]. These compounds were reported to crystallize in the orthorhombic TiNiSi -type structure and are characterized by the ratios $b/a = 0.651$ and $a/c = 0.885$, and 0.630 and 0.916, respectively. Based on the X-ray powder diffraction patterns, we conclude that all the investigated solid solutions of the $URh_{1-x}Ru_xGe$ system crystal-

lize in the same crystal structure as their parent compounds do. The respective lattice parameter ratios are presented in Fig. 1. An Inspection of the lattice parameters reveals that the *a* and *c* parameters change systematically with the concentration, while the *b* parameter remains almost constant. As a result, the unit cell volume seems to deviate from the linear behaviour at = 0.1, though a linear dependence of the *b*/*a* and *a*/*c* ratios with increasing *x* is found (see Fig. 1).

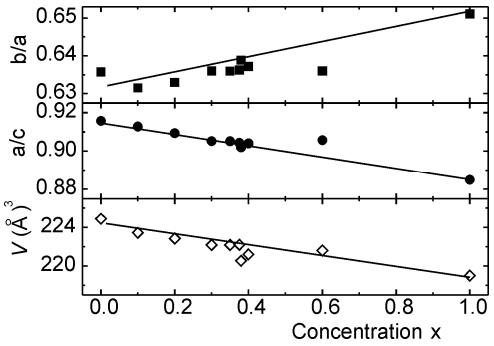

Fig.1. The ratio *a*/*c*, *b*/*a* and unit cell volume of $URh_{1-x}Ru_xGe$ as a function of concentration *x*

3.2. DC magnetization and ac-magnetic susceptibility

Figure 2 shows the temperature dependence of the reciprocal magnetic susceptibility $\chi^{-1}(T)$ for selected $URh_{1-x}Ru_xGe$ samples. Owing to a curvature of the $\chi^{-1}(T)$ curves, an analysis of the data was done with the use of a modified Curie–Weiss law:

$$\chi(T) = \frac{C}{T - \theta_p} + \chi_0$$

where $C = \frac{N_A \mu_{\text{eff}}^2 \mu_B^2}{3k_B}$ and θ_p is the paramagnetic Curie temperature, and χ_0 is a constant term, associated certainly with the Pauli paramagnetic contribution.

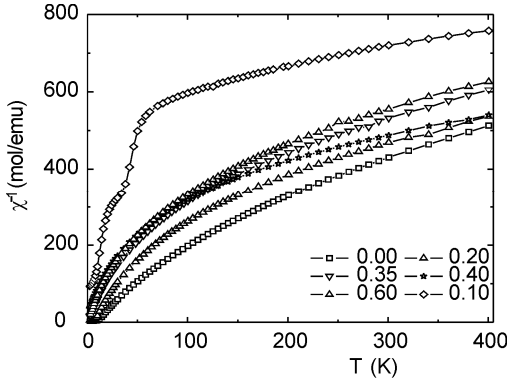

Fig. 2. Reciprocal susceptibility of selected $URh_{1-x}Ru_xGe$ solid solutions as a function of temperature. $\mu_0 H = 0.5$T

The fitted parameters for the data in the temperature range 100–400 K are given in Table 1. One recognizes that the θ_p values are found to be negative for all x. This fact implies the existence of antiferromagnetic interactions in all the studied alloys. A small magnitude of the effective moments hints rather an itinerant electron magnetism of the system.

The dc magnetization of compositions with $x < 0.3$ exhibits an anomaly associated with magnetic order of the U magnetic moments at low temperatures (not shown here). A gradual loss of magnetic order due to Ru substitution is illustrated in Fig. 3, where magnetization collected at 2 K is plotted versus the magnetic field.

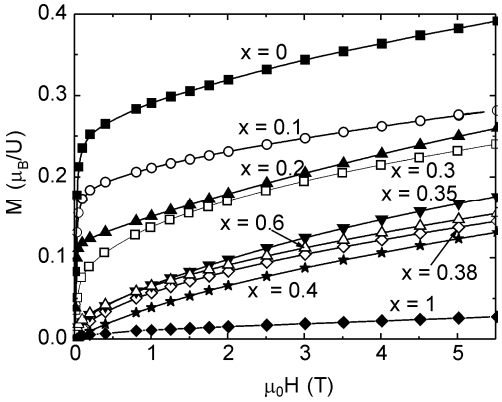

Fig. 3. Magnetization as a function of applied field for selected $URh_{1-x}Ru_xGe$ alloys. $T = 2$ K

Clearly, at 5.5 T the uranium magnetic moment steadily decreases with increasing concentration x. Further, for $x = 0$, 0.1 and 0.2 there is a spontaneous magnetization, that evidences the magnetic order in these alloys. On the other hand, the magnetization of alloys with $0.4 \geq x \geq 0.35$ resembles the behaviour of paramagnets. However, an attempt to fit these data to the expression

$$M = \chi_1 + \frac{\chi_3 H^3}{3!} + \ldots$$

up to the eleventh order failed (not shown here). Furthermore, the magnetic behaviour of $x = 0.3$ bears a resemblance to that of ferromagnets, however, as we show later, this composition does not show any long-range magnetic order at all.

An analysis of the Arrott plots may help us to determinate correctly the values of the Curie temperature for $x = 0$, 0.1 and 0.2. As an illustration, we display such an analysis in Fig. 4a for $x = 0.1$. As can be seen in Fig. 4b, there is no spontaneous magnetization in the sample $x = 0.3$. Therefore, we have no support for a long-range magnetic order in this alloy.

Complementary information on the magnetism of the $URh_{1-x}Ru_xGe$ alloys is provided via the ac-susceptibility measurements (Fig. 4). The maximum at T_{max} found in a real $\chi'(T)$ and imaginary parts $\chi''(T)$ of the ac-susceptibility of $x = 0$, 0.1 and 0.2 confirms the magnetic order in these alloys, but it not the case for $x = 0.3$.

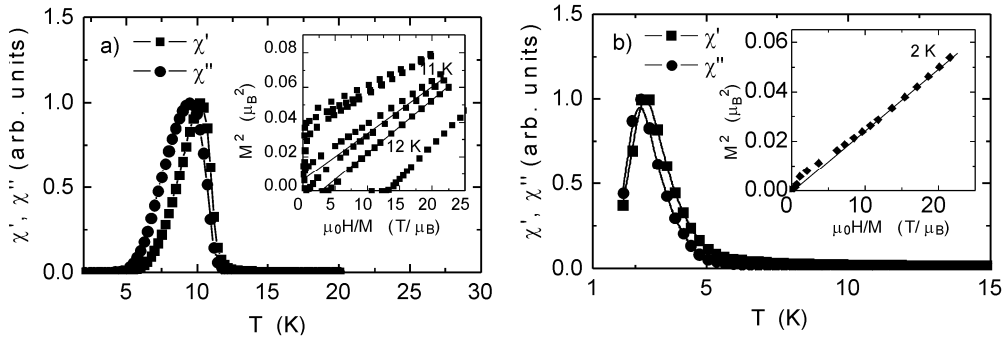

Fig. 4. Temperature dependence of ac-susceptibility for: a) $x = 0.1$ and b) $x = 0.3$. The Arrott plots for these samples are shown in the respective inset.

In particular, a slow decrease in the susceptibility (tail behaviour) in the sample of this composition for $T > T_{max}$ can evidence for some magnetic short-range interactions.

3.3. Electrical resistivity

The temperature dependences of the electrical resistivity of several $URh_{1-x}Ru_xGe$ compositions are depicted in Fig. 5. A characteristic feature of the $\rho(T)$ curves is the appearance of a broad maximum around 130 K. Further investigations are needed to clarify the nature of the observed phenomenon. At present, we tentatively attribute it to a Kondo-like effect. The magnetic order in compositions with $x \leq 0.2$ is supported by a drop in the resistivity at their T_C. No anomaly at low temperatures is visible in compositions with $x = 0.3$, 0.35, 0.38, 0.4, corroborating their nonmagnetic ground state.

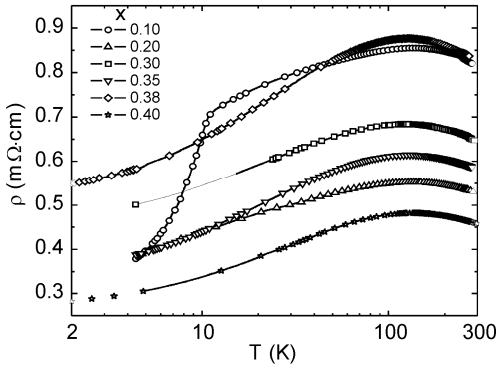

Fig. 5. Electrical resistivities of several $URh_{1-x}Ru_xGe$ solid solutions as a function of temperature

The resistivity data in the low temperature range, shown in Fig. 6, can be fitted well to a power law:

$$\rho(T) = \rho_0 + AT^n$$

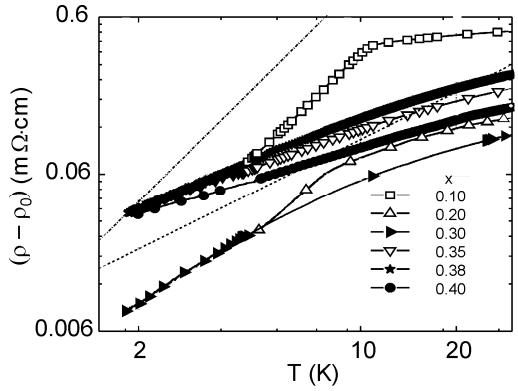

Fig. 6. Low temperature resistivity of selected $URh_{1-x}Ru_xGe$ samples. Dotted lines denote power T^n functions with $n = 2$ and 1, respectively

The change in the coefficient A and exponent n with x (Table 1) reflects changes in the magnetic properties of the alloys. For ferromagnetic alloys, the exponent n attains the value close to 2, characteristic of an electron–electron scattering. On the other hand, for nonmagnetic alloys $x = 0.35$–0.38, we found n close to 1. Such a value of the exponent implies a breakdown of the Fermi liquid behaviour [4]. The composition $x = 0.3$ is characterized by $n = 1.32$, very close to that of spin glasses ($n = 1.5$) [6].

Table 1. Physical properties of selected $URh_{1-x}Ru_xGe$ solid solutions*

x	μ_{eff} (μ_B/U)	θ (K)	T_C (K)			A (mΩ·cm·K^{-n})	n
			Arrott	AC sus.	$d\rho/dT$		
0.00	1.84	–3	9	–	9	0.011	2
0.10	1.51	–10	11–12	10.2	10.7	0.004	1.95
0.20	1.47	–6	7–8	7.5	8	0.002	1.75
0.30	1.60	–31	–	T_{max} =3	–	0.002	1.32
0.35	1.64	–55	–	–	–	0.022	0.99
0.38	1.59	–55	–	–	–	0.019	0.89
0.40	1.54	–59	–	–	–	0.023	0.7
1.00	1.27	–190	–	–	–	–	–

*The values of T_C were determined with various measurement techniques, such as the analysis of the Arrott plots, as well as the ac-susceptibility and electrical resistivity measurements.

4. Concluding remarks

Several samples of the pseudoternary $URh_{1-x}Ru_xGe$ system were synthesized and characterized by X-ray diffraction and EDX analysis. The crystallographic data indicate that the system adopts an orthorhombic TiNiSi-type structure. The lattice parameters seem to deviate from a simple Vegard law around $x = 0.1$. We found that the substitution of Rh by Ru causes vanishing of ferromagnetic order at $x = 0.25$ where the system undergoes a crossover onto a nonmagnetic ground state with short-range

magnetic interactions (x between 0.3 and 0.35). The latter feature differs from that reported by Sakarya et al. [3], who claimed magnetic order even up to $x = 0.325$ (see Fig. 7). As the main result of our study, we found the characteristic features of the non-Fermi liquid state. In summary, we propose a magnetic phase diagram of the $URh_{1-x}Ru_xGe$ system (Fig. 7).

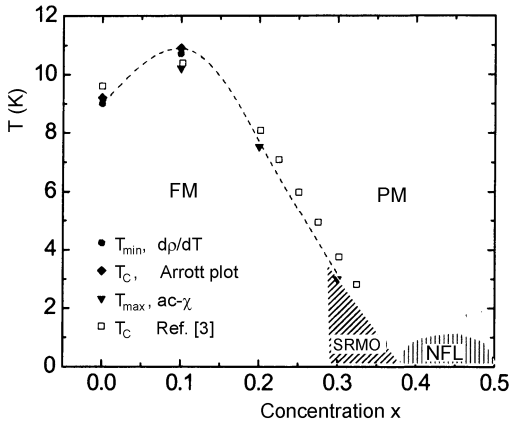

Fig. 7. Tentative magnetic phase diagram of the $URh_{1-x}Ru_xGe$ system. Three different magnetic ground states are shown: FM – ferromagnetic, SRMO – short range magnetic order, and NFL non-Fermi liquid ground state

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