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Specific heat measurements on $U_2(Ru_{1-x}Rh_x)_3Si_5$ ternary silicides

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Abstract

Specific heat (C) measurements have been performed on several compositions of the $U_2(Ru_{1-x}Rh_x)_3Si_5$ system showing an interesting magnetic phase diagram: (i) no magnetic ordering appears above 2 K for $0 \leq x < 0.25$; (ii) on the contrary, two antiferromagnetic ranges exist, respectively, for $0.25 \leq x \leq 0.50$ and $0.50 < x \leq 1.0$. A defined peak seen in $C/T = f(T)$ curve around 8.4(2) and 4.0(5) K for $x = 0.35$ and 0.50, respectively, gives further evidence for a long-range antiferromagnetic order in the concentration range $0.25 \leq x \leq 0.50$. Moreover, for $x = 0.20$ (composition near the non-magnetic→antiferromagnetic transition), a very strong enhancement of C/T is observed below 12 K, reaching a value as high as 255 mJ/U-mol K^2 at 2 K. This behaviour suggests either the occurrence of magnetic ordering below 2 K or that this silicide can be considered as the heaviest electron system. © 1998 Elsevier Science S.A.

Keywords: Uranium; Structural properties; Specific heat

1. Introduction

$U_2Ru_3Si_5$ and $U_2Rh_3Si_5$ ternary silicides which crystallize in the monoclinic $Lu_2Co_3Si_5$-type structure are classified as a non-magnetic ordered Kondo system and an antiferromagnet below $T_N = 26$ K, respectively [1–3]. These behaviours are governed by the strength of the 5f(U)–4d(Ru or Rh) hybridization which decreases with the substitution of Rh for Ru atoms. In this way, the $U_2(Ru_{1-x}Rh_x)_3Si_5$ system exhibits an interesting magnetic phase diagram: two antiferromagnetic phases occur, respectively, for $0.25 \leq x \leq 0.50$ (AF1) and $0.50 < x \leq 1.0$ (AF2) [4]. Neutron diffraction experiments performed on $x = 0.35$ [5] and 1.0 [6]. reveal that the Kondo effect diminishes when x increases since the U-magnetic moment increases from 0.63(16) to 1.75(5) μ_B, respectively, for these two compositions. Also: (i) in $U_2(Ru_{0.65}Rh_{0.35})_3Si_5$ the U-magnetic moments form collinear ferromagnetic 'wavy planes' with an antiferromagnetic coupling between them [5], whereas (ii) in $U_2Rh_3Si_5$ the U-magnetic moments are arranged in a non-collinear antiferromagnetic structure [6]. In other words, the ternary silicides $U_2(Ru_{1-x}Rh_x)_3Si_5$ form an ideal system in order to investigate the competition between the magnetic Ruderman–Kittel–Kasuya–Yosida (RKKY) and non-magnetic Kondo interactions.

The purpose of this paper is to present the structural properties of the $U_2(Ru_{1-x}Rh_x)_3Si_5$ system through specific heat measurements performed on characteristic compositions: $x = 0.20$, 0.35, 0.50 and 0.75. The aim of this study is to characterize the two interesting transitions: non-magnetic→antiferromagnetic (AF1) and antiferromagnetic (AF1)→antiferromagnetic (AF2) occurring, respectively, around $x \approx 0.20$ and 0.50 [4]. We compare, of course, our results with those obtained previously on $U_2Ru_3Si_5$ [2] and $U_2Rh_3Si_5$ [3,7].

2. Experimental details

All samples were prepared by direct melting stoichiometric amounts of the constituents in a cold crucible in an induction furnace in a purified argon atmosphere. The ingots were then annealed in vacuum at 800°C for 1 month.

Structural analyses were performed by means of X-ray powder diffraction using a Guinier camera with Cu Kα radiation. Unit cell parameters were determined by least-squares refinements using an internal 5N silicon standard ($a = 5.43083$ Å).

Magnetization measurements were carried out between 2

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and 300 K using a SQUID magnetometer. The specific heat measurements were performed down to 2 K using a system based on the relaxation-time method [8].

3. Results and discussion

X-ray powder diffraction indicates that all $U_2(Ru_{1-x}Rh_x)_3Si_5$ samples were single phase. All patterns were completely indexed on the basis of a monoclinic cell having the $Lu_2Co_3Si_5$-type structure (space group C2/c) [9]. This structure derives from the tetragonal $ThCr_2Si_2$-type adopted by both URu_2Si_2 and URh_2Si_2 silicides. In these two latter compounds, U-atoms form planes perpendicular to the *c*-axis, whereas in the $U_2(Ru_{1-x}Rh_x)_3Si_5$ silicides, these atoms constitute 'wavy planes' [5]. The composition dependence of the unit cell parameters reveals considerable deviations from Vegard's law (Fig. 1): (i) for $0 \leq x < 0.50$ *a*- and *c*-parameters show a negative deviation from this law, whereas the *b*-parameter exhibits a positive one, and (ii) for $0.50 < x \leq 1.0$ the signs of these deviations are opposite. The value of β-angle increases weakly from 119.3 to 119.5° between $x=0$ and $x=0.45$; β takes a value of 120° for $x=0.50$, and finally keeps a constant value (120.6–120.7°) for $0.50 < x \leq 1.0$. Certainly the structural changes observed near $x=0.50$ (Fig. 1) are associated to the magnetic transition AF1→AF2 evidenced by neutron diffraction experiments [5,6].

T_N temperatures of the $U_2(Ru_{1-x}Rh_x)_3Si_5$ silicides, listed in Table 1, were determined by magnetic susceptibility χ_m measurements in an applied field of 0.01 T. These temperatures correspond to the maxima observed in the $\chi_m = f(T)$ curves.

Figs. 2 and 3 show the thermal dependence of the specific heat, plotted as C/T versus T, for $x=0.20$, 0.35, 0.50 and 0.75.

Table 1
Néel temperature of $U_2(Ru_{1-x}Rh_x)_3Si_5$ ternary silicides

x	T_N (K)		Ref.
	(a)	(b)	
0	-		[1]
0.10	-		—[a]
0.20	-		—[a]
0.25	6.0(5)		—[a]
0.30	9.0(5)		—[a]
0.35	10.0(5)	8.4(2)	—[a]
0.35	9.5(5)		[5]
0.40	9.5(5)		—[a]
0.45	4.0(5)		—[a]
0.50	3.0(5)	4.0(5)	—[a]
0.55	3.0(5)		—[a]
0.60	5.0(5)		—[a]
0.65	8.5(5)		—[a]
0.75	12.0(5)	10.4(5)	—[a]
0.90	21.0(5)		—[a]
1.0	26		[1,2]
1.0	26	25	[7]

(a) and (b), T_N determined, respectively, by magnetization and specific heat measurements.
[a]This work.

The magnetic transition appearing for $x=0.35$ and 0.50 is discerned through a small, but distinct, anomaly which peaks, respectively, at 8.4(2) and 4.0(5) K in agreement with magnetic results (Table 1). In the antiferromagnetically ordered state of $U_2(Ru_{0.65}Rh_{0.35})_3Si_5$ silicide, the data $C/T = f(T)$ below 4 K are consistent with the thermal dependence given by $C/T = \gamma_0 + \beta_0 T^2$ with an extrapolated γ_0 value of 234 mJ/U-mol K^2. This value appears as remarkably enhanced, as is observed in magnetically ordered heavy-fermion systems like $Ce(Ru_{0.4}Rh_{0.6})_2Si_2$ ($\gamma_0 \approx 600$ mJ/Ce-mol K^2) [10] or $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$

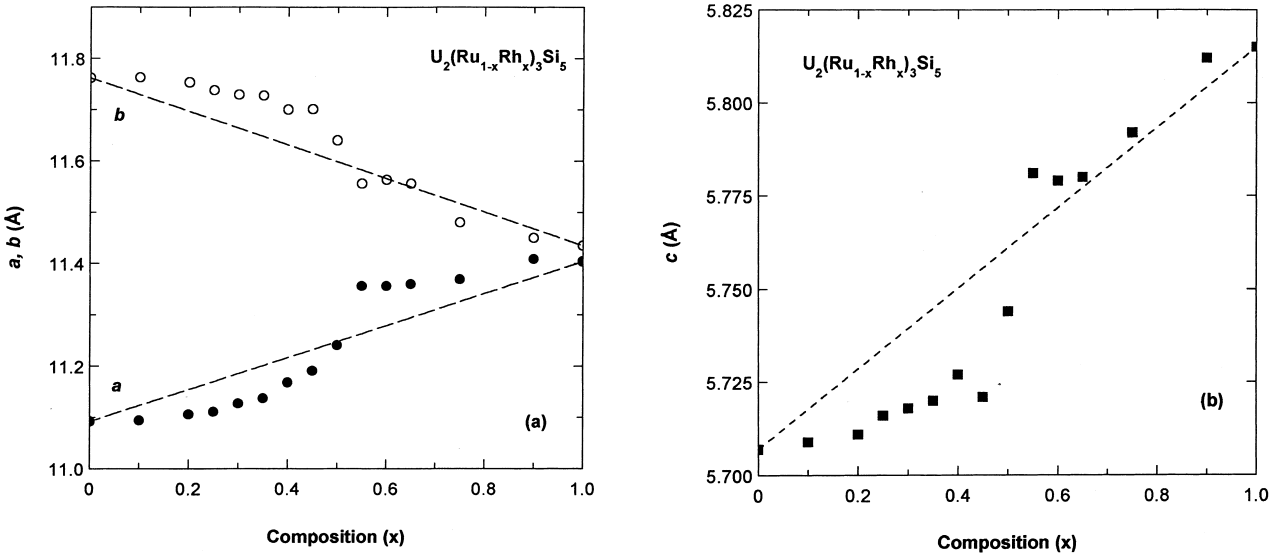

Fig. 1. Dependence of the unit cell parameters of $U_2(Ru_{1-x}Rh_x)_3Si_5$ ternary silicides: (a) *a*- and *b*-parameters; (b) *c*-parameter.

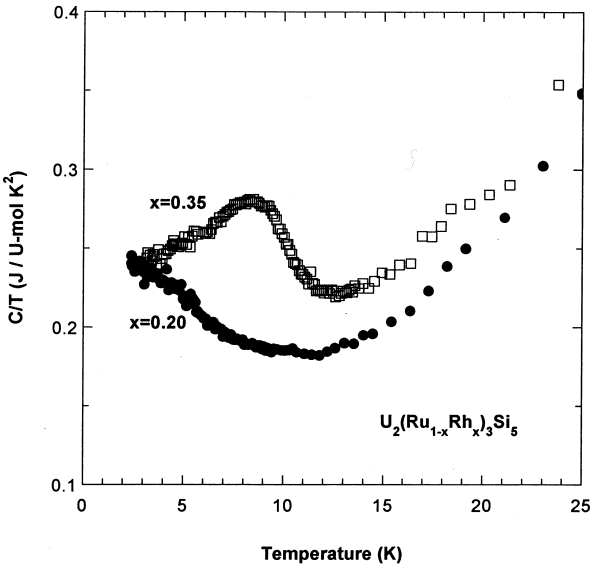

Fig. 2. Dependence of specific heat divided by temperature, C/T, for $U_2(Ru_{1-x}Rh_x)_3Si_5$ with $x=0.2$ and 0.35.

($\gamma_0 \approx 300$ mJ/Ce-mol K^2) [11]. Furthermore, we can note that the relative discontinuity for $U_2(Ru_{0.65}Rh_{0.35})_3Si_5$ at the magnetic transition $\Delta C/T_N$ amounts to 0.11 J/U-mol K^2 which is smaller than that observed for $U_2Rh_3Si_5$ silicide ($\Delta C/T_N \approx 4.5$ J/mol K^2) [7]. This small value points towards a reduced U-magnetic moment in $U_2(Ru_{0.65}Rh_{0.35})_3Si_5$, as indicated by neutron diffraction investigation [5].

For $x=0.75$, we see a distinct jump at 10.4(5) K characteristic of magnetic ordering in the C/T versus T plot (Fig. 3). This result agrees with an increase of the temperature T_N with Rh concentration in the range $0.50 < x \leq 1.0$.

In the system studied, the $U_2(Ru_{0.80}Rh_{0.20})_3Si_5$ compound is close to the crossover between the non-magnetic→antiferromagnetic (AF1) transition. The specific heat data down to 2 K show no sign of magnetic ordering (Fig. 2). Below 12 K, C/T increases with decreasing temperature, reaching a large value of about 255 mJ/U-mol K^2 at 2 K. This C/T value is, respectively, equal to 72 and 22 mJ/U-mol K^2 for $x=0$ [2] and $x=1$ [7]. The behaviour of $U_2(Ru_{0.80}Rh_{0.20})_3Si_5$ can be interpreted as: (i) the beginning of magnetic transition appearing below 2 K; (ii) an enhancement of the effective mass of the quasiparticles, indicating that this silicide can be considered as the heaviest electron system.

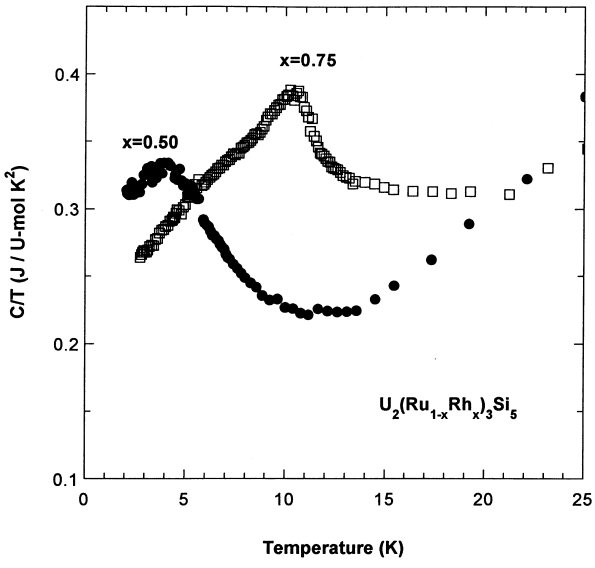

Fig. 3. Dependence of specific heat divided by temperature C/T for $U_2(Ru_{1-x}Rh_x)_3Si_5$ with $x=0.50$ and 0.75.

In conclusion, the present study demonstrates that the replacement of Ru- for the Rh-atoms in $U_2Ru_3Si_5$ leads to an antiferromagnetic order with a strong enhancement of the γ_0 term near the transition from a magnetically non-ordered to a magnetically ordered state. The reduction of the jump ΔC in the specific heat at T_N for $U_2(Ru_{0.65}Rh_{0.35})_3Si_5$, in comparison with that determined for $U_2Rh_3Si_5$, agrees with the existence of two antiferromagnetic phases (AF1 and AF2) in the $U_2(Ru_{1-x}Rh_x)_3Si_5$ system: with increasing x, the 5f(U)-states are more and more localized.

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