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Transport properties of $(U_{1-x}Np_x)Ru_2Si_2$ alloys

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Abstract

Synthesis and resistivity measurements on URu_2Si_2–$NpRu_2Si_2$ solid solutions are reported. $U_{1-x}Np_xRu_2Si_2$ systems, with $x = 0.01$, 0.1, 0.3, 0.5, 0.7 and 0.9, crystallize in the tetragonal $ThCr_2Si_2$-type structure. Resistivity measurements indicate a marked variation of the transport properties with regard to the Np substitution. For low Np dilution (≤ 0.1) considerable changes are observed from the $x = 0$ Cr-like anomaly and a Kondo-like minimum appears below 10 K. In the systems for $0.3 \leq x \leq 0.7$, an increase of T_N is clearly seen and the Cr-like anomaly destroyed in favor of a strong increase of the resistivity suggesting a gap opening. At higher Np-concentration the resistivity is very similar to the pure Np compounds but a large increase in the pure Np-compound 6 K anomaly is observed. © 1997 Elsevier Science S.A.

Keywords: $U_{1-x}Np_xRu_2Si_2$; Resistivity; Kondo systems

1. Introduction

Several uranium-based heavy fermion compounds have attracted large interest due to their unconventional properties. Among these the well-known URu_2Si_2 remains one of the most interesting for which many experimental investigations have been reported [1–4]. In a new attempt to shed light on the anomalous behavior of this compound, a number of investigations have been made of the effects of alloying substitution on the Si, Ru and U sites [5–10]. These last studies were mainly carried out on Ce, La, Y or Th substitutions although in most of them there is no complete range of solid solution.

On the other hand, the isostructural Np compound also exists and displays interesting properties [11]. $NpRu_2Si_2$ shows Kondo behavior and a low-temperature modulated magnetic structure. An antiferromagnetic ordering occurs around 27.5 K and remains incommensurate and modulated down to low temperature, with an ordered moment being approx. 1.5 μ_B and aligned along the c-axis.

It seemed interesting to study the URu_2Si_2–$NpRu_2Si_2$ solid solution systems where a complete range of solubility is observed. Previous studies of this system by Np-Mössbauer spectroscopy have already been reported [12,13]. By investigating the electrical resistivity of $U_{1-x}Np_xRu_2Si_2$ with $x = 0.01$, 0.1, 0.3, 0.5, 0.7 and 0.9, we intend here to contribute to the characterization of their physical properties.

2. Experimental details

Large batches of URu_2Si_2 and $NpRu_2Si_2$ were prepared by arc melting together metals of high purity. Samples obtained were characterized by X-ray diffraction. $U_{1-x}Np_xRu_2Si_2$ solid solutions, with $x = 0.01$, 0.1, 0.3, 0.5, 0.7 and 0.9 were then prepared by arc melting stoichiometric amounts of U and Np compounds. Samples obtained were characterized by X-ray diffraction, metallography and microprobe analysis to verify their homogeneity and the absence of U, Np clusters. No further thermal treatment was found to be necessary.

Bulk samples were then encapsulated for electrical resistivity measurements. Measurements were performed down to 1.4 K with a conventional four-probe

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AC method described elsewhere [14]. Because of dimensional uncertainties, probable presence of microcracks and preferential orientation of the crystallites in the bulk samples, the absolute resistivity values have not been considered here.

3. Results and discussion

All samples were found to be single phase and homogeneous. A $ThCr_2Si_2$-type structure was observed in the whole range of concentration with the lattice parameters listed in Table 1.

The Figs. 1–3 show the resistivity ratio $\rho/\rho_{300\ K}$ curves obtained for each system. Characteristic temperatures are listed in Table 2. From these curves, it clearly appears that a discussion should be divided according to the Np content.

3.1. Low range of Np content, $0 \leq x \leq 0.1$

The resistivity curve obtained for our pure URu_2Si_2 compound is in very good agreement with previous reported data [15]. We may distinguish four aspects: the high temperature Kondo-like negative temperature coefficient, the marked fall in resistivity below a rounded maximum at around 70 K, the Cr-like anomaly associated with the gapped antiferromagnetic ordering and the lowest temperature range.

We observe that the doping up to 10% Np has a little effect on the high-temperature Kondo-like effect and the slope of the curves remains almost unchanged. On the other hand a slight shift of the resistivity maximum is observed towards higher temperatures for 1% Np and lower temperatures for 10% Np (see Table 2 and Fig. 1a). Moreover a marked effect is seen on the slope of the fall in resistivity for 10% Np only. This change of slope may be correlated with a perturbation of the short range correlation preceding the magnetic ordering.

The Cr-like anomaly marked by a minimum (normally associated with the ordering temperature) and a maximum in resistivity is much more sensitive to the substitution effects (Fig. 1b). In $U_{0.99}Np_{0.01}Ru_2Si_2$ a

Table 1
Lattice paramters of $U_{1-x}Np_xRu_2Si_2$ compounds

Compound	a (pm)	c (pm)
URu_2Si_2	413.1(1)	957.4(1)
$U_{0.99}Np_{0.01}Ru_2Si_2$	413.2(1)	957.6(1)
$U_{0.9}Np_{0.1}Ru_2Si_2$	413.1(1)	957.5(1)
$U_{0.7}Np_{0.3}Ru_2Si_2$	413.2(1)	957.8(1)
$U_{0.5}Np_{0.5}Ru_2Si_2$	413.4(1)	958.3(1)
$U_{0.3}Np_{0.7}Ru_2Si_2$	413.6(1)	958.4(1)
$U_{0.1}Np_{0.9}Ru_2Si_2$	413.5(1)	958.8(1)
$NpRu_2Si_2$	413.7(1)	959.3(1)

Table 2
Characteristics temperatures of the anolamies observed in the resistivity of $U_{1-x}Np_xRu_2Si_2$ compounds

Compound	T_N [Ref.]	$T\rho_{max}$	$T_{anomaly}$		
			T_1	T_2	T_3
URu_2Si_2	17.5 K [15]	70 K	17.3 K	16.5 K	—
$U_{0.99}Np_{0.01}Ru_2Si_2$	—	75 K	17.0 K	16.0 K	8.0 K
$U_{0.9}Np_{0.1}Ru_2Si_2$	19 K [12,13]	60 K	18.7 K	14.7 K	9.3 K
$U_{0.7}Np_{0.3}Ru_2Si_2$	22 K [12,13]	40 K	24.5 K	—	—
$U_{0.5}Np_{0.5}Ru_2Si_2$	25 K [12,13]	—	25.3 K	—	—
$U_{0.3}Np_{0.7}Ru_2Si_2$	25 K [12,13]	—	25.5 K	—	—
$U_{0.1}Np_{0.9}Ru_2Si_2$	25 K [12,13]	—	26.7 K	24.0 K	14.0 K
$NpRu_2Si_2$	27.5 K [11–13]	—	27.5 K	26.0 K	6.0 K

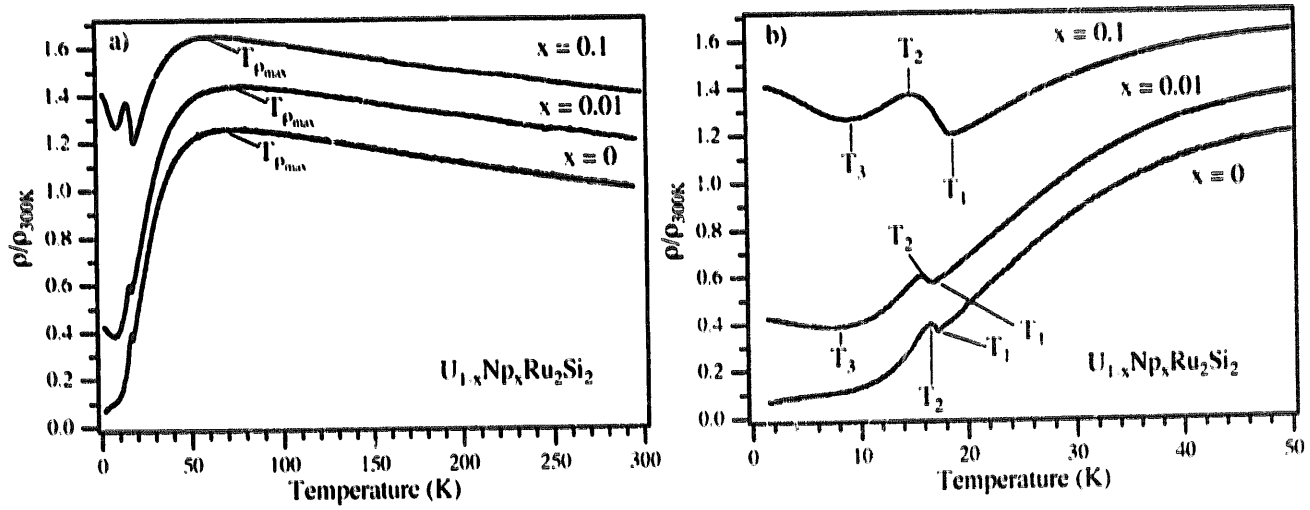

Fig. 1. (a) Normalized electrical resistivity $\rho/\rho_{300\ K}$ vs. temperature for $U_{1-x}Np_xRu_2Si_2$ compounds with $x = 0$, 0.01 and 0.1; (b) enlarged figure for the low temperature. In both figures the curves are displaced for clarity.

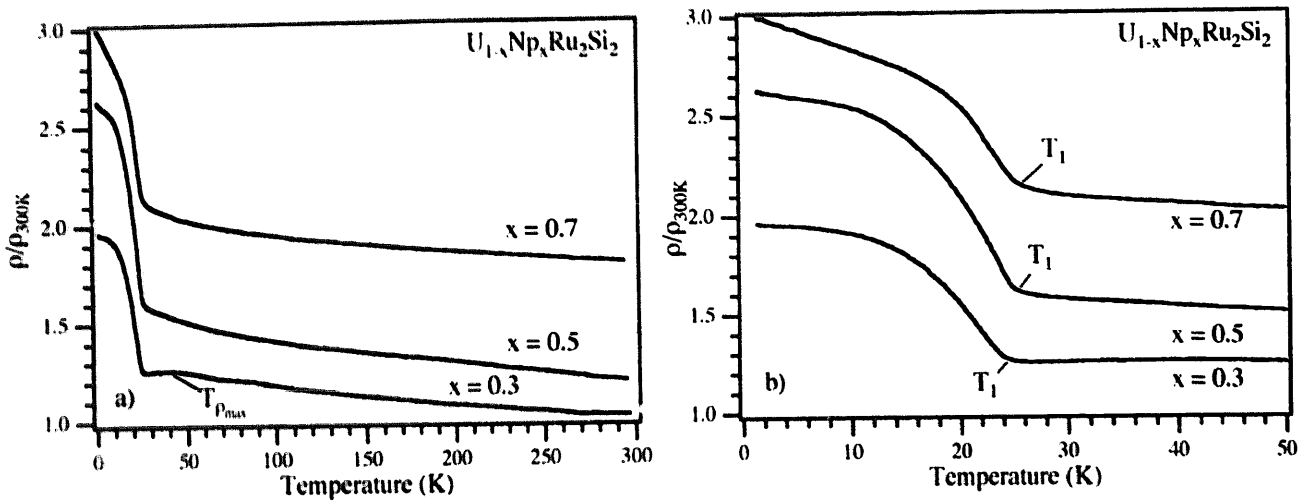

Fig. 2. (a) Normalized electrical resistivity $\rho/\rho_{300\,K}$ vs. temperature for $U_{1-x}Np_xRu_2Si_2$ compounds with $x = 0.3$, 0.5 and 0.7; (b) enlarged figure for the low temperature. In both figures the curves are displaced for clarity.

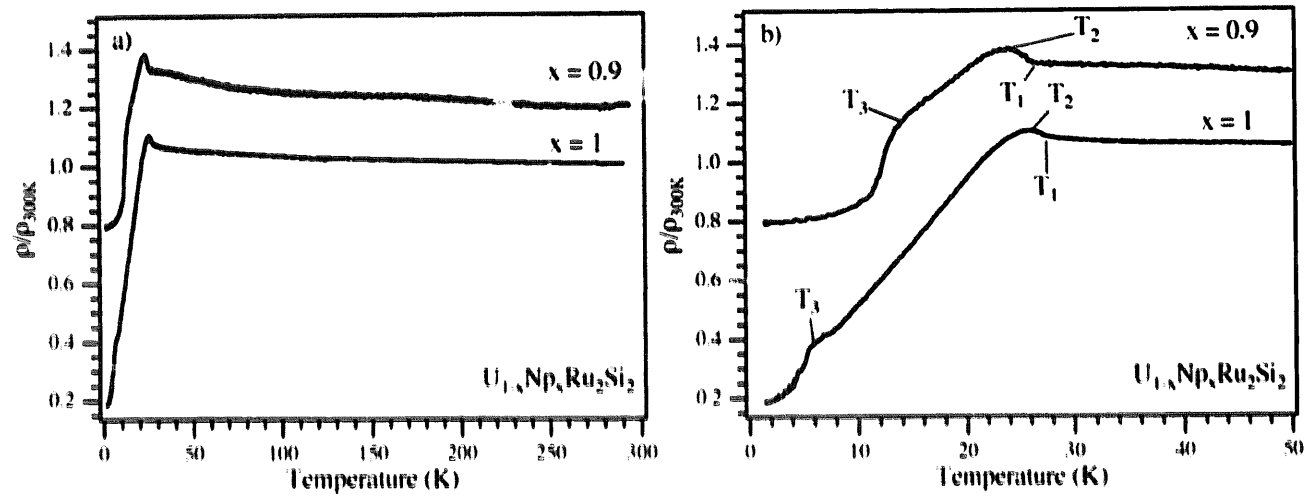

Fig. 3. (a) Normalized electrical resistivity $\rho/\rho_{300\,K}$ vs. temperature for $U_{1-x}Np_xRu_2Si_2$ compounds with $x = 0.9$ and 1; (b) enlarged figure for the low temperature. In both figures the curves are displaced for clarity.

slight decrease and broadening of the ordering temperature (Fig. 1b and Table 2) are observed. This may be understood as a negative chemical pressure effect as the substitution by the Np has the tendency to increase the volume of the cell. In $U_{0.9}Np_{0.1}Ru_2Si_2$ the ordering temperature is shifted to higher temperature and a pronounced increase of the 'size' of the anomaly is clearly seen. This observation may be ascribed to an enhancement of impurity scattering by the loss of Fermi surface, in the same way as observed by substitution with light rare-earths [10].

In both compounds, the slope of the resistivity decreasing after the ordering temperature is less pronounced, which suggests qualitatively a decrease of the energy gap opened over the Fermi surface. Finally, a third anomaly is evident at the lowest temperatures, where the resistivity reaches a minimum around 8 K (Fig. 1b and Table 2). Similar behavior was observed for Th and heavy rare-earths substitution [7,10] and understood as reminiscent of the Kondo resistance minimum that occurs in metals containing small amounts of magnetic impurities. As shown by Mössbauer experiments [12,13], the Np in these alloys carries a magnetic moment and may be considered as 'magnetic impurity' in a nearly non-magnetic surrounding, the moment on the U being very small.

3.2. Middle range of Np content, $0.3 \leq x \leq 0.7$

For the middle range of Np-content (30–70%), the resistivity curves obtained are completely different (Fig. 2). $U_{0.7}Np_{0.3}Ru_2Si_2$ shows a rounded maximum around 40 K while for higher concentration (50%, 70%), a logarithmic linear variation of the resistivity

is observed down to the ordering temperature. The ordering temperature was assigned to the first kink of the resistivity (Fig. 2b) and is in very good agreement with that determined by Mössbauer (see Table 2).

For all systems, we observe, below the ordering temperature, a strong increase of the resistivity with a slope proportional qualitatively to the 'impurity' concentration (U or Np regarding the main component) and maximum for the 50% alloy. No Cr-like anomaly is present.

On the basis of the resistivity alone, an interpretation of this phenomenon is difficult. We may understand this increase of the resistivity as the opening of a gap leading to semimetal type behavior. This could be explained by the opening of a hybridization gap, leading to a decrease of the number of carriers or of their mobility. As we know from Mössbauer spectroscopy that the Np atoms carry moment aligned to the *c*-axis and are likely to show an antiferromagnetic type ordering [13], another hypothesis would be that a change of the magnetic structure periodicity, compared to the crystallographic lattice, leads to the opening of a gap in the band structure (Peierls distortion).

3.3. High range of Np content, $0.9 \leq x \leq 1$

The resistivity curves obtained for $U_{0.1}Np_{0.9}Ru_2Si_2$ and $NpRu_2Si_2$ are shown in Fig. 3. The behavior of the pure Np compound is in very good agreement with previous results [11]. In the paramagnetic range, ρ exhibits a logarithmic T-variation typical of a Kondo effect. The ordering temperature is assigned at 27.5 K, where an anomaly in the curve is seen. In the ordered state, the decrease of the resistivity is understood as a huge magnetic contribution of an energy gap antiferromagnet. In this way the properties of this compound are somehow similar to its pure uranium homologue. At low temperatures (around 6 K) a second transition is observed. This was correlated with a squaring of the modulation of the magnetic structure but no evidence of this could be detected in the neutron diffraction experiment [11].

Doping with 10% of uranium does not greatly modify the high temperature properties of the system. A slight decrease of the ordering temperature and a broadening of the associated anomaly are nevertheless observed and may be explained as for the Np doping in U (Section 3.1). However, a noticeable increase in temperature (to 14 K) of the anomaly associated with the second transition may provide an opportunity of verifying the nature of this transition in the pure Np-compound.

4. Conclusions

URu_2Si_2–$NpRu_2Si_2$ is an interesting system where solid solutions are formed in the whole range of concentration. Moreover, substitution by Np adds a new dimension to doping effects as it is in a trivalent valence state, it contributes to an increase of the total number of 5*f* electrons and carries a magnetic moment. For low Np content ($\leq 10\%$) an increase of the Cr-like anomaly as well as a Kondo type behavior at low temperatures (around 8–9 K) are observed. This is a combination of previous effects separately observed in Th or heavy rare earths and in light rare earth substituted systems. For equivalent U–Np content (30–70% Np) a semi-conductor type behavior is proposed. Finally, for high Np-content ($\geq 90\%$ or U-doped Np-compound) the evidence of a strong increase to 14 K of the $NpRu_2Si_2$'s 6 K anomaly may allow a better understanding of its nature. Complementary studies of these systems, especially by neutron diffraction, are needed.

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