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Journal of Physics: Conference Series **273** (2011) 012024 doi:10.1088/1742-6596/273/1/012024

Crystal structure and physical properties of $NpRh_2Sn$, a new Np-based ternary compound

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Abstract. We report on the synthesis and physical characterization of a new compound, $NpRh_2Sn$, prepared by arc-melting under argon atmosphere. Rietveld analysis of the powder x-ray diffraction pattern reveals an orthorhombic, Fe_3C-type structure, with lattice parameters that are the shortest among the AnT_2Sn series (*An* = U, Np, Pu, and T = Pd, Rh). A fit of the high-temperature magnetic susceptibility curve, $\chi(T)$, gives a Curie-Weiss temperature Θ_{CW} = -29K, and an effective magnetic moment $\mu_{eff} = 2.42\mu_B$. The stabilization of the antiferromagnetic order below T_N = 34 K is revealed by a cusp in $\chi(T)$ and by a small anomaly in the specific heat curve, $C_p(T)$. An enhanced value of the Sommerfeld coefficient, $\gamma \approx 107$ mJ mol^{-1} K^{-2}, indicates a moderate heavy-fermion state. $NpRh_2Sn$ is the first member of the $AnRh_2Sn$ family, and a rare representative of heavy-fermion systems amidst Np- intermetallics.

1. Introduction

The large family of the ternary actinide compounds AnT_2M, where An is an actinide element, T is a transition element, and M is a metalloid, forms mainly in the orthorhombic Fe_3C - type crystal structure (Pnma, s.g. 62). The uranium based alloys (UT_2M) have attracted considerable attention due to a wide range of physical properties, which originates from the sensitive nature of the uranium 5*f* - electrons. Very few compounds with An other than uranium have been reported, and most of them contain palladium as the transition element.

Here, we present the crystal structure and physical properties of a new intermetallic $NpRh_2Sn$ compound, which is the first member of the $AnRh_2M$ family.

2. Experimental

Polycrystalline $NpRh_2Sn$ sample was synthesized by arc-melting stoichiometric amounts of elements under a zirconium gettered ultra pure argon atmosphere. The purity of the product was verified by powder X-ray diffraction (Bruker D8 Focus, CuK_α radiation, graphite monochromator). Data were collected in the 2θ range of 10° – 100° with a step size of 0.01°. The FullProf Suite package was used for Rietveld structure refinement [1]. The magnetic susceptibility was measured using a Magnetic Property Measurement System (MPMS, Quantum Design), whereas the specific heat and electrical

resistivity were determined using a Physical Properties Measurement System (PPMS, Quantum Design).

3. Results and discussion

The as-cast sample extracted from the batch was studied by x-ray powder diffraction and the result is presented in Figure 1. The known crystal structure of URh_2Sn was employed as a starting structural model [2]. As shown in Fig. 1, there is good agreement between the model and the data, suggesting that $NpRh_2Sn$ is isostructural to the U analogue. The lattice parameters for $NpRh_2Sn$ were calculated to be a=9.7208(6) Å, b=4.4221(3) Å and c=6.9043(4) Å and are smaller than the reported ones for $NpPd_2Sn$ [3]. It was found that 8% of Np substitutes the Rh(1) site and 8% of Rh are located on the Np site.

Selected physical parameters for $NpRh_2Sn$ and other members of the AnT_2M series are given in Table 1.

Table 1. Lattice parameters, An-An distance (d_{An-An}) , Néel temperature (T_N) and Curie-Weiss temperature (Θ_{CW}) for 5 members of AnT_2Sn family

	a (Å)	b (Å)	c (Å)	d_{An-An} (Å)	T_N (K)	Θ_{CW} (K)
$NpRh_2Sn$	9.7208(6)	4.4221(3)	6.9043(4)	3.9702	34	-29
$NpPd_2Sn$ [3]	10.004(3)	4.535(2)	6.961(1)	4.0606	15	-80
$PuPd_2Sn$ [4]	10.053(9)	4.502(4)	7.065(6)	---	11	-30
URh_2Sn [2,5]	9.7923(2)	4.37474(7)	6.9639(1)	3.935	---	-200
UPd_2Sn [6]	9.9787(1)	4.58843(5)	6.89166(8)	4.1817	---	-77

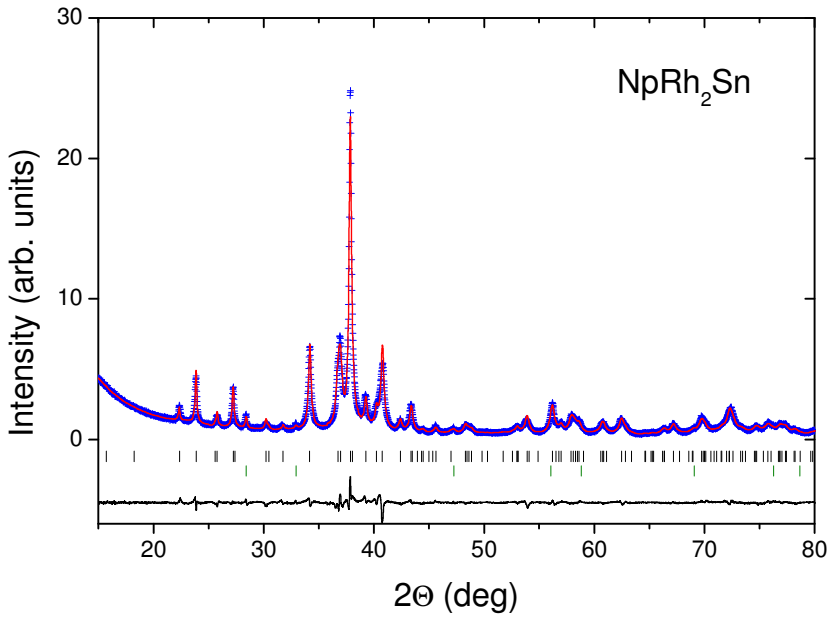

Figure 1. Rietveld refinement of room temperature the x-ray diffraction pattern for $NpRh_2Sn$. Crosses represent observed data, red solid line is the calculated intensity. The black tick marks correspond to $NpRh_2Sn$ and green set refers to the NpO_2 impurity (~2% wt.). Figure of merits: $R_p = 4.04$, $R_{wp} = 5.97$, $R_{exp} = 2.60$ and $\chi^2 = 5.28$.

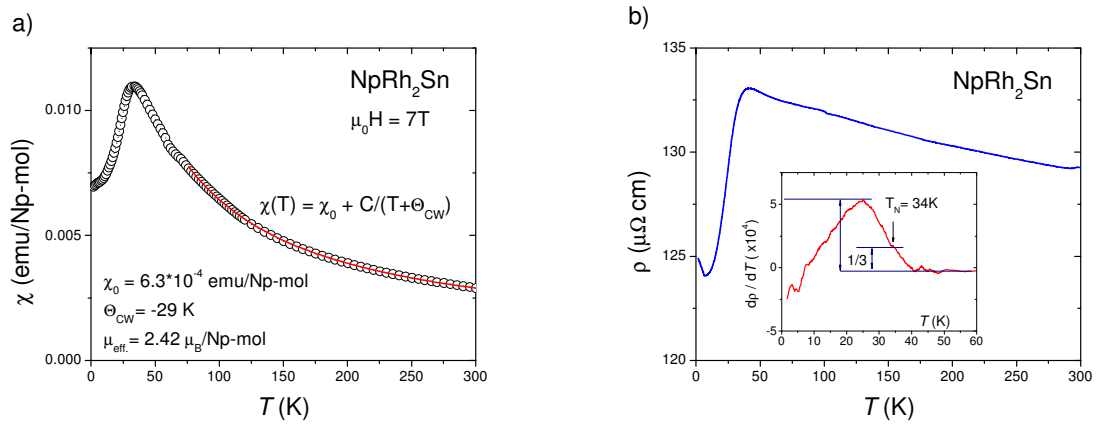

Figure 2. The temperature dependence of the a) magnetic susceptibility (χ(T)) and b) electrical resistivity (ρ(T)) of $NpRh_2Sn$. The solid red line in panel a) is a fit by the modified Curie-Weiss law. The inset in panel b) shows the derivative of the electrical resistivity (dρ/dT) vs. temperature. The Néel temperature, defined as shown in the inset, is 34K.

In Figure 2 we show the temperature dependence of magnetic susceptibility (a) and electrical resistivity (b) of $NpRh_2Sn$. The Curie-Weiss fit of the χ(T) data above 75 K (red solid line) gives a Curie-Weiss temperature Θ_{CW} = -29 K, and an effective magnetic moment μ_{eff} = 2.42 μ_B/Np-mol. This is close to the value expected for Np^{+3} (2.68 μ_B). The measurement indicates an antiferromagnetic anomaly at T_N around 35 K.

Electrical resistivity as a function of temperature (main panel) and the temperature derivative of the resistivity (inset) for $NpRh_2Sn$ are plotted in Figure 2b. At room temperature the resistivity is 129 μΩ cm which is very close to what is observed for $PuPd_2Sn$ [4] and $NpPd_2Sn$ [3]. With decreasing temperature the electrical resistivity slightly increases and at 50 K reaches a value of 133 μΩ cm. Interestingly the non-metallic behavior in the high temperature region was also reported for $NpPd_2Sn$ whereas $PuPd_2Sn$ exhibit metallicity in the whole temperature range. Below 40 K a rapid drop of resistivity, and a pronounced positive peak of dρ/d*T*, are visible. This behavior is caused by the antiferromagnetic transition. Using the derivative of the electrical resistivity we can determine the ordering temperature (1/3 of the dρ/d*T* peak). We found T_N = 34 K, which is very close to the magnetic ordering temperature.

The temperature dependence of the specific heat (C_p) and specific heat over temperature (C_p/T) of $NpRh_2Sn$ are shown in Figure 3. At room temperature C_p reaches a value that corresponds to the Dulong-Petit law: C_p=3nR=99.77 J mol^{-1} K^{-1}, where n=4 and R=8.314 J mol^{-1} K^{-1}. The magnetic phase transition discussed above is not well pronounced in the specific heat measurement. Close to T=30K only a slight change of slope, as shown in the inset of Fig. 3a, is observed on the specific heat curve C_p(T). Similar result, notably a marginal evidence of a specific heat anomaly was reported also for $UPd_{2-x}Sn$ (x=0.05 and 0.15), as discussed in ref. [7].

Figure 3b shows C_p/T versus T^2 (only low temperature region) where the Debye temperature (Θ_D) and Sommerfeld parameter (γ) can be estimated from the linear fit: $C_p/T = C_{el}/T + C_{ph}/T = \gamma + \beta T^2$, where $\beta = \frac{12\pi^4}{5}\frac{nR}{\Theta_D^3}$. Although the experimental data were fitted only up to 10K, which is three times lower than T_N, we are aware of the magnetic contribution (C_{mag}) to the specific heat. Subtracting C_{mag} should not change the value of γ, but might slightly change β, and consequently the Debye temperature. The calculated Sommerfeld parameter for $NpRh_2Sn$, γ = 107 mJ mol^{-1} K^{-2} indicates

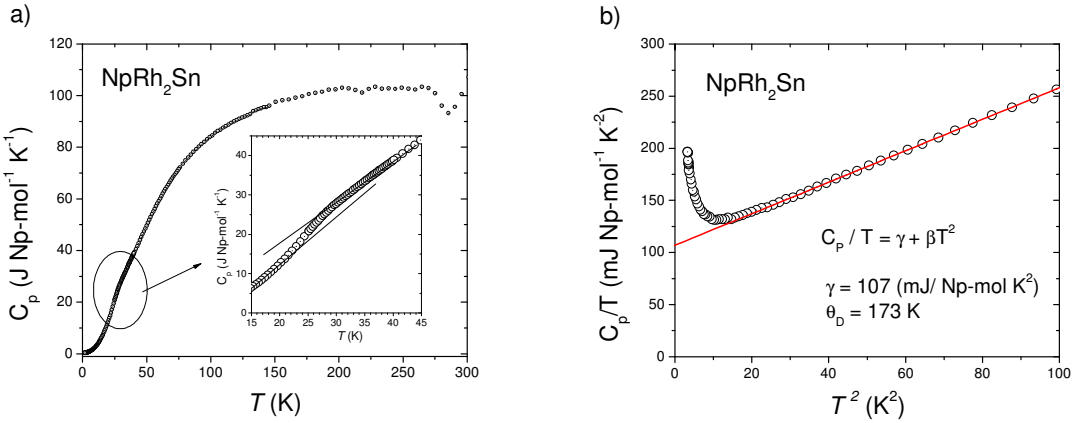

Figure 3. Temperature dependent specific heat (a) and specific heat over temperature (b) of $NpRh_2Sn$. The inset of the figure a) emphasises the magnetic transition region.

moderately heavy fermion state, and is of the same order of magnitude as reported for $PuPd_2Sn$ and $NpPd_2Sn$, which are 180 mJ mol^{-1} K^{-2}, and 400 mJ mol^{-1} K^{-2} respectively.

To summarize, we have synthesized and studied a new Np-based ternary $NpRh_2Sn$ compound. Its properties are similar to those of $NpPd_2Sn$, although a shorter Np-Np distance causes an increase of the Néel temperature. $NpRh_2Sn$ is a rare representative of heavy-fermion systems amidst Np-intermetallics.

Acknowledgements
Np metal required for the fabrication of the compound was made available through a loan agreement between Lawrence Livermore National Laboratory and ITU, in the frame of a collaboration involving Lawrence Livermore National Laboratory, Los Alamos National Laboratory, and the US Department of Energy. T.K. acknowledges the European Commission for financial support.

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