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Magnetic Properties of NpPdSn

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A new compound NpPdSn was prepared and studied by X-ray diffraction, magnetization, heat capacity and electrical resistivity measurements, performed in the temperature range 2–300 K and under magnetic field up to 14 T. The crystal structure determined by single-crystal X-ray analysis is hexagonal with ZrNiAl-type (space group $P\bar{6}2m$). NpPdSn orders antiferromagnetically at 19 K and exhibits a Curie–Weiss behavior with $\mu_{\text{eff}} = 2.66 \ \mu_{\text{B}}$ and $\Theta_{\text{p}} = -47$ K. Bulk properties show temperature variations similar to systems with strong electronic correlations with a large negative paramagnetic Curie temperature and an enhanced low-temperature specific heat ($\gamma \approx 90 \text{ mJ/(mol K}^2)$). It suggests that NpPdSn may be classified as a new Np-based antiferromagnetic Kondo lattice, one of the very few known amidst transuranium-based intermetallics.

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1. Introduction

Uranium-based compounds with the UTX composition, where T is a d-electron transition metal and X stands for a *p*-electron element, have attracted much attention for their large variety of physical behaviors, due to the hybridization of uranium 5f electrons with s, p and/or d electrons of neighboring atoms. In these ternaries itinerant or localized magnetic moment, complex magnetic structures and/or semiconducting or semimetallic-like electrical conductivity have been reported. Moreover, several UTX compounds also show features characteristic of heavy fermion systems. Such a behavior was reported, for example, for magnetically ordered Kondo lattices such as UPdGa (two subsequent antiferromagnetic-like transitions at 30 and 62 K [1]), UPdSi (antiferromagnetic-like transitions at 27 and 33 K [2]), UPdGe (antiferromagnetic transition at 50 K and a ferromagnetic one at 28 K [3, 4]) and UPdIn (antiferromagnetic ordering below $T_{\rm N} = 20$ K with a ferromagnetic component below 7 K [5]). The uranium stannite, UPdSn, exhibits well localized 5f electrons with a small linear specific heat coefficient ($\gamma \approx$ $5 \text{ mJ}/(\text{mol } \text{K}^2))$ [6]. This compound exhibits an antiferromagnetic phase transition at 37 K leading to a magnetic structure with orthorhombic symmetry. A second transition occurs at 25 K, and the magnetic structure becomes of monoclinic symmetry [7]. The ordered magnetic moment on the U site is large $(\mu_{\rm ord} \approx 2 \ \mu_{\rm B})$ [8].

In the course of our systematic studies on the magnetic, transport and thermodynamic properties of transuranium-based $AnPd_2Sn$ intermetallics [9–11], we

have recently focused our attention on the AnPdSn materials. In this paper we report for the first time about crystal structure, magnetic and specific heat properties of NpPdSn.

2. Experimental details

A polycrystalline sample of NpPdSn was prepared by arc melting the stoichiometric amounts of the elements in a Zr-gettered pure argon atmosphere. The sample was examined by X-ray single crystal and powder diffraction methods. Small single crystals were isolated from the



Fig. 1. Crystal structure of NpPdSn.

crushed ingot and used for the data collection on a four--circle X-ray diffractometer. The refined crystal structure was shown to be hexagonal with ZrNiAl-type (space

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group $P\overline{6}2m$) and with lattice parameters a = 7.5076 Å and c = 4.0954 Å. A sketch of the crystal structure of NpPdSn is shown in Fig. 1.

Magnetization measurements were carried out in the temperature range 2–300 K and in magnetic fields up to 7 T using a SQUID magnetometer (Quantum Design MPMS-7). The heat capacity (relaxation method) was measured from 2 to 300 K employing a Quantum Design PPMS-14 system. Due to the radiotoxicity of neptunium, all operations of preparation and encapsulation have been achieved in glove boxes under inert atmosphere (N₂). Moreover, all experimental studies were made using special encapsulation systems due to the contamination risks.

3. Results and discussion

The temperature dependence of the inverse magnetic susceptibility of NpPdSn measured in an applied magnetic field of 7 T is shown in Fig. 2. At low temperature a distinct minimum in the $\chi^{-1}(T)$ curve manifests the onset of antiferromagnetic ordering below the Néel temperature $T_{\rm N} = 19$ K.



Fig. 2. Temperature dependence of the inverse magnetic susceptibility of NpPdSn measured in a magnetic field $\mu_0 H = 7$ T. The solid line is a Curie–Weiss fit. Inset: magnetization versus magnetic field taken at 10 and 25 K.

Above the magnetic transition, the inverse magnetic susceptibility of NpPdSn follows a Curie–Weiss law:

$$\chi(T) = \frac{N\mu_{\text{eff}}^2}{3k_{\text{B}}(T - \Theta_{\text{p}})} \tag{1}$$

with the effective magnetic moment $\mu_{\rm eff} = 2.66 \ \mu_{\rm B}$ and the paramagnetic Curie temperature $\Theta_{\rm p} = -47$ K. The experimental value of $\mu_{\rm eff}$ is similar to the free Np³⁺ ion value expected for the Russell–Saunders coupling (2.68 $\mu_{\rm B}$). It indicates the presence of a well localized magnetic moment on the Np ions. The absolute value of $\Theta_{\rm p}$ is much larger compared to the Néel temperature as usually found in systems with strong Kondo interactions. As shown in the inset of Fig. 2, the magnetization measured in the ordered state is proportional to the applied magnetic field with no sign of any hysteresis effect or metamagnetic-like transition up to 7 T.



Fig. 3. Temperature variation of the specific heat of NpPdSn. The inset shows the low-temperature data in the form C_p/T vs. T^2 . The solid lines mark a straight line behavior.

Figure 3 shows the temperature dependence of the specific heat of NpPdSn. At room temperature, C_p approaches a value expected from the Dulong–Petit law, i.e. $C_p = 3nR = 74.8 \text{ J/(mol K)}$, where n is the number of atoms per molecule (in the case of NpPdSn n = 3) and R is the gas constant. The magnetic phase transition at $T_{\rm N} = 19$ K manifests itself as a small λ -type anomaly in $C_p(T)$. This peak is hardly affected by an applied magnetic field. At 14 T, the maximum in C_p/T shifts only faintly towards lower temperatures and insignificantly decreases in magnitude. The observed behavior is very similar to that found before in NpPd₂Sn [9]. The low temperature electronic contribution to the specific heat of NpPdSn is strongly enhanced. The C_p/T ratio extrapolated to T = 0 K from the regions above and below $T_{\rm N}$ is as large as 380 mJ/(mol $\rm K^{-2})$ and 90 mJ/(mol $\rm K^{-2}),$ respectively (see inset in Fig. 3). These values are comparable with the Sommerfeld coefficients reported for UPdSb and UPdIn, which were 62 mJ/(mol K^{-2}) [12] and 280 mJ/(mol K^{-2}) [13], respectively.

4. Summary

The new Np-based ternary compound NpPdSn was synthesized and studied by X-ray diffraction, magnetization and heat capacity measurements, performed in the temperature range 2–300 K and in magnetic field up to 14 T. The compound crystallizes with hexagonal structure of ZrNiAl-type (e.g. $P\overline{6}2m$) with the lattice parameters: a = 7.5076 Å and c = 4.0954 Å. The magnetic measurements revealed that NpPdSn orders antiferromagnetically at 19 K. The enhanced low-temperature linear specific heat coefficient points to the presence of heavy--quasiparticles in the antiferromagnetic state. It suggests that the compound may be classified as a new Np-based antiferromagnetic Kondo lattice, one of the very few known amidst transuranium-based intermetallics. However, before drawing any firm conclusion about the electronic ground state in NpPdSn future experimental studies are required. Some of them are presently underway.

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