Journal of Nuclear Materials 385 (2009) 46–48

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Magnetic and related properties of PuPdSn

K. Gofryk *, J.-C. Griveau, E. Colineau, R. Jardin, J. Rebizant, F. Wastin, R. Caciuffo

European Commission, Joint Research Centre, Institute for Transuranium Elements, Postfach 2340, 76125 Karlsruhe, Germany

A B S T R A C T

A new phase PuPdSn was prepared and studied by X-ray diffraction, magnetization and heat capacity measurements, performed in the temperature range 2–300 K and in magnetic fields up to 14 *T*. The crystal structure determined from single-crystal X-ray data is the hexagonal ZrNiAl-type [space group $P\bar{6}2m$] with lattice parameters: $a = 7.5057$ Å and $c = 4.0853$ Å. PuPdSn orders antiferromagnetically at $T_N = 21$ K. Moreover, another antiferromagnetic-like transition takes place at 9.6 K. Above T_N the susceptibility follows a modified Curie–Weiss law with $\mu_{eff} = 1.0\ \mu_B$, $\Theta_p = -14$ K and $\chi_0 = 2.1 \times 10^{-4}$ emu/mol. The low-temperature linear specific heat coefficient is small ($\gamma \sim 8$ mJ/mol K^2) pointing to well localized 5*f* electrons.

1. Introduction

During the last years the uranium-based compounds with the composition UPd*X*, where *X* stands for a *p*-electron element, have attracted much attention for their various and intriguing physical behaviour driven by the hybridization of 5*f*-electronic states with *s,p,d*-states of neighboring atoms. Previous studies showed that UPdSn has well localized 5*f* electrons with a small linear specific heat coefficient ($\gamma \sim 5$ mJ/mol K^2) [1]. This compound exhibits an antiferromagnetic phase transition at 37 K leading to a magnetic structure with an orthorhombic symmetry. A second transition occurs at 25 K, and the magnetic structure becomes monoclinic [2]. The ordered magnetic moment on the U site is large ($\mu_{ord} \sim 2\ \mu_B$) [3].

In the course of our systematic studies on the magnetic, electrical and thermal behaviour of transuranium-based *An*Pd$_2$Sn intermetallics [4–6], we have recently focused our attention on the related *An*PdSn materials. Very recently, we have briefly reported the properties of NpPdSn which orders antiferromagnetically below 19 K and shows some features of strong electronic correlations at low temperatures [7]. In this paper we communicate for the first time on the synthesis, structure, magnetic and heat capacity studies of another member of the *An*PdSn family, namely PuPdSn.

2. Experimental details

A polycrystalline sample was prepared by arc-melting stoichiometric amounts of the constituents, and examined by X-ray single crystal and powder diffraction. From the inner part of the specimen small single crystals were selected and examined on a CAD-4 diffractometer.

The crystal structure was refined and shown to be hexagonal of the ZrNiAl-type (space group $P\bar{6}2m$), with lattice parameters $a = 7.5057$ Å and $c = 4.0853$ Å. A sketch of the crystal structure of PuPdSn is given in Fig. 1.

Magnetic measurements were carried out in the temperature range 2–300 K and in magnetic fields up to 7 *T* using a Quantum Design MPMS-7 SQUID magnetometer. The heat capacity was measured from 2 to 300 K by relaxation method employing a Quantum Design PPMS-14 setup. The measurements of the physical properties of PuPdSn (even using very small samples) were limited to temperatures above 2 K due to self-heating effect arising from the radioactive decay of the ^{239}Pu isotope. Moreover, due to contamination risks generated by radiotoxicity of plutonium, all physical measurements were done using special encapsulation systems and all operations of preparation and encapsulation have been achieved in glove boxes under inert pure nitrogen atmosphere.

3. Results and discussion

3.1. Magnetic properties

The temperature dependence of the inverse magnetic susceptibility of PuPdSn measured in an applied magnetic field of 5 *T* is shown in Fig. 2. At low temperatures a distinct minimum in the $\chi^{-1}(T)$ curve manifests the onset of antiferromagnetic ordering below the Néel temperature $T_N = 21$ K. Moreover, another antiferromagnetic-like transition takes place at 9.6 K.

* Corresponding author. Tel.: +49 7247951428.
E-mail address: krzysztof.gofryk@ec.europa.eu (K. Gofryk).

0022-3115/$ - see front matter © 2008 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnucmat.2008.09.049

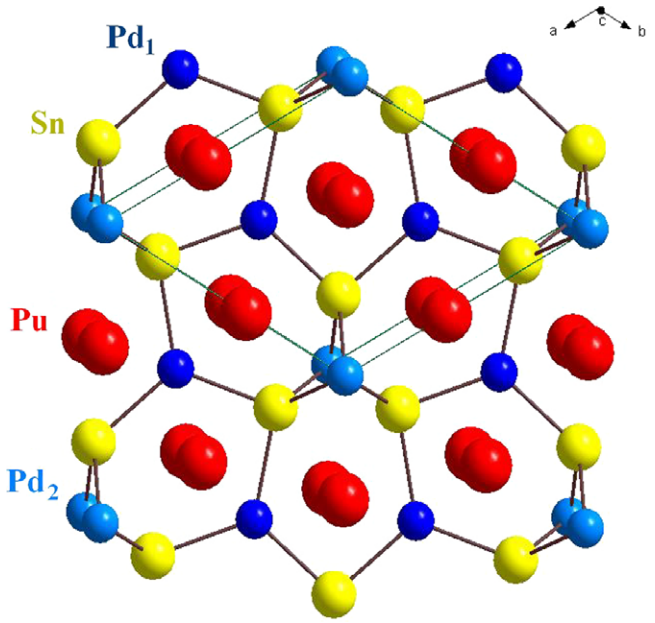

Fig. 1. Crystal structure of PuPdSn.

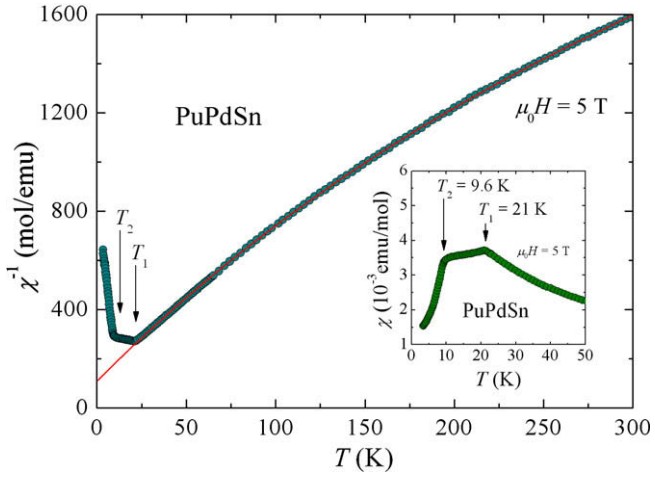

Fig. 2. Temperature dependence of the inverse magnetic susceptibility of PuPdSn measured in a magnetic field $\mu_0H = 5$ T. The solid line is a modified Curie–Weiss fit. Inset: low temperature part of the magnetic susceptibility. The arrows mark the magnetic phase transitions at 21 and 9.6 K.

In the paramagnetic region, the magnetic susceptibility is clearly curvilinear and follows a modified Curie–Weiss law:

$$\chi(T) = \frac{N\mu_{\mathrm{eff}}^2}{3k_B(T-\theta_{\mathrm{p}})} + \chi_0. \quad (1)$$

With an effective magnetic moment $\mu_{\mathrm{eff}} \sim 1.0\ \mu_B$, paramagnetic Curie temperature $\Theta_p = -14$ K and a temperature independent term $\chi_0 = 2.1 \times 10^{-4}$ emu/mol. The value of μ_{eff} obtained is larger than the free Pu^{3+} ion value expected for *LS* coupling (0.84 μ_B), but it is very close to that expected for intermediate coupling (1.01 μ_B). It has been shown recently that the intermediate coupling scheme is appropriate for the 5*f* electrons of Pu metal and Pu-based intermetallics [8,9]. The negative sign of Θ_p refers to antiferromagnetic exchange interactions and is consistent with the antiferromagnetic ordering in PuPdSn.

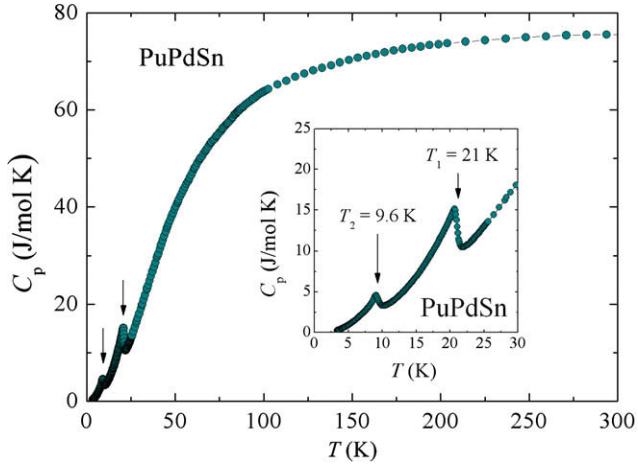

Fig. 3. Temperature dependence of the specific heat of PuPdSn. Inset: low temperature part of C_p. Arrows mark the phase transitions at 21 and 9.6 K.

3.2. Heat capacity

Fig. 3 shows the temperature dependence of the specific heat (C_p) of PuPdSn. Near room temperature C_p is close to 75 J/mole K that corresponds to the Dulong–Petit law i.e. $C_p = 3nR = 74.83$ J/mole K, where n is the number of atoms per molecule and R is the gas constant.

Heat capacity studies confirmed the presence of the two magnetic phase transitions manifested as pronounced λ-type peaks in $C_p(T)$ (see inset in Fig. 3). These peaks are hardly affected by magnetic field. Under a magnetic field as strong as 14 *T*, the maxima in C_p are slightly shifted towards low temperatures and insignificantly decrease in magnitude. The electronic contribution to the specific heat of PuPdSn is quite small at low temperatures. Below 5.5 K the $\frac{C_p}{T}(T^2)$ ratio extrapolated to $T = 0$ K is 8 mJ/mol K^2. Due to magnetic state this value may by treated as an upper limit of the value of the Sommerfeld coefficient in this material. It is also similar to the one previously observed for UPdSn [1].

4. Summary

The new Pu-based intermetallic compound PuPdSn was synthesized and studied by X-ray diffraction, magnetization and heat capacity measurements, performed in the temperature range 2–300 K and in magnetic fields up to 14 *T*. The compound crystalizes with hexagonal structure of ZrNiAl-type (s.g. $P\bar{6}2m$) with the lattice parameters: $a = 7.5057$ Å and $c = 4.0853$ Å. The magnetic measurements revealed that PuPdSn exhibits, as its uranium homologue, multiple magnetic phase transitions at 21 and 9.6 K. The low-temperature linear specific heat coefficient is small, indicating the presence of well localized 5*f* electrons in this material. However, before drawing any firm conclusion about the magnetic and electronic ground state in PuPdSn, future experimental studies are required. Some of them are presently underway.

Acknowledgements

We thank D. Bouëxière for technical assistance. High purity Pu metal was made available through a loan agreement between Lawrence Livermore National Laboratory and ITU, in the frame of a collaboration involving LLNL, Los Alamos National Laboratory and the U.S. Department of Energy. K. Gofryk acknowledges the European Commission for support in the frame of the Training and Mobility of Researchers programme.

References

[1] F.R. de Boer, E. Brück, H. Nakotte, A.V. Andreev, V. Sechovsky, L. Havela, P. Nozar, C.J.M. Denissen, K.H.J. Buschow, B. Vaziri, M. Meissner, H. Maletta, P. Rogl, Physica B 176 (1992) 275.
[2] H. Nakotte, R.A. Robinson, A. Purwanto, Z. Tun, K. Prokeš, E. Brück, F.R. de Boer, Phys. Rev. B 58 (1998) 9269.
[3] R.A. Robinson, A.C. Lawson, J.W. Lynn, K.H.J. Buschow, Phys. Rev. B 45 (1992) 2939.
[4] D. Kaczorowski, K. Gofryk, P. Boulet, J. Rebizant, P. Javorský, E. Colineau, F. Wastin, G.H. Lander, Physica B 359–361 (2005) 1102.
[5] K. Gofryk, D. Kaczorowski, E. Colineau, F. Wastin, R. Jardin, J.-C. Griveau, N. Magnani, J. Rebizant, P. Boulet, P. Javorský, R. Caciuffo, Physica B 403 (2008) 847.
[6] K. Gofryk, D. Kaczorowski, J.-C. Griveau, N. Magnani, R. Jardin, E. Colineau, J. Rebizant, F. Wastin, R. Caciuffo, Phys. Rev. B 77 (2008) 014431.
[7] K. Gofryk, J.-C. Griveau, E. Colineau, R. Jardin, J. Rebizant, F. Wastin, R. Caciuffo, Acta Phys. Pol. A, in press.
[8] K.T. Moore, M.A. Wall, A.J. Schwartz, B.W. Chung, D.K. Shuh, R.K. Schulze, J.G. Tobin, Phys. Rev. Lett. 90 (2003) 196404.
[9] A.O. Shorikov, A.V. Lukoyanov, M.A. Korotin, V.I. Anisimov, Phys. Rev. B 72 (2005) 024458.