



## Crystal structure and physical properties of PuPd<sub>5</sub>Al<sub>2</sub>

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### ABSTRACT

We report on the crystallographic aspects and the basic properties of the plutonium based compound PuPd<sub>5</sub>Al<sub>2</sub>. This material is antiferromagnetic at  $T_N = 5.6$  K and does not present any hint of superconductivity down to 2 K. This material crystallizes in the ZrNi<sub>2</sub>Al<sub>5</sub>-type of structure with lattice parameters:  $a = 4.1302$  Å and  $c = 14.8428$  Å. The magnetization, heat capacity and electrical resistivity measurements indicate clearly antiferromagnetic order at  $T_N = 5.6$  K. This material is compared to the structurally related cerium based material CePd<sub>5</sub>Al<sub>2</sub> presenting superconductivity induced by pressure.

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### 1. Introduction

Superconductivity has been reported recently in NpPd<sub>5</sub>Al<sub>2</sub> [1], a neptunium based intermetallic compound. It has been found that this material displays d-wave superconductivity ( $T_c = 4.9$  K) and heavy-fermion features ( $\gamma = 200$  mJ/mol K<sup>2</sup>) induced by strong electronic correlations [2]. This is the third transuranium based intermetallic compound presenting superconductivity after PuCoGa<sub>5</sub> [3] and PuRhGa<sub>5</sub> [4]. These systems have relatively high critical temperature  $T_c \approx 18.6$  and 9 K, respectively. Their neptunium counterparts NpCoGa<sub>5</sub> [5] and NpRhGa<sub>5</sub> [6] are not superconductors but antiferromagnets at 47 and 36 K, respectively. Taking into account the specificity of the intrinsic properties of the plutonium probably at the origin of superconductivity in the PuTGa<sub>5</sub> family, we decided to synthesize the plutonium counterpart of the NpPd<sub>5</sub>Al<sub>2</sub>, namely PuPd<sub>5</sub>Al<sub>2</sub>.

The compound of our interest was studied by means of X-ray diffraction, magnetization, specific heat and electrical resistivity performed in a wide temperature and magnetic field range (2–300 K, 0–14 T). Preliminary results have been already published elsewhere [7] but here we will focus more on structural and phase aspects (together with preparation and characterization) as well as we present some results of magnetization, specific heat and electrical resistivity of PuPd<sub>5</sub>Al<sub>2</sub>. It appears that the ZrNi<sub>2</sub>Al<sub>5</sub>-structure supports easily the actinide atom and compounds such as ThPd<sub>5</sub>Al<sub>2</sub> [7], UPd<sub>5</sub>Al<sub>2</sub> [8,9] and CePd<sub>5</sub>Al<sub>2</sub> [10,11] have already been reported in addition to NpPd<sub>5</sub>Al<sub>2</sub> [1,2,12]. UPd<sub>5</sub>Al<sub>2</sub> is relatively difficult to obtain a pure phase and seems to do not form by flux [8]. It was then important to examine the quality of the material obtained by arc melting by several techniques (powder and single crystal

X-ray diffraction, scanning electron microscopy). This is particularly challenging in the case of plutonium based materials where radioactivity and self disintegration play an important role. Here, the self-heating effect arising from the radioactive decay of the <sup>239</sup>Pu isotope ( $W = 1.9$  mW/g) prevented measurements below 2 K even in the case of a very small amount of compound (<1 mg).

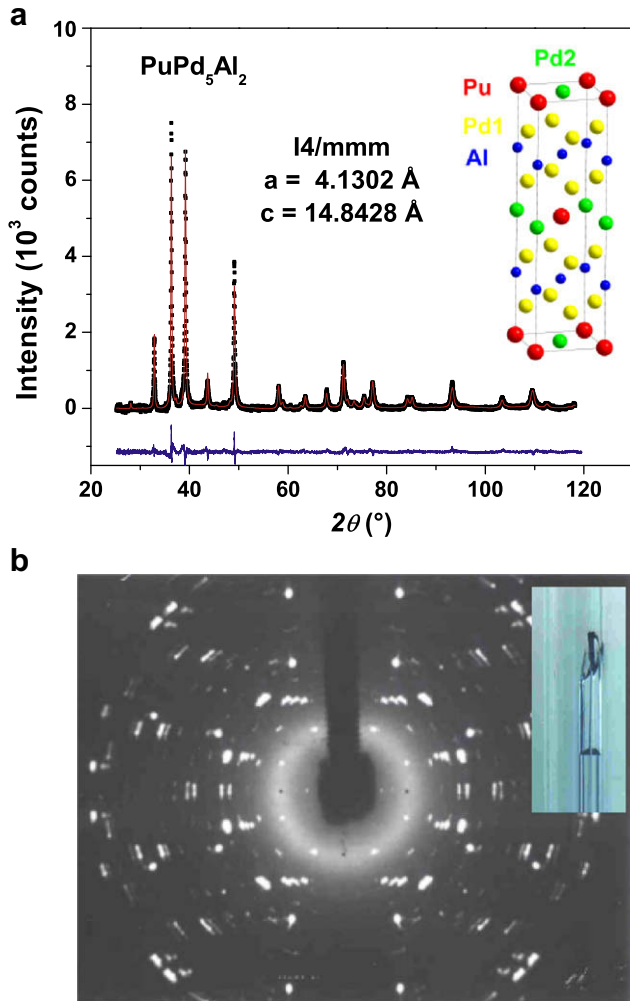
### 2. Preparation and characterization

The polycrystalline samples of PuPd<sub>5</sub>Al<sub>2</sub> were prepared by arc melting the stoichiometric amounts of the pure metals components in the ratio Pu:1 Pd:5 Al:2. Starting materials were used in the form of 4 N Pd pellet and 5 N Al wire as supplied by A.D. MacKay Inc. and 3 N Pu metal (<sup>239</sup>Pu isotope). The arc melting was performed under a high purity argon atmosphere on a water-cooled copper hearth, using a Zr alloy as an oxygen/nitrogen getter. The arc melted buttons were turned over and melted three times in order to ensure homogeneity. The weight losses after melting were smaller than 0.2%. Then the samples were examined by X-ray single crystal, powder diffraction methods and scanning electronic microscopy before physical properties examination.

The powder X-ray diffraction was done using a Bruker D8 diffractometer with the monochromated CuK $\alpha_1$  radiation ( $\lambda = 1.54059$  Å) equipped with a Vantec detector covering 6° in 2 $\theta$ . The powder diffraction pattern of PuPd<sub>5</sub>Al<sub>2</sub> is presented on Fig. 1(a). The powder patterns were recorded in the 2 $\theta$  range of 15–120° (step size of 0.007°), with an exposure of 14 s per step. The diffraction patterns were analyzed by a Rietveld-type profile refinement method [13] using the WinplotR–Fullprof program [14]. The samples were mounted on a rotating device to avoid any preferential orientation effect.

Single crystals suitable for crystal structure determination were mechanically extracted from the button and mounted in a capillary

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**Fig. 1.** (a) X-ray powder diffraction pattern ( $\lambda = 1.54059 \text{ \AA}$ ) recorded for  $\text{PuPd}_5\text{Al}_2$  (DRX). The solid red line through experimental points is the Rietveld refinement profile calculated for tetragonal  $\text{PuPd}_5\text{Al}_2$ . The blue lower curve represents the difference between experimental and model results. A sketch of the crystallographic structure is presented in the inset top left. (b) X-ray single crystal diffraction pattern obtained by rotating a single crystal extracted from the batch (inset top right). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

for X-ray diffraction by rotation (Fig. 1(b)). They were examined on a Enraf–Nonius CAD-4 diffractometer with the graphite monochromatized  $\text{MoK}_\alpha$ -radiation. The lattice constants, determined from least square analysis of the setting angles of 25 X-ray reflections, confirmed the tetragonal unit cell and are similar to those determined by X-ray powder diffraction. The crystal structure was solved by the direct method using SHELX97 [15] and the data processing using the WinGX package [16] are reported in Table 1. The crystal structure (Inset of Fig. 1(a)) was refined from the single crystal X-ray data and was corrected for Lorentz and polarization effects. It appears to be, like  $\text{NpPd}_5\text{Al}_2$  [1], tetragonal with the  $\text{ZrNi}_2\text{Al}_5$ -type (space group  $I4/mmm$ ) with lattice parameters  $a = 4.1302 \text{ \AA}$  and  $c = 14.8428 \text{ \AA}$ . The atomic coordinates obtained are presented in Table 2. Finally, the phase composition was determined by energy-dispersive X-ray (EDX) analysis. The measurements were performed on a Philips XL40 scanning electron microscope (SEM). The microprobe analysis (see Fig. 2) indicates that the prepared and studied samples of  $\text{PuPd}_5\text{Al}_2$  were homogeneous and single phase.

**Table 1**

Crystallographic data for  $\text{PuPd}_5\text{Al}_2$  obtained from a single crystal.

Composition	$^{239}\text{PuPd}_5\text{Al}_2$
Space group	$I4/mmm$ (No. 139)
Lattice parameters ( $\text{\AA}$ )	$a = 4.1302(14)$ $c = 14.8428(46)$
Cell volume ( $\text{\AA}^3$ )	253.20
Formula units per cell	$Z = 2$
Formula mass	825.11
Calculated density ( $\text{g/cm}^3$ )	10.82
Crystal size ( $\text{mm}^3$ )	$0.03 \times 0.20 \times 0.05$
Radiation	$\text{MoK}_\alpha$ ( $\lambda = 0.71073 \text{ \AA}$ )
Scans up to $2\theta$	$60^\circ$
Linear absorption coefficient	$30.38 \text{ mm}^{-1}$
Total number of reflections	1008
Reflections with $F_o > 4\sigma(F_o)$	Unique: 140 ( $R_F = 0.092$ )
Goodness of fit	1.040
Conventional residual $R$	0.0896 ( $F > 4\sigma$ )

**Table 2**

Crystallographic parameters for  $\text{PuPd}_5\text{Al}_2$ .

Atom	Site	$x$	$y$	$z$	$U_{\text{eq}}$
Pu	2a	0	0	0	0.0082
Pd1	8g	0.5	0	0.1456(3)	0.0063
Pd2	2b	0.5	0.5	0	0.0044
Al	4e	0	0	0.2521(20)	0.0156

The structure was refined with anisotropic displacement parameters for all atoms. The last column contains the equivalent isotropic  $U$  values ( $\text{\AA}^2$ ).

### 3. Magnetic properties

The magnetic properties were studied using a Quantum Design (QD) MPMS-7 device in the temperature range 2–300 K and in magnetic fields up to 7 T. The sample selected is presented in the inset of Fig. 3 with encapsulation. The temperature dependence of the magnetic susceptibility of  $\text{PuPd}_5\text{Al}_2$  is shown on Fig. 3. The distinct maximum on  $\chi(T)$  curve refers to the antiferromagnetic ordering at 5.6 K. Above 10 K the magnetic susceptibility follows a modified Curie–Weiss law with an effective magnetic moment  $\mu_{\text{eff}} = 1.05 \mu_B$ , a paramagnetic Curie temperature  $\Theta_p = -12.5 \text{ K}$  and  $\chi_0 = 6.9 \times 10^{-4} \text{ emu/mol}$ . The obtained value of  $\mu_{\text{eff}}$  is close to  $1.01 \mu_B$  as anticipated for  $\text{Pu}^{3+}$  in the intermediate coupling configuration. The negative sign of  $\Theta_p$  refers to antiferromagnetic exchange interactions and it is consistent with the antiferromagnetic ordering in the compound studied.

As shown in the upper inset of Fig. 3, a distinct peak in  $\chi(T)$  is clearly observed. With increasing magnetic field the maximum in the magnetic susceptibility gradually weakens.

### 4. Physical properties at low-temperature

#### 4.1. Specific heat

Heat capacity experiments were performed in the temperature range 2.5–300 K and in magnetic fields up to 14 T on a QD PPMS-14 setup using the relaxation method. The low-temperature dependence of the specific heat of a small polycrystal of  $\text{PuPd}_5\text{Al}_2$  is shown in Fig. 4. Due to contamination risk the sample was encapsulated by Stycast<sup>®</sup> 2850 FT glue (sample mass = 1.44 mg, Stycast mass = 0.381 mg) [17]. Inset in Fig. 4 displays an encapsulated sample mounted on the calorimeter. A distinct  $\lambda$ -shaped anomaly at  $T_N = 5.6 \text{ K}$  confirms the magnetic order in this material. When applying magnetic field the peak gradually weakens and shifts to lower temperatures (see Fig. 4). As shown in the previous

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