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Magnetic structures in TmPdIn and TmAgSn

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ABSTRACT

Low temperature antiferromagnetic structures of TmPdIn and TmAgSn have been derived from powder neutron diffraction data. The magnetic structure of TmPdIn is a commensurate one and related to a propagation vector $\vec{k} = \left[\frac{1}{3}, \frac{1}{3}, \frac{1}{2}\right]$ while the incommensurate sine-modulated structure of TmAgSn is connected with $\vec{k} = [k_x, -k_x, 0]$ where $k_x = 0.1314(2)$. The thulium magnetic moments are constrained within the basal plane and show 'triangular' arrangement. Validity of obtained magnetic structures is discussed on the basis of symmetry analysis.

1. Introduction

Equiatomic intermetallics RTX, where R is a rare earth element, T – d-electron metal and X – p-electron element, have been intensively investigated with respect to their crystal chemistry and physical properties [1,2]. In these compounds, the rare earth magnetic moments order antiferromagnetically at low temperatures.

TmPdIn and TmAgSn, investigated in this work, crystallize at room temperature in a hexagonal crystal structure of the ZrNiAl-type [3–5]. Previously reported magnetic and specific heat data indicated antiferromagnetic ordering below the Néel temperature $T_N = 2.7$ K in TmPdIn [6,7] and 4.2 K in TmAgSn [8], while the paper by Sebastian et al. reports no magnetic ordering in TmAgSn down to 2 K [4].

In this work, crystal and magnetic structures of TmPdIn and TmAgSn are investigated by means of neutron diffraction. It is a part of systematic study of magnetic structures in thulium ternary intermetallics of general composition TmTX. The compounds are investigated in order to determine influence of surroundings atoms on magnetic order. Up to now, antiferromagnetic ordering has been found in TmNiIn [9], TmPtIn [10], TmAgGe [11] and TmAgSi [12] below the critical temperature of 2.5 K, 3.5 K, 4.2 K and 3.3 K, respectively. Complex magnetic structures have been determined in all these compounds. As bulk measurements reported up to now for TmPdIn [6,7] and TmAgSn [8] cannot provide detailed information on magnetic structures, the neutron diffraction data are reported in this work.

2. Experimental details

Polycrystalline samples of TmPdIn and TmAgSn were prepared by arc melting constituent metals, with all stated purity better than 99.9 wt%, under ultra-pure argon atmosphere. Subsequently, the samples were wrapped in tantalum foil, sealed in an evacuated silica tube and heat treated at 973 K for 10 days. Quality of the samples were checked by X-ray powder diffraction (XRD) at room temperature on an X'Pert PRO PANalytical diffractometer with CuK_α radiation. The X-ray diffraction data are analyzed in Refs. [6,8].

Powder neutron diffraction patterns were collected at temperatures ranging from 1.8 to 8.0 K on the G4.1 diffractometer installed at the Orphée reactor (Laboratoire Léon Brillouin, CEA-CNRS Saclay, France). The incident neutron wavelength was 2.426 Å.

The obtained X-ray and neutron diffraction patterns were analyzed using the computer program *FullProf* [13]. Symmetry analysis was performed with the use of the computer program *basireps* distributed together with *FullProf*.

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3. Crystal structure

The X-ray diffraction data collected at room temperature as well as the neutron diffraction data indicate that the investigated samples have the ZrNiAl-type crystal structure with the lattice parameters being in agreement with literature data [3–5].

Fig. 1 shows neutron diffraction patterns taken at paramagnetic state at 6.0 K and 8.0 K for TmPdIn (Fig. 1a) and TmAgSn (Fig. 1b), respectively, together with calculated profiles obtained by Rietveld refinement method. Both patterns indicate presence of weak reflections that cannot be indexed within the main TmTX phases. Limited number of these low intensity reflections made identification of impurity phases impossible. Table 1 contains crystal structure information based on the best Rietveld fits to the neutron diffraction data.

4. Magnetic structure

The neutron diffraction patterns collected below the respective Néel temperatures for TmPdIn and TmAgSn show presence of additional Bragg reflections of magnetic origin (Figs. 2a and 3a, respectively).

In order to facilitate determination of the magnetic structures of both compounds a symmetry analysis was performed. The symmetry analysis method is based on the theory of representation of space groups formulated and developed by Bertaut [14] and Izyumov and Syromyatnikov [15]. A magnetic structure, as an axial vector function $\vec{S}^{\{\vec{k}_L\}}$ localised on the set of equivalent positions of a given space group, may be presented as a linear combination of basic vectors $\vec{\Psi}_{\nu,\lambda}^{\{\vec{k}_L\}}$ of irreducible representations of this space group

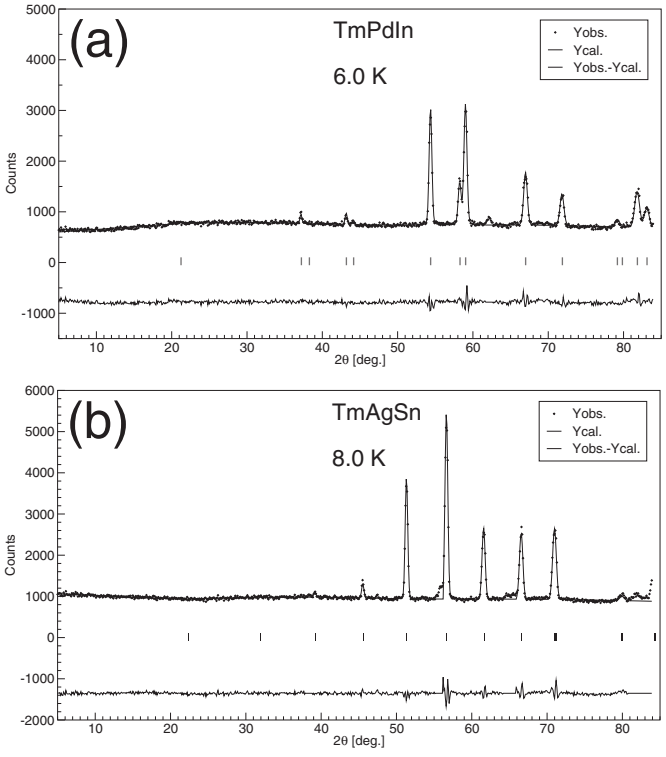

Fig. 1. Neutron diffraction patterns taken at paramagnetic state at 6.0 K (a) and 8.0 K (b) for TmPdIn and TmAgSn, respectively, together with Rietveld fit and difference plot. The vertical ticks indicate positions of Bragg reflections.

Table 1
Structural parameters of TmPdIn and TmAgSn refined from the neutron diffraction data collected at 6.0 K (TmPdIn) or 8.0 K (TmAgSn), and corresponding reliability factors.

	TmPdIn	TmAgSn
Crystal structure	ZrNiAl-type	
Space group	$P\bar{6}2m$ (No. 189)	
T [K]	6.0	8.0
a [Å]	7.617(2)	7.242(1)
c [Å]	3.706(1)	4.422(1)
c/a	0.4865(2)	0.6106(2)
V [Å^3]	186.21(9)	200.85(7)
Tm at 3(g)	$\left(x_{Tm},0,\frac{1}{2}\right)$ $\left(0,x_{Tm},\frac{1}{2}\right)$ $\left(\bar{x}_{Tm},\bar{x}_{Tm},\frac{1}{2}\right)$	
x_{Tm}	0.592(2)	0.570(2)
In or Ag at 3(f)	$(x,0,0)$ $(0,x,0)$ $(\bar{x},\bar{x},0)$	
x_{In} or x_{Ag}	0.264(4)	0.236(4)
Pd or Sn at 1(b)	(0,0,½)	
Pd or Sn at 2(c)	(1/3,2/3,0) (2/3,1/3,0)	
$R_{profile}$ [%]	3.23	2.61
R_{Bragg} [%]	10.2	3.60
χ^2 [%]	1.66	2.01

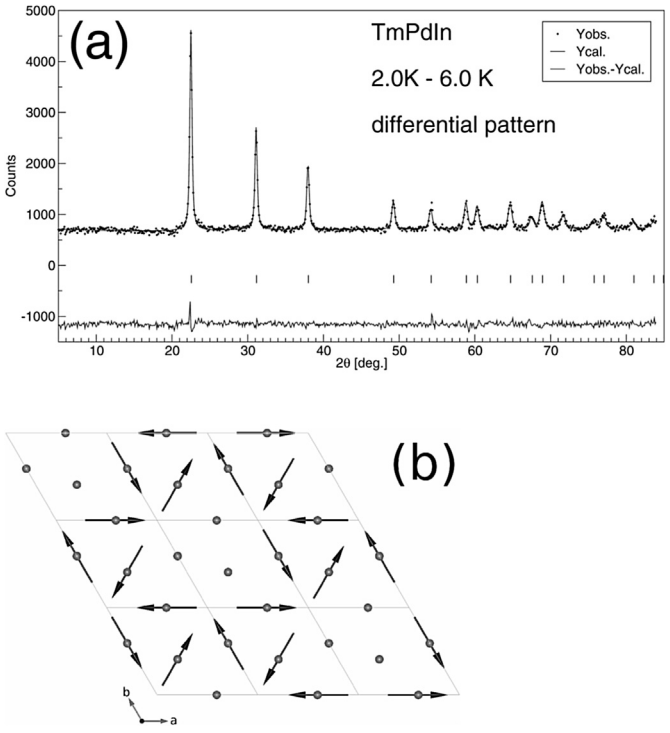

Fig. 2. (a) Differential neutron diffraction patterns of TmPdIn together with Rietveld fit and difference plot. The pattern was constructed as a difference between experimental patterns collected at 2.0 K and 6.0 K. The vertical ticks indicate positions of the magnetic reflections. (b) Magnetic structure of TmPdIn (see the main text for details). The magnetic unit cell of TmPdIn consists of two such layers coupled antiferromagnetically one to another (due to the ½ component of the propagation vector).

$$\vec{S}^{\{\vec{k}_L\}} = \sum_{L=1}^{l_{\vec{k}}} \sum_{\nu=1}^{j} \sum_{\lambda=1}^{l_\nu} C_{\nu,\lambda}^{\{\vec{k}_L\}} \vec{\Psi}_{\nu,\lambda}^{\{\vec{k}_L\}} \quad (1)$$

where L labels arms of the star for a given propagation vector $\vec{k}_L$, ν labels irreducible representations, λ labels dimensions of a particular irreducible representation while $C_{\nu,\lambda}^{\{\vec{k}_L\}}$ are the coefficients which have to be determined from refinement of the experimental