# MAGNETIC PHASE TRANSITIONS IN THE $TbTX_2$ COMPOUNDS (T-*d*-ELECTRON ELEMENTS, X = Sb, Ge)*

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The microscopic nature of the magnetic phase transitions in ordered states observed in $TbTSb_2$ (T = Cu, Pd) and $TbTGe_2$ (T = Pt, Ir) are discussed on the basis of neutron diffraction and magnetometric measurements.

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## 1. Introduction

In this paper the results of the magnetic and neutron diffraction measurements of the $TbTSb_2$ (T = Cu, Pd) and $TbTGe_2$ (T = Ir, Pt) are discussed. These compounds crystallize in different types of crystal structure, such as the tetragonal $ZrCuSi_2$-type (space group P4/nmm) [1] for $TbTSb_2$ compounds and $YIrGe_2$-type (space group Immm) [2] for $TbTGe_2$ compounds. In the first structure the Tb atoms occupy one site while in the second they occupy two nonequivalent crystallographic positions. These compounds are antiferromagnets with a complicated phase situation in an ordered phase.

Magnetic susceptibility and magnetization data were collected using the vibrating-sample and SQUID magnetometers. Neutron diffraction patterns were obtained at the BER II reactor (BENSC, Hahn-Meitner Institute, Berlin). The Rietveld-type program FULLPROF [3] was used for processing these data.

## 2. $TbTSb_2$ (T = Cu, Pd) compounds

With an increasing temperature the following magnetic phases in $TbCuSb_2$ appear:

- between 1.4 and 4.4 K the Tb moments form a collinear antiferromagnetic structure described by the propagation vector $\boldsymbol{k} = \left(\frac{1}{2}, 0, 0\right)$,
- between 4.4 and 5.8 K the magnetic ordering is sine-wave modulated with $\boldsymbol{k} = (0.4227, 0, 0)$,
- between 5.8 K and the Néel temperature equal to 9 K the magnetic order is again a collinear antiferromagnetic described by $\boldsymbol{k} = \left(0, \frac{1}{4}, \frac{1}{2}\right)$.

The magnetic moment of Tb, equal at $T = 1.4$ K to 7.80(5) $\mu_B$ is parallel to the [110] direction in the first two phases and to the [100] for the last one.

Magnetic peaks observed on the neutron diffraction pattern of $TbPdSb_2$ taken at $T = 1.4$ K are indexed as satellite pairs ($\pm\boldsymbol{k}$) with the propagation vector $\boldsymbol{k} = k_y\boldsymbol{b}^* + k_z\boldsymbol{c}^*$ ($k_y = 0.772$, $k_z = \frac{1}{2}$). An analysis of the magnetic intensities reveals that the $Tb^{3+}$ magnetic moments are parallel to the $c$-axis. The magnetic moment is given by $\mu(\boldsymbol{r}) = \mu_0 \sin(2\pi\boldsymbol{k}\cdot\boldsymbol{r} + \varphi_0)$ where amplitude $\mu_0$ equals 8.61(6) $\mu_B$, $\boldsymbol{r}$ is the position of rare earth atom and $\varphi_0$ is the phase. Two different models of the magnetic structure dependent of the phase $\varphi_0$ are possible for $\varphi_0 = \frac{\pi}{4}$ (Fig. 1(a)) and for $\varphi_0 = \frac{\pi}{2}$ (Fig. 1(b)).

The temperature dependence of the magnetic peak intensities $000^{\pm}$ and $010^{-}$ (Fig. 2) gives the Néel temperature at 11 K. Over a whole temperature region at the angles near two narrow magnetic peaks a broad peak connected with the short-range order is observed. An intensity of this peak increases with an increase of the temperature and reaches a maximum above $T_N$.

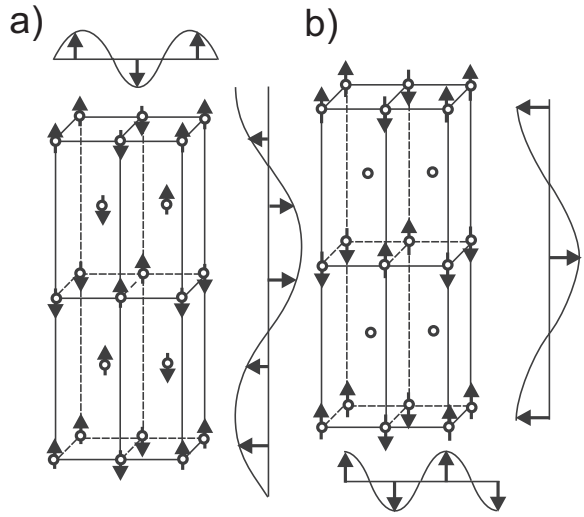


Fig. 1. Magnetic structures of $TbPdSb_2$ for $\varphi_0$ equal to $\frac{\pi}{4}$ (a) and $\frac{\pi}{2}$(b).

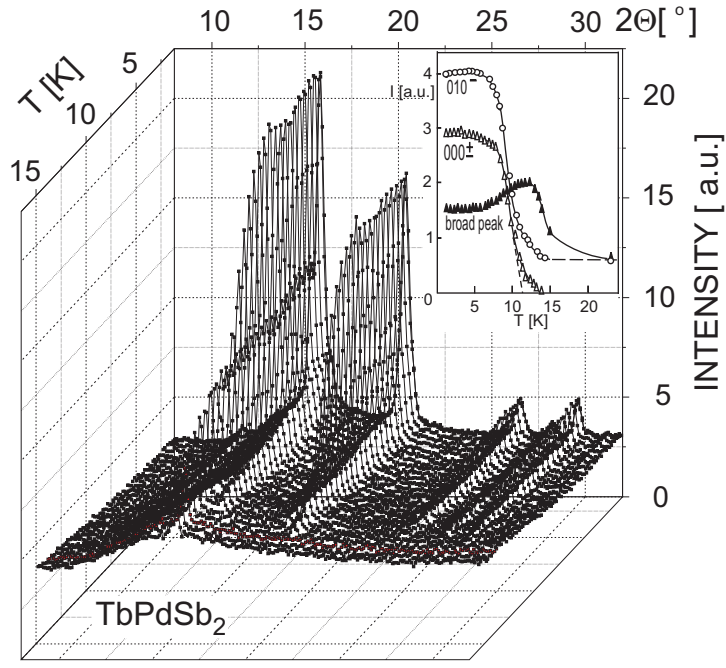


Fig. 2. Temperature dependence of the $TbPdSb_2$ diffraction patterns between 1.4 and 15 K. Angular range $2\theta = 6°$–$24°$. The inset shows the temperature dependence of the magnetic peaks $000^{\pm}$ (△) and $010^{-}$ (○) and the broad peaks (▲).

## 3. $TbTGe_2$ (T = Pt, Ir) compounds

The temperature dependence of the magnetic susceptibility has several following temperatures:

- $T_N = 24.2\,K$, $T_{t1} = 11.4\,K$ and $T_{t2} = 7\,K$ for $TbPtGe_2$ and $T_N = 10.2\,K$ and $T_{t1} = 9\,K$ for $TbIrGe_2$.

Neutron diffraction data indicate that the following models of the magnetic structures can be constructed:

- in both compounds below $T_N$ the Tb moments in the $4(i)$ sublattice: $\boldsymbol{\mu}_1(0,0,z)$, $\boldsymbol{\mu}_2(0,0,\bar{z})$, $\boldsymbol{\mu}_3(\frac{1}{2},\frac{1}{2},\frac{1}{2}+z)$ and $\boldsymbol{\mu}_4(\frac{1}{2},\frac{1}{2},\frac{1}{2}-z)$ form collinear antiferromagnetic arrangement. The orientation of magnetic moments is: $+\boldsymbol{\mu}_1 - \boldsymbol{\mu}_2 + \boldsymbol{\mu}_3 - \boldsymbol{\mu}_4$. The Tb moment is equal to $7.5(1)\ \mu_B$ at 15 K in $TbPtGe_2$ and it is $5.1(2)\ \mu_B$ at 9.4 K in $TbIrGe_2$ and parallel to the $b$-axis. The Tb moments in $4(h)$ site $(0,y,\frac{1}{2};0,\bar{y},\frac{1}{2};\frac{1}{2},\frac{1}{2}+y,0;\frac{1}{2},\frac{1}{2}-y,0)$ are not ordered,

- below $T_{t1}$ the magnetic order in $4(i)$ sublattice does not change while in $TbPtGe_2$ at the $4(h)$ site the Tb moments equal to $6.7(2)\ \mu_B$ at 8.8 K form a sine-wave-modulated structure with the wave vector $\boldsymbol{k} = (0.2677, 0.1312, 0.6989)$; they form an antiferromagnetic arrangement with the orientation of magnetic moments $+\boldsymbol{\mu}_1 - \boldsymbol{\mu}_2 - \boldsymbol{\mu}_3 + \boldsymbol{\mu}_4$ in the crystal unit cell and they lie in the $b$–$c$ plane.

In $TbPtGe_2$ below $T_{t2}$ the Tb moments localized at the $4(h)$ site form a new sine-wave modulated structure with the $\boldsymbol{k} = (0.2584, 0, 0.5895)$. The Tb moment equal to $9.0(2)\ \mu_B$ at 1.5 K lies in the $a$–$b$ plane.

In $TbIrGe_2$ the Tb moments equal to $8.4(2)\ \mu_B$ at 1.5 K form a collinear antiferromagnetic structure with the $\boldsymbol{k} = \left(\frac{1}{2},\frac{1}{2},0\right)$. The Tb magnetic moments lie in the $a$–$c$ plane and have the $+\boldsymbol{\mu}_1 - \boldsymbol{\mu}_2 + \boldsymbol{\mu}_3 - \boldsymbol{\mu}_4$ orientation.

## 4. Discussion

The exchange interactions and Crystalline Electric Field (CEF) are the two factors that influence the stability of magnetic ordering of the rare-earth magnetic moments in these compounds. Because of the large interatomic distances, for example in $TbTSb_2$ compounds a $R$–$R$ distances are about ~4 Å in plane and ~5.5 Å between planes, the exchange interactions between the localized $4f$ electrons are indirect and they are probably mediated via the conduction electrons (RKKY model). This type of exchange interactions favours a long-range oscillator type ordering, while magnetocrystalline anisotropy connected with the CEF favours an uniaxial magnetic ordering.

The magnetic structures of $TbCuSb_2$ indicate a strong magnetocrystalline anisotropy in the basal plane [7]. In the low temperature commensurate and incommesurate phases the Tb magnetic moment is parallel to the [110] direction while in the high temperature phase it lies along the [100] direction.

In $TbPdSb_2$, over a wide temperature range of temperatures up to the Néel temperature a coexistence of long and short range magnetic order is observed. It suggests that the magnetic ordering could be similar to that for $\varphi_0 = \frac{\pi}{2}$ presented in Fig. 1(b). The magnetic moment in center of the tetragonal cell $(\frac{1}{2}; \frac{1}{2}; \frac{1}{2})$ is frustrated and does not give a contribution to the lon g range order. Such a frustration of the magnetic moment is observed in the $CeNi_2Sn_2$ compound [4].

An influence of the Crystal Electric Field (CEF) is described by the Hamiltonian:

$$H_{\mathrm{CEF}} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_4^4 O_4^4 + B_6^0 O_6^0 \,,$$

where $O_n^m$ are the Stevens operators and $B_n^m$ are the CEF parameters defined by Hutchings [5]. The $B_n^m$ values determined for the tetragonal $RT_2Si_2$ compounds indicate that the $B_2^0$ parameter is dominant [6]. The direction of the magnetic moment with reference to the tetragonal $c$-axis is connected with the sign of the $B_2^0$ parameter. The magnetic moment is parallel to the $c$-axis if $B_2^0 < 0$ and it is perpendicular to the $c$-axis if the $B_2^0 > 0$ [7]. The determined direction of the magnetic moment suggests the negative value of the $B_2^0$ parameter for T = Pd and positive for T = Cu. The change of the direction of the magnetic moment with a change of temperature observed in $TbCuSb_2$ indicates an influence of higher-order terms [5].

The different orderings are observed in $TbTGe_2$ (Pt, Ir) compounds. In both compounds the Tb moments in $4(h)$ and $4(i)$ sublattices order magnetically at different temperatures. The $4(i)$ sublattice orders directly below the Néel temperature.

A distribution of the magnetic moments in the $4(i)$ sublattice (see Fig. 3) indicates that the exchange field induced by the Tb moments in $4(i)$ sublattice carries over the $4(h)$ sublattice. It causes that the Tb moments in $4(h)$ and $4(i)$ sublattices order in different temperatures and form a different types of structure.

In both sublattices the Tb magnetic moments have different directions. In the $4(i)$ sublattice the moments are parallel to the $b$-axis which suggests that the $B_2^0$ parameter is negative, while in the $4(h)$ sublattice the Tb moments are in the $a$–$b$ plane in $TbPtGe_2$ and in the $a$–$c$ plane in $TbIrGe_2$ which indicates different values of the $B_n^m$ parameters for different T elements.

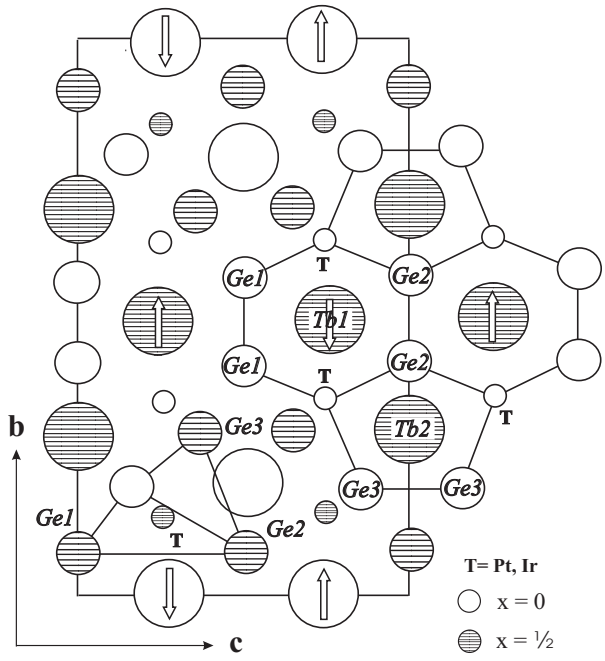


Fig. 3. Projection of the magnetic structure of $TbPtGe_2$ and $TbIrGe_2$ in the temperature range $T_{t1} < T < T_N$.

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