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Magnetic structures of $TbPd_2Si_2$ and $TbPd_2Ge_2$—a redetermination

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Abstract. The magnetic structures of $TbPd_2Si_2$ and $TbPd_2Ge_2$ were redetermined using high-resolution powder neutron diffraction data. $TbPd_2Si_2$ orders below $T_N = 16$ K in a sine-modulated structure with $\boldsymbol{k} = [0, 0.4057(12), 0.1671(16)]$. Magnetic moments of 9.0(1) μ_B at 1.5 K localized on Tb^{3+} ions are aligned along the a-axis. In $TbPd_2Ge_2$ a sine-modulated structure with magnetic moments of 8.9(1) μ_B at 1.5 K on Tb^{3+} ions, parallel to the a-axis, is described by the propagation vector $\boldsymbol{k} = [0, 0.4401(16), 0.1158(37)]$. The analysis of the magnetic peak half-widths observed at 1.5 K indicates that the magnetic order does not extend over the whole crystal, but is confined to clusters.

1. Introduction

Ternary terbium–palladium–silicon (germanium) phases with the 1:2:2 stoichiometry crystallize in the $ThCr_2Si_2$-type structure [1, 2] and often show complex magnetic ordering schemes at low temperatures [3–9]. For the $ErPd_2Si_2$ compound, high-resolution neutron diffraction data give below $T_t = 3.2$ K a square-wave-modulated structure with the wavevector $\boldsymbol{k} = (0.4778, 0, 0.791)$ at $T = 1.5$ K. In an intermediate-temperature range (3.2 K to $T_N = 4.8$ K) the magnetic structure is described by two wavevectors [7]. $TbPd_2Si_2$ and $TbPd_2Ge_2$ were earlier reported to exhibit incommensurate magnetic structures [8, 9]; however, in those experiments the neutron diffraction data were collected using an instrument with poor resolution. We have therefore decided to repeat the study of the magnetic structures of $TbPd_2Si_2$ and $TbPd_2Ge_2$ as it is now possible for us to carry out neutron diffraction measurements on the E6 diffractometer installed at the BERII reactor at the Hahn–Meitner Institute, Berlin. This instrument offers excellent resolution. Additionally, it was possible to obtain full diffraction patterns at selected temperatures, so the temperature variations of the magnetic peak intensities and positions could be traced. These are illustrated in figures 1 and 2.

2. Results and discussion

The experiment was performed on the same samples as were used in our previous studies [8, 9]. The incident neutron wavelength was 2.422(3) Å. The data were processed by the

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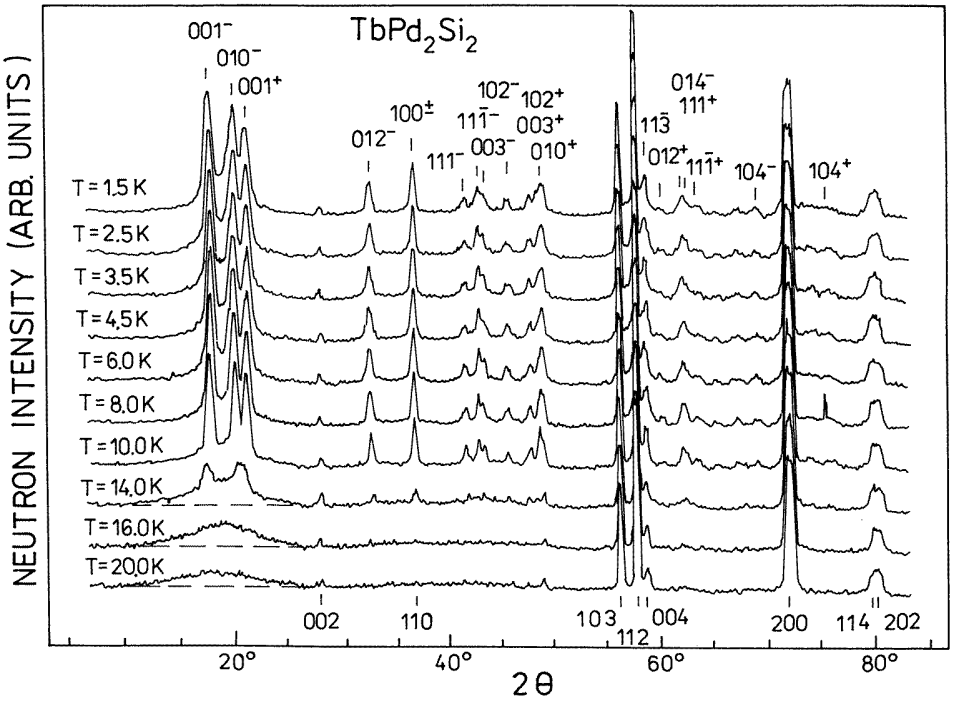

Figure 1. Neutron diffractograms of $TbPd_2Si_2$ recorded at the temperatures between 1.5 K and 20 K.

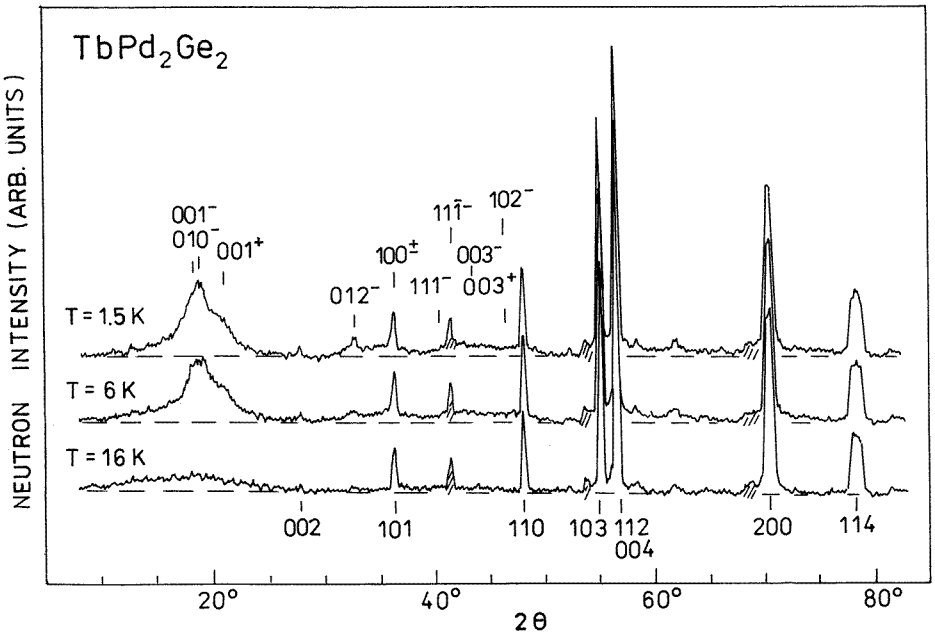

Figure 2. Neutron diffraction patterns of $TbPd_2Ge_2$ obtained at 1.5, 6 and 16 K.

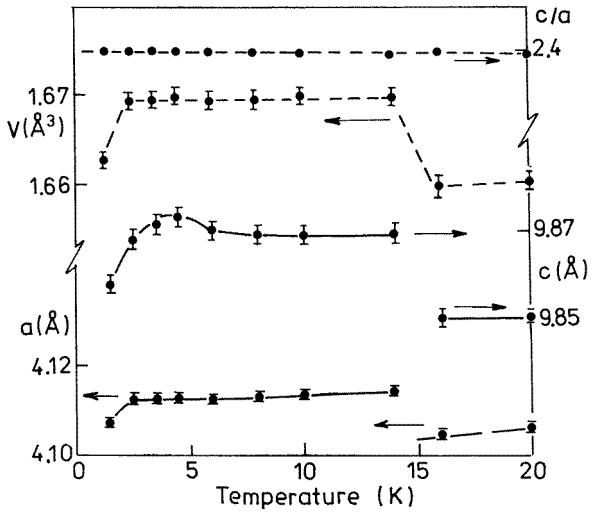

Figure 3. $TbPd_2Si_2$: the temperature dependence of the lattice parameters a and c, the unit-cell volume V, and the c/a ratio.

Table 1. The crystal and magnetic structure parameters for the $TbPd_2Si_2$ and $TbPd_2Ge_2$ compounds.

Compound:	$TbPd_2Si_2$		$TbPd_2Ge_2$	
T (K):	20	1.5	16	1.5
a (Å)	4.1066(8)	4.1072(7)	4.1874(14)	4.1867(10)
c (Å)	9.8510(20)	9.8575(27)	10.0178(39)	10.0185(26)
V (Å^3)	166.13(10)	166.29(9)	175.65(19)	175.61(13)
c/a	2.3988	2.4000	2.3924	2.3929
z	0.3760(16)	0.3780(14)	0.3814(8)	0.3810(6)
U	5.30(0.47)	4.40(0.38)	5.80(0.54)	5.80(0.54)
V	−4.42(0.33)	−4.13(0.32)	−4.71(0.56)	−4.71(0.56)
W	1.02(0.08)	1.16(0.07)	1.04(0.15)	1.04(0.15)
R_{prof} (%)	5.4	5.8	5.3	5.65
R_{Bragg} (%)	9.8	9.3	8.7	7.8
μ (μ_B)		8.9		8.9(1)
θ		92.0(5.4)		87.2(3.2)
φ		1.5(7.2)		2.8(4.0)
U		4.40(0.38)		135.4(18.4)
V		−4.13(0.32)		−97.1(11.6)
W		1.16(0.07)		20.5(1.7)
R_m (%)		14.6		13.8

Rietveld method using the FULLPROF program [10] with neutron scattering lengths taken from reference [11] and the Tb^{3+} form factor adopted from reference [12].

Neutron diffractograms of both compounds recorded above their Néel points contained reflections with $h+k+l$ even, confirming the space group $I4/mmm$, deduced earlier from x-ray diffraction studies to be characteristic for the $ThCr_2Si_2$-type structure [1, 2]. The refined crystallographic data at 1.5 K and above the respective Néel points are collected together in table 1. In addition, figure 3 shows the variation with temperature of the lattice parameters

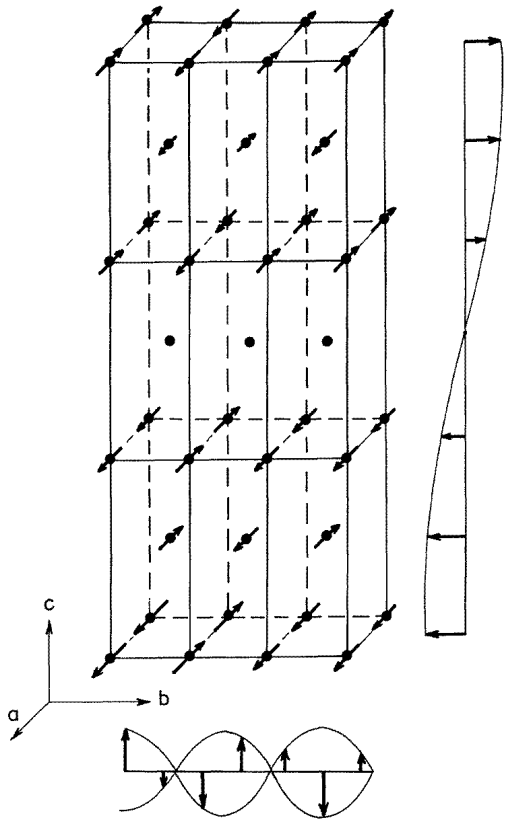

Figure 4. A schematic representation of the magnetic structure of $TbPd_2Si_2$ at 1.5 K.

a and c, of the unit-cell volume V, and of the c/a ratio. An increase at low temperature and a decrease near T_N of the lattice constants a and c, and the unit-cell volume are observed, whereas the c/a ratio is constant over the whole temperature region for $TbPd_2Si_2$.

Magnetic peaks observed in the low-temperature diffractograms of $TbPd_2Si_2$ (see figure 1) were indexed to a magnetic structure defined by the propagation vector $\boldsymbol{k} = |0, 0.4057(12), 0.1671(16)|$. In this model the indices of the observed magnetic peaks are such that $h + k + l$ is odd, indicating that the Tb moments located at the $(0, 0, 0)$ and $(1/2, 1/2, 1/2)$ sites in the $ThCr_2Si_2$-type unit cell are coupled antiferromagnetically. The minimum value for the R-index was obtained for a sine-modulated structure, as schematically displayed in figure 4. The terbium moment of 9.0(1) μ_B at 1.5 K was found to be parallel to the a-axis. Figure 1 shows that the long-range magnetic order in $TbPd_2Si_2$ vanishes at 15 K, but a diffuse magnetic peak is still present at 20 K. The temperature dependence of the magnetic moment value gives the Néel temperature as 16 K (see figure 5). The temperature dependence of the propagation vector components k_y and k_z is shown in figure 6. The value of the k_z-component of the wavevector is practically constant and close to 1/6 below 8 K, whereas the k_y-component increases first and then decreases with increasing temperature. This suggests that at 8 K a change from a structure that is commensurate along the c-axis to an incommensurate structure is observed.

The neutron diffractogram of $TbPd_2Ge_2$ recorded at 1.5 K displays a broad magnetic

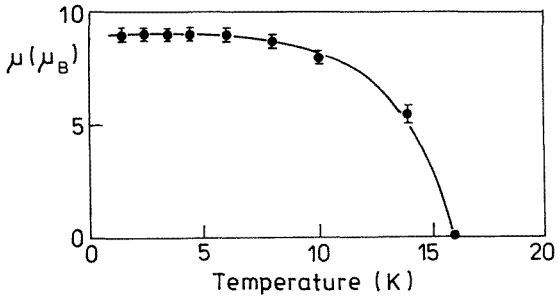

Figure 5. The temperature dependence of the Tb^{3+} magnetic moment in $TbPd_2Si_2$.

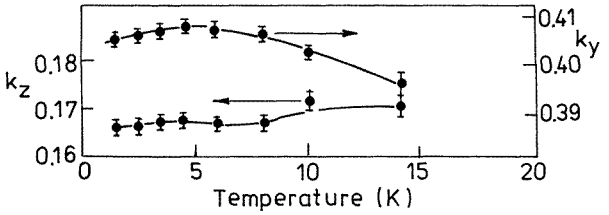

Figure 6. The temperature dependence of the wavevector components k_x and k_y for $TbPd_2Si_2$.

peak at small scattering angles (see figure 2). The FULLPROF analysis of all of the observed magnetic peak positions and intensities gave a model of the magnetic structure similar to that determined for $TbPd_2Si_2$. The structure is sine modulated with Tb^{3+} moments of 8.9(1) μ_B at 1.5 K along the a-axis. The propagation vector components are $[0, 0.4401(16), 0.1158(37)]$. Figure 7 shows the calculated profiles fitted to the observed data, and a difference pattern. This reasonably good agreement has been obtained by performing the analysis of the peak half-widths H_k separately for the magnetic and nuclear reflections.

The half-width parameters U, V, W derived from the expression $H_k^2 = U\tan^2\theta_k + V\tan\theta_k + W$ are listed in table 1. Here θ_k is the Bragg angle at which the reflection k is observed. The parameters U, V, W determined for the magnetic peaks are much larger than those for the crystallographic reflections, suggesting that the magnetic order in $TbPd_2Ge_2$ is confined to clusters and does not extend over the whole crystal.

The half-height of the Bragg reflection H_k is given by the formula

$$H_k = \frac{\alpha\lambda}{D_k\cos\theta_k}$$

where α is the Scherrer constant, λ is the wavelength of the neutron, D_k is the magnitude of the grain (cluster), and θ_k is the Bragg angle. With this relation, it is possible to determine the magnitude of the grain (cluster). Using the values of the half-width parameters listed in table 1, the size of a magnetic cluster in $TbPd_2Ge_2$ has been determined as being close to 100 Å.

The common feature of $TbPd_2Si_2$ and $TbPd_2Ge_2$ compounds is the short-range magnetic order observed at temperatures higher than their Néel points, manifesting itself as broad scattering maxima at angles at which magnetic peaks are observed at low temperatures. In $TbPd_2Ge_2$ long-range magnetic order is still absent at 1.5 K, as indicated by the values of

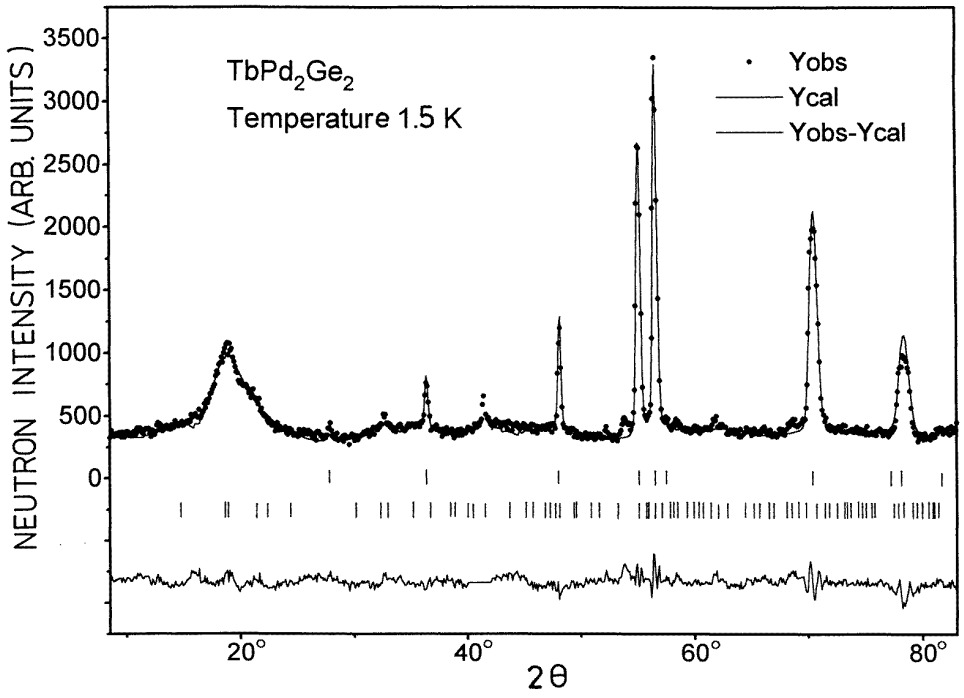

Figure 7. The observed neutron diffraction pattern of $TbPd_2Ge_2$ at 1.5 K, and the calculated profiles represented by solid lines. A difference pattern is shown below. The ticks indicate the positions of the nuclear (upper row) and the magnetic (lower row) reflections used in the profile analysis.

the half-widths of the magnetic peaks recorded at this temperature.

The magnetic structures of $TbPd_2Si_2$ and $TbPd_2Ge_2$ determined in the course of this study show different propagation vector parameters as compared to the data reported in references [8, 9]. The rather low resolution of the instrument used in the earlier experiments prevented us from separating the magnetic peaks observed at small Bragg angles, leading to a different interpretation of the experimental data. The Néel point of $TbPd_2Si_2$ was found to be at 15 K—as in the present study. The earlier $TbPd_2Ge_2$ data show an absence of magnetic order at 4.2 K, while this study reveals its presence at 6 K.

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