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Commensurate–incommensurate magnetic phase transition in $TbCo_2Ge_2$ compound

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Abstract. Neutron diffraction measurements reveal a magnetic phase transition in $TbCo_2Ge_2$ from a collinear antiferromagnetic order with the $\boldsymbol{k}$-vector [001] to an incommensurate sine-modulated magnetic structure described by the wave vector $\boldsymbol{k} = [0, 0, 1 - k_z]$ $k_z = 0.055(2)$. The transition occurs near the Néel point at 34 K. In both phases the magnetic moments localized on Tb^{3+} ions are parallel to the tetragonal axis.

1. Introduction

For many years the magnetic properties of RT_2X_2 ternary compounds (R—a lanthanide element, T—an nd transition metal and X—a p-electron element) have been a subject of continuing interest. RT_2X_2 compounds crystallize in the body-centred tetragonal $ThCr_2Si_2$ type of crystal structure (space group $I4/mmm$) [1]. RCo_2Ge_2 compounds belong to this structural type. Their magnetic properties have been studied by different groups [2, 3]. Among these compounds, $TbCo_2Ge_2$ has been reported to be an antiferromagnet with the Néel temperature of 30 K [4] or 32 K [5]. Neutron diffraction studies have shown that $TbCo_2Ge_2$ exhibits a collinear antiferromagnetic structure described by the wave vector $\boldsymbol{k} = [0, 0, 1]$ with the magnetic moments localized on Tb^{3+} ions and aligned along the tetragonal axis [5, 6]. The temperature dependence of the intensity of the $M100$ magnetic reflection gave the temperature of the transition to paramagnetic state at 30 K [5]. Recently, the results of a neutron diffraction study on a two-phase system of $TbCo_6Ge_6$ and $TbCo_2Ge_2$ have been published [7]. The authors report that below 22 K $TbCo_2Ge_2$ is antiferromagnetic with a collinear $\boldsymbol{k} = [0, 0, 1]$ magnetic structure. Above this temperature, this magnetic structure changes to a longitudinal, sinewave incommensurate phase described by the propagation vector $\boldsymbol{k} = [0, 0, 1 - k_z]$ with $k_z = 0.063(1)$. In both phases the magnetic moments are parallel to the tetragonal c-axis.

Since our neutron diffraction data reported in [5] were collected on an instrument with rather low resolution, we have decided to the measurements on the same sample but using the E6 diffractometer at the Berlin Neutron Scattering Centre which, apart from better incident neutron intensity, offers excellent resolution. The results of this study are presented below.

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2. Experiment and results

Neutron diffraction patterns were obtained on the E6 instrument installed at the BER II reactor on the Hahn–Meitner Institute, Berlin. The incident neutron wavelength was 2.44 Å. Data collection was done at temperature from 1.5 to 35 K (see figure 1). Data analysis was carried out with the FullProf program [8].

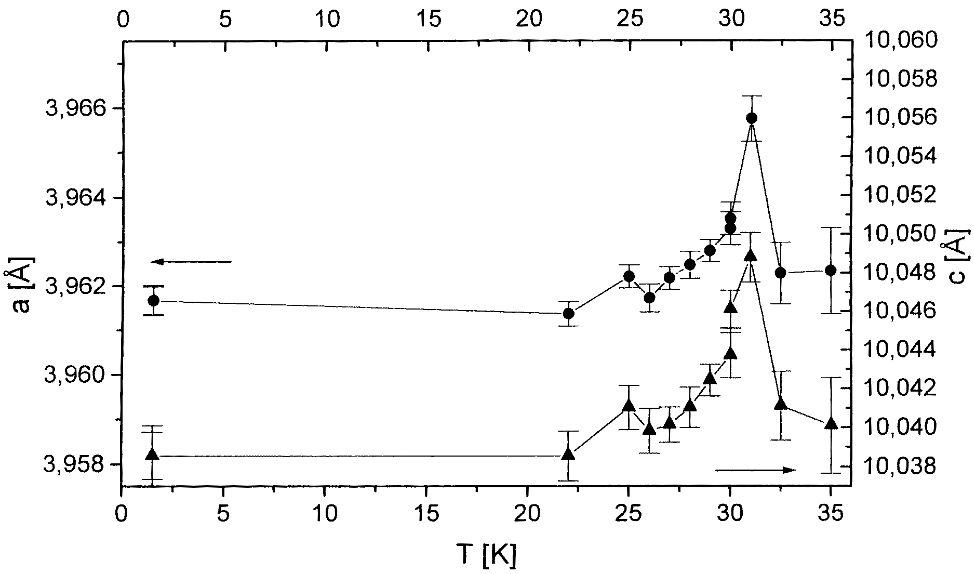

Figure 1. The temperature dependence of the a and c lattice parameters in the $TbCo_2Ge_2$ compound.

Neutron diffraction pattern run in the paramagnetic state at 35 K shows, that all observed reflections are consistent with the tetragonal, $ThCr_2Si_2$-type structure (space group $I4/mmm$). The sample was a single phase. The intensities were processed using atom parameters located in the following sites: Tb at 2(a): $(0, 0, 0)$, Co at 4(d): $(0, \frac{1}{2}, \frac{1}{4})$ and Ge at 4(e): $(0, 0, z)$. The best agreement between observed and calculated intensities was obtained for $z = 0.3725(6)$. Figure 1 shows the variance of lattice parameters a and c with temperature, as determined from neutron diffraction data recorded between 1.5 and 35 K.

The neutron diffraction pattern taken at 1.5 K is displayed in figure 2(a). A number of reflections of magnetic origin are observed. They were easily indexed on the crystallographic unit cell. The indices obeying the $h + k + l$ odd condition, which indicates the presence of antiparallel coupling of the magnetic moments in the $(0, 0, 0,)$ and $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ position. The corresponding magnetic structure is schematically shown in figure 3(a). The magnetic moments are localized on Tb^{3+} ions and parallel to the tetragonal axis. Their magnitude is $9.16(8)$ μ_B at 1.5 K. This type of magnetic order described by the wave vector $\boldsymbol{k} = [0, 0, 1]$, is stable up to 28 K. Neutron patterns obtained at 29 and 30 K reveal, that two satellite reflections around the magnetic peak M102 have appeared (see figure 2(b)). The intensity of the latter decrease as the temperature rises, while the intensities of the satellites rise simultaneously. The above effect indicates that a transition to a new magnetic order takes place. The positions of the satellite reflections are described by the wave vector $\boldsymbol{k} = [0, 0, 1 - k_z]$ with the refined value of $k_z = 0.055$. The inset in figure 4 shows how the magnitudes of k_z change with temperature.

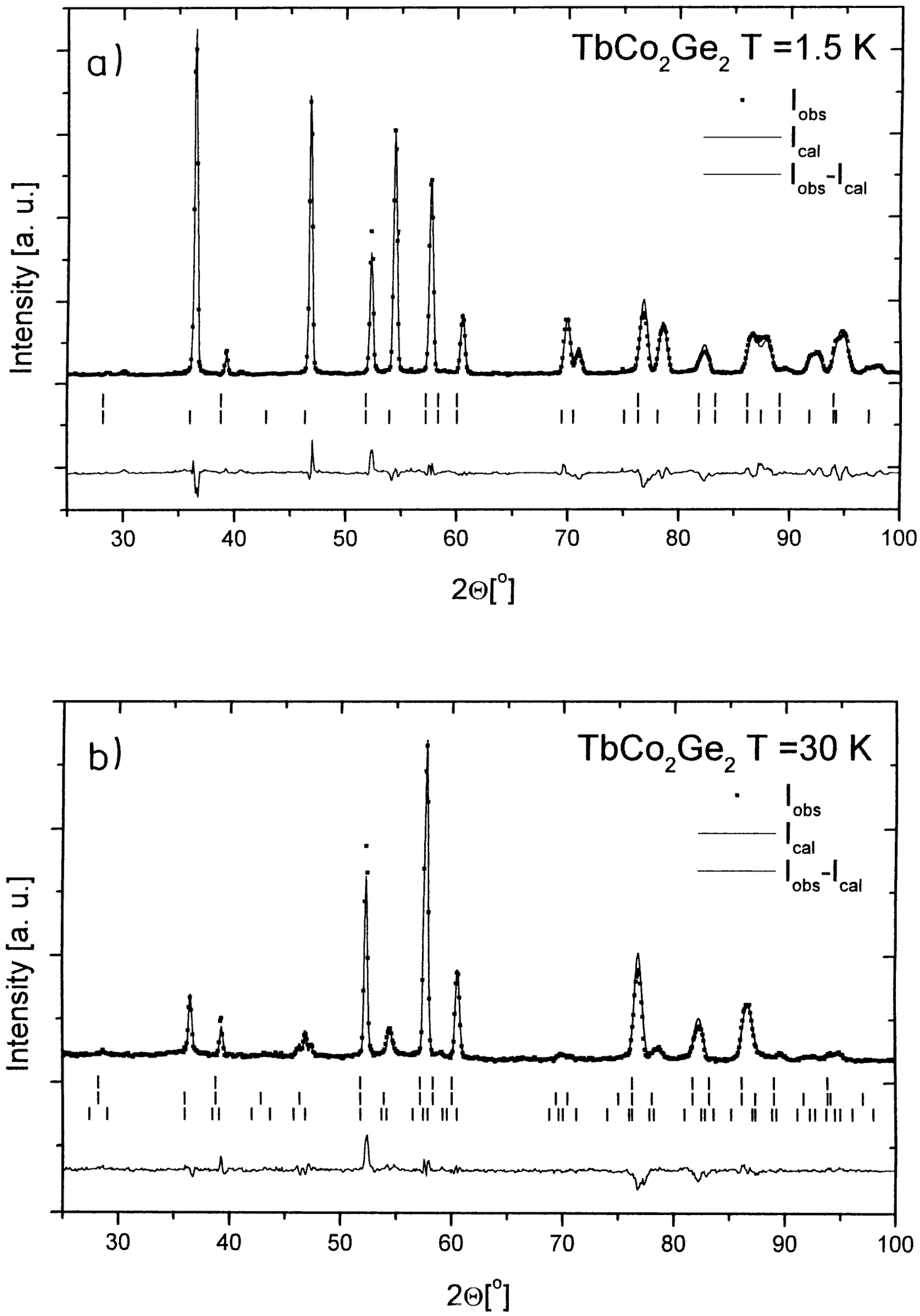

Figure 2. Neutron diffraction patterns of $TbCo_2Ge_2$ (a) recorded at 1.5 and (b) 30 K. The squares represent the observed points, the solid lines the calculated profile for the model crystal and magnetic structures described in the text and the difference between observed intensity and that calculated (below). The vertical bars indicate Bragg peaks for nuclear and collinear and sine-modulated magnetic structures.

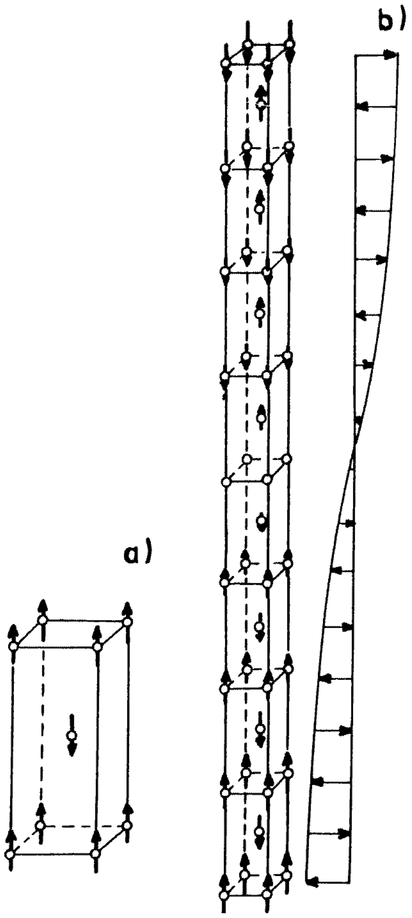

Figure 3. Magnetic structures of $TbCo_2Ge_2$ at (a) low (collinear) and (b) high (sine-modulated) temperature phases.

The best agreement between the observed and calculated intensities was obtained for a sine-modulated structure, schematically shown in figure 3(b). The absence of magnetic reflections $M00l$ in both phases indicates that the magnetic moments are parallel to the tetragonal axis. From a plot of magnetic moment values as a function of temperature, displayed in figure 4, it can be concluded that only the collinear antiferromagnetic phase is present below 28 K. Both phases, collinear and sine modulated, coexist in the temperature range between 28 and 30 K. Above 30 K only the sine-modulated phase is observed. The Néel temperature is at 34 K.

Listing of observed and calculated neutron intensities can be obtained from the corresponding author on request.

3. Comment

The results of the present experiment confirm the existence of a magnetic phase transition in $TbCo_2Ge_2$ near the Néel point, which could not be detected in our previous study due to low resolution of the available diffractometer [5]. The collinear antiferromagnetic order transforms into an incommensurate, sine-modulated structure which exists in a narrow temperature range

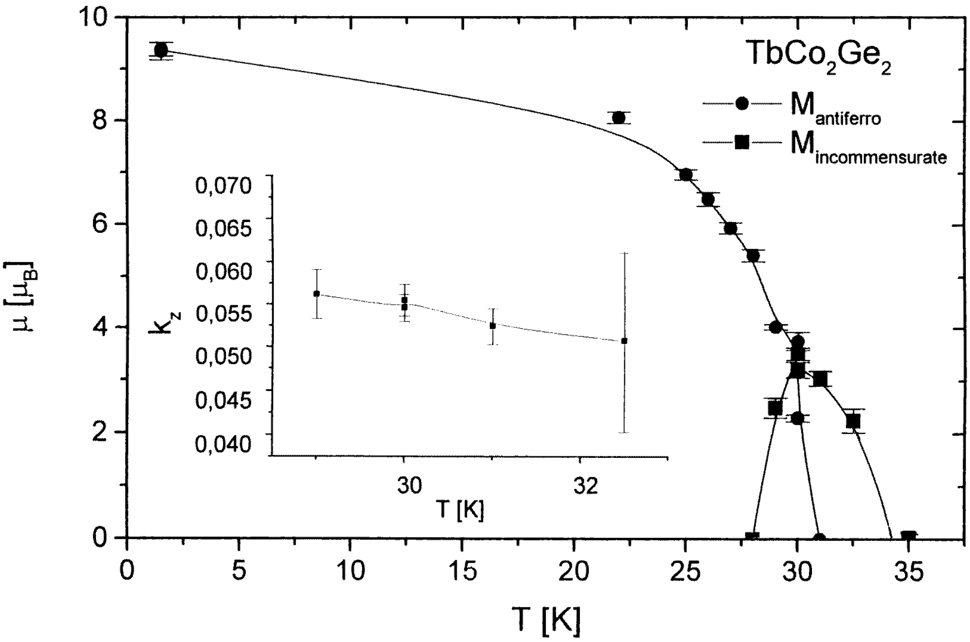

Figure 4. The temperature dependence of collinear (●) and modulated (■) components of the Tb magnetic moment. The inset shows the thermal dependence of the k_z component of the wave vector for $TbCo_2Ge_2$.

from 29 to 34 K (the Néel point), in fair agreement with the data reported in [7]; however, the value of the z-component of the wavevector determined in the course of our study is different from that given in [7]. We have shown that this magnetic phase transition is accompanied by a distinct anomaly in temperature dependence of the lattice parameters (see figure 1).

The transition from a commensurate magnetic ordering at low temperatures to an incommensurate structure in the vicinity of the Néel point has been observed in a large number of RT_2X_2 compounds with $ThCr_2Si_2$-type crystal structure [9], for example, in $TbNi_2Si_2$ [10], $TbCo_2B_2$ [11] and $DyCo_2Si_2$ [12].

Two factors influence the stability of the observed in experiment magnetic structures in RT_2X_2 compounds: exchange interactions via conduction electrons (RKKY model) and the action of crystalline electric field (CEF) of 4f electrons. The former factor favours long range oscillatory magnetic structures, the latter the uniaxial ordering. Both magnetic structures detected in $TbCo_2Ge_2$ exhibit the alignment of magnetic moments along the tetragonal c-axis. This observation is consistent with the positive sign of the second order crystal field parameter A_2^0 determined for RCo_2Ge_2 compounds by a ^{155}Gd Mössbauer spectroscopy experiment [13]. The occurrence of the magnetic phase transition to an incommensurate, oscillatory structure may be explained in terms of the realistic mean field model, which takes into account the temperature dependence of the periodic exchange field and CEF effect [9].

The magnetic phase diagram of $TbCo_2Ge_2$ constructed in the presence of external magnetic field (H, T diagram) shows that the incommensurate magnetic structure appears only when the magnetic field is applied [14]. Our study indicates, however, that the incommensurate phase is stable at $H = 0$ in a narrow temperature range in the neighbourhood of the Néel point. The value of the wave vector component $k_z = 0.052$ reported in [14] for phase III on the (H, T) diagram is close to that found in our study.

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