Proc. Int. Conf. Heavy Electrons (ICHE2010)
J. Phys. Soc. Jpn. **80** (2011) SA083

Powder neutron diffraction study of $TbCoGa_5$

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(Received September 8, 2010)

We report powder neutron diffraction study on $TbCoGa_5$ in order to understand the successive components-separated magnetic transitions. $TbCoGa_5$ exhibits a collinear antiferromagnetic order with the propagation vector $k_1 = (1/2,\ 0,\ 1/2)$ in the intermediate phase II. The magnetic moments are parallel to the *c* direction. In phase III, the *a*- or *b*-component of the magnetic moment is ordered, in addition to the *c*-component order in phase II, resulting in the formation of an antiferromagnetic order canted from the *c*-direction. The results of the present study strongly indicate that, in phase II, the *c*-component of the magnetic moment is ordered but the *a*- or *b*-component of the magnetic moment is non-ordered state.

KEYWORDS: $TbCoGa_5$, powder neutron diffraction, antiferromagnet, magnetic structure, multiply magnetic phase transition, successive components-separated magnetic transitions

1. Introduction

$TbCoGa_5$ adopts the $HoCoGa_5$ structure (space group $P4/mmm$) as shown in Fig. 1. Within this structure the Tb ions can be thought of as forming a two dimensional square lattice in the *ab*-tetragonal plane. Bulk measurements show that this material undergoes two magnetic transitions at $T_{N1} = 36.2$K and $T_{N2} = 5.4$ K. Below T_{N1} the *c*-component of the magnetic moment of Tb^{3+} orders antiferromagnetically but the *ab*-components remain paramagnetic down to T_{N2} where they order. We called the phenomena "successive components-separated magnetic transitions".[1)] This type of components-separated ordering has been previously observed in the 1D-chain triangular magnet $CsNiCl_3$[2–6)] and in DyB_4[7)] in which the arrangement of the Dy atoms in the *ab*-plane is the Shastry-Sutherland lattice. In $CsNiCl_3$, spin frustration gives rise to the unusual successive phase transitions. Furthermore, in DyB_4, the suppression of the magnetic transition temperature for the *ab*-components of the magnetic moment is thought to result from a combination of quadrupolar geometrical frustration and/or competing interactions between the quadrupolar and magnetic dipole moments via the crystalline electric field. On the other hand, Tb ions in $TbCoGa_5$ form the simple rectangular lattice. Therefore, for $TbCoGa_5$, it can be considered that a non-geometrical frustration effect gives rise to the successive components-separated magnetic transitions. It is important to directly confirm the occurrence of the successive components-separated magnetic transitions. Therefore, in order to determine the magnetic structure of $TbCoGa_5$ in phase II and phase III, we have performed a powder neutron diffraction experiment in this study.

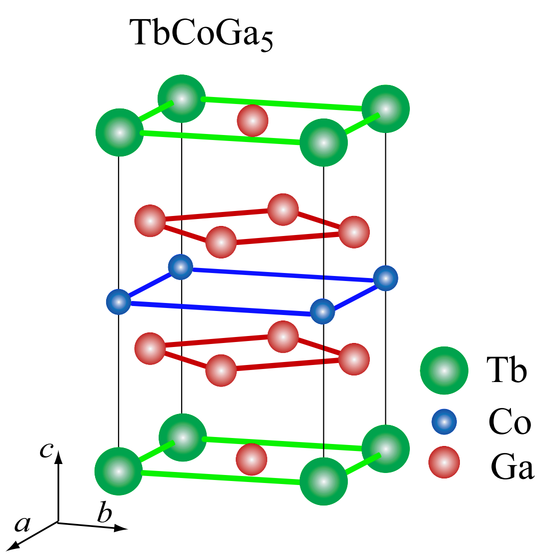

Fig. 1. Crystal structure of $TbCoGa_5$

2. Experimental

Experimental sample of $TbCoGa_5$ was prepared by self-flux method using Ga as a flux. The starting materials were inserted in a quartz tube at a ratio of Tb : Co : Ga =1 : 1 : 30 and sealed under high vacuum. This tube was heated to 750 °C and cooled slowly to room temperature at a rate of −10° C/h.

Powder neutron diffraction experiments were performed in the temperature range 1.5 K–50 K at the Laboratoire Léon Brillouin using the PYRRHIAS (G 4-1) diffractometer ($\lambda = 2.4226$; 800 cells position-sensitive detector). The powder sample of $TbCoGa_5$ was set in a cylindrical vanadium can (10 mm diameter) and held in a liquid helium cryostat. The neutron diagrams have been analyzed with the Rietveld method using the refinement program FullProf.[8)]

3. Results and discussions

3.1 *Magnetic structure in phase II (between T_{N1} and T_{N2})*

Figure 2 shows the powder neutron diffraction patterns of $TbCoGa_5$ at 50 K, 12 K and 1.5 K. All the magnetic Bragg peaks at 12 K can be indexed with $h/2\ k\ l/2$ within a parent unit cell of the crystal structure of $TbCoGa_5$ as shown in Fig. 2 (b). The result indicates that the propagation vector of the magnetic orderings in phase II is $k = (1/2,\ 0,\ 1/2)$. The temperature dependence of the magnetic susceptibility of

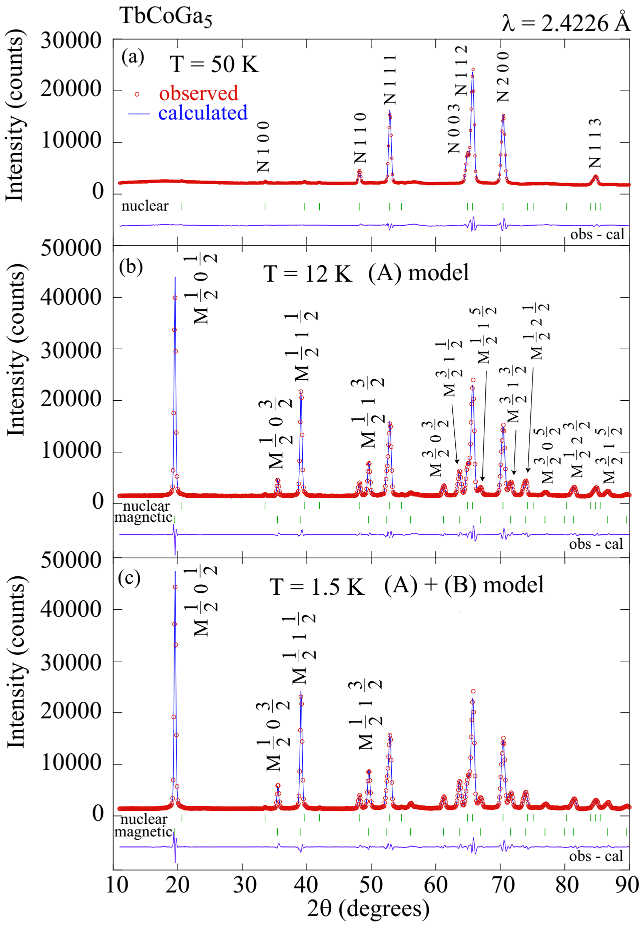

Fig. 2. Rietveld refinement patterns of powder neutron diffraction for $TbCoGa_5$ at 50 K, 12 K and 1.5 K. The open circles represent the observed data. The solid line indicates the Rietveld fitted curve. The upper and lower vertical bars mark the positions of the nuclear and magnetic reflections, respectively. The solid line at the bottom represents the difference between observed and calculated data.

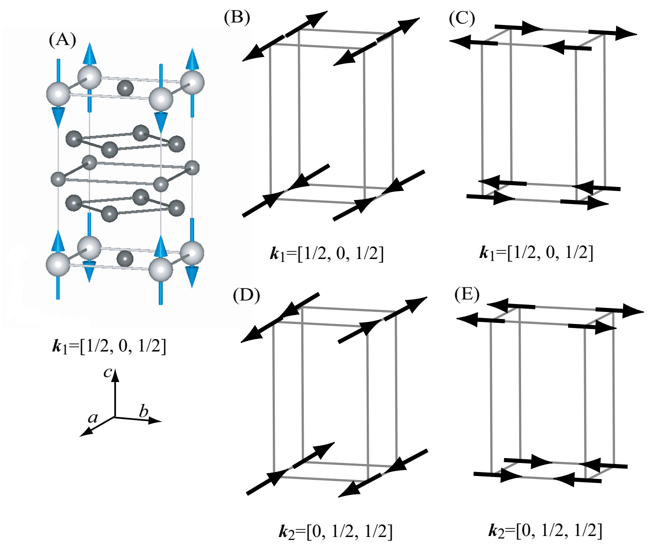

Fig. 3. Five set of possible basis-models for a magnetic structure of $TbCoGa_5$. Model A explains the powder pattern at 12 K. A superposition of model A and one of the other models explain the pattern at 1.5 K.

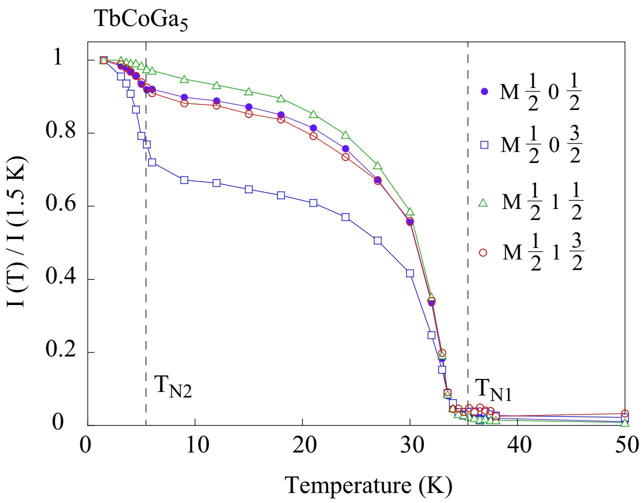

Fig. 4. Temperature dependence of the integrated intensities normalized to 1.5 K values of the M $\frac{1}{2}$ 0 $\frac{1}{2}$, M $\frac{1}{2}$ 0 $\frac{3}{2}$, M $\frac{1}{2}$ 1 $\frac{1}{2}$ and M $\frac{1}{2}$ 1 $\frac{3}{2}$ reflections. The lines are guides for eyes.

$TbCoGa_5$[1)] indicates that the magnetic moments in phase II are parallel to the *c* direction. Moreover, the magnetic unit cell is two times larger in the *a*- and *c*-axis than the crystallographic unit cell. Hence, the basis models are shown in Fig 3. The powder neutron diffraction pattern of $TbCoGa_5$ in phase II was refined by Rietveld method using these magnetic structure model in Fig. 3. Figure 2 (b) shows the best fit of Rietveld analysis in phase II using the model (A). The reliability factors and goodness of fit indicator are R_{wp} = 4.74 %, R_{exp} = 1.14 % and χ^2 = 17.3, respectively. The magnitude of the magnetic moment was deduced to be 8.32(3) μ_B. This value is the 92.4 % of the full moment of the Tb^{3+} ion, i.e., 9 μ_B.

3.2 Magnetic structure in phase III (below T_{N2})

There are no changes of magnetic Bragg positions between phase II and phase III as shown in Fig 2 (b) and (c). Therefore, at first, we performed Rietveld refinement of the data of 1.5 K using the model (A). The reliability factors and goodness of fit indicator at 1.5 K using the model (A) are R_{wp} = 5.53 %, R_{exp} = 1.14 % and χ^2 = 23.5, respectively. The model (A) fit well to data of 1.5 K, however, there are a few reflections whose intensities cannot be explained enough only by the model (A). Figure 4 shows the temperature dependence of the integrated intensities of the M $\frac{1}{2}$ 0 $\frac{1}{2}$, M $\frac{1}{2}$ 0 $\frac{3}{2}$, M $\frac{1}{2}$ 1 $\frac{1}{2}$ and M $\frac{1}{2}$ 1 $\frac{3}{2}$ reflections. The background components were subtracted from the data. Although the M $\frac{1}{2}$ 1 $\frac{1}{2}$ reflection hardly shows an anomaly at T_{N2}, integrated intensities of the M $\frac{1}{2}$ 0 $\frac{1}{2}$, M $\frac{1}{2}$ 0 $\frac{3}{2}$ and M $\frac{1}{2}$ 1 $\frac{3}{2}$ reflections obviously rise up just below T_{N2}. These behaviors can be explained by the orderings of *a*- or *b*-component of the magnetic moment below T_{N2}.

Therefore, we performed the Rietveld refinement on powder neutron diffraction pattern using four mixed models: (A)+(B) model and (A)+(C) model described with a single propagation vector k_1 = (1/2, 0, 1/2), (A)+(D) model and (A)+(E) model described with two propagation vectors k_1 = (1/2, 0, 1/2) and k_2 = (0, 1/2 1/2) which are normal to each other. However, by a powder neutron diffraction experiment, it is impossible to determine whether the magnetic structure of $TbCoGa_5$ in phase III is a single-*k* or a double-*k* structure in principle because of a tetragonal crystal structure. On the one hand, there are hardly any differences between results of Rietveld analysis for (A)+(B) and (A)+(E) model, and between those for (A)+(C) and (A)+(D) model. Therefore, we were not able to determine the the magnetic structure of $TbCoGa_5$ in phase III by this experiment. Accordingly, we should investigate the magnetic structure in phase III by the neutron diffraction and resonant x-ray diffraction study using a single crystal. Moreover, we should perform NMR measurements to confirm whether the magnetic structure in phase III is a collinear structure or a non-collinear structure.

As an example, the result of Rietveld analysis for the (A)+(B) model is shown in Fig. 2 (c). The reliability factors and goodness of fit indicator are R_{wp} = 5.10 %, R_{exp} = 1.14 % and χ^2 = 20.0, respectively. These values better than those by the model (A). The magnitudes of the *c*-component, *a*-component and total value of the magnetic moment were deduced to be 8.56(4) μ_B, 2.54(12) μ_B and 8.92(7) μ_B, respectively. The *a*-component of the magnetic moment order at T_{N2}, in addition to the *c*-component order in phase II, resulting in the formation of an antiferromagnetic order canted from the *c*-direction. It should be noted that the differences between magnitudes of the *c*-component of the magnetic moment in phase II and phase III are very small. In other words, the ordered magnetic moment in phase II does not tilt simply below T_{N2}. In this way, the results of the present study strongly indicate that, in phase II, the *c*-component of the magnetic moment is ordered but *ab*-components of the magnetic moment is non-ordered state.

In an ordinary magnet with successive magnetic phase transitions, second- and first-order transitions appear at upper and lower transition temperatures, respectively. It is to be noted that the magnetic phase transitions at both T_{N1} and T_{N2} in $TbCoGa_5$ are second order ones. It seems unlikely that the unusual successive phase transitions are caused by the ordinary magnetic interactions. The second order–second order phase transitions may result from a strong frustration due to a spin–orbital (quadrupole) interaction.

4. Acknowledgement

This work was supported by a Grant-in-Aid for Scientific Research (C) (No. 21540355) from the Japan Society for the Promotion of Science.

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