Proc. Int. Symposium on Advances in Neutron Scattering Research, Tokai, 2000
J. Phys. Soc. Jpn. **70** (2001) Suppl. A pp. 209–211

Neutron Diffraction Studies of Two-Dimensional Magnetic Ordering in $TbRu_2Ge_2$ at Low Temperatures

Shinji KAWANO, Takeshi KOSUGI[1], Miwako TAKAHASHI[2], Toru SHIGEOKA[3] and Nobuo IWATA[3]

Research Reactor Institute, Kyoto University,Kumatori, Sennan, Osaka 590-0494, Japan
[1]*Graduate School of Human and Environmental Studies, Kyoto University, Kyoto 606-8501, Japan*
[2]*Institute of Materials Science, University of Tsukuba, Tsukuba 305-8573, Japan*
[3]*Faculty of Science, Yamaguchi University, Yamaguchi 753-8512, Japan*

We have investigated the magnetic structure of a single crystal $TbRu_2Ge_2$ at low temperatures, T<4.3 K, by pulsed neutron diffraction, and found out many magnetic satellites to newly distribute at the positions formalistically expressed as $((2n+1)+(2m+1)\tau\ 1\pm\tau\ 0)$ and $(\pm\tau\ 2n+(2m+1)\tau\ 0)$, where n and m are integer and τ=4/17, additionally to the fundamental $(\tau\ 0\ 0)$ and its odd harmonics observed below T_N=37K. From the observed satellites the magnetic structure can be explained with a two-dimensionally squared-up wave model with stacking of the ferromagnetic (2 0 0) plane along the a-axis as a sequence of $\circ 4\bar{4}4\bar{4} \bullet 4\bar{4}4\bar{4}$, where $\circ$ and $\bullet$ denote a mixed plane containing Tb moments up, down and nonmagnetic, while '4' and '$\bar{4}$' the four successive ferromagnetic (2 0 0) planes with moments up and down, respectively.

KEYWORDS: 2D-modulation, mixed plane, pulsed-neutron diffraction, $TbRu_2Ge_2$ single crystal

§1. Introduction

The ternary rare-earth compound $TbRu_2Ge_2$ crystallizes in the tetragonal $ThCr_2Si_2$-type structure (space group: $I4/mmm$). Only Tb ions bear the magnetic moment parallel or anti-parallel to the c-axis, and are simply arranged in a body centered tetragonal lattice. This compound shows a variety of interesting magnetic properties: successive magnetic transitions and a multi-step metamagnetic transition, which are caused by a competition between crystalline electric field effects and long-range RKKY exchange interaction .[1,2] Magnetic measurements have revealed an existence of three antiferromagnetic phases: (1) a high-temperature phase at T_N=37 K>T>$T_{t'}$=32 K, (2) an intermediate-temperature phase at $T_{t'}$ >T>T_t=4.3 K, and (3) a low-temperature phase at T<T_t. Shigeoka *et al.* [3] have reported from neutron diffraction measurements that three antiferromagnetic phases are based on a one-dimensional spin configuration stacking ferromagnetic (1 0 0) planes parallel and/or anti-parallel along the [1 0 0] direction, described by a fundamental propagation vector $\boldsymbol{Q}=(\tau\ 0\ 0)$ with τ=0.235(=4/17), and the coexistence of sinusoidal modulated structures with (0.235 0 0) and (0.247 0 0) is realized for the high-temperature phase. On the other hand, for the low-temperature phase Garnier *et al.* [4] have reported, from the analysis of neutron diffraction and magnetization measurements, that some of ferromagnetic (1 0 0) planes become nonmagnetic with zero-moment, so that a mixed phase occurs, where magnetic and non-magnetic (1 0 0) planes coexist. They have proposed the sequence of Tb planes is $4\bar{4}04\bar{4}$, where 'n' ('$\bar{n}$') stands for n successive planes with moments up (down), and 0 stands for a non-magnetic plane. However, their neutron diffraction measurements were only on the high symmetry line, i.e., scans of the type (1 k 0).

Recently Kawano *et al.*[5] have investigated the magnetic structures of $TbRu_2Si_2$ which shows similar magnetic behavior, to the present $TbRu_2Ge_2$, by neutron diffraction, and revealed that the magnetic structure at low temperatures below 5K, becomes a two-dimensional modulation having many magnetic satellites on low-symmetry lines as well as on high-symmetry lines in the a^*-b^* reciprocal plane.

In the present note we will present the results of pulsed-neutron diffracton of the $TbRu_2Ge_2$ single crystal, in particular for the low temperature phase.

§2. Exprimental

Pulsed-neutron diffraction measurements of the single crystal $TbRu_2Ge_2$ were carried out using a four circle neutron diffractomer for a single crystal (FOX) at the Neutron Scattering Facilities of High Energy Accelerator Research Organization (KENS) at Tsukuba, Japan. Since in the FOX system 36 ^{3}He neutron detectors are one-dimensionally arranged to form a countor bank, one can measure Bragg reflections and/or diffuse scattering widely distributed in a reciprocal plane efficiently and in high signal-to-noise ratio at a time by using pulsed-white neutrons without moving a sample and any counter.

The c-axis of the single crystal was vertically oriented, i.e., only (h k 0)-type reflection data were collected for the a^*-b^* reciprocal plane. The sample was mounted in a liquid helium cryostat and cooled down to 1.7 K. The present interest is concentrated on the low temperature phase for T<T_t=4.3 K. Therefore, the measurements were mainly performed at the temperature range from 4.5 K to 1.7K.

§3. Results and Discussion

Figure 1 gives neutron diffraction patterns in the a^*-b^* reciprocal plane at 4.5 K, just above T_t=4.3 K and at

2.5 K, well below T_t. For the diffractin pattern at 4.5 K (lower figure of Fig. 1) magnetic satellites are mainly observed on the high-symmetry [1 0 0], [2 0 0] and [0 0 1] lines. All of the magnetic satellites are indexed with a propagation vector $\boldsymbol{Q}=(\tau\ 0\ 0)$ with $\tau=0.235(=4/17)$ and its odd harmonics. These results are fully consistent with the previous ones.[3] On the other hand, the diffraction pattern at 2.5 K (upper figure of Fig. 1) newly indicates many satellites on the $(h\ 1\pm\tau\ 0)$ and $(0\pm\tau\ k\ 0)(\tau=0.235)$ lines, and their equivalent lines. Since $\tau=0.235=4/17$

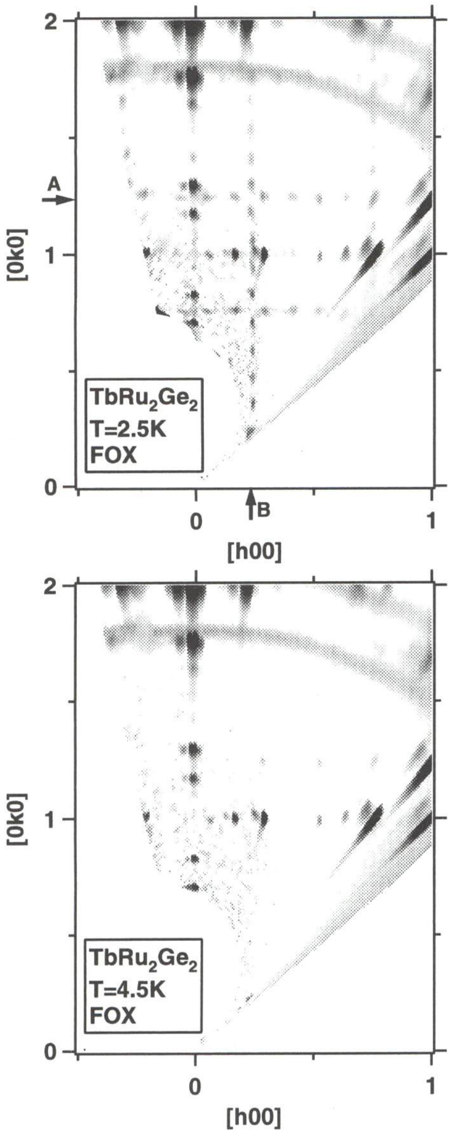

Fig. 1. The observed distribution of pulsed-neutron intensities in the a^*-b^* reciprocal plane for the $TbRu_2Ge_2$ single crystal at 2.5 K (upper) and at 4.5 K (lower). New magnetic satellites on the $(h\ 1+\tau\ 0)$ and $(0+\tau\ k\ 0)$ lines, where $\tau=0.235$, appear along the lines indicated by the arrows A and B in the upper figure, respectively. Two broad arcs in the figures are Debye-Scherrer rings of scattering from an Al cryostat.

for the intermediate-temperature phase, the spin configuration can be expressed as a long period commensurate structure, composed of the (1 0 0) ferromagnetic planes with a sequence of $5\bar{4}4\bar{4}4\bar{4}4\bar{5}$ along the [1 0 0] direction. For this spin structure the distribution of magnetic Bragg reflections can be expressed with the fundamental $(\tau\ 0\ 0)$ wave and its odd harmonics. By the crystal symmetry the [1 0 0] direction is equivalent with the [0 1 0] one, so that the observed distribution of magnetic Bragg reflections in the a^*-b^* reciprocal plane shows the existence of these two domains.

On the other hand, for the low-temperature phase the spin configuration would be given by modifying the magnetic waves of the intermediate-temperature phase to form a two-dimensional modulation. As shown in Fig. 2, magnetic intensities along the A, $(h\ 1+\tau\ 0)$ and B, $(\tau\ k\ 0)$ lines of the upper figure in Fig. 1 can be indexed with odd harmonics.

Figure 3 gives the temperature dependence of the magnetic $(\tau\ 5\tau\ 0)$ and $(-1+5\tau\ 1+\tau\ 0)$ reflections. Both intensities disappear around 4 K, corresponding to T_t, with increasing temperature. The observation of these magnetic satellites on the low-symmetry lines is for the first time in this material, but the appearance of the satellites is very similar in $TbRu_2Si_2$.[4] Shigeoka *et al.*[3] have observed an anomaly in the temperature dependence of the $(1\ 1\text{-}3\tau\ 0)$ reflection; showing almost constant intensities below T_t. Note that this magnetic reflection is on a high-symmetry line. The anomaly comes from the appearance of those new satellites on the low-symmetry lines, additional to the satellites on the high-symmetry lines.

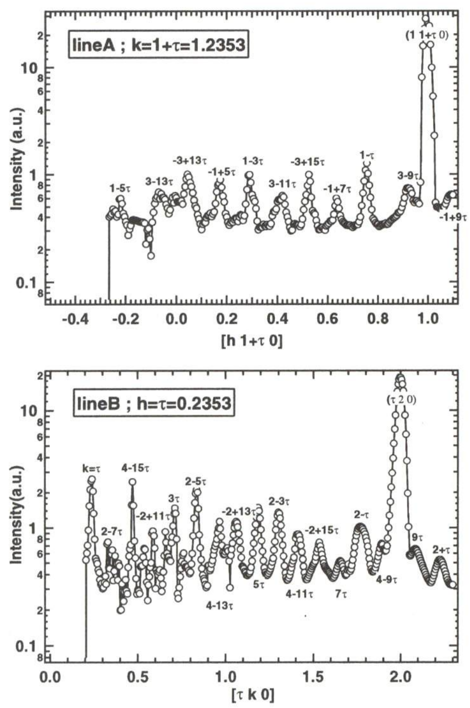

Fig. 2. Pulsed-neutron diffraction patterns along the $(h\ 1+\tau\ 0)$ and $(0+\tau\ k\ 0)$ lines with $\tau=0.235$, showing the arrows A and B lines in the upper figure of Fig. 1. The peaks can be indexed with higher odd harmonics such as $((2m+1)+(2n+1)\tau\ 1+\tau\ 0)$ for the line A and $(\tau\ 2m+(2n+1)\tau\ 0)$ for the line B in Fig. 1.

With respect to the magnetic structure of the low-temperature phase, basically it seems to be modulated from the $(\tau\ 0\ 0)$ structure, because all the satellites observed at the low-temperature phase can be in-

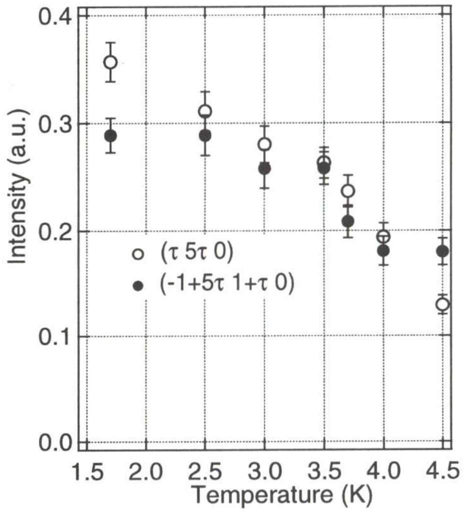

Fig. 3. Temperature dependence of the integrated intensities of the magnetic (τ 5τ 0) and (-1+5τ 1+τ 0) reflections.

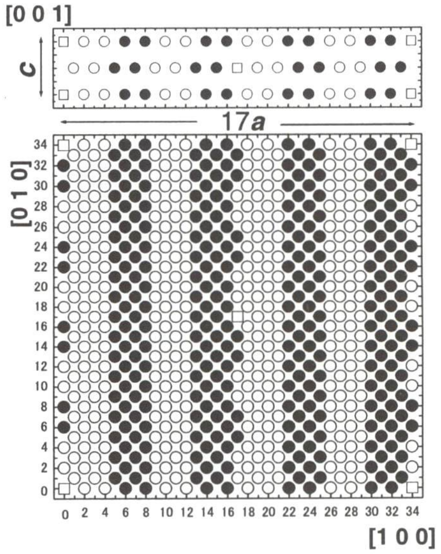

Fig. 4. The proposed magnetic structure model of the low-temperature phase for $TbRu_2Ge_2$. The open circles, solid circles and open squares denote Tb moments up, down and non-magnetic, respectively.

dexed with higher harmonics from the satellites at the intermediate-temperature phase, as shown in Fig. 2. The previously-proposed model[2)] of the mixed phase for the low-temperature phase can never explain the appearance of many satellites on the low-symmetry lines, because the model is one-dimensional with $\boldsymbol{Q}=(\tau\ 0\ 0)$.

Here, we will propose the following model for the magnetic structure; the magnetic unit cell is $17\boldsymbol{a}\times17\boldsymbol{a}\times\boldsymbol{c}$, containing regularly arranged nonmagnetic Tb atoms, as illustrated in Fig. 4. This (1 0 0) plane appears for every 17 layers. The plane contains Tb moments up, down and non-magnetic, so that the plane is, in a sense, a defective plane or a discommensuration, similar to a spin-slip plane, which we have already met the helical phase in Ho[6)] and the sinusoidal phase in Er.[7)] Hereafter we call this plane the mixed plane because of the coexistence of magnetic and non-magnetic Tb ions. The phase for this mixed plane (the 0th (1 0 0) plane in Fig. 4) is shifted by π for the next mixed plane (the 17th plane) along the a-axis. Therefore, if a mixed plane and its π-shifted one, are denoted by ∘ and •, respectively, then the spin configuration can be expressed as ∘$4\bar{4}4\bar{4}$ • $4\bar{4}4\bar{4}$. Kawano[8)] has already given similar phenomenological discussion of the long period spin modulations in $PrCo_2Si_2$ and $NdCo_2Si_2$. For the intermediate-temperature phase the spin configuration is expressed as the sequence of $5\bar{4}4\bar{4}4\bar{4}4\bar{5}$, forming a one-dimensional anti-phase structure along the a-axis. This structure changes to ∘$4\bar{4}4\bar{4}$ • $4\bar{4}4\bar{4}$ at low temperatures. The places of • and ∘ are the positions of very weak exchange interaction, i.e., the strongly frustrated positions, because the plane corresponds to an anti-phase boundary. In such planes non-magnetic Tb may occur. Shigeoka *et al.*[3)] have suggested that this non-magnetic Tb arises from a singlet-ground state due to huge crystalline electric field effects.

Thus, one-dimensional modulation of the intermediate temperature phase is additionally modulated by introduction of the mixed planes to become two-dimensional. Because of tetragonality of the crystal structure, another domain with the same spin configuration exists with $\boldsymbol{Q}=(0\ \tau\ 0)$. Accordingly, the distribution of the satellites reflects these two domains, giving the observed pattern.

For this model a preliminary calculation can reproduce the observed distribution of the satellites in the a^*-b^* reciprocal plane. Since there are some difficulties in the presice estimation of satellite intensities for pulsed-neutron experiments, reactor-neutron experiments are needed. The study along this way is now in progress.

Acknowledgements

This work was performed under the Visiting Research Program of the High Energy Accelerator Research Organization, Tsukuba, Japan.

1) J.K. Yakinthos and E. Roudaut: J. Physique **47** (1986) 1239.
2) A. Garnier, D. Gignaux, D. Schmitt and T. Shigeoka: Physica B **212** (1995) 343.
3) T. Shigeoka, M. Nishi and K. Kakurai: Physica B **237-238** (1997) 572.
4) A. Garnier, D. Gignaux, D. Schmitt and T. Shigeoka: Phys. Rev. B **57** (1998) 5235.
5) S. Kawano, B. Lebech, T. Shigeoka and N. Iwata: Physica B **276-278** (2000) 572.
6) D. Gibbs, D.E. Moncton, K.L. D'Amico, J. Bohr and B.H. Grier: Phys. Rev. Lett. **55** (1985) 234.
7) D. Gibbs, J. Bohr, J.D. Axe, D.E. Moncton and K.L. D'Amico: Phys. Rev. B **34** (1986) 8182.
8) S. Kawano: J. Magn. Magn. Mater. **114** (1992) 258.