Proc. Int. Symposium on Advances in Neutron Scattering Research, Tokai, 2000
J. Phys. Soc. Jpn. **70** (2001) Suppl. A pp. 212–214

Neutron Diffraction Studies for the Magnetic Structures of Tb_7Rh_3

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Neutron diffraction studies on the hexagonal Tb_7Rh_3 have been performed from 1.5 K to 110 K. Tb_7Rh_3 shows the reentrant magnetism containing antiferromagnetic ($T_t = 27$ K), ferrimagnetic ($T_C = 73$ K) and another antiferromagnetic phase ($T_N = 91$ K). The magnetic structure is helical at 1.5 K with propagation vector $\boldsymbol{Q} = (0\ 0\ 1/3)$. In the ferrimagnetic phase, conical magnetic structure with a basal plane helix can be considered; a kind of modification of the magnetic structure occurs at around 50 K. Propagation vector does not change with temperature.

KEYWORDS: neutron diffraction, helical structure, Tb_7Rh_3

§1. Introduction

The rare earth intermetallic compound Tb_7Rh_3 crystallizes in the hexagonal Th_7Fe_3 type crystal structure with the space group $P6_3mc$ in which Tb occupies three non-equivalent sites.[1,2] Magnetic measurements suggest that there exists three magnetic phases; antiferromagnetic ($T_N = 91$ K), ferrimagnetic ($T_C = 73$ K), and another antiferromagnetic phase ($T_t = 27$ K). In the ferrimagnetic phase, we observed small spontaneous magnetization of 0.06 μ_B/Tb in the c-axis at 50 K.[3] Electrical properties are anomalous; the temperature coefficient of electrical resistivity is negative at around room temperature and metallic conductivity is maintained at low temperature. Furthermore, a large increase of electrical resistivity was observed below T_N.[3] We have carried out neutron diffraction studies to reveal the magnetic structures for these phases. In this report we present the experimental results of neutron diffraction studies using powder and single crystal samples of Tb_7Rh_3.

§2. Experimental

Polycrystalline samples of Tb_7Rh_3 were prepared by arc-melting the constituent elements of Tb (99.9%) and Rh (99.96%) under a high purity argon atmosphere. The compounds were found to be single phase with the Th_7Fe_3 type hexagonal structure by powder X-ray diffraction. Single crystals were prepared by the Czochralski method from a single-phase polycrystalline sample using a tri-arc furnace. Single crystals were formed into rectangular shape and annealed at 400°C for 24 hours in an evacuated quartz tube. The crystal orientation was determined by the back reflection Laue method. Powder samples were also annealed at 400°C for 24 hours.

Magnetic susceptibility χ was measured by a vibrating sample magnetometer from 4.2 K to 300 K under the magnetic field of 11 kOe. Powder neutron diffraction experiments at wavelength of 2.462 Å were carried out in a double axis mode using neutron spectrometer HQR installed at JRR-3M in JAERI, Tokai, Japan. Neutron diffraction measurements for single crystals were made at wavelength of 1.006 Å in a double axis mode using neutron spectrometer KUR-TAS of the Research Reactor Institute, Kyoto University, Kumatori, Japan. The data collections were made in the $\boldsymbol{a}^*$-$\boldsymbol{c}^*$ reciprocal plane from 4.2 K to 110 K.

§3. Results and Discussion

Magnetic susceptibilities of Tb_7Rh_3 are shown in Fig. 1 as a function of temperature. The b-axis indicates the (1 2 0) direction. The result of magnetization measurements shows no anisotropy in the c-plane. Magnetic susceptibility along the b-axis increases with decreasing temperature, has a cusp at T_N and decreases. χ along the c-axis shows anomalies at around T_C and T_t. Magnetic transition temperatures were determined from ac magnetic susceptibility measurements. Since magnetic susceptibility along the b-axis is larger than that along the c-axis in paramagnetic region, magnetic moment in antiferromagnetic phase is considered to be aligned in the c-plane.

Figure 2 shows the neutron diffraction pattern at 1.5 K. The strong $(0\ 0\ 0)^{\pm}$ reflection indicates that Tb_7Rh_3 has an antiferromagnetic structure. Since a large $(0\ 0\ 0)^{\pm}$ reflection was also observed in the isostructural compound Nd_7Ni_3,[4] which has a helical structure, we assume that the Tb moment in the basal plane is helically modulated at 1.5 K. The propagation vector of this heli-

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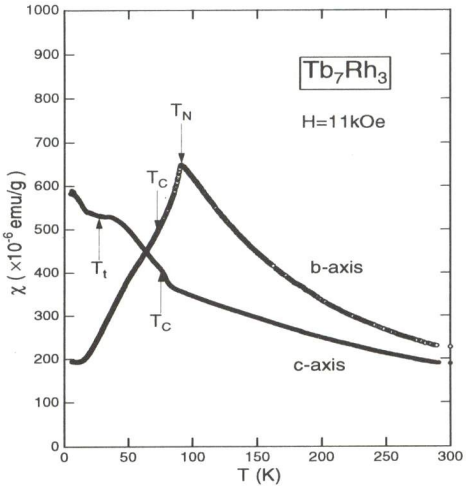

Fig. 1. Magnetic susceptibilities of Tb_7Rh_3 along the b- and c-axes as a function of temperature.

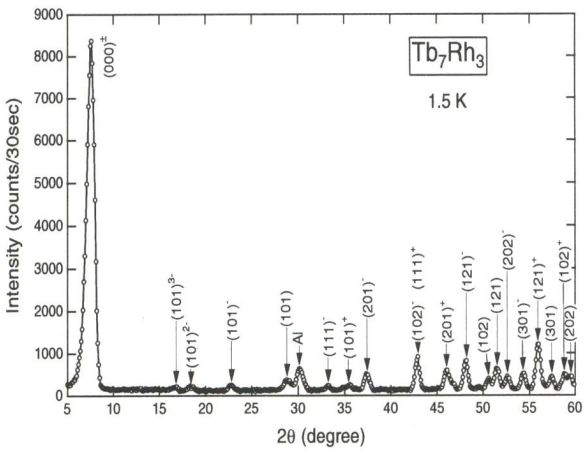

Fig. 2. A powder neutron diffraction pattern of Tb_7Rh_3 at 1.5 K.

cal component was determined to be $\boldsymbol{Q} = (0\ 0\ 1/3)$ from the position of $(0\ 0\ 0)^{\pm}$ reflection. All the peaks at 1.5 K can be indexed nuclear and magnetic Bragg reflections of Tb_7Rh_3 and Al reflections from the cryostat. Observed 2θ values are summarized in Table 1. A good agreement was obtained between the observed and calculated values of 2θ using this helical model. The variation of the diffraction pattern with temperature is shown in Fig. 3. Data are shifted upward for clarity. At 20 K, the same diffraction pattern as that at 1.5 K is observed. New diffraction peaks are observed at 40 K as the antiferromagnetic to ferrimagnetic phase transition. At 60 K, diffraction peaks below $2\theta = 45°$ are invisible except for the $(0\ 0\ 0)^{\pm}$ reflection. At 70 and 80 K, there is only a $(0\ 0\ 0)^{\pm}$ reflection as magnetic reflection below 60°. These results, indicate that the magnetic structure is modified between 40 and 70 K.

The schematic representation of the magnetic and nuclear Bragg spots in the $\boldsymbol{a}^*$-$\boldsymbol{c}^*$ reciprocal plane at 6 K is shown in Fig. 4. All observed magnetic spots can be indexed by the propagation vector $\boldsymbol{Q} = (0\ 0\ 1/3)$.

Tb_7Rh_3 has three non-equivalent Tb sites, in which

Table I. Comparison between observed and calculated values of 2θ at 1.5 K for magnetic Bragg reflections based on the c-axis helical model with $\boldsymbol{Q} = (0\ 0\ 1/3)$.

hkl	$2\theta_{obs.}$ (deg.)	$2\theta_{calc.}$ (deg.)	*diff.* (deg.)
$(000)^{\pm}$	7.51	7.63	-0.12
$(101)^{3-}$	16.66	16.69	0.03
$(101)^{2-}$	18.41	18.38	-0.03
$(101)^{-}$	22.78	22.71	-0.06
$(111)^{-}$	33.20	33.05	-0.15
$(101)^{+}$	35.44	35.31	-0.13
$(201)^{-}$	37.40	37.25	-0.14
$(201)^{+}$	46.09	46.40	0.31
$(102)^{-}$	42.83	42.58	-0.24
$(102)^{+}$	58.87	58.42	-0.44
$(121)^{-}$	48.12	47.97	-0.15
$(121)^{+}$	55.95	55.72	-0.22
$(301)^{-}$	54.33	54.19	-0.16
$(202)^{-}$	52.63	52.42	-0.21

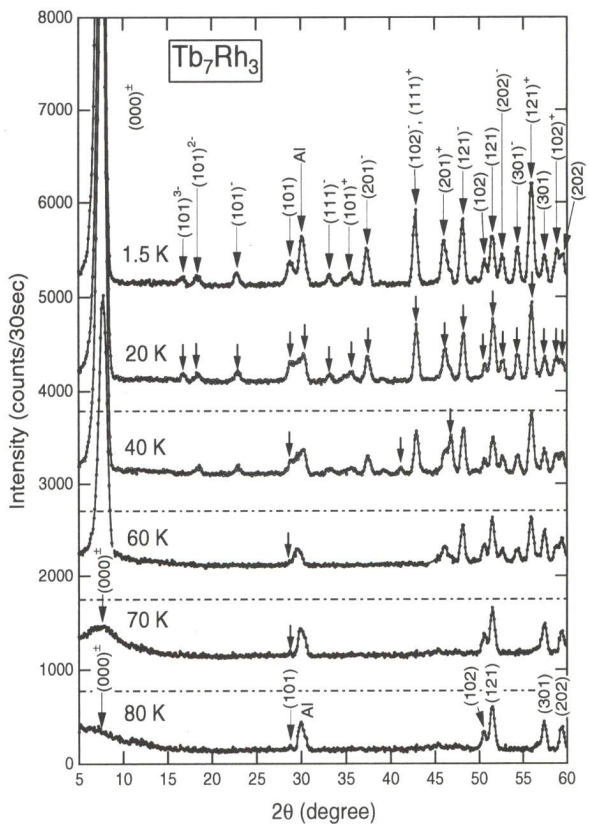

Fig. 3. Powder diffraction patterns of Tb_7Rh_3 at various temperatures.

the site Tb1 (two Tb atoms at 2(b)) has trigonal symmetry and both Tb2 and Tb3 sites (three Tb atoms at 6(c)) have a monoclinic symmetry. Since Tb1 and Tb3 layers are very close to each other, it is considered that one unit cell has four Tb layers along the c-axis. Considering the propagation vector $\boldsymbol{Q}$, magnetic unit cell consists of three crystallographic unite cells. Thus the rotation angle of magnetic moment per layer is estimated to be 30 degrees. Furthermore, we observed second and third harmonics of magnetic reflections in which the third harmonics are overlapped on the nuclear reflections. This

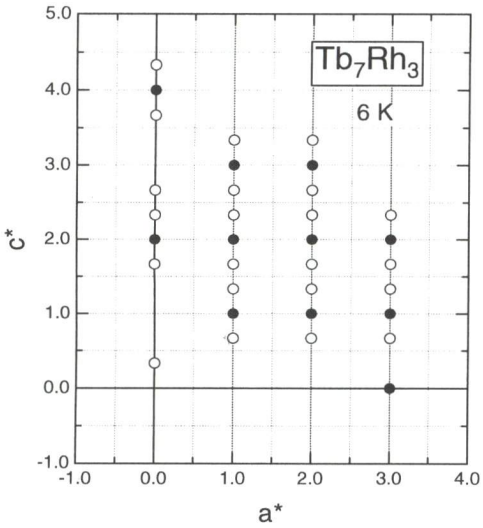

Fig. 4. Schematic representation of the magnetic and nuclear Bragg reflections in the a^* - c^* reciprocal plane at 6 K. Solid and open circles indicate nuclear and magnetic Bragg reflections, respectively.

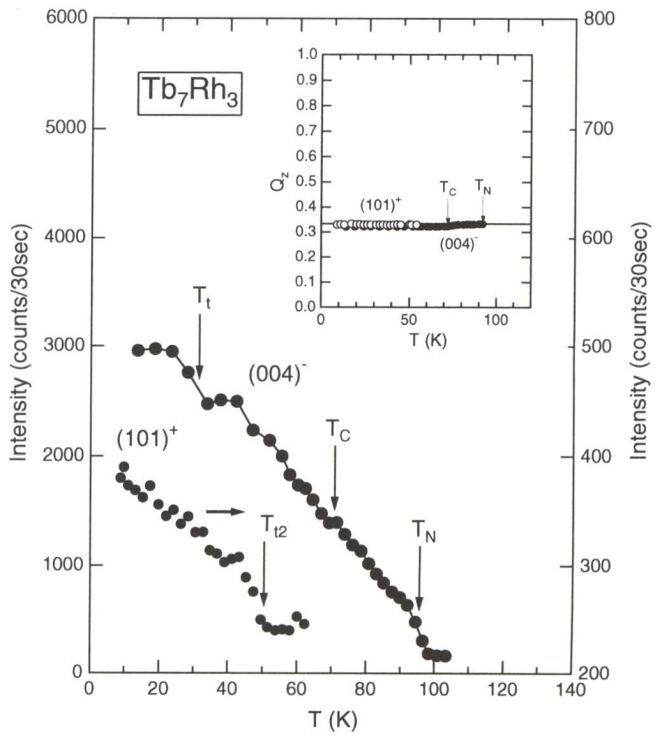

Fig. 5. The $(0\ 0\ 4)^-$ and $(1\ 0\ 1)^+$ peak intensity as a function of temperature. The inset shows the temperature dependence of Q_z.

suggests that the helical structure can be modulated along the c-axis. The component of propagation vector Q_z determined from the peak position of $(0\ 0\ 4)^-$ is 0.322 at 6 K.

Figure 5 shows the $(0\ 0\ 4)^-$ and $(1\ 0\ 1)^+$ peak intensity as a function of temperature. The $(0\ 0\ 4)^-$ peak intensity decreases with increasing temperature and vanishes at around T_N. Anomalies are observed at T_t and T_C, respectively. Between T_t and T_C, since we observed small spontaneous magnetization along the c-axis in the magnetization measurements, a conical structure in which magnetic moment has small inclination angle from c-plane, can be considered. On the other hand, the $(1\ 0\ 1)^+$ peak intensity vanishes at around 50 K which is consistent with the powder diffraction results. Since we have not observed any anomalies at this temperature in magnetic and electrical measurements, a kind of modification of the conical structure might occur at around 50 K. The inset shows the temperature dependence of the propagation vector component Q_z determined from the position of $(0\ 0\ 4)^-$ and $(1\ 0\ 1)^+$ peaks. Q_z is almost independent of temperature. Thus the helical and conical structures are commensurate ones in these magnetic phases. Though, we measured the temperature dependence of (3 0 0) peak intensity which indicates the component of magnetic moment along the c-axis, no pronounced change was observed at T_t and T_C. This is considered to be caused by the small spontaneous magnetization along the c-axis; 0.06 μ_B/Tb. In conclusion, we have studied the magnetic structure of Tb_7Rh_3 by neutron diffraction measurements. The basic magnetic structure of Tb_7Rh_3 is helical and conical one with the propagation vector along the c-axis; $\boldsymbol{Q} = (0\ 0\ 1/3)$. Magnetic structure can be modulated. In the ferrimagnetic phase, a kind of modification of magnetic structures occurs at around 50 K. To reveal the magnetic structure of this compound, more precise neutron diffraction and high field magnetization measurements using single crystals are now in progress.

Acknowledgments

This work was performed under the Visiting Research Program of Institute of Solid State Physics, University of Tokyo and Research Reactor Institute, Kyoto University.

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