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Dynamical Magnetic Properties of $Tb_2Ti_2O_7$

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Dynamical magnetic behavior of one of pyrochlore compounds, $Tb_2Ti_2O_7$ with strong magnetic frustration has been investigated down to the temperature $T \sim 1.5$ K by thermal and cold neutron experiments. The magnetic reflections can be divided into three components, elastic, quasi elastic and inelastic ones. The T- and scattering vector ($\boldsymbol{Q}$)-dependences of the intensity taken with cold neutrons at the elastic position of the spectrometer setting indicate that the correlation of the Tb^{3+} moments becomes appreciable with decreasing T at around ~30 K and grows significantly with further decreasing T down to 5 K. A sharp increase of the intensity observed around $\boldsymbol{Q}$=(0,0,2.1) with decreasing T from 5 K to 1.5 K suggests that large T-dependence exists even in this very low temperature region.

KEYWORDS: $Tb_2Ti_2O_7$, magnetic frustration, neutron scattering

§1. Introduction

$Tb_2Ti_2O_7$ has a pyrochlore type cubic structure (space group $Fd\bar{3}m$) consisting of two individual 3-dimensional networks of Tb_4 and Ti_4 tetrahedra[1)]. Because Tb moments ($\sim 9.4\mu_B$) has the antiferromagnetic nearest-neighbor interaction and do not form a bipartite lattice, the system is expected to have strong magnetic frustration. For this system, magnetic ordering is not observed down to 70 mK[2)], even though the Weiss temperature is estimated to be as high as ~19 K from the magnetic susceptibility data. We have studied the magnetic behabior by means of neutron scattering on a single crystal of $Tb_2Ti_2O_7$, where both thermal and cold neutrons have been used. In the experiments, elastic, quasi elastic and inelastic components of the scattering have been observed. The data of the inelastic scattering taken by thermal neutrons have been reported in ref. 3. In this paper, we mainly report the results of the quasi elastic components.

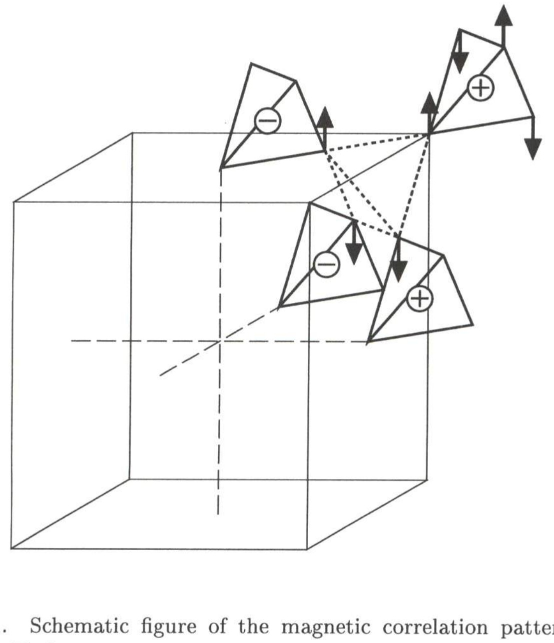

Fig. 1. Schematic figure of the magnetic correlation pattern of $Tb_2Ti_2O_7$.

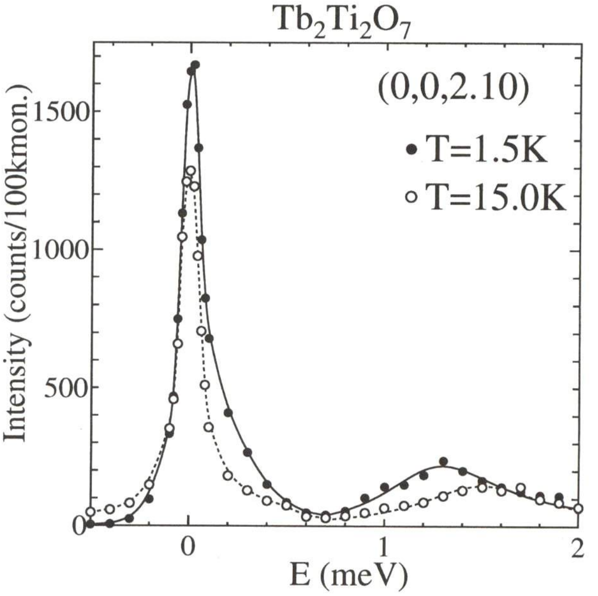

Fig. 2. Examples of the scattering profiles observed for a single crystal of $Tb_2Ti_2O_7$ at $\boldsymbol{Q}$=(0,0,2.1).

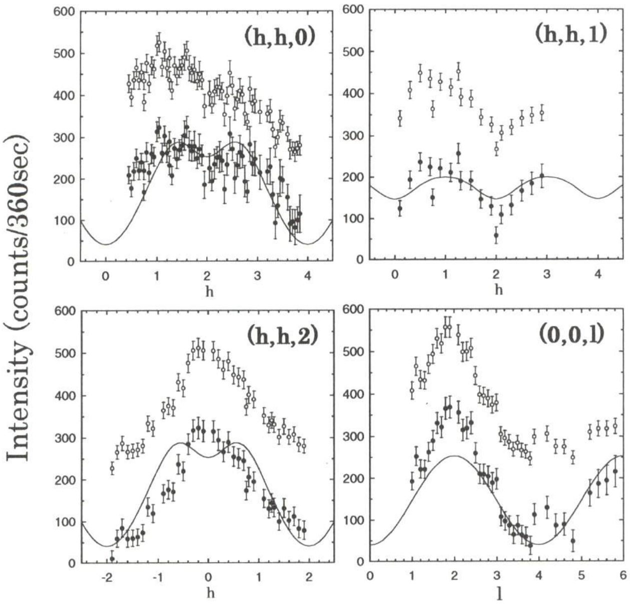

Fig. 3. $\boldsymbol{Q}$-dependence of the magnetic scattering intensity observed at 3.5 K with the spectrometer setting for E=0 meV. Values obtained by subtracting the background counts (which include the nuclear incoherent ones) from the raw data (Open circles), are divided by the square of the magnetic form factor of Tb^{3+} and the results are plotted by the closed circles. The solid lines show the $\boldsymbol{Q}$-dependence of the magnetic intensities calculated by using the correlation pattern similar to that of Fig. 1. See text for details.

§2. Experiments

A single crystal of $Tb_2Ti_2O_7$ with a volume of $\sim$1 cm^3 was prepared by the floating zone (FZ) method. Neutron measurements were carried out by using the triple axis spectrometer at T1-1 of the thermal guide and C1-1 of the cold guide of JRR-3M of JAERI in Tokai. The crystal was oriented with the $[1\bar{1}0]$ direction vertical, where the points (h,h,l) in the reciprocal space can be reached. The 002 reflections of Pyrolytic graphite (PG) were used for both the monochromator and the analyzer. In the measurements at T1-1, the horizontal collimations were 12′(effective)-open-40′-40′ and the initial neutron energy E_i was fixed at 13.57 meV. The full width at half maximum of the energy resolution ΔE determined by the incoherent elastic scattering was about 0.7 meV. In the measurements at C1-1, the horizontal collimations in front of the sample were 12′(effective)-20′ and after the sample were 420′-open, where a horizontally focusing analyzer was used. The final neutron energy E_f was 3.10 meV. The energy resolution ΔE was about 0.08 meV.

§3. Results and Discussion

First, we carried out the measurements at T1-1, where the elastic and quasi elastic components could not be separated. Inelastic magnetic scattering peaks have been observed at energies E=1$\sim$2 meV at 3.5 K at various $\boldsymbol{Q}$-points in the reciprocal space. The dispersion curves are determined along several $\boldsymbol{Q}$-directions as reported in ref. 3. The excitation energy ω_Q is nearly $\boldsymbol{Q}$-independent ($\omega_Q \cong \omega_0$) at T higher than $\sim$30 K, where no appreciable correlation of the Tb-moments exists. As T decreases, the dispersion of ω_Q becomes significant. The peak intensity becomes maximum at $\boldsymbol{Q}=\boldsymbol{Q}_m$ where the peak energy becomes minimum. Because the energy dispersion at 3.5 K is comparable to ω_0, we presume that Heisenberg-like exchange interaction and the anisotropy energy of the system are comparable.

From the distribution of the scattering intensity at the energy E=0.8 meV in the $\boldsymbol{Q}$ space, the pattern of the spatial magnetic correlation was determined as shown in Fig.1 by assuming that the peak is acoustic-like. The spins within a tetrahedron are aligned antiferromagnetically with a collinear structure. The inter-tetrahedra correlation is shown in the figure by the signs. In the analyses, the correlation length was roughly estimated to be the nearest neighbor distance between the Tb moments. However, because the $\boldsymbol{Q}$-dependence of the intensity at E=0.8 meV is affected by that of ω_Q, the estimation may not be accurate. Then, we have performed the measurements at C1-1 with the energy resolution ΔE~0.08 meV.

Figure 2 shows the examples of the energy profiles taken at C1-1 with cold neutrons at $\boldsymbol{Q}$=(0,0,2.1). Both the elastic and quasi elastic components can be observed around E=0 meV and inelastic scattering peaks are separetely observed at E ~1.3 meV and E ~1.5 meV at 1.5 K and 15 K, respectively.

Based on this information on the spectral distribution of the magnetic excitation, we can find that the data taken at 3.5 K at T1-1 with the spectrometer setting of E=0 meV (with the energy resolution ΔE~0.7 meV) and shown in Fig. 3, contain both the elastic and quasi elastic components. We try to use the data in Fig. 3 to determine the correlation pattern of the Tb-moments. In the figure, the raw data are shown by open circles. Because at H=4.5 T, the Tb moments are almost completely aligned, the magnetic scattering intensity can be considered not to exist except the magnetic Bragg points[5], we can roughly determine the background value. The

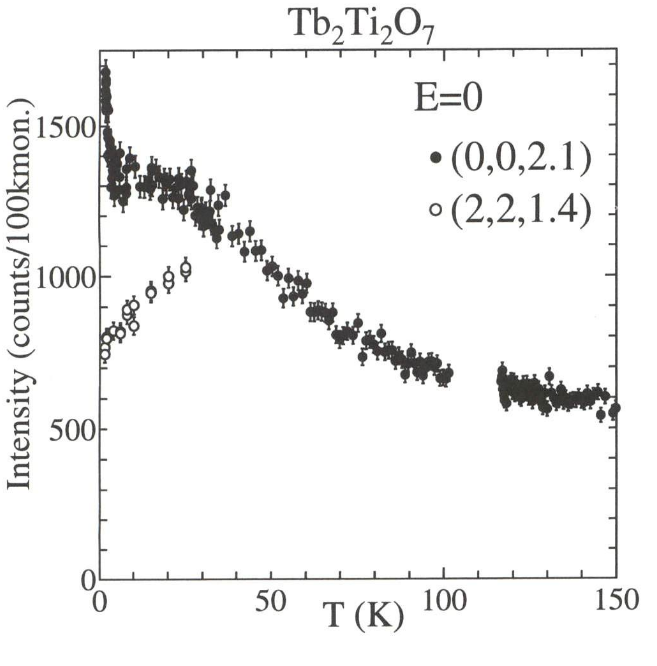

Fig. 4. T-dependence of the scattering intensity at $\boldsymbol{Q}$=(0,0,2.1) and (2,2,1.4) with the elastic position of the spectrometer setting.

net values are obtained by subtracting this background from the raw data.

Then, dividing the net values by the square of the magnetic form factor of Tb^{3+} [4] results shown by the closed circles are obtained. The solid lines show the $\boldsymbol{Q}$-dependence of the scattering intensity calculated by using the correlation pattern similar to that shown in Fig. 1, but with smaller correlation length (The nearest neighbor correlation of the moments m is now of the order of $\sim m^2/4$). The observed data can roughly be explained by the calculation except around $\boldsymbol{Q}$=(0,0,2), where the sharp increase of the intensity at E=0 meV exists with decreasing T below 5 K as shown in Fig. 4.

In the figure, the T-dependence of the scattering intensities at $\boldsymbol{Q}$=(0,0,2.1) and (2,2,1.4) measured at E=0 meV (with the energy resolution of 0.08 meV) are shown . The intensities increase with decreasing temperature down to 30 K in an almost $\boldsymbol{Q}$-independent manner. Below 30 K, the difference of the observed T-dependences between different $\boldsymbol{Q}$-points becomes significant. These results may be understood as follows. As T decreases, the Tb moments tend to be gradually pinned to their local principal axis or the easy axis, and the motion may be become slow, resulting in the increase of the scattering intensity around E=0 meV. Below 30 K, the short range correlation of the moment directions grows, which induces the $\boldsymbol{Q}$-dependence of the scattering intensity. The sharp upturn of the scattering intensity observed below 5 K with decreasing T at $\boldsymbol{Q}$=(0,0,2.1) seems to indicate a change of the correlation pattern even in the region of very low temperatures, much lower than the Weiss temperature of ~19 K estimated for the present system. It may be one of the characteristics of the present frustrating magnetic sysyetm.

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