Quantum spin fluctuations in the spin liquid state of $Tb_2Ti_2O_7$

H. Kadowaki[1], H. Takatsu[1], T. Taniguchi[1], B. Fåk[2], J. Ollivier[3]
[1] *Department of Physics, Tokyo Metropolitan University, Hachioji-shi, Tokyo 192-0397*
[2] *SPSMS, UMR-E CEA/UJF-Grenoble-1, INAC, F-38054 Grenoble Cedex 9, France*
[3] *Institute Laue Langevin, BP156, F-38042 Grenoble, France*

Magnetic systems with geometric frustration have been intensively studied experimentally and theoretically for decades. Spin systems on triangular, kagome, spinel, and pyrochlore [1] lattices play major roles in these studies. Their rich variety of phenomena includes the zero temperature entropy of spin-ice, multiferroics induced by non-collinear magnetic structures, heavy fermion behavior, and unconventional anomalous Hall effect. Subjects that have fascinated many investigators in recent years are classical and quantum spin-liquid states [2,3], where conventional long-range order (LRO) is suppressed to very low temperatures. Quantum spin-liquids in particular have been challenging both theoretically and experimentally since the proposal of the resonating valence-bond state [2].

Among magnetic pyrochlore oxides [1], $Tb_2Ti_2O_7$ has attracted much attention because it does not show any conventional magnetic LRO down to 50 mK and remains dynamic with short range correlations [4,5]. Theoretical considerations of the crystal-field (CF) states of Tb^{3+} and exchange and dipolar interactions of the system showed that it should undergo a transition into a magnetic LRO state at about $T \sim 1.8$ K [6]. Thus the dynamical ground state is a candidate for a quantum spin-liquid [3], but its puzzling origin has been in debate [1]. An interesting theory proposed is that $Tb_2Ti_2O_7$ is quantum spin ice, where a ground state is formed from a symmetrized "2-in, 2-out" classical spin-ice states [7]. Another single-site mechanism was proposed to account for the absence of LRO, in which the CEF ground doublet becomes two singlets owing to a conjectured tetragonal distortion [8].

Several experimental puzzles of $Tb_2Ti_2O_7$ originate from the difficulty of controlling the quality of single crystalline samples, resulting in strongly sample-dependent specific-heat anomalies at temperatures below 2 K [9,10,11]. In contrast, experimental results on polycrystalline samples are more consistent [4,5,9]. Among the experimental results reported to date, an important clue to solve the puzzles of $Tb_2Ti_2O_7$ seems to be a change of state at about 0.4 K suggested by specific heat [9], inelastic neutron scattering [9], and neutron spin echo [5] measurements on polycrystalline samples. At this temperature, a few single-crystalline samples show a peak in the specific heat suggesting a phase transition [11,12], an issue that has not been pursued seriously.

We have investigated the hypothesis that the non-stoichiometry x of $Tb_{2+x}Ti_{2-x}O_{7+y}$ is a tuning parameter for a quantum critical point separating a LRO state from a spin liquid state [13]. We have therefore performed specific heat, magnetization, and neutron scattering experiments on polycrystalline samples of $Tb_{2+x}Ti_{2-x}O_{7+y}$ with different values of x [13]. We find that a minute change of x brings about a systematic change of the specific heat. The ground state goes from LRO with an unknown (hidden) order parameter for $x > x_c$ to a spin liquid for $x < x_c$ [13]. The present inelastic neutron scattering measurements were carried out on the time-of-flight spectrometer IN5 operated with $\lambda =$ 5 and 10 Å at ILL. Samples of $x = 0.005$ and -0.005 with weights of 9 g were mounted in a dilution refrigerator.

To study the spectral change of the magnetic excitations through T_c, we performed inelastic neutron scattering measurements

using the spectrometer IN5 with an energy resolution of $\Delta E = 0.012$ meV (FWHM) [13], which is 5 times better than that in our previous study [9]. Figure 1 shows the temperature dependence of an energy spectrum for the $x = 0.005$ sample at $Q = 0.6$ Å^{-1} [13]. It is evident that the spectrum changes from a continuum ($T > T_c$) to a peaked structure at 0.1 meV ($T < T_c$). The excitation peak at $T \ll T_c$ is weakly Q-dependent, which may possibly be interpreted as a splitting of the CEF ground-state doublet. An energy spectrum of the $x = -0.005$ sample is also shown in Fig. 1 for comparison. Its spectral shape can be approximately expressed by a Lorentzian squared $\mathrm{Im}\chi(E,Q)/E \propto [(\sqrt{2}-1)E^2 + \Gamma^2]^{-2}$ with $\Gamma = 0.1$ meV (HWHM) in $-0.05 < E < 0.3$ meV, revealing quantum spin fluctuations with the same energy scale of 0.1 meV as that of the $x = 0.005$ sample.

The high sensitivity of IN5 enabled us to observe a small Bragg peak for the $x = 0.005$ sample. In the inset of Fig. 1, the intensity of the elastic scattering for $|E| < 0.005$ meV is plotted as a function of Q. Below T_c, a clear Bragg peak at $Q = 0.54$ Å^{-1} is observed, which can be indexed as $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$. The Q-width of this peak is somewhat larger than the instrumental Q-resolution, and corresponds to a correlation length of the order of 100 Å. Although this peak could be of a nuclear (structural) origin, it is more likely an antiferromagnetic (AFM) reflection. In fact, two recent neutron scattering experiments carried out on single-crystalline samples of $Tb_2Ti_2O_7$ showed magnetic short-range order around the same $\mathbf{Q} = (\frac{1}{2}\frac{1}{2}\frac{1}{2})$ [14]. A roughly estimated ordered moment for the $x = 0.005$ sample is 0.08 μ_B at 0.1 K. This ordered moment is much smaller than the magnetic moment ~ 5 μ_B of the ground doublets, which implies that most of the spin fluctuations persist below T_c. In contrast, the entropy change around $T_c = 0.5$ K is $S(T = 0.55) - S(T = 0.38) \simeq 0.25R \ln 2$ [13], which is significant. These probably indicate that there is a major order parameter, which is unknown at present.

Additional experiments and analyses of full IN5 spectra are being performed and will be published elsewhere in the near future. The hidden order parameter of $Tb_{2+x}Ti_{2-x}O_{7+y}$ will be hopefully elucidate by these analyses.

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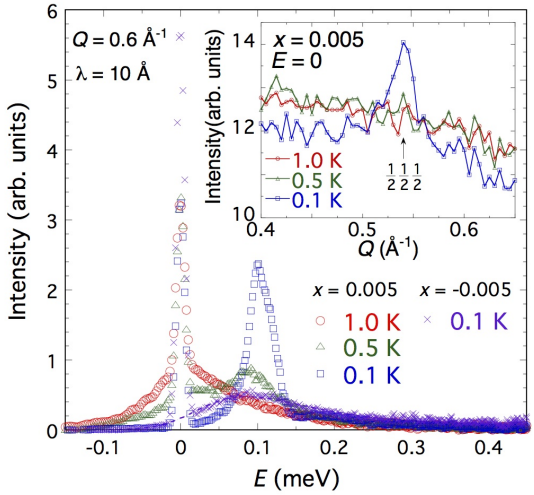

Fig. 1. Energy spectra of polycrystalline Tb2+xTi2-xO7+y with x = 0.005 and -0.005 [13]. The inset shows the Q dependence of the elastic scattering for the x = 0.005 sample. The dashed line is the fit curve.