Experimental report

04/06/2017

Proposal: **4-05-635** **Council:** 4/2016

Title: Quadrupole order of Tb$_{2+x}$Ti$_{2-x}$O$_{7+y}$

Research area: Physics

This proposal is a continuation of 4-05-628

Main proposer: **Bjorn FAK**

Experimental team: Bjorn FAK
Jacques OLLIVIER
Hiroaki KADOWAKI

Local contacts: Jacques OLLIVIER

Samples: Tb2+xTi2-xO7+y

Instrument	Requested days	Allocated days	From	To
IN5	5	5	15/06/2016	21/06/2016

Abstract:

We argue that the frustrated pyrochlore magnet Tb2+xTi2-xO7+y (TTO) has a quantum phase transition between a quantum spin liquid (QSL) state and a hidden order -- probably quadrupolar -- state, as a function of the off-stoichiometry x. The QSL state, referred to as "quantum spin ice", is characterized by an emergent U(1) field and excitations in form of gapped bosonic spinons and gapless photons. This state was recently measured on IN5 using a high-quality single crystal assembly, showing short-range spin-fluctuations that characterize the QSL state. We now propose to study the mixed spinwave-quadrupolar excitations expected in the hidden-order phase using another high-quality crystal assembly on IN5. The data will be analyzed using a mean-field random-phase approximation incorporating planar (anti-)ferropseudospin states. The existence of such excitations would be compelling evidence for quadrupole ordering, and hence provide strong evidence for our conjecture.

Geometrically frustrated magnets have been actively studied in recent years [1]. In particular, pyrochlore magnets [2] showing spin ice behavior [3] have interesting features such as finite zero-point entropy and emergent magnetic monopole excitations. A quantum spin-liquid state (QSL) is theoretically predicted for certain spin-ice like systems [4, 5], where transverse spin interactions transform the classical spin ice into a QSL. This quantum spin ice (QSI), or U(1) quantum spin liquid, is characterized by an emergent U(1) gauge field and by excitations in form of gapped bosonic spinons and gapless photons [4, 5]. By modifying the interactions in some way, the system undergoes a quantum phase transition to a long range ordered (LRO) state of transverse spin or pseudospin.

Among magnetic pyrochlore oxides [2], $Tb_2Ti_2O_7$ (TTO) has attracted much attention, because magnetic moments remain dynamic with short range correlations down to 50 mK [6]. Since TTO has been thought to be located close to the classical spin ice, although clear experimental evidence is still missing, the dynamical low-T behavior of TTO could be ascribed to QSI [5]. Inspired by this intriguing idea, a lot of experimental studies of TTO have been performed, e.g. [7, 8]. However, the interpretation of experimental data remains very difficult [5, 9], yet interesting, partly owing to strong sample dependence [7]. Among these studies, our investigation of polycrystalline $Tb_{2+x}Ti_{2-x}O_{7+y}$ showed that a very small change of x induces a quantum phase transition between the dynamical ground state ($x < x_c = -0.0025$) and a LRO state with a hidden order parameter ($x > x_c$) [8]. We think it is important to clarify the origin of this order parameter, which fluctuates in the spin liquid state ($x < x_c$). Based on theoretical considerations of the crystal-field (CF) states of non-Kramers magnetic ions (including Tb^{3+}) in the pyrochlore structure together with their superexchange interaction [10], a possible answer to the problem of $Tb_{2+x}Ti_{2-x}O_{7+y}$ is an electric multipole (or quadrupole) ordering and a U(1) QSL state [11].

The experimental difficulty of TTO comes from controlling the quality of large crystalline samples for neutron scattering, especially from composition (x) gradient of TTO crystal rods grown by the FZ technique [12]. By using a small crystal with $x \simeq 0.005$, which exhibits a well-defined T_c of 0.53 K, we performed specific heat and magnetization experiments. We analyzed these data together with the magnetic excitation spectra of a polycrystalline sample with $x = 0.005$, and showed that the phase transition at T_c is ascribable to an electric quadrupole order (QO) [11].

By selecting low x-gradient parts of single-crystalline rods, we made a multi-crystal sample in the QO range ($x \simeq 0.001$) with a good mosaicity for the present experiment. Although this QO sample is of high quality, due to the poor thermal conductivity inherent to the multi-crystal sample mount, the cooling of the sample was very slow, taking about 1.5 days between $T = 2$ to 0.1 K. Measurements were performed at two temperatures, $T = 0.1$ and 0.7 K, at a wavelength of $\lambda = 8$ Å, making standard rocking scans with a step of 1°. The main result of the measurements is the existence of excitation with a weakly Q-dependent dispersion relation at about 0.1 meV and excitation continuuum above this dispersion curve. A typical E-Q cut along a (1,1,1) direction measured at 0.1 K is shown in Fig. 1. By assuming the pseudospin-1/2 effective Hamiltonian proposed in Ref. [11], we tried to calculate excitation spectra of the dispersion curve using MF-RPA described in Ref. [13]. A result is shown in Fig. 2 for the (1,1,1) direction, which suggests that MF-RPA can reproduce the observed weakly Q-dependent dispersion relation. Detailed analyses to reveal the QO state are underway.

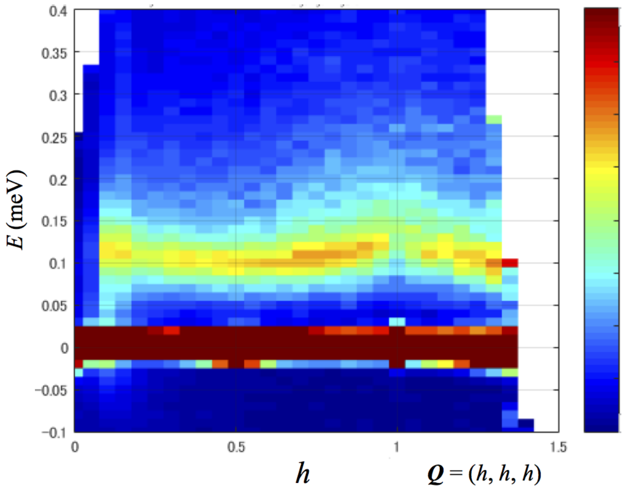

Fig.1 Observed excitation spectra S(**Q**, *E*) along (1,1,1).

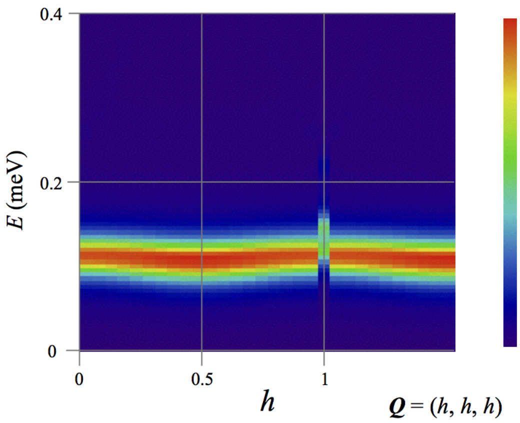

Fig.2 Calculated excitation spectra S(**Q**, *E*) along (1,1,1) using MF-RPA.

References

[1] C. Lacroix, P. Mendels, F. Mila (eds.), Introduction to Frustrated Magnetism (Springer, Berlin, Heidelberg, 2011).

[2] J. S. Gardner et al., Rev. Mod. Phys. 82, 53 (2010).

[3] S. T. Bramwell and M. J. P. Gingras, Science 294, 1495 (2001).

[4] M. Hermele et al., Phys. Rev. B 69, 064404 (2004).

[5] M. J. P. Gingras and P. A. McClarty, Rep. Prog. Phys. 77, 056501 (2014).

[6] J.S. Gardner et al., Phys. Rev. Lett. 82, 1012 (1999).

[7] H. Takatsu et al., J. Phys.: Condens. Matter 24, 052201 (2012).

[8] T. Taniguchi, H. Kadowaki, H. Takatsu, B. Fåk, J. Ollivier et al., Phys. Rev. B 87, 060408(R) (2013).

[9] S. Petit et al., EPJ Web of Conferences 83, 03012 (2015).

[10] S. Onoda and Y. Tanaka, Phys. Rev. B 83, 094411 (2011).

[11] H. Takatsu, S. Onoda, S. Kittaka, A. Kasahara, Y. Kono, T. Sakakibara, Y. Kato, B. Fåk, J. Ollivier, J.W. Lynn, T. Taniguchi, M. Wakita, H. Kadowaki, Phys. Rev. Lett. 116, 217201 (2016).

[12] M. Wakita, T. Taniguchi, H. Edamoto, H. Takatsu, H. Kadowaki, J. Phys.: Conf. Series 683, 012023 (2016).

[13] H. Kadowaki, H. Takatsu, T. Taniguchi, B. Fåk, J. Ollivier, SPIN 5, 1540003 (2015).