Experimental report

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Proposal: **4-05-628**

Council: 4/2015

Title: Quadrupole order of Tb_{2+x}Ti_{2-x}O_{7+y}

Research area: Physics

This proposal is a new proposal

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Samples: Tb2Ti2O7

Instrument	Requested days	Allocated days	From	To
IN3	2	2	14/11/2015	16/11/2015
IN5	7	7	17/11/2015	24/11/2015

Abstract:

This proposal is a part of an investigation of U(1) spin liquid states, quadrupole ordering, and their quantum phase transition in a frustrated pyrochlore magnet Tb_{2+x}Ti_{2-x}O_{7+y}. In spin ices, finite zero-point entropy and magnetic monopole excitations have attracted much attention. Spin ices are well represented by a classical Ising Hamiltonian. When spin-flip terms are added to this spin-ice Hamiltonian, very interesting quantum many-body effects emerge from the macroscopically degenerate ground state. This is referred to as "quantum spin ice", in which the phase angles of the spinors become correlated in such a way that a U(1) gauge field emerges. In the U(1) spin liquid state, the U(1) field is fluctuating down to T = 0, where gapped spinon and gapless photon excitations are predicted. By changing interaction parameters, the system undergoes a phase transition to a LRO states. A simple pseudo-spin ordering is the putative quadrupole ordering of Tb_{2+x}Ti_{2-x}O_{7+y}. The purpose of this proposal is to get convincing evidence for this quadrupole ordering using inelastic neutron scattering.

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 [4]. A quantum spin-liquid state (QSL) is theoretically predicted for certain spin-ice like systems [5–8], where transverse spin interactions drive 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 [5–8]. 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, in analogy to a Higgs transition [6,7].

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 [9]. 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 [10]. Inspired by this intriguing idea, numerous experimental studies of TTO have been performed [11–13]. However, the interpretation of experimental data remains very difficult [8,14], yet interesting, partly owing to strong sample dependence [11]. 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 a dynamical ground state ($x < x_c = -0.0025$) and a LRO state with a hidden order parameter ($x > x_c$) [12]. It is important to clarify the origin of this order parameter, which becomes dynamical in the spin liquid state ($x < x_c$). Based on theoretical considerations of the crystal-field (CF) states of non-Kramers magnetic ions in the pyrochlore structure together with their superexchange interaction [7,15], 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 [16–18].

The experimental difficulty of TTO comes from controlling the quality of large crystalline samples for neutron scattering [18]. 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 [16]. 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 LRO [16,17]. Very recently, we showed that a single-crystalline rod of $Tb_{2+x}Ti_{2-x}O_{7+y}$ grown by the standard FZ technique has a composition (x) gradient, which gives rise to the inhomogeneity problem [18]. By selecting low x-gradient parts of single-crystalline rods, we have made two multi-crystal samples, one in the QSL phase with $x \simeq -0.007$, and one in the hidden order phase with $0 < x < 0.005$, i.e. $x > x_c$. The initial idea of the proposal on IN5 was to measure the hidden-order phase, but due to experimental circumstances, the QSL crystal was used for the measurements.

The crystal assembly had a good mosaicity and was readily aligned on IN5. However, 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°. A complementary measurement was performed at $\lambda = 4.8$ Å to allow comparison with previous measurements. The main results of the measurements is the existence of short-range spin fluctuations around $\boldsymbol{Q} = (1/2, 1/2, 1/2)$, which are characteristic for the QSL state. The temperature dependence of these spin fluctuations shows that the QSL state is developed gradually below 0.4 K (Fig. 1). Detailed analyses to reveal the mechanism of the QSL state are underway.

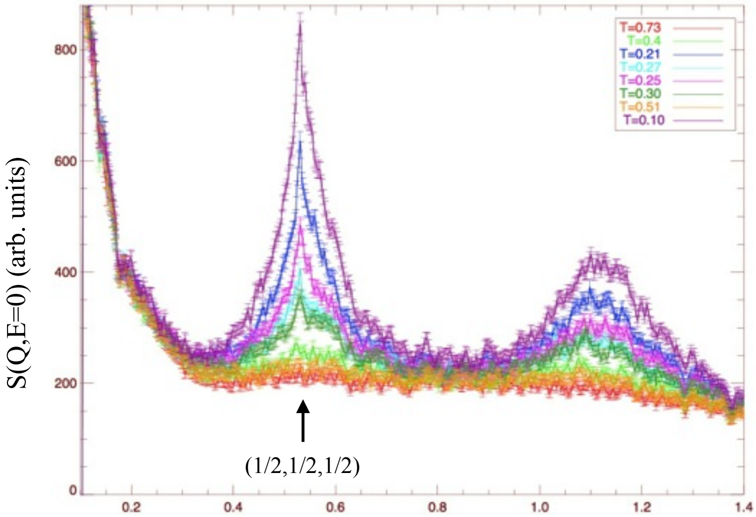

Fig. 1: Temperature dependence of $S(Q, E = 0)$ through $\mathbf{Q} = (1/2, 1/2, 1/2)$.

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