Quadrupole Order in the Frustrated Pyrochlore $Tb_{2+x}Ti_{2-x}O_{7+y}$

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Geometrically frustrated magnets have been actively investigated in condensed matter physics [1]. In particular, spin ice (SI), e.g. $R_2Ti_2O_7$ (R = Dy or Ho) [2], provides prototypical frustrated Ising magnets with the pyrochlore lattice structure [3], consisting of a three-dimensional network of corner-sharing tetrahedra. It displays fascinating features such as a finite zero-point entropy and thermally excited emergent magnetic or SI monopoles [2]. An intriguing theoretical proposal for a U(1) quantum spin liquid (QSL) state, has been made for variants of SI endowed with quantum spin fluctuations [4]. The U(1) QSL state is characterized by an emergent U(1) gauge field producing gapless fictitious photons and by gapped bosonic spinon excitations carrying the SI magnetic monopole charge [4]. By increasing the transverse interaction, the system can undergo a phase transition from the U(1) QSL to a long range ordered (LRO) state [4], being described by a Higgs transition [5] of transverse spins or pseudospins representing electric-quadrupole moments [4].

In a quest to QSL states in frustrated magnetic systems [6], an Ising-like pyrochlore $Tb_2Ti_2O_7$ (TTO) is a potential candidate for a U(1) QSL: it has been reported to remain in a fluctuating spin state down to 50 mK without magnetic LRO [7]. However, the origin of this spin liquid state of TTO has been elusive for more than a decade despite many investigations, and is still under hot debate [8]. To solve this challenging problem of TTO, we start this investigation by postulating that the theoretically-proposed electronic-coupling [9] between electric quadrupole moments of non-Kramers ions including Tb^{3+} is at work for giving the quantum fluctuations to TTO. This postulation is a natural consequence of the previous unsuccessful trial-and-errors of explaining TTO by taking into account only the interactions between magnetic dipole moments and the perturbation through first excited crystal-field (CF) states, and by using another assumption of a Jahn-Teller (JT) effect. Under the present postulation, two ground states of off-stoichiometric $Tb_{2+x}Ti_{2-x}O_{7+y}$ samples [10] can be accounted for by the U(1) QSL ($x < x_c$) and electric quadrupolar ($x > x_c$) states.

We investigate the hidden order of $Tb_{2+x}Ti_{2-x}O_{7+y}$ ($x = 0.005 > x_c$), because the electric quadrupolar order is more tractable than the U(1) QSL by using semi-classical theoretical analyses. Specific heat, magnetization, and neutron scattering experiments were performed, and these experimental data were analyzed using quantum and classical Monte Carlo (QMC, CMC) simulations, and a mean-field random-phase approximation (MF-RPA). The results demonstrate that the hidden order is an electric quadrupolar order [Fig. 1(b)] and that the parameters of the model Hamiltonian is located close to a phase boundary between the electric quadrupole and U(1) QSL states [Fig. 1(a)], which suggests that the elusive spin-liquid state of TTO is the U(1) QSL. We emphasize that a high-quality single-crystalline sample with a well-controlled x value enables us to accomplish this work. Readers are referred to Refs. [11,12] for details.

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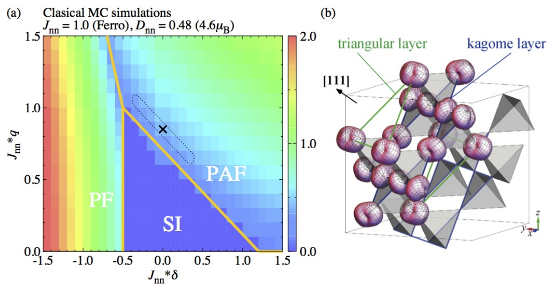

Fig. 1. (a) Phase diagram of the effective Hamiltonian determined from CMC simulations [11]. (b) Schematic view of the deformation of the f-electron charge density due to the PAF order on the pyrochlore lattice [11].