Composite spin and quadrupole wave in the ordered phase of $Tb_{2+x}Ti_{2-x}O_{7+y}$

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Geometrically frustrated magnets have been actively studied in recent years [1]. In particular, pyrochlore magnets [2] showing spin ice behavior have interesting features such as finite zero-point entropy and emergent magnetic monopole excitations. A quantum spin-liquid state is theoretically predicted for certain spin-ice like systems [3], where transverse spin interactions transform the classical spin ice into quantum spin liquid. This quantum spin ice (QSI), or U(1) quantum spin liquid, is characterized by an emergent U(1) gauge field fluctuating down to $T = 0$ and by excitations of gapped bosonic spinons and gapless photons [3]. By changing the interactions of the QSI in some ways the system undergoes a quantum phase transition to long range ordered (LRO) states of transverse spin or pseudospin, being interpreted as Higgs phases [3]. Experimental investigations of the U(1) quantum spin liquid and neighboring LRO states have been challenged by several groups [4]. However it is difficult to characterize the quantum spin liquid states, which preclude standard techniques of observing magnetic Bragg reflections and magnons.

Among magnetic pyrochlore oxides [2], $R_2Ti_2O_7$ (R = Dy, Ho) are the well-known classical Ising spin-ice examples. A similar system $Tb_2Ti_2O_7$ (TTO) has attracted much attention, because magnetic moments remain dynamic with short range correlations down to 50 mK [5]. Since TTO has been thought to be close to the classical spin ice, the low-temperature dynamical behavior of TTO could be attributed to QSI [6]. Inspired by this intriguing idea, many experimental studies of TTO have been performed to date [7] (and references in Refs. [8]). However the interpretation of experimental data has been a conundrum [8], partly owing to strong sample dependence [9]. Among these studies, our investigation [9] 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 spin-liquid state ($x < -0.0025 = x_c$) and a LRO state with a hidden order parameter ($x_c < x$). It is important to clarify the origin of this order parameter, which becomes dynamical in the spin-liquid state ($x < x_c$).

In this work, we try to reformulate the problem of TTO and to reinterpret its puzzling experimental data based on the theoretically predicted [10] electronic superexchange interactions. A novel ingredient of these interactions is the Onoda-type coupling [10] between neighboring electric quadrupole moments of non-Kramers Tb^{3+} ions. The theory [10] proposes an effective pseudospin-1/2 Hamiltonian described by the Pauli matrices representing both magnetic-dipole and electric-quadrupole moments. Depending on the parameters of the Hamiltonian there are two electric quadrupole ordering phases, which are candidates for the hidden order of TTO. These electric quadrupolar orders do not bring about observable magnetic Bragg peaks. However, these orders can be detected by their elementary excitations (inelastic magnetic scattering), and by proper interpretation using a linear spin-wave theory.

In our recent paper [11], starting from the crystal-field (CF) ground state doublet of TTO, we account for its single-site electric quadrupole moments, their LRO, and pseudospin wave excitations in the electric quadrupole LRO. A standard linear spin-wave theory predicts that the pseudospin wave in the electric quadrupole LRO is, in reality, a composite wave of magnetic-dipole and electric-quadrupole moments. We discuss this possibility for $Tb_{2+x}Ti_{2-x}O_{7+y}$ using previously observed [9] low-energy magnetic excitation spectra of a polycrystalline sample with $x = 0.005$ ($T_c = 0.5$ K). The neutron scattering experiment was performed on the time-of-flight spectrometer ILL-IN5 operated with $\lambda = 10$ Å. Fig. 1(b) shows Q-dependent powder spectra taken at $T = 0.1$ K. These data should be compared with powder averaging of the magnetic $S(Q,E)$. Fig. 1(a) shows an example of this powder averaged $S(|Q|,E)$ choosing the parameters $J_{nn} = 1$ K, $q = 0.8$, and $\delta = 0$, which are in the PAF phase. We think that these figures show reasonably good agreement between the calculation and the observation. In this scenario, the hidden order in $Tb_{2+x}Ti_{2-x}O_{7+y}$ samples with $x > x_c$ is an electric quadrupolar LRO, and that $Tb_{2+x}Ti_{2-x}O_{7+y}$ samples with $x < x_c$ are in the U(1) quantum spin-liquid phase. Readers are referred to Refs. [11,12] for details.

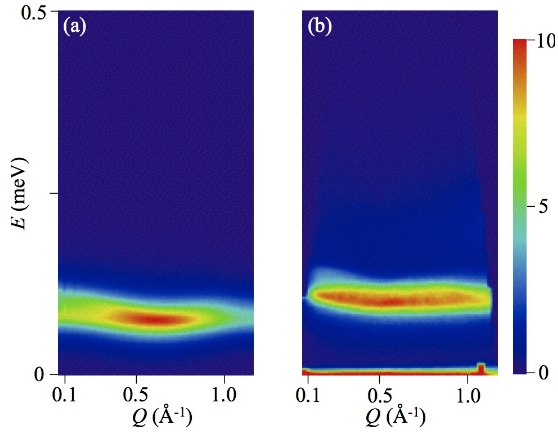

Fig. 1. (a) Powder averaged magnetic S(Q, E) of the PAF ordering using interaction parameters Jnn = 1 K, q = 0.8, and delta = 0. (b) Inelastic neutron scattering spectra of polycrystalline Tb2+xTi2-xO7+y with x = 0.005 at T = 0.1 K.

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