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Spin correlation in kagomé ice state: Neutron scattering study of the dipolar spin ice $Dy_2Ti_2O_7$ under magnetic field along [1 1 1]

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Abstract

We have investigated the kagomé ice state in the frustrated pyrochlore oxide $Dy_2Ti_2O_7$ under magnetic field along a [1 1 1] axis. Spin correlations have been measured by neutron scattering and analyzed by Monte Carlo simulation. The kagomé ice state, which has a non-vanishing residual entropy well established for the nearest-neighbor spin ice model by the exact solution, has been proved to be realized in the dipolar-interacting spin ice $Dy_2Ti_2O_7$ by observing the characteristic spin correlations. The simulation shows that the long-range interaction gives rise to only weak lifting of the ground state degeneracy and that the system freezes within the degenerate kagomé ice manifold .

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Keywords: Geometrical frustration; Spin ice; Kagomé ice; Spin correlations; Neutron scattering

The spin ice model, an Ising ferromagnet with a local ⟨1 1 1⟩ easy axis on the pyrochlore lattice interacting with nearest-neighbor exchange coupling, has attracted much attention because of its intriguing frustration mechanism [1]. It has macroscopically degenerate ground states characterized by the "two-in and two-out" local structure on each tetrahedron, equivalent to the "ice rule" in the water ice [2]. The spin ice behavior, e.g. the residual entropy [3], was discovered in the rare earth pyrochlore oxides, $Ho_2Ti_2O_7$ [4], $Dy_2Ti_2O_7$ [3] and $Ho_2Sn_2O_7$ [5]. However, in these systems Ising spins on the rare earth sites interact via a long-range dipolar interaction, and the mechanism showing the spin ice behavior in these dipolar spin ice was puzzling. A numerical account was provided by the Monte Carlo (MC) simulation studies [1], showing that the ground state manifold is approximately preserved in the dipolar model. More recently an elegant explanation using an analytical method was proposed in a theoretical work [7].

The macroscopic degeneracy of the spin ice model is partly or fully lifted by magnetic fields [8]. Along a [1 1 1] axis the pyrochlore lattice can be viewed as an alternating stacking of kagomé and triangular layers. For field along the [1 1 1] axis, the degeneracy is partly lifted, where random spin configurations are retained only in the kagomé planes, and has been studied by an exactly solvable dimer model [9,10]. Surprisingly, this kagomé ice state based on the nearest-neighbor interacting model, was suggested to be observed experimentally in the dipolar spin ice compound [11,12].

Unlike the case in zero field, the effects of the long-range dipolar interaction have not been clarified in magnetic field, and hence it is still puzzling whether the degeneracy of the kagomé ice manifold is preserved in the dipolar spin ice. To

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resolve this question, we have measured static spin correlations of the dipolar spin ice compound $Dy_2Ti_2O_7$ in magnetic field along the [1 1 1] axis by neutron scattering and observed characteristic spin correlations. By comparing with MC-simulation results based on the dipolar model, we have demonstrated that the kagomé ice state really occurs in the dipolar spin ice.

Neutron scattering experiments were performed using the triple-axis spectrometer GPTAS installed at JRR-3M JAERI. Two single crystalline samples of $Dy_2Ti_2O_7$, which were grown by the floating-zone method using an infrared furnace, were used for the experiments. The samples were mounted in a dilution refrigerator. MC-simulations were carried out using the facilities of the Supercomputer Center at ISSP University of Tokyo, which utilize the standard Metropolis single spin flip algorithm.

We show neutron intensity maps in the scattering plane perpendicular to the [1 1 1] axis in zero and finite ($H = 0.5$ T) fields at $T = 0.3$ K obtained from the experiments and MC-simulations based on the dipolar model in Fig. 1. Experimental results show definite differences between the spin correlations at $H = 0.0$ and 0.5 T, as shown in Fig. 1(I-a) and (II-a), respectively. The characteristic correlations at $\boldsymbol{Q}_0 = (2/3, -2/3, 0)$ ($\boldsymbol{Q}_0$-correlation), the positions are represented by black circles in (II-a), is found at $H = 0.5$ T.

The MC-simulations based on the dipolar model well reproduce the neutron intensity map at $H = 0.5$ T (Fig. 1(II-b)), as well as that at $H = 0.0$ T (Fig. 1(I-b)). The characteristic $\boldsymbol{Q}_0$-correlation observed in the experiment is clearly reproduced in Fig. 1(II-b). The $\boldsymbol{Q}_0$-correlation was proposed by the theoretical work based on the nearest-neighbor model [9], and hence, it is a good signature of the kagomé ice state. The agreement between the experimental and the MC-simulation results on the static spin correlations, especially the observation of the $\boldsymbol{Q}_0$-correlation, indicates that the degeneracy of the kagomé ice manifold is approximately preserved in the dipolar spin ice $Dy_2Ti_2O_7$. Spin correlations at $H = 0.5$ T do not show significant temperature dependence below 0.75 K in both the experiments and the single spin flip MC-simulation. It strongly suggests that the system freezes within the degenerate kagomé ice manifold.

In summary, we have presented the investigation of the neutron scattering experiments of the dipolar spin ice compound $Dy_2Ti_2O_7$ in the magnetic field along the [1 1 1] axis. The analysis of the experimental results by the single spin flip MC-simulations reveals that the kagomé ice state is realized in the dipolar spin ice.

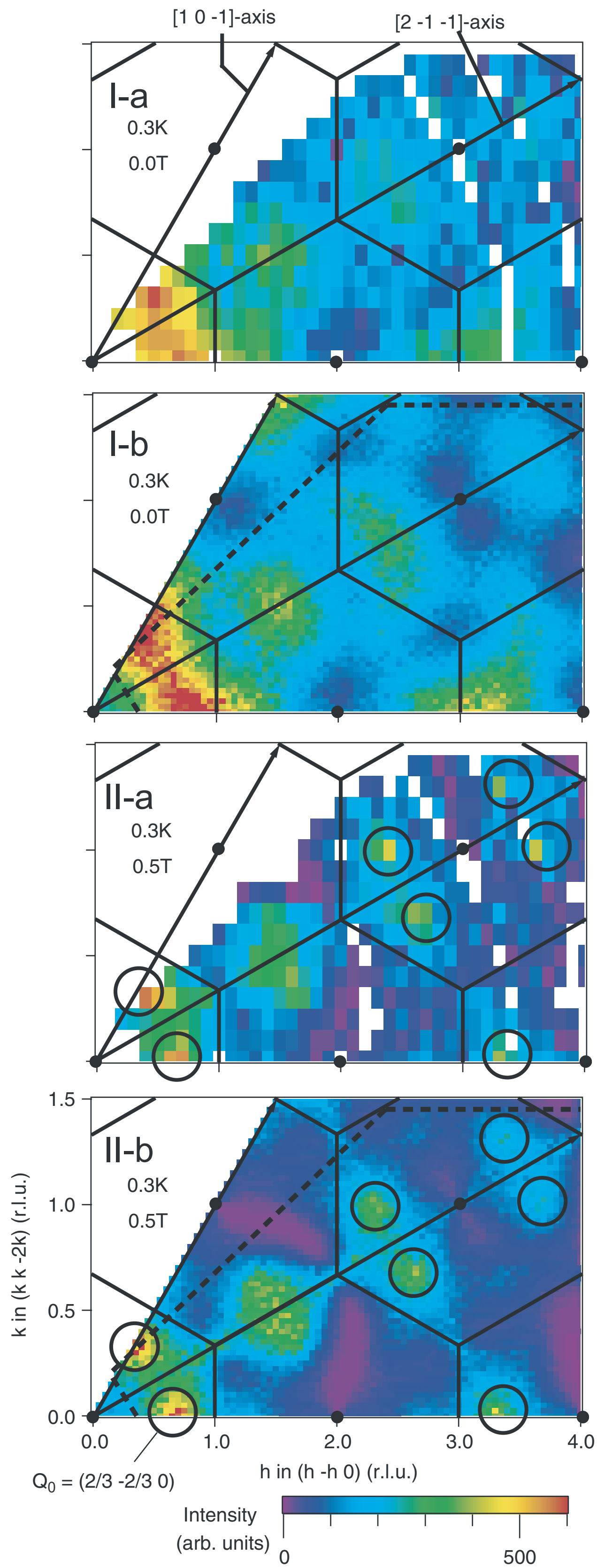

Fig. 1. Neutron intensity maps in the perpendicular plane to the [1 1 1] axis. Experimental results (I-a) at $T = 0.3$ K and $H = 0.0$ T and (II-a) at $T = 0.3$ K and $H = 0.5$ T. MC-simulation results (I-b) at $T = 0.3$ K and $H = 0.0$ T and (II-b) at $T = 0.3$ K and $H = 0.5$ T. Energy parameters used for the simulations in the dipolar model are $J_1 = -3.72$ K, $J_2 = 0.1$ K, $J_3 = -0.03$ K and $D = 1.41$ K, where the first, second, third nearest neighbor exchange interactions and the dipolar coupling constant, respectively. The values of J_1, J_3 and D are assigned to those reported previously [6,13] and the value of J_2 is determined to reproduce the temperature dependences of the specific heat in finite fields [14].

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