Ordered Phase of the Dipolar Spin Ice under [110]-Magnetic Fields

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(Dated: March 22, 2022)

We find that the true ground state of the dipolar spin ice system under [110]-magnetic fields is the “Q = X” structure, which is consistent with both experiments and Monte Carlo simulations. We then perform a Monte Carlo simulation to confirm that there exists a first order phase transition under the [110]-field. In particular this result indicates the existence of the first order phase transition to the “Q = X” phase in the field above 0.35 T for Dy$_2$Ti$_2$O$_7$. We also show the magnetic field-temperature phase diagram to summarize the ordered states of this system.

Keywords: spin ice, Ising chain, Monte Carlo simulation, pyrochlore lattice, Dy$_2$Ti$_2$O$_7$

Spin ice (SI) materials such as Dy$_2$Ti$_2$O$_7$ and Ho$_2$Ti$_2$O$_7$ have been attracting much interest as one of the ideal model materials of geometrically frustrated systems [1, 2]. In the SI materials, the magnetic moments occupy a three dimensional network of corner sharing tetrahedra with cubic symmetry called the pyrochlore lattice (FIG.1). Each magnetic moment is forced to point toward either of two tetrahedron centers by the strong crystal field along the local [111] axes [3, 4]. Then ferromagnetic nearest neighbor interactions lead to a geometrical frustration among four spins in a tetrahedron and the local ground states in each tetrahedron are six-fold degenerated. These local ground states are determined by the rule that two spins point inward and two spins outward. This “2-in & 2-out” rule is called the ice rule [5, 6, 7].

It has been suggested that the large dipole interaction is responsible for the spin ice behavior in the SI materials [8]. In the SI model with long range dipole interaction (the dipolar spin ice, DSI), it would be expected that the degeneracy of the ground state is removed by the long range nature of the dipole interaction and a long range degeneracy of the ground state is removed by the long range nature of the dipole interaction and a long range dipole interaction [8]. In the SI model with long range dipole interaction is responsible for the spin ice behavior in the SI material [8].

Recently, the SI materials under magnetic fields along the [110]-direction have been studied by means of single crystal experiments [8]. Fennel et al. performed neutron diffraction experiments of Dy$_2$Ti$_2$O$_7$ under the [110]-magnetic fields with fixed low temperature ~ 70 mK, which shows magnetic Bragg peaks at the Q = 0 points and diffuse scattering at the Q = X points [9]. From their scattering patterns, they found the coexistence of long-range ferromagnetic order on the field coupled spin chains (called α-chains by Hiroi et al. [10], FIG.4) and short-range antiferromagnetic order on the field independent spin chains (β-chains). They suggested that the true ground state is the Q = X structure as shown in FIG.4 [10], although they could not find a clear evidence for such ordered state. A similar result was found in neutron diffraction experiments of Ho$_2$Ti$_2$O$_7$ by Harris et al [11]. On the other hand, Hiroi et al. performed specific heat measurements of Dy$_2$Ti$_2$O$_7$ in the [110]-magnetic fields. They found a relatively sharp peak at 1.1 K (below the broad peak associated with SI
freezing at low fields) in the field $H \gtrsim 0.4$ T. They suggested that this peak comes from the freezing of $\beta$-chains without long range order. They also argued the frustration between $\beta$-chains and threw doubt on the $Q = X$ structure as the true ground state. In the theoretical point of view, Melko et al performed MC simulations to observe a first order phase transition to the $Q = X$ structure. Thus the $Q = X$ structure is still controversial.

In the [110]-fields, there remains ground state degeneracy on the $\alpha$-chains as far as only the nearest neighbor interaction is taken into account. We expect that this degeneracy is removed by the dipole interaction. In this paper, we give a proof that the $Q = X$ structure is the true ground state configuration under the [110]-fields. We then perform single spin flip MC simulations to show that the relatively sharp specific heat peak originates from the first order phase transition to the $Q = X$ phase and to present the temperature-field phase diagram.

In the spin ice compound, each spin $s_i$ is forced to align along its easy axis $n_i$ due to the strong crystal field, so that it is expressed with an Ising variable $\sigma_i(= \pm 1)$ as $s_i = \sigma_i n_i$, where $n_i$ is one of the four distinct Ising axes $n_\nu; \nu = 0, 1, 2, 3$ corresponding to each site on the basic tetrahedron (FIG.1). Then the Hamiltonian of the Ising DSI system with $N$ spins under an uniform external field $H$ is given as

$$H(\sigma_1, \sigma_2, \ldots, \sigma_N) = -p_{\text{eff}} \mu_B \sum \limits_i \left( H \cdot n_i \right) \sigma_i + (J_{nn} + D_{nn}) \sum \limits_{ij} \sigma_i \sigma_j + 3 \frac{D_{nn} r_{nn}^3}{5} \sum \limits_{ij} \frac{\left| r_{ij} \right|^2 (n_i \cdot n_j) - 3 (r_{ij} \cdot n_i)(r_{ij} \cdot n_j)}{\left| r_{ij} \right|^5} \sigma_i \sigma_j,$$

where the second and the last terms represent the nearest neighbor interactions (both exchange $J_{nn}$ and dipole $D_{nn}$) and the long range part of the dipole interactions, respectively. By using the unit of cell $a$, the nearest neighbor distance $r_{nn}$ is written as $r_{nn} = a/\sqrt{8}$. In this paper we use material parameters of Dy$_2$Ti$_2$O$_7$: $J_{nn} = -1.24$ K, $D_{nn} = 2.35$ K and $p_{\text{eff}} \mu_B = 10.6 \mu_B = 7.09 \text{ KT}^{-1}$. $\mathbb{R}$.

First, we consider the effect of magnetic field along the [110]-direction without the long range interaction term. In the [110]-fields, the pyrochlore lattice is divided into two sets of Ising spin chains named as $\alpha$ and $\beta$-chains, as illustrated in FIG.1. Spins on the $\nu = 0, 3$ sites compose $\alpha$-chains and ones on the $\nu = 1, 2$ sites $\beta$-chains. The [110]-magnetic fields affect spins only on the $\alpha$-chains and tend to align them ferromagnetically. Being free from the [110]-magnetic fields, spins on a $\beta$-chain also align ferromagnetically due to the ice rule when the $\alpha$-chains are ordered ferromagnetically. It should be noted that each ferromagnetic $\alpha$-chain has two possible states and the ground states are $2^{N_\beta}$-fold degenerated, where $N_\beta \sim N^{2/3}$ is the number of the $\beta$-chains. The ground state, the inter-chain configuration of $\alpha$-chains is ferromagnetic (F) while the one of $\beta$-chains is paramagnetic (P). Here we introduce abbreviated notations for inter-chain configurations of the $\alpha/\beta$-chains for convenience. The inter-chain configuration of the $\alpha$-chains is abbreviated as $\alpha$-AF if they are ordered antiferromagnetically (AF). In this notation, the ground state configuration mentioned above is expressed as “$\alpha$-F & $\beta$-AF”. Thus there is no phase transition to the long range ordered phase without the long range interaction.

Now, we consider the effect of the long range interaction. At zero field, the long range interaction removes the macroscopic degeneracy of the ground states and the $q_{\text{rot}} = (0, 0, 2\pi/\alpha)$ configurations become the true ground states. In terms of the inter-chain configuration of the $\alpha/\beta$-chains, these ground states are expressed as “$\alpha$-AF & $\beta$-AF”. In order to show that the $Q = X$ structure, which is expressed as “$\alpha$-F & $\beta$-AF” (see FIG.1), is the true ground state under [110]-magnetic fields, we only consider F and AF inter-chain alignments for the $\alpha/\beta$-chains. By means of the Ewald summation techniques, we calculated the interaction energy of the four configurations for $H = 0$ and obtained

$$\frac{1}{N} E_{\alpha}^{\alpha, \beta} = -1.5060 \text{ K}, \quad \frac{1}{N} E_{\alpha}^{\alpha, F, \beta} = -0.6816 \text{ K}, \quad \frac{1}{N} E_{\alpha}^{\alpha, \beta, \beta} = -1.0938 \text{ K}. \quad (2)$$

Note that the two energy differences $E_{\alpha}^{\alpha, F, \beta} - E_{\alpha}^{\alpha, \beta, \beta}$ and $E_{\alpha}^{\alpha, F, \beta} - E_{\alpha}^{\alpha, \beta, \beta}$ are both 0.4122 K per spin. This result implies that there is no interaction between the $\alpha$ and $\beta$-chains as far as F and AF inter-chain alignment are concerned. Indeed this statement is true since the internal fields acting on the $\beta$-chains from the $\alpha$-F/AF chains are canceled out each other due to the symmetry of the pyrochlore lattice structure. Since “$\alpha$-AF & $\beta$-AF” is the ground state at zero field, we can say that each set of ferromagnetic Ising spin chains tends to align anti-ferromagnetically. In the [110]-field, the energy of the “$\alpha$-F & $\beta$-AF” configuration comes down while the energy of the “$\alpha$-AF & $\beta$-AF” configuration does not change. So there is a ground state transition field $H_c$, at which the following equation holds:

$$\frac{1}{N} E_{\alpha}^{\alpha, \beta} = \frac{1}{N} E_{\alpha}^{\alpha, F, \beta} - \frac{1}{\sqrt{6}} p_{\text{eff}} \mu_B H_c. \quad (3)$$

Using these values given in eq.(2), we obtain the transition point $H_c = 0.142$ T. Thus we conclude that the “$\alpha$-F & $\beta$-AF” ($Q = X$) configuration is the true ground state in the field $H > H_c$.

Next we perform single spin flip MC simulations to show that there exists phase transition to the “$\alpha$-F & $\beta$-AF” phase. Our simulations were carried out on system size up to 5488 spins (cubic unit cell length $L = 7$) with a slow cooling process. The simulation lengths were $\sim 10^5$ MC steps per spin at each temperature. In FIG.
we show our MC results for the temperature dependence of the specific heat for system size $L = 4$ under various magnetic fields $H$. At zero field a broad peak corresponding to the freezing into the SI state is found around $T_{SI} \simeq 1.2$ K, in good agreement with the previous results [8]. As the magnetic field increases, the peak at $T_{SI}$ grows up to $H = 0.30$ T, and then at $H = 0.35$ T a small anomalous peak appears at $T_{\alpha\beta}$ on the low temperature side of $T_{SI}$, and this anomalous peak moves to the high temperature side. At $H = 0.50$T, the peak at $T_{\alpha\beta}$ merges with the broad peak at $T_{SI}$ and there appears a very sharp peak, and then this peak is broken into two peaks at $T_{\alpha}$ and $T_{\beta}$. The peak at $T_{\alpha}$ is broad and shifts to the high temperature side with increasing field. On the other hand, the peak at $T_{\beta}$ is sharp and almost independent of the field. From their field dependences, the peaks at $T_{\alpha}$ and at $T_{\beta}$ must come from the $\alpha$ and $\beta$-chains, respectively. To extract the contribution of the $\beta$-chains, we performed a MC simulation of the system only with spins on the $\beta$-chains. This system equals to the DSI at the high-field limit since the spins on the $\beta$-chains feel no internal field from the $\alpha$-F chains. The temperature dependence is also shown in the FIG. 2 and we see a sharp peak near $T_{\beta} \sim 1.3$ K. This sharp peak corresponds to the peak at $T_{\beta}$ in large but finite magnetic fields as expected. These temperature and field dependences of the specific heat are also seen in the experiments by Hiroi et al [13].

In order to consider what happens at each peak more precisely, we observe order parameters corresponding to the inter-chain orders. For example, the order parameter for the $\beta$-AF order $m_{\beta,AF}$ is the $q_{ord} = (0, 0, 2\pi/a)$ staggered magnetization of the $\beta$-chains. In FIG. 3 we show the temperature dependence of $m_{\beta,AF}$ for system size $L = 6$. We can see the $\beta$-AF order below $T_{\alpha\beta}$ and $T_{\beta}$, and discontinuity of the order parameter $m_{\beta,AF}$ at these temperatures. We also find from the observation of $m_{\alpha,F}$ that the field sensitive broad peak at $T_{\alpha}$ comes from the...
FIG. 5: Magnetic field-temperature phase diagram for $H \parallel [110]$. The open squares, closed squares, open circles and closed circles indicate the characteristic peaks $T_{\alpha\beta}$, $T_\alpha$, $T_\beta$ and $T_{\beta}$, respectively. The solid line shows the first order phase transition and the dotted the crossover.

$\alpha$-F freezing. Therefore the low temperature anomalous peak at $T_{\alpha\beta}$ and the field independent peak at $T_\beta$ are due to the first order phase transition to the $\alpha$-F & $\beta$-AF ordered phase. We also carried out a finite size scaling of the peak height $C_{\text{peak}}$ for the magnetic field $H = 1.00T$ and for the $\beta$-chain system (FIG.1). The scaling data shows that $C_{\text{peak}}$ is proportional to the system size $L^3$ for large systems ($L \geq 5$). These results prove that the phase transition is indeed of the first order.

Finally we summarize our results from the ground state analysis and the MC simulation as a phase diagram in FIG.2. The data points in the diagram indicate the specific heat peak temperatures $T_{\text{SI}}$, $T_{\alpha\beta}$, $T_\alpha$, $T_\beta$ in the MC results with single-spin-flip slow cooling process (FIG.2). The solid line in the diagram corresponds to the first order phase transition. Note that the straight lines shown in the low temperature and in the weak field region are expectation ones from the ground state transition field $H_c = 0.142T$ and the zero field MC results [6]. In the cooling process at high field region, spins on the $\alpha$-chains are almost fixed by the $[110]$-magnetic field below $T_\alpha$, while the spins on the $\beta$-chains still take random configurations ("$\alpha$-F" phase). Then at $T_\beta$ the spins on the $\beta$-chains form the $\beta$-AF order and the phase transition to the $\alpha$-F & $\beta$-AF" configuration takes place due to the ice rule and the long range dipole interaction. In the weak field region $H < 0.30T$, the phase transition has not been observed in our single spin flip MC simulation.

In conclusion, we have studied the effect of the dipole interaction on the DSI system under the $[110]$-magnetic fields through the ground state analysis and the single spin flip MC study. We have found that the "$\alpha$-F & $\beta$-AF" ($Q = X$) configuration is the true ground state in the strong field region $H > H_c = 0.142T$ and the first order phase transition from the "$\alpha$-F" phase to the "$\alpha$-F & $\beta$-AF" phase occurs. We have presented the $[110]$-field-temperature phase diagram. Melko et al. applied the loop algorithm and the multicanonical MC method in order to avoid the freezing at low temperature in the first order phase transition [11, 12]. Such techniques will clarify the detail of phase boundary in the low temperature and the weak field region, and are now under progress.

We are grateful to Z. Hiroi, K. Matsuhira, M. Ogata, and T. Sakakibara for useful comments and discussions. This research was partially supported by the Ministry of Education, Science, Sports and Culture, Grant-in-Aid for JSPS Fellows.

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