Two-stage ordering of spins in dipolar spin ice on kagome

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Spin ice, a peculiar thermal state of a frustrated ferromagnet on the pyrochlore lattice, has a finite entropy density and excitations carrying magnetic charge. By combining analytical arguments and Monte Carlo simulations, we show that a similar magnet on kagome has two distinct spin-ice phases. The lower-temperature spin ice is distinguished by an ordering of magnetic charges. We comment on possible realizations of this model.

Frustrated magnets \[12\] attract attention of both theorists and experimentalists as models of strongly interacting systems with unusual ground states and elementary excitations. One of the recent surprises was a realization that elementary excitations in spin ice \[3, 4\] are quasiparticles carrying magnetic charge \[5\]. Subsequent work \[6\] uncovered signatures of the magnetic monopoles in magnetization dynamics of spin ice. Spin ice is a frustrated ferromagnet discovered in the pyrochlore \[7\], where magnetic \(\text{Ho}^{3+}\) ions form a network of corner-sharing tetrahedra \[8\]. The magnetic moments \(\mu_i = \pm \mu_0 \hat{e}_i\) are forced to point along a \((111)\) axis \(\hat{e}_i\) by a strong crystal field. The easy-axis anisotropy makes the spins Ising-like, so that a microstate of this magnet can be described by Ising variables \(\sigma_i = \pm 1\). The Hamiltonian includes exchange interactions of strength \(J\) for pairs of nearest neighbors \((ij)\) and dipolar interactions between all spins \[4\]:

\[
H = -J \sum_{ij} \sigma_i \sigma_j (\hat{e}_i \cdot \hat{e}_j) \tag{1}
\]

\[
+ \frac{D_{\text{nn}}}{2} \sum_{i \neq j} \sigma_i \sigma_j \left(\frac{\hat{e}_i \cdot \hat{e}_j}{|\mathbf{r}_i - \mathbf{r}_j|^3} - 3(\hat{e}_i \cdot \hat{\mathbf{r}}_{ij})(\hat{e}_j \cdot \hat{\mathbf{r}}_{ij})\right),
\]

where \(D = (\mu_0/4\pi)\mu^2/r_0^3\), is a characteristic strength of dipolar coupling, \(\mathbf{r}_i\) are spin locations, \(\hat{\mathbf{r}}_{ij} = (\mathbf{r}_i - \mathbf{r}_j)/|\mathbf{r}_i - \mathbf{r}_j|\), and \(r_{\text{nn}}\) is the distance between nearest neighbors. In the absence of dipolar interactions, \(D = 0\), and for ferromagnetic exchange, \(J > 0\), the system is strongly frustrated because it is impossible to minimize the energy of every bond \((ij)\). In a ground state, two spins point into every tetrahedron and two point out, which is reminiscent of proton positions in water ice \[8\], where every oxygen has two protons nearby and two farther away. This ice rule is satisfied by a macroscopically large number of microstates, so that both protons in water ice and magnetic moments in spin ice can remain disordered even at low temperatures \[9\].

Large magnetic moments \((\mu = 10\mu_B\) in \(\text{Ho}_2\text{Ti}_2\text{O}_7\)) make magnetic dipolar interactions between nearest neighbors comparable to exchange \[10\]. Together with the long-distance nature of dipolar interactions, the substantial value of \(D\) casts doubt on the usefulness of the short-range \((D = 0)\) model of spin ice. Yet numerical simulations show that, even after the inclusion of dipolar interactions, energy differences between states obeying the ice rule remain numerically small—so small that magnetic order induced by the dipolar interactions is expected to occur only at a rather low temperature, \(T \approx 0.08D\) \[11\] (we set \(k_B = 1\)). The apparent weakness of the dipolar interaction was explained by Castelnovo et al. \[12\], who introduced a “dumbbell” version of spin ice, in which magnetic dipoles are stretched into bar magnets of length \(a\) such that their poles meet at the centers of tetrahedra. The energy of the resulting model can be represented as a Coulomb-like interaction of magnetic charges at the ends of the dumbbells, \(q_i = \pm \mu/a\) \[5\]:

\[
E(\{Q_i\}) = \sum_{\alpha} \frac{Q_{\alpha}^2}{2C} + \frac{\mu_0}{8\pi} \sum_{\alpha \neq \beta} \frac{Q_{\alpha} Q_{\beta}}{|r_{\alpha} - r_{\beta}|},
\]

In this expression, \(Q_{\alpha} = \sum_{i \in \alpha} q_i\) is the sum of magnetic charges at the center of tetrahedron \(\alpha\). In a spin-ice state of the dumbbell model, every tetrahedron will have two north and two south poles with a total magnetic charge \(Q_{\alpha} = 0\), minimizing the first term in Eq. \((2)\). As a result, no magnetic field will be generated and the magnetic dipolar energy will be strictly zero. A similar, albeit incomplete cancellation occurs in the original model, making Eq. \((2)\) a very good approximation for the energy \[11\]. The charge of tetrahedron \(\alpha\), expressed in units of \(\mu/a\), is calculated as

\[
Q_{\alpha} = \pm \sum_{i \in \alpha} \sigma_i\tag{3}
\]

with the plus sign for one sublattice of tetrahedra and minus for the other. Residual interactions, responsible for the formation of magnetic order, are weak and fall off quickly with the distance \[6\]. The resulting energy differences between states obeying the ice rule are only a small fraction of the dipolar energy scale \(D\) \[12, 13\].
short-range version of this model (ways of realizing such a system are discussed below. A microstate obeying the ice rule $Q_\alpha = \pm 1$, shown in Figs. (a) and (b), contains uncompensated charges that generate a magnetic field. To a first approximation, the system energy is given by the Coulomb terms $\frac{1}{2} \sum_{\alpha \beta} Q_\alpha Q_\beta$. Nonzero values of magnetic charges results in substantial energy differences between different states obeying the ice rule. The Coulomb energy is minimized when the charges are ordered in such a way that adjacent triangles carry charges of opposite signs. The charge-ordered states of the dipolar kagome ice are closely related to the ice states of the pyrochlore spin ice in a magnetic field along a (111) axis. The number of such states grows exponentially with the number of spins $N$. They are exactly degenerate in the dumbbell model. In the dipolar spin ice, the degeneracy is broken by small corrections to the Coulomb energy $\frac{1}{2} \sum_{\alpha \beta} Q_\alpha Q_\beta$.

This hierarchy of energy scales suggests the following sequence of thermal transformations in the dipolar spin ice on kagome. As the magnet cools down from the high-temperature paramagnetic state with entropy per spin $s = \ln 2 = 0.693$, it gradually enters the spin-ice state with $s \approx (1/3) \ln (9/2) = 0.501$. At a temperature of the order of $D$, it will enter a distinct phase with ordered magnetic charges, where entropy density is reduced but remains nonzero, $s = 0.108$ per spin; the state is similar to that of the pyrochlore spin ice in a magnetic field along $\langle 111 \rangle$. At an even lower temperature, the system will enter a spin-ordered state with zero entropy density. In contrast, the short-range version of spin ice with nearest-neighbor interactions exhibits neither charge, nor spin order.

The spin order emerging on top of magnetic charge order is expected to be of the $\sqrt{3} \times \sqrt{3}$ type shown in Fig. (c), the same as in the short-range model with antiferromagnetic second-neighbor exchange. This can be understood as follows. In a charge-ordered state every triangle has two majority spins pointing into (or out of) the triangle and a minority spin pointing the other way. Such states can be represented by dimer coverings of a honeycomb lattice, Fig. (d); the dimers indicate locations of minority spins. The energy of such a state is determined by the interactions between minority spins alone. To see that, picture a minority spin $-\mu$ as a superposition of a majority spin $+\mu$ and a minority spin of double strength $-2\mu$; in this representation, majority spins form an inert background. We thus arrive at a model of dimers with point dipoles of strength $2\mu$ directed along the dimers and towards triangles with positive charge. The interaction energy of two dimers depends on their mutual position. It is minimized by increasing the number of second neighbors (distance between centers $\sqrt{3}a_{nm}$) and reducing the number of third neighbors ($2a_{nn}$). The dimer configuration shown

As the magnet is cooled down from a high-temperature paramagnetic state with completely uncorrelated spins, it first gradually enters the spin-ice regime at the crossover temperature $T \approx 2J_{\text{eff}}$, where $J_{\text{eff}} = J/3 + 5D/3$ is the effective interaction for nearest-neighbor Ising spins $\sigma_i \sigma_j$, and then undergoes a phase transition into a magnetically ordered state at $T \approx 0.08D$.

In this Letter we discuss a related problem of dipolar spin ice on kagome, a two-dimensional lattice of corner-sharing triangles. Each spin is constrained to point along the line connecting the two triangles, Fig. (a). Possible ways of realizing such a system are discussed below. A short-range version of this model ($D = 0, J > 0$) was studied by Wills et al.

At a first glance, the dipolar spin ice on kagome is very similar to its counterpart on the pyrochlore lattice and one might expect a similar sequence of states: as the system cools down, it would gradually enter a correlated but still disordered spin-ice state and then make a phase transition into a magnetically ordered state. However, a closer look reveals the existence of an intermediate thermodynamic phase with disordered spins and ordered magnetic charges.

The existence of the intermediate charge-ordered phase depends on the allowed values of magnetic charge $\frac{1}{2}$, which can be even on a tetrahedron and odd on a triangle. Minimization of the first term in Eq. (2) yields $Q_\alpha = 0$ on the pyrochlore lattice and $\pm 1$ on kagome. A microstate obeying the ice rule $Q_\alpha = \pm 1$, shown in Figs. (a) and (b), contains uncompensated charges that generate a magnetic field. To a first approximation, the system energy is given by the Coulomb terms $\frac{1}{2} \sum_{\alpha \beta} Q_\alpha Q_\beta$. Nonzero values of magnetic charges results in substantial energy differences between different states obeying the ice rule. The Coulomb energy is minimized when the charges are ordered in such a way that adjacent triangles carry charges of opposite signs. The charge-ordered states of the dipolar kagome ice are closely related to the ice states of the pyrochlore spin ice in a magnetic field along a (111) axis. The number of such states grows exponentially with the number of spins $N$. They are exactly degenerate in the dumbbell model. In the dipolar spin ice, the degeneracy is broken by small corrections to the Coulomb energy $\frac{1}{2} \sum_{\alpha \beta} Q_\alpha Q_\beta$.

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dependence of the staggered charge order parameter $Q$ in the universality class of the two-dimensional Ising model. However, the long-range nature of the dipolar interactions may change that. A finite-size analysis reveals a universal scaling behavior of the specific heat $c$ and charge susceptibility $\chi_Q$ renormalized at a magnetic charge by Eq. (3). We use the lattice constant $a = 1/\sqrt{3}$ as a unit of length, so that the nearest neighbor distance is $r_{nn} = 1/2$ and the dumbbell length is $a = 1/\sqrt{3}$.

Simulations of the dumbbell model were performed on lattices with periodic boundary conditions and lengths up to $L = 30$, which corresponds to $N = 3L^2 = 2700$ sites. Temperature was measured in units of $D \equiv (\mu_0/4\pi)\mu^2/r_{nn}^3$. The charging energy—the first term in Eq. (2)—had the coupling strength $1/C = 8.264(\mu_0/4\pi)$. The long-range Coulomb interaction in Eq. (2) was truncated beyond the distance of $10r_{nn}$. The temperature dependence of the staggered charge order parameter $Q$ is shown in Fig. 2(a) for various system sizes. The existence of a continuous charge-ordering transition is evidenced by Fig. 2(b), where Binder’s fourth-order cumulants cross at $T_c \approx 0.267D$ for different system sizes.

On the basis of the symmetry of the order parameter, one might expect the charge-ordering transition to be in the universality class of the two-dimensional Ising model. However, the long-range nature of the dipolar interactions may change that. A finite-size analysis reveals a universal scaling behavior of the specific heat $c$ and charge susceptibility $\chi_Q$ renormalized at a magnetic charge by Eq. (3). We use the lattice constant $a = 1/\sqrt{3}$ as a unit of length, so that the nearest neighbor distance is $r_{nn} = 1/2$ and the dumbbell length is $a = 1/\sqrt{3}$.

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FIG. 2: Monte Carlo simulation of the charge-ordering transition in the dumbbell model (2). (a) and (b) show the temperature dependence of the (normalized) staggered charge order parameter $Q$ and Binder’s fourth-order cumulant $B_4 \equiv 1−⟨Q^4⟩/(⟨Q^2⟩)^2$, respectively, for various system sizes $L$. (c) and (d) show the scaling of specific heat $c$ and charge-order susceptibility $\chi_Q$, respectively.

FIG. 3: Temperature dependence of the specific heat $c(T)$ and entropy per spin $s(T)$ of the dipolar spin ice [4] with (a) ferromagnetic exchange $J = 0.5D$ and (b) antiferromagnetic exchange $J = −2.67D$. The linear size of the system is $L = 12$. The dashed lines show levels of entropy $s = 0.693$ (Ising paramagnet), 0.501 (spin ice), and 0.108 (charge-ordered spin ice) per spin.
spin-ordered phase exists at the lowest temperatures.

Spin ice on kagome can be created by substituting 1/4 of magnetic sites of a pyrochlore spin ice with nonmagnetic atoms, as was done in the Heisenberg antiferromagnet herbertsmithite \( \text{ZnCu}_3(\text{OH})_6\text{Cl}_2 \) [17]. Kagome spin ice already exists as an artificial magnetic lattice [18, 19]. Although the energy scale of dipolar interactions in artificial ice greatly exceeds the room temperature [20], it may be possible to introduce thermal motion of spins by agitating them in a manner of granular systems [21].

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[1] H. T. Diep, ed., Frustrated spin systems (World Scientific, 2004).
[2] B. Normand, Contemp. Phys. 50, 533 (2009).
[3] S. T. Bramwell and M. J. P. Gingras, Science 294, 1495 (2001).
[4] M. J. P. Gingras, in Highly frustrated magnetism, edited by C. Lacroix, P. Mendels, and F. Mila (Springer Verlag, in press), arXiv:0903.2772.
[5] C. Castelnovo, R. Moessner, and S. L. Sondhi, Nature 451, 42 (2008).
[6] L. D. C. Jaubert and P. C. W. Holdsworth, Nat. Phys. 5, 258 (2009).
[7] M. J. Harris, S. T. Bramwell, D. F. McMorrow, T. Zeiske, and K. W. Godfrey, Phys. Rev. Lett. 79, 2554 (1997).
[8] V. F. Petrenko and R. W. Whitworth, Physics of ice (Oxford University Press, 1999).
[9] A. P. Ramirez, A. Hayashi, R. J. Cava, R. Siddharthan, and B. S. Shastry, Nature 399, 333 (1999).
[10] B. C. den Hertog and M. J. P. Gingras, Phys. Rev. Lett. 84, 3430 (2000).
[11] R. G. Melko, B. C. den Hertog, and M. J. P. Gingras, Phys. Rev. Lett. 87, 067203 (2001).
[12] R. G. Melko, M. Enjalran, B. C. den Hertog, and M. J. P. Gingras (unpublished), arXiv:cond-mat/0308282.
[13] R. G. Melko and M. J. P. Gingras, J. Phys.: Condens. Matter 16, R1277 (2004).
[14] A. S. Wilks, R. Ballou, and C. Lacroix, Phys. Rev. B 66, 144407 (2002).
[15] M. Udagawa, M. Ogata, and Z. Hiroi, J. Phys. Soc. Jpn. 71, 2365 (2002).
[16] F. Y. Wu, Rev. Mod. Phys. 54, 235 (1982).
[17] J. S. Helton, K. Matan, M. P. Shores, E. A. Nytko, B. M. Bartlett, Y. Yoshida, Y. Takano, A. Suslov, Y. Qiu, J.-H. Chung, et al., Phys. Rev. Lett. 98, 107204 (2007).
[18] M. Tanaka, E. Saithoh, H. Miyajima, T. Yamaoka, and Y. Iye, Phys. Rev. B 73, 052411 (2006).
[19] Y. Qi, T. Brintlinger, and J. Cumings, Phys. Rev. B 77, 094418 (2008).
[20] R. F. Wang, C. Nisoli, R. S. Freitas, J. Li, W. McConville, B. J. Cooley, M. S. Lund, N. Samarth, C. Leighton, V. H. Crespi, et al., Nature 439, 303 (2006).
[21] X. Ke, J. Li, C. Nisoli, P. E. Lammert, W. McConville, R. F. Wang, V. H. Crespi, and P. Schiffer, Phys. Rev. Lett. 101, 037205 (2008).

FIG. 4: Monte Carlo simulations of magnetic phase transitions. (a) and (b) show the temperature dependence of the (normalized) magnetic order parameter \( M \) and Binder’s fourth-order cumulant \( B_4 \equiv 1 - (\langle |M|^4 \rangle /3\langle |M|^2 \rangle)^2 \), respectively, for various system sizes \( L \). (c) and (d) show the data collapsing of scaled specific heat \( c \) and magnetic susceptibility \( \chi_M \equiv \langle |M|^2 \rangle - \langle |M|^2 \rangle^2 / NT \), respectively.