Magnetic monopole and string excitations in two-dimensional spin ice

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We study the magnetic excitations of a square lattice spin ice recently produced in an artificial form as an array of nanoscale magnets. Our analysis, based on the dipolar interaction between the nanomagnetic islands, correctly reproduces the ground state observed experimentally. In addition, we find magnetic monopolelike excitations effectively interacting by means of the usual Coulombic plus a linear confining potential, the latter being related to a stringlike excitation binding the monopoles pairs, which indicates that the fractionalization of magnetic dipoles may not be so easy in two dimensions. These findings contrast this material with the three-dimensional analog, where such monopoles experience only the Coulombic interaction. We discuss, however, two entropic effects that affect the monopole interactions. First, the string configurational entropy may lose the string tension and then free magnetic monopoles should also be found in lower dimensional spin ices; second, in contrast to the string configurational entropy, an entropically driven Coulomb force, which increases with temperature, has the opposite effect of confining the magnetic defects. © *2009 American Institute of Physics*. [doi:10.1063/1.3224870]

I. INTRODUCTION

Geometrical frustration among spins in magnetic materials can lead to a variety of cooperative phases such as spin glass, spin liquid, and spin ice behaving like glass, liquid, and ice in nature. The description and understanding of such states are becoming increasingly important not only in condensed matter but also in other branches such as field theories. In a crystal at low temperature excitations above the ground state often behave like elementary particles carrying a quantized amount of energy, momentum, electric charge, and spin. Several of these objects arise as a result of the collective behavior of many particles in a material, which is most effectively described in terms of the fractions of the original particles. The emergence of these excitations is an example of the phenomenon known as "fractionalization." This occurrence is often tied to topological defects[1] and is common in one-dimensional systems (polyacetylene, nanotubes, etc). Higher dimensional fractionalization is more difficult to be found. In two spatial dimensions the only confirmed case is the involvement of quasiparticles with one third of an electron's charge in the fractional quantum Hall effect in strong magnetic fields. Among several suggestions,[2] there is also the proposal that the merons forming a skyrmion in two-dimensional (2D) Heisenberg antiferromagnets are spinons and therefore, they are neutral spin-half excitations.[3,4] More recently, examples of fractionalization in three-dimensional (3D) systems were provided in spin ice materials.[2,5] Particularly, Castelnovo *et al.*[5] have shown how the famous magnetic monopole may emerge in these materials. Despite some exciting suggestions for its existence from the realms of quantum mechanics, a single magnetic pole remains elusive after decades of searching in particle accelerators and cosmic rays. Now, Castelnovo *et al.*[5] indicated an unexpected but, perhaps, better place to look. Under certain conditions, spin ice magnets behave like a gas of free magnetic poles. There is even a phase transition at which a thin vapor of these poles condenses into a dense liquid. An experimentally measurable signature of monopole dynamics on a diamond lattice in the grand canonical ensemble was presented in Ref. 6. The existence of these excitations in a condensed matter system is exciting in itself. Our aim in this paper is to study spin ice materials, but in two spatial dimensions. Such structures have been artificially produced in a geometrically frustrated lattice of nanoscale ferromagnetic islands.[7–9] Here, we examine the excitations ("magnetic monopoles") and how they interact in this 2D system.

3D spin ice materials have the pyrochlore structure in which magnetic rare-earth ions form a lattice of corner-sharing tetrahedra. To minimize the spin-spin interaction energy, the ice rules are manifested: two spins point inward and two spins point outward on each tetrahedron. A similar system was built in two dimensions with elongated permalloy nanoparticles. This artificial material consists of elongated magnetic nanoislands distributed in a 2D square lattice. The longest axis of the islands alternate its orientation pointing in the direction of the two principal axes of the lattice.[7] The magnetocrystalline anisotropy of permalloy is effectively zero, so that the shape anisotropy of each island forces its magnetic moment to align along the largest axis thus, making the islands effectively Ising-like. The intrinsic frustration on this lattice is similar to that in the spin ice model and can be best seen by considering a vertex at which four islands meet. A pair of moments on a vertex can be aligned either to maximize or to minimize the dipole interaction energy of the pair.

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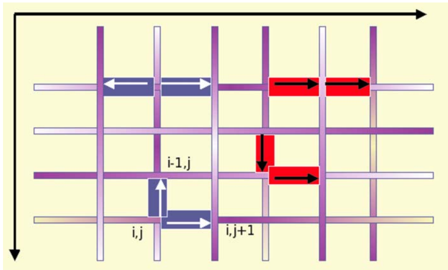

FIG. 1. (Color online) The 2D square lattice studied in this work. Only a few islands are shown. The arrows inside the islands represent the local dipole moments ($\vec{S}_{h(i)}$ or $\vec{S}_{v(i)}$).

As shown in Ref. 7, it is energetically favorable when the moments of a pair of islands are aligned so that one is pointing into the center of the vertex and the other is pointing out (the two arrangements on the right, Fig. 1), while it is energetically unfavorable when both moments are pointing inward or both are pointing outward (the pair of arrangements on the left, Fig. 1). This artificial system exhibits short-range order and icelike correlations on the lattice, which is precisely analogous to the behavior of the spin ice materials. However, it should be stressed that the fundamental interaction among the islands is the long-range dipole-dipole force, once the short-ranged exchange is negligible in this case, where the islands are spaced by around 320 nm, much greater than the permalloy exchange length, around 5–7 nm. Here, we consider an arrangement like that experimentally investigated in Ref. 7. In our scheme the magnetic moment ("spin") of the island is replaced by a point dipole at its center. To do this, in each site (x_i, y_i) of a square lattice two spin variables are defined: $\vec{S}_{h(i)}$ with components $S_x = \pm 1$, $S_y = 0$ located at $\vec{r}_h = (x_i + 1/2, y_i)$, and $\vec{S}_{v(i)}$ with components $S_x = 0$, $S_y = \pm 1$ at $\vec{r}_v = (x_i, y_i + 1/2)$. Therefore, in a lattice of volume $L^2 = l^2 a^2$ (a is the lattice spacing) one gets $2 \times l^2$ spins (see Fig. 2). Representing the spins of the islands by $\vec{S}_i$, which can assume either $\vec{S}_{h(i)}$ or $\vec{S}_{v(i)}$, then the 2D spin ice is described by the following equation:

$$H_{\rm SI} = Da^3 \sum_{i \neq j} \left[\frac{\vec{S}_i \cdot \vec{S}_j}{r_{ij}^3} - \frac{3(\vec{S}_i \cdot \vec{r}_{ij})(\vec{S}_j \cdot \vec{r}_{ij})}{r_{ij}^5} \right], \quad (1)$$

where $D = \mu_0 \mu^2 / 4\pi a^3$ is the coupling constant of the dipolar interaction. The sum is either over all $l^2(2l^2 - 1)$ pairs of spins in the lattice for the case with open boundary conditions (OBC) or over all spins and their images for the case with periodic boundary conditions (PBC). We study these two possibilities; OBC is more related to the artificial spin ice fabricated in Ref. 7, while using PBC we minimize the border effects. In the system with PBC the Ewald summation[11,12] is used.

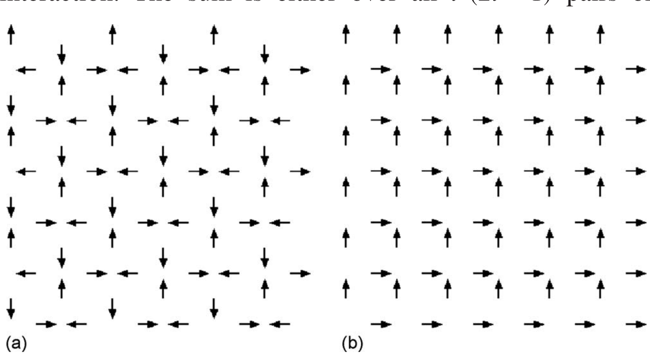

FIG. 2. (a) Configuration of the ground state obtained for $L = 6a$, in exact agreement with that experimentally observed. Note that the ice rules are manifested at each vertex. This is the case in which the topology demands the minimum energy see Fig. 3). (b) Another configuration also respecting ice rules, but displaying a topology which costs more energy.

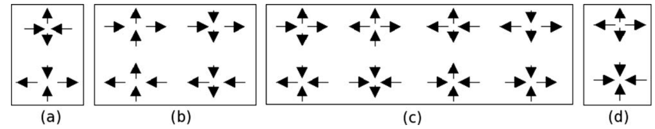

FIG. 3. The four distinct topologies and the 16 possible magnetic moment configurations on a vertex of four islands. Although configurations (a) and (b) obey the ice rule, the topology of (a) is more energetically favorable than that of (b). Equation (1) correctly yields to the true ground state based on topology (a), without further assumptions. Topologies (c) and (d) do not obey the ice rule. Particularly, (c) implies in a monopole with charge Q_M.

II. THE MODEL AND RESULTS

To start, we consider the ground states obtained from Eq. (1) describing the 2D spin ice. To do this we use a simulated annealing process,[13] which is a Monte Carlo calculation where the temperature is slightly reduced in each step of the process in order to drive the system to the global minimum. Our Monte Carlo scheme consist of a simple Metropolis algorithm.[13] In each Monte Carlo step (MCS) we attempt to flip all spins in the lattice sequentially or randomly which gives the same results. Several tests for systems with different sizes $L(6a \leq L \leq 80a)$ were studied. In each simulation $10 \times l^2$ MCSs were done at each temperature starting at $T = 3.0$ and decreasing the temperature in steps $\Delta T = 0.2$ until $T = 0.2$ (the temperature is measured in units of D/k_B). We observed that for $T < 0.4$ the system freezes, in the sense that all trial moves are rejected. The final configuration (ground state) was found to be twofold degenerate [see Fig. 2(a) for a lattice with $L = 6a$]. If we consider the vorticity in each plaquette, assigning a variable $\sigma = +1$ and -1 to clockwise and anticlockwise vorticities, respectively, the ground state looks like a checkerboard, with an antiferromagnetic arrangement of the σ variable. Note that the ground state clearly obeys the ice rule. We remark that it is impossible to minimize all dipole-dipole interactions. Actually, on each vertex there are six pairs of dipoles and only four of them can simultaneously minimize the energy. It is important to mention that although there are other possible configurations that also obey ice rules, these are not the ground state. Indeed, the state shown on the right side of Fig. 2 has energy about four times larger than that of the ground state. The difference between these two states is related to the distinct topologies for the configurations of the four moments (see Fig. 3). It was experimentally shown in Ref. 7 that while the topologies of types (a) and (b) obey the ice rule, case (a) has smaller energy than case (b). Our theoretical calculations confirm this fact. The same ground state was also reported in Refs. 8 and 9. We would like to remark that although this is the ground state, its thermal equilibration in experiments seems to be very difficult.[8–10]

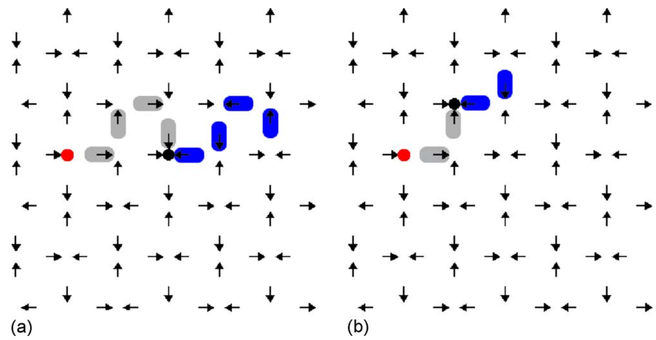

FIG. 4. (Color online) The two basic shortest strings used in the separation process of the magnetic charges: pictures (1) and (2) exhibit strings 1 and 2, respectively. The left circle is the positive charge (north pole) while the right circle is the negative (south pole).

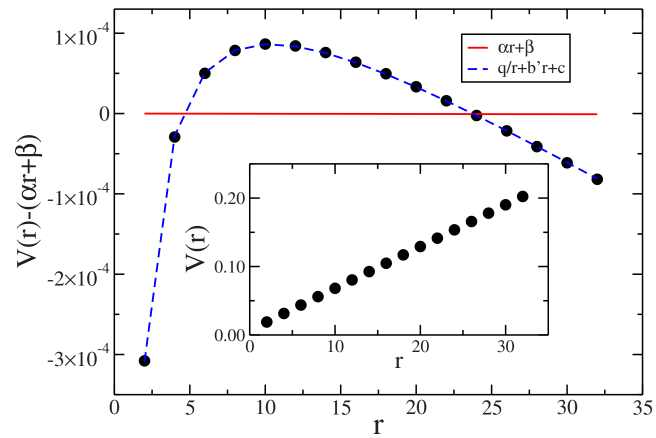

FIG. 5. (Color online) Inset: the interaction potential between two magnetic charges (with opposite signs) as a function of $r=R/a$. The baseline of $V(r)$ is also plotted: the curves are obtained by fitting the data to $\alpha R+\beta$ and $q/R+b'X+c$ minus $\alpha R+\beta$.

Once the system is naturally frustrated, in the two-in/two-out configuration, the effective magnetic charge $Q_M^{i,j}$ [number of spins pointing inward minus the number of spins pointing outward on each vertex (i,j)] is zero everywhere. The most elementary excited state involves inverting a single spin to generate localized "dipole magnetic charges," which implies in a "vortex-pair annihilation." Such an inversion corresponds to two adjacent sites with net magnetic charge $Q_M^{i,j}=\pm 1$, which is like a nearest-neighbor monopole-antimonopole pair. In principle, such "monopoles" can be separated from one another without violations of local neutrality by flipping a chain of adjacent spins. One can easily see that in this process, a "string" of spins pointing from the positive to the negative charge is created (see Fig. 4). The presence of a stringlike excitation joining these poles is evidenced by an extra energy cost behaving as bX, where X is the length of the string and $b>0$ is the effective string tension, as below. In order to establish a link between the monopole-antimonopole distance R and the string length X we choose two basic string shapes to move the charges as shown in Fig. 4. Of course, the shortest strings will be formed around the straight line joining the monopoles and, therefore, we choose two different ways in which they may be created as the charges are separated (see Fig. 4). First, using the string shape 1 and starting in the ground state we choose an arbitrary site and then the gray spins in Fig. 4 are flipped, thus creating a monopole-antimonopole separated by $R=2a$. In sequence, the spins marked in blue are flipped and the separation distance becomes $R=4a$ and so on. In this case $X=4R/2$. Note that the string surges in the system because in the separation process, the topology is locally modified, although still keeping the ice rule; in the region between the two poles, the topology of type (b), which has larger energy than that of type (a), prevails. Being essentially localized along the line joining the monopoles this additional amount of energy increases as the distance between the magnetic charges increases, justifying the bX term.

The potential $V(r)$ (the energy of the excited configuration minus the energy of the ground state) as a function of $r=R/a$ can be obtained by simple evaluation of the energy of each configuration. It is shown in the inset of Fig. 5 for the string shape presented in Fig. 4(a). The behavior is apparently linear but the function $f_q(R)=q/R+b'R+c$, with $q\approx -0.00122Da, b=b'/2\approx 0.00305D/a, c\approx 0.00734D$, fits better the data than the purely linear possibility $g(R)=\alpha R+\beta$, with $\alpha\approx 0.00611D/a, \beta\approx 0.00702D$. This difference becomes clearer when we analyze the χ^2/Dof, which is equal to 1.04×10^{-8} for the linear fitting and 4.5×10^{-13} for $f_q(R)$. Also, in Fig. 5 we draw a baseline of the potential using the linear fit. One can clearly see that $f_q(R)$ describes the data better and, therefore, $V(r)\approx f_q(R)$. The same method was repeated using the string shape 2. In this case, the charges are separated diagonally and $X=2R/\sqrt{2}$. The results are qualitatively the same and the values of the constants are: $q\approx -0.00125Da, b=b'/\sqrt{2}\approx 0.00317D/a, c\approx 0.00724D$. Note that the quantitative changes are small. The results are also qualitatively the same if PBC are used instead of OBC. Furthermore, quantitative differences between PBC and OBC calculations are smaller than 1% for constants b and c, while it is smaller than 9% for q. The larger difference for constant q can be understood if one remembers that the use of PBC will imply that the charges interact also with their images.

Our calculations yield the total energy cost of a monopole-antimonopole pair, separated by R, as the sum of the usual Coulombic-type term, q/R ($q<0$ is a constant), and an extra contribution behaving like bX, brought about from the stringlike excitations that bind the monopoles, so that $V(R)=q/R+bX(R)+c$ [$X(R)$ is the string length, while c is a constant associated to the monopole pair creation]. Until now we have only considered the shortest strings connecting two poles. However, many dipole strings of arbitrary shape and size can be identified that connect a given pair of monopoles. The associated energy cost increases with X and diverges with the length of the string. So, the monopoles should be confined in the artificial material. As we will argue later, it is possible that the string tension vanishes at a critical temperature proportional to b and hence, free magnetic monopoles may also be found in the 2D system.

For concreteness, the magnetic charge may be easily estimated if we take into account experimental values of some parameters. Considering the usual expression for the Coulombic interaction (in mks units) $-\mu_0 Q_M^2/4\pi R$, we get, $|q| =\mu_0 Q_M^2/4\pi$, or $Q_M\approx\pm\sqrt{4\pi|q|/\mu_0}\sim\pm 0.035\mu/a$. Now, using data from Ref. 7 (such as $a \sim 320$ nm and $\mu \sim 2.79 \times 10^{-16}$ JT^{-1}), the fundamental magnetic charge of an excitation in the array of ferromagnetic nanoislands reads $Q_M \approx 3 \times 10^{-11}$ cm/s, which is about 6×10^3 times smaller than the fundamental charge of the Dirac monopole ($Q_D = 2\pi\hbar/\mu_0 e$). Such a charge can even be tuned continuously by changing the lattice spacing.

III. DISCUSSION

Before concluding, it is important to analyze the behavior of the string tension as some parameters are varied in the system. The string tension for the artificial system built in Ref. 7 is approximately given by $b \approx 2.26 \times 10^{-15}$ J/s $\approx 4.5 \times 10^{-3}$ eV/a. Therefore, it is necessary a relatively large amount of energy (about 10^{-3} eV) to separate the "2D monopoles" by one lattice spacing, regardless of how far apart they are. Consequently, at low temperature, there is insufficient thermal energy to create long strings, and so the monopoles would be bound together tightly in pairs. The string tension can be artificially reduced by increasing the parameter $a(b \propto 1/a)$. However, it has also the effect of decreasing the magnetic charge since Q_M is proportional to $1/a$. A way to reduce b without affecting Q_M is increasing the temperature. By using the random walk argument, one can see that the many possible ways of connecting a pair of monopoles with a string give rise to a string configurational entropy proportional to R. Then, as the temperature increases, the string tension should decrease like $b - \epsilon k_B T$, with $\epsilon = O(a^{-1})$. It means that the string may lose its tension by entropic effect and, therefore, it should vanish at some critical temperature $k_B T_c$, of the order of $ba \sim 4.5 \times 10^{-3}$ eV. Another important point in this discussion is that as the temperature increases the monopoles density also increases. Indeed, if the pair creation energy is of the order of $E_c = V(a) \sim 1.3 \times 10^{-2}$ eV, one expects that, for temperatures above this value, the description in terms of monopoles itself could break down (in fact, the Boltzmann factor $\exp(-\beta E_c)$ would increase considerably for $k_B T \gg E_c$). Then, a possible deconfined phase would live between the melting temperature of the ordered and the dense monopole phases. A comparison between ba and E_c suggests that a temperature window between the confined and deconfined phases could be perfectly plausible in the range 4.5×10^{-3} eV $< k_B T < 1.3 \times 10^{-2}$ eV. Our expectation is that the window is still greater (starting at a much lower temperature), since the argument based on the balance of energy versus entropy may overestimate the critical temperature (for instance, the Berezinskii–Kosterlitz–Thouless critical temperature estimated by this argument for the planar rotator model is much higher than the correct value obtained by Monte Carlo simulations, which is $T_{BKT} \simeq 0.89J$,[14] where J is the coupling constant of the model). Once the deconfinement realizes, the question of technological applications of this system is relevant. For instance, learning how to move the magnetic monopoles around would be of importance toward technologies involving magnetic analogous of electric circuits.

However, there is another entropic effect, discussed in previous works of purely ice rule problem and related short-range problems[15–18] for strictly 2D systems, which may change the scenario of free monopoles. That is, the entropic interactions between monopoles due to the underlying spin configuration. Really, two monopoles should be attracted because there are more ways to arrange the surrounding dipoles in the lattice when they are close together. These entropic interactions, in a strictly 2D system, results in a 2D effective Coulomb attraction like $\ln R$, between oppositely charged monopoles, whose strength vanishes proportionally to T, at low temperatures (of course, in 3D materials, such entropic effect should result in a $1/R$ attraction). In our case, this logarithmic interaction could be present in addition to the 3D Coulombic, q/R, and linear bR interactions discussed in this work. Thus, at a temperature high enough to destroy the string tension, this entropically driven 2D Coulomb force would become crucial for keeping the monopole-antimonopole pairs bounded, in such a way that no free monopole phase would occur at any temperature. Nevertheless, how these monopoles precisely experience such an effect in their local dynamics should be investigated in more details. [We should recall that our calculations to obtain the interaction potential between monopoles have been performed at zero temperature and, consequently, this entropic contribution could not be directly (or even indirectly) present in $V(R)$]. The precise effect of the temperature on $V(R)$ is under investigation and will be communicated elsewhere. Here, it should be remarked that the present monopoles are not actually 2D objects: their physical interaction is given by the usual 3D Coulomb force, which means that they should affect magnetic test particles placed at relatively large distances along the direction perpendicular to the plane of islands (we remember that in a strictly 2D space, the magnetic field should be a pseudoscalar field. In addition, a genuine 2D monopole, as a counterpart of the Dirac pole, appears to be not magnetic charge; it rather looks like an exotic electric charge, giving rise to a rotational electric field, instead of radial-like, as usual charges do. For details, see Refs. 19–21). The dipoles forming the 2D lattice are genuinely 3D objects and their long-range dipolar interaction propagates in the 3D space [see Eq. (1)]. It is an important difference of this system when compared to the strictly 2D models such as vertex models and others. Now, it is also important to say that, such entropic interaction will not be accompanied by a magnetic field, it will not renormalize the monopole charge and it will not be felt by a stationary magnetic test particle.[5] Therefore, all calculations concerning the energy scales involved in the physical interactions between the defects will not be altered. In addition, it seems that this force has not been measured directly, yet. The peculiarities between strictly 2D models and the system studied here have not been considered previously. Thus, it is not completely clear if and how the entropically driven 2D Coulomb force acts in the spin ice with an inherent 3D spatial behavior.

Finally, we should remark that the above scenario involving the monopole physical interactions may be drastically changed if one considers these excitations in Fig. 2(b). As experimentally shown in Ref. 8, this metastable state is a very real possibility when magnetic fields are applied. In this case only the topology of Fig. 3(b) is present in the separation process. Further investigation is demanded for shedding extra light on this subject.

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