Conditions for free magnetic monopoles in nanoscale square arrays of dipolar spin ice

L. A. S. Mól,* W. A. Moura-Melo,† and A. R. Pereira‡
Departamento de Física, Universidade Federal de Viçosa, Viçosa 36570-000, MG, Brazil
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We study a modified frustrated dipolar array recently proposed by Möller and Moessner [Phys. Rev. Lett. **96**, 237202 (2006)], which is based on an array manufactured lithographically by Wang *et al.* [Nature (London) **439**, 303 (2006)] and consists of introducing a height offset h between islands (dipoles) pointing along the two different lattice directions. The ground states and excitations are studied as a function of h. We have found, in qualitative agreement with the results of Möller and Moessner, that the ground state changes for $h > h_1$, where $h_1 = 0.444a$ (a is the lattice parameter or distance between islands). In addition, the excitations above the ground-state behave like magnetic poles but confined by a string, whose tension decreases as h increases, in such a way that for $h \approx h_1$ its value is around 20 times smaller than that for $h=0$. The system exhibits an anisotropy in the sense that the string tension and magnetic charge depends significantly on the directions in which the monopoles are separated. In turn, the intensity of the magnetic charge abruptly changes when the monopoles are separated along the direction of the longest axis of the islands. Such a gap is attributed to the transition from the antiferromagnetic to the ferromagnetic ground state when $h = h_1$.

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I. INTRODUCTION

Geometrical frustration in magnetic materials occurs when the spins are constrained by geometry in such a way that the pairwise interaction energy cannot be simultaneously minimized for all constituents. A special example is an exotic class of crystalline solid known as spin ice ($Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$). Recently, Castelnovo *et al.*[1] have proposed that these materials are the repository of some elegant physical phenomena: for instance, collective excitations above its frustrated ground state surprisingly behave as pointlike objects that are the condensed-matter analogs of magnetic monopoles. Some recent experiments[2–5] have reported the observation and even the measurement of the magnetic charge and current of these monopoles in spin ice materials; in addition, simulations also support these ideas.[6,7] Besides, to turn the research of monopoles into a proper applied science, it will be necessary to ask if the basic ideas of dipole fractionalization[1,8] that give an usual spin-ice material its special properties can be realized in other magnetic settings. One of the most promising candidates for accomplishing that, is the artificial version of spin ices recently produced by Wang *et al.*[9] In this system, elongated magnetic nanoislands are regularly distributed in a two-dimensional square lattice. The longest axis of the islands alternate its orientation pointing in the direction of the two principal axis of the lattice.[9] The magnetocrystalline anisotropy of Permalloy (the magnetic material commonly used to fabricate artificial spin ice) is effectively zero so that the shape anisotropy of each island forces its magnetic moment to align along the largest axis making the islands effectively Ising-type. Actually, the fabrication and study of this kind of lower dimensional analogs of spin ice have received a lot of attention.[9–16] Indeed, the ability to manipulate the constituent degrees of freedom in condensed-matter systems and their interactions is much important toward advancing the understanding of a variety of natural phenomena. Particularly in this context, the possibility of observing magnetic monopoles in artificial spin ices[14,16] is a timely problem given that these magnetic compounds could provide the opportunity to see them up close and also watch them move (for example, with the aid of magnetic force microscopy). Very recently, the direct observation of these defects in an artificial kagome lattice was reported by Ladak *et al.*[17] However, there is a stimulating challenging for such an observation (or not) in artificial square lattices as pointed out in advance.

In a previous work[14] we have pointed out that monopoles do not appear as effective low-energy degrees of freedom in two-dimensional square spin ices, as they do in the three-dimensional materials $\{Dy,Ho\}_2Ti_2O_7$. Due to the antiferromagnetic order in the ground state, the constituents of a pair monopole-antimonopole become confined by a string which forbids them to move independently. However, we have also argued that above a critical temperature, the string configurational entropy may lose its tension leaving the monopoles free. The quantitative analysis of such a possible phase transition is under current investigation.[18] Meanwhile, other strategies to find monopoles in synthetic spin ices have been proposed. Möller and Moessner[16] have suggested a modification of the square lattice geometry in which they argue that, considering a special condition, the string tension vanishes at any temperature. This modification in the system produced by Wang *et al.*[9] consists of introducing a height offset h between islands pointing along the two different directions[10,16] (see Fig. 1; such a system is currently under experimental planning[19]). Their idea comprises basically the following: if h is chosen so that the energies of all vertices obeying ice rule become degenerate, then, an ice regime is established leaving the monopoles "free" to move (indeed, there is a Coulombic interaction between the monopoles).[16] For pointlike dipoles they considered that a degenerate state is obtained when the interactions between nearest neighbors (J_1) and next-nearest neighbors (J_2) are equal, leading to the following value for the height offset where free monopoles occur: $h_{ice} \approx 0.419a$ (where a is the lattice spacing).[10] Taking into account the finite extension of the dipoles, the height offset diminishes and as $\epsilon \equiv 1 - l/a \to 0$ (l is the length of the island), the end points of the islands form a tetrahedron so that at $h = \epsilon a/\sqrt{2}$ the ordering disappears and the monopoles become free to move.[16]

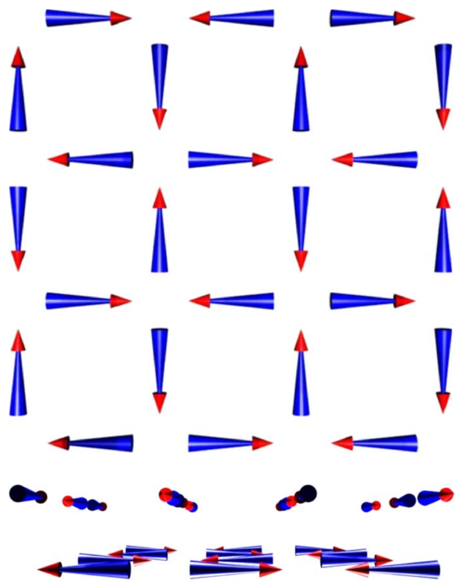

FIG. 1. (Color online) The modified square lattice studied in this work. Top: top view of the system. The arrows represent the local dipole moments ($\vec{S}_{\alpha(i)}$ or $\vec{S}_{\beta(i)}$). Bottom: lateral view of the system showing the height offset between islands. The original material produced by Wang *et al.*(Ref. 9) is two-dimensional with $h=0$.

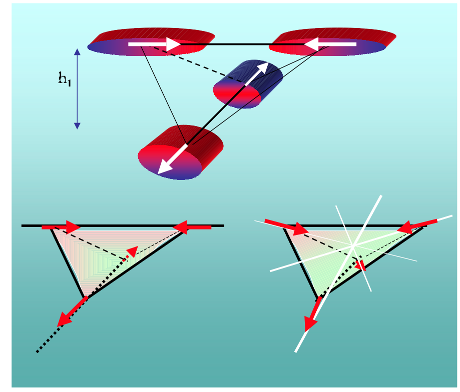

FIG. 2. (Color online) Up: in the artificial spin ice proposed in Ref. 10, the spins obeying the ice rule do not point along directions passing by the center of a tetrahedron as they do in the natural spin ice compounds. Down: configurations of the spins obeying the ice rule in a tetrahedron in the artificial (left) and the natural (right) spin ices. This small distortion of the spins configuration causes a residual ordering and consequently, an outstanding energetic string connects the monopoles in the modified artificial system.

Here we numerically calculate the energetics of the ground states and excitations in the modified square lattice as a function of h. In our calculations we consider pointlike dipoles forming the lattice. Although the main physical aspects of the system must be correct with this approximation, some parameter values (such as magnetic charge, string tension, critical height, etc.) should be quantitatively altered for the realistic case in which l has a finite length. On the other hand since we take into account all the long-range dipole-dipole interactions, it is expected that our results could better describe the actual system. For instance, while in the calculations of Refs. 10 and 16, the ground-state changes its configuration at $h=0.419a$, our results indicate that it occurs at $h=h_1=0.444a$. Besides, we noted that at least one of the several configurations that satisfy the ice rule does not have the same energy of the "ground-states" (GS_1 and GS_2) at this very height, indicating that for $h=h_1$ the system is not in a completely degenerate state. We have also shown that the string tension decreases rapidly as h increases but it does not vanish at any value ($h\leq a$): rather, at $h=h_1$, its strength reads about 20 times smaller than that of the usual case for $h=0$. A possible cause of the finite strength of the string tension even at h_1 is the fact that, concerning the spin configurations in a tetrahedron, the artificial spin ice has a slight difference with its natural counterpart. For the artificial compounds proposed in Ref. 16, the localized magnetic moments forming a corner-sharing tetrahedral lattice are forced to point along the longest axis of the islands (here, x or y directions, see Fig. 2) while in the original $3d$ spin ices, they point along a $\langle 111\rangle$ axis (indeed, in this case, the magnetic dipoles point along axes that meet at the centers of tetrahedra). As a result of this mismatch, there is always a single ordered ground state in the artificial systems, which is responsible for the residual value of the string tension and its anisotropy. Another interesting result obtained here with the pointlike dipole approximation is that the magnetic charge of the monopoles jumps as the system undergoes a transition in its ground state. In addition, in general, this strength of the interaction between a monopole and its antimonopole is anisotropic, depending on the lattice direction and on the type of order. However, as expected from the above discussions, we note that the system anisotropy diminishes as h goes to h_1. Actually, as h increases from zero, the differences found in the values of the "charges" (as distinct directions for the monopoles separation are taken into account) decreases, and they tend to disappear as $h\rightarrow h_1$, i.e., in the ice regime (nevertheless, $h=h_1$ is not really an optimal ice regime, at least for pointlike dipoles).

II. MODEL AND RESULTS

We model the system suggested in Refs. 10 and 16 assuming the magnetic moment (spin) of the island is replaced by a point dipole at its center. At each site (x_i,y_i,z_i) of a "square" lattice two spin variables are defined: $\vec{S}_{\alpha(i)}$ with components $S_x=\pm 1$, $S_y=0$, and $S_z=0$ located at $\vec{r}_\alpha=(x_i+a/2,y_i,h)$ and $\vec{S}_{\beta(i)}$ with components $S_x=0$, $S_y=\pm 1$, and $S_z=0$ at $\vec{r}_\beta=(x_i,y_i+a/2,0)$. Spins pointing along the y direction and spins pointing along the x direction are in different planes, separated by a height h (see Fig. 1). Hence, in a lattice of volume $L^2=n^2a^2$ one gets $2\times n^2$ spins (we have studied systems with $n=20$, 30, 40, 50, 60, and 70). Representing the spins of the islands by $\vec{S}_i$, assuming either $\vec{S}_{\alpha(i)}$ or $\vec{S}_{\beta(i)}$, then the modified artificial spin ice is described by the following Hamiltonian:

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

where $D=\mu_0\mu^2/4\pi a^3$ is the coupling constant of the dipolar interaction. The sum is performed over all $n^2(2n^2-1)$ pairs

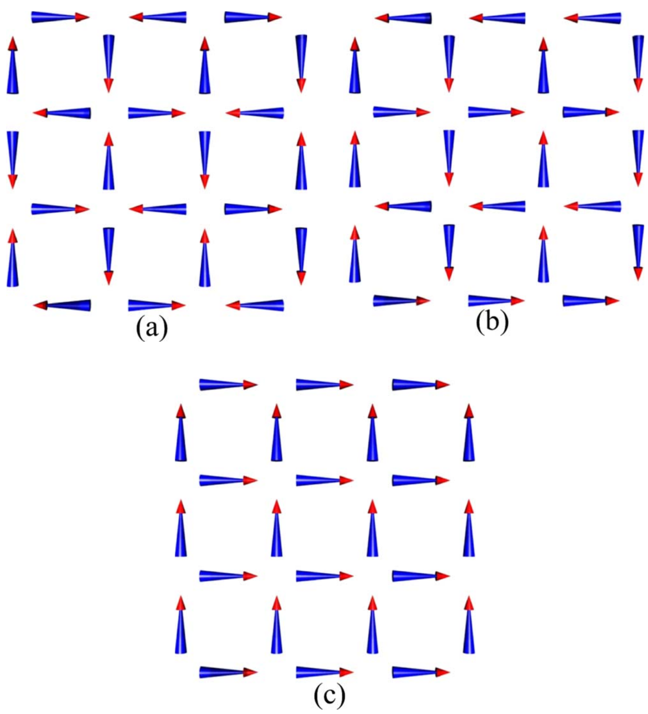

FIG. 3. (Color online) (a) Ground-state configuration for $h<h_1=0.444a$, GS_1. Note that this is exactly the same state obtained in Refs. 10 and 14. (b) Configuration of the ground state GS_2 obtained for $h>0.444$. In GS_2, each vertex has a net magnetization but globally the magnetization vanishes. Note that the ice rule is manifested in every vertex. (c) Another configuration that satisfy the ice rule but has an energy higher than the configurations shown in (a) and (b) when $h=h_1$.

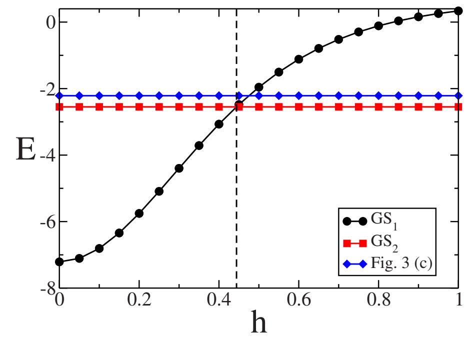

FIG. 4. (Color online) The energy per island of the two ground states (GS_1 and GS_2) and of the configuration shown in Fig. 3(c) (in units of D) as a function of h (in units of the lattice spacing a). Black circles represent the GS_1 energy while red squares concern GS_2 and blue diamonds are for the configuration shown in Fig. 3(c).

of spins in the lattice for open boundary conditions (OBC) while for periodic boundary conditions (PBC) a cut-off radius was introduced when $r_{ij}>n/2a$.

The results presented here consider a lattice with $n=70$, which contains 9800 dipoles (islands) and PBC. We observed exactly the same behavior for OBC and PBC and the size dependence of the results is not appreciable. By using a simulating annealing process (see Ref. 14), the first thing to notice is that the ground-state configuration changes for a critical value of h. Indeed, as shown in Fig. 3(a), for all values $h<h_1=0.444a$, the system ground state (hereafter referred to as GS_1) has exactly the same form as that of the usual case in which $h=0$. However, for $h>h_1$, the ground-state changes to GS_2 [see Fig. 3(b)]. Really, as $h\rightarrow h_1$, the energies of both states are comparable, whereas for $h>h_1$ the state GS_2 is less energetic (see Fig. 4). Such a result is in qualitative agreement with findings of Ref. 10, which presents the transition at $h=h_{ice}=0.419a$. As expected, both configurations obey the ice rule (two spins point in and two point out in every vertex), but while in GS_1 the magnetization is zero at each vertex, in GS_2 it points diagonally, but with net vanishing magnetization. As shown in Fig. 4, the energy of GS_1 increases rapidly as h increases while the energy of GS_2 is constant. Actually, for this latter configuration, the horizontal and vertical sublattices are decoupled. We note that these two ground states are metastable in the sense that they are local minima and cannot be continuously deformed one into another without spending a considerable amount of energy; trying to align the dipoles from one state to another costs the inversion of two spins by vertex (half of the spins have to be inverted in the whole system). This changing has an h-dependent energy barrier which is roughly on the order of 160 D (for $h=0.444a$ and $n=70$), making this process much improbable to occur spontaneously. Thus, considering the system in the GS_1 state and increasing continuously the height from $h=0$, GS_1 may persist even for $h>h_1$ because of the large energy necessary to change to GS_2. Besides, in Fig. 4 we also present the energy of the configuration shown in Fig. 3(c), which also satisfy the ice rule but has an energy higher than those of GS_1 and GS_2 even for $h=h_1$. Consequently, the states satisfying the ice rule are not completely degenerate.

Now, we consider the excitations above the ground state. 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 for $h<h_1$ (GS_1) and for $h>h_1$ (GS_2). The most elementary excited state is obtained by inverting a single dipole to generate localized "dipole magnetic charges." Such an inversion corresponds to two adjacent sites with net magnetic charge $Q_M^{i,j}=\pm 1$ which is alike a nearest-neighbor monopole-antimonopole pair.[1,14] Following the same method of Ref. 14, it is easy to observe that such "monopoles" can be separated from each other without violating the local neutrality by flipping a chain of adjacent spins. We choose four different ways they may be separated (see Fig. 5). First, using the string shape 1 and starting in the ground state GS_1 (for $h<h_1=0.444a$) we choose an arbitrary site and then the spins marked in dark gray in Fig. 5 are flipped, creating a monopole-antimonopole separated by $R=2a$. Next, the spins marked in light gray are flipped and the separation distance becomes $R=4a$, and so on. In this case, the string length (X) is related to the charges separation distance R by $X=4R/2$ (the monopole and the antimonopole will be found along the same horizontal or vertical line). Second, we also consider a string path of form 2 (for $h<h_1$), making the separated monopoles to be found in different lines (diagonally positioned; now we have $X=2R/\sqrt{2}$). More two equivalent ways were studied for $h>h_1$ in which GS_2 is the ground state. In

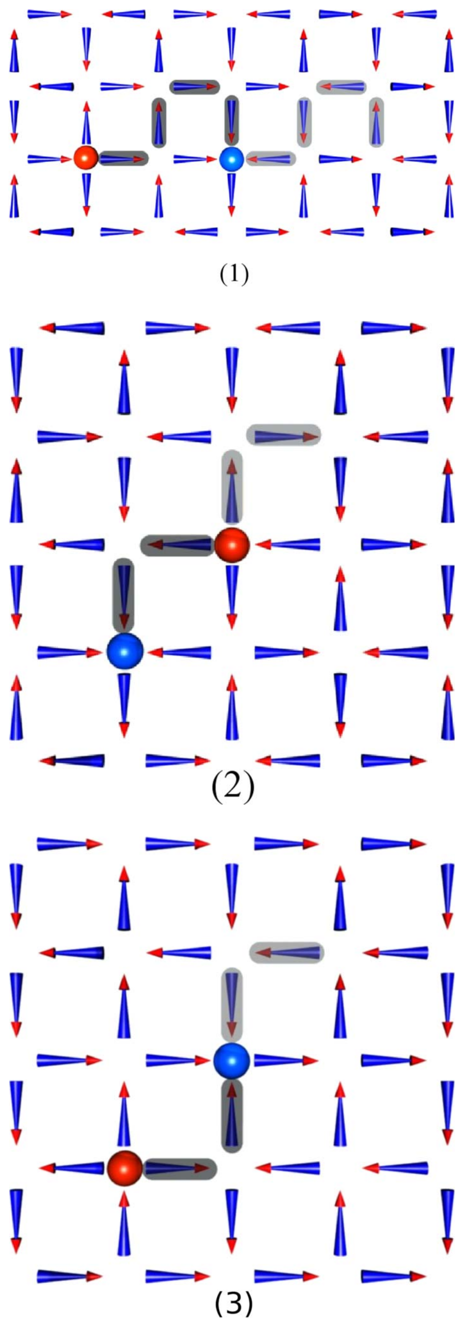

FIG. 5. (Color online) Three of the four basic shortest strings used in the separation process of the magnetic charges. Pictures (1) and (2) exhibit strings 1 and 2, respectively, used for $h<h_1=0.444a$. The red circle is the positive charge (north pole) while the blue circle is the negative (south pole). For $h>h_1$ the ground state is GS_2 and we used a linear string path (not shown above) and a diagonal path [picture (3)].

this case, however, differently from the situation in the GS_1 state, now the monopoles can be separated by using a linear string path (so that $X=R$) without any violation of the ice rule. Finally, another monopoles separation studied for GS_2 is the "diagonal path" (or path 3), in which the charges are put in different lines. Our analysis shows that besides the Coulombic-type term $q(h)/R$ (where $q=\frac{\mu_0}{4\pi}q_1q_2<0$ is the coupling constant which gives the strength of the interac-

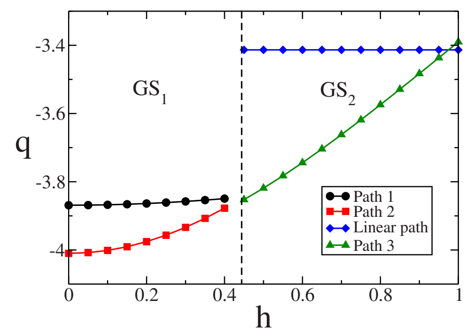

FIG. 6. (Color online) The monopole charge q [see Eq. (2)] obtained analyzing the energy in the separation process of the charges for the two string shapes shown in Fig. 5 for $h<h_1$. When $h>h_1$, the charges variation is shown for a linear and diagonal string paths. Here, q is in units of Da while h in units of a. Note how the anisotropy of the monopole interaction decreases considerably as $h \to h_1$ from below.

tion), the total-energy cost of a monopole-antimonopole pair has an extra contribution behaving like $b(h)X$, brought about by the stringlike excitation that binds the monopoles, say,

$$V(R,h) = q(h)/R + b(h)X(R) + V_0(h), \tag{2}$$

where $V_0(h)$ is a h-depended constant related to the monopole pair creation (for instance, for $h=0$ $V_0(0)\approx 23D$ and $V(a,0)\approx 29D$). The results for the "charge" $q(h)$ are shown in Fig. 6 for the range $0<h<a$. When $h<h_1$, the excitations are considered above GS_1 and we observe that there is a small h-dependent difference in the q value for paths 1 and 2, which vanishes as $h\to h_1$. At higher heights $h>h_1$, q is valued with respect to GS_2 and is h independent for a linear string path. However, for path 3, it comes back to increase as h increases. Therefore, the interaction of a monopole with its partner (antimonopole) is anisotropic in artificial spin ices. Perhaps, it would be more appropriate to redefine things in such a way that $q=\frac{\mu_0}{4\pi}Q_1Q_2\alpha(h,\phi)$, where $q_1q_2 = Q_1Q_2\alpha(h,\phi)$ and the actual value of the charges $Q_1=-Q_2$ is independent of the angle ϕ that the line connecting the poles makes with the x axis. In this case, the anisotropy of the interaction (coming from the background) is implicitly considered in the function $\alpha(h,\phi)$ but its complete expression was not evaluated here. Since $\alpha(h_1,\phi)$ tends to be a constant (independent of ϕ, we set $\alpha(h_1,\phi)=1$ and so, only around the ice regime (i.e., $h\approx h_1$) the interaction tends to be isotropic. Thus we can find the genuine strength of the magnetic charge in this artificial compound as being $Q_1=\pm\sqrt{4\pi|q(h_1)|/\mu_0}\approx\pm 1.95\ \mu/a$, where we have used $|q(h_1)|=3.8$ Da. Just for effect of comparison, using some parameters of Ref. 9 such as $a=320$ nm, we get a charge value which is about 80 times larger than the typical value found for the original $3d$ spin ices[1] (or about 100 times smaller than the Dirac fundamental charge). Besides its anisotropy, another interesting fact about the Coulombic interaction in the artificial compounds is that it jumps at $h=h_1$. Indeed, at this point, q abruptly changes from $q_<\approx -3.8$ Da to $q_>\approx -3.4$ Da when the linear path is taken into account. Such a discontinuity may be attributed to the

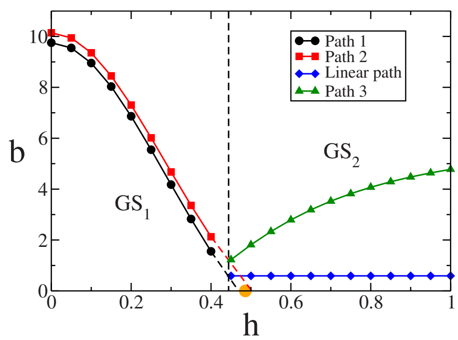

FIG. 7. (Color online) The string tension for the two string shapes shown in Fig. 5 for $h<h_1$ and for a linear string path and path 3 (diagonal) for $h>h_1$. The green dot and the dashed lines represents an extrapolation of our data.

ground-state transition and that above GS$_2$ the Coulombic interaction between a pair somewhat incorporates the residual magnetization stored in each vertex. On the other hand, keeping a diagonal separation of the monopoles along the ground-state transition, the magnetic charge parameter q increases almost continuously.

How the string tension b depends upon h is shown in Fig. 7. Note that while GS$_1$ is the ground state ($h<h_1$), b diminishes as h increases. At higher heights, and being evaluated over GS$_2$, the tension remains a nonvanishing small constant for linear path and turns back to increase for diagonal separation (path 3). In general, since b is also a function of ϕ [i.e., $b(h,\phi)$], it is more favorable energetically that a pole and its antipole reside at the same line in the array. It should be remarked that, near the ice regime, $b(h_1,\phi)$ is almost independent of ϕ (almost isotropic limit) and its value is reduced around 20 times whenever this modified system is compared to its counterpart at $h=0$ (at zero temperature). In principle, this result indicates that free monopoles do not appear in this system. Then, the modified array[16] faces a small obstacle by the fact that the islands are placed in such a way that the spins cannot point to the center of a tetrahedron as they do in the 3d materials. Indeed, as pointed out before, the spins in the artificial compound point along its edges (see Fig. 2); the islands are rigid objects that do permit the spins to point only along their longest axis. This disparity causes an ordering in the artificial material, which diminishes as h increases; eventually it becomes tiny but persists at $h=h_1$. This persistent ordering contributes for the residual string tension at the ice regime and also for the different string tension values as the monopoles are located at different angular positions in the array. Such a difficulty may be overcome when one takes the limit $l\rightarrow a$ in the modified array. As pointed out in Ref. 10, the mechanism responsible for the equivalence between the artificial (2d) and natural 3d spin ices is not operational in $d=2$, as it requires also the dimensionality of the dipolar interaction to coincide with that of the underlying lattice. Here, we have a $d=3$ dipolar ($1/r^3$) and Coulombic ("monopolar," $1/R$) interactions in a two-dimensional array. Independent of this since the state GS$_1$ is metastable one could imagine if the excitations could be considered to lie in GS$_1$ for h slightly greater than h_1. In this case the extrapolation of our results indicate that the string tension may vanish at $h\approx 0.502a$ (see Fig. 7).

III. SUMMARY

In summary, we have investigated the energetics of the modified artificial spin ice expressing several quantities, such as ground-states energy, magnetic charges and string tension, as a function of the height offset h. Our analysis show that the ground-state changes from an ordered antiferromagnetic to a ferromagnetic one at $h=h_1\approx 0.444a$, which is in good agreement with the value obtained in Refs. 10 and 16, $h_{ice}\approx 0.419a$. We claim that such a small difference comes about from the fact that in these cited works, authors assumed equal nearest-neighbor and next-nearest-neighbor interactions, whereas we have taken all the dipole interactions into account. For the excitations above the ground state we have found that the magnetic charges interact through the Coulomb potential added by a linear confining term with tension $b(h)$ which decreases rapidly as h increases from 0 to h_1, assuming a nonvanishing constant value at higher h. Actually, the system presents an anisotropy that manifests itself in both the Coulombic and linear interactions and it tends to diminish as h increases, almost disappearing at $h=h_1$. The source of this anisotropy is a residual ordering, which still persists even in the ice regime (at $h=h_1$ for pointlike dipoles). Ordering and anisotropy may disappear completely in the ideal limit $l\rightarrow a$ and $h\rightarrow 0$. Another interesting result is that the magnetic charge jumps, depending on the direction in which the monopoles are separated, as the system undergoes a transition in its ground state. For a separation of the monopoles, vertex by vertex, along the same line of vertices, which is possible only for the GS$_2$ ground state, the coupling q exhibits considerably discontinuity in relation to its limit value in the GS$_1$ ground state. On the other hand, it tends to grow up continuously for the diagonal path along the transition. Although the residual ordering leads to a confining scenario for monopoles, its very small strength, whenever $h\approx h_1$, signalizes a significant tendency of monopole-pair unbinding at a critical (optimal) height offset, even at zero temperature. Further improvements in model (1), for instance, taking the actual finite size of the dipoles into consideration could shed some extra light to this issue. Additionally, temperature effects may also facilitate the conditions for free monopoles. Indeed, the string configurational entropy is also proportional to the string size and therefore, at a critical temperature,[14] on the order of ba, the monopoles may become free. In view of that, for small b, the monopoles should be found unbind at very low temperatures. As a final remark we would like to stress that these results show that the background configuration of spins has a deep effect in the charges interactions, being responsible for the string tension, anisotropies and a kind of screening of the charges.

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*lucasmol@ufv.br
†winder@ufv.br
‡apereira@ufv.br

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