Magnetic van der Waals-like framework in bilayer artificial spin ice

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We study an artificial spin ice system consisting by two identical layers separated by a height offset $h$. For short separation, the layers are shown to attract each other, provided the whole system is in the ground state. Such an attraction comes about by means of a power-law force compared to van der Waals forces. Whenever magnetic monopoles show up in one (or both) layers, the scenario becomes even more interesting and these layers may also repel each other. By tuning parameters like $h$ and monopole distance, switching between attraction and repulsion may be accomplished in a feasible way. Regarding its thermodynamics, the specific heat peak shifts to lower temperature as $h$ increases.

Keywords: artificial spin ice, geometrical frustration, magnetic monopole, van der Waals forces

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I. INTRODUCTION AND MOTIVATION

Geometrical frustration is an interesting phenomenon which has received a lot of attention recently\(^\text{1}\). In magnetism, it arises whenever interaction between magnetic degrees of freedom is incongruous regarding the lattice underlying the crystal geometry. Frustration emerges in appealing natural materials\(^2\)–\(^4\), and it can also be created by design\(^5\). Indeed, artificial systems have been built in diverse configurations which allow us to control frustration by experimentally tuning suitable parameters. An important class of such designed systems is provided by artificial spin ice (ASI) arrangements\(^6\)–\(^20\), which essentially consists of a planar-type regular array of nanosized elongated ferromagnetic rods where geometrical frustration takes place at the vertices. By virtue of strong shape anisotropy along the major axis, every nanoisland effectively behaves as an Ising-type dipole. Now, the collective interaction among all these dipoles yields surprising emergent phenomena, such as fractionalization. Actually, above the ground state the most elementary excitations show up as magnetic monopoles, coupled in pairs by energetic strings\(^7\),\(^8\), which are flux-carrying magnetized chains. In words, the original degrees of freedom, the usual magnetic dipoles, have been fractionalized into isolated monopoles emerging at ASI vertices. Although they had been originally named Dirac monopoles and strings, it is more suitable to speak about Nambu monopoles and strings as claimed in Refs.\(^\text{21,22}\), after Nambu picture adaptation of Dirac description to a London-type framework\(^\text{23,24}\). Such a magnetic scenery has been observed to occur in distinct ASI lattice geometries including square, rectangular, triangular, and kagome arrangements.

The interest in their physical properties lies in the fact that such systems are promising candidates for new technologies based upon the control of magnetic charges and their currents, something termed as magnetricity and magnetronic. Actually, magnetic charge flow was firstly realized in \(\text{Dy}_2\text{Ti}_2\text{O}_7\) compound, an example of natural three-dimensional spin ice crystal, but at very low temperature \(\sim 200 - 300\text{mK}\), see Refs.\(^\text{25}\). At room temperature, an ordered magnetic current has been observed in an unidirectional arrangement of patterned nanoislands\(^\text{26}\), where no geometrical frustration takes place at all. In turn, even though theoretical studies regarding three-dimensional (3D) ASI appeared more than a decade ago\(^\text{10,12,32}\), their experimental realization took place only very recently\(^\text{30,31}\), which has been achieved due to novel advances in patterning 3D magnetic nanostructures\(^\text{27}\). These
systems consist of only one ASI built by offsetting one of the sublattices by some height $h$ such that the energy of interaction between all nearest neighbours becomes equivalent, allowing this arrangement to undergo a transition to a magnetic Coulomb phase\cite{10,12,30,32}. In order to shed further light onto such a scenery, we investigate a rather similar system whose layers contain nanoislands arranged like a square lattice. Then, we study a bilayer artificial spin ice (BASI, for short), where interactions take place among all the islands of both layers (see Fig. 1). Since each island behaves like an Ising-type dipole, we can envision a framework where van der Waals-like magnetic forces will show up as the collective interaction of these dipoles.

Indeed, van der Waals (vdW) forces arise from the mutual coupling among electric dipoles composing a system. Although two ideal dipoles interact like $r^{-3}$-potential, whenever effects like orientation, induction and dispersion are taken into account the net interaction goes like $V_{vdW}(r) \sim r^{-6}$, see Ref.\cite{27}. Such forces are keystones to understand how atoms and molecules combine to form gaseous, liquid and solid substances\cite{27,28}. More recently, vdW interaction has become increasingly important for a better understanding of layered compounds, including graphene and other 2$D$-materials\cite{29}. Indeed, in such materials the ions are held by strong covalent bonds whereas the layers experience weak out-of-plane vdW forces, making ease their exfoliation. Our main findings give further support to such an envisioning, since the layers composing the BASI interact through a potential, $V(h) \sim h^{-6.4}$, with power-law approximate to vdW potential, $V_{vdW}(r) \sim r^{-6}$. The deviation may be explained by geometrical frustration taking place in each layer, which forbids the Ising-type dipoles of achieving a unique ground-state. We also study the interaction between magnetic monopoles showing up as excitations in the Coulomb phase. In Section II we present our model and methods, while results and discussion is left to Section III. We close our paper by presenting our conclusions and prospects for forthcoming research.

II. MODEL AND METHODS

Every square ASI is described by a dipolar Hamiltonian like follows

$$H_D = D a^3 \sum_{i,j} \left[ \hat{e}_i \cdot \hat{e}_j - \frac{3(\hat{e}_i \cdot \bar{r}_{ij})(\hat{e}_j \cdot \bar{r}_{ij})}{r_{ij}^5} \right] S_i S_j,$$

(1)
FIG. 1. (a): the four possible classes of vertices in a single monolayer square ASI vertex: \( t_1, t_2, t_3 \) and \( t_4 \). Each class comprises different vertex types that share the same energy. The first two classes obey the ice-like rule: two spins point \( in \) while the other two point \( out \) of the vertex center, in short \( 2in-2out \), while the other topologies violate it. (b): sketch of BASI system showing the square arrangement of each layer with lattice spacing \( a \) and separated by height offset \( h \). (c): each layer displays ground state composed only by \( t_1 \) vertices (upper layer is GS1). (d): now, bottom layer presents GS2. (e) potential between the layers is plotted as a function of \( h \). It is noteworthy that GS1 – GS2 is the true combined ground state (for small offset, \( h \lesssim a \)). Indeed, in this case, the layers experience a mutual attraction, while for GS1 – GS1 a repulsive potential tends to keep them each apart other. Note also that this potential decays rapidly, \( V \sim h^{-6.4} \), and practically falls off as \( h \gtrsim 1.5a \).

where \( D = \frac{\mu_0}{4\pi} \frac{\mu^2}{a^3} \) is the dipole-dipole coupling constant, \( \hat{e}_i \) is the local Ising axes of the lattice, \( r_{ij} \) is the distance between \( S_i \) and \( S_j \), and \( S_i = \pm 1 \) accounts for the unity magnetic momenta states which behave as effective Ising spins due to shape anisotropy of elongated nanoislands. [For typical ASI arrangements, each nanoisland carry \( \mu \sim 10^6 - 10^7 \mu_B \) (\( \mu_B \)
is the Bohr magneton) and they are separated by lattice spacing $a \sim 10^2 \text{nm}$, so that $D \sim 10^{-18} - 10^{-20} \text{J}$. In words, if we have a single ASI layer, then such vectors run over directions $x$ and $y$, confined on the layer plane. Whenever a second layer is taken into the game the mutual interaction between the layers must also be accounted. This is accomplished in a simple way just allowing a third component for $r_{ij}$ vectors, so that it may also compute the interaction between pairs of nanoislands belonging to distinct layers as well (clearly, vectors $\hat{e}_i S_i$ are kept on layers planes, say, only with $x$ and $y$ components).

If one intends to bring a third layer, one simply permits $r_{ij}$ to run over its vertices, and so on.

Our simulation is carried out by considering two ASI layers parallel each other and separated by a height offset, $h$. Each layer comprises $29 \times 29 = 841$ vertices comprising a total of $N = 3480$ magnetic moments disposed in a square lattice. Our first task is to determine the combined ground state of the coupled layers as function of $h$. This is done by starting off from a disordered state at a very high temperature; later, the system is driven to very slow dynamics by cooling it to very low temperature, $\sim 0.1D/k_B$.

III. RESULTS AND DISCUSSION

Since $t1$ topology bears the lowest energy, one could thought that any combination of its vertices in both layers would yield the ground state. However this is not true at all. Actually, a single layer achieves its ground state by choosing any combination of $t1$ vertices. But, whenever mutual coupling between them is considered, then things happen in a quite different way. To better realize the scenario, we firstly remark that the ground state of a single ASI is doubly degenerated ($GS1$ and $GS2$) and populated only by $t1$-class. Let $t1$ class split in upper and lower vertices (see Fig. 1); so, in both ground states, the neighbor vertices alternate $t1$-upper with $t1$-lower. In addition, if an arbitrary vertex in an ASI is $t1$-upper in the ground state $GS1$, then, the same vertex would be $t1$-lower in the ground state $GS2$. The first excited state demands the appearance of $t2$ and $t3$ vertices: $t3$ support the monopole-antimonopole pair (with unity and opposite charges $\pm 1$, blue and red spots) joined by an energetic string which is a segment of $t2$ vertices. $t4$ vertices support double-charged monopoles, demanding much higher energies to show up. Clearly, for a single layer
GS1 or GS2 yields one possible ASI ground state. However, whenever taken as a combined system, BASI ground state is achieved in such a way that if one layer is at, say, GS1, the other must be GS2. In this case, the layers experience a considerable attraction, whose potential goes like $V(h) = -78 (h/a)^{-6.4}$ ($V$ is measured in units of the dipolar constant, $D$). On the other, if the layers were put side by side exhibiting the same individual ground state, say, GS1 or GS2 in both, then they would strongly repel each other according to $V(h) = +78 (h/a)^{-6.4}$ (see Fig.1). Other combined ground states are possible, provided that every vertex in one layer alternate as upper/lower types regarding its nearest neighbor lying on the other ASI layer. This is clearly accomplished by the combined GS1 – GS2 ground state. With such an alternating, a similar scenery also holds for $t2$ topology: $t2$-up in a layer combined with $t2$-down in another yields attraction, whereas same type vertex at both leads to a repulsive regime. In these cases, since all vertices in $t2$ class bear net magnetization, attractive or repulsive force acquires an additional contribution due to the mutual interaction among these magnetized vertices. Another example is provided by $t1$ vertices in one layer while $t2$-class in the other, which yields to a repulsive regime. As a whole, it should be emphasized that the true ground state(s) configuration depends upon $h$; here, we have determined it for small $h$. Indeed, as $h$ becomes larger, $h \gtrsim 1.5a$, BASI is practically decoupled and one has two non-interacting ASI systems. [Indeed, its basic thermodynamics puts an even more stringent value, indicating that for $h > a$ one effectively has two decoupled layers, as discussed later].

As a whole, once the original degrees of freedom are dipoles interacting via model Hamiltonian \cite{1}, the above findings suggest that we are faced with a magnetic van der Waals-like framework. In general both layers are expected to experience the potential $V \sim h^{-6.4}$. The deviation from vdW power-law potential, $V_{vdW} \sim \ell^{-6}$ may be attributed to strong Ising anisotropy that just allows each dipole to flip, along with the geometric frustration, coming from the rigid lattice geometry arrangement. Besides this numerical deviation in the potential, in a BASI not only attraction takes place: layers also repel each other depending on their dipole configurations. Additionally, the attraction between ASI layers at ground state (vacuum sate) may be faced as a classical magnetic analog of the famous Casimir effect which describes the attraction of two perfectly conducting neutral parallel plates due to vacuum fluctuations. At zero temperature, Casimir potential also obeys a power
law behavior, \( \mathcal{V}_C = -\left(\frac{\pi^2 \hbar c}{720}\right) A \ell^{-3} \sim 10^{-28} A \ell^{-3} \) \((A\text{ is the area of one of the plates which are separated by } \ell)\). Indeed, Casimir pressure is so tiny, \( F_C/A \sim 10^{-27} \ell^{-4} (\text{N/m}^2) \), which jeopardized its experimental demonstration for around half-century (for a review, see Ref\(^{34}\)). In a BASI, the force between the layers goes like \( F \approx 500 (h/a)^{-7.4} \) (in units of \( D/a \sim 10^{-13} \sim 10^{-11} \text{N} \)). In addition, each ASI vertex in typical arrangements comprises an area \( \sim 10^{-13} \text{m}^2 \) (since the major axis of a nanoisland goes around a few \( a \)), then each ASI layer has a total area of \( A \sim 10^{-10} \text{m}^2 \) (each layer comprises \( 29 \times 29 = 841 \) vertices, as aforementioned), yielding a magnetic pressure \( F/A \sim 1 - 10^2 (h/a)^{-7.4} (\text{N/m}^2) \) which is generally much higher than its Casimir counterpart discussed above. Actually, if BASI layers were considered as neutral plates separated \( \ell = a \sim 10^2 \text{nm} \), then they would attract each other with a Casimir force \( F_C \sim 10^{-17} \text{N} \), while BASI layers at \( h = a \) interact with magnetic force \( F(h = a) \sim 10^{-11} - 10^{-9} \text{N} \).

For practical purpose, the repulsion between BASI layers can be exploited as a kind of magnetic levitation or magnetic damping system at nanoscale. Switching between magnetic attraction/repulsion can be achieved whenever one may drive one of the layers from \( GS_1 \) to \( GS_2 \). Additionally, in the realm of a bilayer system, for instance configurations \( GS_1 \) and \( GS_2 \) may be also viewed as being opposite density of magnetic charges, then reproducing the usual fact that opposite charges attract while like charges repel each other. By choosing those combinations carefully enough, one might design a kind of stable ‘BASI molecule’, where the attractive and repulsive forces balance out. For instance, one may conceive an interesting situation where one layer is fixed while another rotates so that attractive and repulsive forces alternate yielding oscillation to this molecule. These and other proposals are appealing nowadays since actual devices are rapidly shrinking to nanometer scale giving the possibility of tuning the magnitude of BASI magnetic force on demand.

Now, we depart to investigate the appearance of excitations above the ground state and how they change the previous results. At each ASI layer these excitations emerge as magnetic monopole-antimonopole pairs connected by energetic strings. We then start off by considering BASI ground state given by \( GS_1 - GS_2 \), say, \( GS_1 \) in the layer 1 along with \( GS_2 \) in the layer 2. The simplest excited state is obtained by flipping a single magnetic dipole from layer 1 (a discrete rotation of 180 degrees; no flip is performed in layer 2, so it is kept in \( GS_2 \)). This yields a single monopole pair separated by \( s = a \), with \( s \) being the
FIG. 2. (a) A monopole pair lying on layer 2 (upper layer is kept at GS1) reinforce repulsion between the layers, which becomes strengthen as the string tension/size is enlarged. In addition, note that the energy difference between this configuration and GS1 − GS2, ∆E, gets higher as the layers become closer. (b) On the other hand, having one pair by layer with opposite poles closer, layers attract each other, as realized before with GS1 − GS2 configuration.

size of the string. Eventually, successive flips of neighbor dipoles move one of the monopoles away so that the pair is now separated by a larger (higher energetic) string, say $s = 4a$, as depicted in Fig.2(a). The appearance of these excitations imputes in a repulsion between the layers: indeed, even a single monopole pair (along with the smaller string, $s = a$) is enough to overcome the attraction so that repulsive regime dominates. Actually, whenever the string is enlarged and/or more monopoles take place, the layers repel each other with further strength. This is the fact if monopoles and strings appear in one of the layers while another is kept in its ground state. How about we consider a monopole pair in each layer? As we shall see in what follows, depending on the configuration of the monopoles, the attractive regime can be restored. Let us begin by the simplest case with a monopole pair and a string of size $s = a$, say, in layer 1 and a similar configuration in layer 2. In addition, let the poles of layer 2 be inverted with respect to those of layer 1, as depicted in Fig.2(b). In this case, the attraction between opposite and closest monopoles (separated by $d = h$; north/blue and south/red poles) overcome the whole repulsion brought about by the strings
and like poles interaction, so that the layers experience an attractive regime once again. If like poles are moved away their repulsion decreases considerably and attraction between layers is strengthen. On the other hand, if the configuration were like poles closer each other, repulsion would be huge.

Now, we would like to study the case two isolated monopoles, one placed in each layer and, whether and how the surrounding medium affects their interaction. To isolate a monopole in a layer, we should move its partner far away (what has the cost of enlarging the string until the edge of the layer, effectively expelling the moving pole outside the system). This shed further light onto former results and discussion. Indeed, if we consider two isolated like poles, one lying at a fixed position of each layer, as shown in Fig. 3(a), then the layers experience a strong repulsion and tend to keep far apart. On the other hand, if one has opposite poles, their interactions overcome and the layers experience attraction once again.

For the sake of completeness, now the layers are kept at definite and fixed $h$ values and we intend to study the energetic of the system as one of the monopoles is displaced along. For concreteness, let the monopole at bottom layer fixed at $(x_1, y_1, z_1 = 0)$, while the other is initially at $(x_2 = x_1, y_2 = y_1, z_2 = z_1 + h = h)$, but it may be displaced along $xy$ plane. As expected, also in this framework like poles repel whereas opposite monopoles attract.
FIG. 4. (a) Two like monopoles were initially separated by a vertical distance $d = h$. Then, the pole in the upper layer is displaced along $xy$ plane, as indicated. The effective potential, $V_{\text{eff}} = q_m^2 d^{-1} + \kappa s + c$, against $d$ for a number of height offsets. For short monopole separation, Coulomb term dominates, while as $d$ increases the string size $s$ tends to increase even faster and the linear potential dominates. For both cases, the bottom graphics display the Coulomb potential between two magnetic monopoles, $V_C = q_m^2 d^{-1}$, which is clearly repulsive for like charges and attractive for opposite ones.

themselves following a Coulomb potential, $d^{-1}$ ($d$ is the spatial distance between the poles, see Fig. 4). The energetic resembles that obtained for a single ASI layer, like below:

$$V_{\text{eff}}(d, s) = \frac{q_m^2}{d} + \kappa s + E_c.$$ (2)

where, $q_m$ is the charge of an isolated magnetic pole (which may be positive or negative), $\kappa$ is the string tension of size $s$ which accounts for the energy cost of moving the other two
monopoles far away from the remaining ones; $E_c$ is the energy cost to create these excitations, monopole pair and string as well. Table I presents how such parameters vary with $h$: namely, for $h = 0.5a$ one gets $q_m^2 \approx 4.8Da$ (along with $\kappa \approx 10D/a$ and $E_c \approx 450D$). This monopole charge is comparable to that for a single square lattice ASI$^{123}$, $\sim 3.8Da$, but string tension and creation energy are much higher, evidencing the strong coupling between layers at this height offset. As a whole, our findings clearly show that field lines produced by magnetic monopoles lying in a layer spread radially throughout the $3D$ space following a Coulomb-like law.

| $h(a)$ | $q_m^2$ | $\kappa$ | $E_c$ | $q_m^2$ | $\kappa$ | $E_c$ |
|--------|---------|-----------|-------|---------|-----------|-------|
| 0.50   | 4.8     | 16        | 754   | -6.3    | 16        | 334   |
| 0.75   | 5.3     | 11        | 480   | -5.9    | 11        | 410   |
| 1.0    | 5.1     | 10        | 449   | -5.4    | 10        | 432   |
| 1.5    | 4.5     | 10        | 443   | -4.7    | 10        | 439   |
| 2.0    | 4.1     | 10        | 442   | -4.4    | 10        | 440   |

Finally, we deal with the basic thermodynamics of the BASI system. The specific heat, $c = \frac{\langle E^2 \rangle - \langle E \rangle^2}{Nk_B T^2}$ ($k_B$ is the Boltzmann constant), has been obtained by standard Monte Carlo technique along with Metropolis algorithm implemented using Boltzmann distribution, $\sim e^{-\Delta E/k_B T}$, for our original array consisting by 3480 dipoles per layer. We have also implemented $10^4$ Monte Carlo steps to reach a steady state and up to $10^5$ Monte Carlo steps to obtain the averages of thermodynamic variables, each Monte Carlo step corresponding to 3480 single-spin flips. [In order to save time computation, we adopt a cutoff radius $r_c = 6a$ whenever dealing with the dipolar energy. Such a cutoff yields deviations $\leq 0.1\%$ in the total energy of the system]. Fig. 5 depicts specific heat as a function of temperature for distinct $h$ values. First, note that the specific heat peak is shifted to lower temperature as $h$ increases. Indeed, as the layers become decoupled the peak temperature is $T_p \approx 7D/k_B$. This value is slightly smaller than $T = T_c = 7.2D/k_B$ reported in the works of Refs$^{3336}$ for a square ASI at the thermodynamics limit, whereas here we are taking into account the
finite size of BASI layers.

![Figure 5](image)

**FIG. 5.** Simple BASI thermodynamics: (a) Specific heat, $c$, as a function of temperature, $T$. Increasing $h$ the peak is shifted to lower temperature. (b) The peak temperature, $T_p$, against height offset, showing that BASI effectively behaves as two decoupled layers for $h > a$.

### IV. CONCLUSION AND PROSPECTS

Whenever in the ground state, the layers composing our BASI system attract each other with a force which resembles van der Waals power-law forces. When excitations emerge in the system, layers still attract if single opposite charges are disposed in each layer (Fig. 3b), or a monopole pair in each layer but with opposite charges close each other. Other situations favor repulsion, which by itself could be thought as a kind of magnetic levitation or magnetic damping for nanoscaled systems. The switching between attraction and repulsion may be useful to design stable *BASI-type molecule* by balancing these forces. The dependence of specific heat peak with $h$ may be useful to determine optimal heights offset favoring stability.

As prospects, we intend to investigate how BASI behave under translation and rotation of one layer (while the other is kept fixed). As fundamental symmetries they are expected to yield novel effects and to shed further light into system physical properties.
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