Realizing Three-Dimensional Artificial Spin Ice by Stacking Planar Nano-Arrays

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Artificial spin ice is a frustrated magnetic two-dimensional nano-material, recently employed to study variety of tailor-designed unusual collective behaviours. Recently proposed extensions to three dimensions are based on self-assembly techniques and allow little control over geometry and disorder. We present a viable design for the realization of a three-dimensional artificial spin ice with the same level of precision and control allowed by lithographic nano-fabrication of the popular two-dimensional case. Our geometry is based on layering already available two-dimensional artificial spin ice and leads to an arrangement of ice-rule-frustrated units which is topologically equivalent to that of the tetrahedra in a pyrochlore lattice. Consequently, we show, it exhibits a genuine ice phase and its excitations are, as in natural spin ice materials, magnetic monopoles interacting via Coulomb law.

PACS numbers:

Spin ice materials, such as rare-earth pyrochlores and artificial spin ice, are magnetic systems in which frustrated interactions lead to complex (partial) orderings and unusual collective behaviours [1–3]. Magnetic ions in pyrochlore spin ice form a network of corner-sharing tetrahedra whose classical magnetic macro-spins minimize the local interaction energy by obeying the two-in-two-out ice rule proposed by Pauling for the proton orderings in water ice [4]: hence the name, spin ice. It has recently been demonstrated that elementary excitations over the disordered ice manifold of spin ice materials are emergent magnetic monopoles that fractionalize from local dipole excitations [5, 6].

The artificial counterparts of these natural materials, artificial spin ices [3–7], are nanostructured two-dimensional (2D) arrays of single-domain ferromagnetic bars that behave like giant Ising spins. Collective behaviour of the nanomagnets can be controlled through appropriate choices of material, geometry and array topology. Because of their nano-scale interaction energies (∼10−3 − 105 K depending on the size of the nanomagnet and mutual spacing) they reveal, at accessible temperatures, emergent features which in natural materials are seen only at very low temperature. Following the pioneering work of Wang et al. [7, 8] on the two-dimensional square-ice array [7, 8], artificial spin ices have been proposed and studied in diverse types of physical systems [9–12] and geometries such as honeycomb (kagome ice) [13–20], brickwork [21], triangular [22–24], and pentagonal lattices [25]. A systematic approach for designing 2D arrays with emergent ice-type frustration has also been proposed [20, 27], and recently realized experimentally [29].

Most of the experimental efforts in such artificial frustrated magnets has understandably focused on two-dimensional systems: even with mature nanolithography, it remains a great challenge to integrate a full three-dimensional (3D) structure with oblique angles between nanobars such as the pyrochlore lattice. Recently an interesting realization of a 3D artificial spin ice on a opal-like lattice was realized via self-assembly techniques [50, 51], which unfortunately do not allow for control over the lattice geometry and connectivity.

Conversely, planar nano-structured arrays have offered great flexibility in nano-fabrication, which has been recently exploited to produce arrays of exotic geometries [24] or desired super paramagnetic behavior. Maintaining this kind of manufacturing flexibility while producing an arrangement that captures the 3D spin ice behaviours would be ideal. One way to transport the convenience of 2D nano-lithography to 3D fabrication is by stacking 2D structures, thus building the 3D material layer by layer. Then, the essential issue becomes the theoretical design of such layered structure. Essentially that entails tuning Ising interactions between nearest-neighboring (NN) nanobars to mimic spin ice frustration. Finally, it is necessary to verify theoretically that such design would produce the desired ice manifold.

In this manuscript we propose a nano-fabrication oriented design for a multilayer artificial spin ice structure, topologically equivalent to the pyrochlore lattice. We demonstrate that the degrees of geometrical frustration can be controlled by gauging the interlayer spacing. In particular, there exists a critical spacing such that the nearest-neighbor spin-spin interactions is fully frustrated as in the case of pyrochlore spin ice. Through Monte-Carlo simulations we show that it exhibits an ice manifold, as well as lower entropy antiferromagnetic phases. We further show that the effective interaction between monopolar excitations in our 3D design follows a Coulomb law in the ice phase, as in natural dipolar spin ice on the pyrochlore lattice.

Figure 1 shows the proposed multilayer structure. In each layer, parallel ferromagnetic bars are arranged in a rectangular lattice with the long and short lattice constants being 2a and a, respectively; the orientation of the nano islands is aligned with the short axis. The arrays
are rotated by $90^\circ$ from one layer to the next. In addition, the arrays in every other layer are shifted by $a$ along the long axis. The two-dimensional projection of the 3D structure on the $xy$ plane resembles the nano-magnetic arrays in a square ice.

The basic frustration unit shown in Fig. 2(a) consists of two pairs of nano-bars from consecutive layers. The examples of Ising-spin representation of the magnetic state in the unit are shown in Fig. 2(b). These units are analogous to the vertices and tetrahedra in square and pyrochlore ices, respectively. Even though our design is a layered structure, the centers of these frustration units form a 3D lattice that is topologically equivalent to a diamond lattice. Moreover, as shown in Fig. 1 each magnetic bar is shared by two frustration units, exactly as each spin is shared by two tetrahedra in pyrochlore. These observations indicate that the nanomagnets themselves form a lattice that is isomorphic to pyrochlore, and each frustration unit corresponds to a tetrahedron. For convenience, we shall refer to the frustration unit in the multilayer spin ice as tetrahedron. Fig. 2(c) shows a configuration of random magnets satisfying the ice rules; the corresponding spin structure on the pyrochlore lattice is displayed in Fig. 2(d).

Next we consider the energetics of the artificial spin ice. The isomorphism between our tetrahedra and those of a pyrochlore realization is clearly not enough: it is essential that the ice-rule is obeyed at the tetrahedron level. To this end, we first classify the tetrahedra into four types similar to vertices in a square ice [2]: type-I and II refer to 2-in-2-out units defined above, while type-III and IV denote the 3-in-1-out/1-in-3-out and all-in/all-out tetrahedra, respectively; see Fig. 2(b). Assuming a single-domain magnetization for each nanobar [7], the nanomagnets interact with each other through the dipolar interactions:

$$H = \frac{\mu_0}{8\pi} \sum_{i,j} \frac{m_i \cdot m_j - 3(m_i \cdot \hat{r}_{ij})(m_j \cdot \hat{r}_{ij})}{|\mathbf{r}_{ij}|^3},$$  \hspace{1cm} (1)$$

where $m_i = \pm \mu_0 \hat{e}_i$ is the dipole moment of $i$-th nanobar, and $\hat{e}_i$ is a unit vector parallel to the direction of the bar.

The dipolar energy of the four nanomagnets in a unit reaches a minimum when they satisfy the “ice rules”, namely two spins point toward the center and two point outward. However, degeneracy between the two types of 2-in-2-out tetrahedra, I and II in Fig. 2(b), is in general lifted: unlike the equivalent pair-wise spin interactions in a tetrahedron, the dipolar interaction energy between parallel bars (in the same layer) generally is different from that between orthogonal bars (in different layers), a situation similar to the case of square ice [7].

One can, however, restore the degeneracy between the types-I and II tetrahedra by properly adjusting the height $h$ of each layer, as pointed out in Ref. [32, 33] for a two dimensional case. For example, in the point-dipole approximation for the nanobars (with length $l \ll a$), equal dipolar energies for the six 2-in-2-out configurations can be reached by setting $h = h^* = a \sqrt{(3/8)^{1/6} - 1/2} \approx 0.41890 \, a$. In practical realizations the interaction is not exactly dipolar and the required height $h$ can be obtained with the aid of micromagnetic simulations, accounting for the finite extension of the nanobars (see supplementary materials for two cases). Away from $h^*$, the degeneracy is lifted and the lowest energy configurations are type-I (II) for $h < h^*$ ($h > h^*$).

We have thus established the equivalence of the multilayer artificial spin ice and the pyrochlore ice structurally and energetically, at least at the level of the nearest-
neighbor interactions. We therefore expect our design to exhibit a similar ice manifold. Next we consider the thermodynamic transformations as a function of temperature for the 3D artificial spin ice. Indeed, thanks to recent advances in growth 34 and thermal annealing technologies 15, 28, it is now possible to systematically prepare artificial spin ice in thermal ensembles and even probe its low entropy thermal states 15, 28, 33, 67. Our numerical studies focus on multilayer structures with a height \( h \) in the vicinity of \( h^* \), estimated above, and obtain a phase diagram in the \( h - T \) plane. For simplicity, we have used the point-dipole approximation for the magnetic nanobars. The long-range dipolar interactions are implemented using standard Ewald method, and periodic boundary conditions were used in all simulations.

The simulation results for the case with \( h = h^* \) and \( h = 1.013h^* \) are shown in Figs. 3. In both cases, the specific heat shows a broad peak at \( T \approx D_{nn} \), where \( D_{nn} = \mu_0 \mu^2 / 2 \pi a^3 \) is the dipolar interaction energy between nearest neighbors, signaling the crossover into the two-in-two-out ice phase. Indeed, as shown in Fig. 3(b) and (d), population of monopoles (type-III tetrahedra) rapidly tends to zero below the crossover temperature (type-IV are always zero), where most of the tetrahedra are in the 2-in-2-out states (type-I and II). As the temperature further decreases, the system undergoes a discontinuous transition at \( T_N \approx 0.27D_{nn} \) and \( 0.22D_{nn} \), respectively for the two cases, revealed by sharp peaks in the specific heat. For the case with \( h = h^* \), almost all tetrahedra in the ordered phase below \( T_N \) are in the type-I state, similar to the case of square ice 37, 39. On the other hand, the ordered state for \( h = 1.013h^* \) is predominantly composed of type-II vertices, indicating that a different ground state is selected for larger \( h \).

The apparent first-order magnetic transition at \( T_N \) is induced by the dipolar interactions beyond the nearest-neighbor pairs, a situation completely analogous to the low-temperature ordering of spins in the pyrochlore dipolar spin ice 39. In fact, at \( h = h^* \), the extensive degeneracy of the ice phase is already lifted by the second-nearest-neighbor interactions which lacks the pseudo-cubic symmetry of the NN pairs. When \( h \neq h^* \), even the degeneracy at the NN level is lifted, as explained above. In order to obtain the 3D long-range spin order selected by the dipolar interactions, we employed the loop algorithm 39 in combination with single-spin flips in our simulations to avoid the dynamical freezing of spins in the ice regime. The phase diagram in the \( h - T \) plane obtained from our extensive Monte Carlo simulations is shown in Fig. 4. We find two distinct long-range antiferromagnetic orderings at \( T < T_N \). These two ordered phases are separated by a first-order line at \( h_c \approx 0.4194a \), in agreement (only \( 10^{-4} \) larger) with the value \( h^* \) at which the NN interactions are equivalent.

The ground state at \( h < h_c \) is characterized by a \( Q = 0 \) spin structure [AFM-1 in Fig. 4(a)], implying a uniform ordering of type-I tetrahedra. This state is the 3D analog of the staggered arrangements of type-I vertices observed in the ground state of two-dimensional square ice 15, 34. In our 3D multilayer case, tetrahedra of different orientations are in the two different type-I configurations, respectively.

A different ground state was obtained when \( h > h_c \). The second-neighbor interactions in this case favor antiparallel alignment of magnetizations between nanobars of same orientation but in different layers. The resultant 3D spin order, AFM-2 in Fig. 4(a), consists of ferromagnetic (FM) layers of tetrahedra stacked antiferromagnet-
Interestingly, the tension $\sigma$ of a straight Dirac string connecting a pair of monopoles in the two ordered states as a function of $h-h^*$. The two monopoles are separated by a displacement $r = n(a/\sqrt{2},0,h)$ and $r = n(0,a/\sqrt{2},h)$, where $n$ is an integer, in the AFM-1 and 2 states, respectively. The distance $r$ is measured in units of $a$. (b) The effective potential $V_{\text{eff}}(r)$ between a pair of monopoles (3-in-1-out and 1-in-3-out defects) separated by $r = n(a/\sqrt{2},0,h)$. The dashed line corresponds to $V_0/r$, with $V_0 \approx 1.3D_{\text{eff}}$.

![Diagram](image_url)

The ice phase can be described by a plasma of weakly interacting magnetic monopoles.

In summary, we have proposed a 3D layered geometry for a 3D artificial spin ice that captures the fully 3D spin ice behaviour such as an effective Coulomb interactions between monopoles and also provides an accessible and flexible, experimentally realizable geometry. The structure is obtained by layering two dimensional lattices of nano-structures. The interest of this layered design lies in its accessibility via nano-fabrication of successive layers, and promises a viable way to extend artificial spin ice to the third dimension.

Acknowledgment. We are grateful to J. Hollingsworth and S. Ivanov (Center for Integrated Nanotechnologies, LANL and Sandia) for useful discussions on nano-fabrication techniques, and Cynthia Olson Reichhardt for a critical reading of the manuscript. This work was carried out under the auspices of the National Nuclear Security Administration of the U.S. Department of Energy at Los Alamos National Laboratory under Contract No. DE-AC52-06NA25396.

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