FRUSTRATION by design

Ian Gilbert, Cristiano Nisoli, and Peter Schiffer

By fabricating magnetic structures into nanoscale arrays, physicists can directly visualize how condensed-matter systems accommodate competing interactions among dipole moments and other degrees of freedom.

Ian Gilbert is a postdoctoral researcher at NIST's Center for Nanoscale Science and Technology in Gaithersburg, Maryland. **Cristiano Nisoli** is a staff scientist at Los Alamos National Laboratory in Los Alamos, New Mexico. **Peter Schiffer** is a professor of physics and the vice chancellor for research at the University of Illinois at Urbana-Champaign.

Geometrical frustration is a condition that occurs when a material's lattice geometry precludes minimizing the energy of all the interactions among pairs of neighbors simultaneously. The simplest example is three antiferromagnetically coupled Ising spins, pointing up or down, on the corners of an equilateral triangle: It is impossible to arrange the spins so that each pair is antiparallel. In more complex magnetic lattices, the frustrated state can arise from the combination of lattice geometry and the strength and sign of the interactions among the magnetic dipole moments.[1] (See the article by Roderich Moessner and Art Ramirez, PHYSICS TODAY, February 2006, page 24.) A wide variety of exotic and collective phenomena sometimes arises from the competing interactions. A prime example is spin liquids, materials in which the local atomic moments fluctuate down to the lowest accessible temperatures and never settle into a static ground-state configuration.

Among the most intriguing of such geometrically frustrated magnets are the "spin ice" materials. Their three-dimensional atomic lattices situate rare-earth ions with large magnetic moments on the corners of tetrahedra that themselves are arranged in a corner-sharing lattice. The energies of the frustrated interactions between the rare-earth moments are collectively minimized by having two moments point into and two point out of each tetrahedron. Six configurations for each tetrahedron obey that two-in, two-out "ice rule," and spin ice consequently has a macroscopically degenerate low-temperature state. Although the ice rule is obeyed locally at each tetrahedron, no long-range order has ever been observed; the effective long-range disorder among spin states yields an effective finite entropy in the low-temperature limit.

Common frozen water—in which the location of oxygen atoms is periodic but the location of hydrogen ions is not—exhibits the same finite entropy. The ice-rule arrangement of rare-earth moments in spin ice closely mimics the arrangement of the hydrogen ion positions—hence the name spin ice. Those materials exhibit a rich phase diagram that arises from relatively simple interactions between the moments.[2]

Spin ice's local elementary excitations, which occur where the two-in, two-out ice rule is broken, have generated considerable interest in recent years because they can be parameterized as emergent magnetic monopoles.[3] Unlike the theoretical objects Paul Dirac postulated in 1931, the monopole-like excitations arise from the collective behavior of interacting atomic spins in the crystal lattice and correspond to nonzero net numbers of spins that point into or out of the tetrahedra. Created in pairs, individual excitations show indications of moving independently within the confines of the crystal (see PHYSICS TODAY, March 2008, page 16).

In the past decade, a parallel approach to studying frustration has become increasingly common, thanks to modern nanofabrication and microscopy techniques.[4,5] Rather than turning to spin-ice compounds found in nature, which reveal their interesting physics only at cryogenic temperatures and atomic length scales, researchers make their own by patterning a ferromagnetic film into an array of nanometer-scale islands; the arrangements are known generically as artificial spin ice. The island material and dimensions are chosen such that each island constitutes a single magnetic domain whose moment magnitude is typically a million Bohr magnetons or more. (One μ_B is the intrinsic spin magnetic moment of an electron.)

Because such nanoscale ferromagnets are produced lithographically, they can be arranged into essentially any 2D array pattern. Importantly, if neighboring magnetic islands are placed in close proximity, their moments interact with a strength that depends on their separation and relative orientation. Using imaging techniques such as magnetic force microscopy and photoemission electron microscopy, scientists can study the individual moment orientations and collective behavior of the extended system. An artificial spin ice thus constitutes a lattice whose frustrated interactions are both controllable and measurable at the level of a single magnetic moment.

Islands and vertices

Most initial studies of artificial spin ice focused on two simple lattices: the square and the honeycomb, or kagome, both illustrated

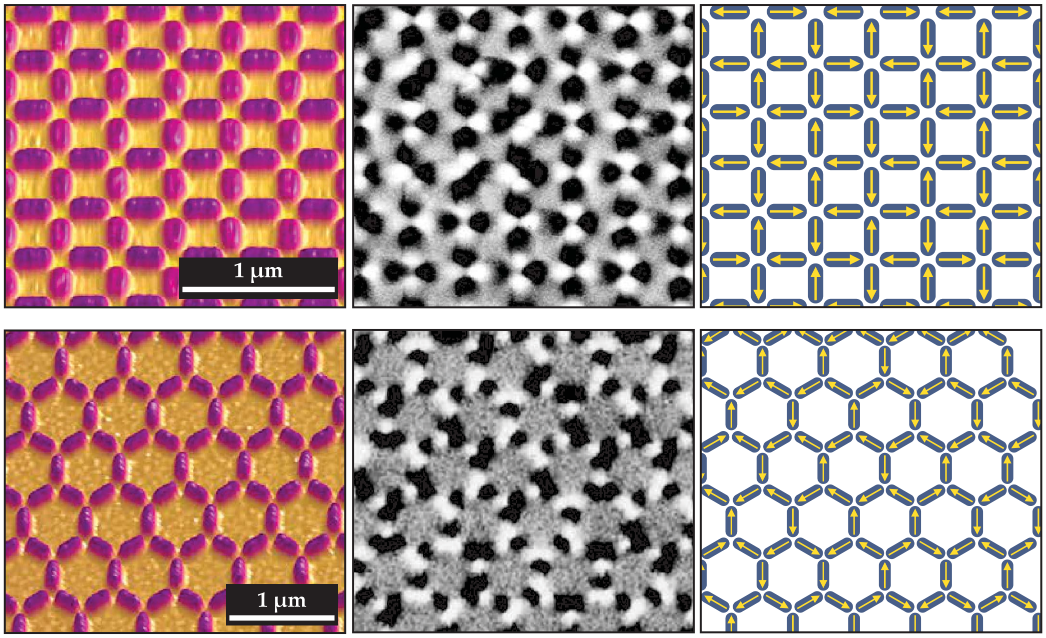


FIGURE 1. ARTIFICIAL SPIN ICE in its most common two-dimensional geometries: **(top)** the square lattice and **(bottom)** the kagome lattice. The islands making up the lattices shown here are fabricated from nickel–iron alloy with lateral dimensions of 80 nm × 220 nm and a thickness of 25 nm. The topography (left) and magnetic field contrast (middle) are imaged using atomic force microscopy and magnetic force microscopy, respectively; black and white indicate the magnetic north and south poles of each island. At right are maps depicting the dipole moments that correspond to the magnetic force microscopy images. (Adapted from ref. 4 and I. Gilbert, "Ground states in artificial spin ice," PhD thesis, University of Illinois at Urbana-Champaign, 2015.)

in figure 1. In each case, the individual structures are elongated islands, in which the shape anisotropy aligns the magnetic moment of each island with its long axis.

In 2006 two of us (Schiffer and Nisoli) and our colleagues created the first instance of an artificial square spin ice.[4] The ferromagnetic islands in those structures, fabricated from permalloy (a common alloy of nickel and iron), had a moment of about $10^7\ \mu_B$. The energy required to reverse a single island's magnetization was substantial—equivalent to about 10^6 K—and made the system highly stable at room temperature. To appreciate the material's local physics, consider its component vertices, where four neighboring islands meet in the shape of a plus symbol. As outlined in the box on page 57, the possible moment arrangements of a vertex in the square lattice fall into four types. In the lowest-energy configurations (types I and II), two moments point in toward the vertex and two moments point out, analogous to the ice rule that governs the local ground states of tetrahedra in natural spin-ice materials.

To circumvent the thermal stability of the moment configurations in the fabricated lattice, we rotated the lattice in an oscillating magnetic field. The goal of that demagnetization process was to make the lattice sample many possible moment configurations to find one that minimized its magnetostatic energy and removed any net magnetization. After the procedure, magnetic force microscopy revealed that many more vertices obeyed the ice rule than would be expected for a random arrangement of island moments; the result demonstrates that Ising spins on an appropriate frustrated lattice suffice to reproduce the ice rule found in natural spin ice.[4]

Because the interactions between pairs of islands at a square ice vertex are not all identical—perpendicular islands interact more strongly than parallel ones—type I ice-rule vertices are lower in energy than type II vertices; the difference lifts the degeneracy of the ice-rule configurations. Indeed, the square lattice possesses an ordered, nondegenerate ground state of type I vertices arranged in a checkerboard pattern. As in rare-earth oxides, each vertex in artificial spin ice can be associated with a net magnetic charge whose magnitude corresponds to the balance of the moments oriented into and out of the vertex. Everywhere in the ground state of the square lattice, the charge is zero.

Unlike in artificial square spin ice, the relative orientation and center-to-center distance between islands in the kagome lattice are the same for all pairs of islands that meet at a vertex, which means that all pair-wise interaction energies are the same. That higher degree of symmetry in the kagome geometry supports numerous thermodynamic phases that arise from the frustration of the interactions between neighboring moments.[6]

John Cumings and his group at the University of Maryland showed that it's easy to obtain the simplest of those phases, a "pseudo-ice manifold," in the kagome lattice by manipulating the moments with alternating magnetic fields, the same demagnetization technique used on the square lattice. Although the island moments do not exhibit long-range order in that simplest phase, they obey a local quasi-ice-rule constraint: One island moment points into and the other two moments point out of each vertex, or vice versa.[7] Furthermore, because each vertex has an odd number of islands, a nonzero magnetic charge appears at every one. As groups led by William Branford from Imperial College London and by Laura Heyderman at the Paul Scherrer Institute in Switzerland have shown, applying an external magnetic field can manipulate the magnetic charges and produce a flow of magnetic-charge current—demonstrations that expand the analogy to monopole behavior.[8]

Thermalization

The energy required to reverse the sign of large, stable moments is, as mentioned earlier, orders of magnitude higher than ambient thermal energies. Although subjecting the sample to a time-varying magnetic field consistently produces statistical ensembles that have a controllable excess of lower-energy vertices, the procedure cannot be said to produce a truly thermalized state and does not allow a square lattice to access its ground state of ordered type I vertices.

In 2011 Chris Marrows and his colleagues at the University of Leeds reported measurements of artificial square spin ice whose "as-grown" state exhibited large domains of type I vertices—patches of arrays whose moment configurations had settled into their collective ground state.[9] Moreover, the team observed small clusters of reversed island moments—the equivalent of excited states or defects in a condensed-matter system—on the background of an ordered ground state. Those clusters followed a Boltzmann distribution, indicative of true thermal equilibrium.

The thermalization in that study took place during the deposition process. When the island thickness was only a few nanometers, the energy barrier to moment reversal was relatively small, and thermal fluctuations allowed the lattice to sample its configuration space well enough that sections found their ground state. As the islands grew thicker, the energy barriers grew larger and froze in place the patches of type I ground-state vertices. The results offered proof that an artificial spin-ice array could be prepared in a thermal ensemble—an essential prerequisite for comparing measurements of the island moment orientations with thermodynamic models of spin systems.

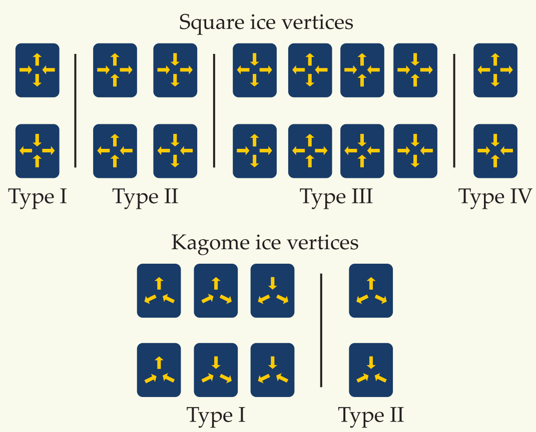
Inspired by that work, other groups developed techniques for making thermalized systems. One method heats an already fully formed artificial spin ice to a high temperature near the Curie point of the islands' ferromagnetic material, above which the material loses its ferromagnetism.[10] At that elevated temperature, thermal fluctuations can rearrange the moment configurations. Upon slow cooling, the interactions between moments determine the collective magnetic state into which the system settles. Once cooled to room temperature, the island moments are again highly stable and immune to perturbations by the imaging tip of a magnetic force microscope.

Björgvin Hjörvarsson of Uppsala University and Heyderman pioneered alternate approaches to thermalization:[11] They produced islands that are thermally active near room temperature, either by controlling the thickness of the permalloy film or by using different materials. To avoid altering the moment configuration by a magnetic force microscope's magnetized tip, the groups turned to photoemission electron microscopy to monitor the samples' microstates. Imaging a lattice using that technique takes only seconds, so the dynamics of moment configurations can be tracked in real time as the artificial spin ice relaxes from an excited state to its ground state. Figure 2 shows a sequence of images of a square lattice during that evolution.

Dedicated geometries

Armed with those methods, researchers can probe geometries deliberately designed to reveal emergent behavior that might only be accessible in a thermalized system. Virtually any imaginable lattice geometry, including the quasicrystalline example based on Penrose tilings[12] that is shown in figure 3, can be fabricated and characterized both locally and in the ensemble limit

VERTICES, ENERGIES, AND MAGNETIC CHARGES



Artificial spin ice is often analyzed in terms of vertices rather than the dipole moments of the individual magnetic islands that compose the lattice. A vertex is a site at which multiple islands converge. In the square lattice, for instance, each vertex consists of four islands arranged in the shape of a plus sign. The vertices are classified according to their energy, which is the sum of all the magnetostatic interactions of the constituent islands and depends on the relative orientations of the island moments. Square ice has 16 possible moment configurations that can be divided into four unique types of vertices, with type I having the lowest energy and type IV the highest. Types I and II both obey the so-called ice rule, in which two moments point inward and two point outward.

In the kagome lattice only two types of vertices exist: Type I vertices obey the pseudo-ice rule (two-in, one-out, or vice versa); the three island moments of the more costly type II vertices point either all in or all out.

At first glance, vertices might seem to be just a convenient way to visualize moment configurations. But models based on assigning energies to different vertex configurations in a lattice of spins are important in statistical mechanics. Many of them are exactly solvable and allow for detailed quantitative analysis of diverse phenomena, including ferromagnetic and antiferromagnetic phase transitions, the residual entropy of frozen water, and crystal growth on surfaces.

Additionally, vertices with an imbalance of inward- and outward-pointing moments possess effective magnetic charges. If you imagine island moments not as point dipoles but as spatially separated north and south magnetic charges, then each vertex may be assigned one charge per moment. For ice-rule vertices with an even number of moments, the magnetic charges of a vertex cancel out in the ground state, but square-lattice type III and type IV vertices possess a net magnetic charge, analogous to a magnetic monopole. In the case of the kagome lattice, because the vertices reside at the center of three moments, they always possess a net effective magnetic charge. The figure on page 54 illustrates one possible charge distribution.

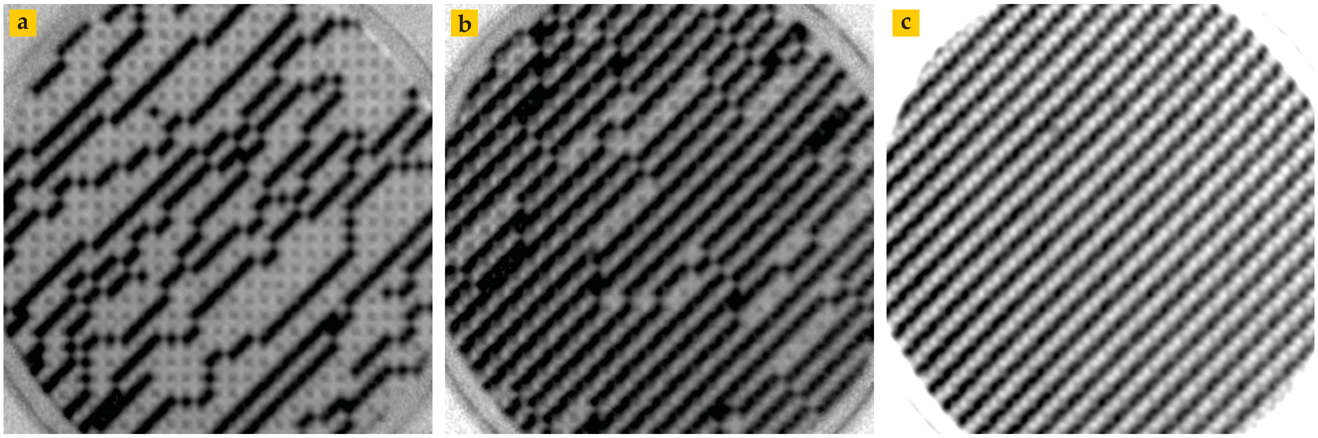


FIGURE 2. PHOTOEMISSION ELECTRON MICROSCOPY images show the dynamical evolution of an artificial square spin ice. The dipole moments of the islands are initially polarized with a magnetic field such that they show white contrast. With the external field removed, thermal fluctuations allowed the moments to reverse sign (to black) during the next several hours as the moment configuration of the sample relaxed to the ground state. In these 20-μm-wide fields of view, **(a)** isolated strings of islands with reversed moments appeared first, and they gradually grew into **(b)** domains of uniformly ordered vertices until **(c)** the entire lattice eventually became perfectly ordered, alternating black and white strips. (Adapted from ref. 11, A. Farhan et al.)

in real space and real time. To date, the most extensively studied novel geometries are those in which the frustration occurs at the level of the vertices rather than at the level of individual island moments. In those vertex-frustrated systems, the lattice geometry forces a subset of vertices into higher-energy states than they would naturally adopt in isolation.[13]

A recent experimental example of vertex frustration is the so-called shakti lattice. Although the geometry does not directly correspond to any known natural magnetic material, it retains the fourfold symmetry of the square lattice and contains vertices that comprise two, three, or four converging island moments. Two years ago the three of us and our colleagues designed, fabricated, and imaged the shakti lattice depicted in figure 4a. Each island grouping, or plaquette, was forced by the lattice geometry to contain two ground-state three-island vertices (open dots) and two excited three-island vertices (filled dots). The four vertices can be arranged in six ways on the four sites of the plaquette. Consequently, the degeneracy of the vertex arrangements on the plaquette is exactly analogous to the degeneracy of spin states on a tetrahedron in natural spin-ice materials. All six configurations are degenerate in energy as well, which means that unlike the collective arrangement of vertices on the square lattice, their arrangement on the shakti lattice faithfully reproduces the collective, disordered ice state of real materials.

When probed experimentally, the shakti lattice exhibits surprising ordering of the magnetic charges of its three-island vertices: The charges alternate sign to form a checkerboard pattern. Furthermore, opposite-signed charges surround, and thus effectively screen, the monopole excitations occasionally found on the four-island vertices in the shakti lattice.

Various geometries, such as brickwork, pinwheel, and pentagonal configurations, can be designed to study chirality, reduced dimensionality, magnetic-charge dynamics, and other interesting effects. Structural defects also produce significant effects. Dislocations introduced into a square lattice, for instance, further frustrate it and produce topological defects even in the otherwise ordered ground state.[14] The dislocations create strings of excited vertices—and a concomitant change in magnetic texture—that will extend either to the edge of the lattice or to another structural defect.

From magnets to superconductors

Microscopic arrays of magnets are not the only intentionally structured materials that follow ice rules and exhibit disordered states. So can closely packed nonmagnetic spheres, for instance. When confined in a quasi-2D plane, the spheres form

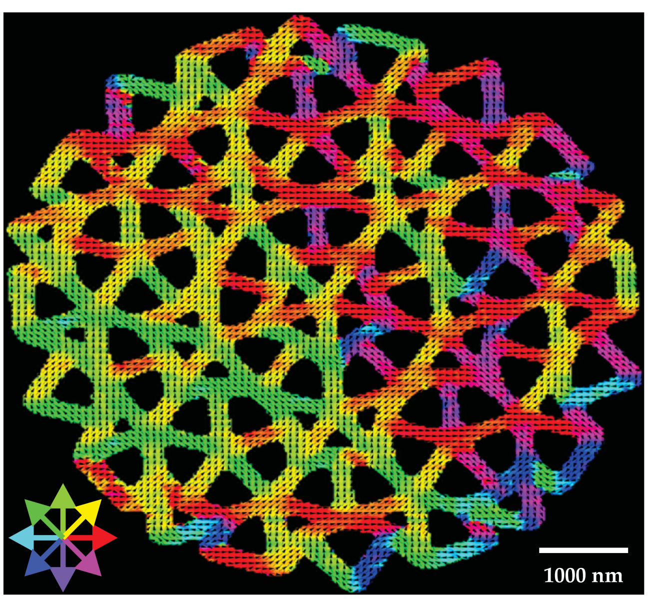


FIGURE 3. QUASICRYSTALLINE SPIN ICE, imaged by a scanning electron microscope. The polarization of the secondary electrons, given by the color scale in the lower left corner, indicates the direction of magnetization and shows the complex magnetic textures a ferromagnetic lattice can adopt in such a geometry. (Adapted from ref. 12.)

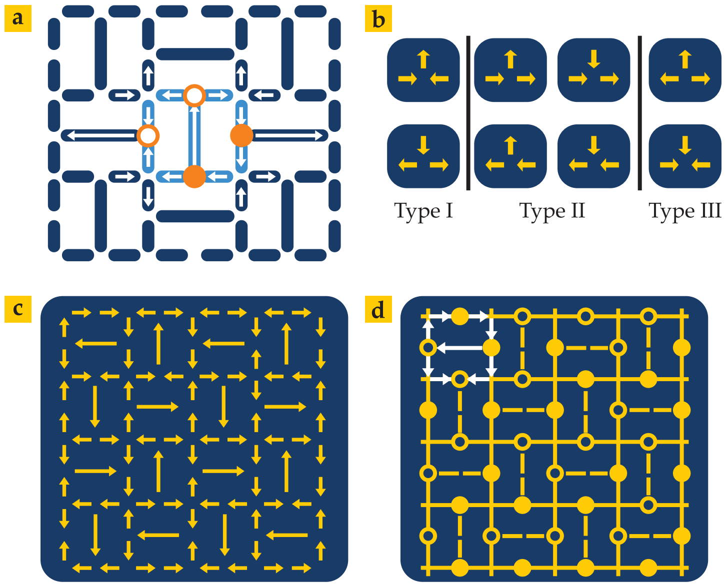


FIGURE 4. THE SHAKTI LATTICE is an artificial spin ice whose geometry combines features of the square lattice, with four moments per vertex, and the kagome lattice, with three per vertex. **(a)** A single island grouping, or plaquette, is highlighted in light blue. In the collective magnetic ground state, the four-island vertices must be in their lowest-energy state. Consequently, the three-island vertices cannot all be configured in their local ground state. The lowest-energy solution to that frustration is to place two (open dots) of the three-island vertices in each plaquette in their ground state and two (filled dots) in their first excited state. **(b)** The ground state of three-island vertices, labeled type I, and the excited states, labeled types II and III (see the box on page 57). **(c)** A representation of the experimentally measured moments extracted from a magnetic force microscope image. **(d)** The same map, redrawn to emphasize the three-island vertex states; open dots represent type I states and filled dots type II vertices. Two vertices of each type reside in each plaquette, an arrangement that matches the collective ice state of real materials. (Adapted from ref. 13, I. Gilbert et al.)

a triangular lattice. But if they are allowed to buckle out of the plane to minimize their volume and thus free energy, the spheres reproduce the frustration of a simple triangular-lattice antiferromagnet.[15]

Similar frustration shows up in other microscopic structures, including hexagonal arrays of Josephson junctions and superconducting rings. Artificial spin-ice structures can be built out of either nonmagnetic colloids or superconducting vortices.[16,17] In both cases, the colloids or vortices can sit at either end of an edge of the lattice, and the lowest-energy configuration corresponds to an ice rule being obeyed at the vertices.

Charge transport and beyond

Artificial spin ice and other engineered frustrated systems are currently reaching a level of complexity and sophistication that goes well beyond the statistical mechanics of simple permalloy square and hexagonal lattices. Researchers are creating systems out of different structures and materials, studying them in different temperature regimes, and even probing them with microwaves that can induce magnetic excitations.

Studies of how electric charges flow through frustrated lattices are likely to be particularly interesting. For example, Branford's group and others have made anisotropic magnetoresistance and planar Hall effect measurements of artificial spin-ice structures, whose short ferromagnetic wires join at the vertices to form a connected network rather than isolated nanoscale ferromagnetic islands.[18] The dynamic behavior of the connected structures turns out to significantly differ from that observed in arrays of nontouching islands. The magnetic moments flip by the nucleation and motion of domain walls rather than by the reversal of individual ferromagnetic islands.

Artificial spin ice can, in principle, also be grown on top of other interesting materials, such as 2D electron gases, superconductors, and topological insulators, to impose complex patterns of magnetic field on the underlying substrate. Scientists are just beginning to explore what phases, patterns, and dynamics emerge. Any connections found with correlated electron physics should soon add an exciting new chapter to the evolving story of frustration by design.

We are grateful for assistance and feedback from our many collaborators and colleagues and for financial support from the US Department of Energy, Office of Basic Energy Sciences, Materials Science and Engineering Division. Alex David Jerez Roman created the title-page image, and Katelyn Gamble prepared other images.

REFERENCES

1. H. T. Diep, ed., *Frustrated Spin Systems*, 2nd ed., World Scientific (2013); C. Lacroix, P. Mendels, F. Mila, eds., *Introduction to Frustrated Magnetism: Materials, Experiments, Theory*, Springer (2011).
2. M. J. P. Gingras, in ref. 1, *Introduction to Frustrated Magnetism*, p. 293; C. Castelnovo, R. Moessner, S. L. Sondhi, *Annu. Rev. Condens. Matter Phys.* **3**, 35 (2012).
3. C. Castelnovo, R. Moessner, S. L. Sondhi, *Nature* **451**, 42 (2008); I. A. Ryzhkin, *J. Exp. Theor. Phys.* **101**, 481 (2005).
4. R. F. Wang et al., *Nature* **439**, 303 (2006).
5. L. J. Heyderman, R. L. Stamps, *J. Phys.: Condens. Matter* **25**, 363201 (2013); C. Nisoli, R. Moessner, P. Schiffer, *Rev. Mod. Phys.* **85**, 1473 (2013).
6. G. Möller, R. Moessner, *Phys. Rev. B.* **80**, 140409(R) (2009); G.-W. Chern, P. Mellado, O. Tchernyshyov, *Phys. Rev. Lett.* **106**, 207202 (2011).
7. Y. Qi, T. Brintlinger, J. Cumings, *Phys. Rev. B* **77**, 094418 (2008).
8. S. Ladak et al., *Nat. Phys.* **6**, 359 (2010); E. Mengotti et al., *Nat. Phys.* **7**, 68 (2011).
9. J. P. Morgan et al., *Nat. Phys.* **7**, 75 (2011).
10. J. M. Porro et al., *New J. Phys.* **15**, 055012 (2013); S. Zhang et al., *Nature* **500**, 553 (2013).
11. U. B. Arnalds et al., *Appl. Phys. Lett.* **101**, 112404 (2012); A. Farhan et al., *Phys. Rev. Lett.* **111**, 057204 (2013); V. Kapaklis et al., *Nat. Nanotechnol.* **9**, 514 (2014).
12. B. Farmer et al., *Phys. Rev. B* **93**, 134428 (2016).
13. M. J. Morrison, T. R. Nelson, C. Nisoli, *New J. Phys.* **15**, 045009 (2013); I. Gilbert et al., *Nat. Phys.* **10**, 670 (2014).
14. J. Drisko, T. Marsh, J. Cumings, http://arxiv.org/abs/1512.01522.
15. M. Skoge et al., *Phys. Rev. E* **74**, 041127 (2006); Y. Han et al., *Nature* **456**, 898 (2008).
16. A. Libál, C. Reichhardt, C. J. Olson Reichhardt, *Phys. Rev. Lett.* **97**, 228302 (2006); A. Libál, C. J. Olson Reichhardt, C. Reichhardt, *Phys. Rev. Lett.* **102**, 237004 (2009).
17. M. L. Latimer et al., *Phys. Rev. Lett.* **111**, 067001 (2013); J. Trastoy et al., *Nat. Nanotechnol.* **9**, 710 (2014).
18. W. R. Branford et al., *Science* **335**, 1597 (2012).

PT