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Editorial

Focus on artificial frustrated systems

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Abstract

Frustration in physics is the inability of a system to simultaneously satisfy all the competing pairwise interactions within it. The past decade has seen an explosion of activity involving engineering frustration in artificial systems built using nanotechnology. The most common are the artificial spin ices that comprise arrays of nanomagnets with competing magnetostatic interactions. As well as being physical embodiments of idealized statistical mechanical models in which properties can be tuned by design, artificial spin ices can be studied using magnetic microscopy, allowing all the details of the microstates of these systems to be interrogated, both in equilibrium and when perturbed away from it. This 'focus on' collection brings together reports on the latest results from leading groups around the globe in this fascinating and fast-moving field.

Keywords: nanomagnetism, frustration, artificial spin ice

1. Introduction

We are surrounded by systems that should exhibit residual entropy at low temperatures, violating the third law of thermodynamics. As Linus Pauling noted in 1935 [1], the possible configurations of protons in crystalline H_2O provide a clear example of how geometrical frustration can lead to vast numbers of energetically similar states, resulting in a non-zero entropy for temperatures arbitrarily close to absolute zero. Frustration occurs in a wide variety of condensed matter systems, and arises when it is not possible to simultaneously satisfy all of the competing pairwise interactions present. This leads to a rich phenomenology, where huge numbers of possible degenerate microstates play important roles in all kinds of complex systems in the physical sciences and beyond [2]. Examples include liquid crystals, magnetic domain patterns, stripe structures in high-temperature superconductors, protein-folding and neural networks.

Conventionally, the study of physical systems is restricted to the investigation of the limited set of naturally occurring materials. The family of rare earth pyrochlore materials closely resemble water ice in their crystal geometry, and equivalent geometrical frustration effects are found in the interactions between the large spins on the rare earth sites: hence they are dubbed ‘spin ices’ [3]. One can mimic their behaviour using nanotechnology, which allows the construction of model systems where the nature of the elements and their interactions can be varied at will to create artificial frustrated systems [4]. Nanomagnetic analogues of spin ice, termed ‘artificial spin ice’ (ASI) [5, 6], have been widely studied in recent years as they provide convenient models for frustration phenomena. A most intriguing aspect is the ability to experimentally observe the details of microstate configurations by defining model length scales so as to be accessible via microscopy and other techniques for probing configurations (examples are shown in figure 1).

In this ‘focus on’ collection, an overview of experimental and theoretical explorations of artificial frustrated systems is profiled. These studies have led to new insights into ordering and other dynamical processes in frustrated and disordered systems. While not exhaustive, this collection showcases the efforts of many authors whose work has helped shape this area, and we present here some of the latest developments in this exciting field.

2. Control of magnetic configurations via topology and geometry

Artificial spin ice consists of elongated nanomagnets positioned either on the sites of square lattice or the kagome lattice, so forming a honeycomb (hexagonal) array. The artificial square ice has four interacting single domain nanomagnets meeting at a vertex [5], whose moments represent the four spins on the corners of a tetrahedron in the bulk spin ice. However, in contrast to its crystal counterpart, the interactions between the four neighboring nanomagnets at a vertex in artificial square ice are not equivalent due to the different distances and angles between them. This lack of equivalence leads to a unique ground state, which can be readily achieved by thermal means [7, 10–12]. In contrast, the artificial kagome (spin) ice, where the magnetic moments now correspond to the spins on the kagome planes of the spin ice crystal, possesses an extensive degeneracy due to the fact that the interactions between the nanomagnets at a three-island vertex are equivalent. In this collection, Morrison *et al* [13] suggest an alternative route to achieve a highly degenerate artificial spin system that is not a result of the degeneracy of the

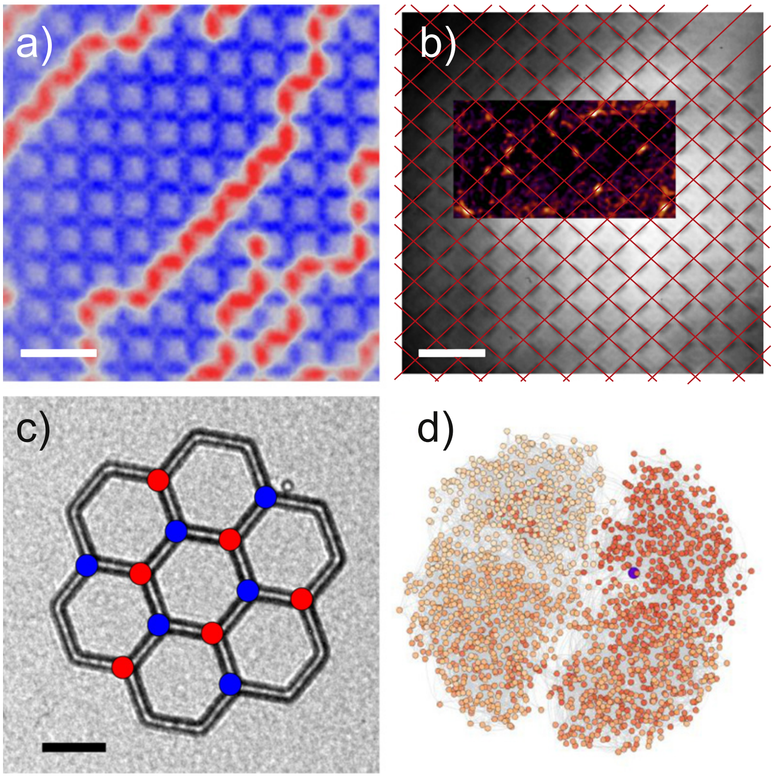

Figure 1. Artificial spin ices. (a) XMCD-PEEM image of artificial spin ice, captured in the so-called 'string regime' [7], while undergoing thermal relaxation from an energetically excited, saturated moment, configuration down to one of the two degenerate ground states. Nanomagnets with moments pointing towards the bottom/left appear in blue contrast, while nanomagnets with moments pointing up/right appear in red contrast. Scale bar 2 μm. (b) X-ray transmission micrograph of a CoFeB artificial square ice with the overlaid red gridlines showing the square lattice. Magnetic contrast is shown in the inset, where islands that have reversed under thermal excitation at 100 °C appear with bright contrast. Scale bar 1 μm. (c) Lorentz transmission electron micrograph of artificial kagome ice after thermal excitation. The magnetization direction within the arms of the array can be determined from the detailed intensity profile across the arm [8], allowing the magnetic charge at each vertex to be inferred. Positive and negative magnetic charges are indicated by the overlaid red and blue dots, showing that the sample is in the charge-ordered, kagome ice-II state. Scale bar 500 nm. (d) The number of configurations that can be created in an artificial square ice by an applied field is very sensitive to disorder. The diagram represents all possible configurations that can be realized by application of the field (with magnitude slightly larger than the mean coercive field) to sixteen elements starting from a saturated type II state with a distribution of switching fields. Each unique configuration is indicated by a circular dot [9].

vertex configurations, but rather a consequence of the arrangement of non-degenerate vertices. The creation of quasiperiodic structures [14], containing varying numbers of neighbouring nanomagnets at the vertices, therefore opens the way to tailor the ground state degeneracy and to discover of novel phenomena in artificial spin ice such as channels of specific vertex types [13] that may act as conduits for transport of magnetic charges [15, 16]. The consequence of these modified lattices for field- and thermally-driven behaviour is a fascinating area for future experimental exploration.

Another way to control the magnetic behaviour of artificial spin ice is to alter the dimensions of the individual nanomagnets. Chopdekar *et al* demonstrate in this collection the possibilities for this kind of control in the building blocks of the artificial kagome ice consisting of one, two and three hexagonal rings of nanomagnets [17]. By manufacturing ring structures containing nanomagnets with two different widths, they showed that it is possible to achieve specific magnetic states during magnetization reversal that have a specific ring chirality, i.e. having either clockwise or anticlockwise circulation of the nanomagnet moments around a ring. Despite the presence of disorder in the form of a variation in the switching field from magnet to magnet, this control is feasible because of the dipolar coupling between closely spaced nanomagnets. These finite nanomagnet systems are of potential interest for spintronic devices because they can be scaled down in size and positioned close together without changing the fundamental magnetic states that can be accessed with fields or currents.

Continuing on the theme of nanomagnet geometry, Schumann *et al* have demonstrated that it is possible to create stable domains of high energy vertices in artificial kagome ice with a magnetic field [18]. The trick here is to use narrow magnets, where the effect of the shape anisotropy dominates over that of the dipolar coupling, and applying a magnetic field parallel to one of the three sets of nanomagnets results in vertices, which alternate spatially between all moments pointing in or all pointing out. Rougemaille *et al* demonstrate with micromagnetic simulations that these unfavourable vertices have an additional degree of freedom that is a result of the curling of the magnetization at the nanomagnet ends [19]. The magnetization curling at a vertex has a clockwise or anticlockwise chirality, which can be set by the application of magnetic field and leads to magnetization reversal via chains of reversed islands with a specific directionality.

It is also important to note that one can achieve specific configurations of magnetic moments in an artificial spin ice with a particular sequence of applied magnetic fields. Budrikis *et al* used numerical simulations to demonstrate that by applying a magnetic field along certain directions, it is possible to control the reversal of specific nanomagnets or chains of nanomagnets [20]. One could now imagine combining these specific field sequences with some of modifications to the spin ice topology described above in order to access specific states that are determined by several parameters: the lattice geometry, the individual switching fields of the elements and the history of applied field. This would lead to possibilities to utilize artificial spin ice as a ‘reconfigurable magnonic crystal’ for future programmable high frequency devices [4].

3. Monopoles and magnetricity

An understanding of dynamics in artificial spin ice can be based solely in terms of single element reversals. An alternate point of view is to consider instead the creation, and flow, of magnetic charge associated with vertices.

The charged vertex picture has great utility for the interpretation and understanding of phenomena occurring in artificial spin ices, and provides a useful starting point for systematic explorations of the processes determining magnetization reversals in them. Using magnetic force microscopy and simulations, Phatak *et al* [21] reveal statistics of vertex type and correlations between vertices that are strongly dependent on interaction strength as controlled by lattice spacing. Effects of disorder are found to be significant, especially for larger lattice spacings. The role of disorder is discussed by Morgan *et al* [22], whose experimental studies provide evidence of the ordering of magnetic charges associated with vertices, as well as indications that charged vertices can interact via long range dipolar forces.

Building upon the concept of mobile magnetic charge carriers, one can imagine the tantalising prospect of 'magnetricity': the creation and flow of mobile magnetic charges that respond to externally applied magnetic fields [23]. The possibility of magnetricity can be examined fruitfully using a monopole model for magnetic charge dynamics [24]. In this picture, magnetization reversal in a single element can create a pair of oppositely charged vertices. The charged vertices appear as defects in the magnetic configuration of the ice. These are defects in the sense that they are spatially localized excitations, with energies above the ground state, that can move and exhibit complex dynamics in response to magnetic fields as well as thermally driven processes [15, 16].

The charged vertex pairs share characteristics associated with magnetic monopoles joined by open ended Dirac strings as introduced by Nambu [25] with reference to Abrikosov flux lines. Nascimento *et al* suggest that this conception of monopole is an appropriate description of the charge pair excitations in artificial spin ices [26]. Using Monte Carlo simulations, Silva *et al* [26] argue that the average separation between paired monopoles in square ice at finite temperature is a weakly increasing function of lattice size, and isolated monopoles can be expected only in the thermodynamic limit. By comparing different lattice geometries, Wysin *et al* [27] present results from simulations employing Langevin dynamics to examine the differences in magnetization processes displayed between Ising and three different lattice models. They explain the differences in monopole separations and magnetic hysteresis in terms of how configurational phase space is explored by thermally driven reversal of element magnetizations. Moreover, their results suggest that careful tailoring of geometry and magnetic material parameters can, at least in principle, allow the realization of ASI with minimal trapping of monopoles.

If magnetricity is to be a true magnetic analogy to electricity, one would want a collection of magnetic monopoles that are able to behave as a gas of interacting quasiparticles. The degree to which the monopoles associated with charged vertices behave as a collection of particles is a subject of investigation in which we confront several intriguing issues: the precise nature and range of interactions governing reversal magnetic elements and the consequent motion of monopoles; the effects of imperfections and disorder on monopole mobilities; and how the ASI network geometry controls the topological structure and formation of emergent monopoles and their ensuing dynamics.

Whereas the studies discussed above are performed for monopoles constructed from the uncompensated magnetic poles at the ends of finite sized elements, Ladak *et al* [28] discuss magnetically charged vertices created in connected networks constructed from magnetic wires. These vertices emerge at the intersections of the magnetic wires when arranged in a honeycomb lattice. Monopoles in these systems are created by the emission and interaction of magnetic domain walls at the wire intersections.

In this scheme, a form of Coulomb blockade can operate, controlling the mechanism through which magnetic switching occurs in the regions between intersections. Ladak *et al* illustrate the sensitive dependence of magnetic switching and monopole formation on the internal structure of the magnetic domain wall. By contrasting results obtained from networks that support transverse walls with results from networks wherein the domain walls are of the vortex type, they find that monopoles form via Coulomb blockade in the transverse wall case, but not the vortex wall network.

Shen *et al* [29] also examine the dynamics of magnetic charges in continuous honeycomb networks using micromagnetic simulations. Their results provide evidence for interactions between charges, and examine effects associated with quenched disorder in the network. Statistical arguments are presented in which an analysis of magnetic reversal events is made based on a distribution of local coercive fields. The authors describe reversal processes in terms of avalanches which propagate between vertices, reporting a $1/n$ distribution of avalanche lengths.

4. Statistical mechanics: from effective to real

Artificial spin ices can be considered to be two-dimensional realizations of the 'toy' models of statistical mechanics that were first considered as early as a few decades ago [30, 31]. In the bulk of work to date, artificial spin ices are almost exclusively athermal systems, since the energy scales for reversal and magnetostatic coupling of the individual magnetic nanoelements are higher than $k_B T$ by orders of magnitude. Nevertheless, even in these athermal studies, they have been shown to obey an effective thermodynamics, within which an effective temperature [32] and entropy [33] can be defined and studied. This is somewhat analogous to the 'effective temperature' that is occasionally utilized in studying the physics of granular systems [34].

Despite the fact that true thermalization in ASIs near room temperature is experimentally very challenging, several early theoretical and numerical studies examined the behaviour of these systems with thermal behaviour. One such study [35], appearing this 'focus on' collection, examines the behaviour of colloidal particles arranged in two-dimensional arrays of bistable traps in the square and kagome lattices. Here, the behaviour shows multi-step ordering with lowering temperature, progressing from an ice-rule state through to a charge-ordered state at the lowest temperatures. One method for simplifying the modelling of thermal behaviour is to consider the system as an ensemble with a flow through an effective potential, a technique known as *Gibbsianization*. This method has provided insights for the theory of thermalized artificial spin ice [36]. Another Monte Carlo study, examining the thermalization of square ice shows the nucleation and growth of ordered vertex domains as the effective temperature is reduced, with possible extensions to ferroelectric spin ice [20].

The study of thermalization in ASI systems took a new direction with the publication, in 2010, of the work of Morgan *et al* [10], which showed that square-lattice ASI samples could indeed be induced past the frustrated state into an ordered ground state. Since this is not expected at room temperature, and no explicit thermalization was carried out, the authors reasoned that the thermalization they observed occurred during the growth of their samples, when the thin magnetic layers provides a reduced barrier allowing thermalization, even at room temperature. A theoretical study, appearing in this 'focus on' collection [37], builds a model to describe this observation. The model predicts a critical line, depending on the growth temperature and lattice constant, which is necessary to produce the ordered state. Surprisingly, the model predicts that samples grown *above* the critical temperature exhibit ordering, while those below remain disordered. Shortly after the work of Morgan *et al*, a second study also showed strong indications of ordering in a square ice sample fabricated from a material with low Curie temperature [38]. This material is fabricated from a thin Fe layer contained within an itinerant Pd host, resulting in a material with relatively high magnetic moment and low Curie temperature. Square ice samples made from this material show a collapse of magnetic moment below the Curie temperature and a dependence of the remanent moment on the direction of applied field between [10] and [11], both indications of interacting thermal behaviour. Shortly after this study, a theoretical article appeared in this 'focus on' collection describing the observed thermal behaviour in both cases [39]. The model is based upon the strength of the dipolar interactions, which is parameterized by an energy D. It predicts a transition temperature, $T_p \approx 7.2D/k_B$, above which free monopoles may appear.

A ground-breaking shift occurred in the study of thermalized artificial systems during 2013, coming from several studies that showed the ability to image individual magnetic elements during the course of controlled thermalization. The first of these, from Farhan *et al*, takes advantage of x-ray magnetic circular dichroism photoemission electron microscopy studies (XMCD-PEEM) to conduct real time observations of the switching of magnetic elements in small kagome ice crystals, where the samples are made intentionally thin to encourage thermal flipping [40]. The study shows that, due to the increasing number of degrees of freedom and complexity of the configuration space, larger and larger crystals are increasingly unlikely to find their respective ground states. Three subsequent studies, appearing in the middle of 2013, all show the ability of sizable crystals of square ASI to achieve their ground states through intentional and genuine thermalization [7, 11, 12]. The study by Porro *et al*—appearing in this 'focus on' collection [11]—used a permalloy material that is deliberately *de-tuned* to be nickel-rich, in order to decrease the Curie temperature. They observe that square ice samples fabricated from this material will anneal into ordered domains of the type I ground state, almost exactly as those observed in the as-grown samples of Morgan *et al* [10]. They further show that—when applying the rotation-demagnetization protocol—the same samples are not able to attain the ground state, cementing that demagnetization protocols used in such systems are athermal. The work by Zhang *et al* [12] showed similar results on optimal-stoichiometry permalloy, with significantly larger domains of the type I-ordered state. In the same study, it was also reported that kagome ice samples, similarly fabricated and studied, would remain disordered, but would enter into a state where the excess magnetic charge at each vertex would exhibit a tendency toward ordering, with alternating magnetic charge on each lattice. Such charge-ordered domains are small, but it stands out as the first observation of such a state, which had been previously predicted [41]. Shortly after, Farhan and coworkers also showed, by XMCD-PEEM, that square ASI samples could attain the ground state, but they added direct and realtime observations, showing a process by which alternating chains could propagate along [11] directions to produce a type I ordered ground state [7]. Taken together, these most recent thermalization studies really throw open the door for the future of ASI work, revealing the true promise of these systems as a statistical mechanics testbed. They allow the exact geometry and structure of the lattice to be controlled, virtually at will and in sharp contrast to other materials emerging from solid-state synthetic methods, and provide the possibility of thermalization of the system and to direct observation of the microscopic effects in realtime.

5. Outlook

The field continues to expand and move forward at a remarkable pace, exploring many new directions. These include the cross-over with magnonics in terms of the analysis of the spectra of high frequency dynamics [42], studies of disorder-induced criticality [43], proposals for three-dimensional systems [44], and the development by Budrikis *et al* of network theories to represent the changes of state in artificial spin ice systems [45]. From a more technological perspective, viewing these systems as information storage [33] or processing technologies could also lead to nanomagnetic logic architectures [46] based on frustrated arrays. These could take on novel forms: for instance, the Ising model provides the theoretical underpinning for the Hopfield model of neural networks [47], suggesting that neuromorphic architectures based on artificial spin ices might be possible. Advances in the field of nanomagnetic logic also reflect back into new experimental opportunities in artificial spin ices, for instance driving out-of-equilibrium dynamics using spin Hall torques [48]. It is clear that this field still contains many more open questions and will lead to several surprising answers in the future.

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