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ARTIFICIAL FERROIC SYSTEMS:
New functionalities from structure, interactions and dynamics

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Contents

1. Introduction

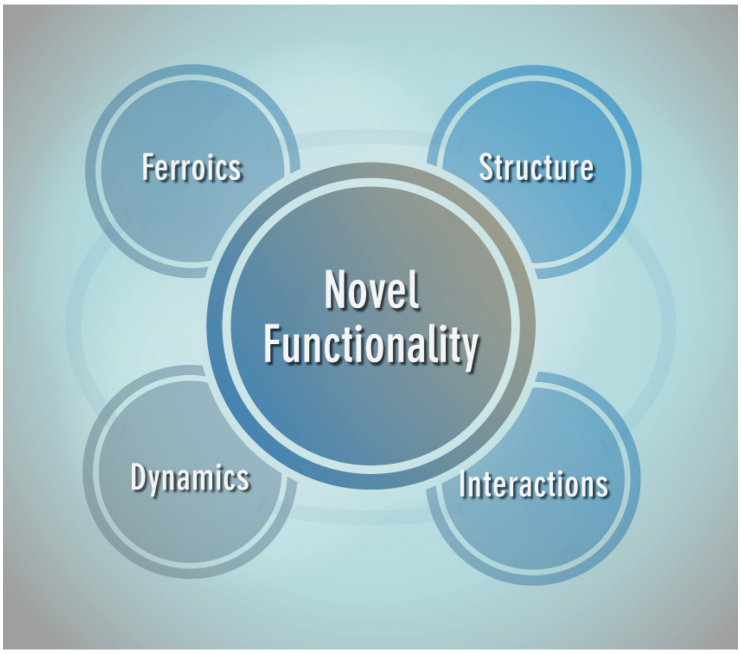

Figure 1: Novel functionality can be gained from artificial ferroic systems consisting of interacting structures with magnetic and/or electric properties. This functionality has the potential for new devices activated by a magnetic and/or electric fields, in either quasi-static or dynamic regimes.

An important but often elusive characteristic of nature is the appearance of new functionalities that emerge from the combination of a variety of interacting elemental building blocks. Here we focus on engineered condensed matter systems, specifically thin film systems made up of several *ferroic* components where new functions and properties appear (figure 1). These *artificial ferroic systems* can be either entirely magnetic or a mixture of magnetic and ferroelectric constituents, and the structure in these systems is created either during thin film growth or with lithographic methods. Such systems have many degrees of freedom that are much more complex and tuneable than simple thin films and, in our approach, we explore how potentially useful properties arise on different time and length scales, and how the timescales for specific dynamic phenomena can be controlled. To be more precise, we mainly discuss the properties of ferroic structures patterned into single domain elements with individual volumes on the order of 10^5 nm^3, and dynamics that occur with characteristic times in the range 10^{-8} to 10^{-10} s.

We begin with *Artificial Spin Systems*, which are arrangements of interacting nanomagnets that provide model systems for their bulk crystal counterparts. A straightforward example would be to represent an antiferromagnet having alternating spins on the crystal lattice with a series of dipolar coupled nanomagnets with out-of-plane anisotropy placed, say, on a two dimensional square lattice. One can also consider more complex arrangements that lead to geometrical frustration such as those found in artificial spin ice, which will be one of the key motivations of the current review. We will describe the ordering and response to slowly varying applied fields and temperature, demonstrating how interactions between elements can lead to novel material behaviour such as non-trivial avalanche dynamics and the appearance of quasi-domains. We also show that these systems can be controlled, which is a pre-requisite for device applications.

We then move on to the implementation of micro- and nanoscale structures as *Tunable Magnonic Crystals,* progressing from the quasi-static behaviour described in artificial spin ice to dynamics at radio frequency timescales. The magnetic properties of the magnonic crystals are determined by the structure in terms of magnetic material, element size and shape. One can thus control magnetization processes and response to applied fields through composition and geometry, and thereby adjust the characteristic timescales governing the creation of new magnetic states and equilibration. Moreover, by controlling the static configurations and their evolution, one also determines many details of the allowed excitation spectra. Indeed, ferromagnets have natural resonances in the microwave region, providing new possibilities to manipulate microwaves in magnonic crystals, a magnetic analogy to photonic crystals, where new and useful dynamic properties emerge by patterning magnetic thin films.

We complete this review with *Functional Ferroic Composites*, in particular combining a ferroelectric material with a ferromagnetic material to give an artificial multiferroic, with improved magnetoelectric properties compared to those of multiferroic single phase crystals. The two ferroic materials can be combined in different ways, for example, either as particles of one material in a matrix of the other or as a layered system. These can be referred to as composite or hybrid systems, which make use of the contrasting properties of the two or more different materials to improve performance or engineer novel properties. For such composites, it is not only the combination of the properties of the individual components of the composite that are important, but more crucially the nature of the coupling at the interfaces that can be mediated by strain but also other phenomena. We discuss here how some of these properties create new possibilities for active control of desired responses to time varying electric and/or magnetic fields. We then introduce microwave frequency properties and present the unique features associated with the new functionalities of the artificial materials.

One might ask why *artificial ferroic systems* have the potential for new possibilities and opportunities, not already considered in the myriad examples of well-studied multiferroic materials and magnetic heterostructures. The first answer to this question is the possibility to discover fascinating, new physics which emerges when micro- and nanometre-sized elements with different properties are brought together and arranged in different geometries in such a way that inter-element interactions generate distinctive, collective behaviour. The second answer, that is more relevant to society, is to create novel physical phenomena that will lead to new and improved devices. Since these materials are polarisable, they are technologically relevant because of their susceptibility to electric and/or magnetic fields, allowing these systems to be implemented in various device applications such as data storage, memory and logic, and as nano-oscillators for communications. In particular, the requirements for information technology and communications include scalability and high density, which necessarily means small size, faster data storage or transfer, higher output signals, low power consumption, low cost, and reproducible control of the magnetic states. Whereas the materials we discuss are well known for their utility in such applications, the emergent collective properties when patterned into the arrays of elements have not been examined with a view towards devices. The concept of artificial ferroic systems that we review here, will not only lead to the creation of high performance materials by design but also holds the promise of enhanced electromagnetic functionality by exploiting the sensitivity of the radio frequency response to the static magnetic configuration. This is an example of how patterned magnetic arrays, in which magnetic configurations can be changed using applied magnetic fields, can provide new functionality, since controlled modification of the magnetic configuration will give a specific change the allowed magnetic excitations. Extension of this to electric field control will be possible by replacing magnetic with multiferroic or ferroelectric/magnetic composites. With these new degrees of freedom in design and construction, and interactions giving rise to novel collective behaviour, artificial ferroic systems have the potential not only to satisfy the above application requirements but also to provide new technological breakthroughs in the coming years.

2. Artificial Spin Systems

We begin by outlining an intriguing class of metamaterials, which consist of particular arrangements of interacting nanomagnets and are referred to here as *artificial spin systems*, with their characteristic length scales and energies distinguishing them from spin systems frustrated at the atomic level due to competing interactions between the individual spins. In the artificial spin ice, the elements of the system are sub-micrometre in size in order to ensure that they are single domain, but have mainly up to now been sufficiently large to suppress thermal fluctuations. The interactions are magnetostatic in origin and can be controlled by simply choosing the distance between nanomagnets, which have lateral dimensions typically on the order of 10's to 100's nanometres. The interactions control how the static order responds to applied magnetic fields or temperature and, the nature of the resulting magnetization dynamics.

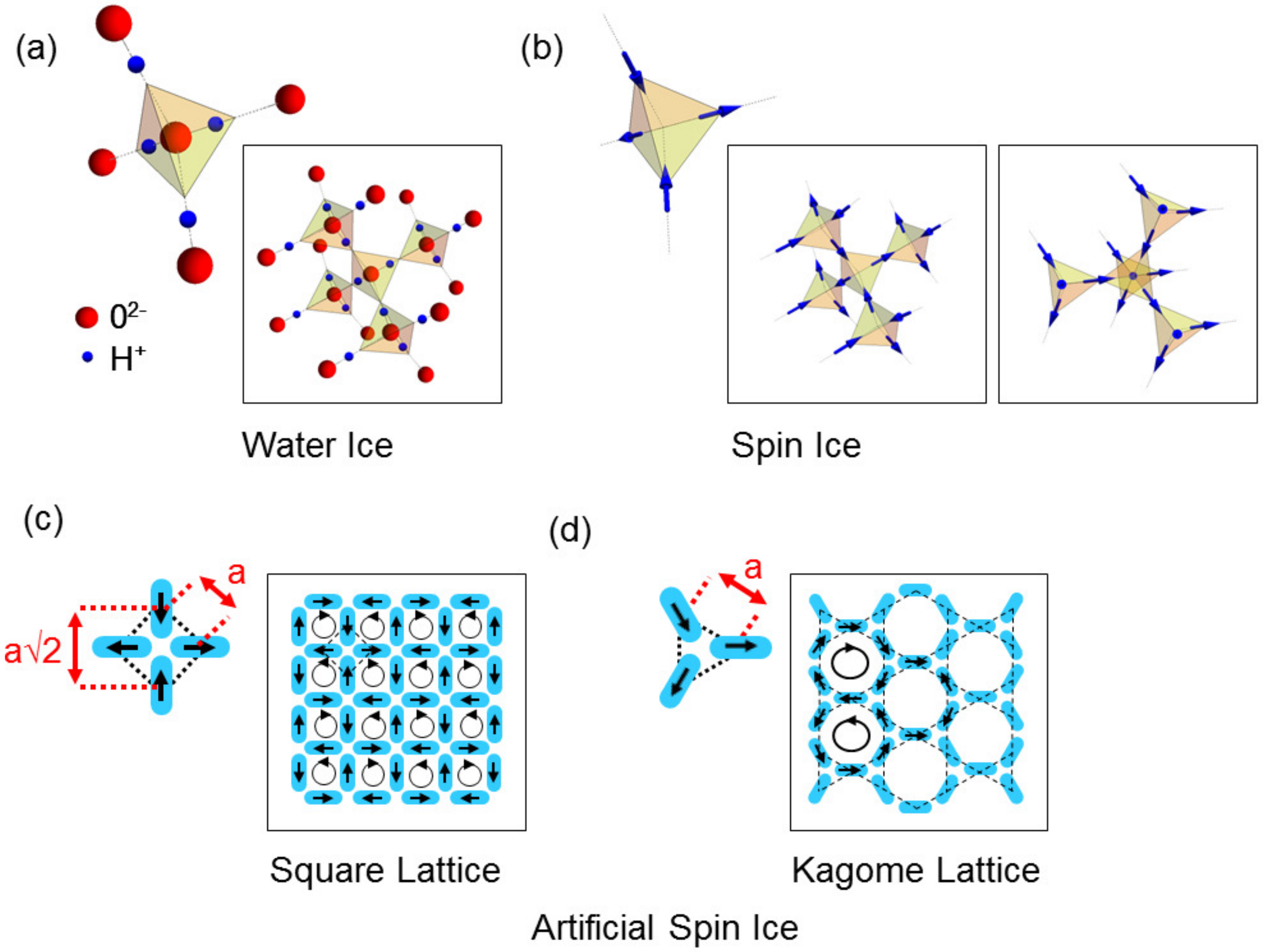

Figure 2: Evolution of artificial spin ice from water ice: (a) Water ice structure with two protons sitting closer to the oxygen atom and two sitting further away (view of single tetrahedron and five corner sharing tetrahedra), (b) the spin structure of the rare earth titanate pyrochlore 'spin ice' (view of single tetrahedron and two views of five corner sharing tetrahedra, with the right hand view emphasizing the kagome planes) with the corresponding artificial spin ice systems: (c) with elongated single domain magnets arranged on the square lattice and (d) with elongated single domain nanomagnets arranged on the kagome lattice. The ground state configuration with the lattice parameter, taken as the distance between the centres of two neighbouring islands, is indicated for each lattice type. Reproduced from [1] with kind permission of Springer Science+Business Media.

Important in these systems is the magnetic polarisation that drives the behaviour and, as mentioned above, their interaction via dipolar coupling, which is mediated by the electromagnetic field. Long range ordering of dipoles is of course an old problem, and has been studied extensively in many contexts. In one and two dimensions, long-range ordering of electric or magnetic moments in lattice geometries is possible [2]. Since the fields from dipolar sources are not simply spherically symmetric, the nature of long range ordering in dipolar lattices is very strongly dependent on details of the geometry [3]. For example, rhombic arrays of dipoles can support either ferro- or antiferromagnetic orderings depending on the vertex angle. There is moreover a strong dependence in two dimensions on array edges, which can destabilise the ordering in favour of vortex or domain formation [4-7]. While early work has primarily been concerned with microscopic dipoles, extension to magnetic particles soon followed [8-11]. There has been a flurry of efforts over the past decade to construct and understand the static and dynamic behaviour of such dipolar coupled nanomagnet systems. In what follows we comment primarily on systems which show potential for emergent behaviour that can arise from frustration and competing interactions. Specifically, we describe systems that have in recent years come to be known as *artificial spin ice*.

2.1 From water ice to artificial spin ice

It is instructive to understand the origins of the term 'artificial spin ice' (figure 2), which began with the discovery of geometrical frustration in the rare earth pyrochlore $Ho_2Ti_2O_7$ [12]. The term 'frustration' in physical systems refers to the inability of the system to satisfy all interactions simultaneously and this leads to a large degeneracy of low energy states and non-zero entropy at absolute zero. In water ice the frustration comes about because of an incompatibility of the bonding distances with the tetrahedral geometry leading to the lowest energy state at a tetrahedron given by the ice rule: two protons (or hydrogen ions) sit closer to the oxygen ion and two are positioned further away (figure 2a).

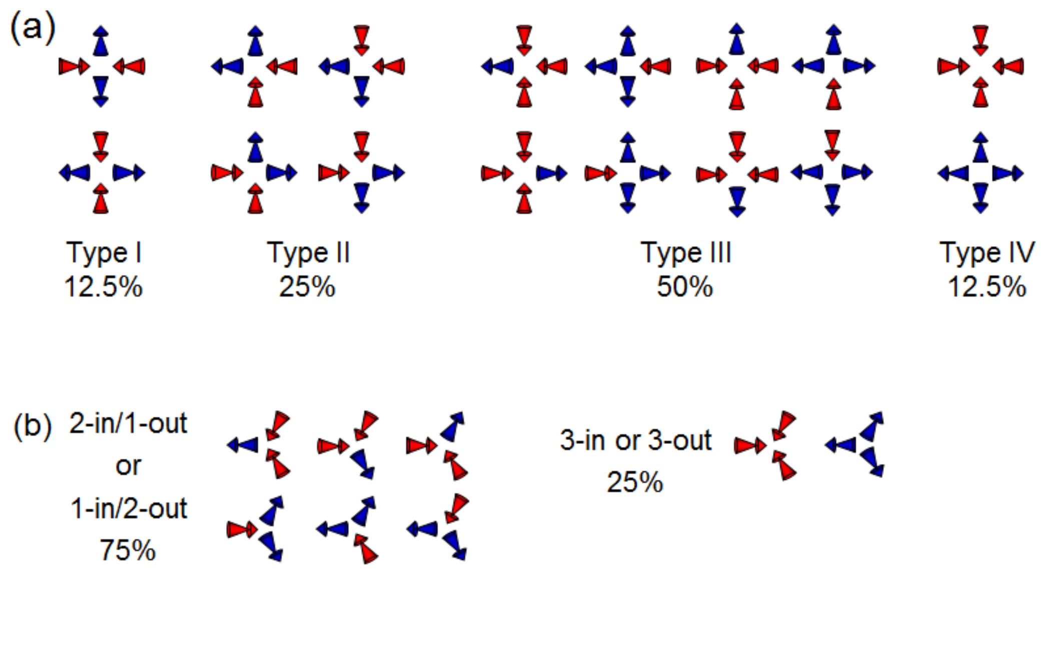

Figure 3: Magnetic moment configurations at island vertices. (a) The sixteen possible moment configurations at a four island vertex in the artificial square ice, sorted into the four different types with increasing energy. (b) The eight possible moment configurations at a three island vertex in the artificial kagome spin ice, sorted into the two different types. The numbers indicate the expected percentage of each type if the individual moment orientations in an array were completely random corresponding to zero dipolar coupling. Reproduced from [1] with kind permission of Springer Science+Business Media.

A corresponding phenomenon in the rare earth pyrochlores comes about due to a combination of the interactions between the spins on the corners of the pyrochlore tetrahedra and the Ising anisotropy along the 111-type directions (figure 2b), which arises from the crystal field and causes the spins to point towards or away from the centre of the tetrahedra. This leads to the equivalent two-spins-in/two-spins-out ice rule for the lowest energy state. For the crystal, which consists of an extended arrangement of corner sharing tetrahedra, the lowest energy (or ground) state is then composed of a large number of such two-in/two-out arrangements. The bulk spin ice crystals display a whole host of interesting thermal and field dependent behaviour that arises from the frustration [12-16]. However, the specific control of the lattice and spin geometry is virtually impossible, and complete knowledge of the orientation of each individual spin is not feasible with current experimental methods.

In 2006, it was the group of Peter Schiffer who came up with the idea of mimicking the behaviour of the three dimensional spin ice crystal with a two-dimensional arrangements of dipolar coupled nanomagnets created with electron beam lithography, so-called artificial spin ice (figure 2c) [17]. These nanomagnet systems can be fabricated from thin film ferromagnetic materials such as Permalloy (an alloy of Ni80%Fe20%) or cobalt, and the size of the nanomagnets is chosen to ensure that they are single domain with the now macroscopic magnetic moments taking on the role of the Ising spins in the bulk crystals. The nanomagnets are elongated and the resulting shape anisotropy causes the moments to align parallel to the long edge of the nanomagnets, so that they can point along one of two directions. The innovative idea of artificial spin systems is that one can tune the array geometry, not only placing the nanomagnets on lattices found in nature, but also in other geometries, and that the magnetic configurations are directly visible using a host of different microscopy techniques sensitive to the magnetisation, magnetic induction or magnetic stray field arising from the nanomagnets [17-21].

As a result of the dipolar interactions, which favour head-to-tail magnetic moment interactions between nearest neighbour nanomagnets meeting at a vertex (i.e. favouring north pole facing south pole), the lowest energy local configuration or corresponding ice rule for magnetic moments in an artificial square ice is two-in/two-out where four islands meet as shown in figure 2c. There is, however, an issue with the square ice, pointed out shortly after the original work was published [22], since the dipolar coupling between the islands at the vertices are not equivalent i.e. neighbouring islands at a vertex are closer together than the islands on opposite sides of the vertices, and the relative orientations between the islands are different (see figure 2c). One could consider lifting one sub-lattice with respect to the other [1, 22], but this is somewhat difficult to construct experimentally. Another possibility is to consider instead the artificial kagome (or hexagonal) spin ice [22, 23], where the elongated magnetic islands are placed on the sites of the kagome lattice to form the links of a honeycomb. This arrangement corresponds to studying the behaviour of a bulk spin ice with a magnetic field applied along the [111] direction [24] and can be visualized by rotating the pyrochlore lattice so that the kagome planes are orthogonal to the line of sight as shown in figure 2b. In such an artificial kagome spin ice, the interactions between the islands at the three-island vertex are the equivalent.

When considering the local magnetic moment configurations at a vertex where the islands meet, one finds that for the square ice there are sixteen possible arrangements (figure 3a) [17], which can be separated into four different vertex types. The vertex energy increases when going from Type I to Type IV although, if one assumes a random distribution of configurations, the percentage of each vertex type is given by the degeneracy of a given vertex type, with the respective values specified in the figure. There are two types of configurations with two moments pointing in and two pointing out, but Type I has a lower energy due to the fact that the interactions between nanoislands at a vertex are not equivalent and leads to a unique ground state comprising a checkerboard of alternating vortices with island moments circulating clockwise and anticlockwise (see figure 2c). For the artificial kagome spin ice there are two vertex types, the first obeys the ice rule given in this case by two-moments-pointing-in/one-moment-pointing-out or vice versa and is the lowest energy configuration. The second is a higher energy state with all moments pointing in (or out) at a three island vertex so breaking the ice rule. The ground state, determined from a long range dipolar calculation which should be taken into account when considering these systems [25], consists of repeated units of a three-ring configuration [26, 27]. Here, two of the rings form vortices of circulating head-to-tail moments, one with the moments circulating clockwise and the other with moments circulating anticlockwise. The third ring has alternating head-to-head and tail-to-tail neighbouring moments (figure 2d). It remains to be seen whether such a configuration can ever be achieved, since the series of head-to-tail moments in the third ring locally forms a very high energy configuration. At this point, it is important to emphasize that in artificial ferroic systems where elements are coupled primarily through dipolar interactions, one should consider the long range nature of the dipolar coupling. There can be significant differences between models that consider all elements when calculating the long range dipole interactions in artificial spin systems and those which consider only elements within a certain distance range, with a near neighbour only model being the smallest range [28]. It is therefore important to remember that predicted magnetic configurations, and the behaviour in response to external stimuli, can in some systems depend strongly on approximations made in modelling of long range interactions [5].

The particular magnetic configuration observed in artificial spin ice is dependent on the field and thermal history. Here, the lattice type dictates the extent of frustration and therefore the extent of the magnetic order, and also the detailed geometry in terms of island size and lattice parameter (or distance between neighbouring islands), determines whether it is the dipolar coupling or the shape anisotropy, or a combination of both, that dictates the magnetic behaviour of the array. For example, in the artificial kagome spin ice, the ice rule in demagnetised systems is robust for strong dipolar coupling [18, 19]. However, the ice rule can be broken on application of a magnetic field [21, 29], leading to a regular array of ice rule defects at low dipolar coupling [30, 31]. It should also be pointed out that, while arrangements of dipolar coupled nanomagnets are thought to be a good way to mimic the behaviour of Ising spins, one can also consider systems made up of connected nanowires that, for example, form a honeycomb network with the artificial kagome spin ice geometry [19, 21, 32-34]. In the connected systems, the fundamental reversal processes are different from those of the individual nanomagnets. Here the reversal occurs via domain walls passing through the system and one can consider these walls to be acting as charge carriers [33, 35]. The connected artificial square ice is often referred to as an antidot array [36], since one can think of the structure as a magnetic thin film with an array of holes in a magnetic thin film. Such antidot arrays have been of recent interest for use as magnonic crystals [37], described in Section 3, and for the generation of antivortices [38].

Artificial spin ice is a particular realisation of arrangements of magnetic moments that mimic the spins in the crystal counterpart, and is part of a much larger family of *artificial spin systems* that encompasses several different arrangements of dipolar coupled nanomagnets. This includes a whole host of other planar geometries such as modified basic spin ice lattices [39], those that use a triangular lattice as a template [40, 41], and those that place the nanomagnets on the links of a Kagome lattice [42] or on quasiperiodic lattices [43]. One can also study magnetic elements with perpendicular anisotropy, which are interesting for applications such as bit patterned media [44], and correspond to perpendicular Ising spins placed, for example, on a triangular, kagome or honeycomb lattice [28, 45]. Each of these systems has its own unique behaviour in terms of the ordering of the magnetic moments and, because one can fine-tune the geometry, in terms of the lattice, disorder and defects, they provide ideal systems for studying return point memory [46]. It should also be mentioned here that one can create pseudo-spin systems from non-magnetic components, for example using colloids [46-50], by constructing a potential landscape for trapping superconducting vortices [51], or by fabricating an array of dipolar coupled superconducting rings [52].

One can also study systems based on magnetic elements, which interact at very different length scales. At the smallest length scales, clusters of carefully positioned antiferromagnetically coupled atoms are coupled by Ruderman–Kittel–Kasuya–Yosida exchange interactions [53]. In contrast, macroscopic magnetic systems [54, 55] have much slower reversal timescales on the order of seconds, allowing the dynamics to be effortlessly recorded, and offer the possibility to generate an effective temperature by applying random local fields, with an electromagnet assigned to each macroscopic magnet. Despite the variety, all of these systems provide a way to study the effects of frustration, each offering its own advantages in terms of length and time scales, the measurement techniques available to record the behaviour, options for applying external stimuli and possibilities to tailor the systems.

In the next section, we will describe various areas of particular interest to the artificial spin ice community. We first discuss attempts to reach the ground state primarily using field anneal protocols, but also more recent work involving thermally activated systems, and then describe the emergence of various phenomena from the collective behaviour of the magnetic moments in such two dimensional dipolar coupled systems including magnetic monopoles, avalanches and quasi-domains with their accompanying domain boundaries.

2.2 Approaching the ground state with magnetic field protocols

One key ambition since the first proposal to use two dimensional arrangements of nanomagnets as a model system for bulk spin ice [17] has been to study behaviour analogous to the thermally active spins in their crystal counterparts, and to determine how far it is possible to achieve the lowest energy configurations using an effective thermal anneal involving demagnetization with an applied magnetic field. Such demagnetisation methods have involved rotating the sample either about an in-plane or out-of-plane sample axis in a magnetic field that is applied orthogonal to the axis of rotation. During rotation, the applied field is reduced from a value above saturation down to zero and following such a demagnetization of artificial square ice samples, it has been observed that the nearest neighbour correlations dominate the behaviour of the system as in the bulk counterpart, but that the absolute ground state was not achieved. In finite systems consisting of clusters of dipolar coupled nanomagnets [18], it has been found that the ability to achieve the ground state via demagnetization reduces as the extent of the clusters increases and the ground state is only reproducibly achieved in the smallest clusters consisting of a ring of nanomagnets with no frustrated vertices. One might ask whether this lack of ground state in larger systems is an indication of the increasing frustration or just a matter of increasing complexity since, for larger clusters, the pathways to the ground state are more intricate. In order to address this question, clusters of islands based on the square ice have been studied where all clusters include the same number of islands but with varying degrees of frustration depending on the island arrangement [56]. In this work, it was indeed observed that the ability to achieve the ground state in clusters increases as the degree of frustration increases.

During demagnetization of an artificial spin ice, the orientation of the axis about which the sample is rotated does not appear to affect to the ability to achieve the lowest energy states [18]. However, the use of finer steps on reducing the magnetic field does lead to a reduction in the energy of the final state, and even produces a zero magnetic moment [57]. However, extrapolation of the dipolar energy to the values expected when the field step is reduced towards zero indicates that using such demagnetization methods will ultimately not lead to the ground state. It may turn out to be more effective to rotate the sample in a magnetic field that is maintained at a constant value, [58] which is predicted to give a maximum of ground state vertices. Here the field should not be set too high, otherwise the moments would simply follow the field, but should rather be set just above the field at which the moments freeze in to ensure a dynamic response. Another possibility is to vary the sense of rotation during demagnetization so that, in combination with disorder resulting from the switching field distribution of the nanomagnets, the system is prevented from getting stuck in higher energy local minima [59].

It is instructive at this point to consider the athermal ice problem within the context of population dynamics using a type of mean field theory. In this approach one can identify allowed processes described in terms of vertex interactions [58]. This approach provides estimates of vertex populations that can be achieved by specific field protocols. The method is fruitful in that the average populations of different vertex types can be compared with those obtained from images taken in experiment.

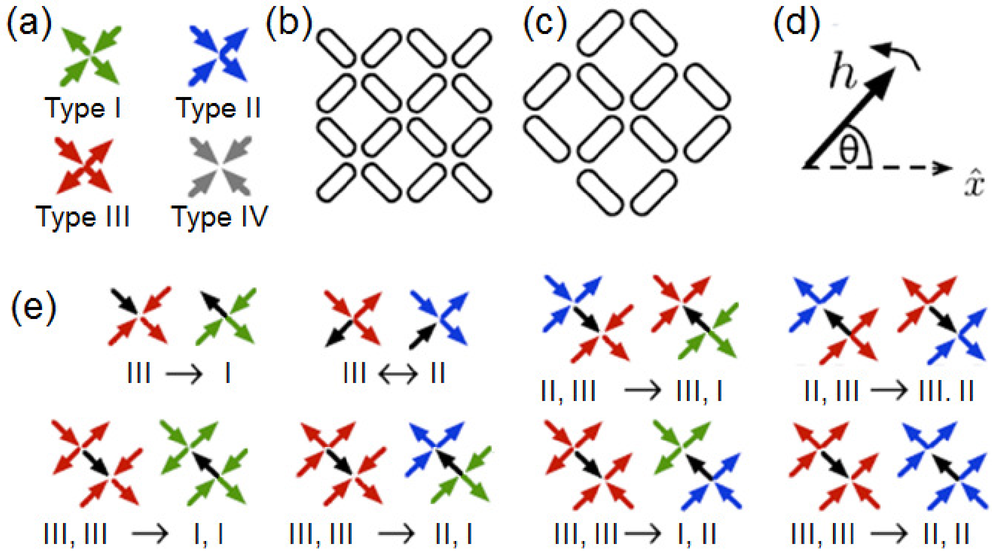

Figure 4: The geometry of artificial square ice and vertex interactions. In (a) examples of each of the four types of vertices are shown with Type I having the lowest energy and Type IV having the highest energy. In (b) "open" and (c) "closed" edge geometries for a finite array are depicted. In (d) the magnitude and direction of an applied field in a rotating protocol is defined. Examples of the dynamics involved in creating configurations are shown in (e). The first two are examples of how a single spin flip transforms one vertex type into another, and the other diagrams illustrate how single spin flips drive vertex pair processes. Reprinted with permission from [58]. Copyright 2010 by the American Physical Society.

One begins by defining transition rates Γ_{ij} of transformations between vertex types i and j via single magnetic element reversals for a specific configuration. We note that reversal of the magnetization of an element can be a complicated process. However this process occurs quickly when compared to the rates at which applied magnetic fields are changed. As such, we consider the reversals as simple reversals of magnetic elements, and we refer to these as 'spin flips'. The transition rates therefore describe in a simple way the dynamic processes associated with reversals of moments in individual magnetic elements that result in our so-called 'spin flips'. The characteristic time for such reversals is determined by the processes of reversal. Typically reversal will occur on time scales roughly determined by the rate at which energy from the magnetic spin system dissipates into the electronic and lattice systems. This occurs on timescales of the order of tens of nanoseconds, and is sensitive to details of the reversal process. Examples of some of the possible spin flip processes are sketched in figure 4. In this figure, the square lattice geometry and its vertex types are shown in (a). One needs also to consider the edges of the array, with two possibilities shown in (b) and (c). In the rotating field protocol, a fixed magnitude magnetic field is rotated with respect to the array through an angle θ (see figure 4d). Depending on magnitude and angle, different processes can occur, as shown in (e). The circled numbers denote the vertex type, and reversal of the black spins drives the transitions between single vertices or pairs of vertices.

A mean field theory describing the evolution of the populations to a steady state under a field protocol is made by first supposing that reversal of an element's magnetization s_i occurs when

$-\vec{H}_{loc}\cdot\vec{s}_i > h_c$, where H_{loc} is the sum of all dipolar and external fields acting locally on the element, and h_c is the coercive field for the element. Next, one can consider all possible spin flip driven transitions for a given field magnitude and orientation that transforms a vertex of type *i* to a vertex of type *j*. By averaging over these possible spin flip driven processes one is able to construct an average transition rate ν_{ij} that describes the overall change in vertex populations n_i and n_j. The same is done for the ν_{ijk} that describe correlated vertex transitions (as shown for six examples in figure 4). The population equations then take the form:

$$\frac{d}{dt}n_i = \sum_{ij} \upsilon_{ij} n_j + \sum_{ijk} \upsilon_{ijk} n_j n_k \qquad (1)$$

These define a set of coupled nonlinear population equations, and can be solved numerically for the stationary populations.

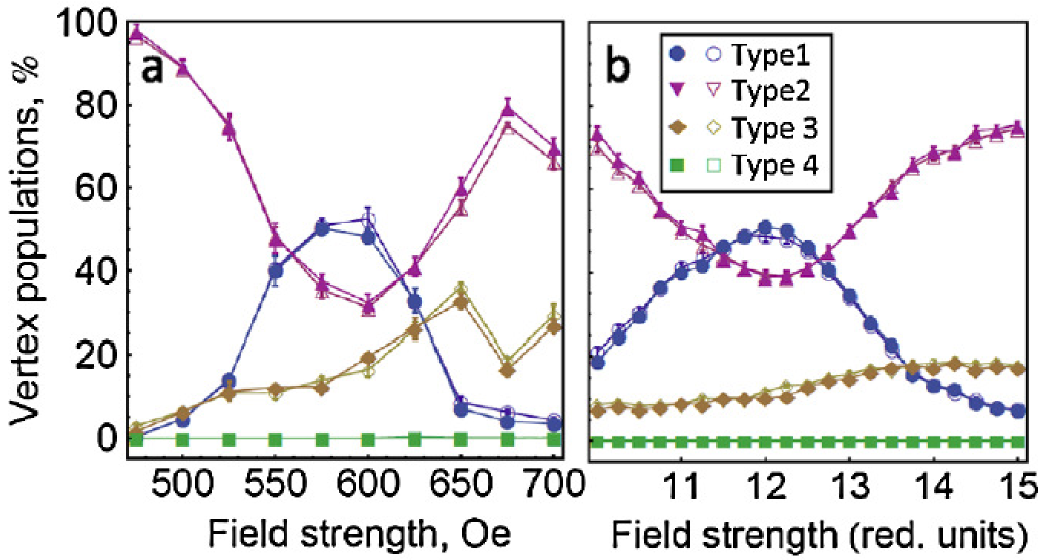

Figure 5: Vertex populations versus applied magnetic field strength from (a) experiment and (b) simulation of artificial square ice manufactured from Permalloy islands (thickness 30 nm, length 280 nm, width 85 nm, and lattice parameter a√2 = 400 nm). In both cases a magnetic field of fixed magnitude was applied and rotated in the plane of a square ice array. A distribution of switching fields is assumed, with a standard deviation equivalent to the near-neighbour interaction field. Symbols represent vertex types as shown in the legend, with open (closed) symbols for open (closed) edge arrays. The disorder given by the switching field distribution leads to very little difference in results for open and closed edges. Each data point is the average over several runs and error bars represent the standard error. The populations are sensitive to the applied field only within a range of applied field strengths, and this range is determined by the strength of the switching fields and inter-element interactions. The agreement between experiment and theory is obtained by assuming a Gaussian distribution of switching fields and using the width of the distribution as a fitting parameter. In this way, agreement provides a unique measure of the switching field distribution. Reprinted with permission from [60]. Copyright 2012 by the American Physical Society.

It should be noted that in order to approach real experimental systems, the effects of disorder must be taken into account. In a perfect system, the energies associated with the transition of one vertex type to another are distinct. However, the inclusion of disorder in either interaction strength or coercive field modifies the transition processes so that previously distinct processes can occur at similar energies. Such inclusion of disorder was shown by simulations to provide an excellent agreement with experimental determination of vertex populations under rotating field protocols for square ice [60]. Example results are shown in figure 5, with the results from experiment shown in figure 5a and the corresponding simulations shown in figure 5b where a disorder in coercive field was assumed. This fitting allows one to extract a value for the spread in coercive fields simply by counting vertices and, in the next section, we will describe work that demonstrates how this disorder is critical for defining the behaviour of avalanches of flipped nanomagnet moments [29].

In this section, we have described how demagnetization methods have been used as an effective thermal anneal in attempts to achieve the ground state in static artificial spin ice systems, where the shape anisotropy of the individual nanomagnets results in a high energy barrier to moment flips at around 10^5 K. Despite having static moments, one can consider such systems to be a thermal ensemble having an effective temperature [61, 62] and calculate the system entropy by considering configurations of smaller units in the arrays [63]. However, ultimately field protocols do not provide the stochastic dynamics associated with in a true thermal anneal. They are comparatively deterministic in nature, which comes about because the orientation of the moments is given not only by a combination of the local dipolar fields from neighbouring magnets but also the instantaneous applied field magnitude and orientation. This becomes most obvious for systems with large lattice parameters, and therefore low dipolar coupling, where all of the moments end up pointing towards the orientation of the applied magnetic field just before they freeze in [18].

2.3 Emergent magnetic monopoles

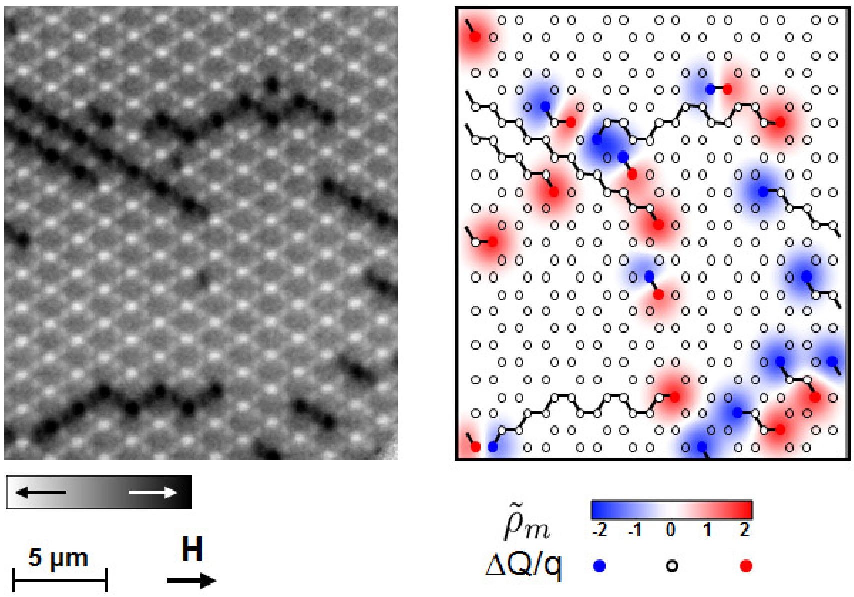

Figure 6: Experimental observation of emergent monopoles and Dirac strings in a quasi-infinite artificial kagome spin ice. To the left is a photoemission electron microscopy image of an artificial kagome spin ice consisting of Permalloy islands (thickness 20 nm, length 470 nm, width 160 nm, and lattice parameter 500 nm) together with the associated ΔQ map to the right. The Dirac strings connect monopole–antimonopole pairs with ΔQ = +2q and −2q indicated with red and blue dots. The ΔQ map is shown together with the dimensionless coarse grained magnetic charge density, ρ_m, both of which give signatures for nonzero charge density. Adapted by permission from Macmillan Publishers Ltd: Nature Physics. [29], copyright 2011.

The collective behaviour of the dipolar coupled nanomagnets in artificial spin ice results in emergent phenomena including magnetic monopoles and avalanches, described in this section, and quasi-domains described in the next section. Such emergent physics has been rarely used to describe the behaviour of an array of nanomagnets. In terms of fundamental science, this provides a new way to think about these systems that may in turn lead to the discovery novel physical phenomena. An additional ambitious goal would be to invent and design novel spintronic devices that take advantage of emergent phenomena, for example making use of magnetic charge transport.

We begin with emergent magnetic monopoles, which are quasiparticles predicted to occur in the pyrochlore spin ice as monopole-antimonopole pairs by Castelnovo et al. in 2008 [64]. The monopole pairs are connected via a so-called 'Dirac String', which refers to the concept used by Dirac to take into account Maxwell's equations ($\nabla \cdot \vec{B} = 0$) [65] with an arbitrarily narrow flux tube transferring magnetic flux between them. This prediction was followed by a series of experiments using Neutron Scattering to detect the signatures of the emergent magnetic monopoles and Dirac strings in the pyrochlore systems at low temperature [66-68]. Taking a natural step, it then became an important challenge to try to find corresponding charge defects in artificial spin ice [29, 32]. However, before proceeding, it is important to note that these *emergent* magnetic monopoles are captured in a condensed matter system and one should take care to distinguish them from their elementary particle counterparts [69].

At this point, it is important to know how to identify such charged quasiparticles in an artificial spin ice system. For this one can consider the dumbbell charge model introduced by Castelnovo et al. [64], and envisage replacing a magnetic dipole with two charges, one positive (+1q) and one negative (-1q), residing at neighbouring vertices. If one considers then the net charge at each vertex, Q, the ground state arrangement of charges for the artificial square ice consists of zero charge at each vertex, whereas for the artificial kagome spin ice, this becomes an ordered array of alternating Q = ±1q charges. In order then to locate the magnetic monopoles (antimonopoles) in a given magnetic configuration, one needs to consider the charge at each vertex in relation to the ground state of charges, ΔQ. The emergent magnetic monopoles (antimonopoles) can be found at the sites where ΔQ = ±2q [1, 29].

In quasi-infinite artificial kagome spin ice, the magnetization reversal is characterised by lines of islands with reversed magnetic moments [29] as shown in figure 6a. These lines of islands with reversed moments can be considered to be mesoscopic approximations of the so-called 'Dirac string', with a monopole-antimonopole pair defining the two ends and the Dirac string giving a record of the monopole motion [29]. The Dirac strings are indicated in figure 6b by a continuous line connecting a red-blue monopole–antimonopole pair corresponding to vertices with ΔQ = +2q and −2q. In order to confirm the presence of monopoles, one can consider the coarse grained charge density, ρ_m, [64] which, as can be seen in figure 6b, is indeed consistent with the position of the charge defects ΔQ/q = ±2. Interestingly, the Dirac strings in this two dimensional system are one dimensional [29], which is a signature of 'dimensional reduction due to frustration' [70], and is a consequence of the fact that the strings do not branch which, along with the fact that they do not propagate around corners, can be understood by considering the local dipolar fields [71]. The direction of Dirac string propagation can be controlled, for example, by local chirality of the spins, given by the local curling of the spins at the ends of the islands interacting at a vertex [72] and the chirality of the moving domain wall [73] in a connected system. In the end, the prevailing propagation mechanisms will depend on the orientation of the applied field and the detailed geometry of the system. For example, the extent of the strings is dependent on the amount of disorder given by the distribution in switching fields of the islands. A system with high disorder is characterized by short avalanches, whereas low disorder results in avalanches whose length approaches the sample size [71, 74].

The displacement of charge defects leaving lines of islands with reversed moments can also be observed in square ice arrays as the motion of Type III vertices (see figure 3a) [20, 75], but for them to be 'freely moving', the relative heights of the sublattices should be modified [1, 76]. Indeed, as the positive and negative charges separate in the artificial square ice systems fabricated to date, the relevant energy term does not always behave as expected for a Coulomb interaction [76]. Therefore, in order to confirm real quasipartile behaviour, one would have to demonstrate appropriate quasipartile interactions, i.e. that oppositely charged monopoles attract and like monopoles repel. A first hint of such interactions has been observed during magnetization reversal [77] and also as a preponderance of closed looped configurations minimising the number of monopole-antimonopole pairs in thermally annealed samples [78]. In addition, it is appealing to think of employing such quasiparticles in devices that make use of *magnetic charge* transport without an associated electric charge current. Such devices would operate on 'spin currents' in which angular momentum is used to transport information, with a minimum of ohmic losses that are associated with electron scattering.

2.4 Thermal behaviour: novel phases and dynamics

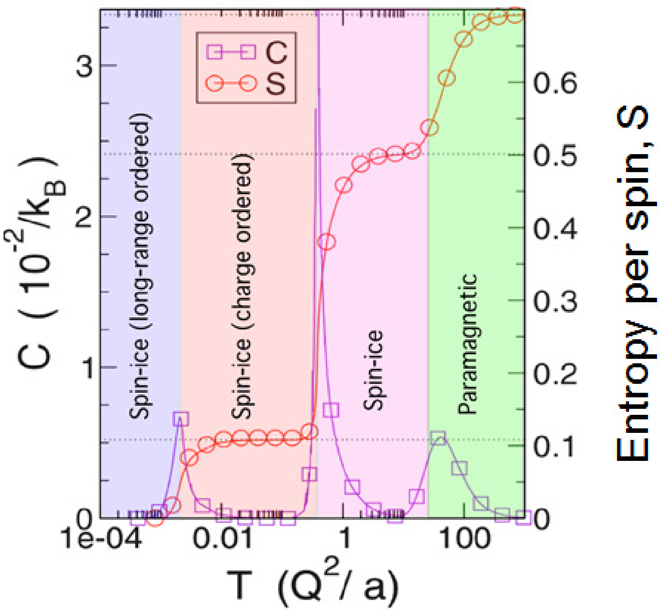

Figure 7: Theoretical phase diagram for an artificial kagome spin ice system. Both the changes in entropy per spin, S, and associated specific heat, C, are shown. Reprinted with permission from [26]. Copyright 2009 by the American Physical Society.

The first publications that addressed the possibilities of artificial spin ice in a thermal regime were theoretical, demonstrating that when considering the long range dipolar interactions in an equivalent spin system there are a number of transitions on reducing the temperature [26, 27]. The transitions are observed as distinct peaks in the heat capacity, C, with the entropy, S, having broad plateaus for the different phases (figure 7). These transitions bring about changes in the vertex charge order, the spin ordering or both. At high temperature the system resembles a paramagnetic phase with neither charge nor spin order, and on cooling there is first a transition to a phase with a gas of ±1q charges, followed by a charge ordered phase with +1q, -1q charges on alternating vertices. While the charge order at this temperature is associated with a large number of spin configurations, on decreasing the temperature further it was found that the lowest temperature phase consists of a configuration with both spin and charge order as shown in figure 2d.

Along with more recent work [79], these theoretical results have provided insight into what one might expect to observe in a real thermal system with fluctuating magnetic moments. In terms of experimental work, a first indication that thermal annealing will provide the long sought after route to the low energy states was uncovered in an as-grown artificial square ice consisting of permalloy islands [78]. The appearance of the ground state in this sample (see figure 8a) was linked to its formation in the first stages of film growth when the layer was thin enough (sub-1 nm) to support thermal fluctuations. As the layer became thicker, the moments froze into the ground state consisting of vortices with head-to-tail moments of alternating chirality as shown in figure 2c, with boundaries separating domains of opposite chirality. Local excitations consisting of clusters of islands with moments flipped compared to the background were also present and the frequency of these excitations decreased exponentially with their excitation energy above the ground state (figure 8b). This was found to follow a Boltzmann distribution, so providing a signature for thermal excitations and therefore 'true thermodynamics'. This experimental work was followed by a theoretical treatment that provided a phase diagram of the ordering as a function of thickness and lattice constant [80]. Here it was demonstrated that the lowest energy state can be reached for sufficiently high dipolar coupling (low lattice constant) and suitably high temperatures. More recently it has been demonstrated experimentally that thermally induced ordered states can be readily obtained in artificial spin-ice systems by heating the sample close to the material Curie temperature and then freezing them in by cooling back to room temperature [81].

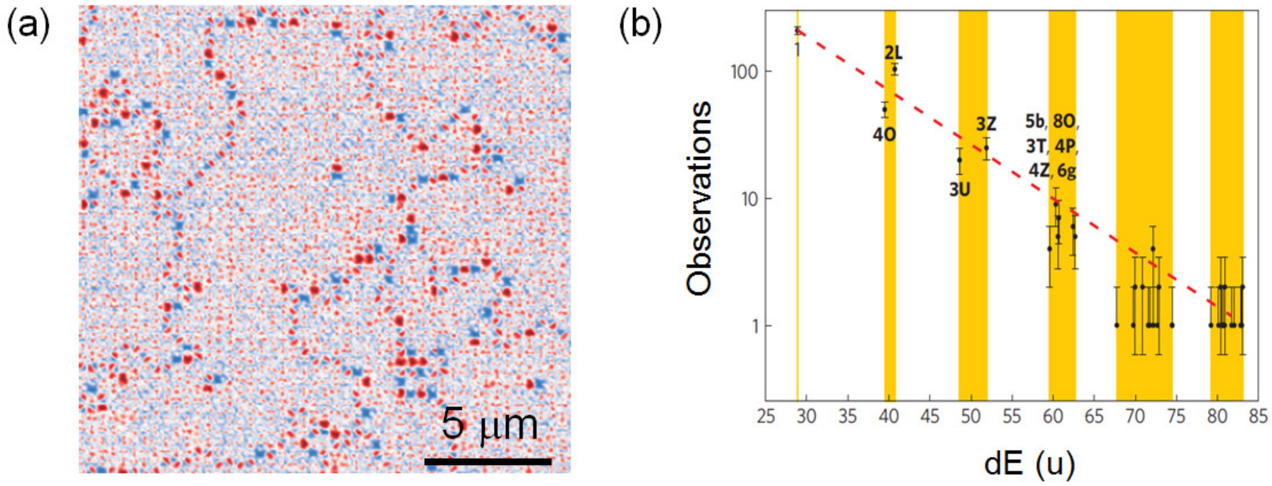

Figure 8: First indications of thermal behaviour in artificial spin ice. (a) Magnetic force microscopy image of long range ordering in the artificial square ice with Permalloy islands (thickness 26 nm, length 280 nm and width 85 nm, and lattice parameter a√2 = 400 nm) with quasi-domains of opposite chirality separated by boundaries and local excitations consisting of small groups of moments flipped against the low energy background. (b) Abundance of square ice excitations plotted against the energy of observed excitations with respect to the background. The fact that the frequency of excitations decreases exponentially with the excitation energy indicates a Boltzmann distribution that is a signature for thermal excitations. Adapted by permission from Macmillan Publishers Ltd: Nature Physics [78], copyright 2011.

While the artificial spin ice display many features that approximate those discovered in microscopic spin ice, they are especially interesting when viewed from the perspective of novel magnetic materials. In this regard, they can be understood as an artificial two dimensional multi-sublattice magnet. In particular, as discussed above, the square ice supports two degenerate and incompatible ground states. This leads to the possibility to have quasi-domains of ground state ordering separated by boundaries. When one leaves the athermal regime, domain growth should occur spontaneously at temperatures large enough to cause individual elements to fluctuate and this has been demonstrated in numerical simulations [82] (see figure 9). The boundaries between the domains contain Type II and III vertices, and the wall modifications have been observed to occur by Type III vertex nucleation and propagation. Since the domain boundaries are associated with a higher energy than the Type I background, the system tries to minimize their length, so causing the small domains to shrink at the expense of the larger ones until they eventually disappear.

Finally, indications of real thermal dynamics have been observed in ultrathin layers of iron sandwiched between palladium layers [83, 84]. For these monolayer systems, temperature-dependent magnetization measurements indicated that on increasing the temperature, a decrease in the macro-spin order or a 'pre-melting' occurred at a temperature well below the intrinsic ordering temperature of the island material [84]. In addition, the recent imaging of thermally active artificial spin ice manufactured from permalloy films only a few nm's thick [85], opens the way to completely new directions of study, allowing the direct observation of configurational dynamics and thermal relaxation, and providing a controlled route to the lowest-energy state.

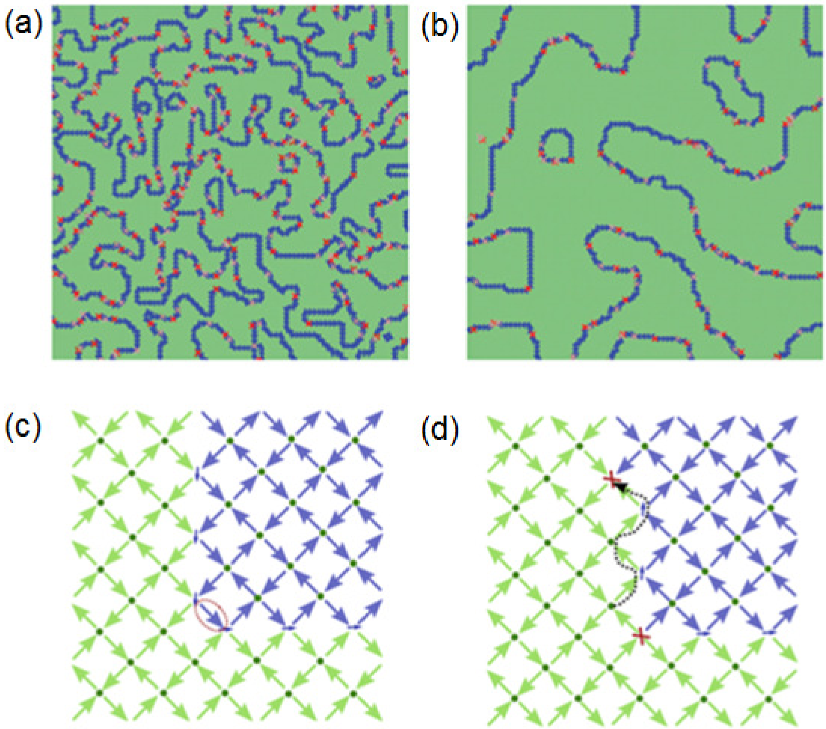

Figure 9: (c) Different stages in the evolution of artificial square ice domain boundaries calculated using Monte Carlo simulations. The green regions are domains containing Type I vertices and the domain boundaries consist of Type II vertices (blue arrows) and Type III vertices (red crosses). Small clusters and domains disappear as the large domains grow. (d) Simulations demonstrate that the domain wall modifications are driven by Type III vertex motion, so that domains shrink via Type III vertex creation and propagation. Reprinted with permission from [82]. Copyright 2012 IOP Publishing Ltd.

2.5 Control of artificial spin ice with a view to devices

If we are to use such frustrated arrangements of dipolar coupled magnets for applications such as logic devices [86], then a first important step is to be able to control their behaviour. One way to do this is to modify individual islands, for example making them narrower (wider) than the rest of the islands in the array so that the shape anisotropy, and therefore the island switching field, is higher (lower). In this way, one can determine in a controlled manner where Dirac string avalanches in quasi-infinite artificial spin ice arrays start and where they stop (see supplementary material of ref. [29] and Ref. [71]). In small clusters, one can use this to control the field induced states and vortex chirality [87].

Another intriguing approach is to ask whether it is possible to create a specified configuration of magnetic moments with a sequence of applied magnetic fields. In particular, a field oriented along certain directions can drive specific element reversals and avalanches, where the reversal details depend upon the magnitude of the field and disorder in the system. An analysis of this concept was made in simulations whereby specific configurations were tracked for sets of possible field sequences [88]. It was found that a sequence of such fields can access a small subset of states, but not all states. However it is possible to increase the number of states accessible to a finite sequence of fields by controlling a small number of element orientations. An example is shown in figure 10.

In figure 10a, the four configurations in a sixteen spin square array that can be accessed for a certain applied field strength are shown. The starting point is a fully polarised lattice and for this particular field strength there are only two configurations available to evolve into. Which configuration appears depends on the direction of the applied field. One additional configuration is possible from each state and arrived at by applying the field in a new direction. The exact configurations are shown in figure 10b, where the top and bottom pairs of configurations in figure 10a correspond to the two possible evolutions. Controlling one of the corner spins, indicated by the circular frame in figure 10a, allows one to access many more configurations, as shown in figure 10c, where now one is able to change 128 different configurations through alignments of the control spin and different orientations of the applied field.

Finally, at the sizes investigated today, quantum tunnelling between magnetic states is not feasible, but if the nanomagnets could be scaled down towards the size of single-molecule magnets, one could start to consider the manipulation of spin waves and the generation of novel quantum states [89].

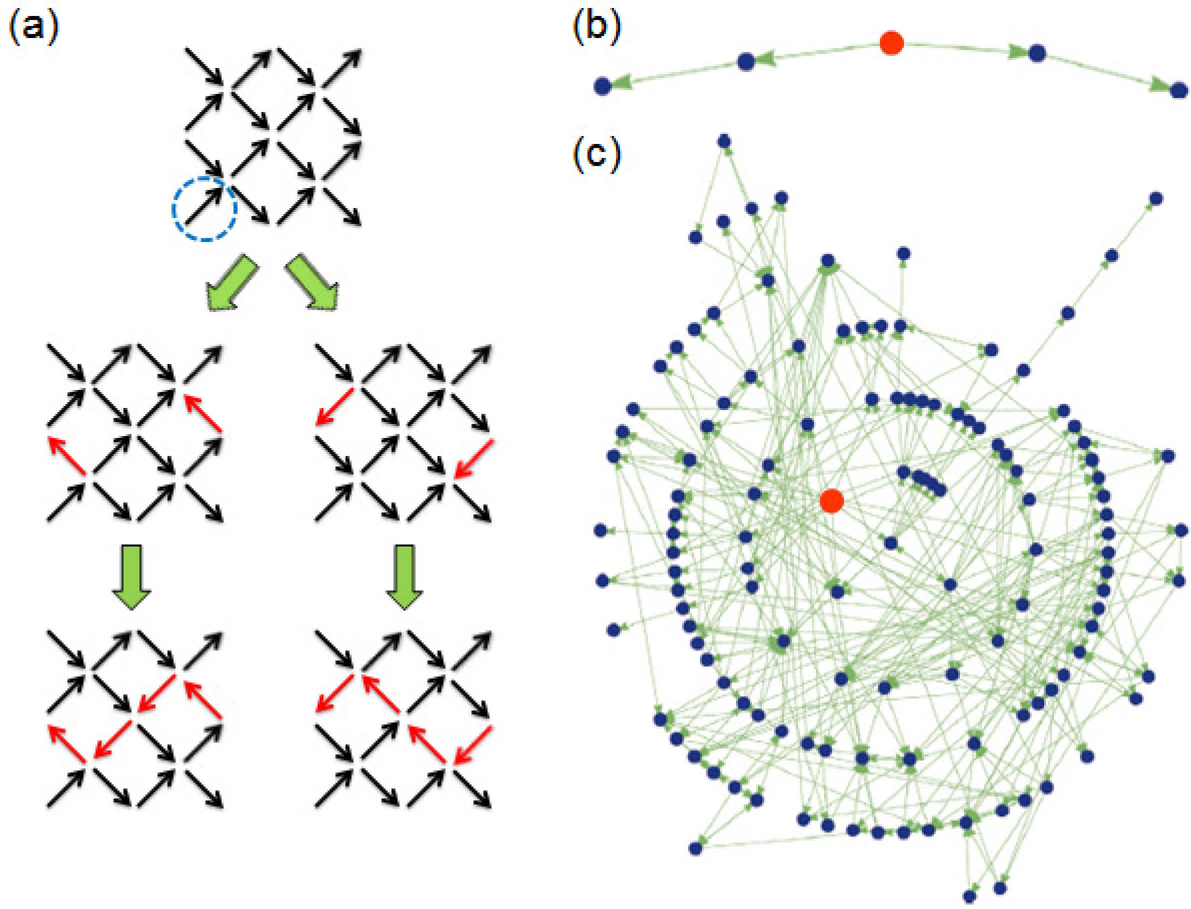

Figure 10: Effect of a control spin on allowed configurations of a sixteen element array. The reference configuration for the array is with all spins pointing to the right, i.e. the black arrows. Without a control spin, a field strength of h = 11.5 is large enough to access only four configurations. These configurations are shown in (a), begin with reversals of edge element spins, and are accessed by applying the field along specific directions. The corresponding map of allowed configurations is shown in (b) where each dot represents a unique configuration of element spins, and each line represents an orientation of the field (with fixed magnitude h). To illustrate the effect of including a control spin, we specify the orientation of an element spin at the lower-left corner of the array [indicated with a circular frame in (a)] in addition to, and independently of, the applied field. This allows in principle two different flipping processes for each field orientation corresponding to the two different orientations of the control spin. The cumulative effect of these different flipping processes is that a multitude of configurations can be accessed depending on the orientation of the control spin and the direction the field is applied, even though its magnitude is still h. In (c) the corresponding configurations are shown. Reprinted with permission from [88]. Copyright 2012 IOP Publishing Ltd.

3. Tunable Magnonic Crystals

3.1 Dynamics at shorter timescales

Up to this point we have considered relatively slow processes in artificial spin ice, occurring over times on the order of 10 ns or longer. These time scales are long enough to allow configurations to change in a quasi-adiabatic manner, as is the case for domain wall motion in continuous ferromagnets. While we have seen that it is possible to access a number of magnetic configurations, we have discussed dynamics only with regards to how the interacting system of magnetic elements can relax into a static arrangement. We now consider the response to magnetic fields applied at significantly higher frequencies than the fields used to determine static configurations. The timescale for the high frequency dynamics is set by the characteristic frequencies for spinwave excitations that can exist within the magnetic materials. For ferromagnets with small anisotropies, these are on the order a few GHz and higher, so that the relevant timescales are on the order of 100 ps and shorter. We will see that these excitations are strongly affected by the underlying magnetic configurations, and change when the configuration changes. This leads to a new functionality for arrays of interacting magnetic elements, namely reconfigurable microwave circuits as described in Section 3.3.

The *slow dynamics* is a result of the rate at which a system can change from one local minimum to another, and is for this reason strongly linked to the rate at which energy leaves the magnetic system as heat. In the example sketched in figure 11, such a process corresponds to a transition over an energy barrier. If this process is driven thermally it is then stochastic by nature. The probability of the transition depends upon the height of the barrier and the magnitude of fluctuations. In a real material, the energies of the fluctuations are determined by the local curvature at the stable and the unstable extrema of the potential landscape associated with the magnetic configurations.

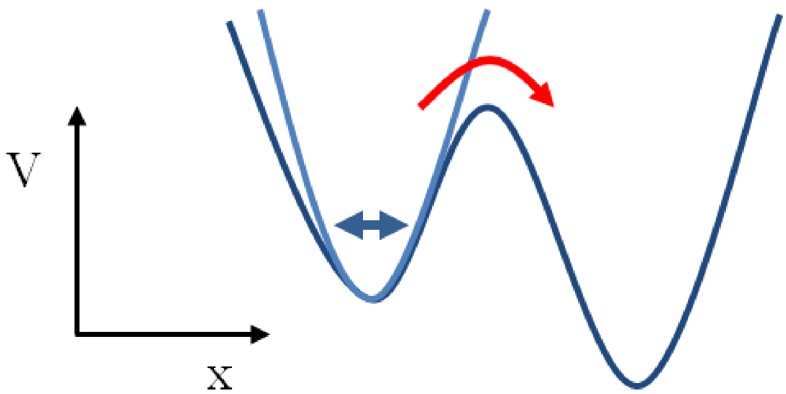

Figure 11: Different time scales for a double well potential describing a two state problem. Slow dynamics corresponds to processes usually driven by field or thermally, that take the system from one equilibrium point to another. This is illustrated in the two potential wells described by the potential energy V(x), where thermal fluctuations can take the system across the barrier from the metastable state into the stable state over the saddle point as indicated by the red arrow. Fast dynamics are associated with fluctuations about equilibria, such as the oscillation indicated by the blue arrow on the left. The frequencies of these fluctuations depend on the local curvature of V(x).

In this part of the review, we are particularly concerned with the resonant dynamics associated with small amplitude magnetic excitations of the system, and such *fast dynamics* can also be understood by considering figure 11. While the potential well minima are defined by the static magnetic configurations, the excitations correspond to small fluctuations of the spin system away from the static equilibrium, as indicated by the blue arrow in the left-hand well, and have frequencies that depend upon the curvature of the potential well. These represent harmonic fluctuations in the magnetization density that are analogous to elastic waves in solids. It is important to understand that the shape of the wells and the position of the barriers for individual elements are controlled by the magnetic properties of the material. In a strongly interacting array of elements, the interactions will also modify the effective potential landscape. Changes in the shape of the landscape of the potential energy occur on timescales determined by the dissipation dominated *slow dynamics*, and the path through the energy landscape can be chosen by the strategies discussed earlier in which magnetic fields are applied in certain directions, and certain key elements are controlled in specific ways. The dynamics involved in the fast response certainly play a role in the slow, stochastic processes but, because spinwave dynamics usually involve small amplitude fluctuations at GHz frequencies, the slow dynamics generally occur over timescales one or two orders of magnitude larger, at least for moderate temperatures relative to the barrier height.

In what follows we focus on processes associated with fluctuations, which occur on time scales at least an order of magnitude shorter than the slow relaxational dynamics. As noted earlier, these *fast spinwave dynamics* occur at GHz frequencies and above, and the corresponding excitations in magnetic materials are very sensitive to the geometrical structure of the material. For this reason, arrays of magnetic elements in which structure controls the spectrum of allowed excitations are called *magnonic crystals* and have analogies with photonic crystals in which the structure affects the optical properties. In magnonic crystals, magnetic excitations are typically at microwave frequencies in the materials used to date in artificial spin ice such as NiFe alloys. However, applications of magnetic materials for microwave frequencies have traditionally involved low loss materials such as the ferrites and garnets. The wavelengths of low GHz frequency spinwaves in these insulator materials are typically on the order of 1-10 μm. These materials are ferrimagnets with small net magnetic moments, and higher frequency operation in moderate applied magnetic fields is difficult to realise. These factors, together with challenges in micro- and nano-fabrication for these materials, limit possibilities for scaling into microcircuit implementation. Instead, we discuss some aspects of metal-based magnetic nanostructures, as used for artificial spin ice, which have much larger magnetic moments and support excitations in the 10-100 GHz range at μm wavelengths (and shorter) for small applied magnetic fields. These wavelengths are orders of magnitude smaller than those possible in air, and the relative ease for patterning in μm and sub-μm geometries make the metallic magnetic systems of high interest for magnonics because of the possibility construct a new class of μm-sized radio frequency components. In the next section we discuss high frequency spinwave excitations, including their special properties when studied in patterned metallic structures.

3.2 Spin waves in element arrays

As mentioned above, spin waves are dynamic magnetic excitations with energies above the static ground state configuration of the magnetization. Their study in magnetic materials patterned into μm and sub-μm sized elements is a relatively new and rapidly expanding area. Rather than providing a detailed review here, we note that excellent summaries of recent developments can be found in the references [37, 90-93], and we refer the reader to the references therein for relevant original papers that have appeared in the vast history of the topic. We concentrate here on the key concepts of spinwaves in patterned films, and provide some examples illustrating progress and directions for future developments for magnonics that make use of arrays of patterned magnetic elements in which the magnetic configuration of an array of elements can be modified.

We consider first the general problem of spinwaves through ferromagnetic media. Spinwave energies and properties can be deduced readily from suitable equations of motion. Towards this end, it is useful to consider a phenomenology commonly used to describe magnetic spin systems, and the corresponding equations of motion that have been successfully applied to model the dynamics. The relevant energies are the local magnetic anisotropy, and exchange and dipolar energies, and can be described in terms of an effective field $\vec{H}_{eff}$ acting on a magnetic moment $\vec{m}$.

Equations of motion for magnetization at a position r contain a precessional torque term and a term that describes energy loss. An often used form is called the Landau-Lifshitz-Gilbert equation:

$$\frac{\partial M}{\partial t} = -\gamma M \times H_{eff}(r) + \frac{\alpha}{M_S} M \times \frac{\partial M}{\partial t}, \qquad (1)$$

where H_{eff} is the sum of all local fields (including external applied magnetic fields), $\gamma = g\mu_B / \hbar$ is the gyromagnetic ratio, and α is a damping constant. The effective field contains also contributions associated with magnetocrystalline anisotropy and magnetic exchange. The first term on the right of equation 1 describes precessional motion of the magnetisation and the second term is a form often used for dissipative relaxation. These equations underpin most micromagnetic simulations designed to predict and explain dynamics in magnetic materials, single elements and arrays of dipolar coupled elements.

It is important to note that the equations of motion naturally lead to nonlinear dynamics and that, in general, linear excitations exist only for small amplitude oscillations. For example, suppose that the equilibrium configuration of magnetization M_S in a ferromagnetic is uniform along the z direction. Decomposing the magnetization into dynamic and static components, we can write $\vec{m}(x,t) = \vec{s}(x,t) + \hat{z}M_S$, where $s(x,t)$ is a small fluctuating component transverse to the static component. Small amplitude precessions mean that $|\vec{s}| << M_S$. The torque equations can then be solved for a magnetic resonance in an external applied magnetic field $\hat{z}H_0$, which corresponds to precessional motion, described as transverse components of s that oscillate in time. For example, if the static component is along the z direction then the transverse components obey $(s_x\hat{x} + is_y\hat{y})\exp(-i(\omega t))$, and the resonance condition is given by

$$\omega_o = \sqrt{H_o(H_o + 4\pi M_S)}, \qquad (2)$$

where H_o is the magnitude of the applied field. This is a linear response, and exists only for small amplitude excitations. Large amplitude excitations, where $|\vec{s}| << M_S$ does not hold, can be nonlinear.

Continuing with the small amplitude limit, there are travelling wave solutions that exist with amplitudes of the form $(s_x\hat{x} + is_y\hat{y})\exp(i(\vec{k}\cdot\vec{r} - \omega t))$ whose energies involve additional contributions from dipolar and exchange interactions. In the long wavelength limit, the dipolar interaction can be dominant, and lead to a variety of interesting features. For example, propagation along the direction of the magnetization can have a negative group velocity, and such excitations are called ‘backward travelling’ waves since their group velocity is negative. Propagation perpendicular to the magnetization includes the possibility of surface localised waves for suitable geometries. These modes propagate in only a limited range of directions with respect to the magnetization, and are used in insulating magnetic materials for circulators and mode isolators. Surface localised excitations are particularly relevant for the arrays of magnetic elements considered here, as will be seen later.

In addition to allowing for localised excitations, boundaries at film surfaces and element edges place conditions on the allowed wavelengths of travelling spinwaves. For example, a wave travelling along the normal direction relative to the film plane in a thin film will be reflected at the surfaces. This results in counter-propagating waves along the direction normal, which constructively interfere only for the set of wavelengths which ‘fit’ inside the film. Perfect pinning at a surface places a node of the spinwave at each surface, so that the allowed wavelengths are multiples of the thickness.

In this way surfaces determine and ‘quantize’ the allowed wavevectors for travelling spinwaves. From this point of view, small magnetic elements can be thought of as waveguides for spinwaves which permit only wavelengths satisfying certain conditions [93-95]. Collective excitations in arrays of elements are formed from excitations with large fields that stray outside individual elements. As explained later, such modes are typically surface and edge localized resonances associated with individual elements since these excitations can have significant dipolar fields at distances away from the element [96, 97].

From the above considerations, we see that spinwaves in individual magnetic nanoelements are reflected by the boundaries and appear as standing waves or resonances, plus some modes localised to the edges. When μm-sized soft iron elements are within 100 nm of one another, they are close enough to interact significantly through dipolar interactions. In this case, the edge localised modes can hybridise through the interaction, and create bands for collective spinwaves. In terms of patterned thin films, standing and collective spinwaves have been observed in dots and arrays of wires with individual element sizes and spacings in the sub-μm regime [37, 90-94, 98-105]. Experiments have been made to explore how spinwaves propagate when the wires are deformed [106, 107] and how spinwaves travel along bends [108]. Another comprehensive study of spinwaves in wires was made using Brillouin light scattering in longitudinally magnetized array of magnetic stripes of rectangular cross section and is reported in Ref. [109]. There it was shown that the distance between adjacent elements controls the strength of interaction due to dynamic dipole coupling between nearest neighbours, and evidence of the band structure formation was found for sufficiently closely spaced stripes.

A distinctive feature for spinwave excitations is that at microwave frequencies the propagation is so slow that the waves are magnetostatic, without an oscillating electric field component, and have wavelengths that can be diffracted by metallic magnetic elements with sub-micrometre dimensions. As discussed above, oscillations of the magnetization can create evanescent magnetic fields that couple neighbouring elements [110, 111]. One can understand the formation of spinwave bands on bringing individual elements close together in terms of overlap of the dynamic fields from neighbouring elements. As noted earlier, bands of collective excitations can exist for strongly interacting elements.

The idea of collective mode formation in arrays of coupled metallic magnetic elements is illustrated conceptually in figure 12. The excitations in magnetic materials are complex in detail, but do display some general features that can be captured simply by thinking of oscillation amplitudes in confined geometries. The sketches for the two elements in figure 12a and b illustrate some allowed excitations characterised by their amplitude profiles. In these sketches, one can associate the amplitude profiles with a single component of the precessing vector magnetisation. An example of an edge localized profile is shown in the element in figure 12b and is characterised by having its largest amplitude at the edges. The large amplitude at the edge corresponds to a large dipolar field for a spinwave, and this field can extend well outside the element. Having a large dynamic stray magnetic field, this mode can hybridise with similar modes on neighbouring elements when the elements are spaced sufficiently close together. An example of overlap is shown in the three elements in figure 12c. The dotted lines represent schematically the dipolar field amplitude associated with the edge localised modes, and overlaps correlate magnetizations oscillating in separate elements. There can be a phase difference between the fluctuations in the neighbouring elements that corresponds to a collective mode with a wavelength determined by the array geometry rather than the element size. In this way a band of collective excitation energies can form in much the same way that the overlap of electron wavefunctions creates a band of electronic states in a crystalline structure. This concept of collective modes is a general phenomenon that has been realised in superlattices and multilayers for elastic waves, electronic states, magnetic spinwaves, and optical waves.

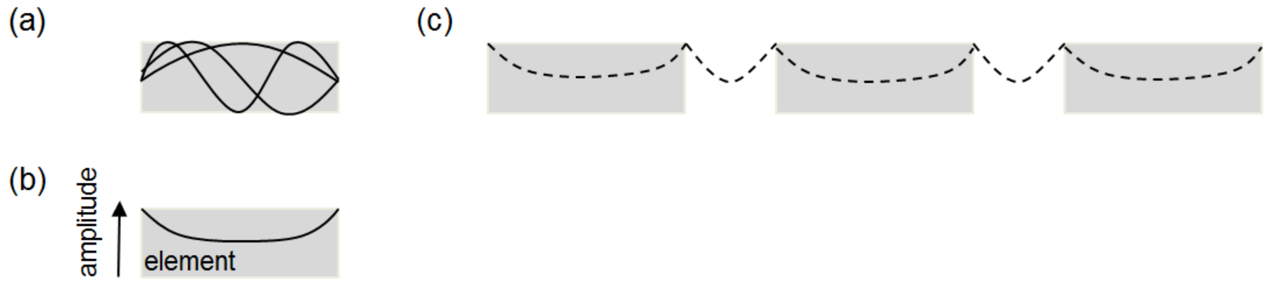

Figure 12: Conceptual sketch of mode overlap between elements in an array of ferromagnetic elements. Some possible standing waves, allowed in the magnetic 'waveguides' are shown in (a). The profile of an edge localized excitation is shown in (b). The solid lines are profiles corresponding to a transverse component of the precessing magnetization. The standing modes have nodes near the edges, but the edge localized modes have large amplitudes near the edges. Because the precession generates oscillating magnetic fields, the fields associated with the edge localized modes can extent significant distances outside the elements. These fields can serve to correlate precessions between neighbouring elements, and thereby create 'collective' excitations. An example is shown in (c). The associated magnetic fields (shown as dashed lines) extend outside the three elements to significant distances, and overlap between neighbouring elements. Wide bands of collective excitations can exist with bandwidths determined by the degree and strength of overlap.

3.3 Spinwave bands in reprogrammable crystals: artificial spin ice

The dependence of the microwave frequency properties on the magnetic configuration present provides an example of new functionalities derived from interacting arrays of magnetic particles. The artificial spin ice systems discussed earlier exhibit complex configurations that can be modified and controlled using temperature and external applied fields. This suggests that they may also be interesting for their emergent microwave properties in that new spinwave bands will arise through the interactions coupling the elements.

The appearance of frequency bands in which collective spinwaves exist is possible through dipolar fields emanating from edge localized spinwave modes, as discussed above in relation to the example depicted in figure 12. We also noted that individual elements display a variety of standing spinwave modes and can display more complex behaviour in realistic geometries. We now present some results from micromagnetic modelling, and show that some standing waves can display localisation features, and thereby have substantial stray fields extending outside the element. We show in figure 13 an example of such a standing spinwave mode for an isolated magnetic element with a geometry similar to that used in an athermal artificial spin ice. The saturation magnetisation in this element is aligned along the long axis, and in figure 13a, a contour map of a single spinwave mode is shown, calculated using micromagnetic simulation tools. Large amplitude precession is indicated with red and yellow contrasts, while small amplitude precessions are in blue and black. While several modes exist at low frequencies (in the 1-10 GHz range), this particular mode illustrates a standing wave mostly confined to the centre of the element. Nevertheless there is an appreciable amplitude near the edges, with correspondingly large stray dipolar fields that can overlap with similar fields from neighbouring elements.

In figure 13b band formation is shown as elements in Type I and Type II vertex configurations are brought close together in the manner sketched in the left side of the figure. The spectra associated with an array of vertices with infinite spacing and with close spacing between vertices are shown on the right. When far apart, the modes from each vertex are degenerate, and form narrow peaks. The two dominant peaks correspond to the lowest frequency, largest wavelength, standing modes with significant edge localizations. The peaks split as the vertices are brought close together, indicating the formation of a band structure as degeneracies are removed by inter-island interactions.

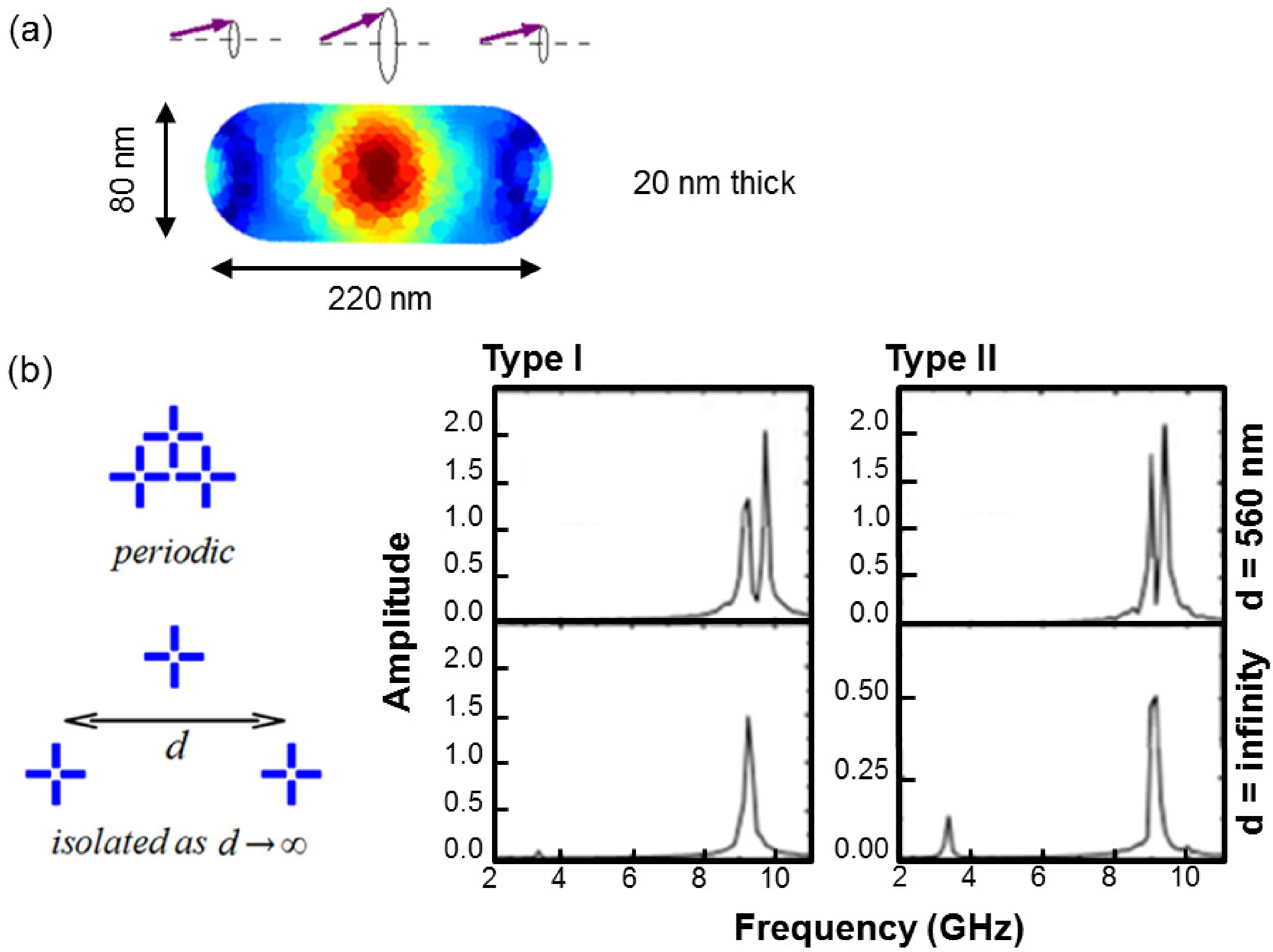

Figure 13: (a) Profile of spinwave mode in a single magnetic element. Blue corresponds to small amplitude precession, and red corresponds to large amplitude precession. Note the increased amplitude at the element edges. (b) Formation of spinwave bands in an artificial spin ice as elements in a Type I configuration (a ground state) or a Type II configuration (saturated) are brought together. Generated using Nmag [112].

It should be noted that very different mode spectra can exist for different vertex types. The corresponding band structures are also quite different, with structure that depends strongly on the detailed magnetic configuration. This feature suggests an interesting possibility for utilising artificial spin ice in magnonics. As we saw, a variety of magnetic configurations in artificial spin ices are possible depending upon how static magnetic fields are applied. Since spinwave spectra depend sensitively on the precise configuration of elements, it will be possible to control details of the microwave frequency response by changing the configuration. Just as there are many configurations possible, there are likewise many possible spectra associated with the configurations. For this reason we can consider the artificial spin ice as a "reconfigurable magnonic crystal". Specifically, we can establish a specific magnetic configuration through a sequence of applied fields and therefore "program" the reconfigurable crystals, so referring to artificial spin ice as *reprogrammable crystals*. It is also worth noting that configurations that consist of multiple vertex types display spectral features that can be associated with contributions from different vertices, in addition to new features arising from interactions between dissimilar vertices. One consequence is that dynamic changes in the magnetic configuration should also appear as transient features in the mode spectra.

Combining artificial spin ice with magnonics, therefore provides a new direction of scientific and technological endeavour, which needs to be explored in more detail. Indeed, recent new developments in the study of the microwave frequency response of artificial spin ice with micromagnetic simulations demonstrates the possibilities for controlling spinwave propagation in such a reconfigurable magnonic crystal [113], and the mode spectra indicate features that may be experimentally observable as local modes. Indeed there will be unique features associated with of possible configurations that affect and control details of the spinwave spectra. This added control and flexibility can be exploited to create a new class of magnonic materials with possible applications to radio frequency communications and information processing technologies.

3.4 Additional magnonic structures

As mentioned earlier, construction of magnonic crystals using magnetic metals is motivated by the short wavelengths (compared to vacuum and insulators) of magnetostatic waves in conductors. This allows for scaling of elements in magnonic crystals down to μm and sub-μm dimensions for device applications. However, there are difficulties with large damping and low group velocities. In order to overcome these issues, antidot arrays, consisting of an array of holes in a magnetic thin film, offer a promising geometry which has been explored by several groups. In these structures the periodic array of holes are used as scattering centres for spinwaves [114-119]. The spinwaves can travel easily through the medium surrounding the holes, but are nevertheless affected by the holes. The effect of periodic patterning is to modify the spinwave mode profiles, creating a band structure in the spinwave manifold that can be controlled by the size and spacing of the holes.

The mechanism for determining the spinwave modes in the antidot arrays involves again dipolar fields. The magnetization at the hole edges consists of uncompensated magnetic poles, and these produce large fields within the hole and, more importantly, between the holes in the material. This results in local magnetic fields between the antidots that can be strongly nonuniform. These nonuniform fields in turn result in local variations of the resonance condition (as given, for example, by equation 2). As a consequence, spinwaves can be channelled along regions where the resonance conditions are matched to the distribution of local magnetic fields.

In this context, it should be noted that the connected artificial spin ice systems [19, 32, 33] are equivalent to antidot arrays, and the various geometries will provide new mechanisms for controlling the propagation of spinwave excitations, in particular with the possibility for creating reprogrammable crystals as described in the previous section.

Advances in lithography in terms of array construction and composition also bring new directions. For example, large scale arrays of bi-component systems have been reported recently, and offer possibilities for additional functionality [120, 121]. In these systems, electron lithography is used to create an array containing two different magnetic metals. One way to do this is to fill the dots in an antidot array with a different material, thus creating a two dimensional composite. Another way is to use lithography and lift-off to construct interwoven arrays of different materials. In each case a complex magnonic band structure is predicted [122]. To date, only a subset of predicted modes has been observed, with some evidence of band gap formation created by scattering from the periodic structure.

3.5 Applications and other phenomena: magnetic logic and nonlinear excitations

A promising direction for magnonic crystals is their implementation for magnetic based logic. This is a large and growing field and, while a complete review is beyond the scope of the present article, we mention a few key ideas in order to illustrate the breadth of potential existing in this area.

Strategies for magnetic logic include the control of domain wall movement and special geometries of strongly coupled magnetic nanoelements [86, 123, 124]. Spin wave based logic gates are also envisaged, utilising spinwave interferometry, and an example of a one-input NOT gate was presented in [125], and is based upon manipulation of spinwave phase [93, 126]. The key challenges in this area include controlling the phase and amplitudes of the spinwaves [125, 127-129], and finding materials and geometries with long spinwave propagation path lengths that are also scalable for use in integrated circuits.

As noted earlier, the equations of motion for spin waves are in general nonlinear, and are only approximately linear for small amplitude excitations. Therefore, another important topic for magnonics is in the general area of nonlinear excitations. Nonlinear phenomena associated with spin wave interactions are well known in terms of dissipation and linewidths, but there are also nonlinear excitations of interest for applications [130-133]. Experimental investigations of nonlinear effects can be made by using spin torque transfer or large amplitude GHz frequency magnetic fields to drive resonance or other magnetic excitations. Nonlinearities appear as the amplitude of the driving field is increased [133].

A low order process that can be affected by confined geometries for high power microwave absorption by ferromagnets is called parametric decay [134]. This process is the decay of a zero wavenumber ($k = 0$) resonance into two oppositely travelling spin waves, each with half the frequency of the parent resonance. This process conserves energy and momentum since the $k = 0$ resonance has zero momentum. The decay is possible for a resonance at frequency ω_o only if travelling states exist at $\omega_o / 2$. Whether these states exist or not depends upon the magnitude of any applied magnetic fields, and also on any confinement quantisations. Most significantly, the existence of states with frequency less than resonance in a ferromagnet require dipolar contributions to the energy, and are possible only for waves with negative group velocity. The key point here is that these waves exist only in a limited range of wavelengths where dipolar energies dominate over exchange energies in the travelling wave. Moreover, they may exist only for a limited range of propagation directions in finite structures due to the existence of surface states and other localized modes. This gives rise to some exotic phenomena, such as analogies to Bose-Einstein condensation [135].

The relevance to our present review is seen by noting that effects due to geometrical constraints can be exploited in patterned arrays. Magnetic element dimensions can be adjusted such that the availability of spinwave states governing nonlinear processes, such as parametric decay, depend on element size. This opens the possibility to, for example, control threshold amplitudes by designing the element dimensions and provides new opportunities for integration into spin electronic devices, with the possibility to detect nonlinearities via an inverse spin Hall effect [136].

3.6 Fast switching and spin wave interactions

The need for high speed operations involving field or current driven high speed magnetic reversal has led to extensive investigations of reversal dynamics of magnetisation in metallic ferromagnets. Metals are typically much more lossy than insulating magnets, due to additional channels for dissipation through interaction of dynamic magnetization with conduction electrons. High speed switching involves a very nonlinear dynamics of the precessing magnetization. Moreover, reversal of magnetization into a new static direction requires dissipation of spin precessional energy, and is thereby strongly affected by nonlinear spinwave interactions that redistribute precessional energy into spinwave excitations.

For an intuitive picture of high frequency dynamics, one can think of the spins as classical angular momenta that precess about their equilibrium direction. Small angle precession corresponds to linear resonance and spinwave propagation, and large angle precession can result in magnetic reversal. Large amplitude precession is of particular interest for achieving the energy efficient switching in magnetoresistive sensors and memory elements.

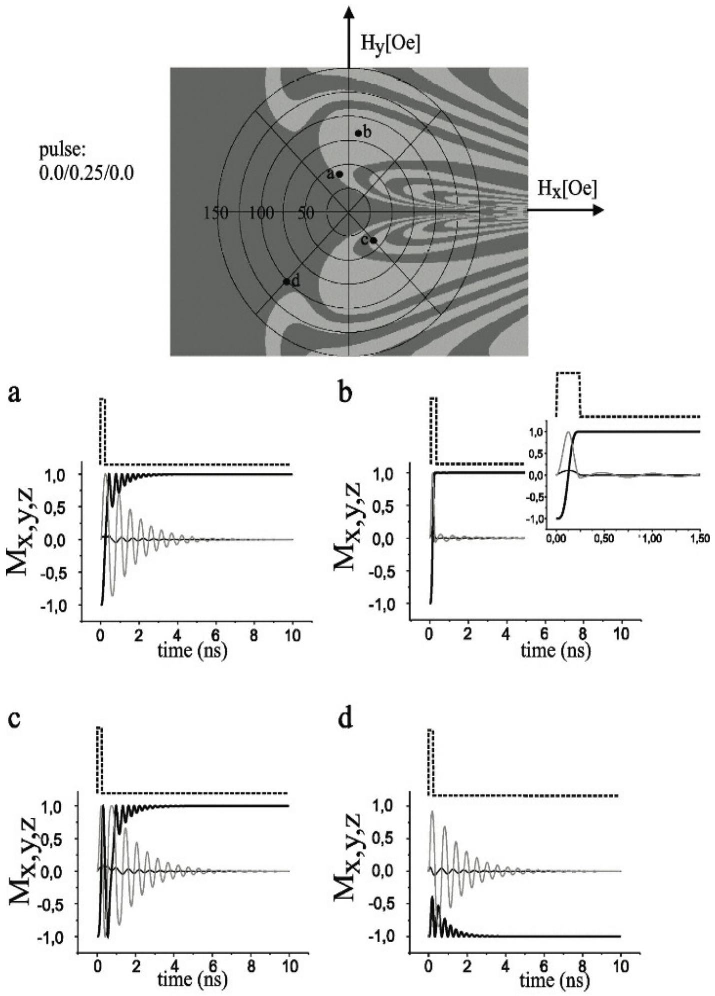

Figure 14: Diagram indicating field orientation and magnitude values for complete reversal of an ellipsoidal particle driven by a magnetic pulse. At the top, a polar graph is shown indicating under what conditions switching of an ellipsoidal particle can be achieved using a pulsed magnetic field. The axes of the graph are the components of the pulsed applied field with the major axis of the particle aligned along the x direction. Light regions indicate complete reversal, and dark regions indicate no reversal. Examples of the magnetization precessions associated with pulse application are shown in (a)-(d) where the magnetization components are plotted as a function of time. The pulse is indicated above each graph. Each example corresponds to a specific pulse orientation as specified by the corresponding point indicated on the top polar graph. These show how a very small pulse width drives precession in all components of the magnetization, and that very small changes in pulse width can lead to very different final configurations. In particular, one sees that switching occurs in (a)-(c) when the z component (darkest line) goes from -1 to +1, but not in (d). The inset shows how a wide pulse can suppress oscillations and lead to a relatively slow switching process. Reprinted with permission from [137]. Copyright 2000 by the American Physical Society.

Dynamics for different geometries of single domain nanomagnets have been studied by many authors [137, 138]. Switching is achieved by application of a short magnetic field pulse at some angle with respect to the initial orientation of the magnetization. The motion is strongly damped as appropriate for precession in a lossy material such as the metallic magnet Permalloy. Precession is highly elliptical and controlled by dipolar fields and confinement effects, and magneto-crystalline anisotropies created by the local atomic environment.

The highly nonlinear dynamics complicate the understanding of which conditions will give magnetic switching. Not all magnitudes or orientations of the applied field lead to switching. An example of a phase diagram for switching in a magnetic element is shown in figure 14 as a polar plot that illustrates this point. The dynamics summarised in figure 14 were determined by integrating the torque equations of motion in Eq. (1) for an ellipsoidal shaped magnetic element exposed to a pulsed magnetic field. The light regions indicate values and orientations of the applied field that lead to switching, and the dark regions indicate values that do not lead to switching. Examples of how the individual magnetization components oscillate in time are shown in figure 14(a)-(d). Each of the examples correspond to one of the labelled points on the polar plot, and illustrate how different orientations of a short pulse field produce different resulting precessions. Some of these precessions create trajectories that terminate with a switched particle, while others do not. The inset in figure 14b shows how a wide pulse can suppress oscillations and result in a comparatively slow reversal of the magnetization.

It should be noted that switching may be affected by excitations during high speed reversal processes in real elements and so do not behave as block spins. Recently it has been shown that there is a pseudo-threshold for high order spin wave interaction processes during fast switching in thin films [139]. This may explain the apparent violation of spin conservation observed at nanosecond time scales. In these experiments, vector magneto-optics was used to track all components of a precessing magnetization driven by a pulsed field, and evidence was presented that the magnitude of the magnetization was not conserved over timescales on the order of a nanosecond. This can be understand if precession of the magnetization created spinwaves that effectively reduce the magnitude of the observed magnetization during the experiment. These results illustrate a very interesting, and technologically relevant, area for exploration: namely, spin wave turbulence.

One of the key challenges here is to achieve exceptionally efficient switching of magnetic elements with minimal energy lost to heat [140-144]. It has been proposed in fact, that magnetic logic based on switching of magnetic elements can approach the so-called Landauer limit [145]. This limit describes processes in which work done on the magnetic system goes directly into changing the information content of a magnetic configuration. The aim here would be to implement reversal processes based on controlled precession, in order to reverse spins with minimal loss to heating through dissipation, driving reversal of elements in logic arrays with carefully constructed magnetic field pulses. One of the key goals for the future is therefore to be able to drive reversal of elements in logic arrays with carefully constructed magnetic field pulses. In particular, simulations for basic logic gate configurations constructed from magnetic element arrays suggest that it may be possible to approach the Landauer limit with suitably constructed pulse sequences and array geometries [145]. Important here is to devise schemes in which certain sequences and configurations of pulsed fields can be used for information storage or transfer with the theoretical minimum energy lost to heat. Experimental proof of minimum energy switching accomplished by applying carefully shaped field pulses, with a minimum of heat loss by the reversing element, would open a door to creating exceptionally efficient logic schemes for low power consumption computation.

4. Functional Ferroic Composites

So far in this review we have illustrated the how new functionalities can appear in systems constructed from dipolar coupled ferromagnetic elements. Introduction of geometrical constraints can create frustration and competition that lead to non-trivial configurational dynamics, which in turn create new resulting magnetic properties. We then illustrated how an associated property, radio frequency response, can be manipulated and controlled as a new derived functionality.

There are also exciting possibilities for new derived properties, which arise when new functionality is introduced with materials combining different interacting ferroic components. These materials could be used in arrays of interacting elements, such as those in artificial spin ice, for example to actively control the magnetic properties of particular elements and for controlling the motion of domain walls [146]. We present below a summary of some of the interesting and unique properties, along with future challenges, and propose here that an exciting direction for future work will be found with the construction of patterned arrays from these new materials. Indeed, it should be possible to create artificial spin ice using elements with both ferroelectric and magnetic components, providing very new and exciting possibilities using electric fields to drive and control configurable properties.

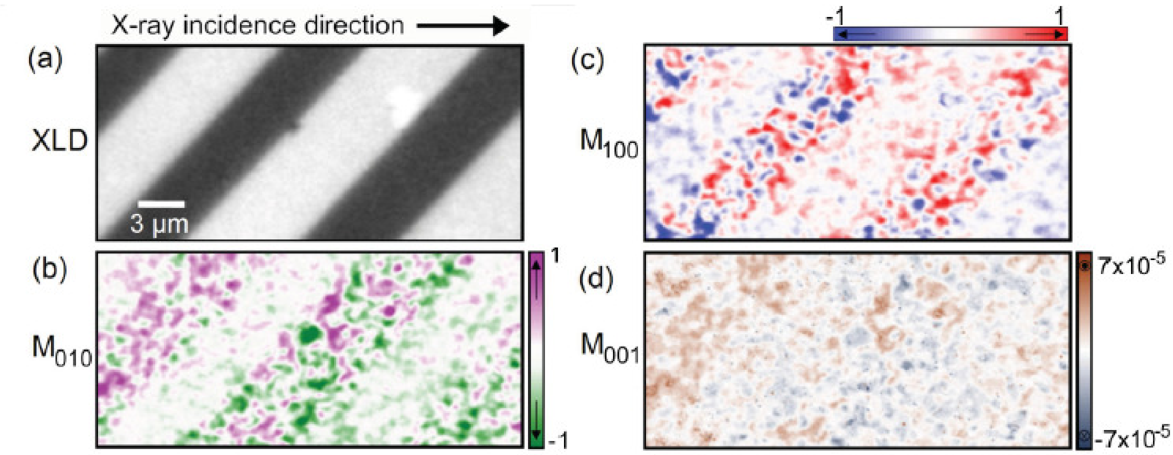

Figure 15: The relationship between the strain domains and magnetic domains in $CoFeO_3$ on a $BaTiO_3$ substrate using photoemission electron microscopy. (a) x-ray linear dichroism (XLD) reveals the strain contrast and (c-d) x-ray magnetic circular dichroism (XMCD) reveals the orientation of the magnetization. Reprinted with permission from [147]. Copyright 2010 by the American Physical Society.

4.1 Materials and Interactions in Multiferroic Composites

Here we specifically address multiferroic composites (or artificial multiferroics) [148-150], where the magnetoelectric effect is generated by combining a magnetostrictive material with a piezoelectric material, either as a thin film stack or particles of one material in a matrix of the other. For device applications, one important aim is to be able to apply a voltage to the piezoelectric material, so inducing a strain that is transferred via elastic coupling to the magnetostrictive material. It is this transferred strain that gives rise to a modification of the magnetic state. Such multiferroic composite materials provide a magnetoelectric response much higher than in single-phase materials and has ordering temperatures suitable for practical applications such as sensors, transducers, filters, oscillators, and spintronic devices for logic and information storage [151-155]. We outline here the key challenges associated with these functional ferroic composites and, if the reader would like more information, there are several excellent detailed reviews of the field [149, 150, 156-162]. We then discuss the important, new direction of high frequency response of functional ferroic composites and heterostructures in the microwave region, and the unique aspects possible for electric field control of dynamic, as well as static, ferroic properties, in particular in artificial spin ice.

The overall properties of multiferroic composites very much depend on their composition (material and thickness) and microstructure. For the ferromagnetic layers, current promising choices of materials include Terfenol-D, $CoFe_2O_4$, and $NiFe_2O_4$, and for the ferroelectric layers $BaTiO_3$ (BTO) and $Pb(Zr,Ti)O_3$, as well as $BiFeO_3$, (BFO), which is the only room temperature single-phase magnetoelectric multiferroic reported to date [163], while a future challenge would be to discover completely new classes of materials with enhanced ferroelectric/magnetic properties. In the end, however, the determining factor for the performance of such hybrid materials is the coupling across the magnetic-piezoelectric interface, which defines how strong the magnetoelectric response will be. Therefore the understanding of the coupling mechanism is crucial in order to create a perfect interface with optimal magnetoelectric response for device applications, particularly at the microscopic level [147]. A technique that can identify the effectiveness of the coupling is photoemission electron microscopy, which can be used to map both the magnetic and strain domains. This has been shown for a $CoFe_2O_4$/ $BaTiO_3$ multiferroic sample with example results presented in figure 15. A stripe contrast corresponding to the modulation in the magnetic anisotropy (figure 15a) was found that is directly correlated with the modulation in the magnetic orientation (figure 15b-d). In addition, since it is the coupling between the components of the system that is critical to the magnetoelectric response, great care should be taken with the conditions under which the materials are grown, since this will determine the interface microstructure and chemistry.

As mentioned above, the magnetoelectric effect is typically generated via elastic coupling between the magnetostrictive and piezoelectric layers. However, additional effects such as exchange coupling/bias, charge mediation and interface roughness may play important roles and it is critical to be able to distinguish between these effects and utilize them to advantage. Indeed, if the piezoelectric is replaced by a multiferroic, one can exploit the exchange interaction between magnetic order in the multiferroic and the ferromagnet. In this case, an electric field can influence the magnetic state of the multiferroic, in turn modifying the ferromagnetic state in the magnetic layer via exchange coupling [164, 165]. In addition, the use of $La_{0.7}Sr_{0.3}MnO_3$ in multiferroic composite devices is of interest because it is half-metallic [166], providing the potential to create electrically controlled fully spin-polarized currents [158].

4.2 Electric field control

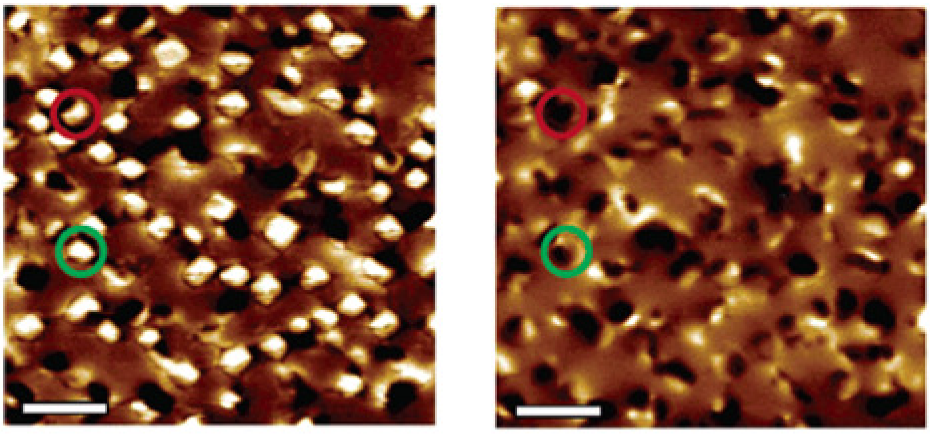

Figure 16: Changes in the magnetic configuration of a $(BiFeO_3)_{0.65}$-$(CoFe_2O_4)_{0.35}$ film upon electrical poling. Magnetic force microscopy image taken (a) after magnetization in an upward oriented 20 kOe perpendicular field, and (b) after electrical poling at +12 V. The scale bars are 1 μm. Reprinted with permission from [167]. Copyright 2005 American Chemical Society.

Of paramount interest to scientists working in the field of multiferroic composites is control of magnetic states with the electric field. While there have been a few demonstrations of electric field modification of the magnetic properties, a reproducible and controlled reversal of magnetic states is still to be shown. Nevertheless, manipulation of magnetic domains in thin film bi-layers such as $Fe_{0.7}Ga_{0.3}$/BTO [168] has been demonstrated, as well as magnetization reversal in $CoFe_2O_4$ pillars in a BFO matrix by electrical poling with a scanned Piezo-Force Microscopy tip [167, 169, 170] as shown in figure 16. Here the film was first magnetized out-of-plane with the $CoFe_2O_4$ columnar structures appearing white in figure 16a. After electrical poling (figure 16b) a large fraction of the magnetic columnar structures fully reversed contrast from white to black (see pillar marked with red), some partly changed colour (see pillar marked with green), and only a few remained unchanged.

In terms of first demonstrations of a switchable device, Chu et al. [171] have shown that the ferromagnetic domain structure of CoFe microstructures on a BFO film can be modified. The modification of the magnetic state in a nickel nanomagnet via the application of an electric field to a PZT film device [172], and reversible electric-field-induced switching between two reversible and orthogonal magnetic easy axes in a Ni/PMN-PT device have also been demonstrated [173]. Now electric field modification of the magnetization in isolated elements has been shown, one key future challenge would be to demonstrate the controlled and reproducible switching of arrays of nanomagnets [174], possibly dipolar coupled as in artificial spin ice, using an electric field.

Spurred on by the possible implementation of multiferroic composites in magnetic tunnel junction (MTJ) devices [175], one important question is the robustness of ferromagnetic and ferroelectric phases at low film thicknesses, since very thin films are required as tunnel barriers. Recently it has been shown that the ferroelectric polarization can be stabilized in ferroelectric film thicknesses of a few unit cells [176-178]. Moreover, it has been predicted that the ferroelectricity in a Co/BTO/Co MTJ can be maintained at BTO thicknesses down to 1.6 nm [179]. Interestingly, in a recent study employing Piezo-Force Microscopy and soft x-ray resonant magnetic scattering [180], it has been demonstrated that both magnetism and polarization occur in ultrathin BTO films coupled to a ferromagnet such as Fe or Co. Therefore it appears that multiferroicity can be generated in a diamagnetic ferroelectric at room temperature, opening the way to the creation of novel multiferroic materials.

4.3 Electromagnons: multiferroic composites at high frequency

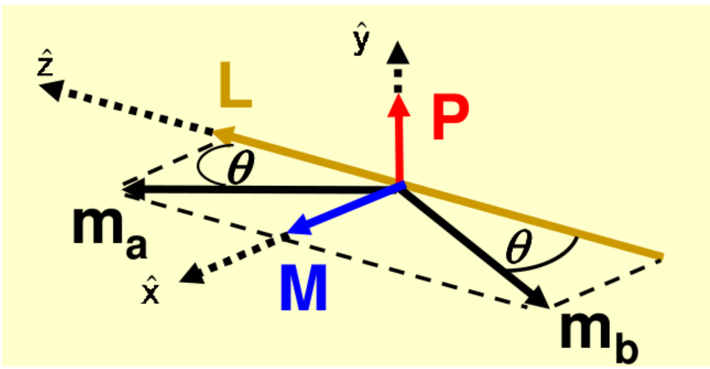

Figure 17. Orientation of electric and magnetic polarizations in a single phase multiferroic material with a canted spin structure such as that expected for $BaMnF_2$. Magnetically there are two sublattices of spins, labeled $\vec{m}_a$ and $\vec{m}_b$. These sub-lattices are canted away from antiparallel order by an angle θ that depends on the strength of magnetoelectric coupling compared to the electric polarization P and magnetic anisotropies. This results in a weak ferromagnetism indicated by $\vec{M}$. Vectors $\vec{L}$ and $\vec{M}$ represent the sum and difference of the two sub-lattice magnetizations.

Linking back to Section 3 of this review, we now turn to excitations in multiferroics. We begin by considering single phase materials, and then discuss composites. Single phase materials displaying simultaneous ordering of magnetic and electric polarizations have been extensively studied for possible electric field control of magnetic properties. At present the most interesting of these materials for applications have at least two magnetic sublattices that are antiferromagnetically coupled, and display weak ferromagnetism through canting of the sublattices. An example of this spin structure is sketched in figure 17 for a weakly coupled two sublattice multiferroic. This material is antiferromagnetic, and can be described as two interpenetrating lattices of spins. Spins within each of these “sub-lattices” are parallel, but spins from different sublattices would be antiparallel except for some canting due to a competing exchange interaction associated with the magnetoelectric energy. The sublattices of spins are indicated as $\vec{m}_a$ and $\vec{m}_b$. The canting corresponds to a weak ferromagnetism and is depicted by the vector $\vec{M}$. It is common also to define a vector $\vec{L}$, which is formed by the difference between $\vec{m}_a$ and $\vec{m}_b$ perpendicular to the electric polarization $\vec{P}$. The utility and interest in such single phase materials derives from the magneto-electric coupling between $\vec{M}$ and $\vec{P}$.

The simultaneous existence of, and coupling between, magnetism and ferroelectricity appears in a number of complex oxides, but its manifestation is subtle and complex, and unfortunately for applications, weak in known room temperature single phase multiferroics. It is now understood that the electronic states responsible for magnetism and ferroelectricity are associated with different atoms in the oxides, and are in some sense incompatible. Magneto-electric coupling in these materials arises primarily through spin orbit effects and is typically weak and strongly dependent upon local site symmetry.

The weak ferromagnetism in a single phase material can support GHz frequency resonances and spin waves [181]. An alternative approach is to consider composites based on ferromagnetic materials which have technological applications in the MHz and GHz regimes. This approach allows tuning of electromagnetic response by combining materials with dissimilar dielectric properties. Such constructions can be used to create "artificial" materials where the resultant electromagnetic properties are greatly enhanced compared to the properties of the constituents. This strategy provides options to create materials which interact in such a way as to allow electric fields to strongly affect magnetic order even when magnetoelectric coupling is not present or too weak to be useful.

We can then link back here to multiferroic composites in which strong interactions between components can drastically modify the properties of magnetic excitations. In particular, an effective form of magneto-electric coupling appears in the spin wave dynamics of multiferroic composites can appear. Consider a heterostructure composed of alternating ferromagnetic and weak ferromagnetic ferroelectric films. The long wavelength, average magnetic and electric response of the heterostructure is determined by Maxwell's electromagnetic wave equations with boundary conditions on the dynamic electric and magnetic fields. Even if exchange coupling is neglected between the ferromagnetic and multiferroic films, one finds that magnetic dipolar fields serve to mediate an effective magneto-electric interaction in the radio frequency response [182].

As a concrete example, using parameters appropriate for $BiFeO_3$ and NiFe, and taking into account interfacial strain, one finds a magnetoelectric excitation at 4.07 GHz for films of equal thickness [182]. This is a shift downward of the bare NiFe resonance by nearly 2 GHz, which is a significant result since the magneto-electric coupling is transferred to the NiFe via dipolar interactions. A magneto-electric spin wave resonance is thereby created, whose frequency is sensitive to the magneto-electric coupling strength and which will be directly affected by electric as well as magnetic fields.

The multiferroic heterostructure provides an interesting analogy to the artificial spin ice magnonics discussed earlier in which magnetic dipole coupling between individual elements provides a means to affect GHz frequency excitations with magnetic fields in new ways. In the multiferroic heterostructure, the dipolar coupling creates a material in which *electric* fields can be used in new ways to affect GHz frequency excitations.

4.4 The future of multiferroic composites

The future of multiferroic composites depends greatly on the ability to transfer the knowledge gained so far into novel device applications. As mentioned above, it is important to find ways to control reproducibly the switching of the magnetic state with an electric field. This is non-trivial, for example in strain driven samples, since an electric field applied to the ferroelectric on its own will not bring about magnetic reversal. Since the strain drives anisotropies, in the elongated elements found in artificial spin ice discussed in the second section of this review, it would be possible to rotate the easy axis direction, but this would result at best in a rotation of the magnetization by 90°, but not achieve 180^{o} reversal. Therefore, either some asymmetry needs to be included, such as an additional magnetic field, or ingenious electric field pulse schemes need to be created, such as those for magnetic field switching described in section 3.6, in order to drive the system reversal. Thinking further along these lines, electrical field controlled magnetic elements could be implemented in logic devices, for example providing a control element as described in section 2.5 where the anisotropy can be actively tailored with an applied electric field. Likewise control elements could be implemented as an aid to programming the reconfigurational magnonic crystals described in section 3.3.

In terms of low and high frequency properties, multiferroic composite systems are interesting in terms of providing static electric and/or magnetic field control. Simple capacitive geometries, for example, can be used to modify the frequencies of microwave resonances due to screening at ferroelectric/ferromagnetic interfaces [183]. The low frequency response to alternating fields, typically operating between 10 mHz and 100 Hz, is also interesting for implementation in magnetic sensors. For example, electric current sensing in the kHz range is possible by detecting the vortex field surrounding a d.c. line current. Also related to this, one can think of using magnetoelectric transformers for high voltage gain.

The multiferroic composite systems represent a new and fascinating class of materials with electromagnetic properties that can be optimized for particular applications through the composition and design of the heterostructure. In order to obtain a high frequency response in the 10 to 100 GHz range, ferrite-piezoelectric ceramic composites are also of interest and need to be further investigated. These also allow for electric field tunable devices, for example with YIG films grown on PMN-PT or some other piezoelectric material, and applications include band-stop and bandpass filters, phase shifters and delay lines [160].

5. Summary and Outlook

The purpose of this review has been to provide a picture of how controllable functionality can emerge in artificially created ferroic materials with mesoscopic structure and interactions between the components. We have provided a comprehensive picture of the relationship between configurational order and emergent dynamic material properties within the context of magnetic arrays, starting with a section on *artificial spin systems,* and then moving onto dynamics and materials in sections on *tunable magnonic crystals* and *functional ferroic composites*. The functionality of these *artificial ferroic systems* ultimately rests upon the material properties of the constituent elements, and also on the ability to fabricate high quality arrays of elements.

In order to have a picture of this concept, one can think about a musical instrument such as a violin. In order to build such a string instrument, one has to first find appropriate materials, for a violin spruce or maple wood for the body and gut, steel or other synthetic materials for the strings. The material is crafted into a suitable geometry to produce the required performance. Critical here is tuning of the strings by slow movements of the bow before the violin can be put to use in a virtuoso performance. In this way, one can create an instrument with an audible frequency response that is an example of controlled functionality emerging from individual elements with quite disparate properties. In order to attain a particular response in the artificial ferroic systems discussed in this review, a similar strategy is followed. We begin with suitable materials and a well-defined geometry, tune the system with the application of slow varying electric and magnetic fields, and achieve the final device performance with microwave excitation.

With this in mind, we then summarised new functionalities for electric field control of magnetic materials possible with composites involving ferroelectric and multiferroic materials. The technology to achieve high density patterning at the nanoscale is most advanced for magnetic metals, but still needs to be developed for multiferroic composites. We believe that the paradigm offered by the magnetic patterned arrays, if expanded to encompass ferroic composites, has a great potential to yield important new and useful phenomena, due to the additional control of the magnetic state with an electric field. This concept of engineering of materials and system properties at the mesoscopic scale is in the process of development for materials with magnetic and/or dielectric polarisations and we have pointed in this review to interesting directions for future developments.

We have illustrated how the patterning of magnetic materials into periodic arrays can lead to magnetic configurations that exhibit a rich and complex dynamics. Mobile magnetic charges appear in strongly interacting arrays, and possibilities may exist to harness them as a novel type of charge flow, or ‘magnetricity’ [184]. Moreover, novel approaches to configuration control may allow a multitude of magnetic states to be addressed. A particularly exciting direction for this topic is to explore the dynamics possible in the presence of thermal fluctuations. First reports of ‘thermal artificial ice’ are now appearing [78, 81, 85], and with them the technology to make nanoscale elements with extremely well defined properties is developing, as well as ideas of how to incorporate nanoscale magnets for computing at thermal equilibrium [140]. Building on the idea of reconfigurable magnetic states, it is important to look to high frequency dynamics that may lead to new types of logic and microwave frequency devices. Such developments are just now being pursued [111, 113, 123-129, 185] and there are extensions into other areas of magneto-electronics and spintronics such as sensors and biomedical applications.

Following the concept of new functionality, an important class of polarisable materials incorporating ferroelectric and magnetic materials has emerged from ferroic systems that respond to electric fields through dielectric polarisation and strain effects. Here the control of interface structure is critical in order to introduce desired electric and magnetic field responses, and continued refinements are required, including: tuning of materials properties, use of new materials and improvement of coupling at the ferroelectric/magnetic material interface. While such composite systems have been studied for some time, this area has great scope for development, particularly with regard to GHz and THz functionality, as it becomes possible to affect and detect high frequency magnetic response with electric fields. Advancement here depends very much on improved control of interfaces, construction of heterostructures, and development of robust lithographic techniques.

Related to this are topics involving charge and spin transport. This is a very large and important area not covered in the present review, and has several opportunities for future exploration of transport effects in dipolar coupled magnetic arrays. A new direction particularly relevant for this review is the possibility to utilise spinwave excitations in new ways, for example in spin caloritronics in which spin currents can be created by thermal gradients. Since spinwaves carry angular momentum, it has been argued that spin-orbit coupling can affect the transport of angular momentum via spin currents across interfaces in some materials. As a consequence, mechanisms originating from spin orbit coupling at interfaces may give rise to spin Seebeck and spin Hall effects, equivalent to their conventional charge transport analogues [186-189]. This is an exciting and very active area of development with potentially broad impact on many aspects of magnetization dynamics in nanostructures and thin films.

Further directions include several associated areas which are growing quickly. In plasmonics and metamaterials, structure is used to exploit and control the interaction of electromagnetic waves with matter. For plasmonics, the characteristic length scales of features are in the millimetre to micrometre range. One important area to be explored is the incorporation of magnetically polarisable elements in a plasmonic array in order to tune different properties such as negative refraction with external applied fields [91, 190-192]. Further directions include laser induced switching [193], coupling light to charge and spin currents for information encoding and transfer, optical trapping and manipulation, nanoplasmonics and imaging, to name just a few, with the potential to generate and steer spinwaves in an all optical set-up [194].

As a result of the nearly boundless possibilities for manipulating element structure and composition with nanoscale precision, the developing horizons for new ideas and approaches are broad and largely unexplored. Many of the most recent ideas are based on analogies to intriguing phenomena studied in very different contexts. Electrical conduction in topological insulators, for example, is a phenomena postulated only a few years ago and the experimental study is an exciting and developing area, particularly with regards to the search for suitable materials and systems The topological phase responsible for this state does not only have to be realised in electronic systems. Analogies have been proposed for electromagnetic waves, i.e. plasmonics, and also for spinwaves in magnetic materials [195, 196], offering numerous opportunities for creating new concepts and devices that can exploit topological protection of propagating excitations.

The complex interplay between structural symmetry, interactions and physical properties, reasonably well understood for materials at the atomic level, is only now being explored for materials defined at the micro- and nanoscale. We are now approaching a regime in which we can harness a wide palette of responses: electric, magnetic, thermal, elastic and electromagnetic, both in quasi-static and dynamic regimes. Here we have outlined a number future directions for scientific endeavour in the field of *artificial ferroic systems*, and it is clear that what began more than a decade ago with nanotechnology and first attempts to understand the slow dynamics of patterned polarisable materials, is now developing into an exciting field that combines materials, structure and interactions at mesoscopic length scales and brings new functionalities at a range of timescales, going from slow dynamics to the high frequency regime.

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