Disorder-independent control of magnetic monopole defect population in artificial spin-ice honeycombs

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Abstract. Breakdown of the ice rule in artificial spin-ice nanostructures results in magnetic monopole defects with zero magnetic moment. Such defects exist during the magnetic switching process in some nanostructures and yet are absent in other apparently similar arrays having the same geometry and made from the same material components. One explanation proposed for this discrepancy is that it is due to the variation of disorder across samples, with monopole defect formation occurring only in highly disordered samples. Although disorder can indeed play a role in the determination of monopole density, in this paper we show, by experiment and simulation, that in samples of similar, low disorder, the factor controlling the nature of magnetic switching is whether the domain walls are in the transverse wall regime or in the vortex wall regime. This work illustrates that monopole formation can be controlled by intrinsic micro-magnetic behaviour as well as by extrinsic quenched disorder.
1. Introduction

Artificial spin-ice materials [1] are arrays of frustrated nanomagnets and have recently been shown to be ideal tuneable systems enabling unparalleled studies of frustration. Two different artificial spin-ice systems have been studied to date and these consist of nanomagnets arranged in a square [1] and a Kagome lattice [2–5]. The magnetization of each bar is single domain and points along the long axis of the bar and thus can be considered an Ising macrospin. Each bar is well described as a dumbbell with a magnetic charge of \( +q \) at its head and \( -q \) at its tail, where \( q \) is the magnetization of the bar divided by its length. Within the array this effectively quantizes the local magnetic charge at each vertex, \( Q = \Sigma q \), into integers of \( q \). The ice rule in spin ices, in general, can be understood as local magnetic charge minimization. In the Kagome artificial spin-ice system, three bars meet at each vertex leading naturally to magnetic frustration with a finite magnetic charge at each vertex [3]. The ice rule local ordering principle favours the set of six possible vertex configurations with \( Q = \pm 1q \), and suppresses the unique ice-rule-violating configurations known as monopole defects that possess \( Q = +3q \) and \( Q = -3q \) and zero dipole moment. The ice-rule-obeying configurations occur when one spin points into a vertex and two spins point out (1-in, 2-out; \( Q = -1q \)) or the opposite (2-in, 1-out; \( Q = +1q \)).

The nanoarrays we consider here are composed of individual bars of length \( l = 1500 \pm 10 \text{ nm} \) and width \( w = 100 \pm 10 \text{ nm} \). These bars are of sufficient dimension and shape anisotropy for magnetic reversal to be mediated by domain wall motion (rather than coherent rotation [6]). Head-to-head domain walls have charge \( Q = +2q \) and tail-to-tail domains walls carry charge \( Q = -2q \).

Recent studies [4, 6–8] have shown that cycling a Kagome artificial spin-ice system through its hysteresis loop leads to the generation of magnetic monopole defects. However, these defects have been absent in equivalent switching experiments of nominally similar structures [2]. When previously we considered connected cobalt [3] honeycombs, we found that the parameters that enter the problem of creation and imaging of monopole defects can be mapped onto a simple model that requires only the ice-rule violation field (\( H_I \)) and the distribution (standard deviation, \( \sigma \)) of bar coercivities. Our simulated monopole defect population matched the experimental observation (\( \sim 1\% \) of vertices) for \( \sigma/H_I = 1 \). The monopole defects observed in this study were formed in an extrinsic process involving a single domain wall and a change in vertex charge of \( \pm 2q \). A simple, purely Coulombic model treating domain walls as circular discs of magnetic charge with diameter \( w \) has subsequently been developed [5, 9, 10], which provides an elegant picture of the physical origin of \( H_I \) and predicts well the occurrence or absence of extrinsic monopole defects [5]. Thus in that model the presence [3, 7] or absence [2, 5] of monopole defects was attributed solely to the disorder of the system [5].
However, this argument ignores a second mechanism of monopole defect formation which, in contrast to the above, is *intrinsic* to the system. We have previously observed monopole formation \[4\] in connected permalloy honeycomb arrays with disorder comparable to those characterized in \[5\]. In these connected structures with thickness 8 and 16 nm, we found that the monopole defect formation occurred by a cooperative two-domain wall process, described as magnetic Coulomb blockade \[4\]. High-resolution scanning transmission x-ray microscopy (STXM) images of these intrinsic monopole defects revealed the richness of the near-field magnetic charge structure at the vertex, which appeared to contain two undistorted transverse domain walls \[4\]. It is well established that in Ni$_{81}$Fe$_{19}$ nanostructures of wire width 100 nm, there is a transition from transverse domain walls to vortex domain walls at a wire thickness of approximately 20 nm \[11–13\]. Furthermore, these two domain wall types carry quite different magnetic charge distributions (as shown in figure 1) and have been shown to respond differently to a variety of pinning potentials \[11\]. It is our hypothesis, which we expand below, that the second mechanism responsible for monopole defect formation relates to the energy scales associated with these different domain wall types.

To date, all experimental studies in which monopole defects were not observed in connected Kagome structures have been carried out on significantly thicker samples (>23 nm) and so are likely to be in the vortex wall regime. In order to study the role of domain wall type systematically, we have prepared artificial spin-ice Kagome structures with low disorder (in terms of edge and surface roughness) and of thicknesses 18 ± 1 and 36 ± 1 nm. We have studied the switching process using high-resolution STXM and we demonstrate unequivocally that monopole defects are readily observed in the transverse wall (18 nm) sample, but are completely absent from the equivalent vortex wall (36 nm) case.
We use micro-magnetic simulations to investigate the pinning potentials in the transverse wall and vortex wall regimes. The striking difference between the two domain wall regimes highlights the limitations of models which treat the domain walls as simple spheres of magnetic charge \[9\]. In fact, we also observe strong differences in the cascade behaviour of the walls as they flow through the lattice. In general, we find that the simple picture of a spherical charge carrier works well in the vortex regime but, in the transverse regime, locally correlated domain wall processes, such as cascade branching and magnetic Coulomb blockade, play a significant role.

2. Method

Samples were fabricated by e-beam lithography on SiN membranes, evaporation of Ni\textsubscript{81}Fe\textsubscript{19} and lift-off. The array had a triangular injection pad on the left-hand side in order to study controlled domain wall injection. The results of this study will be published elsewhere. Atomic force microscopy (AFM) was used to measure the sample thickness and surface roughness. Samples were fabricated at thicknesses of 36 ± 1 nm (sample V) and 18 ± 1 nm (sample T), in order to probe the reversal of systems in the transverse and the vortex domain wall regime, respectively. The rms surface roughness calculated over an area of \((100 \mu \text{m})^2\) is 2.27 nm for sample V and 1.12 nm for sample T. Room-temperature STXM studies were carried out on beamline 11.02 at the Advanced Light Source (Berkeley, CA, USA). The sample was mounted in the STXM chamber between the pole pieces of an electromagnet, which allowed the application of an in-plane field of \(\pm 60 \text{ mT in situ}\). The chamber was pumped down to a pressure of approximately 100 mTorr before filling with He gas. Elliptically polarized x-rays were provided by an undulating beamline, after which they were focused to a spot size of approximately 30 nm using a Fresnel zone plate. The sample and electromagnet were mounted at approximately 30° with respect to the x-ray propagation vector, enabling the in-plane component of the magnetization to be probed, using the x-ray magnetic circular dichroism effect. In order to study the switching of the array, the sample was first saturated in the positive field direction, and small field increments were applied in the negative field direction. Micro-magnetic simulations were performed with the OOMMF \[15\] package with a lateral mesh size of 5 nm. The magnetocrystalline anisotropy of Ni\textsubscript{81}Fe\textsubscript{19} was assumed to be zero, the exchange stiffness was taken to be \(3 \times 10^{-11} \text{ J m}^{-1}\), and the saturation magnetization was taken to be 800 emu cm\(^{-3}\).

3. Results

Figure 2 shows a series of partial hysteresis loops obtained from STXM images during multiple field cycles within sample V (blue line) and sample T (red line). The hysteresis loops were calculated from series of sequential images taken from similar arrays on samples T and V. The same sample is studied in Run A and Run B and so the quenched disorder is unchanged; the small differences between runs for each sample are caused by other types of randomness in the switching process (such as whether the domain wall propagates diagonally up or diagonally down after exiting a horizontal bar). We find a striking difference in the magnetic reversal characteristics of the two arrays. In sample V, there is a sharp onset of switching between the fields of \(-9\) and \(-11\) mT. The rate of switching with respect to field strength then changes and the region from \(-11\) to \(-13\) mT shows very little change in magnetization. A second onset of
Figure 2. Experimental room-temperature hysteresis loops generated from STXM images. Normalized x-component of magnetization versus applied field stepped in the $-x$-direction after saturation in the $+x$-direction for 36 nm sample V in the vortex regime (blue circles) and 18 nm sample T in the transverse regime (red squares). The same sample is studied in run A and run B. The error bars are small compared to the symbols and the lines are a guide to the eyes. The switching is fundamentally different in the two regimes, yet it is stochastic in both.

switching occurs at $-13 \text{ mT}$ and the array is completely switched at $-15 \text{ mT}$. Sample T shows very different characteristics. There is a sharp onset of switching at $-12 \text{ mT}$ and this continues unhindered until, at $-14 \text{ mT}$, the array is saturated. In order to characterize the distribution of bar switching fields, in a comparable way to Daunheimer et al [5], the hysteresis loops were fitted to

$$M(H) = \sum m_i \text{erf} \left( \frac{H - H_c}{\sigma_i \sqrt{2}} \right),$$

(1)

where $m_i$ is the amount of magnetic moment that is switched in each event, $H_c$ is the mean coercive field and $\sigma_i$ is the standard deviation. In the transverse case, we see only one switching event in the loop, and fitting the experimental $M(H)$ curve to equation (1) yields $H_c = 12.98 \text{ mT}$, $\sigma = 0.69 \text{ mT}$, which leads to a disorder parameter $\sigma/H_c$ of 0.049. In the vortex regime there are two switching events in the loop and fitting leads to $H_{c1} = 13.41 \text{ mT}$, $\sigma_1 = 1.12 \text{ mT}$, a disorder parameter of 0.084 and $H_{c2} = 10.25 \text{ mT}$, $\sigma_2 = 0.86 \text{ mT}$, which also corresponds to a disorder parameter of 0.084. It therefore appears that both samples have a relatively low disorder with $\sigma/H_c < 10\%$.

In order to understand the remarkable differences in the array switching characteristics, analysis of STXM images is necessary. Figure 3 shows a series of STXM images taken of sample V (vortex regime) at 0.5 mT intervals in the negative field direction. Black colour contrast indicates that the magnetization has a component in the positive $x$-direction and white colour contrast indicates the magnetization has a component in the negative $x$-direction. Figure 2(b) shows the onset of switching in the array, occurring at a field of $-10.5 \text{ mT}$.
Figure 3. Switching of the Kagome artificial spin-ice system in the vortex domain wall regime under stepwise application of magnetic fields in the \(-x\)-direction after saturation in the \(+x\)-direction. White contrast indicates positive \(x\)-component of magnetization and black contrast indicates negative \(M_x\). The scale bar represents 10 µm.

Switching starts at the edges of the array and is mediated by domain walls creating long cascades that cover the entire length of the array. As the field becomes more negative (figures 3(c)–(f)) more cascades are created until the vast majority of the edge bars have switched, leaving isolated
bars that have not yet switched within the bulk of the array. Further switching within the array requires the nucleation of domain walls from within the bulk of the array, for which there is a substantial energy cost. In the region from $-13$ to $-15$ mT, vortex domain walls are nucleated in the bulk of the array and the remaining bars within the array switch. Switching within sample V was studied multiple times, and at no point were monopole defects observed. Where cascades terminated in an unswitched region, an ice-rule-obeying vertex of opposite magnetic charge is observed, as shown in figure 4.

Figure 5 shows some example STXM images taken of sample T (transverse regime) at 0.5 mT intervals in the negative field direction. Switching in the transverse regime also starts at the edges of the array. However, the single-chain cascade behaviour observed in the vortex regime does not occur and multiple branches appear quickly in the reversal process. This can also be seen in the partial hysteresis loop shown in figure 2. Switching takes place rapidly within a 2 mT window. The last few bars to switch are those in which the opposite direction of magnetization was stabilized by a magnetic Coulomb blockade effect [4] at a monopole defect. Monopole defects are seen during the switching process of sample T, as shown by red circles in figure 5(c), but are relatively rare and occur on less than 1% of the vertices.

4. Discussion

A simple model in which the domain walls are considered to be magnetically charged circular discs of diameter $w$ explains well the occurrence of extrinsic monopole defect formation [9]. The model results suggest that switching from a $Q = +1q$ to $Q = -1q$ vertex requires the emission of a $Q = +2q$ DW, so the coercive field can be approximated as the field required to overcome the Coulombic attraction between the wall and the oppositely charged vertex at the separation $w$. If all vertices are considered to be structurally identical, then any applied field that causes this depinning of the wall from the vertex is then also sufficient to push the domain wall to within a distance $w$ of an oppositely charged vertex and more than enough to depin it from that
Figure 5. Switching of the Kagome artificial spin-ice system in the transverse domain wall regime under stepwise application of magnetic fields in the $-x$-direction after saturation in the $+x$-direction. White contrast indicates positive $x$-component of magnetization and black contrast indicates negative $M_x$. The scale bar represents $5 \mu m$. 
like-charged vertex. Therefore, the $Q = +3$ state formed by this process is not a stable intermediate in the switching of a perfect array. Thus, two regimes can be defined by the relative magnitude of the quenched disorder in the distribution of bar coercivities within the array. Only in the strong disorder case, in which the total spread of coercive fields of bars within the array is greater than the fields associated with magnetic charge considerations, can monopole defects readily form by this extrinsic process.

Our STXM images indicate fundamental differences in the switching processes of an artificial Kagome spin-ice in the transverse and vortex domain wall regimes. Micro-magnetic simulations were performed in order to investigate the difference between intra-array domain wall nucleation fields for transverse and vortex domain walls. In our previous study, we found that there was a large disparity (approximately a factor of six) between measured room-temperature switching fields and zero-temperature simulations, which is simply a function of temperature, and that the simulations gave excellent agreement with images of the local magnetic structure with a simple linear scaling of the applied field [4]. Zero-temperature simulations were performed upon a single vertex, in which a domain wall was placed close to the vertex centre, on the end of the horizontal bar (oriented in the same direction as the applied field). A field was then applied to push the domain wall through the vertex and switch the structure. We find that switching occurs in two distinct stages. At the first stage, the domain wall was pushed into the vertex, resulting in the switching of one of the diagonal bars ($H_d$). In the second stage, a domain wall actually has to nucleate close to the vertex to switch the remaining diagonal bar ($H_n$). We found that in the transverse regime, $H_d$ was equal to 29.5 mT and $H_n$ was equal to 56.5 mT, whereas in the vortex regime $H_d$ was equal to 33.0 mT and $H_n$ was equal to 84.5 mT. When a domain wall reaches a vertex it is presented with a choice of two equivalent paths; in the vortex wall regime the domain wall acts as a single conserved magnetic charge carrier and selects one path; there is then a large barrier to the switching of the remaining bar, which must switch by propagation of a second domain wall of opposite charge or by intra-array domain wall nucleation. In figure 3(c) at 11 mT all the available edge nucleation points in sample V have switched, and each has produced a chain of switched bars right across the sample. A large number of bars (some in quite large domains) have been missed by the edge-nucleated walls, and so switching there can only begin by intra-array domain wall nucleation at higher fields, which can be identified as a clear step in the measured hysteresis loop. As there is one nucleation point per edge hexagon and the domain wall is choosing one path of two, approximately half of the bars in the array are switched by the edge-nucleated domain wall and the observed plateau in the MH curve is close to zero magnetization.

In the transverse regime the micro-magnetic simulations indicate that the transit of the domain wall through the vertex apparently reduces the energy barrier to nucleation of a second domain wall at that vertex, causing a branch in the cascade of switched bars from the original edge nucleation site. This effective amplification of the number of edge-nucleated domain walls gives a measurable difference to the hysteresis loop structure because the edge nucleation can assist the switching of all of the bars. The only pinning mechanism effective at fields slightly above the mean coercive field from edge-nucleated domain walls is the formation of monopole defects, which are stabilized by a magnetic Coulomb blockade effect between two walls [4].

The other clear difference between the two samples is that monopole defects are readily observed in the 18 nm sample (T) in the transverse wall regime, and are not observed in the 36 nm sample (V), which is in the vortex wall regime. Monopole defect formation is at least somewhat statistical in nature and always comparatively rare, so the data presented here are not
statistically sufficient to claim that monopole defects can never occur. However, given the strong similarity of the MH loop of the 36 nm sample in figure 2 to that of Daunheimer et al [5], where many more extensive studies have shown that monopole defects are convincingly absent, it is very likely that we are in the same regime, and the absence of monopole defects is not merely chance. Note that if we attribute the presence of monopoles to high disorder, and parameterize the two samples with the disorder parameter, $\sigma/\bar{H}_c$, we would obtain disorder parameters of 0.049 for $T$ and 0.085 for $V$ and then our predicted monopole populations would not match the experiment. In fact, the samples were fabricated in parallel processes and the only evidence of any difference at all in the extrinsic disorder is the greater surface roughness in sample $V$, which cannot account for either the unusual shape of the hysteresis loop of sample $V$ or the observation of monopole defects only in sample $T$. This illustrates that this parameterization method can give artificially high estimates of the extrinsic disorder for 180° reversals [5]. It is our hypothesis that the presence or absence of monopole defects is instead controlled by the stability of the magnetic Coulomb blockade between two domain walls in close proximity [4]. We have previously shown that a modified version of the simple Coulombic argument which takes into account the very different transverse domain wall magnetic charge distribution (figure 1) can explain the stability of the Coulomb blockade vertex between two transverse walls understanding the analogous process in the vortex regime requires some knowledge of the pinning potential at the vertex for vortex domain walls and the magnetic charge distribution of vortex domain walls of different chirality. In order to determine the pinning potential at the vertex, micro-magnetic simulations were performed upon a single vertex that had been preset to a Coulomb blockade-type monopole defect state where it is the bar parallel to the field direction that is unswitched. A field was then applied in the positive $x$-direction in order to determine the field needed to depin a domain wall and propagate through the vertex. For transverse domain walls ($t = 18$ nm) the depinning field here is greater than $H_d$ for the ice-rule vertex, whereas for vortex domain walls ($t = 36$ nm) the monopole depinning field is less than that of the ice-rule vertex. Thus the simple charge argument for no monopole defects of Mellado et al [9] works well in the vortex case. This observation is unsurprising as the magnetic charge distribution in a vortex domain wall is much closer to the charged circular disc approximation.

The enhanced tendency for monopole defect formation is also illustrated by micro-magnetic simulations of 180° reversal in a single vertex. The structures were first saturated in the positive field direction, after which 0.1 mT steps were applied in the negative field direction. This was carried out on samples of 18 and 36 nm thickness. Figure 6 shows the hysteresis loops generated for samples of 18 nm thickness (a) and samples of 36 nm thickness (b) with micro-magnetic configurations (inset). Switching in the 18 nm vertex leads to the creation of a monopole defect, stabilized by the magnetic Coulomb interaction, as we have seen previously [4]. This gives a noticeable step in the loop, since a finite amount of field is needed to overcome the mutual magnetic Coulomb repulsion of the two transverse walls. The simulation in the 36 nm structure led to an abrupt switching, and no monopole defect was seen.

In conclusion, we have carried out an extensive study of the magnetic reversal process in the transverse and the vortex domain wall regime for an artificial Kagome spin-ice system. Monopole defects are absent from the reversal in the vortex domain wall regime and this seems to be due to the difference in magnetic charge configurations of the two wall types. The highly asymmetric magnetic charge distribution in the transverse walls gives a much stronger inter-wall Coulomb repulsion than in the vortex walls, in which the charge is more diffuse. Hence,
monopole defects can form by the Coulomb blockade mechanism in the transverse case, but not in the vortex regime. In both regimes we find that the intra-array domain wall nucleation field ($H_N$) is large compared to both the edge nucleation field and the intra-array domain wall depinning fields ($H_P$). Thus, magnetic switching is mediated by long cascades carried by edgenucleated domain walls. We find that the larger intra-array depinning field within the vortex regime is responsible for a distinct plateau in the hysteresis loop. Our results imply that the vortex regime provides a better model system for the study of ‘magnetricity’ effects [14] where conserved magnetic charge carriers are injected into the structure, whereas the transverse regime appears to offer some more exotic functionality, including storage of stable monopole defects in a low-disorder system and the amplification of the number of injected carriers in the array.

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