Magnetic dipole configurations on honeycomb lattices: effect of finite size and boundaries

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Artificial dipolar spin-ice patterns have attracted much attention recently because of their rich configurations and excitations in the form of Dirac strings connecting magnetic monopoles. We have analysed the distribution of excitations in the form of strings and vertices carrying magnetic charges \( Q = \pm 3q \) in honeycomb artificial spin-ice patterns. Two types of patterns are compared, those that terminate with open hexagons and those with closed hexagons. The dipole configurations and the frequency of spin-ice rule-violating \( Q = \pm 3q \) vertices depend slightly on the boundary conditions of the pattern. Upon rotation of the patterns by \( 2\pi \) in a coercive magnetic field of 500 Oe, complete reversibility of the charge and string configuration is observed.

**Keywords:** artificial spin ice; magnetic force microscopy; magnetic dipole interaction

1. Introduction

Lateral patterns of magnetic dipoles arranged onto square or kagome lattices have recently attracted much theoretical and experimental attention because of their intriguing excitation properties above the ground state [1–5]. In particular, these studies report on the excitation of magnetic monopoles and their separation, forming Dirac strings of bound and opposite magnetic charges [3,5]. Artificial magnetic dipole patterns are often referred to as spin-ice patterns. Depending on the symmetry of the pattern, we distinguish between square ice, following the spin-ice rule ‘two-in–two-out’, or kagome spin ice, with the modified spin-ice rule ‘two-in–one-out’ or vice versa. In the latter case, magnetic islands decorate an underlying honeycomb lattice. Assuming that the shape of the islands can be described as an infinitely thin needle in a single magnetic domain state, we can regard such a needle as a magnetic dipole. According to Castelnovo et al. [6] and Möller & Moessner [7], such magnetic dipoles can be described as pairs of opposite magnetic charges \( \pm q = \pm \mu/l \), where \( l \) is the length of the dipole or the charge separation, and \( \mu \) is the magnetization of the needle (island). Then, in a honeycomb lattice, at each vertex, three dipoles meet with total magnetic charge \( Q = \sum_{i=1}^{3} q_i \). Vertices fulfilling the ice rule have a total charge of \( Q = \pm 1q \), and those violating the spin-ice rule have total charges \( Q = \pm 3q \). In a high saturating magnetic field, all magnetic dipoles will preferentially point

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Figure 1. (a) Schematic of the charge-ordered state of the kagome spin-ice pattern with $Q = \pm 1q$ at each vertex, reminiscent of the charge order in an ionic crystal such as NaCl. The charge-ordered state is realized in a saturating magnetic field with magnetic dipoles pointing predominantly into the field direction oriented parallel to the horizontal dipoles. (b) Flipping of one horizontal dipole in a descending field corresponds to a local magnetic charge excitation of $\Delta Q = \pm 2q$. (c) The local magnetic charge excitation may propagate by further dipole flips, forming a Dirac string of separated magnetic charge excitations. (i) Dipole model; (ii) dumbbell model of magnetic charges at each end of a dipole. (Online version in colour.)

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into the field direction, thereby forming a magnetic-charge-ordered $Q = \pm 1q$ state, resembling an ionic crystal such as NaCl. This ordered state is also referred to as the ‘ice II’ state \cite{7,8} and is schematically shown in figure 1a. Figure 1a(i) shows schematically a dipole model of the kagome lattice, whereas figure 1a(ii) reproduces a dumbbell model \cite{6,7}, which is closer to what one observes by imaging the patterns with magnetic force microscopy. Returning to coercivity in a descending field, it is likely that one dipole oriented antiparallel to the field direction reverses in order to align parallel to the external field, creating a local excitation with magnetic charge change $\Delta Q = \pm 2q$ and resulting total magnetic charges $Q = \pm 3q$ (figure 1b). Via dipole interaction with the neighbours in this vertex, the local excitation may propagate by flipping further dipoles (figure 1c). After a few flips, the local excitations have separated, forming a Dirac string of separated magnetic charges with diminishing dipole interaction. These excitations have indeed recently been observed by Mengotti et al. \cite{5}.

Although, in reality, islands in artificial patterns have finite width and higher multipole contributions \cite{9}, in the following, we follow the idealistic scheme and discuss our experimental results in terms of local magnetic charge per vertex in honeycomb lattices.

Recently, we have reported on the excitation of single monopole–antimonopole pairs in honeycomb patterns forming a gas of magnetic charges that condense into a charge-ordered state of $Q = \pm 3q$, when sweeping the external magnetic field through coercivity \cite{10}. Although the charge ordering is similar to the one observed in the ice II phase, the local charge per vertex is tripled, causing a much higher charge repulsion. For this reason, the $Q = \pm 3q$ charge-ordered state
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is not a ground state but can be realized whenever the magnetization reversal energy of individual islands dominates over the dipole–dipole interaction between neighbouring islands [11]. As the lateral extensions of the patterns were rather large (200 × 200 μm containing up to 400 vertices), in the statistical analysis of the magnetic charges the boundaries can safely be ignored. However, in smaller patterns, the boundaries should play a crucial role, and the frequency of \( Q = \pm 1q \) versus \( Q = \pm 3q \) vertices may be affected by the terminating hexagons, i.e. whether they are open or closed. When patterns are terminated by open hexagons, all vertices including those at the border comprise three islands. On the other hand, only two islands meet in a vertex in the outermost hexagons if they are closed. In a recent publication, Budrikis et al. [12] have calculated and simulated the effect of open and closed squares in square spin-ice patterns. In particular, they find that, upon rotation of the pattern in a magnetic field corresponding to the coercivity of the pattern, the frequency of type I vertices increases successively upon each complete rotation. Type I states in square ice pattern are those with two opposite dipoles pointing into the vertex and two pointing out, yielding a total charge of \( Q = 0 \). Similar conclusions should also apply for the kagome spin ice, and one may expect a reduction of high-energy \( Q = \pm 3q \) vertices by rotating the pattern in a constant field close to the coercive field value.

2. Experimental procedures

We have prepared six different kagome patterns in sets of three that are distinguished by their terminating hexagons either being open or closed at the border of the patterns. The island–island separation in each pattern was either 200, 400 or 800 nm. All six patterns consist of polycrystalline Fe. Thin Fe films with a thickness of 20 nm were deposited by ion beam sputtering (Roth and Rau) onto a Si(100) substrate. To improve the adhesion of Fe on Si, a 5 nm thick Ta film was deposited first. Finally, the Fe film was covered with a 2 nm aluminium oxide layer for oxidation protection. As the Si surface is terminated by a natural silicon oxide layer without preferred orientation, the deposition of Ta and Fe results in a polycrystalline structure of these films. Poly-crystallinity was intentionally aimed for in order to eliminate, on average, crystal anisotropy contributions and to enhance the magnetic dipole character of the islands. Subsequently, the films were spin-coated with a negative resist and exposed by electron beam lithography (Raith ELPHY Quantum) to define islands with an aspect ratio of 10 : 1. For the present study, the island dimension was 1 × 0.1 μm. This aspect ratio is sufficient to achieve a single-domain state in polycrystalline Fe islands at remanence. Scanning electron microscopy (SEM) images (FEI Quanta 200FEG) of two representative patterns with open and closed hexagons are shown in figure 2.

In order to analyse the effect that the boundary of the honeycomb lattice has on the dipole configurations, we used a magnetic force microscope (MFM; HV-Solver of NT-MTD) with a scan range of 100 μm. In MFM images, the contrast is provided by the emanating stray fields at each end of the magnetic islands. Domain states reduce the contrast and hamper the assignment of the magnetic charge on either end. The pattern size of 50 × 50 μm was kept smaller than the scan range, such that the entire pattern can be scanned even with different

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azimuthal angles. On the other hand, the patterns still contain a sufficient number of magnetic islands to account for a representative analysis. A magnetic field was applied in the azimuthal plane and in a field range of ±1000 Oe. For the sample rotation, we used a small rotational stage with angle positioning via a piezodrive (Attocube ANR30).

In the SEM images of the honeycomb patterns (figure 2), the crystallographic directions are indicated for characterizing the orientation of the patterns. For an orientation [10] parallel to the external field, all islands are oriented either perpendicular to the field or have an angle of inclination of 30°. We call this direction the hard-axis orientation. For a [11] orientation of the pattern, one of the island sublattices is aligned parallel to the field direction, and the other two have an inclination angle of 60°. This orientation is referred to as the easy-axis orientation.
For processing MFM images, a program was developed in order to generate ‘digital’ magnetic hysteresis curves and for counting the number of vertices having either charges $Q/q = \pm 1$ or $Q/q = \pm 3$ [10,11]. Islands in a domain state with unclear magnetic charges were discarded.

3. Experimental results

(a) Field dependence

We usually start recording MFM patterns from the demagnetized state of the pattern. Demagnetization is achieved by rotation of the pattern in a stepwise reducing magnetic field, following a procedure described by Wang et al. [13]. Then, the pattern is oriented along the [10] or [11], orientation and the magnetic field is swept first to positive values up to saturation, returning to remanence and reversing the field direction up to negative saturation, etc. Changes in the magnetic charge configuration occur only with increasing field. In minor loops, the remanent magnetization is always identical to the one that was reached along the hysteresis. This shows that any change in the magnetic charge configuration has a large potential barrier, much higher than thermal energies. Each configuration is basically frozen in. Changes of the respective configuration can be achieved only by applying the next higher field value if saturation has not yet been reached, or by reversing the field direction.

Upon crossing the coercive field $H_c$ at about 400–500 Oe, the magnetic charge configuration becomes very sensitive to external field variations and may change its value quite dramatically in response to small field changes. When crossing the coercive field point, we observe rather different configurations, depending on whether the pattern is terminated with open or closed hexagons. In figure 3, we show MFM maps of both patterns in the [11] orientation taken on the ascending branch crossing the coercivity at an external field of 500 Oe. In patterns with closed hexagons at the edges, only a few dipole strings become visible; one of them is marked by white lines in figure 3a. By contrast, the pattern with open hexagons at the edges exhibits many more strings at the same external field, and their length and number increase dramatically up to saturation (figure 3b). The strings contain dipole arrays with alternating $Q = \pm 1q$ magnetic charges at each vertex. These strings are very similar to those observed by Mengotti et al. [5] via photoemission electron microscopy. They indicate excitations of magnetic monopoles, which become separated by the length of the strings.

Furthermore, we have analysed the frequency of $Q = \pm 3q$ vertices in patterns with open and closed boundaries as a function of the external field. The results are plotted in figure 4 for the pattern with 800 nm island separation. We find that the frequency of $Q = \pm 3q$ vertices is much less than in patterns that we have investigated previously using bigger islands [10]. Still, the highest frequency of about 10–20% always occurs at the coercive field values. In patterns with only 400 nm separation between the islands, the abundance of $Q/q = \pm 3$ vertices is even lower. The difference is probably due to the size of the dipoles used in these patterns ($1 \times 0.1 \mu m$) when compared with the earlier ones ($3 \times 0.3 \mu m$), favouring inter-island interaction over the intra-island reversal energy. These findings are in accordance with the results of Mengotti et al. [2,5].
Figure 3. Honeycomb patterns with island separation of 800 nm in a field of 500 Oe applied in the [11] direction (0° orientation), corresponding to the coercivity point of the magnetization reversal. Strings of \( Q = \pm 1 \) dipoles marked by white boxes can be recognized in honeycomb patterns terminated by open or closed hexagons. The strings are more abundant in patterns with open hexagons at the borders (b) than with closed hexagons (a). (Online version in colour.)

(b) Rotational dependence

Next, we discuss the abundance of \( Q = \pm 3q \) vertices of the same patterns but in a constant magnetic field close to the coercivity of 500 Oe and during clockwise rotation from 0° (magnetic field parallel to the [11] direction) in 15–20 steps up to 360°. Representative examples for the 800 nm pattern with closed hexagons at the border are shown in figure 5 at rotation angles 0°, 94°, 180° and 360°. The
rotation of the pattern can be recognized by the low contrast area. At rotation angles $0, 0.33\pi, 0.66\pi$, etc., one of the sublattices of the pattern is aligned parallel to the field direction. Thus, one would expect a sixfold symmetry expressed in the distribution of $Q = \pm 3q$ vertices during rotation of the pattern. The results from the evaluation of the 800 nm patterns are plotted in figure 6 for open and closed hexagons at the border. The expected sixfold symmetry cannot be recognized. Surprisingly, the pattern with closed hexagons starts with a very low number of $Q = \pm 3q$ vertices at zero angles and goes through a maximum at $180^\circ$, returning to the same low frequency upon full rotation. By contrast, the pattern with open hexagons exhibits a bimodal distribution together with some asymmetry, which may be an artefact and due to a slight off-centre positioning of the sample. Again, we notice that the frequency of $Q = \pm 3q$ vertices is lower in the 400 nm patterns than in the 800 nm patterns, a trend already seen for the field dependence (not shown here).

Aside from some experimental artefacts and limited statistics, we can clearly confirm three main observations from the rotational dependence of the patterns in a constant coercive magnetic field. First, the frequency of $Q = \pm 3q$ vertices...
Figure 5. MFM maps of the 800 nm pattern taken in a constant magnetic field of 500 Oe and for different rotation angles from $0^\circ$ (field parallel to the [11] direction) in a clockwise fashion to $360^\circ$. Representative examples are shown for four angles completing one full rotation. The rotation of the pattern can be followed by identifying the low contrast area, which is located on the left-hand side in the $0^\circ$ orientation. (Online version in colour.)

is much reduced compared with patterns that we have investigated previously consisting of larger islands. Second, the rotational dependence does not show any sixfold symmetry. And third, the vertex configuration appears to be reversible. Upon rotation by $2\pi$, the initial state is recovered. In general, the recovery of the pattern should be understood in a statistical sense, meaning that the statistical abundance of $Q = \pm 3q$ vertices is essentially equal to the one observed at the starting point, but may be different with respect to the exact local configuration. In figure 7, we have analysed the abundance and location of the $Q = \pm 3q$ vertices. The maps are similar to those shown in figure 5, but with a slightly different selection of angles of rotation and zoomed in on the right-hand part of the pattern. Furthermore, the $Q = \pm 3q$ vertices are highlighted by transparent discs. From $0^\circ$ to $180^\circ$, we notice a substantial increase of $Q = \pm 3q$ vertices, which bunch together into extended domains. After further rotation to $315^\circ$, the domains disintegrate and the number of $Q = \pm 3q$ vertices is again drastically reduced. Finally, after a full rotation to $360^\circ$ we observe not only the same number of $Q = \pm 3q$ vertices as at $0^\circ$, but also that their locations are identical. Thus, the recovery of the pattern after one full rotation is not only present in a statistical sense, but indeed also—as far as we can see—given in a configurational sense. No annihilation of $Q = \pm 3q$ excitations with higher energy could be discerned upon rotation. Instead, there appears to be a configurational memory for the number and location of these $Q = \pm 3q$ excitations upon complete
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Figure 6. Abundance of $Q = \pm 3$ vertices in a pattern with 800 nm separation of the magnetic islands with (a) open and (b) closed hexagons at the borders as a function of rotation angle with respect to an applied constant field of 500 Oe. Zero radians corresponds to the $[11]$ direction as shown in figure 2.

rotation. The $Q = \pm 3q$ excitations are usually starting or end points of Dirac strings, as sketched in figure 1. Indeed, these strings can also be observed in the patterns shown in figure 7. As a guide to the eye, these strings are marked with white semitransparent bars. After a complete rotation, the same strings reappear (aside from one), confirming the recovery of the charge distribution and their spatial configurations. This reproducibility in a rotational field is astounding and unexpected. It is not in line with predictions by Budrikis et al. [12], according to which a progressive change in the pattern should take place upon multiple rotations, favouring the low-energy vertices at the expense of the high-energy vertices. Instead, we find essentially a complete reversibility and no annihilation of the $Q = \pm 3q$ vertices upon rotation.

4. Summary and conclusion

We have fabricated, by lithographic means, honeycomb patterns decorated with magnetic islands that show, by virtue of their aspect ratio, dominating single-domain and dipolar character. The dipole configurations were imaged by MFM as a function of several parameters: distance of islands in the vertices of 200, 400 and 800 nm; external field; and orientation of the patterns with respect to the field direction. Furthermore, the patterns are distinguished by their different edge geometries, i.e. those with open hexagons and those with closed hexagons at the borders of the pattern.
Our investigations showed that the boundary condition of the patterns, i.e. whether they are terminated with open or closed hexagons, affects the charge distribution and the abundance of $Q = \pm 3q$ vertices in the pattern to some extent. During magnetization reversal, their frequency has a peak at the coercivity, independent of the boundary condition. Overall, their frequency is much less (10–20%) when compared with patterns of similar island separation but larger dimension of the dipoles (60–80%) \[10,11\]. There is, however, a difference with respect to the formation of Dirac strings when crossing the coercive field: many more strings emanate from the border to the interior of the pattern when the hexagons are open at the border than when they are closed.

When rotating the pattern with closed hexagons, large domains of $Q = \pm 3q$ vertices occur over a large angular range and finally annihilate after completing a full rotation. In fact, we noticed that the initial and final charge distributions

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Figure 7. MFM images of the 800 nm pattern similar to the ones shown in figure 5 but now zoomed into the right-hand part of the 0° pattern. The $Q = +3q$ vertices (white) are marked with white transparent discs, and those with $Q = -3q$ (dark) are marked with yellow transparent discs. In the 180° pattern, the $Q = \pm 3q$ vertices form large domains in the upper right corner. Dipole strings in the 0° and 360° patterns are marked by white transparent bars. Note that the charge and the string configurations are identical in the 0° and 360° patterns. (Online version in colour.)
are not only the same on average. They indeed exhibit essentially identical charge configuration, including the location of $Q = \pm 3q$ vertices and the dipole strings that are attached to them. During rotation of patterns with open hexagons, domain formation of $Q = \pm 3q$ vertices was observed to a much lesser degree. Overall, rotation of honeycomb patterns at the coercive field was found to be very rich in the excitation of many different dipole configurations. But in the end, after a complete cycle, the dipole configurations are identical to the starting position. Therefore, we cannot confirm the predictions of Budrikis et al. [12] for square ice patterns, according to which a progressive change in the pattern should occur upon multiple rotations, favouring the low-energy vertices at the expense of the high-energy vertices. Instead, we find a complete reversibility and no annihilation of the high-energy $Q = \pm 3q$ vertices upon rotation.

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