Magnetic dipole configurations in honeycomb lattices: order and disorder

Alexandra Schumann\textsuperscript{1}, Philipp Szary\textsuperscript{1}, Elena Y Vedmedenko\textsuperscript{2} and Hartmut Zabel\textsuperscript{1,3}

\textsuperscript{1} Institut für Experimentalphysik/Festkörperphysik, Fakultät für Physik and Astronomie, Ruhr-Universität Bochum, 44780 Bochum, Germany
\textsuperscript{2} Institut für Angewandte Physik, Universität Hamburg, Jungiusstrasse 11, D-20355 Hamburg, Germany
E-mail: hartmut.zabel@rub.de

\textit{New Journal of Physics} 14 (2012) 035015 (19pp)
Received 17 November 2011
Published 20 March 2012
Online at http://www.njp.org/
doi:10.1088/1367-2630/14/3/035015

\textbf{Abstract.} Dipolar spin ice has attracted much attention because of its intriguing ground state ordering and elementary excitation properties. We present experimental realizations of magnetic dipolar spin ice on periodic lattices with honeycomb symmetry. We have analyzed in particular the evolution and distribution of excitations with magnetic charges $\pm 3$ per vertex as a function of magnetic field and the distance $b$ between the dipoles ranging from $b = 0.4$ to $1.7 \, \mu m$. In all the dipole patterns investigated, we observe a surprisingly high abundance of $\pm 3$ magnetic charges at coercivity in the descending and ascending branches of the magnetic hysteresis. At the same time, these $\pm 3$ vertices form a charge ordered state with large domains, resembling an ionic crystal. Monte Carlo simulations of the magnetization reversal confirm in the framework of a macrospin model an enhanced abundance of $\pm 3$ magnetic charges at the coercive field. But much better agreement is achieved by taking into account the micromagnetic reversal mechanism, which proceeds via nucleation and domain wall propagation for dipoles aligned with the field and via coherent rotation for all others.

\textsuperscript{3} Author to whom any correspondence should be addressed.
1. Introduction

Lateral patterns of magnetic dipoles arranged onto square or triangular lattices have recently attracted much theoretical and experimental attention because of their intriguing elementary excitation properties above the ground state [1–5]. In particular, in these recent publications the excitation of magnetic monopoles was reported and how these separate to form Dirac strings of bound and opposite magnetic charges [3, 5]. Artificial magnetic dipole patterns are often referred to as spin-ice patterns. Those are two-dimensional (2D) counterparts of 3D pyrochlore spin-ice lattices. The term ‘spin ice’ refers to the ice rule of water ice, where on average two hydrogen atoms point towards an oxygen ion in a tetrahedral environment and two point away [6]. In 3D spin-ice lattices, such as Dy$_2$Ti$_2$O$_7$ [9, 18] and Ho$_2$Ti$_2$O$_7$ [8], the spin-ice rules govern the ordering of Ising spins in the corners of tetrahedra [7, 10, 19, 20] implying that two out of the four spins on the vertices are directed toward the tetrahedral center, whereas the other two spins point away. The two-in–two-out rule can be mapped onto 2D square lattices. Here the Ising spins are replaced by single-domain macro-spins with magnetic dipole character, which interact at each vertex via dipole–dipole interaction [1, 11, 12]. For triangular kagomé or honeycomb lattices decorated with magnetic dipoles the corresponding ice rule applies: two-in–one-out or vice versa two-out–one-in [3, 5, 13]. If we assign according to Castelnovo et al [10] to each magnetic dipole $p$ equal and opposite magnetic charges $\pm 1q = \pm 1p/l$, where $l$ is the charge separation, then in the kagomé lattice each vertex fulfilling the ice rule has a total charge of $\pm 1$ in units of $q = p/l$. Vertices with three dipoles pointing into a vertex or three dipoles pointing out of a vertex violate the ice rule. Their local charge correspondingly is $\pm 3$ (see figure 1). Vortices with local charge $\pm 3$ are termed type I vortices, whereas the six configurations that fulfill the ice rule are collectively referred to as type II vertices.

In the following, we will limit our discussion to the honeycomb lattice. The honeycomb lattice consists of three sublattices. If one of the sublattices is aligned parallel to the external magnetic field (referred to as the easy axis), in saturation the dipoles in all three sublattices will align such that a charge ordered state with alternating charges $\pm 1$ is generated at each vertex. This $\pm 1$ charge order is reminiscent of an ionic crystal. On reversing the field direction to the opposite saturation field, at each vertex the charge is exchanged by flipping each individual dipole of the pattern. The charge ordered state in saturation is shown schematically in figure 2(a).
Figure 1. Possible configurations of magnetic dipoles on a honeycomb lattice. The configurations are separated into type I, which violate the spin-ice rule, and type II, which obey the spin-ice rule. Each configuration is assigned to a charge value, which is the total charge per vertex.

Figure 2. (a) Charge order in saturation; (b) the flipping of a single dipole parallel to the applied field causes the excitation of single magnetic dipole; (c) a sequence of flipped dipoles form a Dirac string.

If an individual dipole parallel to the field direction is flipped, then the charge changes from $+1$ to $+3$ on one side and from $-1$ to $-3$ on the other side. Thus, at both participating vertices the charge change is $\pm 2$ (figure 2(b)). As this flip costs magnetic energy, it corresponds to a local excitation of a new $\pm 2$ monopole–antimonopole pair [5]. Individual and uncorrelated flips may be considered as a gas of local excitations. However, if the flipping of the first dipole causes subsequent dipoles to flip in a domino-like effect, then the monopole–antimonopole pair becomes separated, forming a Dirac string of charged monopoles, where the vertices at both ends have changed their charge state by $\pm 2$; see figure 2(c). The total charge difference remains zero, but by the sequence of flips two magnetic monopoles have separated. Mengotti et al [5] have observed and analyzed the formation of Dirac strings during reversal at the coercive field by photoemission electron microscopy (PEEM) using the XMCD effect for the magnetic contrast. Ladak et al [3] have imaged honeycomb lattices by magnetic force microscopy (MFM) for different magnetic fields close to coercivity. Similar to the work of Mengotti et al, they have observed the excitations of local ice rule violating $\pm 3$ monopole pairs, which then become separated into Dirac strings by changing the magnetic field. In contrast, Qi et al [14] have never observed local $\pm 3$ excitations in their connected honeycomb patterns, nor emerging strings. In continuous honeycomb patterns where the dipoles are physically connected in each vertex, local $\pm 3$ states are very difficult if not impossible to excite [15].
Recently, we reported on the excitation of single monopole–antimonopole pairs in honeycomb patterns forming a gas of local $\pm 3$ pairs that condense into a charge ordered state, when sweeping the external magnetic field through coercivity [16]. As the magnetic dipoles are not connected, our experimental conditions can be characterized as the weak interaction limit. The honeycomb lattice with connected magnetic dipoles as investigated by Tanaka et al [13], Ladak et al [3] and Qi et al [14] may be characterized as the limit of strong interaction. The experimental conditions realized by Mengotti et al [2, 5] lie intermediate between the strong and the weak interaction limit. The weak interaction limit is analogous to a van der Waals potential that has only a hard core repulsion radius but no specific minimum in the interaction potential. Nevertheless, a van der Waals gas may condense into an ordered state under appropriate conditions. Here we provide a more detailed account of the charge ordering effect of magnetic monopoles in honeycomb patterns, scrutinizing in detail the conditions for its occurrence from an experimental point of view as well as using Monte Carlo and micromagnetic simulations.

This paper is organized as follows. We first describe in section 2 the experimental procedures for pattern preparation and for image processing. In section 3.1, we report on the reversal of single elements and in section 3.2 on demagnetization procedures. The reversal of complete patterns is described in 3.3, followed by a discussion of the effect of local defects in the patterns (section 3.4). In section 4, we present Monte Carlo simulations of the patterns that are close to the experimentally studied patterns. We conclude in section 5 and provide a summary of the results.

2. Experimental procedures

All our magnetic dipole 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 film 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 polycrystalline structures of these films. Subsequently, the film was spin coated with a negative resist and exposed by electron beam lithography using a Raith ELPHY QUANTUM lithography unit to define islands with an aspect ratio of 10 : 1. After development, the surplus material was removed by means of ion milling. A final removal of the residual resist on top of the magnetic islands facilitates the force microscopy imaging process. MFM images were taken with a set of samples that have an edge length of 3 $\mu$m and a width of 0.3 $\mu$m. Preliminary studies showed that this aspect ratio is sufficient to achieve a single domain state in polycrystalline Fe islands at remanence.

The magnetic islands were positioned on triangular lattice sites decorating a honeycomb lattice such that three dipoles meet at any vertex at an angle of 120°. Three different interparticle distances $b$ were used in order to vary the strength of the dipolar interaction: 0.4, 0.8 and 1.7 $\mu$m. Each writing field of a pattern has the lateral extension of $200 \times 200 \mu$m$^2$ and contains from 100 up to 400 vertices, depending on the separation of the dipoles. This is sufficient for an analysis of the vertex states inside a pattern, as the boundary effect does not propagate far into the pattern according to our analysis of boundary effects reported elsewhere.

For topological imaging of the patterns, we used a scanning electron microscope (SEM) with a Schottky emitter (FEI Quanta 200FEG). An MFM was used for magnetic imaging.
Figure 3. SEM images of a small section of a much bigger honeycomb pattern with different separations and periodicities between the islands, consisting of polycrystalline Fe with a length of 3 $\mu$m and a width of 0.3 $\mu$m. Bars indicate the scale and arrows show the basis vectors of the honeycomb lattice.

(HV-Solver of NT-MTD), retrofitted by a rotational stage (attocube ANR30). This was necessary for proper alignment of the samples with respect to the applied magnetic field provided by an electromagnet, and for carrying out rotational studies. The magnetic tip consists of CoCr alloy and can sustain fields of up to 1000 G without reversing. MFM images were taken in far field at a few hundreds of nm distance from the sample surface, where the magnetic dipole interaction dominates over van der Waals forces for topographical imaging at smaller distances. The lateral resolution of the MFM images is of the order of 200 nm. All imaging experiments were carried out at room temperature.

Representative SEM images of the honeycomb patterns are shown in figure 3. The two basis vectors of the honeycomb lattice are indicated, which are used for characterizing the orientation of the patterns. For an orientation [10] parallel to the external field, all dipoles are oriented either perpendicular to the field or have an inclination angle of 30°. We call this direction the hard axis orientation. For a [11] orientation of the pattern, one of the dipole sublattices is aligned parallel to the field direction; the other two have an inclination angle of 60°. This orientation is referred to as the easy axis orientation.

For processing the MFM images, a program was developed in order to generate 'digital' magnetic hysteresis curves and for counting the number of vertices, being either in a type I or a type II state. Dipoles that are oriented parallel to the field are assigned the normalized value $\pm 1$. All other dipoles have the values $\cos(\pm 30^\circ; \pm 60^\circ) = 0.866; 0.5$, depending on their inclination angle. Also the vertices can be assigned to discrete values depending on the respective configuration. First the number of white ($-1$) and black ($+1$) charges are counted in a vertex. The sum yields the total charge of a vertex. Summing up all charges from all vertices, the total charge, i.e. the charge neutrality, can be verified. The configuration of charges within a vertex is also recorded and magnetic dipole values are assigned to the respective vertex configuration according to figure 4, where all possible configurations together with their assigned values are listed for the easy and the hard axis direction. In the hard axis direction, the vertical dipoles do not contribute to the magnetization determined in the horizontal direction. The assigned dipole values should not be mixed up with the charge values per vertex. For instance in the top two rows the sum of the magnetic dipole values are zero, as the magnetic dipoles cancel each other out. However, the charge value is +3 in the first row and −3 in the second row.
3. Results

3.1. Single, double and triple dipoles

Before starting the analysis of complete patterns, we have performed magnetization reversal studies of single, double and triple dipoles of the same aspect ratio as that used in the extended patterns.

For single dipoles oriented parallel to the field direction, a switching field of $400 \pm 50$ G was determined. The magnetization reversal takes place via domain wall nucleation and propagation. In the case of the double-dipole structure, one dipole was aligned parallel to the field direction and the other one is $60^\circ$ inclined. In this case, the switching field is $400$–$500$ G for dipoles with a separation of 0.4 and 0.8 $\mu$m, but it is only 300 G for a separation of 1.7 $\mu$m. For the triple dipoles, the switching fields are essentially the same with a slight tendency to lower values. A representative example is shown in figure 5 of a double dipole with a separation of 0.4 $\mu$m at different field values before and after switching.

3.2. Demagnetization protocol for honeycomb patterns

Next we present the magnetization reversal of complete honeycomb patterns. In order to generate the same initial condition for all patterns, we have first demagnetized the pattern...
Figure 5. Magnetization reversal of a double-dipole arrangement. The dipoles have a length of 3 μm and a width of 0.3 μm and the interparticle distance is 0.4 μm. MFM images from left to right are taken at zero field, at 400 G and at 500 G. In the right panel, only the horizontal dipole has switched, but not the inclined one.

via a standard protocol. Starting from a saturation field of 1000 Oe, the polarity of the field was successively switched and the magnitude was slowly ramped down in steps of 10 Oe to zero. This demagnetization protocol was first proposed by Wang et al [1, 17] and also produced satisfactory results in our case. MFM pictures of all three honeycomb lattices in the demagnetized state and with easy axis orientation are shown in figure 6. A statistical analysis of the patterns with respect to the relative abundance of type-I vertices versus type-II vertices shows that for an interparticle separation of 0.4 μm, 8% of the vertices have a type-I configuration and 85% are in one of the six type-II configurations. For a separation of 0.8 μm these numbers are 13 versus 80%, and for a separation of 1.7 μm, we find 21 versus 73%. The relative abundances for each pattern should add up to 100%; however, some of the vertex configurations cannot clearly be identified, either due to resolution or due to local defects. Without correlation the statistical distribution should be 25 versus 75%. The pattern with 1.7 μm separation comes close to this value as one would expect for an increasing separation of the dipoles. In the case of the [10] oriented patterns, this trend is not as clear, and even for the smallest interparticle separation a much larger abundance of type-I configurations of 14% can be recognized in the demagnetized state compared to the [11] orientation. After demagnetization the average digital magnetization of the patterns is close to zero. Deviations from zero can be explained by uncertainties in the assignment or by defects.

### 3.3. Remagnetization and magnetization reversal of honeycomb patterns

#### 3.3.1. Honeycomb patterns with [11] easy axis orientation

After demagnetization of the patterns we have remagnetized them in steps of 50–100 Oe up to 900 G. For the 0.8 and 1.7 μm patterns, this field is sufficient to reach saturation. In saturation a ±1 charge-ordered state is prevalent. It is stable and also remains the same after removing the field. In figures 7(b) and (c), we show the ±1 charge-ordered state after returning to remanence. This state is characterized by the highest magnetization and on the average zero magnetic charge. It is clearly distinguished from the charge-disordered state after demagnetization, see figure 6. For the pattern with an interparticle separation of 0.4 μm, a field of 900 G is not sufficient for reaching saturation. At least two domains coexist at this field, which can also be recognized at remanence; see figure 7(a).
**Figure 6.** Three honeycomb lattices with different dipole separations and oriented along the easy [11] axis are shown in the demagnetized state: top, 0.4 µm, middle, 0.8 µm and bottom, 1.7 µm.

**Figure 7.** Honeycomb lattices with different dipole–dipole separations at remanence after saturating in a field of 900 Oe parallel to the [11] direction: top, 0.4 µm, middle, 0.8 µm and bottom, 1.7 µm.
Figure 8. The virgin curves and the digital magnetic hysteresis of three honeycomb lattices with different separations (top: 0.4 µm; middle: 0.8 µm; bottom: 1.7 µm) are plotted for the easy axis orientation.

Figure 9. The relative abundance of the type-I state during the magnetization reversal starting with the virgin curve (black solid triangles), followed by the descending branch (red solid squares), and the ascending branch (blue solid circles) for all three honeycomb lattices with different separations (top: 0.4 µm; middle: 0.8 µm; bottom: 1.7 µm) with the easy axis orientation.

For each field value, MFM images were recorded and analyzed according to the digital image processing described in section 2. Starting from the demagnetized state and magnetizing along the virgin curve, we have taken complete hysteresis loops of all patterns with the field oriented parallel to the easy $[11]$ axis of the patterns. The hysteresis curves are shown in figure 8. They confirm that in the demagnetized state the magnetization is essentially zero, deviations can
be explained by uncertainties in the identification of some vertices and by some lithographic
defects in the pattern. The coercivity for all three patterns is about 400–500 G. In all cases the
change of magnetization from the saturation value to zero at the coercive field is fast, but from
coefficacy to the opposite saturation the hysteresis is more rounded. This is due to the fact that
coefficacy can be reached by switching the horizontal dipoles at a rather low field of 400–500 Oe,
whereas the switching of the inclined dipoles requires higher fields due to the reduced torque
causing some coherent rotation before switching takes place. Thus, the hysteresis reflects the
different switching mechanisms and coercive values for the horizontal dipoles and the inclined
dipoles. For further discussion of the switching mechanism, see section 4.

Next, we analyze the relative statistical abundance of vertex configurations according to
type I and type II. As the relative abundance of type I and type II is complementary, it is
sufficient to show only the field dependence of the type I states with magnetic charges $\pm 3$.
The results are shown in figure 9. For all three patterns we recognize that the type-I state
is present in the demagnetized state as already mentioned above, but becomes predominant
at the coercive field, reaching values from 60% for the 1.7 $\mu$m pattern up to 75% for the
0.4 $\mu$m pattern. The high abundance of the type I state, which violates the spin-ice rule, is
quite surprising but can be rationalized considering the difference in the coercive fields and the
different switching mechanism for the horizontal dipoles as compared to the inclined dipoles
(see section 4). Consequentially the type I state predominates at the coercive fields and forms
a charge-ordered state, as shown already for the 0.8 $\mu$m pattern in [16]. All three patterns with
different interparticle separations at coercivity are compared in figure 10. These patterns indeed
exhibit an almost perfect regular pattern of alternating $\pm 3$ charges. Thus the highest degree of
frustration is at the same time combined with a highly charge ordered state.

As this charge-ordered pattern occurs at the coercive field, one may wonder whether it is
stable or not with decreasing applied field. It turns out that the stability of the charge ordered
pattern is very high and it continues to exist, even after switching off the magnetic field. In
fact, an opposite field of $-200$ G is required to destroy the charge order in the 0.4 and 0.8 $\mu$m
patterns, whereas $-80$ G are required for the 1.7 $\mu$m pattern. This shows again that the patterns
with smaller separation of the dipoles are more strongly coupled than the patterns with larger
separation between the dipoles.

The high stability of the charge-ordered pattern is very surprising and is not supported by
the Monte Carlo simulations discussed in section 4. It may only be understood if we assume
that the alternating $\pm 3$ charges form new magnetic dipoles, which stabilize themselves through
dipole–dipole interaction. We will come back to this point in section 3.3.3.

Closer inspection of figure 9 shows that the abundance of type-I states is not symmetric
for descending and ascending fields. The biggest asymmetry is seen for the 1.7 $\mu$m pattern. It is likely that the asymmetry may be caused by a slight misalignment of the pattern with
respect to the external field. For instance, if the effective field at the sample position is higher
in the positive field direction than in the negative field direction, then the dipole pattern is more
perfectly ordered for positive saturation, which in turn would result in a higher $\pm 3$ charge order
upon reversal at negative coercive fields.

3.3.2. Honeycomb patterns with [10] hard axis orientation. During the magnetization reversal
of the honeycomb pattern in the [10] orientation only the dipoles in two of the three sublattices
reverse their orientation, whereas the dipoles in the third sublattice are not affected by the
applied field. Their random orientation remains up to the saturation field of 1000 Oe. The
Figure 10. MFM images of the three honeycomb patterns with [11] orientation and with different interparticle separations at the coercive field value. In all three patterns, a charge-ordered state with $\pm 3$ charges at the vertices can be recognized. Local defects may disturb the order occasionally.
other two sublattices consist of dipoles which are oriented at angles $\pm 30^\circ$ and which behave symmetrically in the external field. Therefore, the magnetic hysteresis for the $[10]$ hard axis orientation is more symmetric than in the $[11]$ direction. We note that during the reversal process the relative abundance of type I states is much less for the $[10]$ orientation than in the $[11]$ orientation, 20–30% instead of 60–75% in the $[11]$ direction. Furthermore, the coercive field clearly scales with the separation of the dipoles from 500 Oe for the 0.4 $\mu$m pattern, to 400 Oe for the 0.8 $\mu$m pattern and 300 Oe for the 1.7 $\mu$m pattern. For the $[10]$ orientation, a charge-ordered state does not occur at the coercive field. In fact, the type-I states can be further suppressed by saturating the sample first in the $[11]$ orientation and subsequently rotating the pattern back into the $[10]$ orientation. With this procedure all dipoles perpendicular to the applied field form a ferromagnetically aligned sublattice. Upon magnetization reversal only the $\pm 30^\circ$ inclined dipoles switch their orientation more or less simultaneously at the coercive field, leaving no room for type-I formation. After saturation in positive or negative applied fields, a $\pm 1$ charge ordered state is generated, which is also stable after returning to remanence, as can be seen in figure 11.

3.3.3. Domain formation in honeycomb patterns with $[11]$ easy axis orientation. In section 3.3.1, we noted that the charge-ordered state occurring at the coercive field is surprisingly stable. It can be destroyed by either increasing the field toward saturation, or by applying a reversal field. If the dipole–dipole interaction is responsible for the stability, then one would expect the formation of domains to also occur during the initial magnetization and not only by sweeping through the coercive field. We have thus analyzed the honeycomb patterns with respect to charge $\pm 3$ domain formation starting from the demagnetized state to saturation. In figure 12, an example is shown for a domain state at 600 G along the initial magnetization curve. The domains are highlighted by continuous lines. A histogram of domains is provided in figure 13, which shows the number of domains containing a specified number of $\pm 3$ charge pairs as a function of the external field. At small initial fields small domains dominate containing only one up to three charge pairs. As the field increases a few domains grow to large sizes containing more than 30 charge pairs. This becomes even more pronounced in patterns with 0.4 $\mu$m separation, where domain sizes containing more than 200 charge pairs are found. From this we infer that indeed the $\pm 3$ charge pairs are coupled and form domains.

In section 3.3.2, we have also noted that the formation of a charge-ordered state is unlikely for the $[10]$ hard axis orientation. Therefore, we were curious to explore whether domains of $\pm 3$ charge pairs also form along the initial magnetization curve in $[10]$ orientation. Indeed this is the case for all three patterns. Representative for all patterns, we show in figure 14 the histogram for the 0.8 $\mu$m pattern for better comparison with the $[11]$ orientation. All this indicates that the highly frustrated type-I state appears to stabilize itself by organizing into $\pm 3$ charge pairs analogous to an ionic crystal lattice. This is quite a surprising result, which obviously cannot be observed for patterns with much smaller magnetic dipoles where the formation of Dirac strings prevails [3, 5]. At this point we would like to point out that the observed charge ordered state is a state of very high symmetry. It is characterized by zero charge and zero magnetization per honeycomb ring, while each of the three sublattices is ferromagnetically aligned, yielding a vanishing chirality. Apart from the $\pm 1$ charge order realized in saturation, the $\pm 3$ state is the second and only other configuration that yields a complete order with translational symmetry.
3.4. The role of single-dipole defects

Local defects in the honeycomb lattice may change the configurations of the surrounding vertices. This will shed light on the interaction of the dipoles, and if the disturbance is sizeable, then a healing length can be defined. Therefore, we have analyzed in detail the surroundings of missing dipoles. Missing dipoles frequently occur during pattern fabrication. Before analyzing patterns with defects it is advantageous to label the different neighboring vortices as indicated in figure 15. If $A_1$ and $A_2$ define the location of a dipole that is removed, then $B_1$ to $B_4$ are the nearest-neighbor vertices and $C_1$ to $C_8$ are the next-nearest-neighbor vertices which we will consider in the following analysis. This is sufficient as the correlation will not reach any further. Using this nomenclature we have analyzed the abundance of defects in honeycomb patterns with dipole separations of 0.4, 0.8 and 1.7 $\mu$m and for both [11] and [10] orientations. In particular,
we have compared the abundance of type-I states on B and C sites in patterns with and without defects.

In patterns with a dipole separation of 0.4 µm, we note that the frequency of type-I states on B-sites is reduced if \(A_1-A_2\) contains a defect. On C sites no difference beyond statistical error can be discerned. For the patterns with 0.8 and 1.7 µm separation, the frequency of type-I states on B and C sites is within error bars identical for patterns with and without local defects. This shows that only for patterns with the smallest dipole separation, local defects affect the type-I statistics over a very short range not reaching beyond the nearest-neighbor sites. On the one hand, this finding ensures that patterns with some local defects are still representative of the statistics of more perfect patterns. On the other hand, the slight reduction of type-I states found on B-sites indicates that magnetic flux closure is required to stabilize ±3 vertices.

4. Monte Carlo simulations

In order to understand an astonishing abundance of type-I vertices for all studied interparticle distances, we have performed Monte Carlo simulations of 40 × 40 honeycomb arrays. In the first set of simulations, the particles were regarded as macroscopic magnetic moments, which are allowed to take only two Ising-like orientations determined by the geometry of the array. Similarly to previous calculations [2], the standard Ewald’s summation of dipolar sums and the Gaussian distribution of the switching fields (coercivities) were utilized. In contrast to [2], the variation of interparticle distances was taken into account by considering higher-order magnetic contributions [11, 22, 23].

*New Journal of Physics* 14 (2012) 035015 (http://www.njp.org/)
The dipolar magnetic moment of an individual island was found to be $\mu \approx 1.33 \times 10^{-14}$ A m$^2$, while its octopolar counterpart is of the order of 0.5 $\mu$m. Arrays with the largest interparticle separation of $b = 1.7 \mu$m were approximated as pure dipolar ensembles. For smaller interparticle distances, octopolar contributions were also taken into account. The dipole–dipole interaction between two mutually parallel moments, which are perpendicular to the connection line, is $E_{dd} \approx 0.71 \times 10^{-35}/b^3$ J ($E_{dd} = 1.44 \times 10^{-17}$ J for $b = 1.7 \mu$m).

**Figure 13.** Histogram of domain sizes containing $\pm 3$ charge-ordered pairs for a pattern with a [11] orientation and a dipole separation of 0.8 $\mu$m. The number of charge pairs is plotted versus domain size and external field.

**Figure 14.** Histogram of $\pm 3$ charge-ordered pairs for a pattern with [10] orientation and with a dipole separation of 0.8 $\mu$m. The number of charge pairs is plotted versus domain size and external field.
The average number of ±3 magnetic charges for interparticle distances $b = 1.7 \, \mu m$ and $b = 0.8 \, \mu m$ is shown in figures 16(a) and (b), respectively. In good qualitative agreement with experimental data, we find an increase of the density of magnetic monopoles at coercivity in both arrays. The frequency of magnetic type-I charges in looser packed arrays is larger than in their denser counterparts. However, the density of ±3 states in the calculations is significantly smaller than expected from the experiments and therefore similarities to the Dirac strings observed by Mengotti et al [5] are found, instead of the magnetic charge ordering.

The dipolar interactions in magnetic arrays act on two different scales: the microscopic scale of atomic spins and the macroscopic scale of an array. In the simulations described above, the microscopic magnetic structure of each individual particle is neglected. Instead, the Gaussian distribution of coercive fields was implemented similarly to all previous calculations [2]. The microscopic details of the magnetization switching, however, are of decisive importance for the coercivity of each individual particle. To account for the microscopic magnetization reversal we have performed a second set of simulations. Using the coarse-graining procedure described in [21], each rectangular element of an array was built out of $\approx 1400$ cells (cell size $\approx 25 \, \text{nm}$). The cells are coupled by ferromagnetic exchange as well as by dipolar interaction. The complete honeycomb array possesses $10 \times 10$ rows of macroscopic hexagons, i.e. of the order of $\approx 5 \times 10^5$ dipoles in total with open boundaries. Within this computational scheme we no longer need an artificial distribution of the switching fields. Instead, to account for imperfections in the experimental arrays in some of the calculations a random distribution of magnetic inhomogeneities was included.

The introduction of the microscopic structure of the elements strongly changes the distribution of magnetic charges during the hysteresis. The most striking feature is that the frequency of the ±3 magnetic charges increases to 100% at coercivity in perfect agreement with the experimental findings. The reason for this behavior is hidden in the microscopic mechanism of the magnetization reversal. Due to the microscopic structure of each individual rectangular element the coercive fields are not random any more. Starting with complete saturation at +800 Oe along the easy direction, figure 16(c) shows the magnetization structure of a portion of an array at −350 Oe (close to coercivity). The azimuthal magnetization direction is visualized via
Figure 16. MC simulations of honeycomb arrays. (a, b) Density of ±3 monopoles as a function of applied field in looser packed ($b = 1.7 \, \mu m$) and denser packed ($b = 0.8 \, \mu m$) arrays of macroscopic magnetic moments; (c) a portion of an array where each element consists of 1440 microscopic dipoles. The color scheme denotes the azimuthal orientation of magnetization. The arrows emphasize the homogeneously magnetized rectangles; (d) the microscopic magnetization pattern of the element A, marked in (c). For clarity only each fourth microscopic spin is shown.

The color map. One sees that while the canted elements of the array are still homogeneously magnetized (except for negligible inhomogeneities at the ends of the elements), the horizontal particles are either completely reversed or show domain patterns, i.e. reversal is in course. The inner structure of the bar marked in figure 16(c) is augmented in figure 16(d). The stray fields of the neighboring elements and the external magnetic field induce vorticity at the
ends of the bars which are parallel to the applied field. The vortex transforms into a domain wall, which, in turn, propagates through the bar. The domain wall energy is of the order of $E_{\text{dw}} = S \sqrt{A \cdot K} \approx 3.6 \times 10^{-17}$ J, where $S$ is the area of a domain wall, $A = 21 \times 10^{-12}$ J m$^{-1}$ the exchange stiffness and $K = \mu_0 M_s^2 / 2 = 1.7, \ldots, 1.9 \times 10^6$ J m$^{-3}$ the shape anisotropy energy density [24]. Of course, this energy is identical for any of the elements within the array. However, the energy developed by the magnetic field is proportional to the angle it makes with the magnetization. Therefore, the field needed to overcome the barrier $E_{\text{dw}}$ is lower for particles oriented parallel to the easy [11] direction. Hence, the coercive field of those particles is also lower and they switch first. These results are in very nice agreement with the experimental findings. Even the artificial introduction of inhomogeneities does not change the magnetization reversal. We always find a perfect ordering of the type-I vertices at $H \approx 400$ Oe.

5. Conclusion

We have fabricated by lithographic means honeycomb patterns decorated with magnetic dipoles. The dipole configurations were imaged by MFM as a function of several parameters: the external field and orientation of the patterns with respect to the field direction, referred to as the easy axis and hard axis orientation, respectively. The patterns are distinguished by their distances between the dipoles in each vertex, which are 0.4, 0.8, or 1.7 µm. Furthermore, we have analyzed the effect that local defects have on the distribution of type I states. Unlike honeycomb patterns that are investigated by other groups, we noted for all our patterns a high abundance of type-I states with three dipoles pointing either in or out of a vertex. The abundance of type-I states is particularly high for patterns with easy axis orientation at coercive field values. In fact, at coercivity the high abundance of type I states results in a charge order of alternating ±3 charges. Type-I states violate the spin-ice rule and vertices with type-I configurations, representing magnetic monopoles, are points of high energy. Nevertheless, we find that the charge ordered state is rather stable and does not easily decompose by removing the field. This high stability and the fact that the charge ordered state forms domains let us speculate that alternating magnetic monopoles condense into an ordered state with enhanced dipole–dipole interaction between the vertices with ±3 charges.

Monte Carlo simulations shed more light on the distribution of the vertex states in the honeycomb lattices. The high abundance of type-I states at coercivity can be reproduced. But much better agreement is found when taking the microscopic dipole interaction during the reversal process into account. In fact, we identify the competition between the microscopic dipolar interactions within individual elements and the macroscopic dipolar coupling between the elements as the physical reason for the formation of a charge ordered state at coercivity, i.e. an ordering of magnetic monopoles.

Acknowledgments

The authors are grateful to Björn Sothmann for support in data evaluation procedures and to Peter Stauche and Jürgen Podschwadew for technical support. We thank the Deutsche Forschungsgemeinschaft for financial support of this work within the SFB 491. EV acknowledges support from Deutsche Forschungsgemeinschaft (SFB 668) and the Cluster of Excellence ‘Nanospintronics’.
References

[1] Wang R F, Nisoli C, Freitas R S, Li J, McConville W, Cooley B J, Lund M S, Samarth N, Leighton C, Crespi V H and Schiffer P 2006 Nature 439 303
[2] Mengotti E, Heyderman L J, Fraile Rodriguez A, Bisig A, Le Guyader L, Nolting F and Braun H B 2008 Phys. Rev. B 78 144402
[3] Ladak S, Read D E, Perkins G K, Cohen L F and Branford W R 2010 Nature Phys. 6 359
[4] Morgan M J P, Stein A, Langridge S and Marrows C H 2011 Nature Phys. 7 75
[5] Mengotti H E, Heyderman L J, Rodriguez A F, Nolting F, Hugli R V and Braun H B 2011 Nature Phys. 7 68
[6] Pauling L J 1935 Am. Chem. Soc. 57 2680
[7] Morris D J P et al 2009 Science 326 411
[8] Fennell T, Deen P P, Wildes A R, Schmalzl K, Prabhakaran D, Boothroyd A T, Aldus R J, McMorrow D F and Bramwell S T 2009 Science 326 415
[9] Kadowaki H, Doi N, Aoki Y, Tabata Y, Sato T J, Lynn J W, Matsushira K and Hiroi Z 2009 J. Phys. Soc. Japan 78 103706
[10] Castelnovo C, Moessner R and Sondhi S L 2008 Nature 451 42
[11] Remhof A, Schumann A, Westphalen A, Zabel H, Vedmedenko E Y, Mikuszeit N, Last T and Kunze U 2008 Phys. Rev. B 77 134409
[12] Zabel H, Schumann A, Westphalen A and Remhof A 2009 Acta Phys. Pol. A 115 59
[13] Tanaka M, Saitoh E, Miyajima H, Yamaoka T and Iye Y 2006 Phys. Rev. B 73 052411
[14] Qi Y, Brintlinger T and Cumings J 2008 Phys. Rev. B 77 094418
[15] Mellado P, Petrova O, Shen Y and Tchernyshyov O 2010 Phys. Rev. Lett. 105 187206
[16] Schumann A, Sothmann B, Szary P and Zabel H 2010 Appl. Phys. Lett. 97 022509
[17] Wang R F et al 2007 J. Appl. Phys. 101 09J104
[18] Ramirez A P, Hayashi A, Cava R J, Siddharthan R and Shastry B S 1999 Nature 399 333
[19] Möller G and Moessner R 2009 Phys. Rev. B 80 140409
[20] Wills A S, Ballou R and Lacroixet C 2002 Phys. Rev. B 66 144407
[21] Vedmedenko E Y 2007 Phys. Status Solidi b 244 1133
[22] Vedmedenko E Y and Mikuszeit N 2008 Chem. Phys. Chem. 9 1222
[23] Westphalen A, Schumann A, Remhof A, Zabel H, Karolak M, Baxevanis B, Vedmedenko E Y, Last T and Kunze U 2008 Phys. Rev. B 77 174407
[24] O’Handley R C 2000 Modern Magnetic Materials (New York: Wiley)