

Controlling magnetic charge landscapes in nanostructures

Novel approaches to nanomagnetic writing

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Abstract

New tools for the fine control and manipulation of magnetic charge landscapes are developed and presented. Scanning probe systems fitted with magnetic tips are used to write 360° domain walls, complex topological defects, and entire system-wide microstates in nanomagnetic arrays including artificial spin ice. These new nanomagnetic writing techniques open previously inaccessible regions of the microstate space in artificial spin ice, including states exhibiting negative effective temperatures and the elusive long-range ground state of kagome artificial spin ice.

0.1 Declaration of originality

I declare that the contents of this thesis are my own work and contain no material which has been previously published or written by another person, or substantial proportions of material which have been accepted for the award of any other degree or diploma at Imperial College London or any other educational institution, except where acknowledged. All figures and data reproduced from the published work of other persons are explicitly referenced as citations and, where necessary, the appropriate copyright and intellectual property permissions have been obtained and stated as such. All contributions to this thesis which have been carried out by others, with whom I have worked at Imperial College or other institutions, have been clearly acknowledged as such. The intellectual content of this thesis is the product of my own work, except where assistance from others in the conception, design and vital discussions, leading to its claims and conclusions, in addition to assistance to the presentation and style of the thesis, has been acknowledged.

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0.4 Publications and awards

0.4.1 Publications

J. C. Gartside, D. M. Arroo, D. M. Burn, V. L. Bemmer, A. Moskalenko,
L. F. Cohen and W. R. Branford.

‘Realising the kagome ice ground state and thermally inaccessible states via topological defect-driven magnetic writing’

Nature Nanotechnology (2017)

doi: 10.1038/s41565-017-0002-1

J. C. Gartside, D. M. Burn, L. F. Cohen and W. R. Branford.

‘A novel method for the injection and manipulation of magnetic charge states in nanostructures’

Scientific Reports **6** 32864 (2016)

doi: 10.1038/srep32864

0.4.2 Awards

Anne Thorne Prize for best PhD thesis concerning development of new experimental techniques,
Imperial College London 2018

Best poster presentation, Joint European Magnetic Symposium Glasgow 2016

Nature Physics best poster presentation, IOP magnetism Sheffield 2016

Best oral presentation, Imperial College London post-graduate research symposium 2016

Best poster presentation, International conference on magnetism Barcelona 2015

Best poster presentation, Imperial College London post-graduate research symposium 2015

1 | Introduction

1.1 Motivation

Gaining enhanced control over the magnetic charge landscapes of nanopatterned systems has long been a goal of the nanomagnetic research community. The abstracts and introductions of literature in the field are littered with allusions to the rewards offered if one could freely sculpt and manipulate distributions of magnetic charge in complex nanostructures. These rewards include realising low-power, state-persistent reconfigurable magnonic crystals²⁻¹² - vital functional elements for the nascent field of magnonics - and functional processing units for neuromorphic computation¹³⁻²⁰ - a potential means to circumvent the von Neumann bottleneck²¹ limiting conventional CMOS logic systems. However, the lack of simple, powerful means with which to manipulate magnetic charge landscapes has kept these outcomes out of reach, severely limiting the utility and fundamental scope of nanopatterned magnetic systems.

One nanomagnetic system consistently providing fertile ground for researchers is the family of geometrically-frustrated magnetic arrays termed ‘artificial spin ices’ (ASI). Since its 2006 inception¹³ ASI has played host to several important fundamental physical studies, investigating the dynamics and origin of phenomena such as ground-state degeneracy and corresponding zero-point entropy in ice-like systems^{2,14,22,23}, magnetic monopole-like quasi-particles and their associated ‘Dirac strings’²⁴⁻³⁰ and system-wide statistical behaviours including exotic spin-gas to spin-liquid or spin-solid phase transitions^{2,10,31-36}. The majority of these phenomena emerge from the complex magnetic charge landscape and vast microstate space of ASI, borne out of the

lack of a well-defined ground state (a characteristic of geometrical frustration).

The above mentioned studies have lead to a great deal of understanding concerning the huge host of possible micromagnetic configurations (equivalent to magnetic charge distributions) populating the ASI microstate space. This knowledge has prompted the research community to begin to cast its gaze beyond purely fundamental studies and identify hypothetical ASI-based applications, leveraging the rich microstate space to potentially provide impressive functionality. Speculative theoretical work has been performed on harnessing collective excitations of periodic spin-wave oscillations throughout the system, with the exact frequency response or ‘magnonic band structure’ finely tunable via modifying the magnetic charge landscape. Several groups have also noted the potential of ASI to provide the underlying system for hardware realisations of neural-networks and neuromorphic computation. Critically however, these designs hinge on the ability to freely reconfigure the magnetic charge landscape, switching the orientation of any single magnetic element within the ASI array at will without altering the state of other elements. In fact the need for a capable nanomagnetic writing solution extends beyond applied purposes, fundamentally-relevant microstates such as the ‘spin-crystal’ long-range ground state of kagome (hexagonal) ASI have so-far eluded repeated attempts at realisation from a number of research groups. Without complete microstate-writing capability, extracting advanced functionality from ASI remains a theoretical curiosity.

At the outset of this thesis, the writing requirement was not well-met. State writing was accomplished through either the application of globally-uniform saturating magnetic fields - accessing a mere handful of possible states - or thermalisation protocols randomly orienting all elements within the array - unsuitable for repeatable studies or intricate low-entropy states. If real progress is to be made in ASI and related nanopatterned magnetic arrays, I recognized that this gap in the experimental toolset available to the research community needed to be urgently addressed.

1.2 Goals and challenges

This thesis sets out to resolve the issue set out above, providing the community with powerful, flexible tools for the fine manipulation of magnetic charge distributions. It is my intention that any tools created must stand a real chance of being adopted by the wider research community. It is still an admirable scientific effort to introduce methodologies requiring expensive, hard-to-access equipment or extensively modified homebrew systems, difficult to replicate without specialist knowledge, but these techniques more rarely find favour outside of their ‘home’ research group. In light of this, this thesis will take the approach of developing techniques using only non-modified, widely available equipment.

The thesis aims to create tools satisfying the following conditions: they substantially broaden the accessible region of the total microstate space - ideally opening the entire range of states, they require no specialist or modified equipment which may prove a barrier to access, they must have a relatively low level of experimental complexity and must operate on a reasonable time-scale such that researchers may prepare and analyse multiple states within a single experimental session.

Of course, it was not clear if it would be possible to meet all (or indeed any) of these conditions, but these are the goals which I set myself in order to direct the research. If these criteria were satisfied and it did prove possible to develop a nanomagnetic writing system, the final goal was to employ the new writing method to realise previously unobserved (or indeed inaccessible) states of fundamental or applied interest.

1.3 Thesis overview

Over the course of this thesis, the reader will see that the endeavour to develop novel magnetic writing methodologies was a success. After a description of the key physical concepts underpinning the thesis (chapter 2) and a discussion of the experimental and theoretical methods used (chapter 3), chapter 4 will introduce the first original writing technique I developed - a

method for inserting bundles of magnetic charge (360° domain walls) into nanostructures. This is followed in chapter 5 by a description of the second writing technique developed, a tool for the complete reconfiguration of magnetic charge landscapes in nanopatterned magnetic systems. The technique detailed in chapter 5, ‘topological defect-driven magnetic writing’ (TMW), is able to access the entire ASI microstate space. We demonstrate its capabilities by realising two significant microstates: the ‘spin-crystal’ long-range ordered ground state of kagome ASI and thermally-inaccessible high energy, low entropy states exhibiting negative effective temperatures.

2 | Key concepts

2.1 Micromagnetics

The work in this thesis focuses on the local manipulation of micromagnetic spin textures, so a discussion of the energetics responsible for stabilising these textures is in order. The total micromagnetic energy ϵ_{tot} of a spin system is given by the following expression (after Coey³⁷):

$$\epsilon_{tot} = \epsilon_{ex} + \epsilon_d + \epsilon_a + \epsilon_Z + \epsilon_{me} \quad (2.1)$$

The five terms refer respectively to the exchange, demagnetisation, magnetocrystalline anisotropy, Zeeman and magnetoelastic energies. The first three terms are intrinsic to ferromagnets and always present to some degree, while the Zeeman and magnetoelastic energies are responsive energies to applied magnetic field (ϵ_Z) and strain (ϵ_{me}).

Before describing the origin and effects of each term in turn, we will briefly describe their significance to this thesis. The exchange and demagnetisation terms are the key drivers behind stabilising the various spin textures considered in this thesis, which are generally determined by minimising $\epsilon_{ex} + \epsilon_d$. The Zeeman energy is crucial in this thesis in determining how the field of an MFM tip may locally upset the balance between ϵ_{ex} and ϵ_d , introducing new defects to the spin texture in the process. The majority of systems considered here have negligible contributions from ϵ_a and ϵ_{me} due to the low magnetocrystalline anisotropy of $\text{Ni}_{81}\text{Fe}_{19}$ and may

be ignored in the majority of cases.

2.1.1 Exchange energy

The exchange energy is a purely quantum mechanical phenomenon which may be seen as the root cause of ferromagnetism. At its core, it is driven by the Pauli exclusion principle (PEP) forbidding indistinguishable fermionic particles from occupying the same quantum state. For our purposes, these indistinguishable fermions are the electrons in our metallic magnetic structures, with their quantum state described by their spin vector and spatial position. To satisfy the PEP between electrons we must either have matching spins and differing spatial positions, or unaligned spins and similar spatial positions. This means that for electrons with aligned spin vectors, it is energetically favourable for them to be well-spaced out as far as the system permits. We can now consider another interaction between electrons on neighbouring atoms in a metallic crystal, the electrostatic repulsion between like-charges. When arranging electrons on atoms in crystal lattice, the electrostatic energy is minimised by maximising the separation between their spatial charge distributions. In the case of the 3d transition metals considered in this thesis (Fe, Ni and Co) the overlap between the electric charge distributions of partially filled d-orbitals on neighbouring atoms is reduced by aligning the spins of unpaired electrons in the d-orbitals, forcing the electrons to remain further apart due to the PEP forbidding electrons with matching spin from occupying the same spatial position. This alignment of spins propagates through the metal and leads to ferromagnetic spin textures, with the energy saved by aligning the spins termed the exchange energy.

The classic Heisenberg Hamiltonian describing the exchange energy of two spins is

$$H = -2J\hat{S}_1 \bullet \hat{S}_2 \tag{2.2}$$

where J is the material-dependent exchange constant with units of energy. This may be

extended across a many site lattice containing j sites as

$$H = -2 \sum_{i \rightarrow j} J_{ij} \hat{S}_i \bullet \hat{S}_j \quad (2.3)$$

When dealing with the smoothly varying magnetisation textures observed in the thin ferromagnetic nanostructures considered in this thesis, it is generally preferable to replace the picture of discrete spins on a fixed lattice with that of a continuous magnetisation texture $\mathbf{M}$. In this case we may write the exchange energy in integral form as

$$\epsilon_{ex} = \int A (\nabla \mathbf{e}_M)^2 d^3r \quad (2.4)$$

where $\mathbf{e}_M$ is a unit vector denoting the local magnetisation direction, defined as $\mathbf{e}_M = \mathbf{M}(\mathbf{r})/M_s$, and A is the exchange stiffness. The exchange stiffness may be approximated from the Curie temperature T_C as $A \approx k_B T_C / 2a_0$ where a_0 is the lattice parameter. A microscopic picture of the exchange stiffness is given by the expression

$$A = \frac{J \langle S^2 \rangle Z_c}{a} \quad (2.5)$$

where J is the exchange constant, Z_c is the number of spins per unit cell and a the lattice parameter. The exchange stiffness of Ni₈1Fe₁9 is $A = 10$ pJ/m, about half that of Fe and a third of Co. The exchange stiffness A and saturation magnetisation M_s may be combined to give the exchange length

$$l_{ex} = \sqrt{\frac{2A}{\mu_0 M_s^2}} \quad (2.6)$$

l_{ex} is a useful parameter for understanding how the exchange energy shapes micromagnetic structures, it describes the shortest length across which magnetisation may be twisted without

incurring very high exchange energy penalties. Regions of magnetic material with radius $r \leq l_{ex}$ may be assumed to have a near-isotropic magnetisation, allowing for computationally-cheap micromagnetic simulation where magnetic samples are discretised into cells with side-length $a \sim l_{ex}$. Py has an exchange length of $l_{ex} \sim 5$ nm.

In a 2D x,y spin system, which the ~ 10 nm thick structures studied in this thesis are to good approximation, this simplifies to

$$\epsilon_{ex} = \int A(\nabla\theta)^2 dxdy \quad (2.7)$$

where θ is the angle between spins. The exchange energy penalty for sharp variations in the magnetisation is high, favouring collinear spins or smooth, gradually rotating spin textures. This energy cost leads to the spontaneous alignment of spins throughout regions of material that we call ferromagnetic.

2.1.2 Demagnetisation energy

We have seen that the exchange energy favours a spin system where all spins point in the same direction. However, we know that in real systems this isn't always the case. One may 'demagnetise' a ferromagnet by subjecting it to alternating or rotating field protocols, and ferromagnetic materials will often spontaneously assume multi-domain states where the magnetisation of each domain isn't aligned with its neighbours. To understand this, we can consider the basic relation between B , H and M

$$\mathbf{B} = \mu_0(\mathbf{H} + \mathbf{M}) \quad (2.8)$$

Taking the divergence of both sides and using Maxwell's 'no magnetic monopoles' relation

$\nabla \bullet \mathbf{B} = 0$, we arrive at the relation

$$\nabla \bullet \mathbf{H} = -\nabla \bullet \mathbf{M} \quad (2.9)$$

From this we can see that within the volume of a magnetic sample where $\mathbf{M} \neq 0$, there must exist some finite $\mathbf{H}$ field to satisfy the above relation. This is termed the ‘demagnetising field’ $\mathbf{H}_d$ and takes the form

$$\nabla \bullet \mathbf{H}_d = -\nabla \bullet \mathbf{M} \quad (2.10)$$

The corresponding demagnetisation energy ϵ_d may be written as

$$\epsilon_d = -\frac{1}{2} \int \mu_0 \mathbf{H}_d \bullet \mathbf{M} \, d^3r \quad (2.11)$$

if considering the volume of the magnet itself, or

$$\epsilon_d = \frac{1}{2} \int \mu_0 H_d^2 \, d^3r \quad (2.12)$$

over all space.

2.1.3 Magnetic charge (or pole) avoidance

We can see from the H_d^2 dependence that demagnetisation field is to be avoided as far as possible. The largest source of demagnetisation field is ‘magnetic surface charge’ $\mathbf{M} \bullet \hat{\mathbf{e}}_n$, where $\hat{\mathbf{e}}_n$ is the surface normal. Magnetic charge is akin to magnetic poles, but typically a more convenient term especially when systems may contain a multitude of poles rather than the familiar ‘north’ and ‘south’. North and south poles may be thought of as essentially positive and

negative magnetic charges. Wherever possible, $\mathbf{M}$ will avoid surface charge by lying parallel to the surface rather than pointing out of the magnetic material. In some cases it is possible to have zero surface charge throughout the sample by fracturing the magnetisation into several ‘flux-closure’ domains, but in smaller nanostructures (such as the ones studied in this thesis) some surface charge is typically unavoidable. Figure 2.1 shows two potential magnetisation configurations in a thin rectangular ferromagnet, a single domain (a) and flux-closure domain pattern (b). In a system with only exchange energy, the single domain state is energetically favourable as there is no angular variation between spins. However, once demagnetisation energy is introduced there is a large penalty for the magnetic charges at either end of the structure length. The demagnetisation energy favours a multi-domain state as shown in fig. 2.1 b), and for large enough structures the magnetisation will spontaneously fracture into such a state to balance the exchange and demagnetisation energies. For Py, the crossover between single domain and flux closure states occurs at structure dimensions of $2\mu\text{m}$ ³⁸. Single-domain states may in theory adopt any magnetisation direction relative to the host structure length. However, in practice simple ferromagnetic materials such as Py or Co will always orient their single-domain magnetisation parallel to the structure long-axis to minimise surface charge unless forced otherwise by an externally applied field. Figure 2.1 c) demonstrates the large amount of surface charge arising from a magnetisation perpendicular to the structure width. This tendency for M to lie along the length of structures is known as ‘shape anisotropy’.

Multi-domain states are characterised by discrete magnetisation regions of magnetisation, typically with large angles between the magnetisation of adjacent regions. For instance, the two long domains in fig. 2.1 b) are rotated 180° relative to each other and 90° relative to the shorter domains at the structure ends. As we have seen, the exchange energy abhors an abrupt rotation in magnetisation and as such, adjacent domains are separated by distinct spin-textures known as ‘domain walls’ (DWs). While fracturing the magnetisation into multiple domains may reduce the magnetic surface charge $\mathbf{M} \cdot \hat{\mathbf{e}}_n$, the domain walls themselves may possess magnetic volume charge $\nabla \cdot \mathbf{M}$ which carries its own associated demagnetisation energy penalty. Whether multi-domain states will form is decided by balancing the volume charge creation cost against the surface charge reduction benefit.

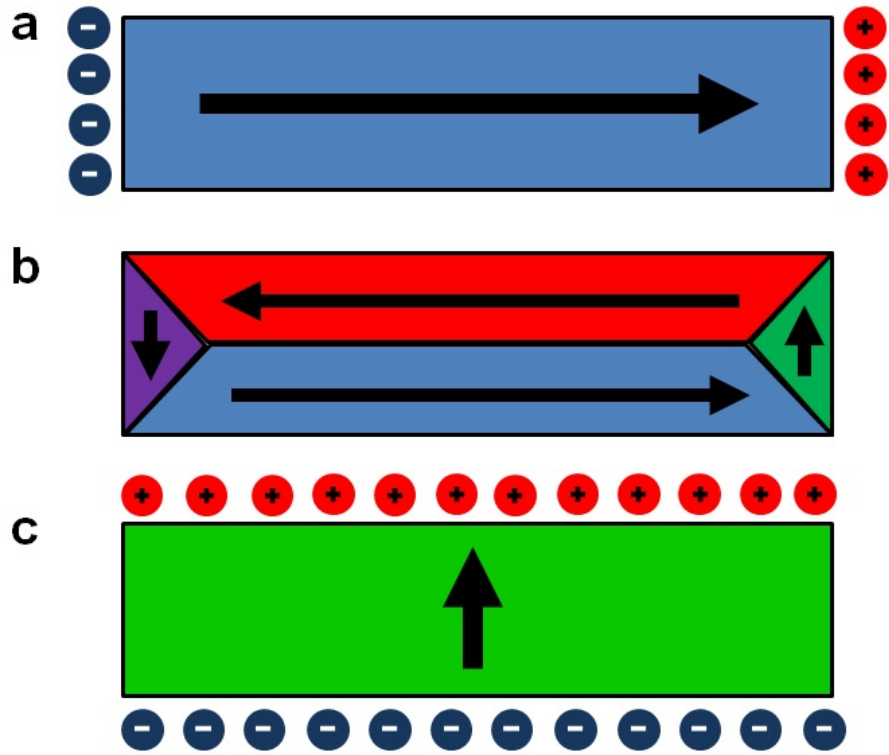


Figure 2.1: Single domain (a) and flux-closure (b) magnetisation states of a thin rectangular slab of ferromagnet. The single domain state has opposite polarity magnetic poles, or magnetic surface charges, at either end while the flux closure state has no magnetic surface charge. (c) Single-domain state with magnetisation perpendicular to structure length. Highly unfavourable due to large amount of surface charge.

For the nanostructures considered in this thesis, film thicknesses range from $\sim 8 - 15$ nm and structure widths from $\sim 20 - 200$ nm. For these spatial dimensions, the exchange energy penalty of spontaneously forming DWs is higher than the demagnetisation energy cost of having magnetic charges at the structure ends. This is due to the magnetisation being forced to rotate sharply across small lengthscales, and leads to structures assuming single domain states unless DWs are otherwise forced into them.

2.2 Domain walls

Creating, manipulating and exploiting DWs within magnetic nanostructures are the core aims of this thesis, so a proper discussion of them is useful.

Domain walls come in a few configurations, which one is energetically favourable in a ferromagnetic element is typically decided by the spatial dimensions of the element. The magnetisation rotation in a DW may occur parallel or perpendicular to the plane of the DW. These DWs are termed Néel (perpendicular rotation) and Bloch (parallel). For Py, structures thinner than 30-60 nm support Néel walls with Bloch walls found in thicker structures^{37,39}. Bloch walls have no associated magnetic volume charge, but contain a surface charge as the magnetisation is forced to rotate out of the plane of the sample between domains. Néel walls contain a volume charge, but no surface charge along the length of the wall. However, where Néel walls meet sample edges some surface charge exists as stray field is projected out of the magnetic material. This becomes more significant in nanopatterned samples (especially thin nanowires) where the sample edges form a large fraction of the total sample.

The DWs in this thesis are all ‘transverse’ Néel walls, meaning the magnetisation rotates monotonically between adjacent domains, remaining in the plane of the sample while doing so as shown in fig. 2.2^{37,40}. A more complex form of Néel wall known as a ‘vortex’ wall is also possible, and is energetically favourable when the relation $\frac{wt}{l_{ex}^2}$ between the structure width w , thickness t and exchange length l_{ex} exceeds 100³⁷. The nanopatterned structures considered in this thesis are too narrow to support multiple parallel domains so DWs always span the width of

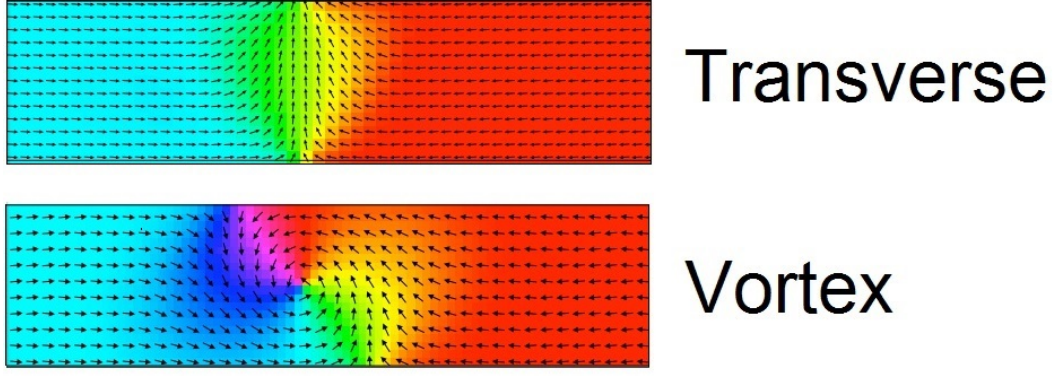


Figure 2.2: Micromagnetic structure of transverse and vortex configuration Néel domain walls.

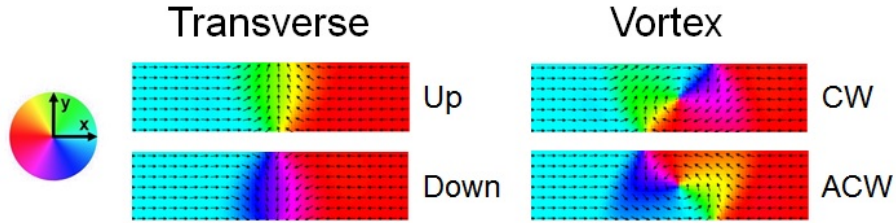


Figure 2.3: Micromagnetic structure of transverse and vortex configuration Néel domain walls showing both possible chiralities for each DW type.

nanostructures rather than the length. The magnetisation is constrained to lie along the nanostructure length by surface charge avoidance (also termed ‘shape anisotropy’ when driven by the dimensions of a patterned sample) so adjacent domains are magnetised anti-parallel to each other and separated by 180° DWs. Micromagnetic structures of transverse and vortex 180° Néel wall configurations are shown in figure 2.2, with both possible chiralities shown in fig. 2.3.

The magnetic charge magnitude and polarity of DWs may be understood from a simple toy model treating each domain as a separate ‘bar’ magnet. The magnetic charge associated with the structure ends of a single-domain magnet is given by $q_m = \frac{M}{l}$, or $q_m = \frac{M_s V}{l} = M_s w t$ where M_s is the saturation magnetisation, V the volume and w, t, l the width, thickness and length respectively. Figure 2.4 a) shows the position of negative (positive) polarity magnetic charges at magnetisation ‘tails’ (heads) in single domain magnets. When combining multiple domains, the magnetic charges sum at the domain wall. A ‘tail-to-tail’ DW has a magnetic charge of $-2q_m$, as seen in fig. 2.4 b). These magnetic charges may be treated in many ways as analogous to electrical charges, they are sources and sinks of H field and exert Coulombic attractive and repulsive forces on each other.

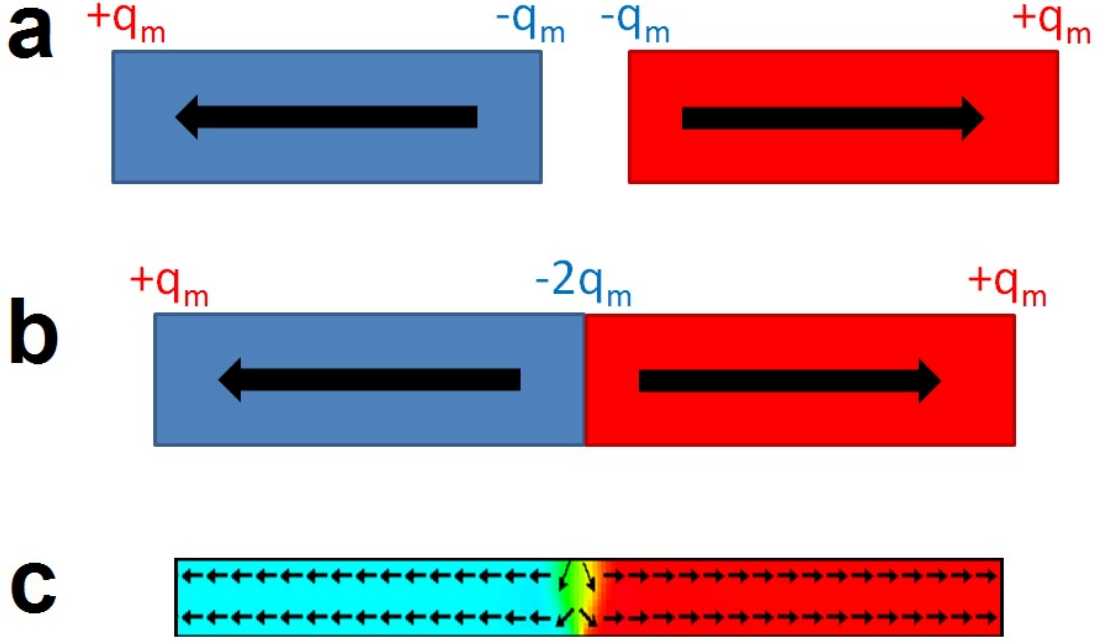


Figure 2.4: Schematic of magnetic charge polarity and magnitude in single domain magnets and DWs.
a) Separate single domain magnets with anti-parallel magnetisation showing location and polarity of magnetic charge q_m .
b) Single magnet consisting of two domains separated by ‘tail-to-tail’ DW.
c) Micromagnetic picture of two-domain state shown in b).

Composite structures comprising of multiple bound DWs are possible and one such structure, the 360° DW, is intrinsic to this thesis. As shown in figure 2.5, adjacent 180° DWs must always have opposite polarity magnetic charge. This leads to a Coulombic attraction between the DWs and they will move towards each other, at which point the chirality of the walls becomes important. Two 180° DWs of the same chirality will annihilate on contact, smoothly unwinding into a collinear state and lowering exchange and demagnetisation energy in the process. If the DWs are of opposite chirality, any attempt to rotate the magnetisation such that the DW unwinds serves to increase the angle between spins within the DW, increasing exchange energy and preventing annihilation. This causes the two DWs to form a metastable bound state and act as a single coherent quasiparticle.

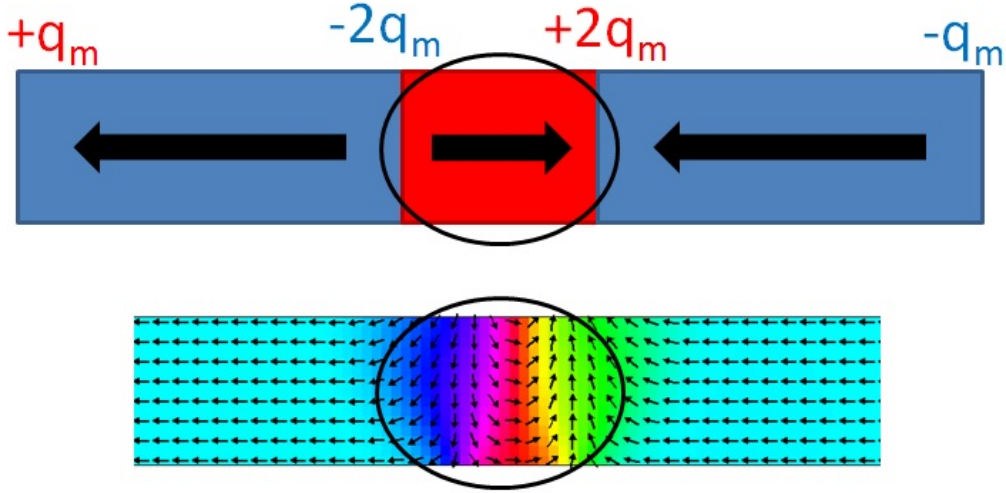


Figure 2.5: Schematic and micromagnetic structure of 360° DW showing magnetic charges.

2.3 Topological charge or winding number

So far we have considered DWs as bundles of magnetic charge, but from our brief discussion of 360° DWs we can see that charge alone does not provide an adequate description for understanding DW behaviour. Additional properties such as the chirality play an important role in determining DW dynamics, such as deciding whether adjacent DWs will form a stable bound state or annihilate each other. The internal structure of a DW is non-trivial and a clear means of understanding it is vital for any detailed micromagnetic work. One may glean a lot of information from looking at micromagnetic magnetisation maps of the type produced by simulation packages such as OOMMF or mumax, but it is useful to have a simple and rigorous language in which to define and discuss the fine structure of DWs and other related spin textures.

The language of topological charges⁴¹, or winding numbers, provides an elegant framework in which to describe DWs. In an otherwise smooth spin system, any point where the magnetisation locally diverges is considered a ‘topological defect’ and may be assigned a winding number describing exactly how the spin texture diverges at that point. The net winding number of a closed spin system is rigorously conserved, and by counting the winding numbers of all topological defects, either in static states or at various stages of dynamic processes, one can get a good feel of how DWs form, annihilate and are stabilised.

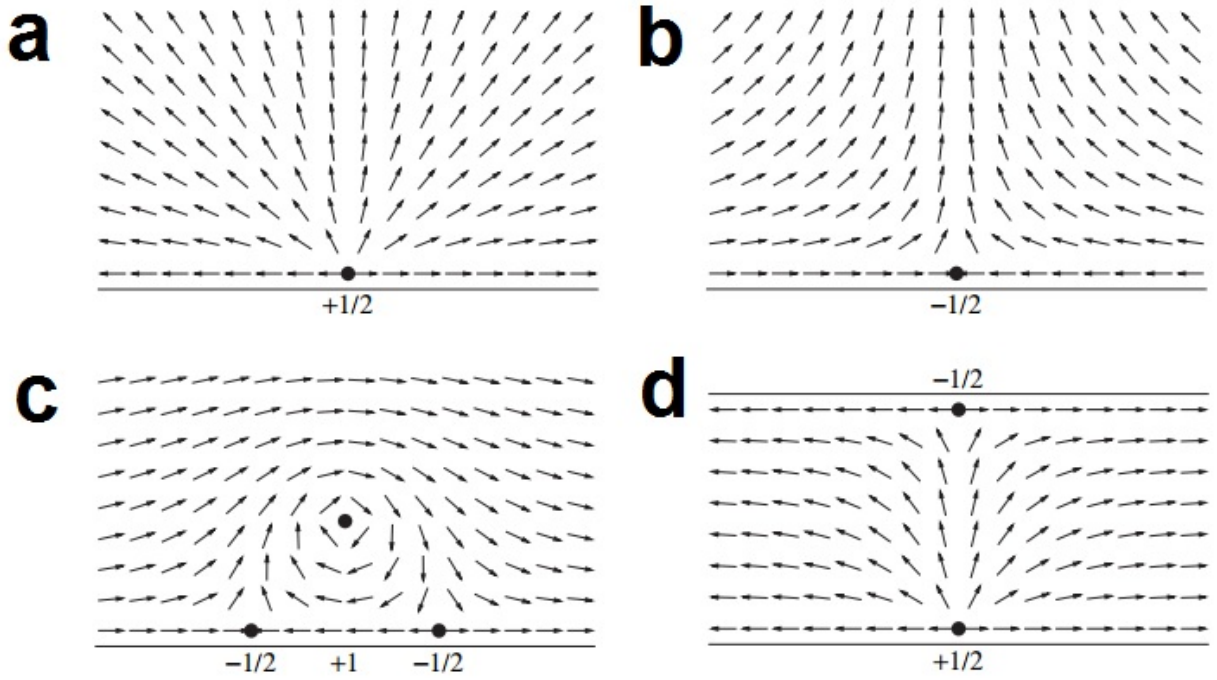


Figure 2.6: Illustration of various topological defects in a nanomagnetic spin system.

a) edge-bound $+1/2$ topological defect

b) edge bound $-1/2$ topological defect

c) $+1$ vortex topological defect in the bulk of the magnet. The winding number of the vortex is balanced by the formation of two edge-bound $-1/2$ defects.

d) pair of $\pm 1/2$ defects joined by a continuous line of spins. This configuration is a 180° transverse Néel DW.

Figure adapted from¹.

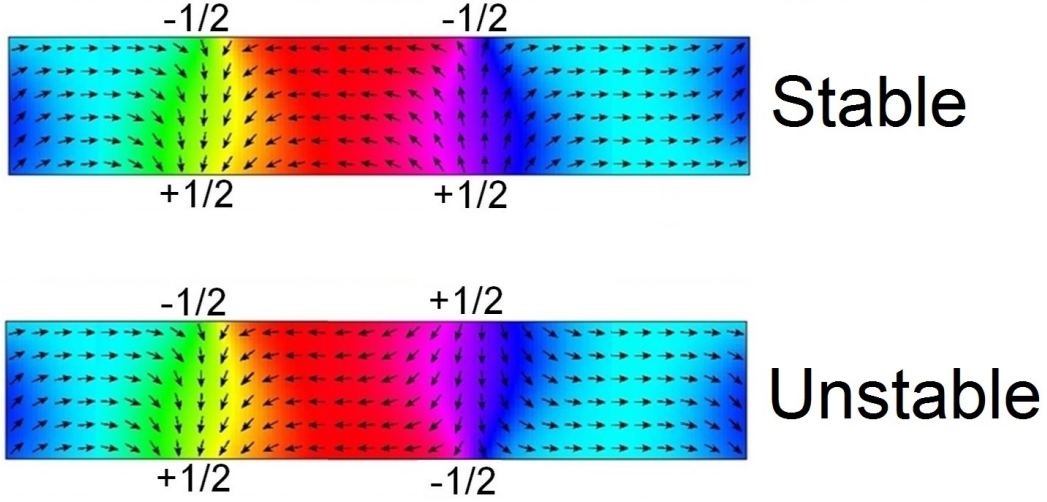


Figure 2.7: Two possible chirality configurations for adjacent 180° DWs, leading to a stable 360° DW or mutual DW annihilation.

Tchernyshyov and Chern provide an excellent treatment of how topological charge may be applied in a nanomagnetic context to describe DWs¹ and the approach used here follows from their work. Let us first consider the basic topological defects which arise in spin systems, defects with winding numbers of $\pm 1/2$ and ± 1 . Figure 2.6 a)-b) shows the micromagnetic structure of the half-integer defects. Defects with half-integer winding numbers may only exist on the system edges; bringing them into the bulk would create a discontinuity or ‘tear’ in the magnetisation along a line between the defect and the nearest system edge, carrying a high exchange energy penalty^{1,42,43}. Figure 2.6 c) shows a $+1$ winding number defect (termed a ‘vortex’) in the bulk of a system. The net winding number of a system is conserved and as such, the introduction of the vortex is balanced by two $-1/2$ winding number defects bound to the system edge (the additional balancing defects in panels a) and b) are out of shot). Figure 2.6 d) shows how multiple topological defects may be combined into stable micromagnetic structures, in this case an 180° ‘up’ chirality transverse Néel DW. It is comprised of a bound pair of $\pm 1/2$ defects, connected by a continuous chain of spins. The chain is formed to prevent a magnetisation discontinuity between the edge-bound defects and has an exchange cost per unit length¹. This causes the edge-bound defects to orient themselves directly opposite each other, minimising the connecting spin-chain length. By reversing the position of the positive and negative edge defects, the chirality of the DW is also reversed. The DW chirality may be considered a shorthand for the relative orientation of the edge defect polarity.

The tendency of the opposite polarity defects to minimise distance the distance between them can be treated as a quasi-Coulombic ‘topological attraction’⁴⁴. This picture can be helpful in understanding the formation of more complex micromagnetic structures, such as the 360° DW mentioned above. Figure 2.7 shows two possible arrangements of adjacent 180° DWs in a nanowire. The ‘stable’ arrangement has a down-chirality DW (left) next to an up-chirality DW, while the unstable arrangement contains two DWs of matching chirality. As described above, the adjacent DWs must be of opposite polarity magnetic charge, and will feel a Coulombic attraction. In the stable case, the topological defects have matching winding number on each edge. This leads to a ‘topological repulsion’ which prevents the DWs from unwinding into each other and stabilises a 360° DW. In the unstable case, the topological defects have opposite polarity winding numbers on each edge and a topological attraction is added to the magnetic charge attraction, bringing the defects into direct contact and causing mutual annihilation. By reversing the position of the positive and negative edge defects, the chirality of the DW is also reversed. One can think of DW chirality as a shorthand for the relative orientation of the edge defect polarity.

The net winding number n_{net} of a system containing ‘ g ’ holes (g is chosen for ‘genus’, the topological terminology for a hole in a surface) is given by

$$n_{net} = 1 - g \quad (2.13)$$

The systems considered in this thesis are generally formed of nanowires, rectangular slabs of material with no hole - hence $n_{net} = 1$. Working out where the topological defects are in a nanowire containing no DWs may be initially confusing, the answer lies in the fine structure of the magnetisation at the wire ends. Figure 2.8 shows how the asymmetric magnetisation states in the corners of the nanowire provide two separate contributions of winding number $+1/2$. In the case illustrated in fig. 2.8 the $+1/2$ defects are in opposite corners, a configuration known as an ‘s-state’. The $+1/2$ defects may also both occur in corners on the same edge of the wire, which is termed a ‘c-state’^{45–47}.

The methodology behind assigning a winding number to an arbitrary topological defect can often

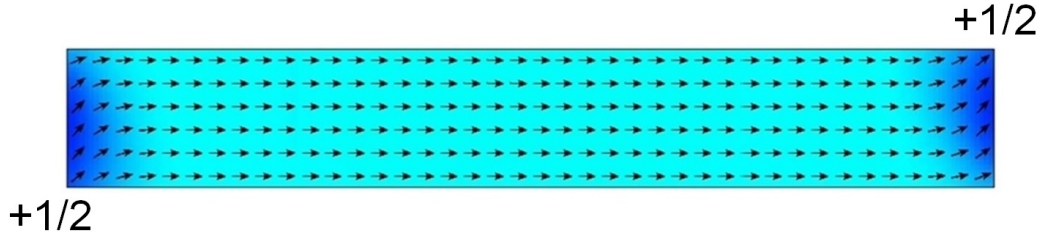


Figure 2.8: Magnetisation structure of a single domain nanowire. Half-integer topological defects at wire ends are marked with their winding numbers.

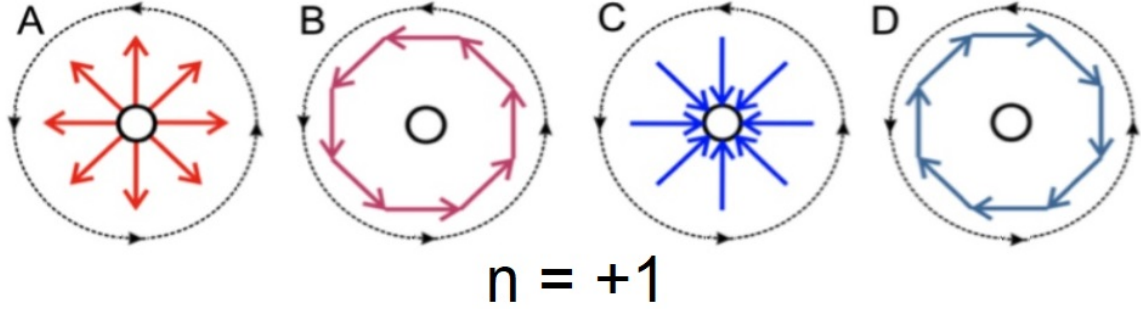


Figure 2.9: Various magnetisation textures containing $n = +1$ topological defects. Solid coloured arrows represent magnetisation direction, dashed black line represents direction of integral path around defect. In all four cases, the angle between the integral path direction (here CCW-pointing small black triangles) and the magnetisation texture remains constant at all points along the integral path. Adapted from⁹³.

seem opaque to those approaching it for the first time. For an excellent in-depth treatment which avoids overly-specialised terminology, the reader may refer to the supplementary information of ‘Domain Wall Trajectory Determined by its Fractional Edge Defects’ (Pushp, Parkin et al, Nature Physics, 2013)⁹³. Here we will outline a brief explanation following the argument of the above mentioned work that should prove sufficient in the context of this thesis.

If we take the magnetisation vector as being constrained in the x,y plane (a reasonable assumption for the nanostructures investigated in this thesis), we may map its possible values of $\theta = 0 \rightarrow 2\pi$ onto the circumference of a circle. If we then trace a closed path in real space around our topological defect of interest, we may measure how many complete traversals the magnetisation vector makes of the $\theta = 0 \rightarrow 2\pi$ circle as we traverse the closed path. The number of traversals is the winding number n .

Perhaps easier to visualise, figure 2.9 shows four example magnetisation textures (magnetisation direction represented by the solid coloured arrows) inside the same circular closed integral path

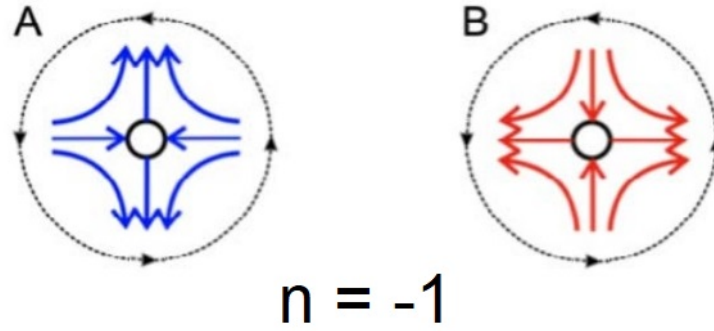


Figure 2.10: Various magnetisation textures containing $n = -1$ topological defects. Solid coloured arrows represent magnetisation direction, dashed black line represents direction of integral path around defect. In both cases, the angle between the integral path direction (here CCW-pointing small black triangles) and the magnetisation texture continuously rotates, opposite to the path direction. Adapted from⁹³.

(dotted black line with CCW triangular arrows). In each case the magnetisation texture contains a single topological defect (solid black circle at the centre). If we observe the magnetisation texture while following the integral path round, we see that in each case A-D it performs a single complete $0 \rightarrow 2\pi$ rotation - hence the winding number of the central topological defect is $n = 1$.

Unfortunately, if we look now at some alternative magnetisation textures shown in figure 2.10 we can see that it is not always this simple! In each example if we observe the magnetisation direction along a circuit of the integral path, we can count a single complete $0 \rightarrow 2\pi$ rotation. Following our above argument, this should give a winding number of $n = 1$. However, for both cases in fig. 2.10 the winding number is in fact $n = -1$. The subtlety which we have neglected so far is that we must also observe the direction which the magnetisation texture rotates *relative* to the direction along which we traverse the closed integral path. If we compare the angle between the arrows denoting the direction of the integral path and the arrows denoting the magnetisation texture direction at various points along the integral path, the difference between the examples in figures 2.9 and 2.10 become apparent. In the $n = +1$ cases shown in fig. 2.9, the angle remains fixed at all points along the integral path. This shows that the magnetisation texture is rotating at the same rate as the integral path vector itself, resulting in positive values of n . In the examples shown in fig. 2.10, the angle between the integral path vector and magnetisation texture rotates in the *opposite* direction to the path itself, leading to negative values of n - in this case $n = -1$ as the magnetisation texture performs a single $0 \rightarrow 2\pi$ along the closed integral path.

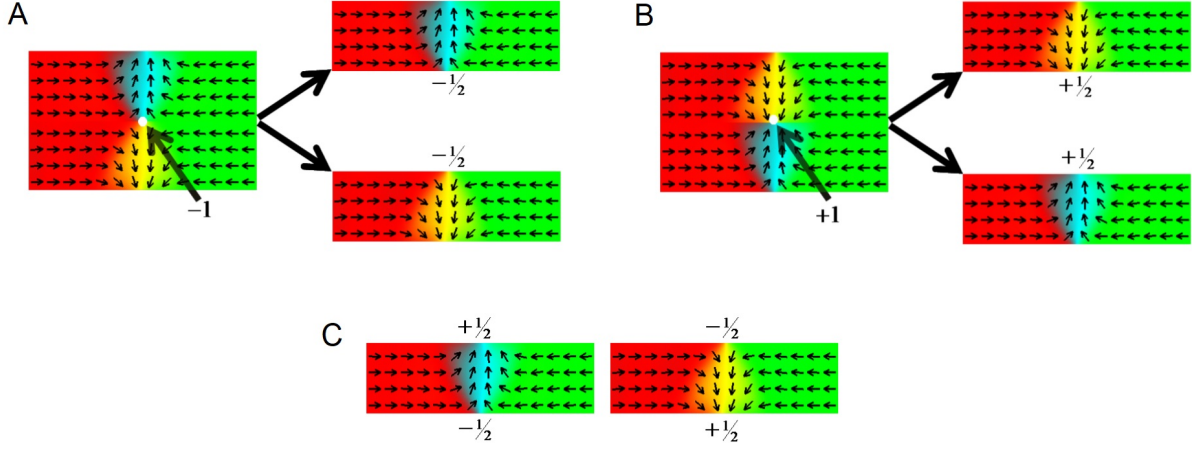


Figure 2.11: Magnetisation textures (magnetisation direction denoted by colour) containing topological defects with $n = -1$ (A) and $n = +1$ (B) winding numbers are split in half to produce pairs of edge-bound topological defects with half-integer winding numbers. C illustrates how 180° domain walls are formed from pairs of opposite-polarity half-integer topological defects. Adapted from⁹³.

So we have now seen how to discern the sign and magnitude of n for topological defects existing in the bulk of a magnetisation texture. How about for the case of the edge-bound ‘half-integer’ defects we mentioned at the start of this section? Luckily, armed with the technique we have just learnt it is quite simple.

Figure 2.11 shows the splitting of magnetisation textures containing $n = -1$ (A) and $n = +1$ (B) topological defects into half. Applying our closed path integral technique as before, the resultant halved topological defects (now sitting on the system edge) and surrounding textures are seen to possess winding numbers of $n = \pm 1/2$. Figure 2.11 C shows the combination of two opposite polarity $n = \pm 1/2$ defects on either side of a rectangular strip. This configuration corresponds to the 180° domain walls introduced in section 2.2.

The topological defects considered in this thesis all have winding numbers of $n = +1$ or $n = \pm 1/2$, which we have now covered and thus may proceed.

2.4 Artificial spin ice

The work in this thesis concerns the development of novel means for controlling and manipulating magnetic charge landscapes in nanopatterned systems. Artificial spin ice (ASI) boasts one of the

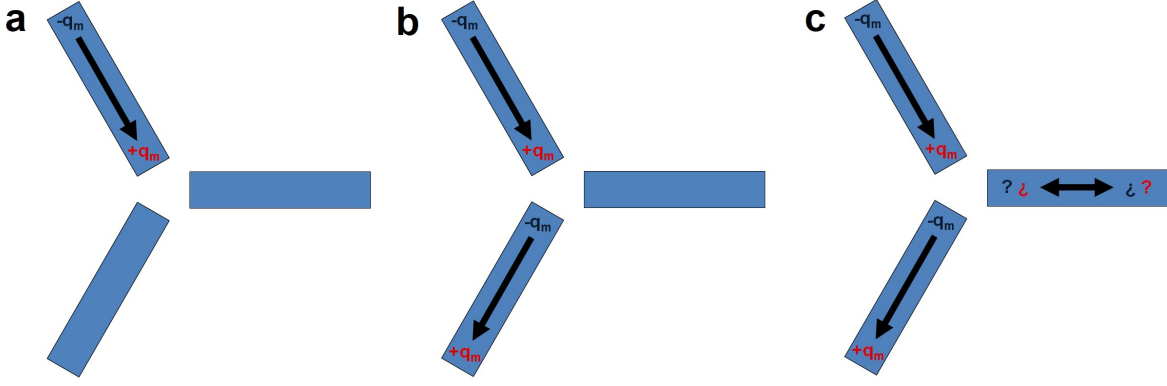


Figure 2.12: Schematic of magnetic charge arrangement at kagome ASI vertex highlighting origin of geometrical frustration. Both possible orientations of the ‘horizontal’ bar have equal energy.

richest magnetic charge landscapes in currently studied systems, and as such represents a perfect candidate in which to develop and demonstrate new tools for interfacing with magnetic charge.

Artificial spin ices are 2D arrays of nanopatterned single-domain magnetic elements, typically nanowires or slightly ovoid ‘stadia’. The magnetisation of the elements is confined to lie along the long-axis by shape anisotropy, leading to Ising-like ‘macrospin’ behaviour with the only possible magnetisation states oriented along the wire length. This allows for just two potential orientations (i.e. ‘left’ and ‘right’ pointing in the case of figure 2.8), hence the comparison to Ising spins. The key points differentiating them from some arbitrary grid of nanowires are twofold: First, that they exhibit strong inter-element interactions arising from significant stray-field overlap between adjacent elements, typically achieved through gaps of less than ~ 150 nm between the ends of adjacent elements. Second (and contingent on the first condition), the magnetic charge distribution of the system must exhibit geometrical frustration. In the case of ASI, geometrical frustration means that the geometry of the array is designed such that there exists no unique arrangement of magnetic charge at each vertex which minimises the system energy - i.e. there must always be some unfavourable contribution to the stray-field interaction between adjacent bars at each vertex.

This can be visualised simply if we consider a triangular three-element vertex, as found in ‘kagome’ (named for a Japanese basket-weaving design) ASI. Scanning electron microscope images of kagome ASI samples fabricated for this thesis are shown in figure 2.13. Figure 2.12 shows magnetisation schematics of a kagome vertex. Let us attempt to populate the magnetic

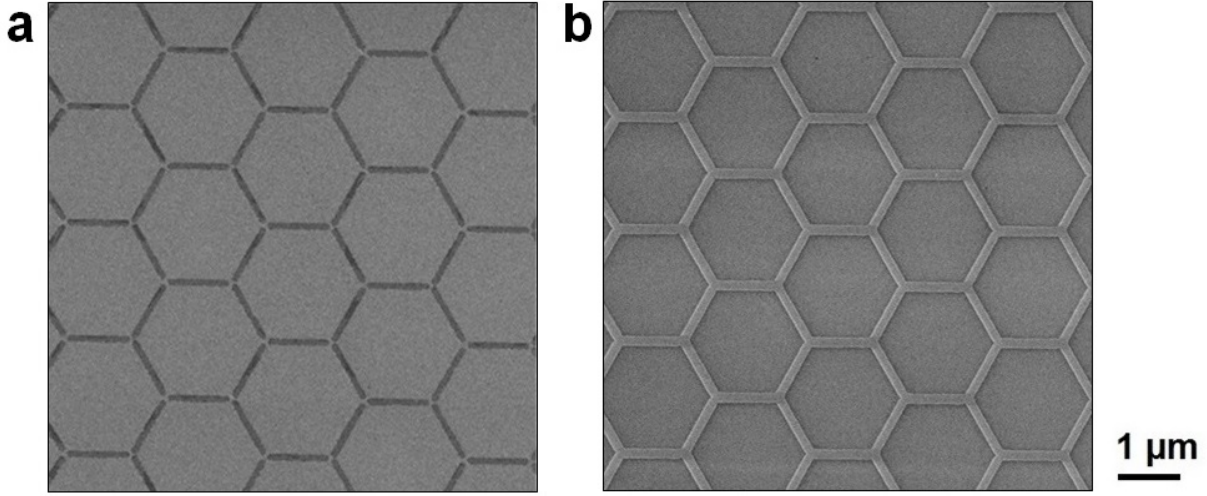


Figure 2.13: Scanning electron microscope images of disconnected (a) and connected (b) kagome artificial spin ice. Structures are composed of 10 nm thick Py.

charges of the vertex such that the dipolar energy is minimised. One may choose the orientation of the first bar freely (a), the second bar must be oppositely oriented to avoid an unfavourable repulsive interaction at the vertex (b), and then we are left with a conundrum for the third bar! (c). Both possible orientations contain a favourable attractive interaction with one bar and a repulsive interaction with the other, often termed ‘two-in, one-out’ (or vice versa) states or $\pm q_m$ states due to the net magnetic charge. This results in a degenerate ground state, the degeneracy of which scales with system size as $\frac{3}{2}^{N/2}$ where N is the number of vertices. It is worth noting that many ASI designs exist, with ‘square’ and ‘kagome’ (hexagonal) the most popular alongside an ever-increasing host of more exotic geometries such as ‘tetris’, ‘shakti’ and Penrose-tiling. This thesis focuses on the hexagonal kagome geometry as its odd co-ordination number ensures that a net magnetic charge is always present at vertices, aiding the imaging and mapping of charge landscapes.

The vast ground state degeneracy and corresponding extensive entropy mirror that of water ice as determined by Pauling⁴⁸, hence the ‘ice’ in artificial spin ice. In H₂O the magnetic charge polarity is replaced by the relative distance between neighbouring hydrogen and oxygen ions (‘close’ and ‘far’ positions are possible, mapping onto $\pm q_m$), with both systems exhibiting remarkably similar residual or ‘zero-point’ entropy as $T \rightarrow 0$ - $S_0 = 0.72R\ln(2)$ and $S_0 = 0.71R\ln(2)$ for kagome ASI and H₂O respectively^{49,50}.

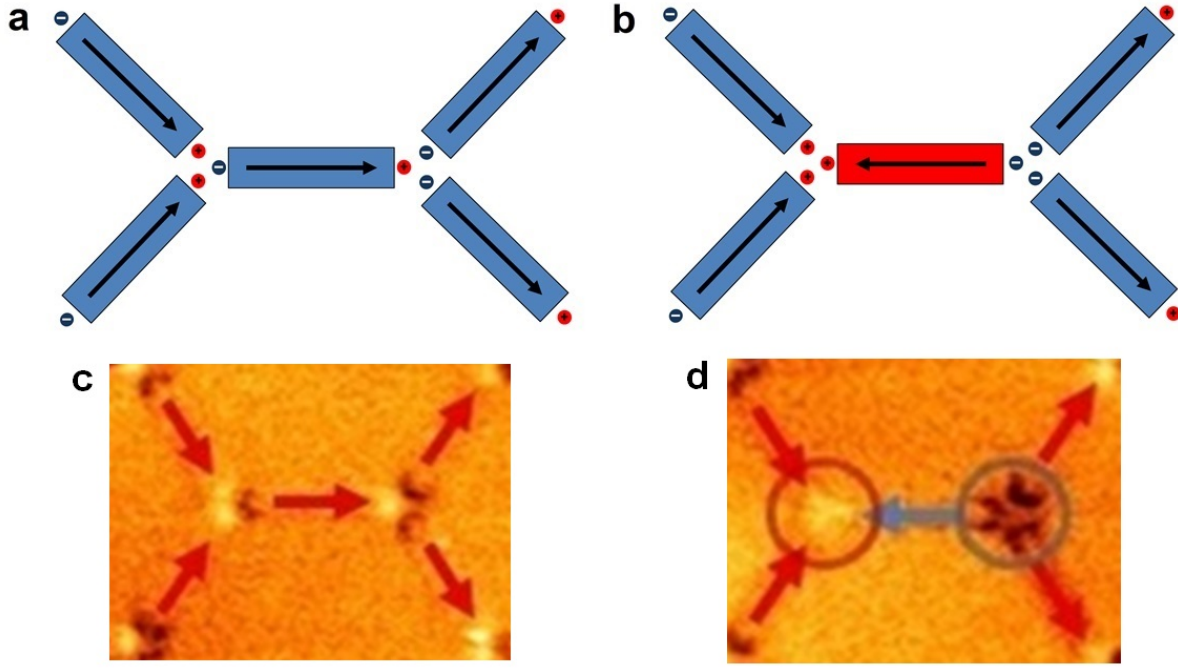


Figure 2.14: Schematics (a, b) and MFM images (c, d) of kagome ASI vertices in low-energy $\pm q_m$ ‘two-in, one-out’ (a, c) and high energy $\pm 3q_m$ ‘three in’ (b, d) states. The $\pm 3q_m$ vertex state is often termed a ‘monopole defect’ in the ASI community.

It is possible to force ASI vertices to break their tendency towards low-energy $\pm q_m$ states and arrange them into energetically unfavourable excited states where all magnetic charges are of matching polarity. These ‘three-in’ or ‘three-out’ ($\pm 3q_m$) states have garnered much attention, in part due to their tantalising similarity to the long-sought Dirac magnetic monopole^{51,52}. These $\pm 3q_m$ vertices act as total sinks or sources of H field, akin to a Dirac or ‘cosmological’ monopole - but crucially are not monopoles in B . Typically a ‘monopole-defect’ creation event will reverse a single macrospin such that excited $\pm 3q_m$ vertices are created at either end, as shown in figure 2.14. Once the initial energy cost of creating a monopole pair has been paid, each monopole may move freely through the ASI system by ‘hopping’ from vertex to vertex, reversing one macrospin at a time. As the total monopole number is conserved during these hopping events there is no additional energy cost and the monopole-defects may move freely. This is an important point and distinguishes the two ‘monopoles’ from a well-separated dipole due to the absence of a separation energy cost.

The chain of reversed macrospins created by monopole hopping events forms a continuous line of magnetisation between the negative and positive monopoles. The line is stabilised as breaking it

would result in an additional monopole-pair creation event with a high energy penalty. This line is somewhat analogous to the ‘Dirac string’ thought to link real cosmological monopoles⁵³, and adds additional weight to ASI’s utility as a model system for studying magnetic monopole-like dynamics.

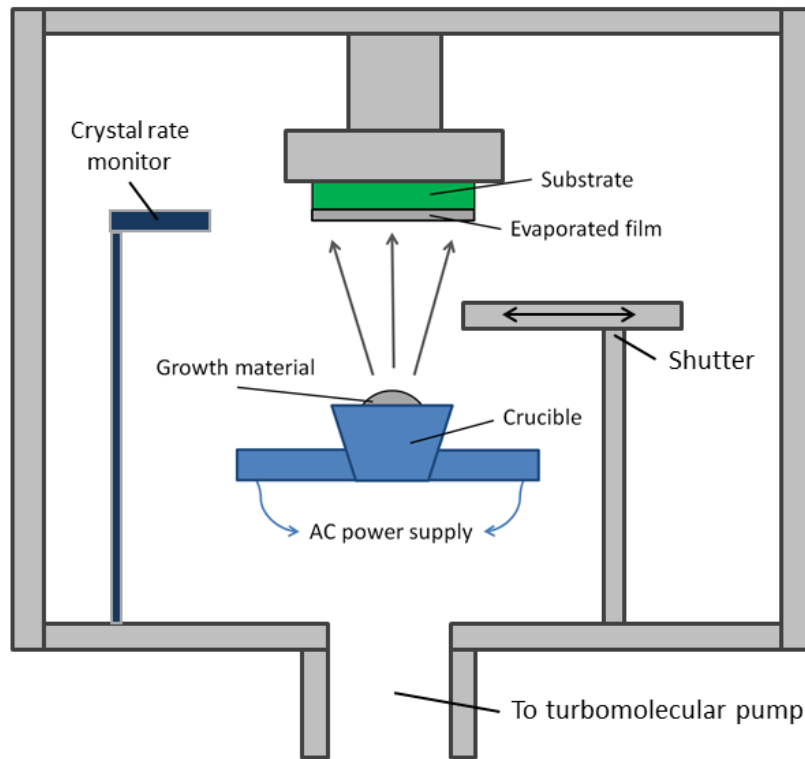


Figure 3.1: Schematic of thermal evaporator.



Figure 3.2: Photographs of thermal evaporation system.

- a) Entire system. UHV growth chamber on the left with viewing window on front face and rotating stage motor visible on top of the chamber. Inficon deposition controller, turbomolecular pump controller and vacuum gauge are visible to right of chamber.
- b) Interior of growth chamber. AlO crucible is at the bottom of the chamber, quartz crystal deposition-rate monitor and sample stage are at the top.
- c) Detail of control electronics, including high-tech equipment booking system.

3 | Methods

3.1 Fabrication

3.1.1 Thermal evaporation

Thin-film growth and metallisation of patterned structures were achieved via thermal evaporation. The desired growth material is placed in a crucible formed of coiled tungsten wire coated with alumina. Above this a substrate, either blank or covered with patterned resist, is suspended on a sample stage with the ability to rotate from 0.1-2 Hz. Substrates for this thesis

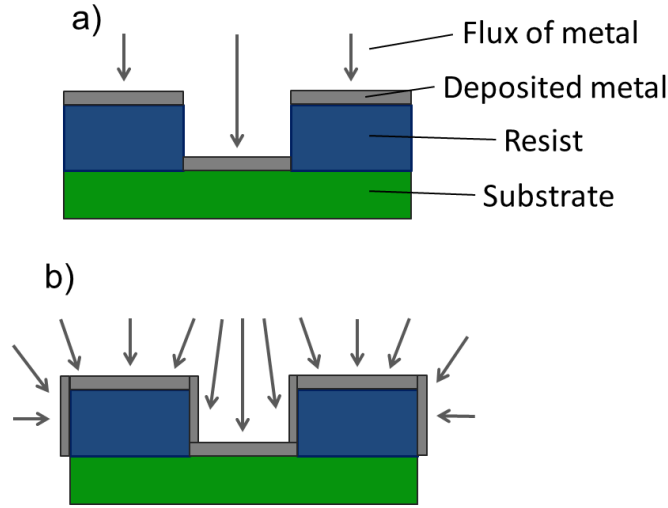


Figure 3.3: Comparison of material deposited on patterned resist by thermal evaporation and sputtering techniques.

- a)** Directional coverage produced by thermal evaporation operating in the ballistic regime. Resist sidewalls are left uncovered.
- b)** Isotropic coverage produced by magnetron sputtering. Resist structures are entirely covered in metal.

were Si[100] with a 300 nm top layer of SiO₂. The entire assembly is pumped down to ultra-high vacuum (UHV) with a base pressure of 5×10^{-7} mBar via a two-stage rotary and turbomolecular pump system. Evaporation is achieved by passing current through the crucible, resistively heating the tungsten wire which in turn heats the growth material through the electrically insulating alumina. The growth metal is heated to boiling point, causing a stream of evaporated atoms to spread through the growth chamber. Upon reaching the substrate atoms are kinetically trapped and adhere on collision, forming a continuous thin film. Film thickness is controlled by a quartz crystal monitor. The crystal is oscillated electronically and as material is deposited onto it its resonant frequency decreases. The net decrease and rate of decrease are used to compute the thickness and deposition rate respectively. Once the growth material is hot enough to achieve the desired deposition rate a shutter between the crucible and substrate is opened and the thickness reading from the crystal monitor is reset. During growth the sample stage is rotated to prevent a wedge-shaped film if the substrate is not mounted perfectly normal to the flux of evaporated material. The shutter is closed once the desired thickness is reached and growth is complete.

The structure of evaporated material can differ substantially from that of bulk. Atoms may not be able to assume their ideal lattice positions before they are trapped in local energy minima

after expending their initial kinetic energy or obstructed by further material reaching the surface. Additionally the vacuum is not perfect and a flux of undesired material will be incident on the substrate throughout growth. These factors lead to lattice dislocations\vacancies and embedded impurities respectively, both of which reduce desirable material characteristics such as saturation magnetisation and conductivity. Purity of deposited material may be improved by lowering the pressure inside the chamber during growth. This 'working' pressure is significantly higher than the base pressure, partly due to the desired stream of evaporated atoms but also due to the release of gases from the crucible and chamber walls due to the elevated temperature during growth. The working pressure benefits from a pre-growth 'outgassing' procedure whereby the crucible is heated and maintained slightly below the growth temperature with the substrate shutter closed while the pumping system removes impurities released into the chamber. This procedure improves the working pressure from $\sim 5 \times 10^{-5}$ mBar to $\sim 5 \times 10^{-6}$ mBar.

Lattice imperfections may be reduced by lowering the deposition rate such that material reaching the surface has more time to assume optimal crystal position before it is trapped under subsequent atoms. Use of a heated substrate also helps by providing atoms at the surface with additional thermal energy with which to escape local positional energy minima and reach optimal position within the lattice. Our system is not equipped with substrate heaters but we may vary deposition rate via the power supplied to the crucible. Lowering the deposition rate has the drawback that the flux of impurities incident on the substrate increases relative to that of the growth material, resulting in loss of magnetism at rates below $0.1 \text{ \AA/s}$ in pressures comparable to ours⁵⁴. As such a balance must be struck between allowing atoms sufficient time to order and maintaining high purity. For the structures in this thesis the system was operated at a rate of $0.8 \text{ \AA/s}$ which is found to reliably produce good quality material.

Thermal evaporation is suited to liftoff-based nanofabrication as the low chamber pressure during growth means that evaporated material travels directly to the substrate in a ballistic regime. This gives highly-directional deposition as opposed to the isotropic coating provided by magnetron sputtering, an alternative widely-used thin-film growth technique. A directional coating is important for lithographic patterning as it leaves the resist sidewalls metal-free and

open to attack and removal by the liftoff solvent. An illustration of this is given in figure 3.3. Our evaporator is confirmed to function in the ballistic regime by calculating the approximate mean free path λ of evaporated material. By taking the ideal gas law in the single particle limit where $N = 1$

$$PV = k_B T \quad (3.1)$$

and treating the volume as a cylinder with cross-sectional area equal to that of an evaporated metal atom, the cylinder's length then represents the unoccupied distance in space through which the atom may travel before any gas-phase collisions occur. This length is equivalent to λ and allows us to write

$$\lambda = \frac{k_B T}{\pi r_{atom}^2 P} \quad (3.2)$$

Taking $r_{atom} = 5 \text{ \AA}$, $P = 5 \times 10^{-6} \text{ mBar}$ and $T = 300 \text{ K}$ a mean free path of $\lambda = 10.5 \text{ m}$ is found. This is much larger than the crucible-substrate separation of $\sim 0.2 \text{ m}$ hence the evaporation system is well within the ballistic regime, even for clumps of several atoms emitted together.

Both Py (NiFe) and Co were thermally evaporated to form the nanostructures considered in this thesis. The vast majority of structures were Py, and unless Co is explicitly mentioned the reader can assume that structures are composed of Py.

3.1.2 Electron beam lithography

Electron beam lithography (EBL) operates on much the same principle as SEM and was performed in this thesis using the same Raith E-Line system. The key difference is that rather than raster scanning a square imaging window, the beam is directed along a path defined using computer aided design (CAD) software which describes the desired structures in either positive or negative tone. Patterning occurs in a layer of electron-sensitive positive or negative tone resist

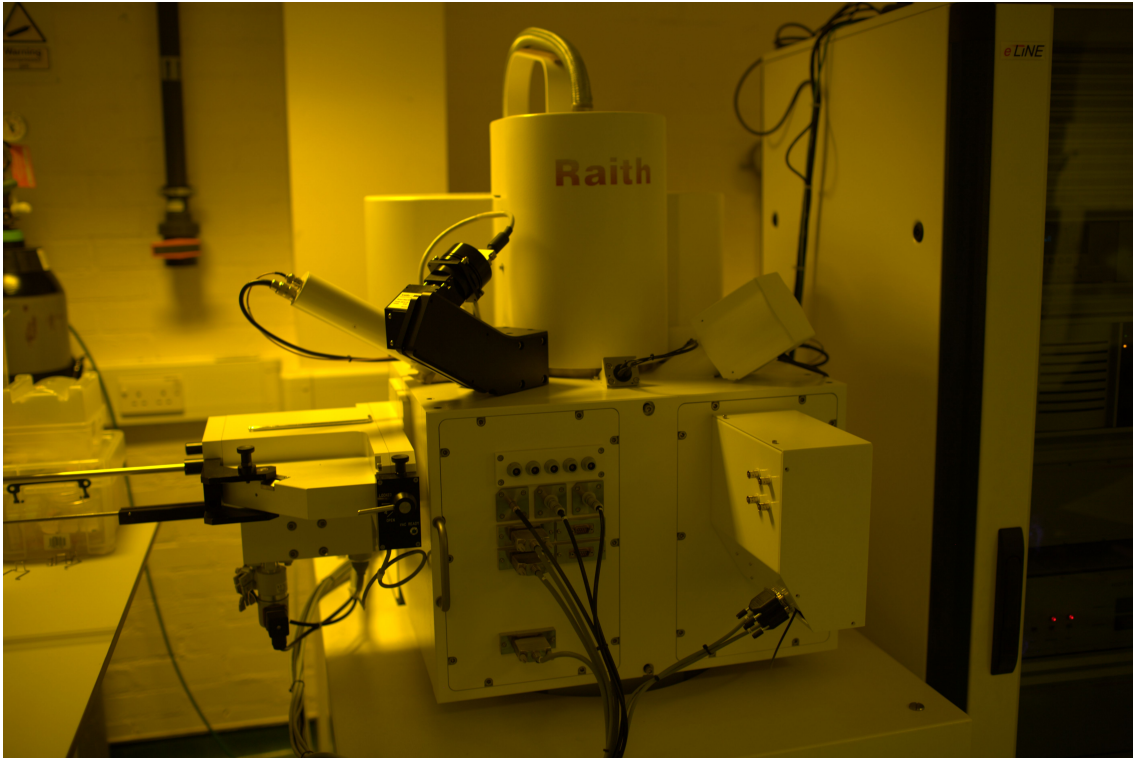


Figure 3.4: Photograph of Raith E-Line combined scanning-electron microscopy and electron-beam lithography system.

which is applied to a blank substrate or continuous thin film.

Polymethyl methacrylate (PMMA) is a model positive tone resist. Stable for long periods of time, easy to handle and apply and providing good quality resolution down to 10-20 nm it is the most widely used positive tone EBL resist and was employed exclusively for this thesis.

Essentially short chains of perspex suspended in anisole solvent, PMMA is applied to substrates by spin coating and then baked for 5-10 minutes at 180° C to evaporate off solvent and promote polymer crosslinking, leaving a hardened layer of resist. Electron beam doses of 200-450 $\mu\text{C} / \text{cm}^2$ are then locally applied to cause de-crosslinking of polymer chains, allowing the exposed regions to be removed via a 3:1 mixture of isopropanol (IPA) and methyl isobutyl ketone (MIBK). This leaves bare substrate along the beam's path and is suited to structures composed of mostly empty space if using a liftoff process or mostly continuous film if milling.

Two negative tone resists were employed, Hydrogen silsesquioxane (HSQ) and Microchem 'Ma-N 2400'. EBL doses of 1000-2500 $\mu\text{C} / \text{cm}^2$ and 200-500 $\mu\text{C} / \text{cm}^2$ were used for HSQ and Ma-N respectively. In both these resists incident electrons promote cross-linking, leaving exposed

regions unaffected while unwritten resist is removed via solvent (tetramethylammonium hydroxide (TMAH) solution for HSQ, acetone and Microchem 'Ma-R' for Ma-N. This leaves resist along the beam's path and is best suited for structures composed of mostly empty space if ion milling from a sheet film or mostly continuous film if metallising post-lithography. HSQ has been shown to achieve feature sizes of 2 nm and 10 nm pitch and unfortunately it has issues with polymer agglomeration or 'glassing out' where the polymer chains suspended in liquid begin to spontaneously cross-link, spoiling the stock. Ma-N has no such issues and is also much faster to write due to the lower beam doses required. While both resists were employed, the majority of successful work in this thesis was accomplished using Ma-N.

3.1.3 Ion beam milling

Ion beam milling (IBM) is a technique used to remove unwanted material from a thin film. IBM operates by accelerating ions (here Ar^+) towards a sample with sufficient energy that they kinetically liberate atoms from the sample surface upon collision. By coating regions of the film with a patterned resist, IBM may be used to define nanostructures as material covered by the resist is not removed. After removing unprotected material, the remaining resist may be dissolved by solvent to leave the patterned structures on top of bare substrate. Unlike EBL one needs to be aware of the risk of ion implantation, whereby the milling ions become embedded in the thin film. This can negatively affect certain material properties, including lowering saturation magnetisation and introducing pinning sites capable of trapping DWs. While these IBM introduced DW pinning sites may be exploited to great functional effect⁵⁵⁻⁵⁷ (especially when using locally-focused IBM, known as 'FIB'), it is generally undesirable and care must be taken to avoid it by optimising resist thickness, milling time and structure dimensions (generally some implantation occurs at structure edges so one way add 'sacrificial' buffer regions by slightly increasing structure widths). For this thesis IBM was employed solely with negative tone resists.

Initially, hydrogen silsesquioxane (HSQ) resist was employed. It has high resolution (down to 2-4 nm) and good IBM resistance. Unfortunately due to ageing the resist stock had polymerised before I started my thesis, leading to months of lost time as the resist would still appear to

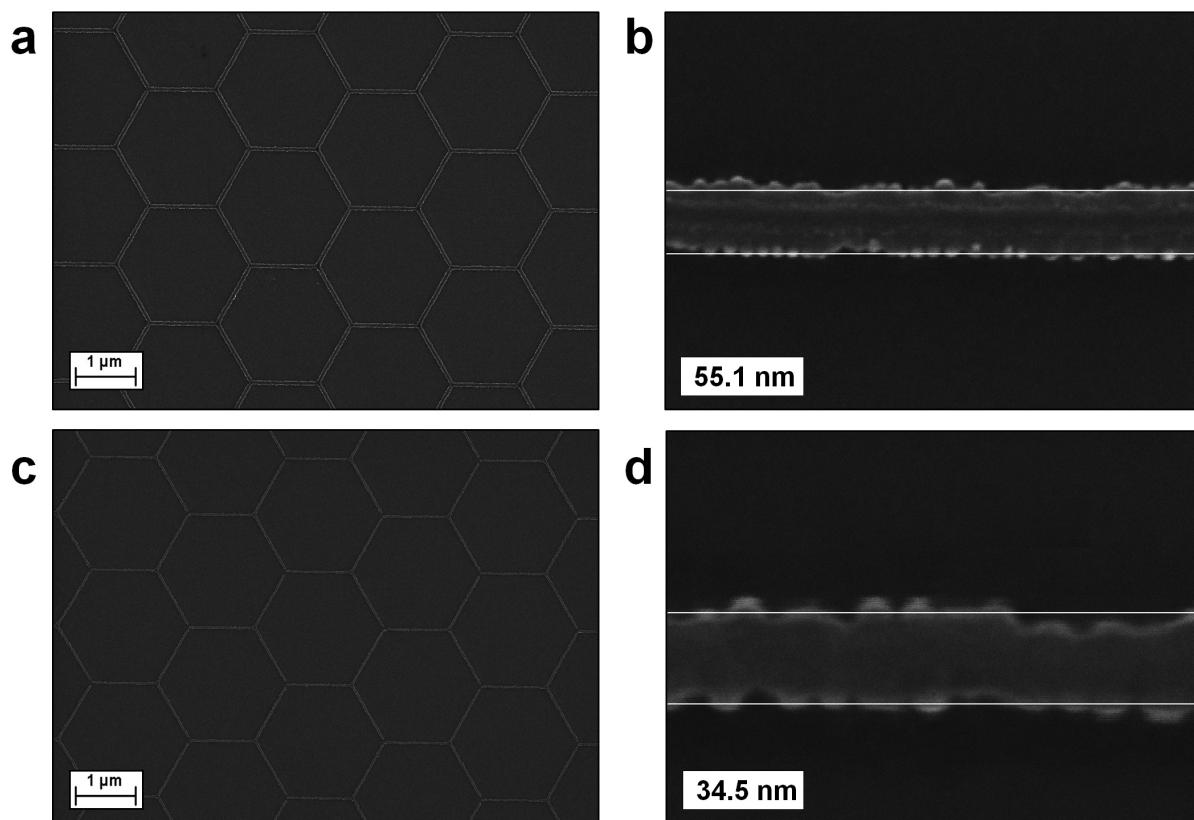


Figure 3.5: SEM images of poor-quality, first-generation IBM-patterned Py nanostructures. a),b) 1 μm long, 55.1 nm wide nanowire ASI. Edge roughness is poor. c),d) 1 μm long, 34.5 nm wide nanowire ASI. Edge roughness is extremely poor relative to wire width and several broken wires are observed around vertices.

spincoat well but not develop patterned structures after writing. Eventually, the cause of the issue was discovered and the decision was made to move to Ma-N resist, a more user friendly resist in terms of patterning time, development and resist-removal.

The first IBM patterned structures using Ma-N were of poor quality. Edge roughness was extremely poor, with some Ar^+ ions able to penetrate the resist edges due to unoptimised EBL dosage and structure design leading to a sloped resist profile that thinned at structure edges. Ion implantation was rife and MFM of ASI arrays showed no clear magnetic charges at vertices. An example of the first-generation structures are shown in figure 3.5.

The IBM patterning process was then optimised, with resist thickness and EBL dose increased. Milling acceleration voltage was reduced and the milling process broken up into several 90s stages (number of stages depended on film thickness, with a milling rate of 3.5 nm/min for Py)

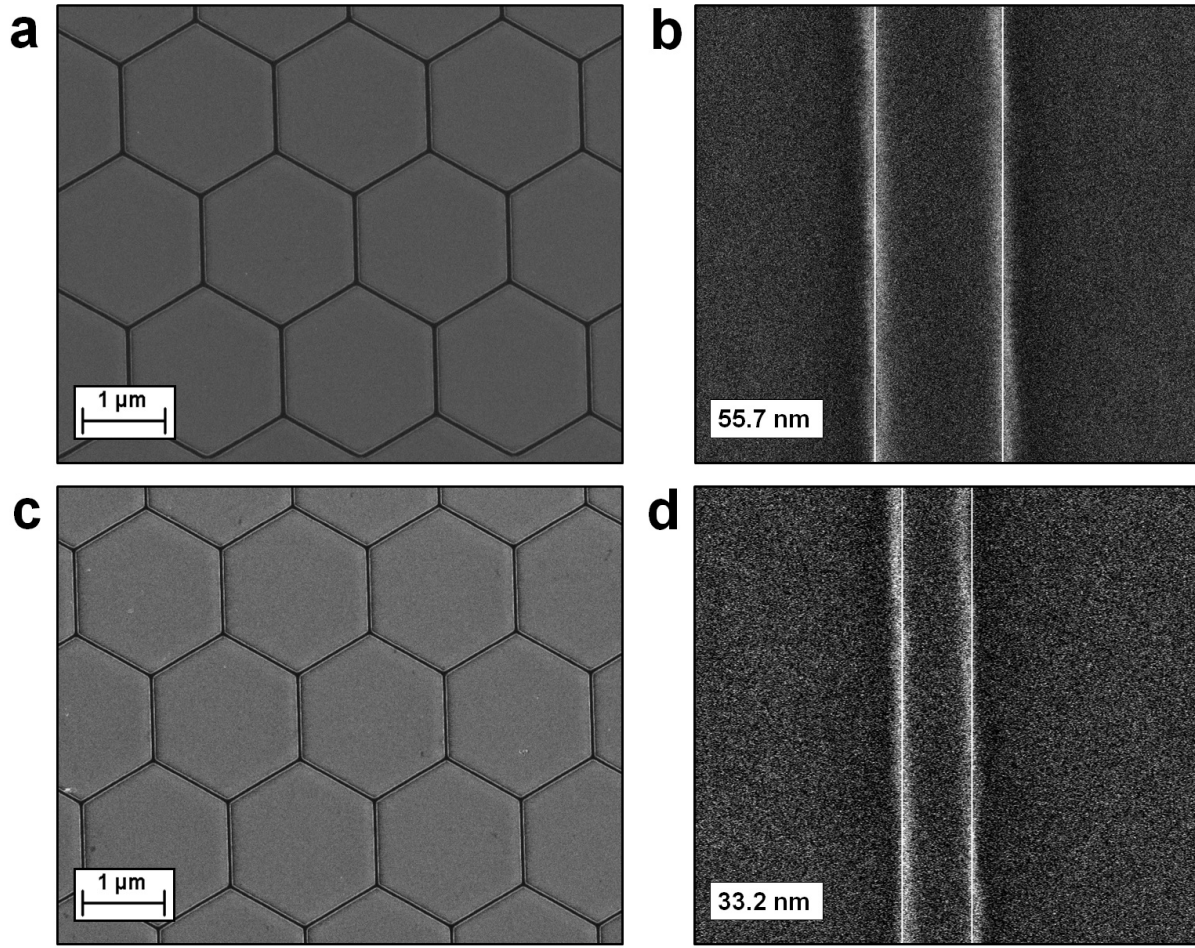


Figure 3.6: SEM images of optimised IBM-patterned Py nanostructures.

a),b) 1 μm long, 55.7 nm wide nanowire ASI. High quality structures are observed throughout ASI lattice.
c),d) 1 μm long, 33.2 nm wide nanowire ASI. High quality structures are observed.

rather than a continuous mill. Breaking up the milling reduces heating of the sample which was previously leading to brittle and hard to remove resist. Examples of post-optimisation structures are shown in figures 3.6, 3.7 and 3.8 below. High-quality structures with wire widths down to 17.4 nm were reliably produced, with a plot of EBL dose against measured structure width shown in figure 3.9 for a range of CAD structure widths. Magnetic properties of the structures were good, with figure 3.10 showing example MFM images.

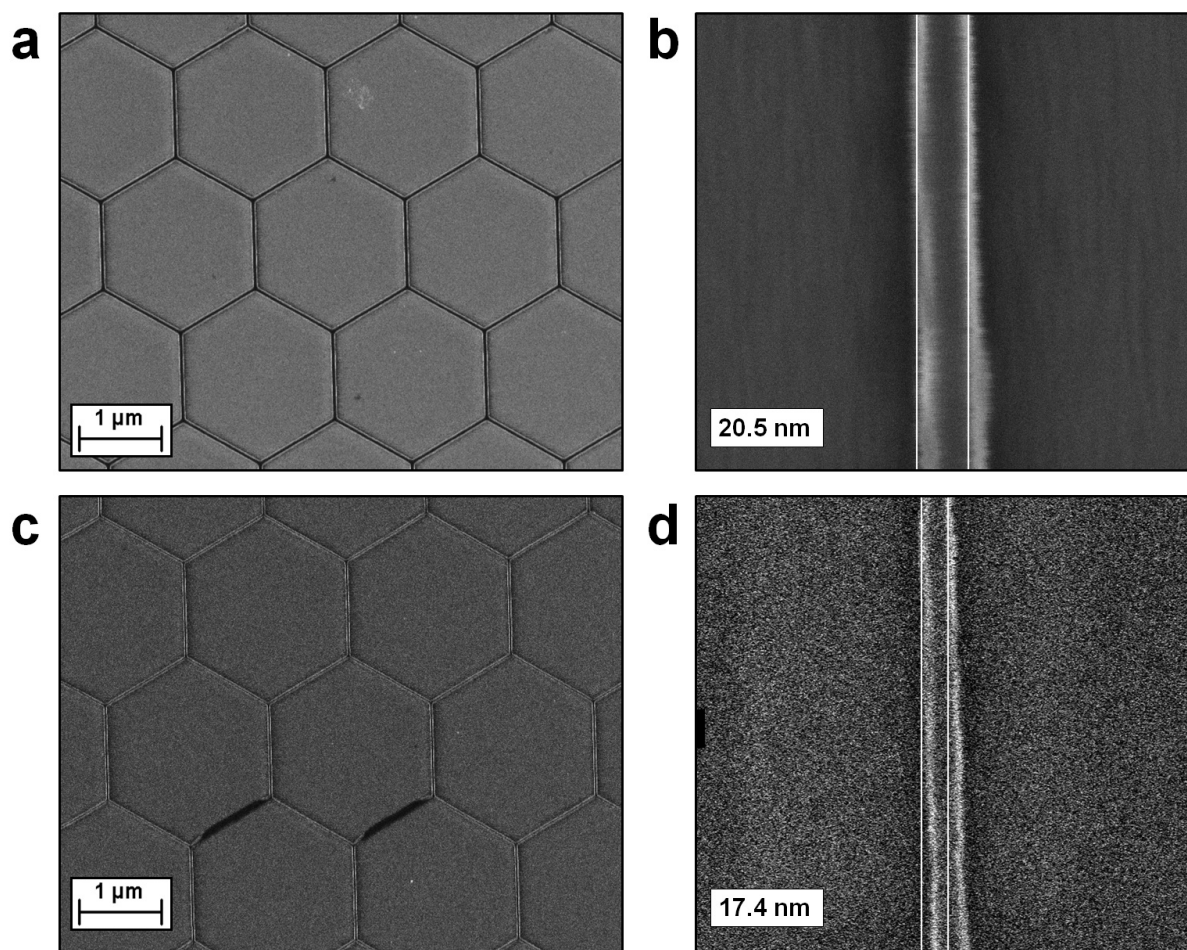


Figure 3.7: SEM images of optimised IBM-patterned nanostructures.

a),b) 1 µm long, 20.5 nm wide nanowire ASI. High quality structures are observed throughout ASI lattice. c),d) 1 µm long, 17.4 nm wide nanowire ASI. Generally ASI is high quality, though two nanowires appear to have collapsed. Presumably due to patterned resist being too thin to survive milling and resist removal processes.

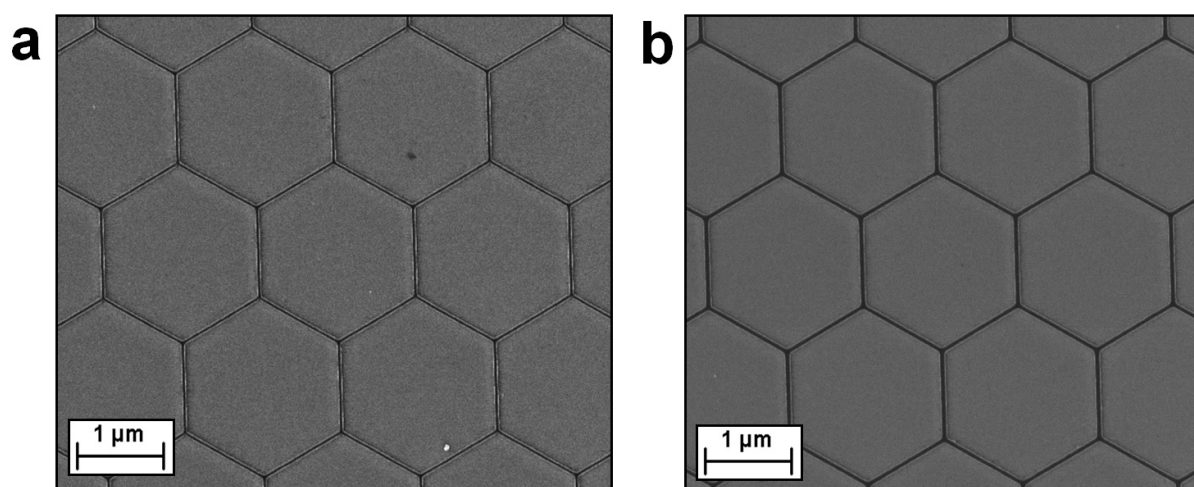


Figure 3.8: SEM images of optimised IBM-patterned nanostructures. a) is 21.5 nm wide, b) is 30.7 nm wide.

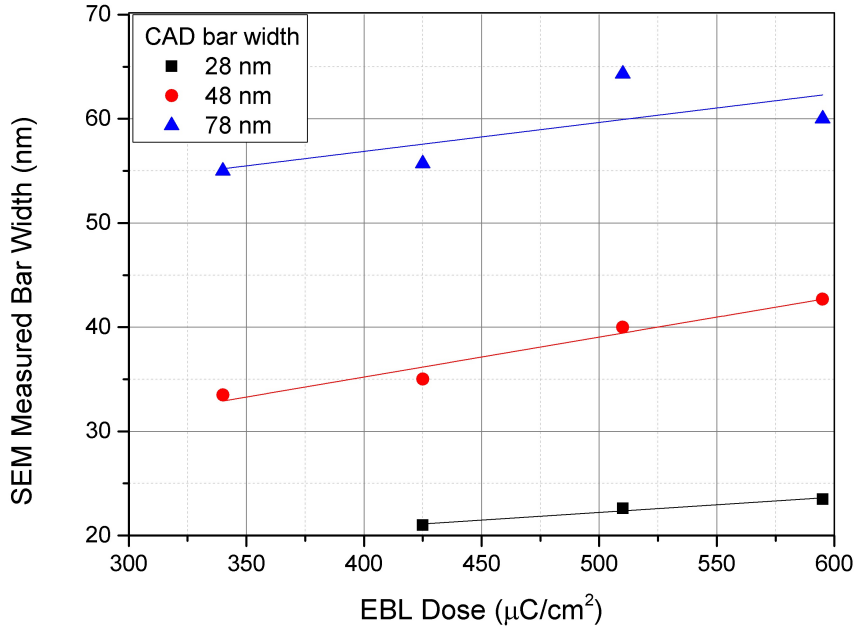


Figure 3.9: Width of final ion-beam milling patterned nanostructures (measured by SEM imaging) as a function of electron dose. Three CAD-defined nominal structure widths (28 nm, 48 nm and 78 nm) are examined. The resultant fabricated structures were found to be thinner than their nominal CAD-defined widths, with an approximately linear relation between resultant structure width and beam dose.

3.1.4 Liftoff process

All nanostructures composed of material we were able to deposit in-house were fabricated using positive-tone electron beam lithography followed by a liftoff process. Positive tone liftoff has fewer steps relative to IBM with negative tone resist and lower scope for process failure. As our thermal evaporator has no problem producing good quality permalloy (nominally $\text{Ni}_{81}\text{Fe}_{19}$), the simple ferromagnet par excellence for nanopatterning due to its near-zero magnetocrystalline anisotropy, the vast majority of samples were fabricated via liftoff. Co was also thermally evaporated when a higher saturation magnetisation or coercivity was required. The liftoff process comprises four distinct steps: First, a layer of PMMA resist is spun-coat over a blank Si/SiO₂ substrate and a CAD-defined pattern is described using EBL. The resist is then developed, leaving empty channels in an otherwise continuous sheet of resist where metallic elements are to be added. The third step entails loading the patterned resist into the thermal evaporator and coating the sample with a metallic film of the desired thickness. Finally, the metal coated sample is placed into acetone solvent for 1-2 days with a few minutes of ultrasonic agitation if the

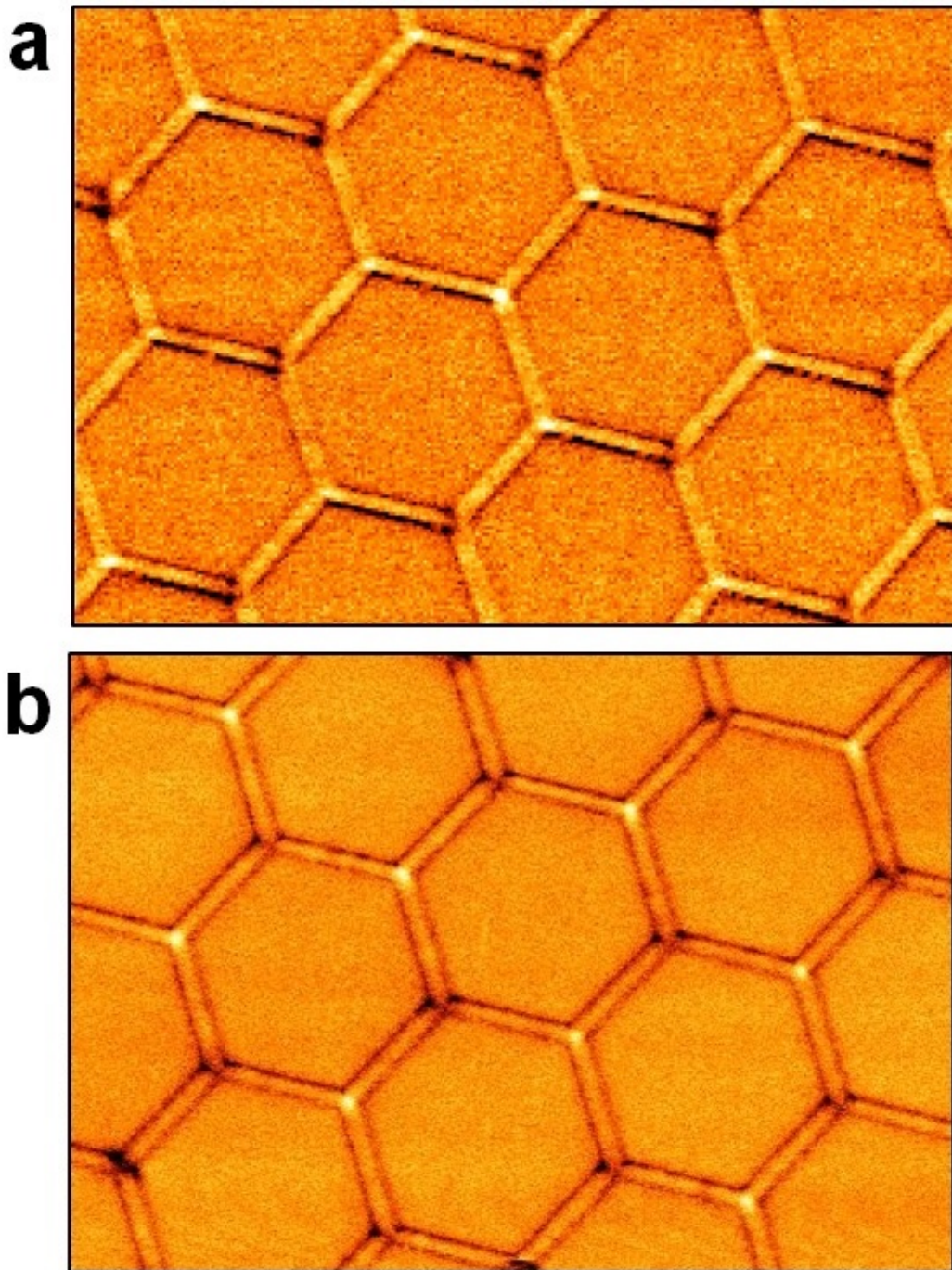


Figure 3.10: MFM images of 100 nm wide, 15 nm thick Py nanowires showing reversal of magnetic charge polarity while imaging. ‘Fast’ axis of raster-scan is horizontal in all images.

pattern contains closed spaces of below 700 nm diameter. This removes the metal-coated resist and any other organic residues which may be present, leaving well-defined nanopatterned metallic elements on an otherwise bare substrate.

Feature sizes down to 17.5 nm were successfully fabricated during this thesis which is at the bottom end of the capability of our EBL system, showing a high-quality process was developed. On average, nanowire widths of 70-100 nm were employed which could be reliably fabricated with few or no defects.

3.2 Characterisation

3.2.1 Atomic force microscopy

When working with nanopatterned structures it is often desirable to obtain a detailed picture of the surface topography. Nanofabrication techniques rarely produce perfect structures and the manner in which they deviate from their ideal form offers useful clues as to how to adjust the fabrication process to obtain better structures. Atomic force microscopy (AFM) is ideally suited to this purpose, providing sub-nm resolution in $\hat{z}$ and 10-30 nm lateral resolution.

AFM was developed at IBM in 1986⁵⁸ and involves bringing a sharp (ideally atomically sharp but typically 10-30 nm wide at its end) tip mounted on a flexible cantilever close to the surface of interest. The surface is then raster-scanned in the $\hat{x}\hat{y}$ plane under a static tip using a piezoelectric sample stage. Short-range forces between the tip and sample dominated by repulsive Coulombic and attractive Van der Waals interactions are very sensitive to the distance between tip and surface. These interactions cause the tip to closely follow the topography of the moving surface underneath, bending the flexible cantilever as shown in figure 3.11. To convert the movement of the tip and cantilever into a more easily measurable quantity a laser beam is directed at the reflective top side of the cantilever close to the tip location with an angle of incidence such that the beam is reflected onto a photodiode array some distance away from the cantilever. The cantilever bends up and down as the tip tracks the varying surface topography

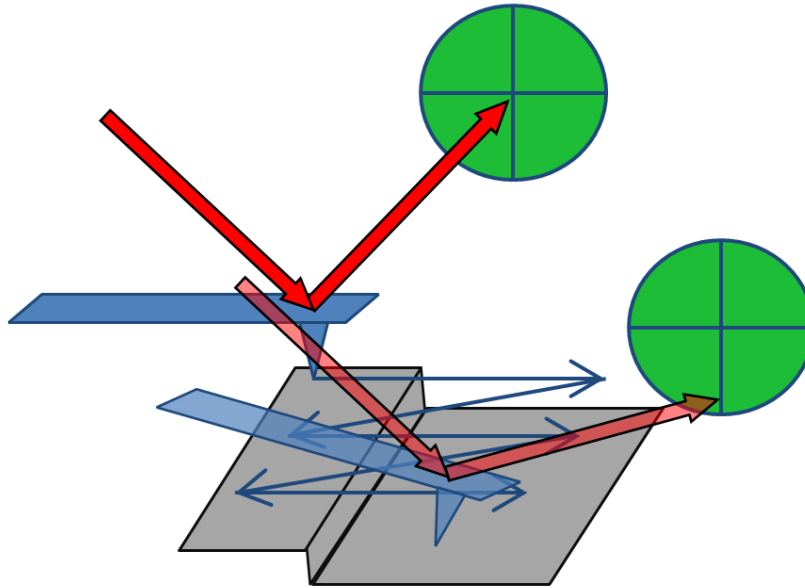


Figure 3.11: Schematic of atomic force microscopy operation. The tip and cantilever are depicted in light-blue following a dark-blue raster path over the grey sample surface. A red laser beam strikes the back of the cantilever and is deflected onto the green photodiode array. Two positions in the raster scan are shown, illustrating the effect of varying surface topology on the position of the laser beam on the photodiodes. The lower surface to the right of the image leads to the laser striking a lower point on the photodiode array. The movement of the laser spot across the array is much larger than the movement of the tip across the step in the sample due to the distance between the cantilever and photodiode. As such it is much easier to measure and convert into surface topography data.



Figure 3.12: Photographs of Dimension 3100 SPM system used for AFM and MFM.

- a) Entirety of system with vibration isolation housing and cover visible as well as electronic control hardware rack.
- b) Scan-head removed from system with tip-holder facing camera. Tip is visible at very centre of image, held in place by metallic spring-clamp.
- c) Scan-head in place over sample stage. Microscope used for scan-area positioning visible at left of image.

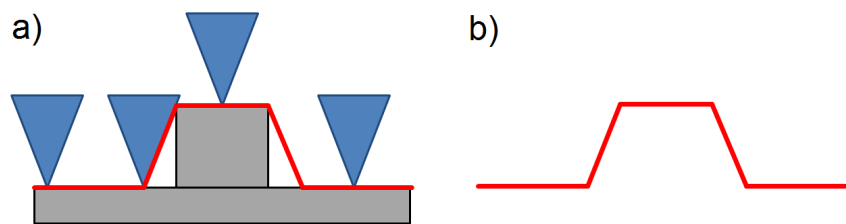


Figure 3.13: Illustration of AFM tip convolution.

a) Tip path across a stepped surface. The tip-path is illustrated in red and does not perfectly track the surface due to the finite tip-width.

b) Surface height data recorded from the scan in **a)** illustrating convolution error.

which causes the reflected laser spot to move across the photodiode array. The movement of the spot is much greater than that of the tip itself due to the distance between the cantilever and photodiodes, an arrangement known as an 'optical lever'⁵⁹, allowing accurate measurements of tip displacement and therefore surface topography to be made with relative ease via the distance between illuminated photodiodes within the array. The measured deviation can be used with an electronic feedback system to maintain a constant tip-sample separation by varying the $\hat{z}$ -position of the sample stage so that the laser spot remains in the centre of the photodiode array, minimising stress on the cantilever. This allows surfaces with sharper gradients and higher aspect ratios to be measured with reduced risk of damage to the tip and cantilever. Tips can be made from a range of materials but are typically Si or SiN.

While AFM has good $\hat{z}$ -resolution its lateral resolution is limited by the tip's radius of curvature. As depicted in figure 3.13 the tip may not fully track the topography of a raised area due to collision with the wider upper region of the tip. As such a sharp step will always appear as a slope with gradient proportional to the tip radius. More accurate determination of the width of stepped features such as nanowires is possible using scanning electron microscopy.

The act of physically dragging a tip over a sample while in contact can be detrimental to both tip and sample. AFM resolution is limited by tip sharpness and contact forces can cause the fragile end of the tip to break off or attract debris, increasing the effective tip radius and lowering resolution. In some cases the tip can even break off parts of the sample itself, usually in soft or biological materials but also in nanopatterned metal. To mitigate these effects AFM systems may be operated in a 'tapping mode' whereby the cantilever is oscillated at its resonant

frequency by a piezoelectric actuator at amplitudes around 100 nm. Here the tip is only in contact with the surface for very brief periods (a typical resonant frequency is 180 kHz) and while in contact there is negligible dragging of the tip across the sample. All AFM results presented below were obtained in tapping mode.

AFM was performed on two hardware systems, the vast majority on an old (1995) but reliable Dimension 3100 with a few high-resolution scans taken using a modern Asylum MFP-3D. The Asylum was only employed when its enhanced x, y stage positioning (± 2 nm accuracy) was necessary to perform intricate writing operations as described in chapter 5.

3.2.2 Magnetic force microscopy

AFM systems provide a versatile workhorse upon which to base methods for obtaining spatial surface maps of various physical properties. One such method which requires very little modification of the basic AFM system is magnetic force microscopy (MFM). MFM provides a map of the magnetisation state of a sample's surface using an AFM tip which has been coated with a magnetic material, typically evaporated CoCr in commercially available tips. A typical tip field is around 40 mT at a tip-sample separation of 50 nm⁶⁰. Unlike AFM, the tip is held at a distance of 20-200 nm above the sample surface while the surface is raster scanned.

The force on the MFM tip from a magnetic sample's stray field is given by

$$\mathbf{F} = \mu_0(\mathbf{m}_{tip} \cdot \nabla \mathbf{H}_{sample}) \quad (3.3)$$

While MFM systems can measure this force directly by tracking changes in the deflection of a static (non-oscillating) tip a more common technique is to measure changes in the resonant frequency of a cantilever in driven oscillation ω_r where⁶⁰

$$\omega_r \approx \left(1 - \left(\frac{1}{2k} \right) \frac{\delta \mathbf{F}_z}{\delta z} \right) \quad (3.4)$$

and

$$\frac{\delta \mathbf{F}_z}{\delta z} = \mu_0 \left(\mathbf{m}_{tip} \cdot \frac{\delta^2 \mathbf{H}_{sample}}{\delta z^2} \right) \quad (3.5)$$

As the resonant frequency is shifted by the stray field $\mathbf{H}_{sample}$ of the sample a feedback system varies the driving frequency of the cantilever ω_d such that the cantilever oscillation is maximised. This yields a stronger signal than direct measurements of the force as while $\mathbf{F}_z$ is small the stray field of the sample has a sharp spatial gradient and $\frac{\delta \mathbf{F}_z}{\delta z}$ is larger.

MFM may be operated in either constant height or constant tip-sample separation mode.

Constant height is simpler to perform as the sample stage is left in a fixed $\hat{\mathbf{z}}$ -position ('height') but has the drawback that a region of magnetisation at a low tip-sample separation will appear stronger than an identical region of magnetisation at a higher tip-sample separation. This is avoided in constant tip-sample separation mode whereby a preliminary AFM scan of the surface is used to dynamically adjust the $\hat{\mathbf{z}}$ -position of the sample stage during the MFM scan. Constant separation mode also allows the user to scan at lower tip-sample distances without fear of crashing the tip into raised surface features. Smaller separations are also useful as the spatial resolution of MFM is roughly equal to the tip-sample separation down to a minimum of around 20 nm. At low separations the tip experiences well-defined stray fields above the sample's poles magnetic charges which then smear out and overlap at larger tip-sample separations, decreasing image resolution. A drawback of constant separation mode is that bringing the MFM tip close enough to the sample to perform AFM can alter the sample's magnetisation state.

A key part of this thesis involved developing a 'magnetic writing' technique, using the local magnetic field of MFM tips to achieve controlled writing of the magnetic landscape in nanopatterned ferromagnetic structures. That such a system could be conceived and developed without need for modification of system hardware is testament to the versatility and power of surface-probe based systems. This technique will be discussed in-depth in later chapters.

MFM was performed on the same two hardware systems as AFM, namely an old but stable and functional Dimension 3100 and a modern Asylum MFP-3D. The Dimension 3100 was used for all

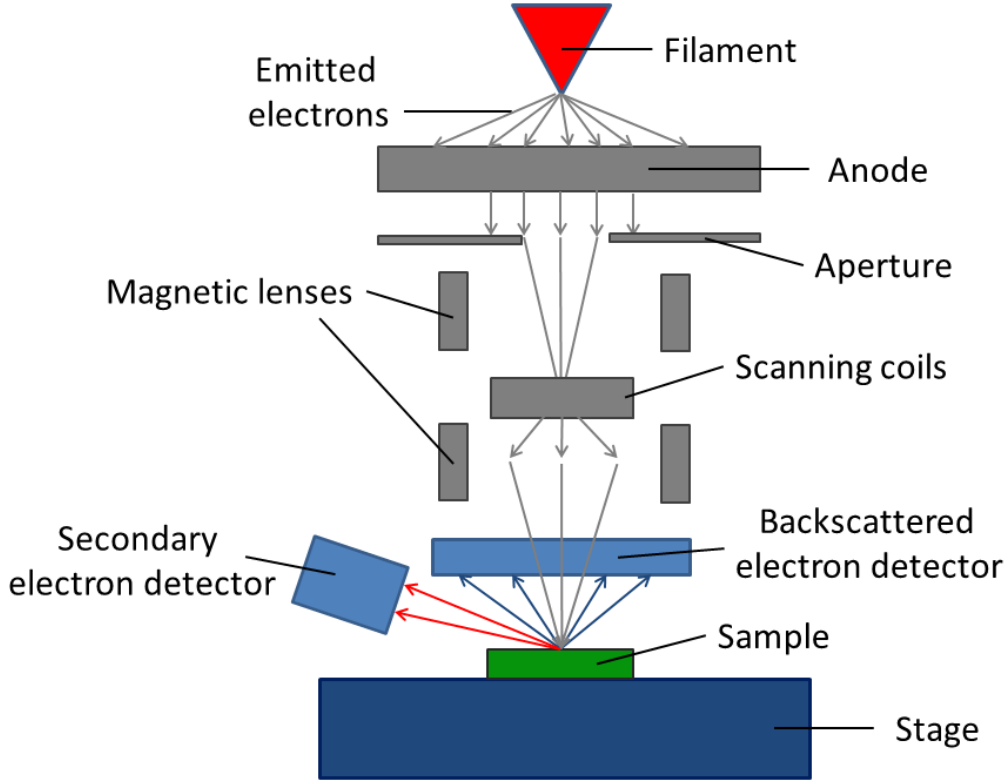


Figure 3.14: Schematic of a scanning electron microscope.

AFM and MFM imaging save for figure 5.3, showing the ground state of kagome ASI. The reason for this is that the manual writing of the ground state required accurate positioning of the tip along complex paths around the sample, with an x, y position error of less than $\pm 50 - 100$ nm. The tip-movement piezos in the Dimension 3100 are around 20 years old, and while fine for accurately following a continuous raster-scan path for imaging, have an unacceptable level of drift for intricate patterning duties. The Asylum MFP-3D is quoted by the manufacturer as x, y accurate down to ± 2 nm and was able to write the ground state without issue.

3.2.3 Scanning electron microscopy

Scanning electron microscopy (SEM) is a powerful sub-optical imaging tool. The Raith E-Line system installed at Imperial College is capable of 5-10 nm resolution with a sample loading time of only 10 minutes. SEM functions in a similar manner to optical microscopy, replacing photons with electrons and glass lenses with precisely-shaped magnetic fields. The shorter wavelength of

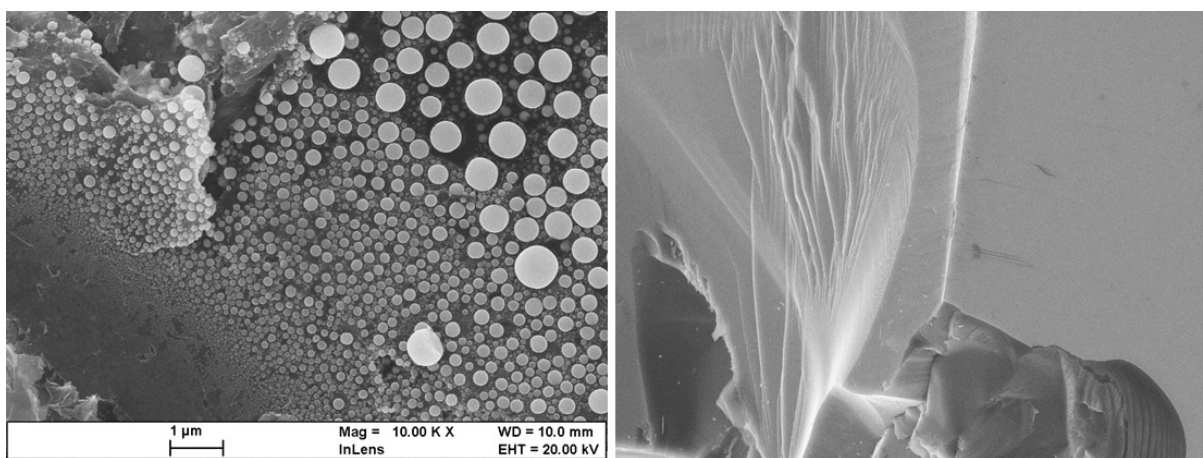


Figure 3.15: Example images taken using a scanning electron microscope. Left panel shows Sn spheres deposited on a Carbon film. Their highly regular round geometry allows for accurate setup and focusing of the electron beam. Right panel shows cleaved edge of Si/SiO₂ substrate.

electrons relative to optical photons allows for greatly enhanced resolution and depth of focus.

Rather than capturing an image instantaneously as in optical microscopy SEM builds an image by raster-scanning a beam of electrons over an area. The beam is moved by a set of scanning coils which deflect the beam by subjecting it to time-varying magnetic fields similar to the process used in cathode-ray televisions. SEM is performed under ultra high vacuum (UHV) and requires that samples be conductive. Insulating samples may be imaged by first depositing a thin layer of metal via sputtering or thermal evaporation.

A schematic of an SEM is shown in figure 3.14 illustrating the path of the electron beam.

Electrons are emitted from a LaB₆ filament and accelerated to 20 keV by an anode. The beam then passes through a 10 μm aperture before being focused onto the sample by condenser and objective magnetic lenses. Electrons incident on the sample are subject to a range of interactions leading to distinct types of electrons leaving the surface. The main component of an SEM image typically comes from low-energy (below 50 eV) 'secondary' electrons which are emitted from the top few nm of the sample. Secondary electrons are released from the sample by inelastic collisions with electrons in the accelerated beam. The flux of secondary electrons at the detector is sensitive to the sample-detector separation and the electronic structure of the sample, allowing the detected intensity to be used to build an image of the sample topography and material composition. The material sensitivity is not exceptionally high in comparison with other electron microscopy techniques such as Auger electron spectroscopy or transmission electron microscopy,

but metallic structures show significantly different contrast to an insulating substrate, which for the purposes of this thesis is ideal. Secondary electrons are collected by a scintillator-photomultiplier based Everhart-Thornley detector⁶¹. Backscattered electrons arise from elastic collision with higher energy electrons a few microns within the sample. The interaction is sensitive to the local electron density of the sample and therefore atomic number, allowing a map of sample composition to be produced. In this thesis SEM is performed only in secondary electron mode.

SEM has the benefit over AFM of not suffering from tip convolution and can thus obtain more accurate measurements of patterned nanostructure dimensions, a key analytical requirement for this thesis. If images are not focused and captured fairly rapidly (30-60 s depending on beam current) there is a risk that metallic structures may accumulate charge, distorting the image due to deflection of incoming electrons, but this is easily avoided with practice. SEM imaging is also much faster than AFM, images are captured in seconds rather than tens of minutes.

3.3 Micromagnetic simulation

Much of the work in this thesis involved attempting to find explanations for confusing or unexpected experimental results, or studying highly-dynamic micromagnetic processes occurring on nanosecond timescales. In both these cases, micromagnetic simulation proved an invaluable tool; both to understand what experiment was telling us and to provide insight into the fascinating nanosecond-scale world, painstakingly difficult to observe experimentally.

Micromagnetic simulations were performed using the object-oriented micromagnetic framework⁶² (OOMMF), a software package developed by Porter and Donahue at the US National Institute of Standards and Technology (NIST) in 1999 and continually updated since. OOMMF is beloved throughout the micromagnetic community for providing a powerful suite of accurate and reliable tools, capable of simulating a bewildering range of systems and conditions while retaining a high degree of user accessibility. Micromagnetic simulations played a key role in this thesis, which would not have been possible without it. Their role in elucidating otherwise unobservable nanosecond-scale dynamic processes often provided the key to understanding otherwise inscrutable behaviours.

OOMMF iteratively solves the Landau Lifshitz Gilbert equation for a finite number of three-dimensional cells or elements which the system is divided up into. The LLG equation is shown below

$$\frac{d\mathbf{M}}{dt} = -\gamma\mathbf{M} \times \mathbf{H}_{eff} + \frac{\alpha}{M_s}\mathbf{M} \times (\mathbf{M} \times \frac{d\mathbf{M}}{dt}) \quad (3.6)$$

where $\mathbf{M}$ is the sample magnetisation, M_s the saturation magnetisation, γ the gyromagnetic ratio and α a phenomenological damping parameter. H_{eff} is the effective field, arising from the demagnetising field of the sample and any external applied fields. The LLG describes the precession of spins around H_{eff} and OOMMF implements it in simulated nanopatterned systems using a stepwise Euler or higher-order Runge-Kutta method. This method assumes a constant

$\frac{d\mathbf{M}}{dt}$ for the duration of each time-step ($\delta t = 10$ ps), with the $\frac{d\mathbf{M}}{dt}$ value updated between each time step.

It can be assumed to a good approximation that spins with less than one exchange length between them will behave coherently. As such, if each cubic simulation cell is defined with a side length below the exchange length then the discretisation holds well and the finite difference approach used by OOMMF is capable of providing good agreement with experiment. At each time-step ($\delta t = 10$ ps) the magnetisation vector of each cell is perturbed relative to its six nearest-neighbours, with the lowest-energy configuration selected and carried forward to the next time-step. This process is repeated until the global change in magnetisation $\frac{\partial M}{\partial t}$ falls below a user-defined limit, at which point the system is assumed to be in a stable or meta-stable state.

The computation time for a given simulation is dependent largely on two factors, the number of simulated elements and the choice of Gilbert damping factor α . The exchange length l_{ex} is given by^{63,64}

$$l_{ex} = \sqrt{\frac{2A}{\mu_0 M_s^2}} \quad (3.7)$$

where A is the exchange length, μ_0 is the vacuum permittivity and M_s the saturation magnetisation. Taking typical parameters for NiFe of $A = 1.3 \times 10^{-11}$ J/m and $M_s = 8.6 \times 10^5$ A/m, we arrive at a value of $l_{ex} \approx 5.5$ nm. Trial simulations taking cell side-lengths of 4 nm and 2.5 nm were found to converge to the same final states with only minor superficial deviations in dynamic pathways. For thin-film based samples, computation time varies approximately by the element side-length squared, as such a value of 4 nm was generally employed throughout this thesis. The Gilbert damping factor of thin-film NiFe has been measured at $\alpha = 0.006$ using ferromagnetic resonance⁶⁵. However, realistic values of α require significantly higher computation time than higher values assuming a greater degree of damping. Simulations taking $\alpha = 0.1$ tend to produce the same final state as the realistic value for most processes and require many fewer computation cycles. The drawback is that the dynamic pathway taken between the initial and final states of the system can vary considerably for different α values. As the work in this thesis concerns highly dynamic processes, we do not have the luxury of increased damping.

As a compromise, a damping parameter of $\alpha = 0.01$ was employed, with the dynamic pathways observed showing inconsequential variation from $\alpha = 0.06$.

One key drawback of OOMMF is that it performs simulations at an effective temperature of 0 K. In practice this means that states which appear stable in simulation may be collapsed by thermal fluctuations under experimental conditions, though this is generally only the case when close to phase boundaries or with weakly-stabilised states. The micromagnetic states studied in this thesis under ambient conditions mapped closely onto their simulated counterparts produced by OOMMF.

4 | A novel method for the injection and manipulation of topological defects in ferromagnetic nanostructures

The following chapter concerns the development and implementation of a novel method for introducing and manipulating topological defects in an otherwise smooth ferromagnetic spin-system. Topological defects are fascinating physical phenomena in their own right but can also form higher-order structures containing several defects such as magnetic domain walls or Skyrmions. These superstructures provide the basis for several promising technological applications and also mediate a variety of physical processes in nanopatterned ferromagnetic systems.

Isolating new and exotic combinations of topological defects is currently of great importance. Much has been achieved with simple 180° magnetic domain walls. Designs for next-generation ‘racetrack’ data storage systems, spatial manipulation of magnetic particles and cellular automata have all been presented using 180° DWs as the fundamental functional topological defect. A 180° DW is one of the more simple stable topological defects supported in ferromagnetic spin-systems, consisting of a pair of bound $\pm 1/2$ topological charge defects. Given the results achieved by manipulating these relatively simple topological defects, researchers have been motivated to design systems exploiting the properties of more complex defects. Work examining ‘Skyrmionic’ systems has flourished in the last five years, with potential benefits

including a 10^5 reduction in required current densities relative to 180° DW based approaches. Skyrmions have the drawback of requiring exotic and high quality single-crystal materials or multi-layered systems to host them and are difficult to stabilise at room temperatures. Another promising topological defect is one lying somewhere between the simple 180° DW and the exotic but challenging-to-realise Skyrmion. Consisting of a bound pair of opposite chirality 180° DWs, the 360° DW adds additional functionality not observed in a single 180° DW and is stable in conventional ferromagnetic materials at ambient conditions. An issue currently impeding research into 360° DWs is the lack of a powerful and flexible means to insert them into a system. The following chapter lays out and demonstrates a remedy for this; a novel SPL-based approach for the injection and spatial manipulation of 360° DWs in nanowire-based systems composed of NiFe and Co.

4.1 Abstract

Realising the promise of next-generation magnetic nanotechnologies is contingent on the development of novel methods for controlling magnetic states at the nanoscale. There is currently demand for simple and flexible techniques to access exotic magnetisation states without convoluted fabrication and application processes. 360° domain walls (metastable twists in magnetisation separating two domains with parallel magnetisation) are one such state, which is currently of great interest in data storage and magnonics. Here, we demonstrate a straightforward and powerful process whereby a moving magnetic charge, provided experimentally by a magnetic force microscope tip, can write and manipulate magnetic charge states in ferromagnetic nanowires. The method is applicable to a wide range of nanowire architectures with considerable benefits over existing techniques. We confirm the method's efficacy via the injection and spatial manipulation of 360° domain walls in Py and Co nanowires. Experimental results are supported by micromagnetic simulations of the tip-nanowire interaction.

4.2 Introduction

Magnetic nanostructures can be characterised by maps of their magnetic charge distribution. Magnetic charges arise from a combination of geometric patterning⁶⁶ and internal micromagnetic structure such as that found in magnetic domain walls (DWs)^{67,68}. Gaining a thorough understanding of magnetic charge distributions in nanostructures and techniques for their control is essential for next-generation magnetic technologies. In particular, it is important to develop powerful and flexible methods for the manipulation and control of magnetic charge. Existing methods for the injection of magnetic charge into nanostructures rely on complex solenoid-based write heads⁶⁹, subjecting whole devices containing injection pads to global field sequences^{70–75} or using complex nanowire geometries to locally apply pulsed Oersted fields^{44,76–78}. These methods are non-trivial in both fabrication and field-protocol stages and other than solenoidal write-heads inflexible in that they demand spatially fixed charge injection points, predetermined at the device design stage.

Here we report a novel method for the injection and control of magnetic charge in nanostructured magnetic material using the stray field of an external moving magnetic charge. The moving charge is provided here experimentally by a magnetic force microscopy (MFM) tip and allows for the flexibility in spatial injection point whilst providing highly precise, local injection of magnetic charges over a wide range of nanowire geometries, in addition to the spatial manipulation of existing charge structures. To confirm the viability of the method we demonstrate the injection and subsequent manipulation of a bound magnetic charge pair composed of two transverse DWs, known as a 360° DW⁷⁹. 360° DWs are currently of great interest in data storage and magnonics, as well as intriguing topological defects in their own right. They have been proposed as candidates for high-density data storage⁸⁰ including magnetoresistive random access memory (MRAM) systems^{76,81,82} and as phase-shifting⁸³ and frequency doubling⁸⁴ magnonic circuit elements as well as spin-wave generators⁸⁴. Experimental progress of such applications has been so far hampered by a lack of simple and versatile means to access 360° DW states, an issue which this method addresses. Additionally, we demonstrate an

elegant extension of the injection technique to achieve the controlled motion of 360° DWs through nanowires. Spatial manipulation of 360° DWs is not possible using uniform global fields as opposite forces are generated on each composite magnetic charge^{85–87} and current-driven motion requires high current densities^{84,88} or multiple-wire geometries⁸⁹ as well as the incorporation of macroscopic electrodes within the device architecture. A tip-based technique for writing magnetic charge structures into continuous films has recently been described⁹⁰, relying on a heated tip in conjunction with a complex multi-layered film and requiring a considerable applied global magnetic field to function. Here we provide a simple and versatile alternative to existing techniques. While this work was under review, a methodology for reconfiguring a nanopatterned magnetic charge array using an MFM tip along with global magnetic fields was described⁴. The technique is essentially global field mediated with a localised field enhancement provided by an MFM tip raised a distance above the sample. The technique described in this work requires no global field and as such offers considerable benefits.

4.3 Initial observation of tip-nanowire interaction

Before embarking on a detailed description of the 360° DW injection and manipulation technique, it may be interesting to describe the circumstances leading to its discovery. As is the case for many scientific endeavours, this one was born purely from accident. While attempting to MFM image 180° DWs in Py nanowire-based architectures for an unrelated project, it was noticed that on occasion the magnetic charges at nanowire ends or vertices would reverse polarity while under the MFM tip. Deciding to examine this further, it was noticed that the magnetic charges would reverse both while directly under the MFM tip and also when the tip was further from the magnetic charge.

Figure 4.1 shows various MFM images from the initial session where magnetic charge switching was observed. Figure 4.1 a) was performed with the ‘slow’ axis (i.e. the axis along which the tip will incrementally step only after completing each constituent scan line of the raster image) of the MFM raster scan directed downwards, or in the negative y-direction. The first magnetic

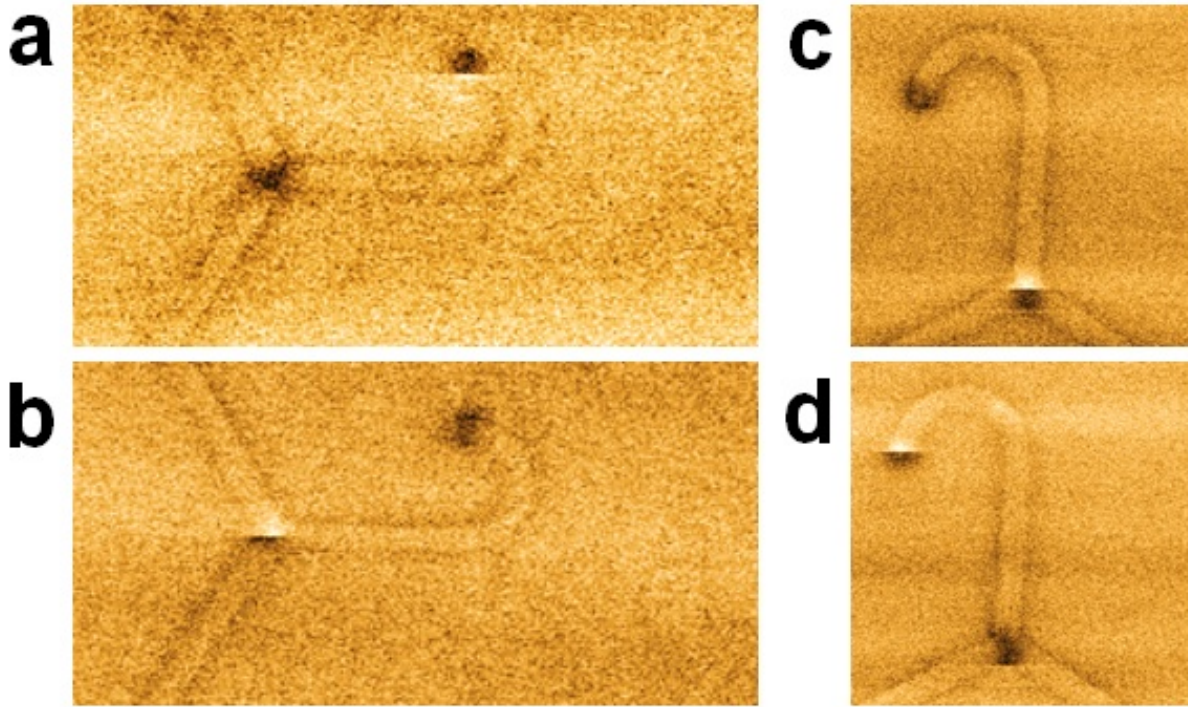


Figure 4.1: MFM images of 100 nm wide, 15 nm thick Py nanowires showing reversal of magnetic charge polarity while imaging. ‘Fast’ axis of raster-scan is horizontal in all images.
a) Magnetic charge at the end of curved nanowire reverses while imaging.
b) Magnetic charge at the nanowire vertex reverses while imaging.
c) Scan-angle is now rotated 90 ° relative to a) and b). Vertex magnetic charge reverses while imaging.
d) Magnetic charge at end of curved nanowire reverses while imaging.

charge encountered is the one at the end of the curved ‘walking stick’ style nanowire. At first the magnetic charge contrast is observed to be dark, indicating a negative magnetic charge exerting an attractive force on the MFM tip. After several raster-scan passes of the tip over this charge, its contrast changes to light, indicating the tip-charge interaction has now become repulsive and the tip and charge are now of matching polarity. The magnetic charge at the vertex remains dark throughout the scan. Figure 4.1 b) was performed immediately after this, but with the slow-axis of the raster scan now directed upwards or in the positive y-direction. The magnetic charge at the vertex is initially dark, as expected, but then reverses its contrast after a few passes of the MFM tip. The magnetic charge at the curved-wire end is now observed to be dark, rather than light as it was last observed.

Figure 4.2 a) and b) show another example of this behaviour at higher magnification. Figure 4.2 a) was performed first with the slow-axis of the scan directed ‘up’ the page, with the vertex charge last observed with light contrast. A ‘down’ scan was then performed, shown in figure 4.2

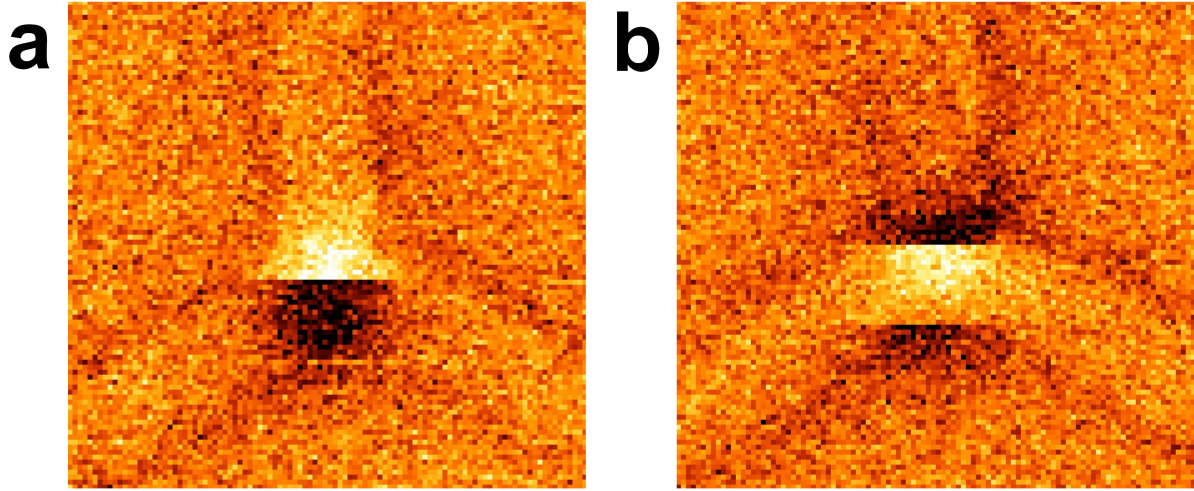


Figure 4.2: MFM images of 100 nm wide, 15 nm thick Py nanowires showing reversal of magnetic charge polarity while imaging. ‘Fast’ axis of raster-scan is horizontal in both images.

- a)** Magnetic charge at nanowire vertex reverses while imaging.
- b)** Magnetic charge at nanowire vertex reverses twice while imaging.

b). The vertex is now initially observed to be dark, before switching polarity twice as the MFM tip passes over it.

This phenomenon could be caused by one of two processes - either the MFM tip has had its magnetic charge polarity reversed, or the magnetisation of one or more nanowires in the system has been reversed. To test which one of these cases was occurring, the MFM scan angle was rotated such that both the magnetic charges at the vertex and curved wire-end were being imaged at the same MFM scan line. Figure 4.3 shows a scan performed at such a scan angle. The wire-end charge can be seen to reverse polarity after several passes of the MFM tip, while the vertex charge remains constant. This shows the MFM tip magnetisation is not reversing, as this would cause the simultaneous reversal of both wire-end and vertex charge polarity. Instead, the magnetisation reversal of one or more nanowires in the system must be responsible for the observed polarity switching. This realisation piqued my interest, which physical processes could be responsible for the local reversal of magnetisation in a nanowire? The nanowires seemed too large for the highly-localised field of the MFM tip to be driving a conventional field-based reversal. One idea which seemed plausible was that the tip may somehow be locally distorting the magnetisation such that topological defects were being introduced to the spin-system, which may then play a role in driving the magnetisation reversal process. Arrays of isolated nanowires

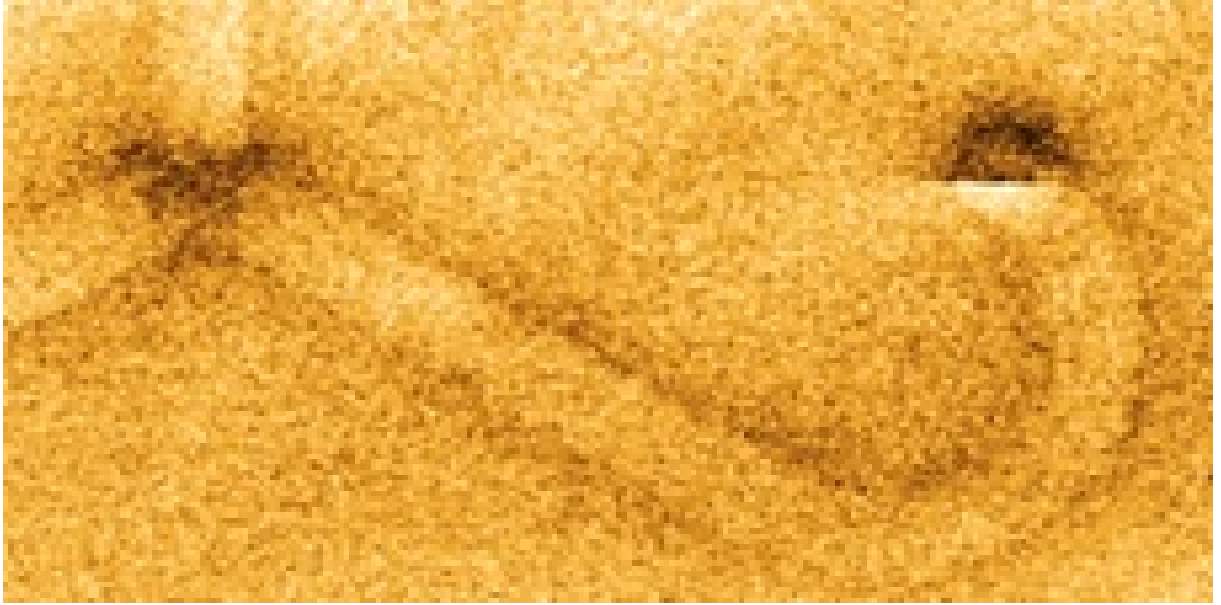


Figure 4.3: MFM image of 100 nm wide, 15 nm thick Py nanowires showing reversal of magnetic charge polarity in a single magnetic charge while imaging two charges. This shows that magnetic reversal of the nanowire rather than the MFM tip is responsible for magnetic charge polarity switching. ‘Fast’ axis of raster-scan is horizontal.

were then fabricated to simplify observation and analysis of any magnetic switching processes which may occur. The results of the following study are detailed below.

4.4 Experimental demonstration of the injection process

Initially the nanowires were saturated using a global magnetic field applied along their axis.

Figure 4.4 a) shows a low-moment (LM) tip MFM scan of a typical nanowire (here Co) after the application of this global field. The dark and light contrast at the ends of the wire show the location of the magnetic charges and in this case confirm the expected single-domain state prior to injection.

Figure 4.4 b) shows an MFM scan of the same nanowire performed using a high-moment (HM) tip. In this case the dark contrast throughout the wire shows the tip is attracted to the wire, suggesting that a magnetic charge is present under the tip at all points of the scan along the nanowire. A line profile taken along the wire is shown below the MFM image. The continuous contrast throughout the centre of the wire is seen to have twice the magnitude of the end

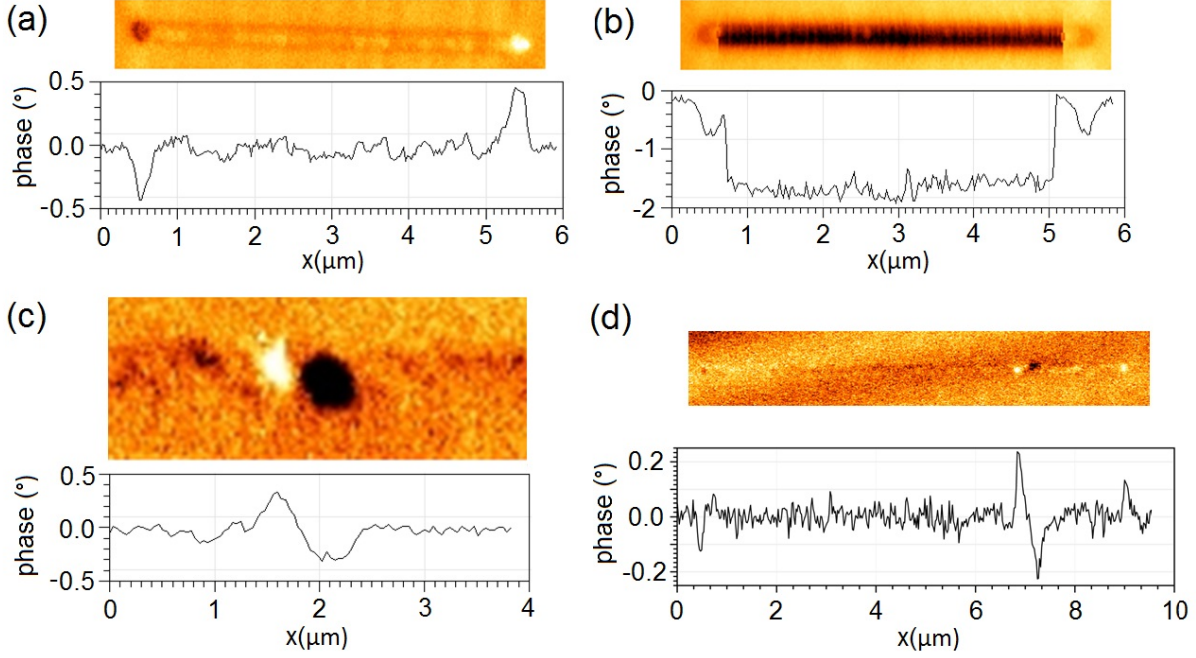


Figure 4.4: MFM images of Co nanowires in imaging (LM) and injection (HM) modes. Dark contrast marks regions where a negative phase shift was required to maintain constant phase in the cantilever oscillation. This corresponds to an attractive tip-sample interaction, with light contrast indicating a repulsive interaction. **a)** LM-MFM image of a 5 μm nanowire magnetised in-plane. No DWs are present. **b)** HM-MFM image of same 5 μm nanowire. Dark contrast throughout the wire indicates DW injection. **c)** LM-MFM image taken after HM injection process. Adjacent regions of opposing magnetic charge are observed, corresponding to an injected 360° DW. **d)** LM-MFM image after HM injection process showing a whole nanowire containing a 360° DW. Wire end charges are seen to have half the magnitude of the injected DW. **a)-c)** show 150 nm wide nanowires, **d)** shows a 100 nm wide wire. The HM injection scan leading to the state shown in **d)** was performed in a contact mode with no oscillation of the tip-height while the injection scan leading to the state shown in **c)** was performed in a tapping mode with resonant oscillation of the tip-height, no difference is observed between the resultant 360° DW states. Further discussion of tapping and contact modes is provided in the methods section.

charges. As DWs possess twice the magnetic charge of a geometrical wire-end charge, this suggests a DW has been introduced to the wire (the central portion of the wire was confirmed to be initially charge free by a prior LM-MFM scan). After the HM scan, the tip is retracted at the midpoint of the wire. In figure 4.4 c) a subsequent LM-MFM scan is performed, showing a pair of opposing polarity magnetic charges present in the wire. Again, these are each twice the magnitude of the end charges, indicating head-to-head and tail-to-tail 180° DWs in close proximity. The two 180° DWs must have the necessary opposing chiralities to form a stable 360° DW state as oppositely charged DWs of matching chirality will simply attract and annihilate⁸⁷. Figure 4.4 d) shows an entire nanowire with an injected 360° DW. The constituent 180° DWs can clearly be seen to possess twice the magnitude of the end charges and no other magnetic charges are present in the wire.

These three stages demonstrate experimental confirmation of the injection process: LM-MFM imaging of the wire prior to injection, an HM-MFM scan to inject magnetic charges and a post-injection LM-MFM imaging of the wires showing the injected charge structure. It was found that allowing the HM tip to perform subsequent scan lines in a raster fashion did not lead to the injection of additional 360° DWs with each line. This has the benefit that 360° DWs may be precisely placed by allowing the tip to raster scan until the desired injection location is reached at which point the HM-tip is retracted, leaving a single 360° DW. The scan shown in figure 4.4 c) was performed at the retraction point of a previous HM scan. Micromagnetic simulations of scans on wires containing an existing 360° DW show the tip field causing the DW to collapse before the tip reaches the wire (discussed further in the simulation section below). The tip then injects a new 360° DW as it crosses the wire, leaving a single 360° DW in the wire as observed experimentally. Web-links to videos of the time evolution of this process is included in the ‘micromagnetic time-evolution videos’ section at the end of this thesis.

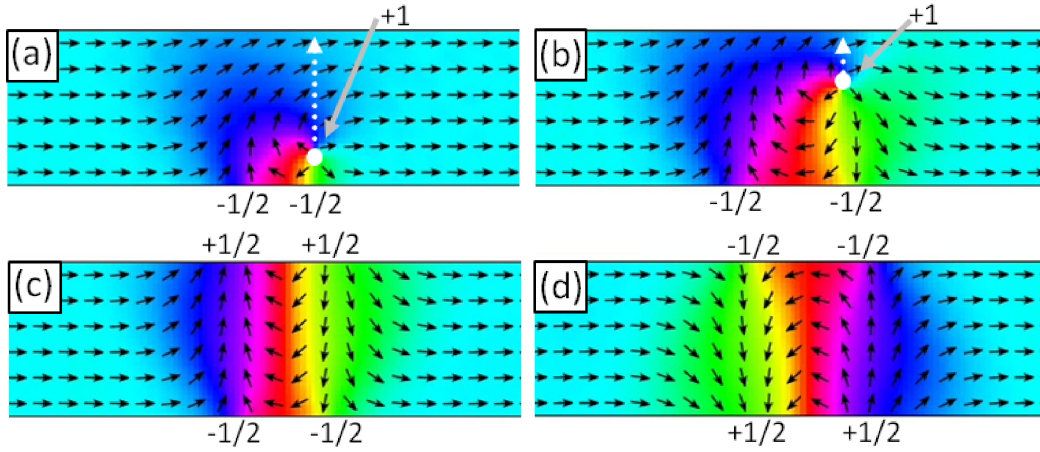


Figure 4.5: The micromagnetic structure of a nanowire distorted by the movement of a magnetic charge. Topological defects present are labelled with their winding numbers. The moving magnetic charge and its direction of motion are represented by the white arrow and dotted line respectively. The wire simulated here is 150 nm wide by 10 nm thick, with the material properties of Py. (a) and (b) show how the arrangement of topological defects within a nanowire can be manipulated by the passing charge. (c) and (d) show the final micromagnetic structures of 360° DWs with opposing chirality, resulting from the charge moving across the wire in up and down directions respectively. Web-links to videos of the time evolution of this process is included in the ‘micromagnetic time-evolution videos’ section at the end of this thesis.

4.5 Micromagnetic simulation of the injection process

Micromagnetic simulations provide further insight into the physical behaviour taking place whilst the field from a monopole-like tip interacts with the nanowire structure. Figure 4.5 shows a series of micromagnetic configurations of a nanowire as (a) and (b) a magnetic charge passes over the nanowire and (c) and (d) 360° DW structures stabilise in the nanowire following the interaction with the moving magnetic charge. Web-links to videos of the time evolution of this process is included in the ‘micromagnetic time-evolution videos’ section at the end of this thesis.

We find that the MFM tip locally distorts spins in the nanowire out of the homogenous ferromagnetic state and into a conformation following that of the monopole-like tip field as described by Magiera et al^{91,92}. This introduces a magnetic vortex-like structure into the magnetisation texture which stabilises into a 360° DW once the tip has crossed the wire.

It is perhaps clearest to describe the injection process in terms of dynamic topological defects⁴¹, both those with integer winding numbers which are free to move within the wire and those with fractional winding numbers, bound to the wire edges. Topological defects in thin-film

ferromagnets are points at which spins diverge from a uniform collinear texture in a manner which cannot be smoothly unwound. Each defect has an associated winding number describing the manner in which spins locally diverge. The net winding number of a system is rigorously conserved and in a nanowire must sum to zero¹. In figure 4.5 a) the monopole-like field of the moving magnetic charge (illustrated by the solid white circle) distorts the uniformly magnetised micromagnetic structure of the nanowire. A spin rearrangement to lower the Zeeman energy in the vicinity of the magnetic charge results in the formation of a topological defect with a +1 winding number^{91,92} directly under the moving charge. The introduction of this defect at the wire's lower edge is accompanied by the formation of two additional topological edge defects with $-1/2$ winding numbers in order to conserve the net winding number of the wire. These fractional defects remain bound to the lower edge of the wire, this can be understood by examining the energetics as their leaving the edge would create a discontinuity in the magnetisation along a line between the wire edge and the defect with a large associated energy penalty¹. As the magnetic charge moves across the nanowire, figure 4.5 b) shows the +1 defect following the movement of the charge. On reaching the upper edge of the wire the +1 defect can no longer follow the motion of the magnetic charge and separates into two +1/2 topological defects, bound to the wire edge as seen in figure 4.5 c). Lines of spins oriented perpendicular to wire's length are seen connecting each $-1/2$ defect to the corresponding +1/2 defect on the opposite edge of the wire. Each line and connected pair of defects represents a 180° DW, hence the resultant micromagnetic structure created by the injection process is a bound state of two 180° DWs forming a stable 360° DW, here with an anti-clockwise chirality. The movement of the tip across the wire in the downwards direction results in the injection of a 360° DW with reversed chirality. The final state of this process is shown in figure 4.5 d). The sign on the topological defects is inverted with respect to c) and the magnetisation now rotates in a clockwise direction. As such our method provides chirality-selective injection, key to controlling DW dynamics in nanowire-network systems^{93,94}.

Simulations were performed here with the moving magnetic charge (modelled as a point-source monopole) at a constant height above the nanowire. Experimental injection scans were performed with the tip in both a non-tapping contact mode and in tapping mode. The contact mode simplifies the dynamics of the injection process and better corresponds to simulation. Figure 4.4

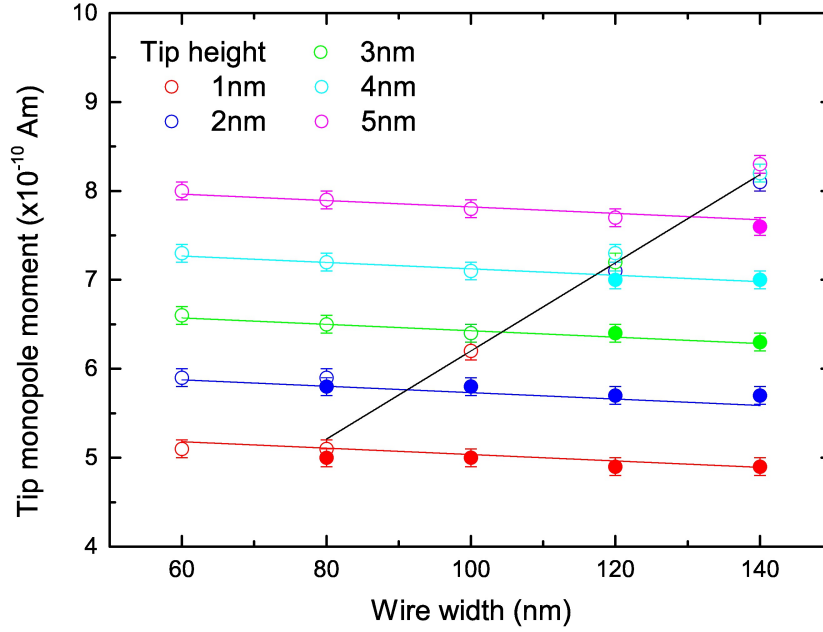


Figure 4.6: Magnetic charge magnitude required to inject a 360° DW into a 10 nm thick Py nanowire. Points represent boundaries between regions of differing injection behaviour. Open points represent the lowest tip monopole moment required for successful injection of a stable 360° DW structure. Closed points indicate the lowest tip monopole moment required for injection of unstable 360° DW structures, which then collapse once the moving tip charge has passed the wire. The critical tip charge strength required for stable injection is denoted by the black line.

d) shows a 360° DW state injected using a contact-mode injection scan. However, to improve imaging quality and reduce tip damage most injection scans were performed in a tapping mode where the tip height is constantly varied by oscillating the MFM tip cantilever at its resonant frequency. Figure 4.4 c) shows a 360° DW injected using a tapping-mode injection scan. No difference in the resultant 360° DW state was observed between tapping and contact modes.

The dependence of the nucleation process upon the properties of the MFM tip was investigated through further simulation. Figure 4.6 plots the final micromagnetic state of the nanowire following the pass of the magnetic charge for a range of nanowire widths, magnetic charge magnitudes and charge-wire height separations.

For small magnetic charges, the injection of a 360° DW does not occur but above a critical charge magnitude a 360° DW is injected. The points in figure 4.6 represent the minimum tip moment required to inject a 360° DW into the nanowire, with the open points denoting 360°

DWs which collapse after injection once the moving tip charge has passed the wire and closed points denoting 360° DWs which remain stable. Let us first consider the tip strength $P_{injection}$ required for the initial injection of a 360° DW, its trends shown by the coloured lines with slight negative gradient. This critical injection strength increases with increased tip-wire separation, understandable as the tip-field strength at the wire surface decreases as the tip charge is raised higher above the wire. The critical injection strength decreases with increased wire width, potentially due to reduced exchange energy (or shape anisotropy if one prefers) penalty at the nanowire edge as the edge spins constitute a smaller fraction of the wire for greater wire widths. The coloured lines in figure 4.6 show a fit to the data using the simple phenomenological form $P_{injection} = \alpha h + \beta w + \gamma$ where P is the critical charge strength, h the height of the charge above the wire and w the wire width. Here $\alpha = 69.6 \pm 0.8$ A, $\beta = -360 \pm 40$ μ A and $\gamma = 4.70 \pm 0.05$ Am.

The second, more useful from a practical standpoint, critical strength is that required to introduce a 360° DW which will remain stable in the wire long after the tip charge has passed. This critical stable injection strength P_{stable} is denoted by the solid black line and increases with nanowire width but as expected shows no magnetic charge / nanowire separation dependence as the 360° DW stabilisation process occurs once the tip has already passed the nanowire. This dependence is of the form $P_{stable} = \beta w + \gamma$ where $\beta = 5.0 \pm 0.2$ A and $\gamma = 1.2 \pm 0.2$ Am.

To investigate why each raster line of the HM-MFM injection scan does not inject an additional 360° DW an injection process was simulated with an existing 360° DW located at the point where the tip-charge crosses the wire. Figure 4.7 a) shows a nanowire containing a stable 360° DW. When a moving magnetic charge approaches the wire, its local field forces the spins forming the fractional edge defects on the near edge of the wire to align with the monopole-like field, the reduction in Zeeman energy overcoming the potential binding the fractional defects to the wire edge. The fractional defects then combine into an integer vortex defect, shown in figure 4.7 b). The vortex defect is forced across the wire as the spins around it relax to a collinear ferromagnetic state, lowering their exchange energy. Upon reaching the far side of the wire the vortex meets and annihilates the opposite polarity fractional defects bound to the far edge,

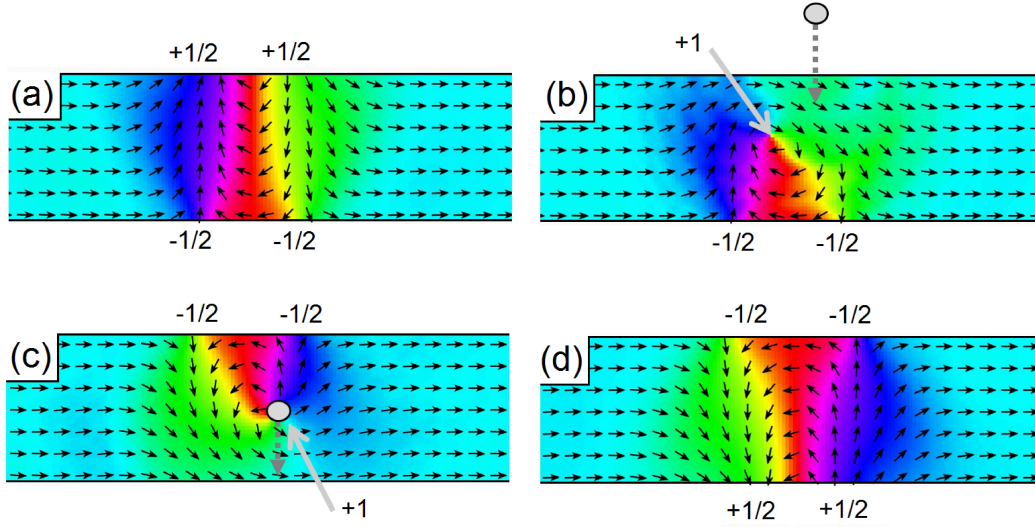


Figure 4.7: An existing 360° DW is destroyed by an approaching magnetic charge before a new 360° DW is injected. Topological defects are labelled with their winding numbers, the moving charge and its direction of motion are represented by the grey circle and dotted arrow respectively. **a)** shows an initial stable 360° DW which is caused to collapse by an approaching magnetic charge in **b)**. The charge then crosses a DW-free wire **c)**, effecting the same injection process depicted in figure 4.5 and leaving a new stable 360° DW **d)** after passing the wire. Web-links to videos of the time evolution of this process is included in the ‘micromagnetic time-evolution videos’ section at the end of this thesis.

leaving no DW in the wire. This process occurs before the moving magnetic charge reaches the wire, hence the charge encounters a wire in a collinear spin state and a new injection process occurs as normal **c)**, leaving a single stable 360° DW in the wire **d)** after crossing. The tip-mediated collapse of existing DWs presents itself as a DW deletion method if the tip is halted after collapsing the existing DW, but before reaching the wire.

4.6 Experimental demonstration of spatial manipulation

While the weaker magnetic charge associated with a low moment tip is not sufficient to nucleate 360° DWs, it can still influence pre-existing DW structures. Here we present further MFM data and micromagnetic simulations showing the controlled movement of DWs using lower magnetic tip charge values. Note that the overall magnetic charge of a 360° DW is zero as its composite 180° DWs carry equal and opposite charge. However the 180° DW closer to the tip-charge will experience a stronger interaction with the tip, resulting in a net force on the 360° DW and thereby facilitating tip-mediated motion.

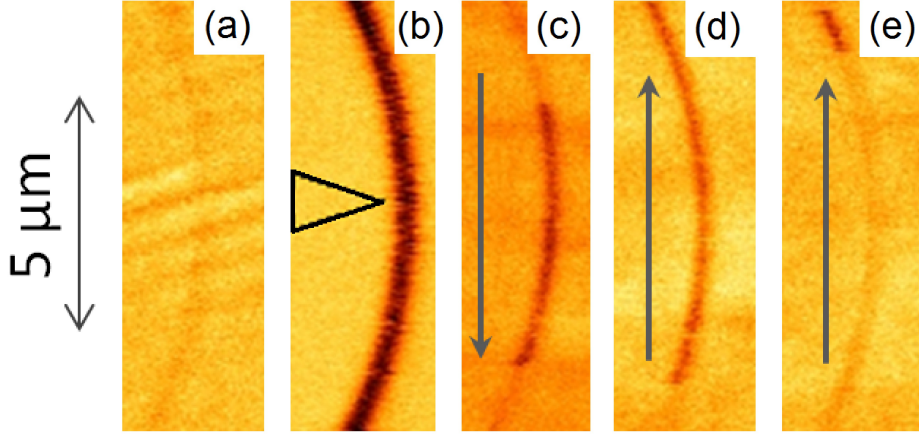


Figure 4.8: Series of MFM images illustrating 360° DW injection and spatial manipulation in a Py nanowire. The scale bar to the left corresponds to all images. **b)** uses an HM-tip, other images LM. The black triangle in **b)** indicates the point of tip retraction. Prior to retraction, the wire was raster scanned from the top to the bottom of the image frame to collect the image data before scanning half-way back up the frame and retracting at the point marked by the black triangle. The grey arrows in **c)-e)** indicate the slow-axis of the raster scan direction. Scans were performed in chronological order from **a)-e)**. Wire curvature is not a necessary condition for 360° DW injection and manipulation, the same dynamics are observed in straight wires.

Figure 4.8 shows a sequence of MFM images of a 150 nm wide wire of radius of curvature $10\ \mu\text{m}$. The sequence depicts the same initial LM imaging **a)** and HM injection **b)** processes as described in the injection section above, the black arrow overlaid in **b)** indicates the final position of the HM tip prior to retraction. However, here the nanowire material is Py rather than Co. As Py is a softer magnetic material than Co, the LM tip previously used for imaging injected DWs instead achieves spatial manipulation of injected magnetic charges. Note that MFM images are performed in a raster fashion with each subsequent horizontal line measured at a later time. This allows the imaging of dynamic processes as seen below. Figure 4.8 **c)** shows a scan moving from top to bottom of the image (direction indicated by the overlaid grey arrow) and shows the LM tip collecting a DW at $2\ \mu\text{m}$ from the HM retraction point via an attractive magnetostatic interaction between tip and DW magnetic charges. The DW charge then follows the motion of the tip down the wire before stopping a few μm from the bottom of the image, potentially due to pinning at a defect. Figure 4.8 **d)** is a subsequent scan moving up the wire. We observe a magnetic charge being collected by the tip close to its final position in **c)** before moving with the tip up the length of the wire. To rule out the possibility that our contrast shows the wire becoming magnetised out of plane rather than a dynamic image of a moving DW we retracted the tip to $200\ \mu\text{m}$ above the sample where the tip-sample interaction is negligible before moving

the tip to the bottom of the wire. We then perform a second upwards scan, shown in e). As expected, the wire remains in an in-plane state. No magnetic charges are observed until 1 μm from the tip-retraction point at the top of d) where the charge is again collected by the tip. This shows we have successfully moved an injected magnetic charge through a nanowire and are able to deposit it at a desired position before re-collecting later if desired. This feature allows for the flexible re-configuration of a device's magnetic charge distribution with an accuracy of 1-2 μm (the distance at which charges are attracted to the tip), a considerable advantage which is not achievable using existing DW injection techniques.

Here we have used a LM-tip to achieve spatial manipulation in Py. The same process is observed in Co wires using slightly stronger commercially available 'normal' moment (NM) MFM tips.

4.7 Micromagnetic simulation of spatial manipulation

Figure 4.9 plots the displacement of a DW along a wire following an interaction with a moving magnetic charge which crosses the wire at varying lateral distances from the DWs initial position. The displacement of the 180° DW is found by measuring the change in M_x for the wire whilst the position of the 360° DW is found from the final micromagnetic configuration by taking the point of maximum M_y magnetisation component along the nanowire axis. Web-links to videos of the time evolution of this process is included in the 'micromagnetic time-evolution videos' section at the end of this thesis.

The 180° DW has a magnetic charge which is repelled from the moving magnetic charge representing the tip (an oppositely magnetised tip or opposite polarity domain wall would lead to an attractive interaction). When the scan is near the DW location, the DW can be displaced by distances up to 800 nm. As the point at which the moving charge crosses the wire moves further from the DW, the magnitude of this displacement decreases. For the 360° DW structure, while a uniform magnetic field cannot induce a net force on the DW (due to opposite forces generated on each composite magnetic charge as described above) the localised field from the tip has a stronger influence on the closest of the two magnetic charges, leading to a weak displacement

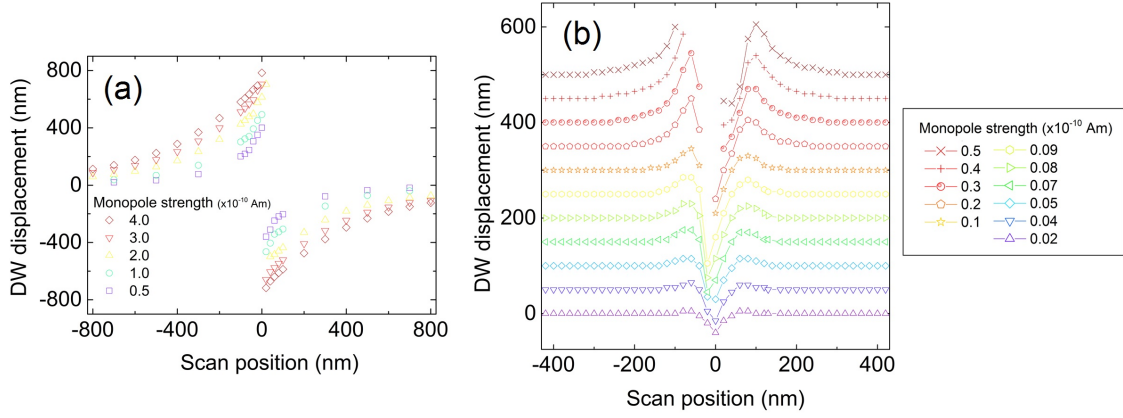


Figure 4.9: Spatial manipulation of a) an up-chirality transverse head-to-head domain wall and b) a clockwise-chirality 360° domain wall in a 100 nm wide nanowire by a passing monopole-like magnetic charge. In the 360° DW case the majority of the wire is magnetised in the positive x-direction. Lines representing different monopole strengths in b) have been separated along the y-axis for ease of viewing, all monopole strengths give a DW displacement of zero at scan positions of ± 425 nm. Missing points in b) denote a destruction of the existing 360° DW by the magnetic charge. A scan position of 0 indicates that the magnetic charge crosses the wire directly at the location of the existing DW. The magnetic charge is suspended 5 nm above the wire.

effect. This effect is evident in figure 4.9 b) where tip-crossing operations performed to the left and right side of the DW both result in displacements in the positive x-direction (this is reversed to displacements only in the negative x-direction by reversing the tip-charge polarity or the initial magnetisation of the nanowire). Again, the propagation distance decreases when the moving magnetic charge is further from the DW structure. This behaviour holds for scan positions greater than around the nanowire's width away from the existing DW, however below this limit different dynamics are observed. The central region about the y-axis in figure 4.9 b) shows maximum positive displacements either side of zero, between which negative displacements are observed. The negative displacement reaches a maximum magnitude close to zero with a slight asymmetry caused by the moving magnetic charge experiencing attractive and repulsive interactions respectively to the opposite polarity constituent 180° DWs. The asymmetry is reversed about the y-axis by switching the polarity of the moving magnetic charge or the whole 360° DW. The shape of the central region of figure 4.9 can be understood as the 360° DW experiencing a point of minimum energy when the constituent 180° DW of opposite polarity to the moving magnetic charge lies directly under the moving charge. At tip-crossing points within the finite width of the 360° DW, the DW will shift its position to reach this minimum energy point. The 180° DW with a matching polarity to the tip will move away while the opposing

polarity 180° DW will approach the tip-charge, allowing for motion of the 360° DW in both directions along the wire. For moving magnetic charge strengths above 9×10^{-12} Am the field of the moving charge can cause the $\pm 1/2$ topological edge defects on the near side of the wire to become unbound, leading to a collapse of the 360° DW as described above and a reversion to a uniformly magnetised state. These events occur when the moving charge crosses the wire close to the DW and are represented in figure 4.9 by missing points in the central region. Experimentally, this DW deletion behaviour was not observed while using LM-MFM tips and as such we believe the spatial manipulation operations demonstrated are in the sub- 9×10^{-12} Am regime.

4.8 Angular dependence of injection process

Investigations were made into varying the angle of the tip scan vector during the injection process. Figure 4.10 a) shows an array of six 150 nm wide wires scanned at angles from 0° to 30° in 10° increments. The tip scan vector is represented by the blue arrow and remains constant as the wires are rotated. At a scan angle of 0° the tip is moving perpendicular to the length of the wire. Dark contrast indicating injection is occurring throughout the wire, with DWs injected during the AFM scan-line remaining in the wire for the duration of the MFM scan-line except at short $\sim 1 \mu\text{m}$ regions at either end of the wire, presumably due to magnetostatic attractions from the wire end charges interfering with the injection process or pulling injected DWs into the ends where they will annihilate before the MFM scan is completed. As the scan angle is increased, the light region at the top of the wire remains unchanged whereas the bottom light region increases in length. Interestingly, negative scan angles cause a lengthening of the light regions at the top of the wires while the bottom regions remain fixed. A plot of the no-injection length against scan angle is shown in figure 4.11.

This phenomenon was investigated further through simulation. Scan angles of -60° to 60° were simulated in $\pm 10^\circ$ increments. Moving the scan angle away from 0° was seen to vary the distribution of momentum imparted to the component 180° DWs. At a scan angle of 0° , a combination of positively charged tip and left-magnetised wire (as depicted in 4.12 resulted in all

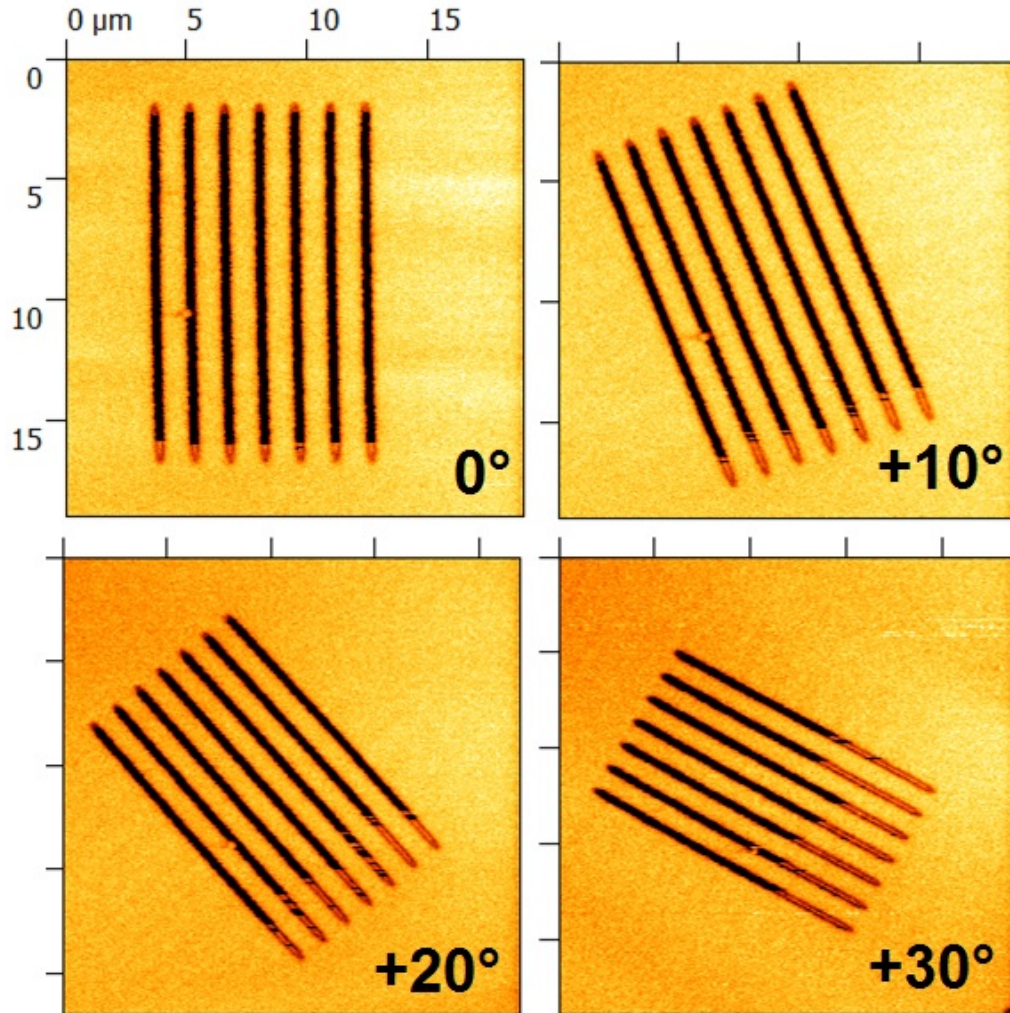


Figure 4.10: a) MFM images with the tip scan angle varied from 0° to 30° in $+10^\circ$ increments. Light regions of wire indicate no magnetic charge is present during MFM scan line. Wires are magnetised towards the top edge of the image and the MFM tip is magnetised into the page.

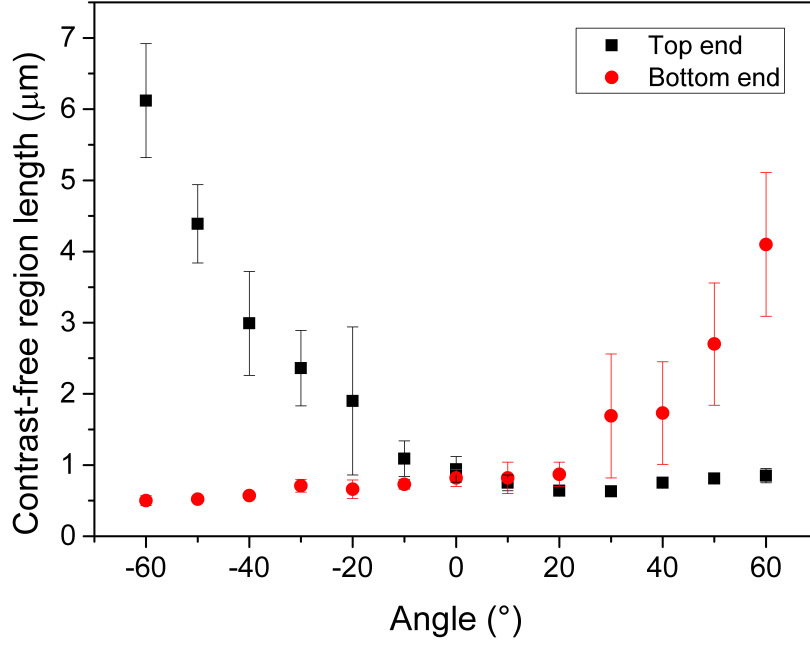


Figure 4.11: Length of wire end-regions exhibiting no dark MFM contrast versus MFM scan angle. For positive scan angles (as shown in the figure 4.10 MFM series) the bottom ends of the wire become increasingly contrast free, while for negative scan angles the top of the wires gain a magnetic charge free region.

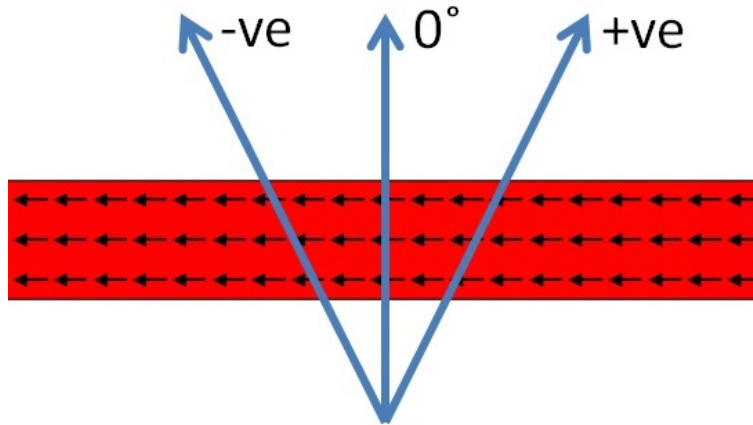


Figure 4.12: Schematic illustrating how negative and positive MFM scan angles have been defined. A positive angle has the component of the tip-velocity along the wire length directed anti-parallel to the initial magnetisation of the wire.

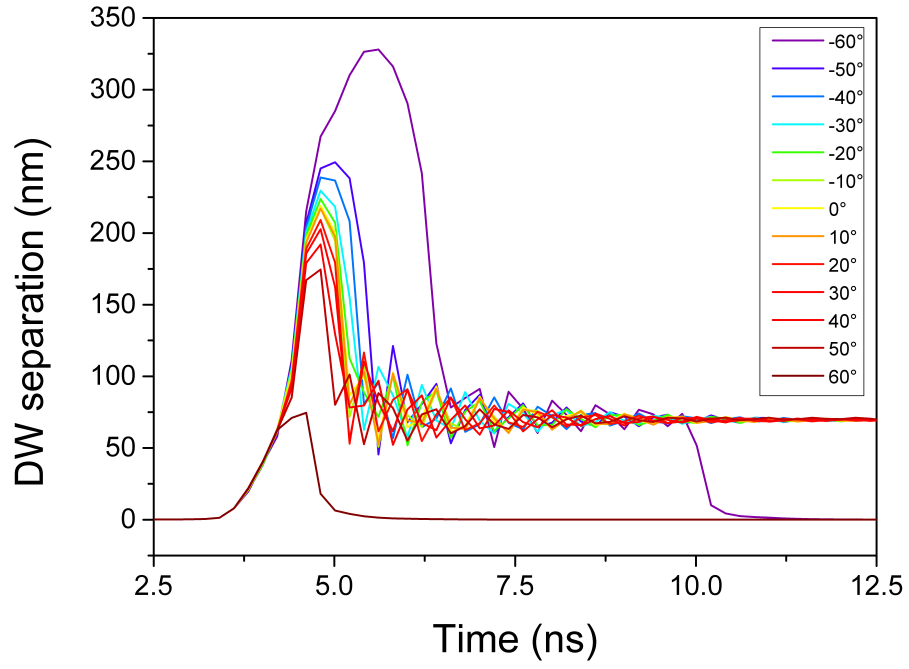


Figure 4.13: Time-varying inter-DW separation during OOMMF-simulated injection process for $2.5\ \mu\text{m} \times 100\ \text{nm} \times 10\ \text{nm}$ nanowire. MFM scan angles of -60° - 60° were simulated with larger peak separations observed at negative scan angles.

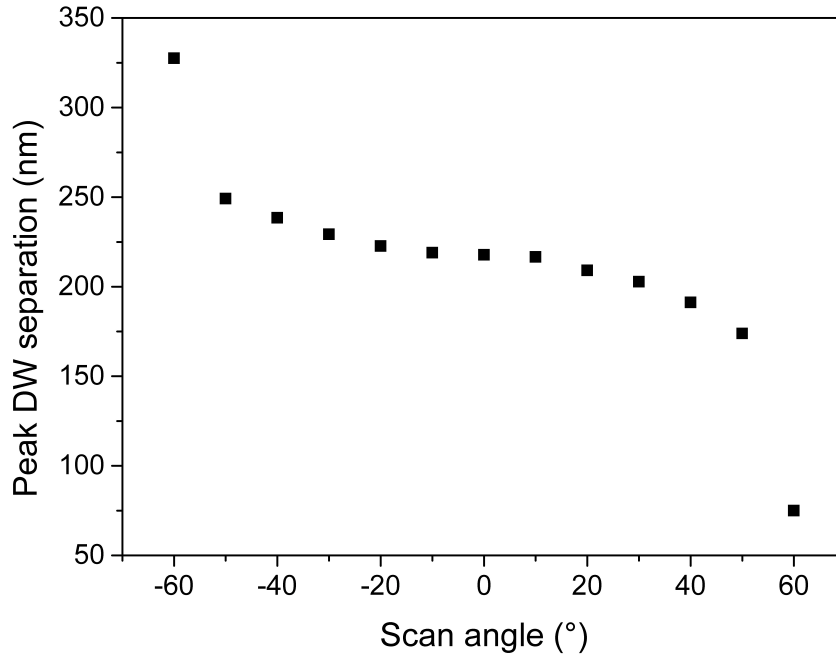


Figure 4.14: Peak DW separations reached during simulated injection events at various scan-angles. Data from same simulations shown in figure 4.13.

momentum being imparted to the right-hand DW, which is accelerated in the positive x-direction towards the right-hand end of the wire. The left-hand DW remained essentially static, save for a slight motion to the right towards the end of the injection process, arising out of magnetostatic attraction to the right-hand DW. As the scan angle was increased in the positive direction, i.e. with a component in the positive x-direction being added to the tip velocity, the left-hand DW begins to gain momentum directed along the x-component of the tip velocity. As both DWs now have momentum in the positive x-direction, the peak DW separation is decreased. This can be seen from the varying peak heights in figure 4.13, with peak separation values for each scan angle extracted and presented in figure 4.14. The peak separation decreases with increasing scan angle up to 50° as the left-hand DW receives more momentum in the direction of the tip-velocity. At 60° the left-hand DW receives enough momentum such that it collides with the right-hand DW, resulting in the collapse of both DWs. Considering now negative scan-angles, the same process occurs by which the left-hand DW receives momentum directed along the x-component of the tip-velocity. This leads to increased DW separations as the two DWs now move in opposite directions along the wire.

The observed results have two important implications, one is that the inter-DW separation during the injection process is tunable to a good degree just by varying the angle at which the MFM tip crosses the wire. The second is that the net momentum of the two DWs (and therefore the final resting position) may be controlled. At a scan angle of 0° the net momentum is in the positive x-direction, meaning that the resultant 360° DW will come to a rest to the right of the tip-crossing point. This may be undesirable if one wishes to insert a 360° DW at the exact point the tip crosses the wire. In this case, one has only to use a negative scan angle to impart some momentum to the left-hand DW in the negative x-direction, resulting in a net momentum of zero and a resultant stable 360° DW placed at the point the user desires.

The simulation results seem to explain the experimentally observed behaviour. If one accepts that there is a threshold distance from the wire end-charges, below which a DW will be pulled into the wire end via magnetostatic attraction and annihilated as opposite topological charges meet, then it stands to reason that DWs passing this point will not survive long enough after

injection to be imaged on the interleaved MFM scan line. In figure 4.10 the combination of an MFM tip magnetised into the substrate plane and wires magnetised towards the top of the image will result in a net DW momentum directed towards the bottom of the image. This matches with the experimentally observed behaviour. At a 0° scan angle the charge-free region at the bottom of the wires is longer than at the top, suggesting that the DWs are crossing the magnetostatic attraction threshold at injection points further from the wire ends due the momentum imparted to DWs during injection. As the scan angle varies, this effective magnetostatic attraction threshold moves further from the wire ends as additional momentum is imparted to the DWs during the injection process. At negative scan angles the process occurs in reverse, with the charge-free area at the top of the wires increasing in length as momentum directed towards the top ends of the wires is imparted during the injection process. If either the tip or nanowire magnetisation direction is reversed, the relation between scan angle and imparted momentum is reversed.

4.9 Effect of nanowire width on width of 360° DWs

Finally, injection simulations were performed on wires of varying widths. Figure 4.15 shows the variation of inter- 180° DW spacing or 360° DW width with nanowire width. A linear relationship between the two was observed of the form $w_{360^\circ DW} = 0.89w_{wire}$.

4.10 Discussion

In conclusion, we have performed experimental and micromagnetic studies on the interactions between a moving magnetic charge and a ferromagnetic nanowire with in-plane anisotropy. The spins in the nanowire experience the localised monopole-like field of the moving charge, leading to a change in spin orientation. With the movement of this charge over boundaries at the nanowire edges we show how topological defects can be introduced to a nanowire, creating micromagnetic structures such as 360° DWs. Our experimental work shows this process can be

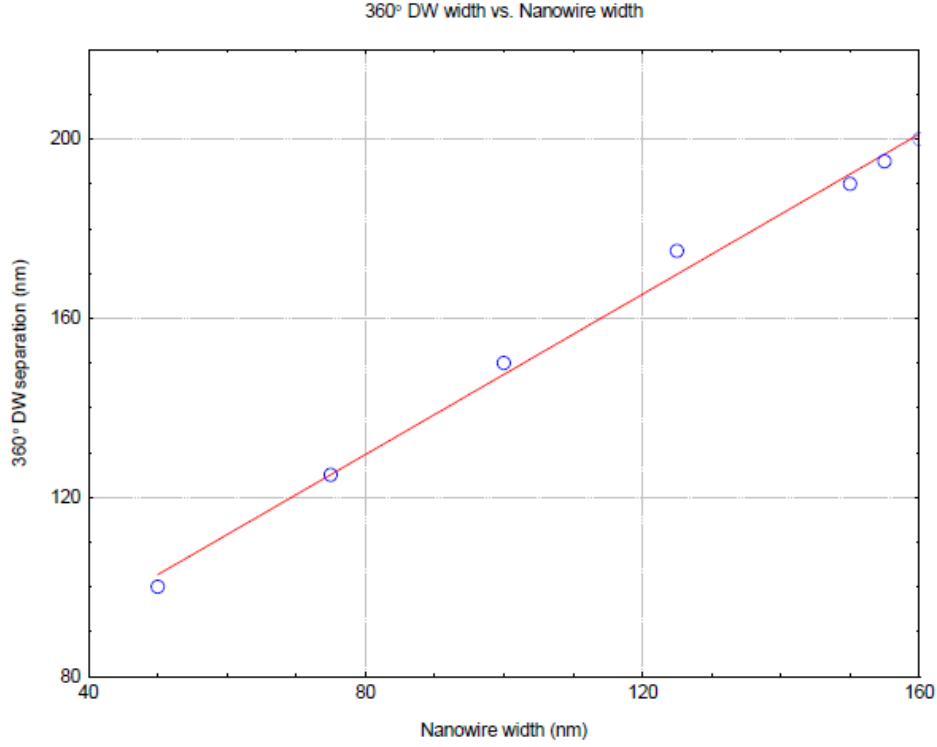


Figure 4.15: Plot of equilibrium 360° DW width against nanowire width.

achieved using the magnetically charged tip of an MFM.

The methodology described here serves to enhance the degree and sophistication of control available to researchers working on nanostructured magnetic systems, representing a significant addition to the existing toolbox of methods in its flexibility and ease of use. Particularly notable is that it does not build incrementally on existing electrical-stripline or global-magnetic-field based protocols as in the majority of recent advances, instead providing a substantially novel approach. The method allows for simple chirality control and spatial manipulation of injected 360° DWs, as well as the removal of the spatially fixed injection points and constraints on device architecture inherent in existing techniques. It is also possible to inject several 360° DWs at points along the same wire (confirmed experimentally through MFM imaging) or perform injection across arrays of parallel wires using a single scan. The added spatial flexibility opens up a host of previously inaccessible device designs, creating promising avenues for future work.

5 | Topological defect-driven magnetic writing: A new approach to magnetic SPL

The following chapter details the development and application of a novel scanning probe lithography (SPL) technique, allowing the reliable writing of complex magnetic states in nanopatterned systems. For some time, claims have been made about the potential benefits afforded by the ability to write any conceivable magnetic microstate into systems comprising dense arrays of nanostructured magnetic elements, such as ASI. These claims include timely and relevant applications, such as exploiting ASI and related systems as hardware neural networks^{33,95} or reconfigurable magnonic crystals^{3,11,96–98}. However, until recently there were no means by which to accomplish accurate magnetic nanopatterning, and hence other than a few proof-of-concepts or theoretical studies these claims have gone unsubstantiated.

In 2016 a methodology was presented⁴ combining a globally-uniform external magnetic field (applied via macroscopic electromagnet) with the local field of an MFM tip to achieve targeted switching of magnetic elements in a limited range of systems. While effective under specific conditions and a much-needed step towards a universal magnetic patterning solution, the method has significant drawbacks including an inability to operate in strongly-interacting systems. Here we present ‘topological defect-driven magnetic writing’ - a novel approach to magnetic writing requiring no system-wide external magnetic fields and offering significantly enhanced utility and

flexibility relative to existing methods.

5.1 Abstract

Non-interacting nanomagnetic arrays are widespread in data storage and processing. As current technologies approach fundamental limits on size and thermal stability, embracing the strong interactions present at high array densities becomes attractive. One design exploiting the enhanced magnetic interactions in dense arrays to enhance functionality is the geometrically-frustrated metamaterial ‘artificial spin ice’ (ASI). Frustrated systems offer vast untapped potential arising from their unique microstate landscapes, presenting intriguing opportunities from reconfigurable logic to magnonic devices or hardware neural networks. However, progress in such systems is impeded by the inability to access more than a fraction of the total microstate space. Here, we present a powerful scanning-probe writing technique, topological defect-driven magnetic writing, providing access to all possible microstates in ASI and related nanomagnetic arrays. We demonstrate the creation of two previously elusive states; the spin-crystal ground state of dipolar kagome ASI and high-energy, low-entropy ‘monopole-chain’ states exhibiting negative effective temperatures.

5.2 Introduction

Artificial spin ices (ASI) are magnetic metamaterials comprised of strongly-interacting nanomagnets, each acting as a single ‘macrospin’, in geometrically frustrated arrays. The macrospins obey magnetic equivalents of Pauling’s ice-rules⁴⁸, arranging themselves to minimise net magnetic charge q_m at each vertex. ASI⁹⁹ has provided vital physical insight on diverse topics from magnetic monopole-like excitations^{25,100} to fundamental thermodynamics^{14,16,22,31,101,102}. Key to its promise is the enormous number of unique magnetic charge configurations (or microstates) and the possibility to directly image the magnetic

structure. A kagome (honeycomb) ASI array containing N vertices possesses $\sim \frac{3}{\sqrt{2}}^N$ microstates satisfying the ice-rules, yielding vast microstate landscapes. The ice-rule obeying states all possess near-equivalent energies leading to massive degeneracy, inviting deep thermodynamic studies¹⁰² as well as device-focused applications exploiting the microstate landscape to store information^{95,103} or function as a hardware neural-network^{14–16} or reconfigurable magnonic crystal^{3,104}. However, the full potential of ASI is unrealised as conventional state-preparation techniques access only a tiny fraction of the total microstate space. Consider a six-macrospin hexagon of kagome ASI, possessing $2^6 = 64$ metastable magnetic configurations (local minima in the potential energy surface). Typically a global magnetic field initialises state A and then drives magnetic reversal to state B . As each bar switches only once, six additional states are sampled and so 57 states are never accessed. Energy barriers between states vary largely so few low-energy pathways from A to B exist. Most states do not lie on any of these pathways, rendering them inaccessible via global magnetic field protocols. In a typical 10^3 macrospin array just $10^{-296}\%$ of possible states are sampled in a global-field driven magnetic reversal. Thermal activation and subsequent annealing of ASI can randomly sample low-energy microstates inaccessible using global fields^{18,102,105–107}. However, the exact final state cannot be specified and to-date it has not been possible to explore the network of microstates sufficiently to prepare a system in the dipolar kagome ASI ground state¹⁸. The limited state-writing functionality has impeded progress in ASI, therefore developing a protocol for the precise control of individual macrospins is crucial to revealing the entire ASI manifold and the rich physics within.

Scanning-probe lithography (SPL) offers an ever-broadening suite of techniques for exquisite nanoscale patterning¹⁰⁸, including topographical modification^{109,110}, molecular synthesis¹¹¹ and recently nanomagnetic writing. Patterned modification of magnetic anisotropy in thin-films via a heated tip and global H field⁹⁰, magnetic-force microscope (MFM) tip-mediated domain wall (DW) injection¹¹² and a combined MFM-tip and global-field protocol for selective remagnetisation of nanowires⁴ represent key advances in the field, forging paths for new work in fundamental and device physics. Here, we present ‘topological defect-driven magnetic writing’ (TMW) - a novel, powerful magnetic SPL technique for directly injecting topological defects⁴¹ in magnetic nanowires. These defects propagate through structures as DWs, achieving controlled

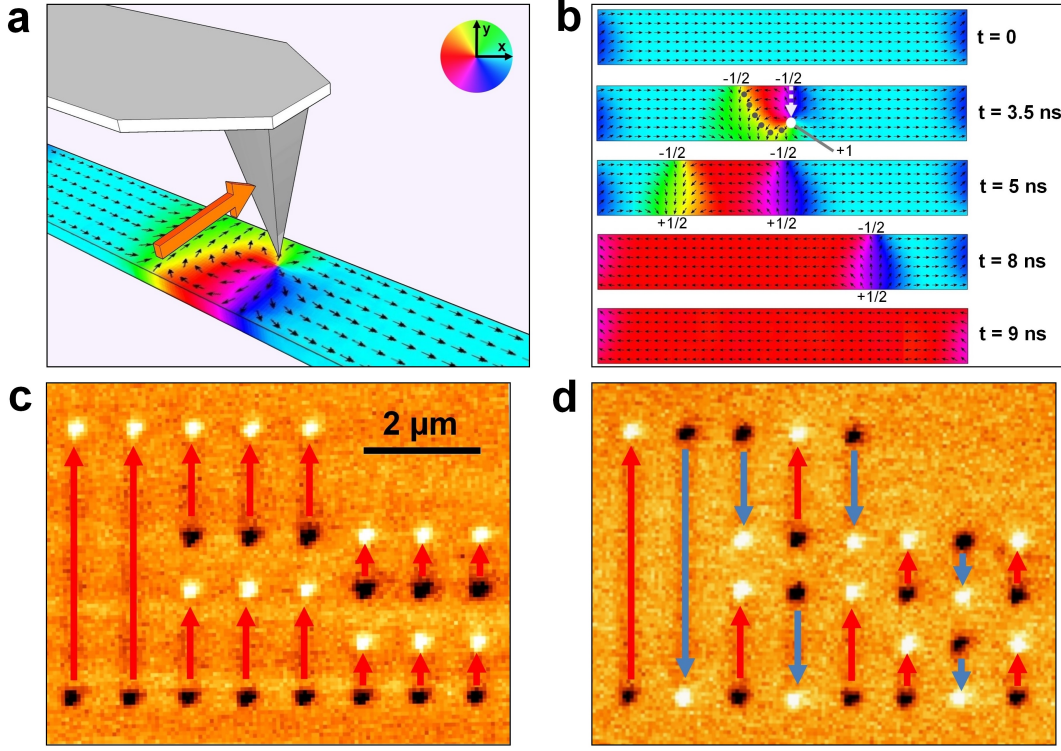


Figure 5.1: The TMW writing process. **a)** Schematic of TMW operation. An MFM-tip induced topological vortex defect is moved through a nanostructure spin texture by the motion of the tip. Colour wheel represents magnetisation direction in panels a) and b), orange arrow denotes v_{tip} . **b)** Micromagnetic simulation of the time-evolution of the TMW write-function used to reverse the magnetisation of a $500 \times 75 \times 10$ nm nanowire. The MFM tip and its direction of motion are represented by the white circle and dashed arrow respectively. Topological defects are labelled with their winding numbers. Web-links to videos of the time evolution of this process is included in the ‘micromagnetic time-evolution videos’ section at the end of this thesis. **c, d)** MFM images of 10×100 nm² cross-section NiFe nanowires of 1, 2 and 4.8 μ m length, (c) after initialisation with global field and (d) after TMW writing of selected wires. Light and dark contrast indicate positive and negative magnetic charge respectively. Overlaid arrows show magnetisation direction as a guide to the eye, red arrows indicate unwritten wires, blue arrows show TMW-written wires.

and reversible writing of magnetisation states in the process with no global fields required.

Working in ambient conditions, TMW reveals the entire microstate landscape of ASI and related nanopatterned magnetic systems. We demonstrate several previously unobserved states including ‘monopole chains’, high-energy low-entropy ASI configurations inaccessible by conventional means and exhibiting negative effective temperatures²³, and the spin-crystal ground state of kagome ASI^{18,105,113}.

5.3 Working principle of TMW

TMW is depicted schematically in fig. 5.1 a) with a corresponding time evolution shown in fig. 5.1 b) (micromagnetic simulations performed using OOMMF⁶²). We demonstrate TMW experimentally first by writing states in NiFe nanowires. Figure 5.1 c) shows MFM images of nanowires after initialisation in global field H_{sat} , 1 d) shows MFM after selective TMW reversal. TMW begins in the exact same manner as the 360° DW injection technique described in the previous chapter, consisting of passing a commercially available high-moment MFM tip over a nanostructure at a tip-sample separation of $r \approx 5 - 10$ nm with the tip moving perpendicular to the long-axis of the structure. At these small tip-sample separations, the tip field H_{tip} may be treated using a point-probe approximation^{114,115} as having a monopole-like form $H_{tip} = \frac{\mu_0}{4\pi r^2} q_{tip}$ where q_{tip} is the magnetic charge of the tip. The nanowire begins the process in a collinear ferromagnetic state ($t = 0$ in fig. 5.1 b). The tip then approaches the structure and at sufficient values of H_{tip} the spins in the wire rearrange themselves to lie along the radial tip field, lowering their Zeeman energy. Around the apex of the tip ($t = 3.5$ ns, represented by the white circle) these realigned spins form a vortex configuration due to the locally divergent shape of H_{tip} . In an otherwise smooth spin system, a vortex is a topological defect with an associated winding number¹ of $+1$. The net winding number of a ferromagnetic spin system is rigorously conserved and as such, the formation of the vortex is balanced by creating a pair of half-vortex topological defects with winding number $-1/2$. Defects with fractional winding number are forbidden in the bulk so the half-vortices remain bound to the edge of the structure¹. The $+1$ vortex remains under the tip as it moves through the nanostructure, maintaining a connection to the edge-bound half-vortices via two continuous chains of reversed spins to avoid a magnetisation discontinuity. The chains are highlighted in the $t = 3.5$ ns panel by the dotted grey line (left of tip) and the dashed white line (in line with tip motion). The tip effect is asymmetric: On one side (right of tip in fig. 5.1 b) the component of H_{tip} along the wire-length is parallel to the wire magnetisation, so any disturbance to the micromagnetic structure is minimal. On the other side H_{tip} is anti-parallel to the magnetisation and drives local magnetic reversal. This forces the left-hand chain into an elongated curve around a nascent left-magnetised domain created

between the $-1/2$ defects ($t = 3.5$ ns) in the wake of the tip. As the tip finishes crossing the structure, the vortex is brought into contact with the structure's lower edge. Whole integer defects may only exist in the bulk, so the vortex decomposes into a pair of edge-bound $+1/2$ defects to conserve the net winding number¹. The chains of reversed spins now each connect a $+1/2$ topological defect on the bottom edge of the structure to a corresponding $-1/2$ defect on the top edge ($t = 5$ ns), each chain-defect pair forming an independent 180° DW¹. The distorted shape of the left-hand chain of spins has an associated micromagnetic exchange and demagnetisation energy penalty relative to the straight, shorter right-hand chain. In the absence of the tip the left-hand chain straightens out, adopting a lower energy conformation and gaining momentum in the process. The DW accelerates towards the left-end of the wire, aided by a magnetostatic attraction between the DW magnetic charge and the magnetic charge associated with the wire-end. On reaching the left-end of the structure, the DW is free to unwind into a collinear spin state ($t = 8$ ns), lowering the total system energy. The remaining right-hand DW also experiences a magnetostatic attraction to the structure ends. Providing the tip crosses the structure slightly to the right of centre, the net force on the remaining DW is directed to the right. This pulls the DW into the right-hand end of the structure where it unwinds on contact, leaving the wire DW-free, magnetised anti-parallel to its initial state. The TMW writing process is now complete. Whether the right or left-hand DW forms in a high-energy state is decided by a combination of the nanostructure's initial magnetisation and the magnetisation of the MFM tip, allowing the user to tailor the writing dynamics simply by remagnetising the tip. The combination depicted in fig. 5.1 b) arises from a right-magnetised nanostructure and MFM tip magnetised into the page. Reversing either magnetisation will result instead in the right-hand DW forming in a high-energy state and accelerating once the tip passes. Reversing both magnetisations returns to a high-energy left-hand DW. Web-links to videos of the time evolution of this process is included in the 'micromagnetic time-evolution videos' section at the end of this thesis. Crucially, the TMW process relies solely on the local field of the tip with no additional global H field required, enhancing utility and flexibility relative to existing techniques⁴.

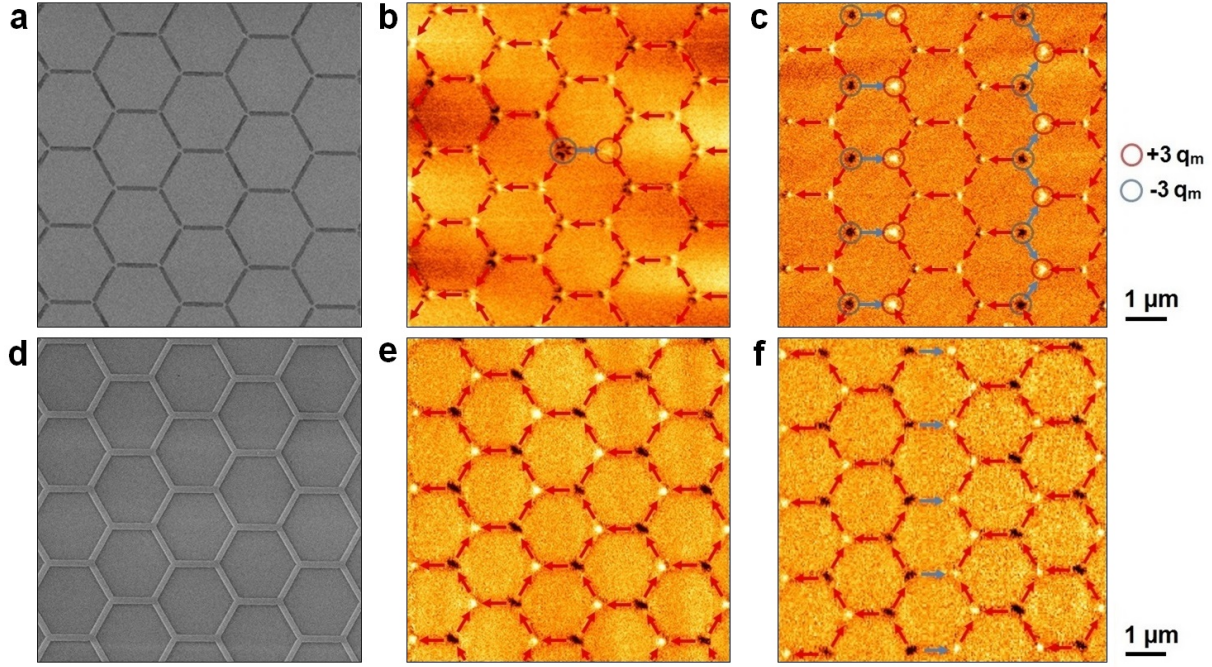


Figure 5.2: Realisation of TMW read/write functionality in ASI. SEM (a,d) and MFM (b,c,e,f) images demonstrating TMW written states in various ASI arrays. NiFe nanowires are $1\ \mu\text{m}$ long, $10\ \text{nm}$ thick. a,b,c) wires are $75\ \text{nm}$ wide, disconnected at vertices with $30\ \text{nm}$ gaps. d,e,f) are $100\ \text{nm}$ wide, connected at vertices. MFM images are overlaid with arrows indicating magnetisation direction of each macrospin. Prior to imaging and writing, structures were saturated by an external field H_{sat} . Red arrows denote unwritten nanowires still magnetised along H_{sat} , blue arrows show selectively remagnetised wires. **b)** MFM image showing a single tip-reversed nanowire, creating a pair of $\pm 3\ q_m$ monopole-defects. **c)** MFM image of TMW-written ‘ladder’ and zigzag ‘monopole-chain’ states. System was re-magnetised along H_{sat} between images (b) and (c). **e)** MFM ‘before’ image showing all wires aligned to H_{sat} . **f)** MFM ‘after’ image of (e) showing a column of TMW-reversed nanowires.

5.4 TMW state writing in ASI

To examine how the TMW state-writing functionality performs in an ASI system, $1\text{ }\mu\text{m} \times 75 \times 10\text{ nm}^2$ nanowires were arranged into kagome ASI arrays with wires disconnected at vertices. Vertex separations of 30 nm were used, defined from the wire end to the vertex centre. Arrays were confirmed to be in a strongly-interacting ASI regime via MFM imaging of their as-grown states, with only ice-rule obeying vertices observed. Structures were initially saturated along a globally-applied field H_{sat} . TMW was then performed in zero-field to selectively reverse the magnetisation of specific macrospins until the desired microstate was reached. Figure 5.2 shows scanning-electron microscope (SEM) images of the ASI arrays (left-hand column) along with MFM images of initialised and TMW-written states. Figure 5.2 b) shows a single TMW-reversed macrospin against a uniform H_{sat} aligned background, creating a pair of oppositely charged $\pm 3\text{ }q_m$ monopole-defect vertices without disturbing surrounding macrospins. This demonstrates both the spatial accuracy of TMW, allowing control of each individual macrospin, and its ability to overcome the significant energy-barriers required to create ice-rule breaking states in strongly-interacting systems, thus granting access to the full ASI manifold. Figure 5.2 c) shows a more complex TMW-written state containing long strings of adjacent ice-rule violating vertices, previously unobserved ‘monopole-chains’. Demonstrated here are both ‘ladder’ (left) and zigzag (right) monopole-chains, high-energy states (due to the large number of excited $\pm 3\text{ }q_m$ vertices) with low entropy (due to the relatively few ways to arrange such configurations on an ASI array). This combination of high energy with low entropy results in a state exceedingly difficult to realise via conventional means and also fulfills the criteria for exhibiting ‘negative temperature’ - predicted to exist in ASI²³ and discussed further below. Such states highlight the novel and unexplored regions of the ASI manifold now accessible using TMW. Even in these high-energy states the system retains its written configuration with high stability, undisturbed by the MFM imaging process and maintaining microstate fidelity over several months.

So far, only disconnected nanowires have been considered. To test the flexibility of TMW in

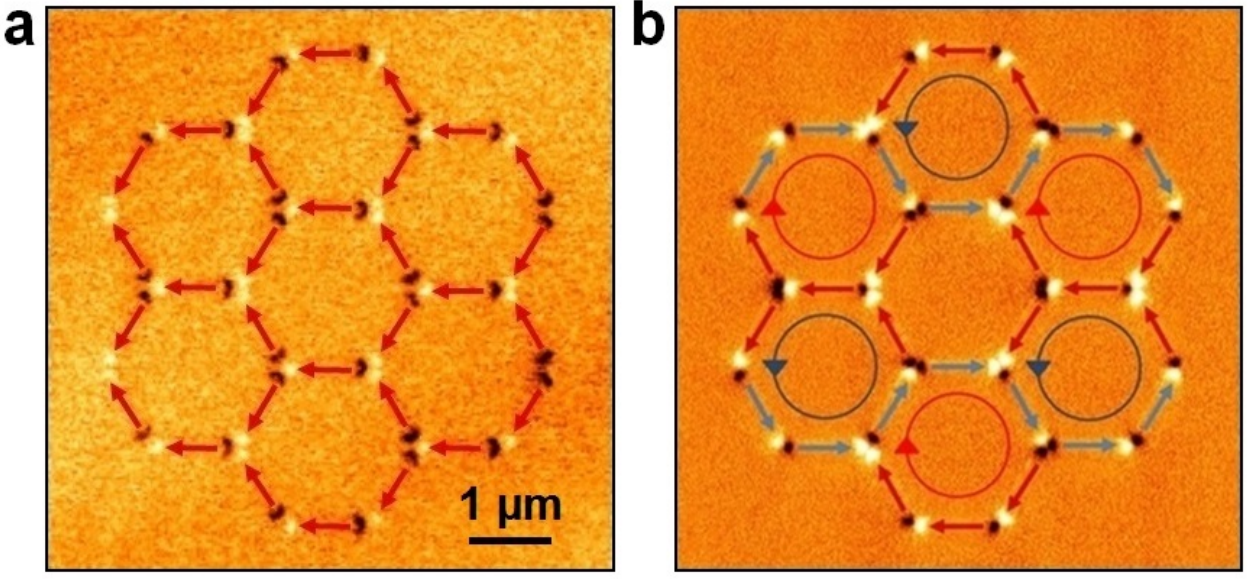


Figure 5.3: TMW-realised access to the kagome ASI ground state a) MFM image of $1\ \mu\text{m} \times 75\ \text{nm} \times 10\ \text{nm}$ NiFe nanowires in an ASI 'rosette' with 30 nm vertex gaps. All wires are aligned to H_{Sat} . b) MFM image of the chiral spin-crystal ground state of kagome ASI, accessed via topological defect-driven magnetic writing. Reversed wires are highlighted by blue arrows. Circular arrows inside hexagons highlight the alternating-chirality magnetisation loops characterising the ground state.

varied architectures, ASI arrays were fabricated with nanowires connected at vertices. Figure 5.2 d) is an SEM image of $1\ \mu\text{m} \times 150\ \text{nm} \times 10\ \text{nm}^2$ nanowires in a connected kagome ASI array. Figure 5.2 e) shows MFM imaging of the same array with macrospins saturated along H_{sat} . MTL was then used to reverse a central column of macrospins, with fig. 5.2 f) showing the successfully-written resultant state. The TMW writing process is seen to retain its functionality in systems comprising connected nanowires, opening up microstate writing in a huge range of potential systems.

5.5 TMW realisation of the kagome ASI ground state

One ASI microstate which has received significant attention while eluding observation is the spin-crystal ground state of dipolar kagome ASI. The ground state of the frustrated kagome lattice has been of interest in both natural and artificial systems, with the ground state of natural systems consisting of point-spins only recently reported after years of investigation¹¹⁶. Point-spin systems interact via nearest-neighbour exchange interactions, hence the difficulty in

identifying a long-range ordered ground state. However, when considering dipoles of finite length (i.e. artificial macrospins), further neighbour interactions (multipolar terms) break the degeneracy of ice-rule states as long-range magnetic charge and spin order are imposed^{24,117,118}. This leads to a well-defined ground state in ASI, existing when both charge and spin order are present throughout the entire system. A quasi-ground state in a partial ‘building block’ kagome ASI system comprising just 1-3 hexagons has been demonstrated³⁴, but the smallest system supporting a true ground state unit-cell which can tile infinitely over the kagome lattice is the 30 macrospin ‘rosette’ shown in figure 5.3. Attempts have been made to thermally activate then anneal into the ground state on a rosette-sized system¹⁸, but the near-degenerate energies of all ice-rule obeying microstates and the low entropy of the ground state (2 configurations in 2^{30}) have rendered efforts ineffectual. A simple and convenient means to reliably access the spin-crystal ground state will allow closer examination of the divergence between true and artificial ice-like systems, a major point of interest in the field. As such, demonstrating the first experimental realisation of the spin-crystal kagome ground state is a perfect opportunity for testing the capabilities of TMW.

Figure 5.3 a) shows an MFM image of a kagome ASI rosette comprising $1\text{ }\mu\text{m} \times 75 \times 10\text{ nm}^2$ nanowires with all macrospins aligned along H_{sat} . TMW was then performed in zero-field to selectively remagnetise 14 of the 30 macrospins, with the resultant state shown in fig. 5.3 b). The macrospins in each of the outer hexagons now lie in closed continuous spin-loops, with chirality alternating between adjacent hexagons (highlighted by circular arrows within hexagons). This configuration not only satisfies the ice-rules but also exhibits magnetic charge and spin ordering, with the observed charge and spin configurations exactly matching those previously reported in theoretical schematics of the spin-crystal ground state^{18,24,113,117,119}. As such, we conclude that the TMW-written state presented here represents an experimentally-realised kagome ground state. That states eluding observation up to this point, despite direct efforts to prepare them, may now be written to order using readily-available equipment is an important threshold development for the fields of ASI and SPL.

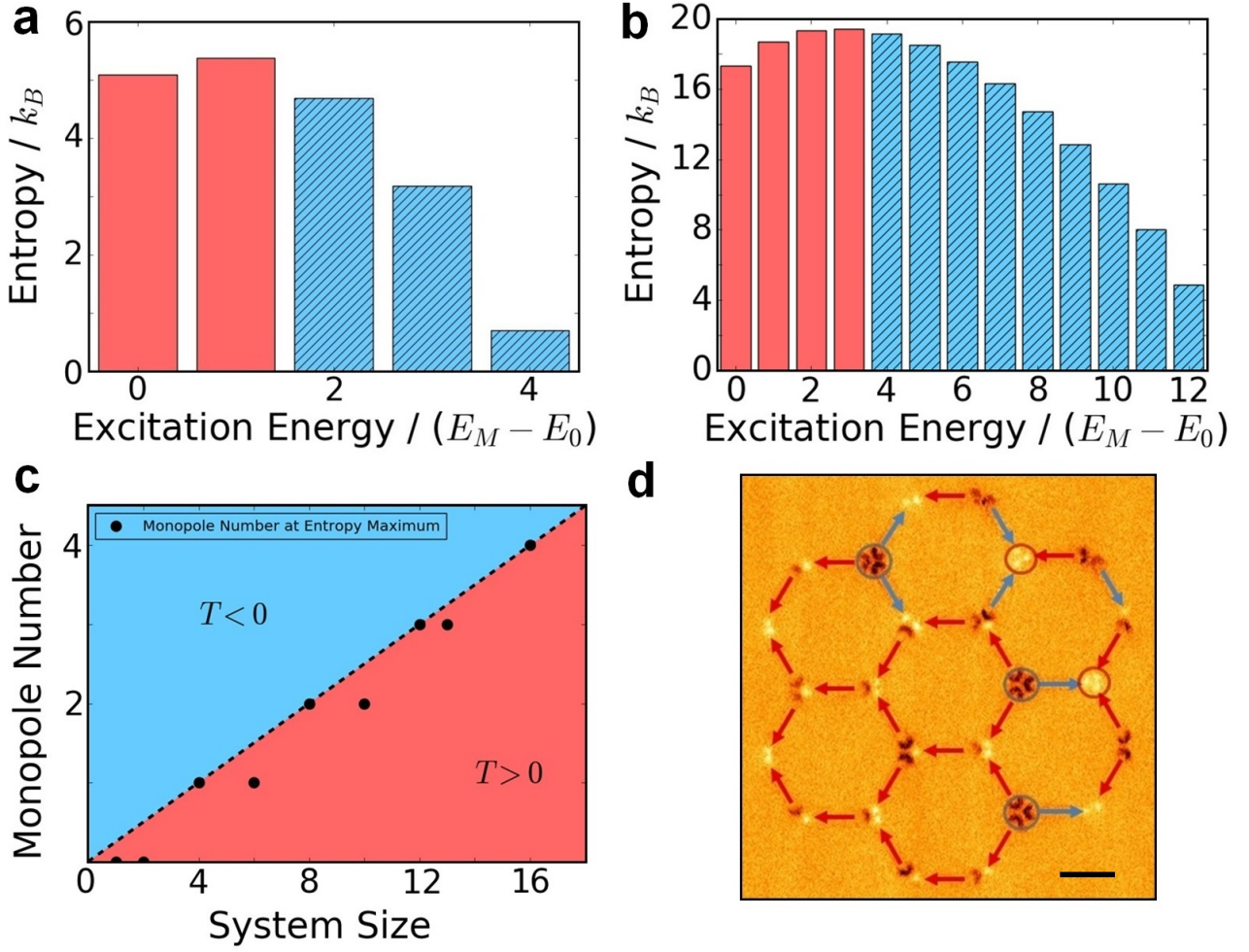


Figure 5.4: Negative effective temperature in ASI

Entropy vs. excitation energy histograms for **a)** a four-vertex and **b)** a twelve-vertex kagome ASI system. Excitation energy normalised by $E_M - E_0$ corresponds to number of monopole vertices present. Red bars denote positive temperature states, shaded blue bars indicate negative effective temperatures.

c) Effective temperature phase diagram for one-to-eighteen vertex kagome ASI systems. Negative effective temperatures arise when more than one-quarter of total vertices are in monopole-defect configuration.

d) MFM image of a twelve-vertex (counting three-macrospin vertices only) kagome ASI rosette. Five monopole-defect vertices are observed, indicating a negative effective temperature state. Scale bar indicates 1 μm .

Enumeration of effective temperatures performed by D. M. Arroo, sample fabrication and TMW writing of negative effective temperature states performed by J. C. Gartside.

5.6 Negative effective temperature states in ASI

As well as providing reliable access to low-energy states, TMW also significantly expands the accessible ASI microstate space by allowing preparation of high-energy, low-entropy microstates such as the monopole-chains discussed above. In general, the thermal anneals used to prepare ASI states work by keeping ASI systems in thermodynamic equilibrium with an (effective, in the

case of field protocols) heat bath. As the temperature of the heat bath is reduced, the ASI settles into a low-energy state with relatively few ice-rule defects. Rotating-field protocols may be thought of in a similar way by assigning them an effective temperature based on the field step-size and treating them as a heat bath²³. In contrast to these methods, TMW can reliably prepare ASI in states that are inherently out-of-equilibrium. For example, it is vanishingly improbable that a microstate with 19 monopoles on a system of 40 honeycomb vertices as shown in figure 5.2 c) would occur through any sort of equilibration process with a heat bath of finite temperature. To formalise this idea, it is useful to invoke the concept of an effective spin temperature for ASI as introduced by Nisoli *et al*²³. In statistical mechanics, the temperature of a system is formally defined as a measure of how the energy of that system varies with disorder $T = \frac{\partial U}{\partial S}$. Taking into account only nearest-neighbour interactions (which dominate ASI energetics), the energy of an ASI system is determined by the number of monopoles (ice-rule violations) in the system, with each monopole raising the total energy by the difference in energy between a monopole and ice-rule vertex, $\Delta E = E_M - E_0$. The total energy of the system is thus given by $E = E_{ice} + n\Delta E$, where E_{ice} is the system energy when all vertices obey ice-rules and n is the number of monopoles. Since the fully ice-rule compliant state is highly degenerate but the fully excited state (where every vertex hosts a monopole) has just two possible configurations (spin-degeneracy), there must exist a range of microstates with negative effective temperatures^{120,121}. Such states are precisely those difficult to access via thermal and rotating field protocols, but trivial to realise via TMW.

By directly enumerating the number of monopoles (and therefore system energy) for all microstates in small kagome ASI systems and plotting monopole number versus entropy (figure 5.4 a-c), the sign of the effective temperature may be clearly visualised from the gradient $\frac{\partial U}{\partial S}$. Figures 5.4 a) and b) show this relation between entropy and energy/monopole-number in four and twelve-vertex systems respectively, with a clear change in gradient and associated transition to negative temperature occurring in both when over a quarter of vertices host monopole defects. The effective temperatures of all microstates in 1-18 vertex systems were calculated, with results shown as a phase diagram in fig. 5.4 c). For all system-sizes negative temperatures were seen to correspond to microstates in which over a quarter of total vertices were monopole-defects. The

five-monopole microstate on a 12-vertex honeycomb rosette shown in figure 5.4 d) thus corresponds to a negative temperature state directly written by TMW, as does the monopole-chain state shown in fig. 5.2 c). By comparison, the rosette system in fig. 5.4 d) would have to be heated to almost 10^5 K and cooled near-instantaneously to have a 0.1% chance of leaving the system in a five-monopole state. ASI enables direct room-temperature access to the complex and exotic physics of frustrated magnetism, with system energetics and time-scales easily tunable via fabrication. Adding to prior observations of otherwise-elusive ice-rule behaviour¹⁰¹ and monopole-defects²⁵, TMW now introduces ASI as a model-system in which to study the dynamics and evolution of negative temperature states.

5.7 Micromagnetic dynamics of the TMW writing process

To gain further insight into the dynamics of the writing process, micromagnetic simulations were performed using OOMMF. To isolate the intrinsic MFM tip specific dynamics from the effects of the magnetic charges at the nanowire ends, a semi-infinite wire of 10 μm length was simulated with plates of fixed magnetisation employed at either end to effectively remove the magnetic charges¹²². A magnetic charge q_{tip} and associated monopolar H field representing the MFM tip were passed over a $75 \times 10\text{nm}^2$ cross-sectional wire at a tip-wire separation of 5 nm, locally distorting the spin system and introducing dynamic topological defects as described above. The time dependent separation between the two 180° DWs formed by these defects was measured for a range of q_{tip} values, with the results shown in figure 5.5 a). The tip first reaches the wire at $t = 2.5$ ns, soon after which the injected DWs are seen to rapidly separate. In the absence of magnetic wire-end charges the dipolar attraction between the two DWs eventually brings them back together, forming a stable composite 360° DW¹¹². The peak inter-DW separation reached was found to monotonically increase with q_{tip} . To investigate the mechanism by which this increasing separation occurs, a method is required for quantifying the amount of potential energy being stored in the contorted high-energy DW during the injection process. The key micromagnetic terms governing such dynamics are the exchange and demagnetisation energies. In order to separate the energy cost of the stable, non-distorted DWs from the additional

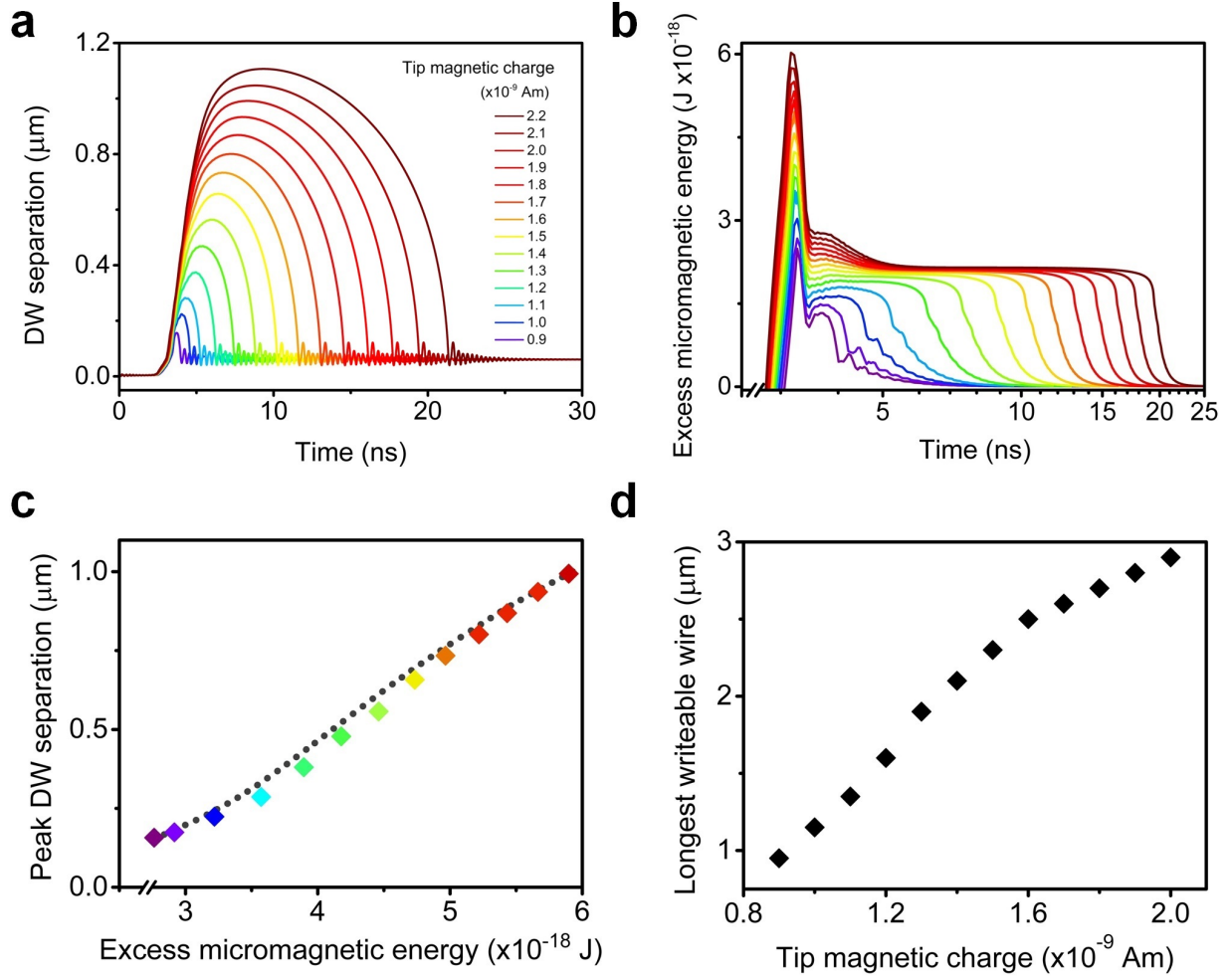


Figure 5.5: Dynamics of the TMW writing process.

a) Time-dependent separation between domain walls injected in an infinite 75 nm wide nanowire by varying strength MFM tips. Tip reaches the wire at $t = 2.5 \text{ ns}$.

b) Excess micromagnetic energy versus time with colours corresponding to the same range of tip strengths as **a)**. **c)** Peak inter-DW separation for different strength MFM tips as a function of peak excess micromagnetic energy. Coloured diamonds are data taken from the OOMMF simulations shown in panel **a)** with colours corresponding to the same tip strengths. Grey dotted line shows data given by a simple semi-analytical kinematic model.

d) Longest simulated TMW-writeable nanowire versus MFM tip magnetic charge strength q_{tip} .

potential energy driving the DW separation, the combined exchange and demagnetisation energies were measured from OOMMF simulations relative to the energy of a stable 360° DW where the spin-chains of both composite 180° DWs lie in a straight, low-energy conformation perpendicular to the long-axis of the wire. Any increase in micromagnetic energy relative to this can be linked to the distortions induced in the spin-chains by H_{tip} , with a value of zero reached at the end of the injection process corresponding to the formation of a stable 360° DW. The time-evolution of the resultant composite energy, termed the ‘excess micromagnetic energy’, is plotted in fig. 5.5 b) for the same time period and q_{tip} values as fig. 5.5 a) with a matching colour scheme indicating q_{tip} for each curve. Time is presented on a logarithmic scale as the dynamics of interest occur in the initial stage of the injection process when the tip is close to the wire. Figure 5.5 b) shows an initial sharp increase in the excess micromagnetic energy as the vortex defect is introduced to the wire and the spin-chains are formed, peaking abruptly at $t = 3.2$ ns where the MFM tip finishes crossing the nanowire. At this point where the vortex defect decomposes and the DWs are no longer bound, the excess micromagnetic energy rapidly drops, coinciding with a large increase in the DW separation seen in fig. 5.5 a). This correlation serves as a good indication that the MFM tip is indeed inducing a large amount of micromagnetic potential energy in the nascent contorted DW, which is then converted to DW kinetic energy in the absence of the tip as the DW violently straightens out, generating momentum in the process. To better understand the relationship between the excess micromagnetic energy and the momentum imparted to the dynamic DW, figure 5.5 c) plots the peak DW separation reached in fig. 5.5 a) against the peak micromagnetic energy from fig. 5.5 b). The diamond markers represent data extracted from the OOMMF simulations shown in fig. 5.5 a) with colour codes corresponding to the same q_{tip} values as in the previous two panels. A smooth monotonic increase in DW separation is observed with increasing excess micromagnetic energy, further suggesting that the release of pent-up micromagnetic potential is driving the DW separation. However, as micromagnetic dynamics are complex with multiple competing interactions present, it is not immediately clear that the excess micromagnetic energy is the dominant component driving the injection dynamics. In order to test whether the qualitative behaviour observed in the OOMMF simulations can be reproduced by considering just the excess

micromagnetic energy, a simple semi-analytical model was employed, treating each 180° DW as a quasiparticle with effective mass $m = \frac{h^2 N}{4\pi^2 K \lambda^2}$ where N, K and λ represent the number of spins in the DW, the transverse magnetic anisotropy energy of the wire and the DW width respectively^{123–125}, and magnetic charge $2q_m$ where $q_m = \pm M_s w t$ with M_s, w and t being the saturation magnetisation, wire width and thickness respectively¹²⁶. DW width was measured from OOMMF simulations as the distance throughout which the wire magnetisation direction varied from the long-axis of the wire by greater than 4° . Here, values of $m = 3.41 \times 10^{-23}$ kg and $2q_m = 1.29 \times 10^{-9}$ Am were determined. The two opposite magnetic charge polarity DWs are confined to a one-dimensional line representing the nanowire at an initial separation r_0 , measured from the OOMMF simulations at the moment the vortex topological defect decomposes into half-vortex defects and the DWs become unbound. One DW remains fixed at $r = 0$, representing the low-energy DW formed from the straight spin-chain, while the other representing the high-energy DW formed from the contorted spin-chain is initialised moving away from $r = 0$ with a velocity v_0 given by converting the peak excess micromagnetic energy (taken from OOMMF at the point the tip-localised vortex defect decomposes) to kinetic energy via $\frac{1}{2}mv_0^2$. The moving DW feels an attractive magnetic Coulomb force $F_{DW} = \frac{\mu_0}{4\pi r^2}(2q_m)^2$ directed towards the static DW at $r = 0$ and a viscous Gilbert-damping drag force¹²⁷ $F_{drag} = -vb$ where b is a phenomenological damping coefficient, here 1.53×10^{-14} Nsm⁻¹. The equation of motion for the moving DW is then solved for DW position r at each time step $\delta t = 1$ ps using numerical integration. The results of this model are plotted in figure 5.2 b) as the dotted grey line and can be seen to closely reproduce the behaviour observed in the OOMMF simulations. This provides further evidence supporting our hypothesis that micromagnetic potential energy stored in the shape-distortion of the forming DW by H_{tip} is responsible for driving the momentum of the dynamic DW and therefore the writing process, rather like drawing back a pinball-machine plunger which is released once the tip completes its crossing of the wire. TMW injection on ‘quasi-infinite’ nanowire arrays was performed with subsequent MFM imaging supporting the dynamics observed in OOMMF (see fig. 5.6 in the following section), suggesting that the picture of one high-energy DW ‘fired’ down the wire and a second relatively static low-energy DW is indeed physically accurate. The results of this experiment are discussed below.

To now examine the writing dynamics of TMW in a finite-length wire with end charges present, simulations were performed on $10 \text{ nm} \times 75 \text{ nm}$ cross-sectional wires of $0.5\text{-}3 \text{ }\mu\text{m}$ length. As before, a magnetic charge and associated monopolar field representing the MFM tip were passed across the wire close to its central point. The topological defects and associated DWs were injected into the wire as described above, but for wires below a certain length threshold the dynamic DW now makes on contact with the wire end, unwinding to a smooth spin state before the remaining DW is pulled into the opposite end, also unwinding and leaving a collinear reverse-magnetised state. The longest reversible wire was determined for a range of q_{tip} values by simulating injection on increasingly longer wires until the DWs were no longer able to reach the wire ends and reversal did not occur. The relation between q_{tip} and the longest writeable wire is shown in fig. 5.5d), with a monotonic increase observed in maximum writeable wire length with increasing q_{tip} values. Wires several μm in length are switchable by q_{tip} values corresponding to typical high-moment MFM tips ($\sim 10^{-9} \text{ Am}$), matching well with experimentally observed behaviour. The trend between longest writeable wire and q_{tip} suggests that TMW may be tailored to switch wires of a desired length by selecting appropriate strength MFM tips, achieved practically by varying the thickness of the magnetic coating layer on the tip. Potentially allowing for longer wires to be written using thicker magnetic coatings ($> 60 \text{ nm}$) than are commercially available.

5.8 TMW in quasi-infinite nanowires

To ascertain whether the injection dynamics observed in micromagnetic simulation represent a realistic picture of the injection process, 'quasi-infinite' nanowires were experimentally fabricated. In such wires where the injection process can be considered as independent of influences from the wire-end charges, the TMW writing process essentially simplifies back to the 360° DW writing process described in the previous chapter. However, to fully understand the dynamics of the TMW writing process it is important to first get a handle on what is occurring in the absence of the end-charges before complicating the process by adding them in.

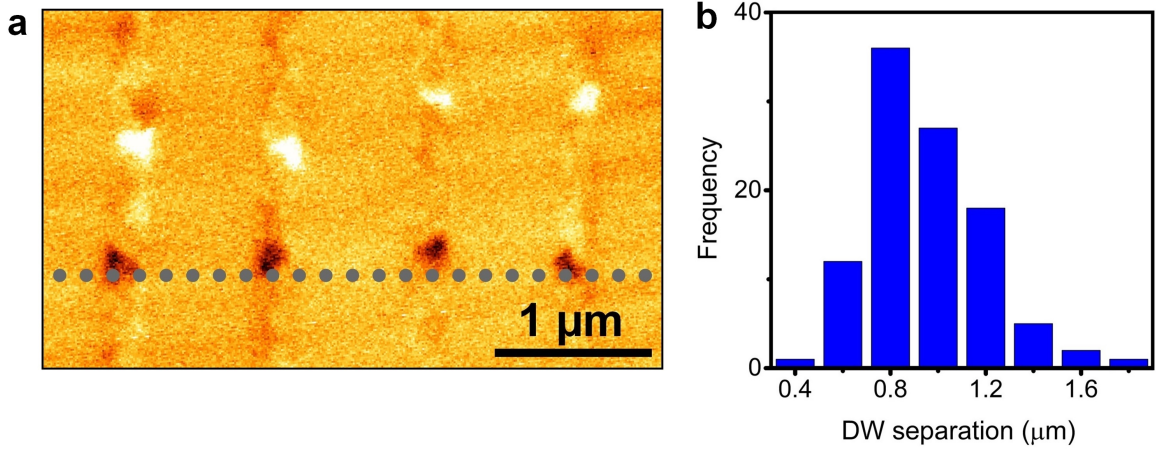


Figure 5.6: Results of TMW performed in ‘quasi-infinite’ nanowire arrays

a) MFM image of 100 nm × 10 nm cross-section Co nanowire array after TMW injection. Dotted grey line represents path of MFM tip during writing.

b) Histogram showing separation between injected DWs in nanowire arrays shown in a), measured over 101 TMW writing events.

Arrays of 20 μm long nanowires were fabricated, such that at their central points the effects of the wire-end magnetic charges were negligible. The wires were initially magnetised along their long-axis by a global H field. TMW injection was then performed across the mid-point of the wires and the resultant state imaged via MFM, with Co used rather than NiFe as the harder magnetic material allows for non-invasive MFM imaging of DWs without H_{tip} disturbing them. A high deposition rate and coarse liftoff process were employed in an attempt to generate a high frequency of DW pinning sites in order to ‘trap’ DWs at various stages of the dynamic injection process. Fig. 5.6 a) shows a nanowire array after TMW injection. A 180° DWs is observed in each wire under the path of the tip, with a second 180° DW displaced 0.8-1.2 μm along the wire. In each case the second DW is displaced to the same side of the DW lying under the tip path. The observed behaviour correlates well with our simulation results, suggesting that two DWs are created by the passing tip, one in a low-energy state which remains close to its initial location and one high-energy DW with a significant amount of momentum, travelling a distance away from its injected position. It is important to note that the DWs imaged here have become pinned at some point during the injection process (the lowest-energy state being a stable 360° DW with around 90 nm separation between DWs), each wire represents an effective snapshot taken at some point during the injection process and as such the DW separations can be taken as a minimum separation reached. However, previous work has identified that pinning is more likely

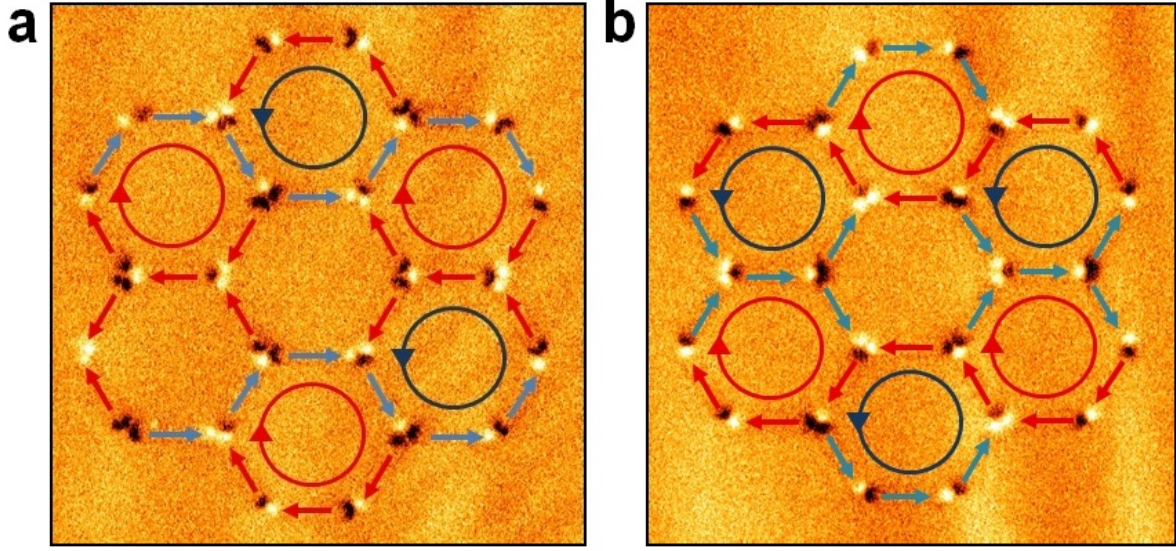


Figure 5.7: TMW-written states in disconnected ASI rosettes

a) MFM image of a single bar (bottom-left loop) reversed from an otherwise complete ground state, highlighting ‘erase’ functionality.

b) MFM image of alternate-chirality ground state. All 30 macrospins have been reversed from the original ground state.

to occur while DW velocity is low (i.e. towards the stationary point before DWs begin moving back together), so measured DW separations may well be relatively representative of the maximum distance reached¹²⁸. Figure 5.6 b) is a histogram of DW separations, measured over 101 separate TMW injection events in the same nanowire shown in fig. 5.6 a). A separation range of 0.44-1.66 μm is observed, corresponding well to the separations observed in OOMMF simulations and suggesting that dynamic topological defects induced by the tip are indeed responsible for the TMW writing process.

5.9 TMW ‘erase’ functionality

To demonstrate the ‘erase’ functionality of TMW along with the ease by which complex states may be transitioned between, every macrospin in the spin-crystal ground state shown in fig. 5.3 b) was reversed via TMW, preparing the alternate chirality ground state. Figure 5.7 a) shows a single TMW-reversed bar in an otherwise complete ground state configuration. The remaining 29 macrospins were then reversed using TMW. Figure 5.7 b) showing the resultant

alternate-chirality ground state, highlighting the accuracy and flexibility of the writing technique.

5.10 TMW on ASI with smaller feature size

So far we have considered only ASI comprising 1 μm long nanowires. This length was chosen for two reasons. First, to give a good spatial separation between the magnetic charges at the wire ends, allowing for well-resolved MFM imaging clearly illustrating the written ASI microstates. Second, we wanted to prevent doubts that the TMW writing process was essentially a conventional field-mediated magnetisation reversal using the local MFM tip-field in lieu of a global magnetic field. At wire lengths of 1 μm , the tip-field will have spatially decayed to well below the coercive field of the nanowire throughout the majority of the wire length. For shorter nanowires this is not necessarily the case, and as such it is harder to make the argument that TMW is a novel reversal process driven by twin 180° DWs.

Now that TMW has been successfully demonstrated in 1 μm nanowire systems, it is worth examining its performance in systems comprising smaller features. An ASI lattice formed of $400\text{ nm} \times 80\text{ nm} \times 10\text{ nm}$ nanowires was fabricated using an EBL liftoff process and thermal evaporation of Py. The lattice was initially prepared in a saturated magnetic state via a global field applied in the negative x-direction along the ‘armchair’ lattice vector. TMW was then performed in zero external field to reverse two central columns of nanowires, one of ‘ladder’ conformation and one of ‘zigzag’. Resultant TMW-written magnetic states are shown in figures 5.8, 5.9 and 5.10 with the initial saturating global field applied towards the left of the image frame. Figures 5.8 and 5.9 both show the same two TMW-written states, with a) the first state written after the initial field-saturation and b) a subsequently written state formed by reversing additional bars from the state shown in a). Figure 5.9 is just a magnified image of the central region of figure 5.8 with overlaid arrows denoting the magnetisation direction of written (blue) and unwritten (red) bars as a guide to the eye. Figure 5.10 is a further magnified view of the central region of panel b) of the previous two images, allowing a clearer view of the TMW-written state.

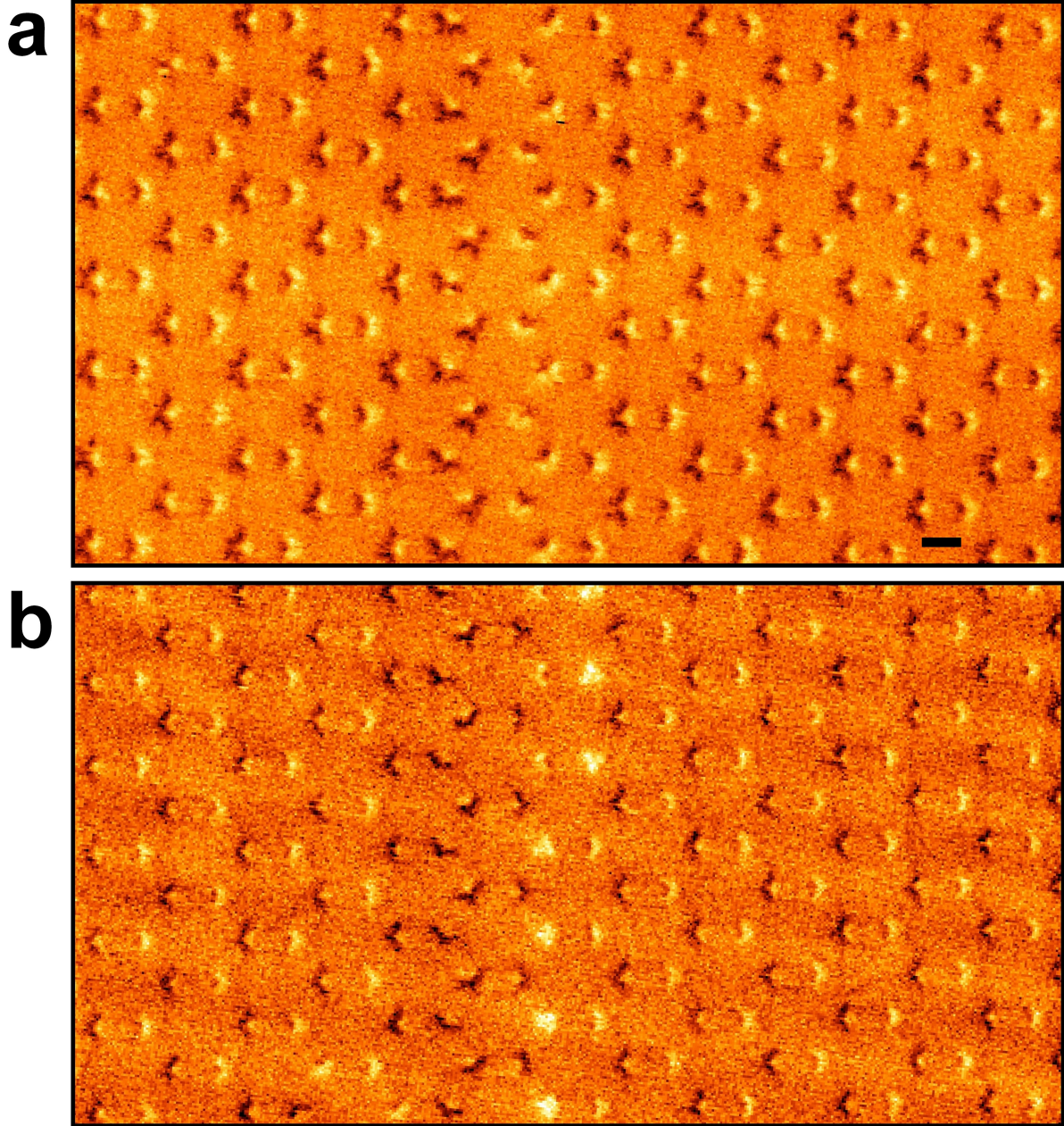


Figure 5.8: TMW-written states in ASI lattice comprising $400 \text{ nm} \times 80 \text{ nm} \times 10 \text{ nm}$ nanowires. Nanowire separation at vertices is 50 nm . Scale bar is 400 nm long.

a) An ASI lattice initially magnetised to the left of frame with two central columns of nanowires (one ‘zigzag’ and one ‘ladder’) magnetically reversed via TMW.

b) Starting with the magnetic state shown in a), further nanowires were TMW-written to create a central column of monopole-defect vertices.

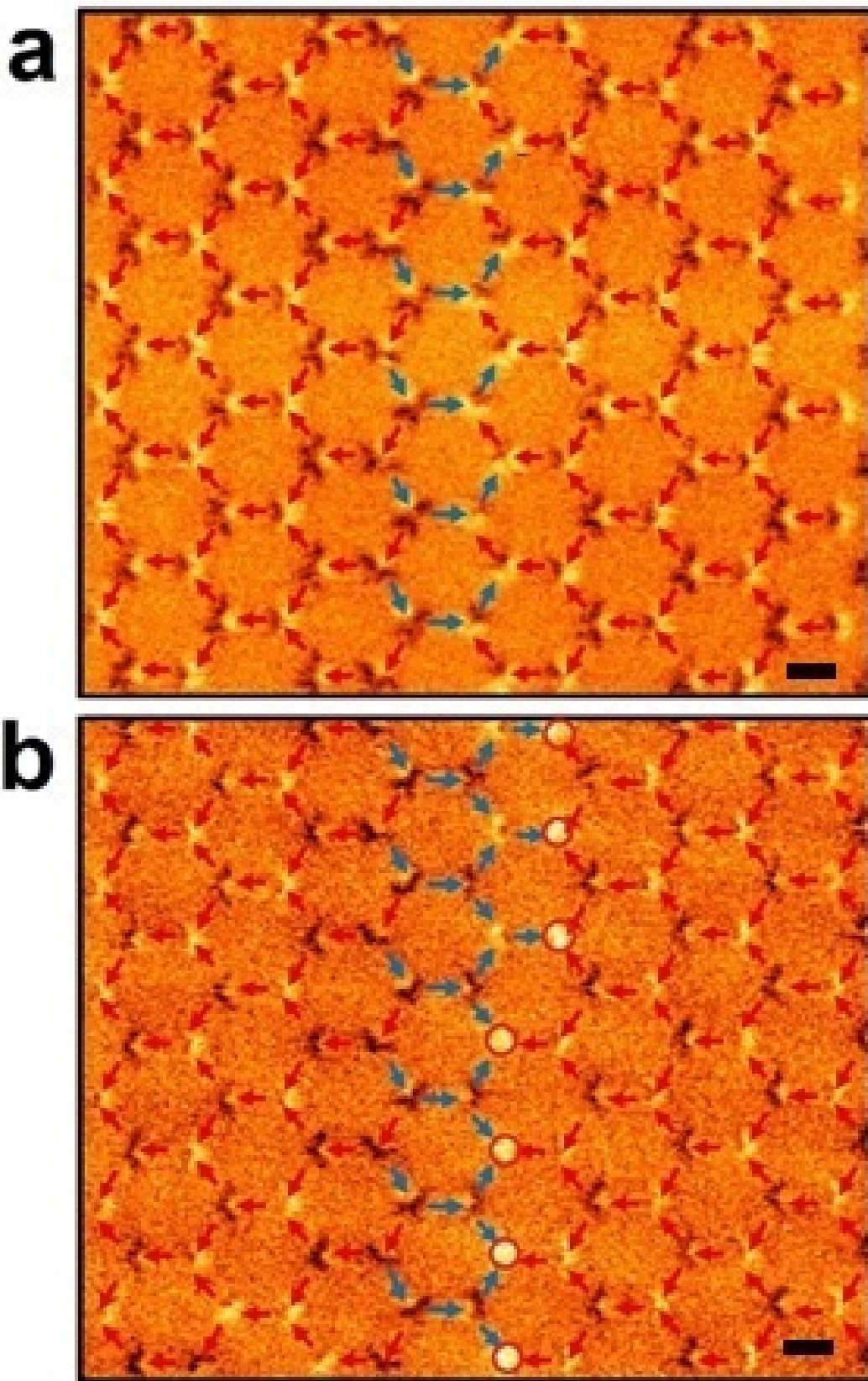


Figure 5.9: Central region of same TMW-written states shown in 5.8 with overlaid colour denoting unwritten (red) and written (blue) bars. Scale bar is 400 nm long.

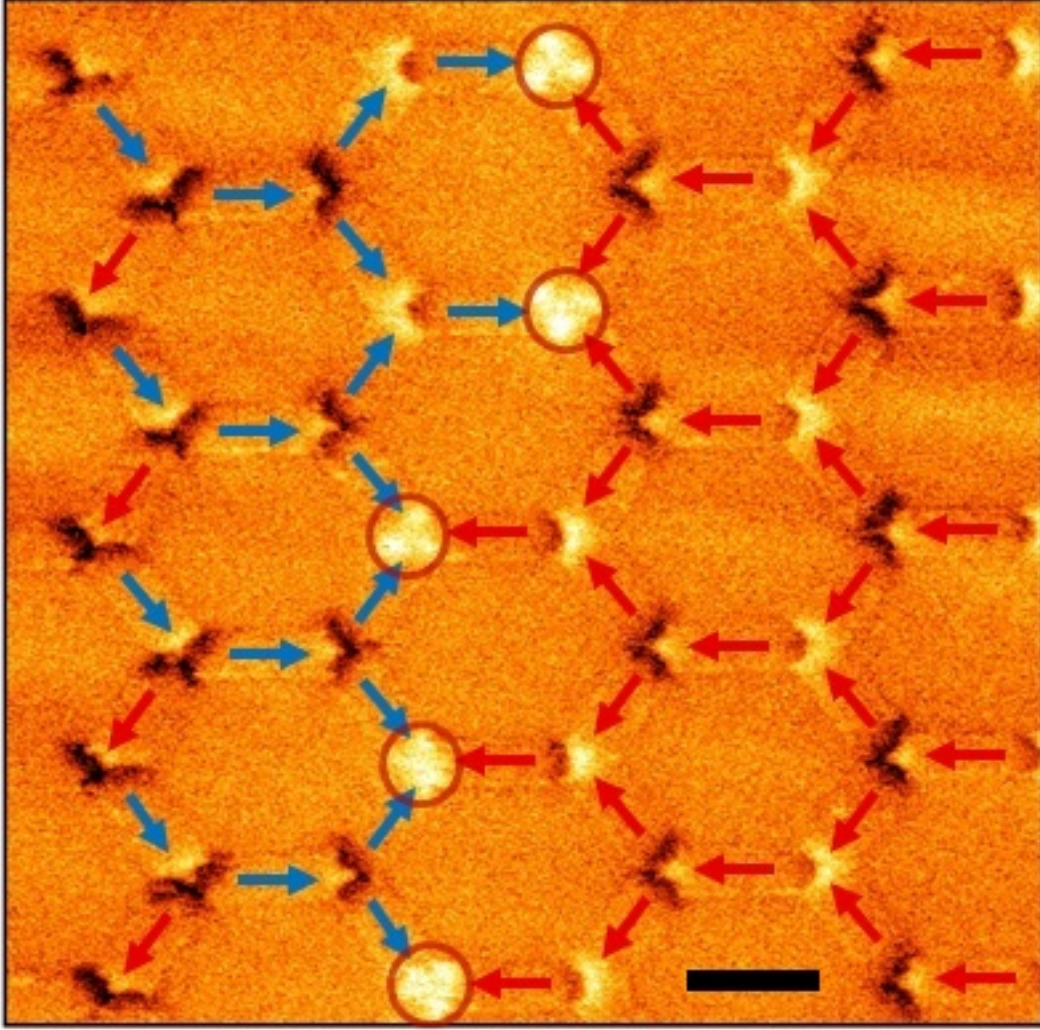


Figure 5.10: TMW-written state in ASI lattice comprising $400 \text{ nm} \times 80 \text{ nm} \times 10 \text{ nm}$ nanowires. Nanowire separation at vertices is 50 nm .

Zoomed-in view of the central region of figure 5.8 b). Magnetisation directions of unwritten (red) and written (blue) bars are denoted by overlaid arrows, monopole-defect vertices are highlighted by red circles. At the the right side of the frame the system is still in its field-saturated state. An TMW-written central column of $+3q_m$ vertices is observed, along with an TMW-written 'downwards'-tracing zigzag column to the far-left of frame.

Even smaller ASI was then fabricated, using $300\text{ nm} \times 80\text{ nm} \times 10\text{ nm}$ nanowires. TMW was used to write excited $\pm 3q_m$ vertex states throughout the lattice, with figure 5.11 showing the successfully written state.

TMW is seen to retain its functionality down to 300 nm long ASI, with writing of intricate states containing excited and non-excited vertices possible. The increased inter-bar interaction strength arising from the smaller 50 nm vertex separations does not present a barrier to writing excited states. This is a promising result highlighting the capability of TMW to function in high-density strongly-interacting arrays, a crucial capability for any competitive magnetic writing technique. Sub-300 nm arrays have not yet been experimentally examined, doing so is a clear candidate for future work.

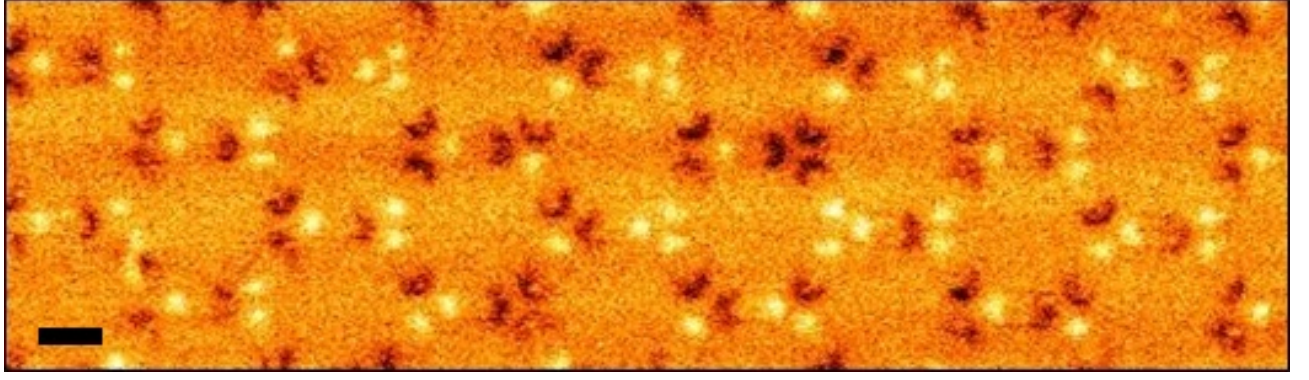


Figure 5.11: TMW-written $\pm 3q_m$ states in ASI comprising $300 \text{ nm} \times 80 \text{ nm} \times 10 \text{ nm}$ Py nanowires. Nanowire separation at vertices is 50 nm . Scale bar is 300 nm

While on the subject of shrinking nanowire sizes, it is worth considering the fundamental dimensional limits of TMW. While instrumental limits on lateral tip-positioning (for reference, the Asylum MFP-3D system used for the more intricate state writing specifies an x, y accuracy of $\pm 0.1 - 1 \text{ nm}$) and MFM imaging resolution (roughly equivalent to the tip-sample lift-height and hard to improve beyond $20\text{-}30 \text{ nm}$ using current equipment) are most likely to define the workable resolution of TMW, hardware capabilities constantly improve. Micromagnetic simulation, where tip positioning and ‘imaging’ resolution are perfect, allows us to examine the underlying physical limitations on TMW.

Figure 4.15 shows that the width of a 360° DW in a nanowire is $\sim 0.9\times$ the width of the wire. As such, TMW as described above cannot really function in magnetic islands with aspect ratios of above 0.9 , additionally aspect ratios above this cease to have a well-defined shape anisotropy so ‘writing’ a stable magnetisation direction becomes unrealistic. Two wire widths (60 nm , 30 nm) were selected and for each width simulations were run on a range of wire lengths. In the 60 nm wide case, TMW functioned down to wire lengths of 140 nm . Below this, 180° reversal of magnetisation still occurred controllably down to wire lengths of 90 nm , but in a process more akin to coherent Stoner-Wohlfarth rotation with no well-defined DWs formed. For the 30 nm case, TMW also occurred down to wire lengths of 90 nm . Again controllable 180° magnetisation reversal continued below this, even down to 30 nm by 32 nm elements.

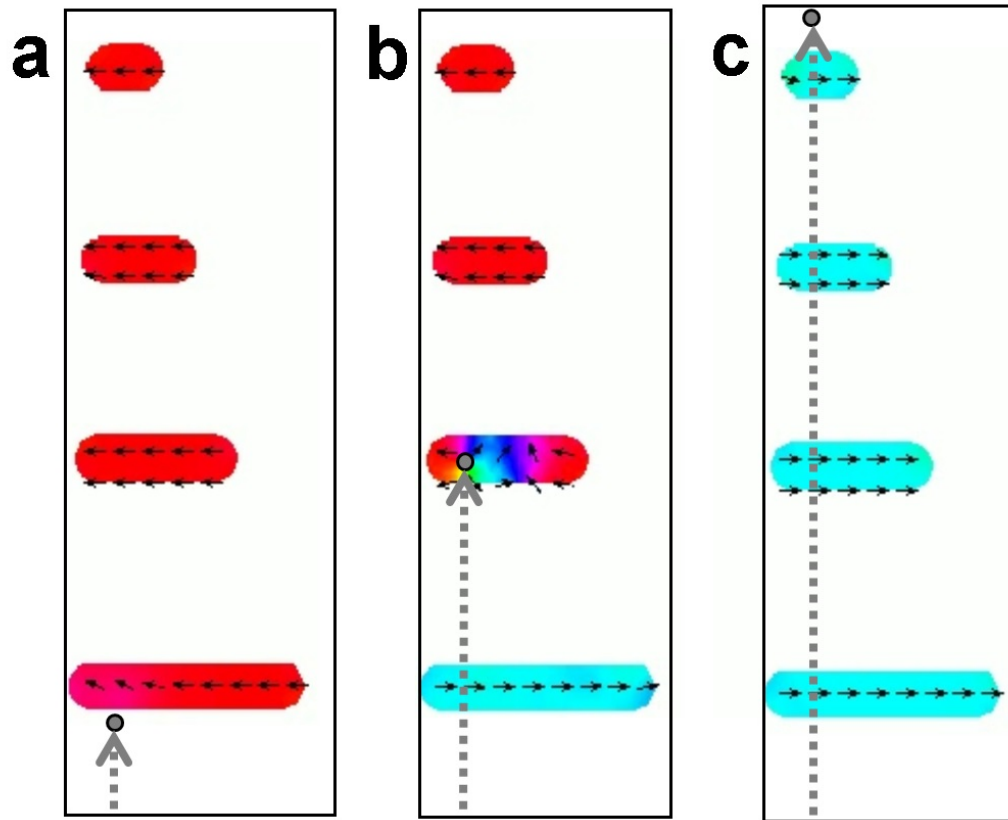


Figure 5.12: Time-evolution series of a micromagnetic simulation of TMW-writing in 60 nm wide, 10 nm thick nanowires of 300 nm, 200 nm, 140 nm and 90 nm length. TMW is seen to retain writing functionality for all nanowires.

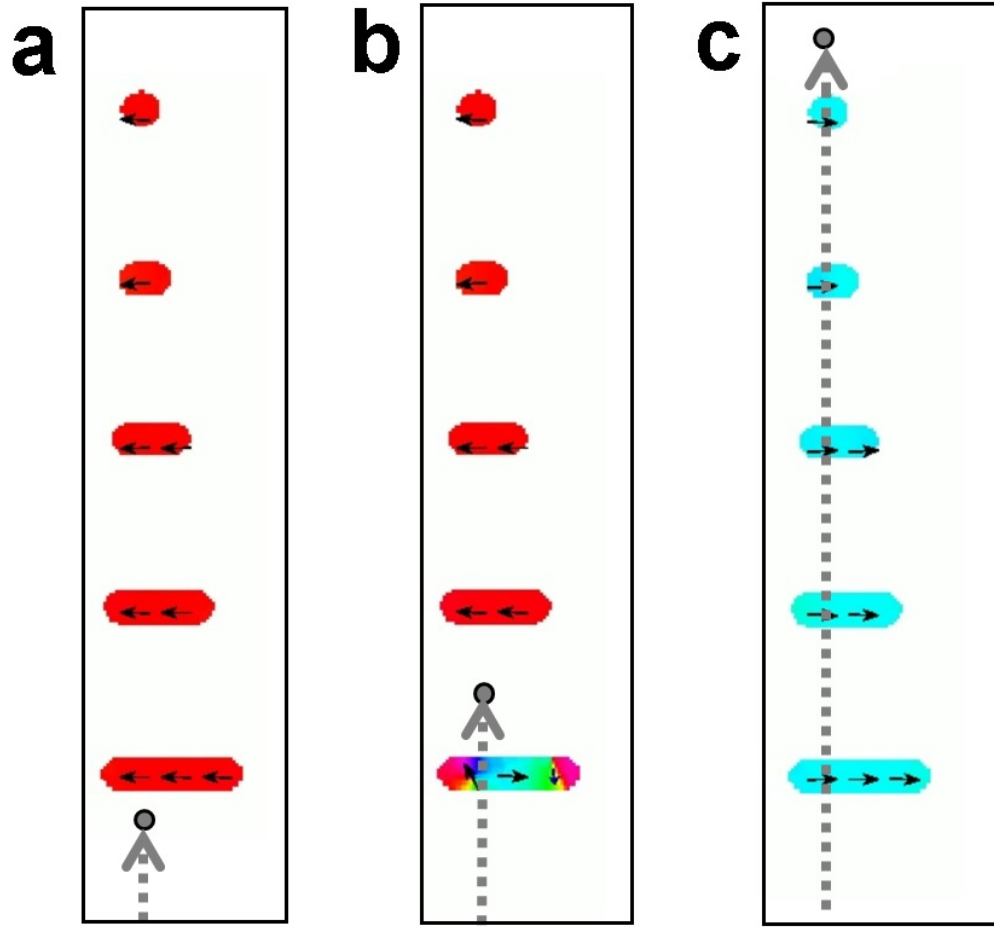


Figure 5.13: Time-evolution series of a micromagnetic simulation of TMW-writing in 30 nm wide, 10 nm thick nanowires of 120 nm, 90 nm, 65 nm, 40 nm and 32 nm length. TMW is seen to retain writing functionality for all nanowires. However, for nanowires shorter than 90 nm the switching mechanism more closely resembles Stoner-Wohlfarth rotation than a dynamic injection of two distinct 180° DWs as seen in longer wires throughout this thesis.

5.11 TMW-written trapped DW states in continuously connected ASI

As shown in figure 5.2, TMW can write states in ASI with no gaps between nanowires at vertices. Figure 5.2 shows the writing of ice-rule obeying $\pm 1q_m$ vertex states. To ascertain the capability of TMW to create excited $\pm 3q_m$ vertices in connected ASI, a write operation was performed on the $1\text{ }\mu\text{m} \times 75\text{ nm} \times 10\text{ nm}$ ASI lattice shown in figure 5.2. The array was prepared with an initialising global field H_{Sat} such that the tip path illustrated by the grey dotted line in figure 5.14 a) would create a chain of monopole-defect vertices along the ‘armchair’ lattice column crossed by the tip. Figure 5.2 shows the resultant state after the TMW write, with fig. 5.2 b) showing the same state with overlaid arrows showing magnetisation direction of written (blue) and unwritten (wires). Interestingly, rather than a chain of excited vertices, a chain of dark contrast $-2q_m$ charges are observed in each wire crossed by the tip (marked by yellow circles in fig. 5.14 b)). The $-2q_m$ charges are very close to a vertex in each case and by labelling the magnetisation directions of surrounding nanowires as in fig. 5.14 b), we can see that the nanowires containing the $-2q_m$ charges have the same polarity magnetic charge at either end. The $2q_m$ charge magnitude and the 180° magnetisation reversal occurring within the ‘host’ wires is a strong suggestion that the TMW-injected charges are 180° DWs, presumably left over from the writing process and unable to unwind into the vertex due to the high energy barrier of creating an excited monopole-defect vertex. This energy barrier is considerably higher in connected ASI due to a large added exchange energy penalty.

Also, the ‘vertical’ (aligned with y-axis) bars to the right-hand side of the lattice column crossed by the tip have been switched by the write operation. As only one of the two TMW-injected 180° DWs is still visible, it appears that the second 180° DW was able to pass through the right-hand vertex and mediate switching of the vertical bars. The fact that only the bars to the right of the tip path have switched suggests a systematic process by which one DW and not the other is permitted to pass through the vertex. As the chirality of each DW is decided by the initial magnetisation direction of the nanowire, and so will be the same in each case, the vertex is

potentially acting as a chirality-selective filter.

While monopole-defect vertex states were not observed, the ability to write bound DWs into a continuous nanowire architecture is a potentially useful one. DWs act as phase shifters for spin waves passing through them^{83,129} ($\delta\phi = \frac{\pi}{2}$ for each 180° DW, so $\delta\phi = \pi$ for a 360° DW) and ASI has been identified as a potential next-generation reconfigurable magnonic crystal^{3,96,97,130}. Also, strings of bound DWs could be inserted into an array to disrupt its intrinsic switching behaviour under globally applied field, allowing access to otherwise hidden regions of the microstate space.

The majority of the experimental work on TMW during this thesis was focused on disconnected ASI arrays, largely because this allows the exact magnetic charge state of each vertex (and therefore the full x,y magnetisation map of the system) to be read out via MFM, rather than just the average vertex charge as in connected systems. As such there is still a good deal to understand about the exact functionality of TMW in connected systems, which are more complex from both an imaging and also a magnetisation dynamics point of view. For example vertices in connected systems have well-defined chiralities which alter the path and behaviour of DWs passing through them, as alluded to above^{93,94}. There may well be a means to write excited vertices in connected systems using TMW, whether by using a stronger tip or even something as simple as choosing a tip path such that write events with the tip moving perpendicular to the target nanowires, rather than at $\pm 60^\circ$ as in 5.14.

5.12 Enhanced control over TMW writing behaviour via tip scan-angle and tip-nanostructure crossing point

So far, only MFM tip velocity vectors oriented perpendicular to the long-axis of the wire have been considered. In many cases there is no need to complicate things further and indeed it has been demonstrated that any conceivable state may be written in disconnected ASI using only perpendicular write-scans. However, as discussed in 4.8 the injection process is highly dependent on the exact manner in which the tip crosses the wire. In this section we investigate potential

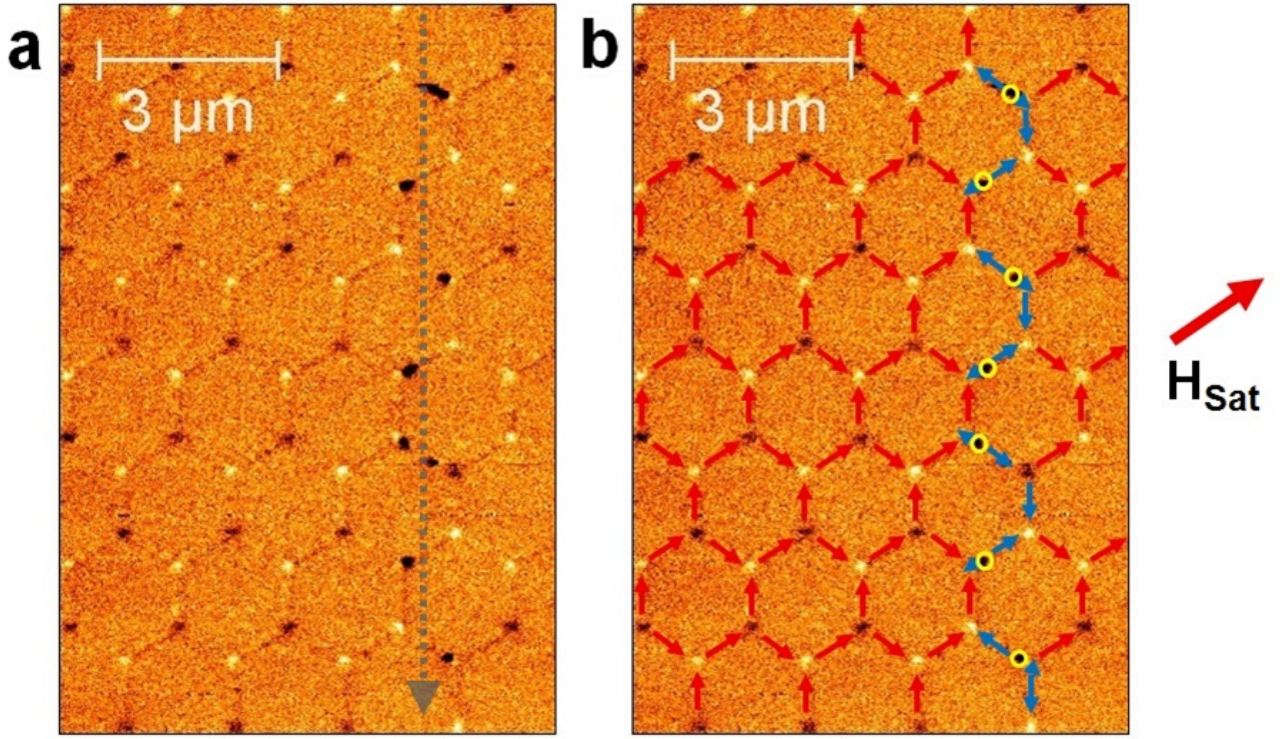


Figure 5.14: Trapped DWs in continuously-connected kagome artificial spin ice. Path and direction of the MFM tip during writing is shown by the dotted grey line and arrow. Panel b) adds overlaid arrows illustrating magnetisation direction, red and blue arrows denote unwritten and written wires respectively, yellow circles indicate position of injected DWs.

ways to leverage these variations in injection behaviour in order to achieve enhanced control over the TMW writing process.

Figure 5.5 shows that as the peak inter-DW separation increases, so does the length of the longest writeable nanowire. This stands to reason as for TMW reversal to occur, the two injected 180° DWs must reach opposite ends of the nanowire. If both DWs unwind into the same wire-end, the wire is returned to its initial state. The further the DWs are separated during the injection process, the easier it is to avoid this and unwind them into opposite ends. One way to vary the inter-DW separation is to increase the magnitude of the moving magnetic charge driving the injection, as demonstrated in figure 5.5. However, if the moving magnetic charge is being supplied by an MFM tip the only means to vary the magnetic charge magnitude is to deposit a thicker CoCr layer on the tip. Unless tips are manufactured in-house or custom-ordered, both of which are expensive and time-consuming options, the only CoCr thickness available suitable for TMW is the 'high-moment' option. Also, varying the inter-DW separation using this method

would require changing the MFM tip each time a different DW separation was desired. As such, it is impractical to employ a range of tips with varying coating thicknesses.

In section 4.8 we have seen that by varying the injection scan angle we may control the momentum imparted to the DWs during injection. As discussed previously, a positively charged tip will impart essentially no momentum to the DW on the side the wire's magnetisation is initially directed, i.e. for a left-magnetised wire the left-hand DW will receive next to zero momentum and the right-hand DW will be accelerated off to the right of the wire. By varying the injection scan angle such that the tip velocity possesses a component directed towards the side of the static DW (in this case to the left, or negative x-direction), momentum may now be imparted to the otherwise-static DW. This results in an increased DW separation, with figure 4.14 showing that a scan angle of -60° will result in a $\sim 1.45\times$ increase in the peak DW separation reached relative to a perpendicular scan. As such, it is possible to achieve a substantial increase to the maximum writeable wire-length with no modification to the tip hardware necessary. The increase in DW separation also varies monotonically with scan angle, allowing an exact desired separation to be achieved by selecting the corresponding scan angle.

Having just considered enhanced functionality granted by varying the scan angle, it is now worth considering what degree of functionality may be retained if the angular degree of freedom is removed from the injection process. In the case of writing ASI states using only perpendicular injection scans, three different global scan angles are required: 0° , 60° and -60° . If using an SPL capable system equipped with an MFM tip there is no issue with this and unless a scan-angle increased DW separation as described above is desired, varying the scan angle to allow for perpendicular crossing of each nanowire in the lattice is the simplest means of writing states. However, SPL systems are expensive, physically cumbersome and inherently limited in their writing throughput due to the need to move a single, relatively large physical structure to each writing site with high precision. One can envision an alternate system whereby the magnetic charge is provided by a topological defect in an adjacent magnetic structure, for instance a current-driven DW in a wire suspended over the ASI lattice. Figure 5.15 shows a schematic of how this may be achieved, with arrays of injection lines each addressing a column of nanowires.

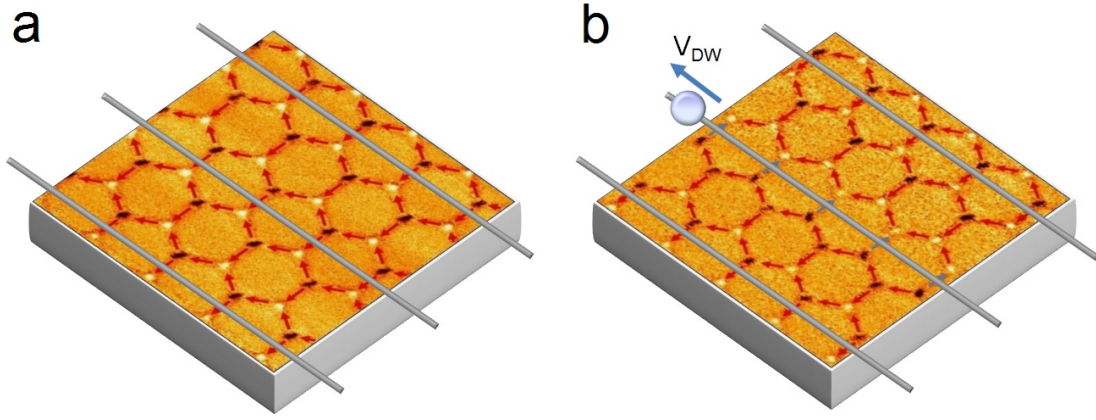


Figure 5.15: Schematic of DW injection striplines used to write ASI states

- a)** ASI lattice with suspended parallel magnetic injection striplines. ASI lattice is initially in a field-saturated state.
- b)** A current pulse is used to move a DW along the central stripline. The stray field of the DW behaves in the same manner as that of a high-moment MFM tip, achieving TMW reversal of a column of nanowires below the stripline.

Such a configuration would benefit from vastly increased write speeds and alleviate the need for a dedicated SPL system and fragile tips. The key drawback here is that pathways for the magnetic charge must be hard-coded into the system at the design stage, also unless a zig-zag style stripline is employed (which has associated DW pinning issues) only a single global scan-angle is allowed. As such, it is worth investigating how TMW performs in an ASI system under these restrictions.

Simulations of various TMW write-scans were performed on a single y-shaped kagome ASI vertex using OOMMF. Initially continuously connected nanowires were considered. There are greater constraints on the unwinding of DWs into the vertex centre here than for disconnected nanowires due to the sharing of topological charges between wires where they meet. The disconnected case may be treated as having the same switching behaviour as the connected system but with no DWs remaining stuck next to the vertex centre due to repulsive topological charge interactions. Nanowires simulated here have dimensions of $1 \mu\text{m} \times 75 \times 10 \text{ nm}^2$.

Six TMW write-scan cases were considered, arising from a combination of three tip-structure crossing points and two scan directions. The six configurations are illustrated schematically in figure 5.16. Starting with 'up' directed injection scans, figure 5.17 a)-d) depicts a time-evolution series of a tip-structure crossing point 100 nm to the left of the vertex centre. As the tip crosses the 'horizontal' wire, a 360° DW is injected. The right-hand 180° DW is then propelled through

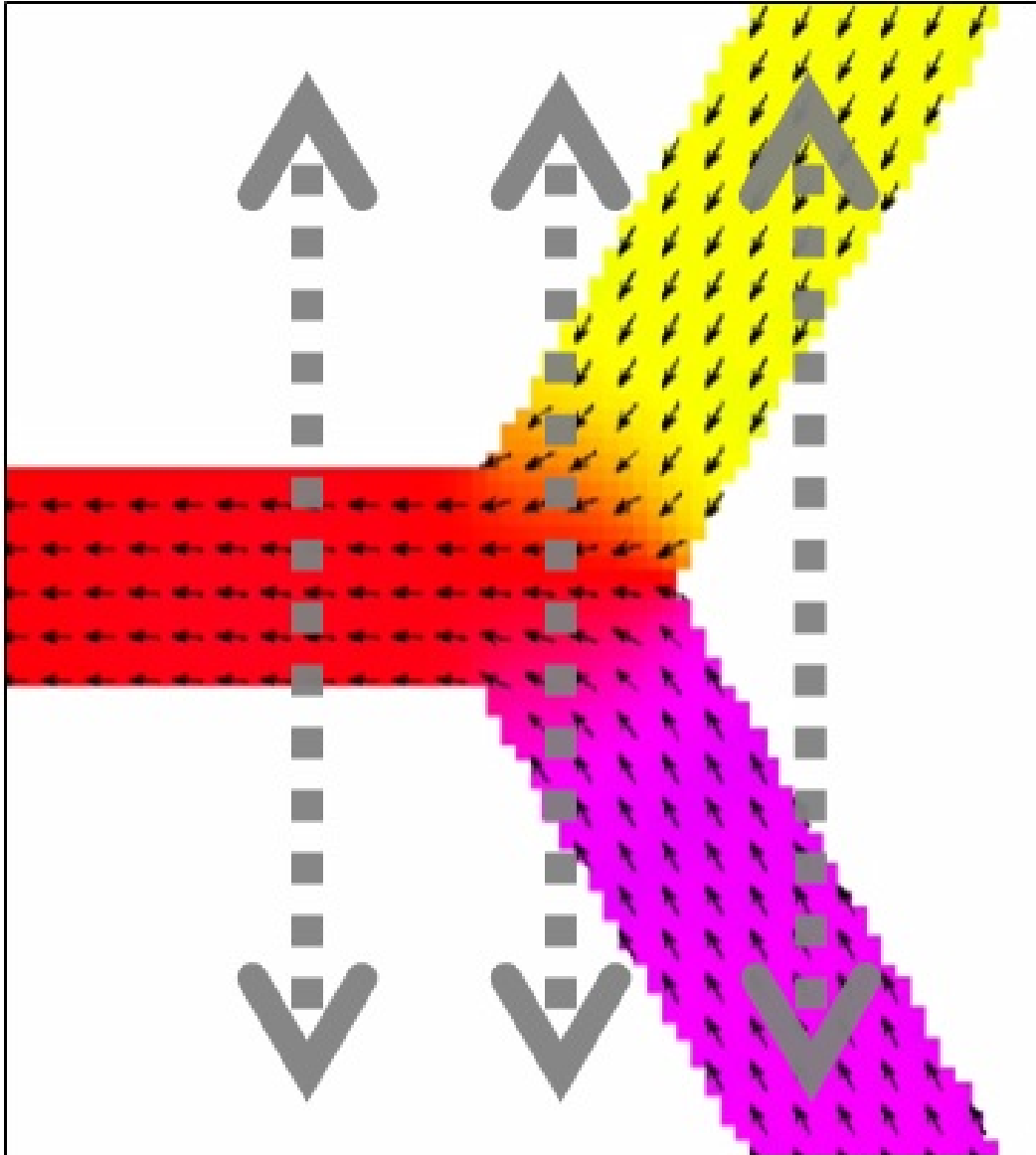


Figure 5.16: Schematic of the six TMW write-scan configurations considered for the OOMMF-simulated restricted scan-angle injection study. Tip-vertex crossing points with lateral displacements of -100 nm (left), 0 nm and 100 nm (right) relative to the vertex centre were studied. For each lateral displacement, both 'up' and 'down' tip-scans were performed.

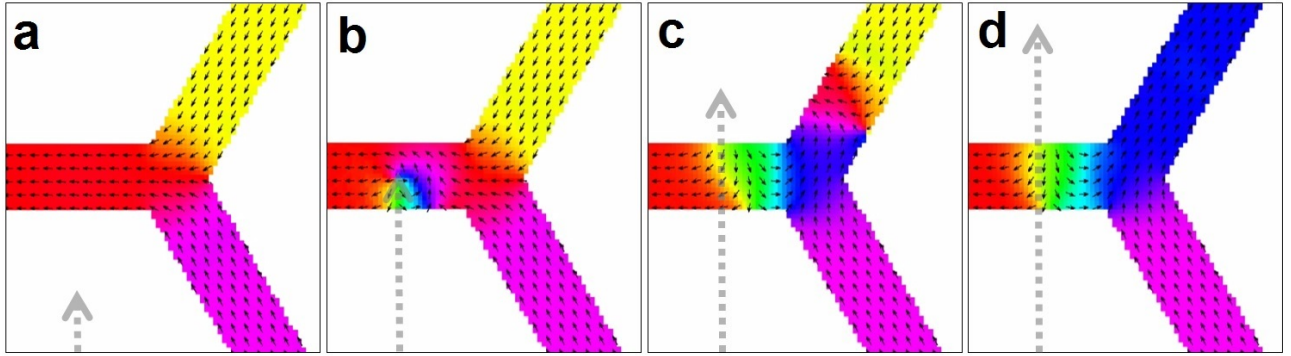


Figure 5.17: OOMMF time-evolution series of TMW write-scan performed on a kagome ASI vertex. Tip-vertex crossing point is 100 nm to the left of the vertex centre. MFM tip path and position are illustrated by dashed grey arrow. Nanowires are $1 \mu\text{m} \times 75 \times 10 \text{ nm}^2$.

a)-d) TMW write-scan performed 100 nm to the left of the vertex centre. The resultant state is a reversed top wire and a static down-chirality 180° DW in the horizontal wire.

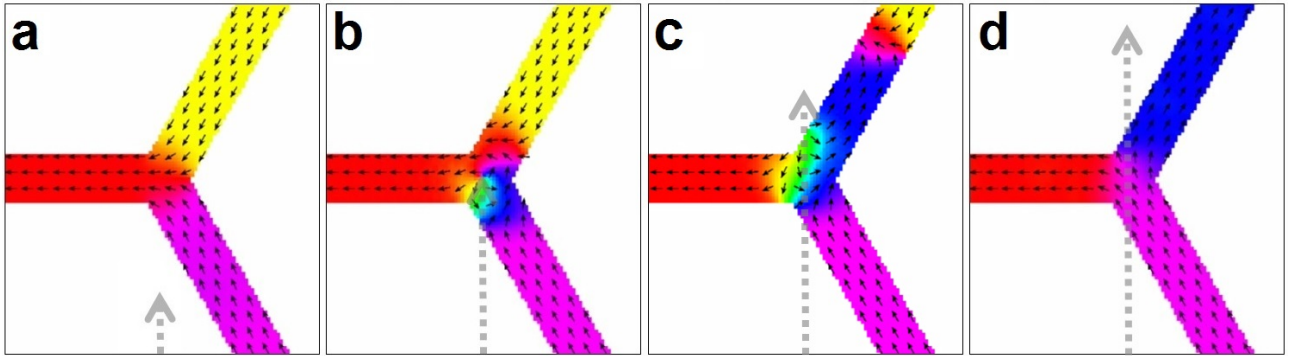


Figure 5.18: OOMMF time-evolution series of TMW write-scan performed on a kagome ASI vertex. Tip-vertex crossing point is directly over vertex centre. MFM tip path and position are illustrated by dashed grey arrow. Nanowires are $1 \mu\text{m} \times 75 \times 10 \text{ nm}^2$.

a)-d) TMW write-scan performed at vertex centre. A 360° DW is nucleated in a distorted state spanning the vertex. The left-hand 180° DW collapses soon after injection due to it spanning too long a distance across the vertex. The right-hand 180° DW is accelerated to the right by the intrinsic injection momentum, selecting the top wire due to its 'up' chirality. The resultant state is a magnetically-reversed top wire.

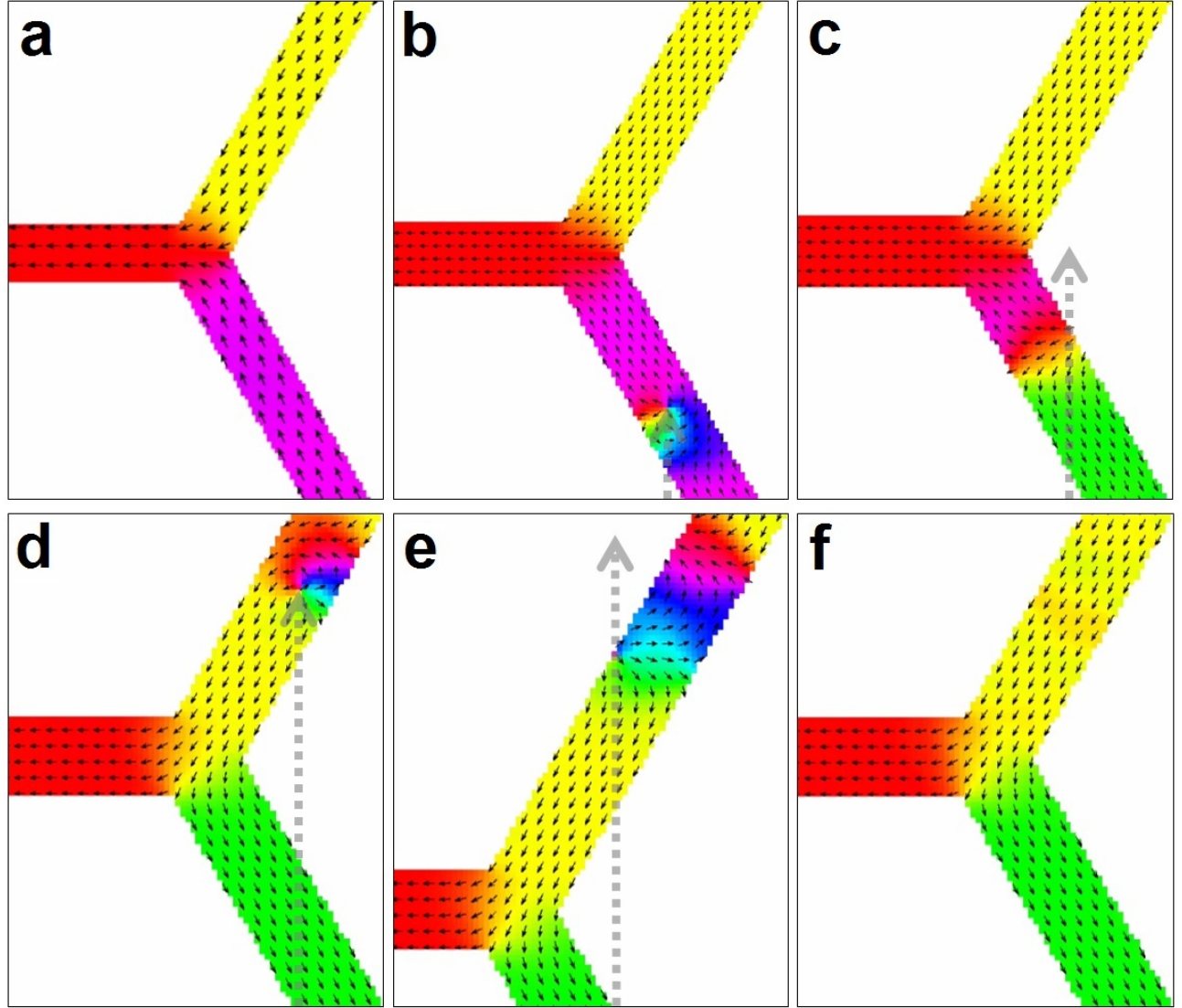


Figure 5.19: OOMMF time-evolution series of TMW write-scan performed on a kagome ASI vertex. Tip-vertex crossing point is 100 nm to the right of the vertex centre. MFM tip path and position are illustrated by dashed grey arrow. Nanowires are $1 \mu\text{m} \times 75 \times 10 \text{ nm}^2$, as in 5.17. a)-f) MFM tip progresses upwards, injecting a 360° DW in the bottom wire as it passes. The injection process here has an effective scan angle of -60° , meaning that as described in section 4.8 the left and right 180° DWs are imparted with momentum directed towards the vertex centre and bottom-right of frame respectively. The right-most 180° DW receives sufficient momentum from the injection process to propagate to the end the wire. The left-most 180° DW moves into the vertex centre (aided by magnetostatic attraction as well as the injection-imparted momentum) and unwinds, leaving the magnetisation of the bottom wire reversed. The MFM tip then encounters the top wire. Here the scan angle is $+60^\circ$, meaning that both DWs are propelled away from the vertex centre towards the top-right of frame. This leads to both 180° DWs unwinding into the top end of the wire, leaving the top wire magnetisation unreversed.

the vertex by the intrinsic momentum of the injection process, continuing through the top wire due to its 'up' chirality (this chiral selectivity has been previously discussed in depth by Zeissler et al⁹⁴). The DW then unwinds at the end of the top wire, effecting magnetisation reversal of the wire. The resultant state of the vertex is now an TMW-reversed top wire with a 'stuck' 180° DW in the horizontal wire, unable to unwind into the vertex. It is worth noting that this arises in the case where reversing the horizontal wire would create a 'monopole' $\pm 3 q_m$ vertex. If the vertex was not a single, isolated vertex but instead part of a larger ASI lattice that had been uniformly magnetised left or right, both 180° DWs would remain bound in the horizontal bar. This allows for the injection bundles of bound magnetic charge into the system, even creating a 'ladder' of trapped 360° DWs from a single TMW write-scan. This could potentially be of interest for magnetotransport experiments, investigating how the behaviour of a large number of bound topological defects effects the collective transport properties of an ASI system.

Figure 5.18 a)-d) depicts a time-evolution series for a write-scan passing directly over the vertex centre. A 360° DW injection process initially occurs as normal, but due to the vertex geometry the MFM tip has to cross a region of Py film around twice the width of the nanowire width. The positive tip-charge used here causes the left-hand 180° DW to form in a straight line following the path of the tip, seen in figure 5.18 b) to c). This means that the DW attempts to span the width of the vertex, placing an increased energetic strain on the DW due to the finite DW energy per unit length¹²². For the dimensions considered here the DW becomes unstable at the increased vertex-width length and collapses immediately after the tip finishes crossing the vertex. The nascent right-hand 180° DW forms in a curved state spreading out to the right of the MFM tip due to the combination of tip-polarity and wire magnetisation. This causes the nascent right-hand DW to spread into the 'top' nanowire as it forms, meaning that this 180° DW needs only span the normal nanowire width. Figure 5.18 b) shows the right-hand 180° DW forming across the 'top' wire. The injection process imparts the DW with momentum propelling it up through the top wire, its progress can be seen in figure 5.18 c). The DW then unwinds into the end of the top wire, effecting magnetisation reversal. The resultant vertex state of a reversed top wire and no 'stuck' DWs can be seen in figure 5.18 f).

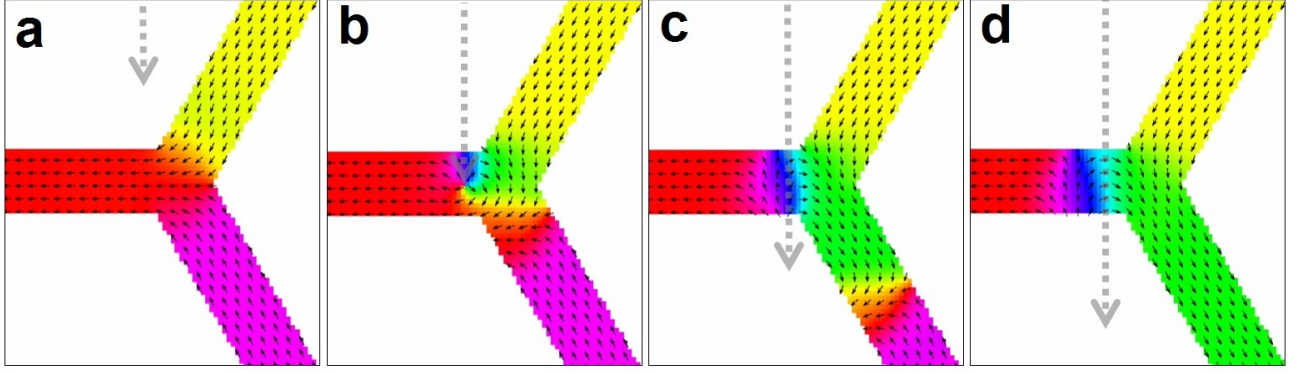


Figure 5.20: OOMMF time-evolution series of ‘downwards’ TMW write-scan performed on a kagome ASI vertex. Tip-vertex crossing point is 100 nm to the left of the vertex centre. MFM tip path and position are illustrated by dashed grey arrow. Nanowires are $1\text{ }\mu\text{m} \times 75 \times 10\text{ nm}^2$. a)-d) TMW write-scan performed 100 nm to the left of the vertex centre. The resultant state is a reversed bottom wire and a static up-chirality 180° DW in the horizontal wire.

As the tip-vertex crossing point is moved right past the vertex centre, the TMW write process becomes slightly more complex. The MFM tip now crosses the ‘top’ and ‘bottom’ wires separately, causing two distinct injection events. An OOMMF time-evolution series for a tip-vertex crossing point 100 nm to the right of the vertex centre is shown in figure 5.19. Figure 5.19 shows the MFM tip progressing upwards through the bottom wire, initiating a 360° DW injection event as it passes. The injection process here has an effective scan angle of -60° , meaning that as described in section 4.8 the left and right-hand 180° DWs are imparted with momentum directed towards the vertex centre and bottom-right of frame respectively. The right-hand 180° DW receives sufficient momentum from the injection process to reach to the bottom-end of the wire, unwinding into the wire end on contact. The left-hand 180° DW is propelled into the vertex centre, aided by magnetostatic attraction to the vertex-centre charge. Upon reaching the vertex-centre it unwinds, leaving the magnetisation of the bottom wire reversed. Figure 5.19 d) shows the MFM tip then encountering the top wire. Here the scan angle is $+60^\circ$, meaning that both DWs are propelled away from the vertex centre towards the top-right of frame. This leads to both 180° DWs unwinding into the top end of the wire, leaving the magnetisation of the top wire unreversed. The resultant vertex state of a magnetically reversed bottom wire and no ‘stuck’ DWs can be seen in figure 5.19 f).

We have now seen that slight variations of the tip-vertex crossing point can have pronounced effects on the TMW writing behaviour. One may select whether to switch the top or bottom

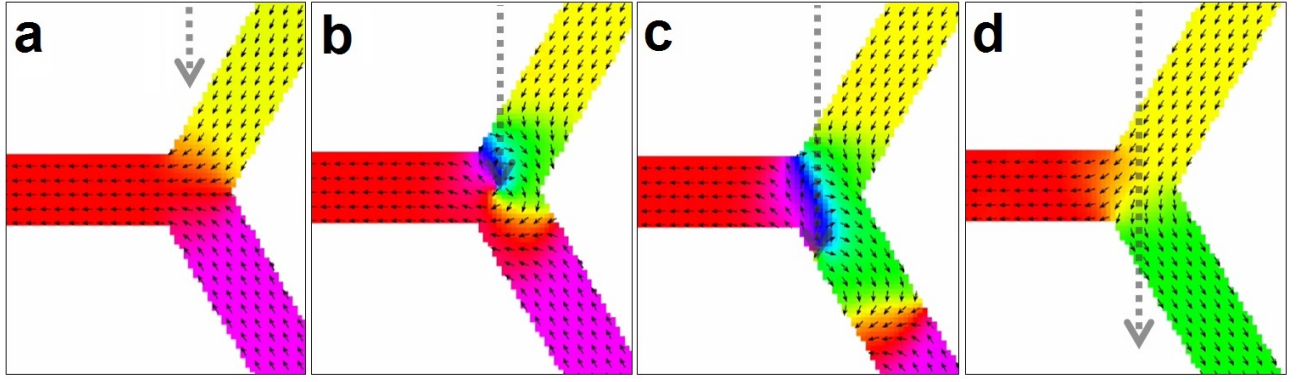


Figure 5.21: OOMMF time-evolution series of TMW write-scan performed on a kagome ASI vertex. Tip-vertex crossing point is directly over vertex centre. MFM tip path and position are illustrated by dashed grey arrow. Nanowires are $1\ \mu\text{m} \times 75 \times 10\ \text{nm}^2$. a)-d) TMW write-scan performed at vertex centre. A 360° DW is nucleated in a distorted state spanning the vertex. The left-hand 180° DW collapses soon after injection due to it spanning too long a distance across the vertex. The right-hand 180° DW is accelerated to the right by the intrinsic injection momentum, selecting the bottom wire due to its 'down' chirality. The resultant state is a magnetically-reversed bottom wire.

angled wires or, in the case where reversing the 'horizontal' wire would result in monopole creation, injecting a bound DW. Let us now consider if it is possible to gain additional control from also varying the scan-direction of the MFM tip. So far all write-scans have been performed with an 'upwards' motion of the tip. Figure 5.20 shows a downwards TMW write-scan with a tip-vertex crossing point 100 nm to the left of the vertex centre. The write behaviour is seen to be a horizontally-reflected version of that observed for an upwards writescan, as seen in figure 5.17. By reversing the motion of the MFM tip, it is now the bottom nanowire which switches magnetisation and the stuck 180° DW is of 'down' chirality instead of 'up' as before.

Figure 5.21 shows a downwards writescan passing directly over the vertex centre. Similarly, the behaviour here is the reverse of that observed for an upwards write-scan at the same tip-vertex crossing point. Figure 5.21 shows the bottom wire being magnetically reversed instead of the top wire for the upwards scan case shown in figure 5.18.

Finally, figure 5.22 shows a downwards write-scan with a tip-vertex crossing point of 100 nm to the right of the vertex centre. Once again, the magnetic switching behaviour is horizontally reflected relative to an upwards write-scan at the same crossing point. The resultant state for the downwards scan is a magnetically-reversed top wire.

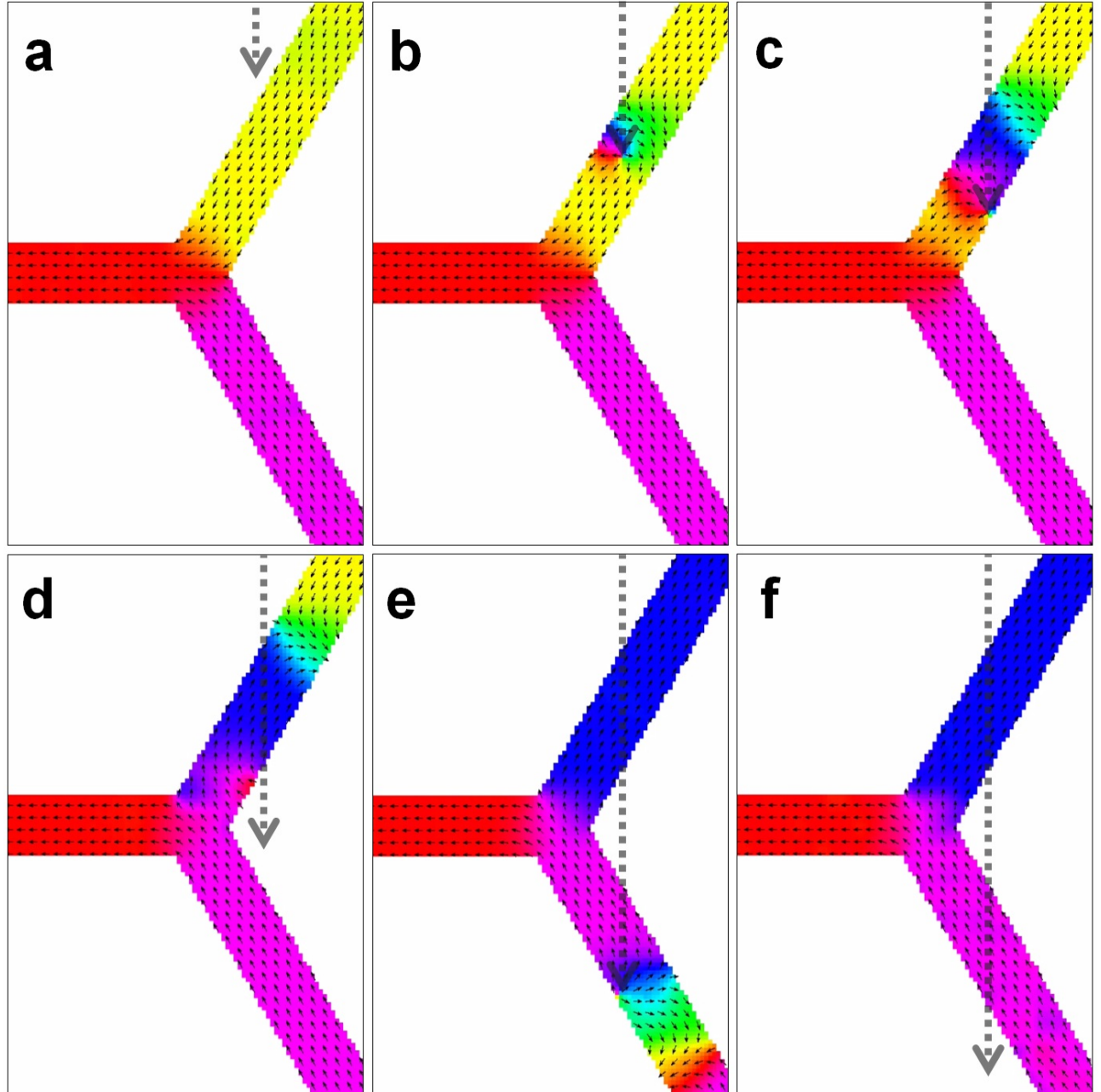


Figure 5.22: OOMMF time-evolution series of TMW write-scan performed on a kagome ASI vertex. Tip-vertex crossing point is 100 nm to the right of vertex centre. MFM tip path and position are illustrated by dashed grey arrow. Nanowires are $1 \mu\text{m} \times 75 \times 10 \text{ nm}^2$.

a)-f) MFM tip progresses downwards, injecting a 360° DW in the top wire as it passes. The injection process here has an effective scan angle of -60° , meaning that as described in section 4.8 the left and right 180° DWs are imparted with momentum directed towards the vertex centre and top-right of frame respectively. The right-most 180° DW receives sufficient momentum from the injection process to propagate to the end the wire. The left-most 180° DW moves into the vertex centre (aided by magnetostatic attraction as well as the injection-imparted momentum) and unwinds, leaving the magnetisation of the top wire reversed. The MFM tip then encounters the top wire. Here the scan angle is $+60^\circ$, meaning that both DWs are propelled away from the vertex centre towards the bottom-right of frame. This leads to both 180° DWs unwinding into the bottom end of the wire, leaving the top wire magnetisation unreversed. The resultant state is a reverse-magnetised top wire.

Having now considered all six TMW write configurations at an ASI vertex, we can see that TMW retains a large degree of flexibility and power even when confined to a single scan-angle. One may select which branch of the vertex is magnetically reversed through a combination of varying the tip-vertex crossing point and the scan-direction of the tip. This demonstrates that using the architecture proposed in figure 5.15, it remains viable to reverse both the ‘horizontal’ and $\pm 60^\circ$ nanowires without having to pattern DW-carrying wires at multiple angles. This is a key point as angled DW-carrying wires will eventually meet and become electrically continuous, preventing individual electrical address of each wire and severely restricting device design.

It is worth noting that for the case of a tip-vertex crossing point 100 nm to the left of the vertex-centre, the vertex essentially performs the role of a chirality-filter for the 180° DWs which pass through it. Figure 5.17 and figure 5.20 each show a 180° DW passing through the vertex-centre before propagating along an angled nanowire branch. In the case of figure 5.17 an up-chirality DW selects the top branch while for figure 5.20 a down-chirality DW selects the bottom branch. This chirally-defined vertex branch selectivity matches with previous findings^{93,94}, however until now for a DW to reach a specific vertex within an ASI lattice it would have to travel through the entire system from an injection point at the lattice edge, likely becoming blocked along the way due to incompatible topological charges at a vertex in the ‘wrong’ magnetisation state¹⁵. If a global magnetic field was used to nucleate DWs within the lattice, no spatial selectivity would be possible and DWs would nucleate everywhere, making controlled studies extremely difficult. With TMW, one may now controllably introduce DWs of desired chirality into any single wire or vertex.

Enhanced flexibility of TMW relative to global field based writing protocols

As we have seen, TMW removes the need for any uniformly applied external global fields. While this has the benefit of both simplifying the experimental setup and allowing integration with systems disrupted by applied fields, in the following section we show that removing the need for

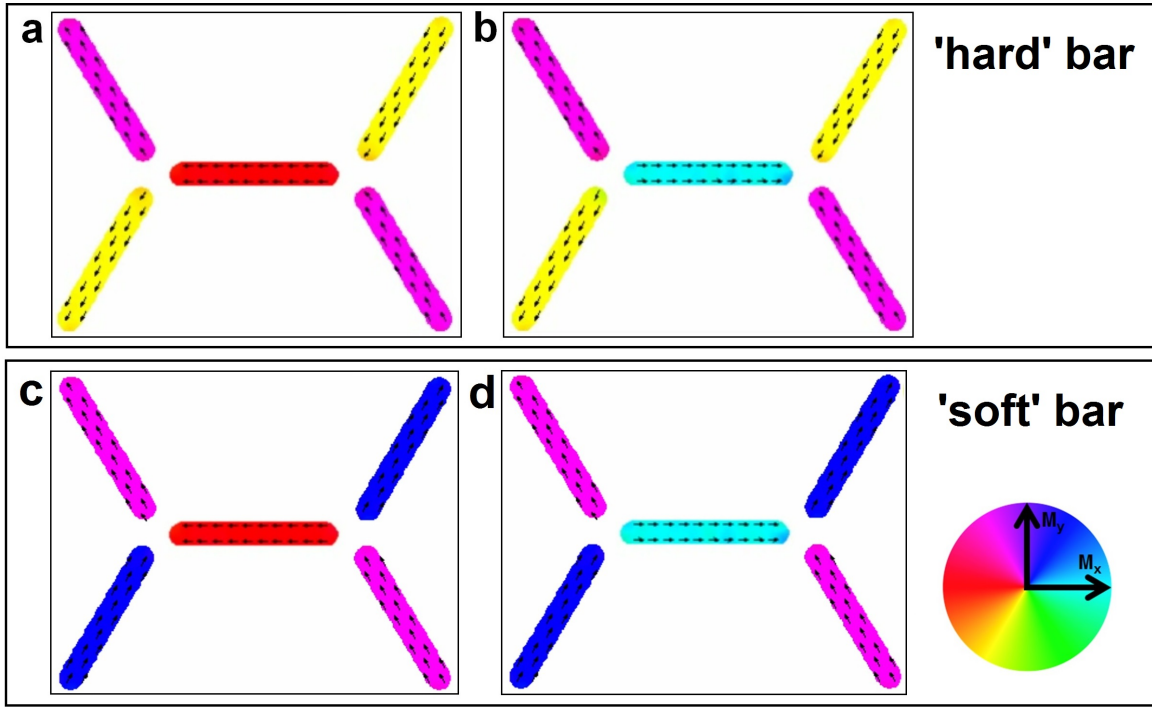


Figure 5.23: Switching events creating excited monopole-defect vertices (a-b) and non-excited vertices (c-d).

a-b) Switching of a ‘hard’ bar resulting in the creation of excited $\pm 3q_m$ vertices.

c-d) Switching of a ‘soft’ bar leaving non-excited vertices.

global fields is essential for any widely applicable magnetic writing technique. Global field based protocols using an MFM tip to locally boost a uniform external field set to 90% of an element’s switching field have shown impressive initial results⁴. However, global field techniques have the critical drawback of being unable to operate in strongly interacting systems where the switching field of an element is significantly modified by the magnetic states of its neighbouring elements. As technological progress necessitates that magnetic arrays operate at the highest possible densities, strong interactions between elements become unavoidable and competitive technologies must retain functionality in strongly interacting systems.

In strongly interacting systems, bars that would create excited vertices when switched have substantially increased switching fields relative to bars creating non-excited vertices. We will refer to these bars as ‘hard’ and ‘soft’ respectively with switching examples shown in figure 5.23 a)-b) (hard) and c)-d) (soft). We performed micromagnetic simulations on the two-vertex kagome ASI configuration shown in figure 5.23 to quantify the range of switching fields in typical strongly and weakly interacting arrays. Bar dimensions of 400 nm X 60 nm X 10 nm were used and vertex

separations giving strong (50 nm) and weak (300 nm) interactions. The system was initialised as in figure 5.23 a). Initially we start by forcing the $\pm 60^\circ$ bars to remain fixed, to find the switching field of the hard central bar. We then relax that condition to find the switching field of the $\pm 60^\circ$ bars. The process was then repeated for soft bars with the system initialised as in figure 5.23 c).

<i>Hard bar switching fields</i>	Central bar switching field	$\pm 60^\circ$ bars switching field	Percentage difference	Difference in switching field
Strongly interacting case (50 nm gap)	97.5 mT	72.5 mT	26%	25 mT
Weakly interacting case (300 nm gap)	87.5 mT	72.5 mT	17%	15 mT
Single isolated bar	82.5 mT	NA	NA	NA

<i>Soft bar switching fields</i>	Central bar switching field	$\pm 60^\circ$ bars switching field	Percentage difference	Difference in switching field
Strongly interacting case (50 nm gap)	84.5 mT	72.5 mT	14%	12 mT
Weakly interacting case (300 nm gap)	84.5 mT	72.5 mT	14%	12 mT
Single isolated bar	82.5 mT	NA	NA	NA

The tables above show the observed switching fields. It can be seen that for a strongly interacting system, the switching field of a hard bar is increased such that a global field tuned to 90% of its switching field will switch all soft bars in the system. As such, existing global field based techniques are seen to be inoperable in strongly interacting systems. Indeed as array densities increase, necessary for technological progress, the difference in switching field for hard bars will rise even higher¹³¹ and supplying sufficient local field boosts without writing adjacent bars becomes an untenable proposition. As TMW operates on a distinctly different concept, directly manipulating topological defects to achieve controlled switching with no need for external global fields, the above issues do not apply as demonstrated by our TMW-writing of complex excited states in strongly-interacting systems.

5.13 Confirmation of strongly-interacting ASI

A key benefit to TMW relative to previous methods for writing magnetisation states in nanopatterned magnetic arrays is its ability to function in strongly-interacting systems. Up to this point it has essentially been left to the reader to trust that the ASI arrays considered in this

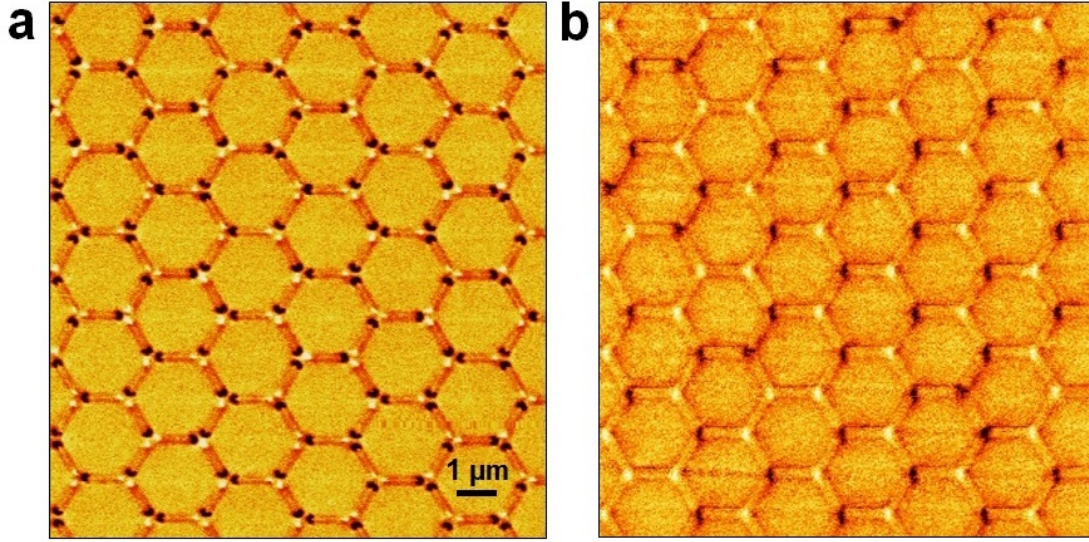


Figure 5.24: MFM images of as-grown ASI arrays.

a) Disconnected and **b)** connected ASI arrays immediately after deposition. The ice-rules are obeyed at every vertex in both arrays, indicating a strongly-interacting system. The disconnected array has vertex gaps of 50 nm.

thesis fall within the strongly-interacting regime. For such an integral part to the claims of TMW's novelty relative to prior art, it is important to provide experimental verification that the ASI arrays in this thesis are indeed strongly interacting.

To confirm that ASI arrays were in the strongly-interacting regime, structures were imaged via MFM immediately after deposition. In a randomly prepared non-interacting ASI system where the orientation of each macrospin is independent of its neighbours, the chance of observing no $\pm 3 q_m$ monopole-defect vertices is given by the ratio of ice-rule obeying microstates $\frac{3}{\sqrt{2}}^N$, where N is the number of vertices, to the total number of microstates 2^n where n is the number of macrospins. A non-interacting $15 \mu\text{m} \times 15 \mu\text{m}$ array containing 118 vertices and 222 macrospins therefore has a 2.1^{-11} probability of containing no $\pm 3 q_m$ vertices. Figure 5.24 shows MFM images of disconnected (a) and connected (b) as-grown arrays with no ice-rule violations observed, showing structures are in the strongly-interacting ASI regime.

5.14 Practicalities of experimentally performing TMW

To the end-user, utilising the TMW process is extremely simple. An interface is provided to define the path of the tip relative to the nanopatterned structures (and therefore the magnetic state to be written) graphically, by drawing over an existing AFM/MFM image or CAD schematic, or by defining an (x, y) position list. Writing does not occur when the tip-sample separation is over 5-10 nm, allowing for free movement of the tip around samples between writing events by raising the tip.

For both systems, control protocols were developed whereby the user may specify a set of (x, y) position co-ordinates to describe the path of the tip relative to the nanopatterned structures to be written along with commands to raise the tip in z to a non-interacting height above the surface to allow for free movement around samples or lower the tip such that TMW writing occurs. Alternatively, the user may load an image of the structures to be written and graphically define the tip path with a simple point and click interface. The image may be a previously collected AFM or MFM image, CAD schematic or any commonly used image file format.

Writing has been successfully performed on wire-like structures of widths and lengths up to 150 nm and 4 μm with tip speeds of up to 400 $\mu\text{m/s}$. Significantly faster tip speeds of up to 200 m/s show successful writing in micromagnetic simulations but cannot yet be experimentally tested using our current equipment.

5.14.1 Micromagnetic simulation details

Additional insight into the reversal dynamics of the nanowires was provided by performing a series of micromagnetic simulations using the object-oriented micromagnetic framework (OOMMF)⁶². Typical micromagnetic parameters for permalloy were used, i.e. saturation magnetisation, $M_S = 860 \times 10^3$ A/m, exchange stiffness, $A = 13 \times 10^{-12}$ J/m, zero magnetocrystalline anisotropy and a Gilbert damping parameter, $\alpha = 0.01$. The point probe approximation (that at small tip-sample separations an MFM tip may be described by a point

monopole moment^{114,115}) was used. This approximation is widely used in MFM simulations and previous work analysing systems with similar dynamics has shown that both dipolar and monopolar-modelled tip fields induce tip-localised magnetisation vortices in ferromagnetic thin films^{91,92}.

The simulated nanowires were 10 nm thick with widths of 75 nm and were divided into $2.5 \times 2.5 \times 10$ nm cells. Finite-length wires of 0.5-3 μm length were studied along with a semi-infinite wire of 10 μm length where the demagnetisation effects from the wire ends were corrected for by the inclusion of plates of fixed magnetic charge at the nanowire ends¹²².

The field from the MFM tip was modelled as a single magnetic charge, q_{tip} , producing a radial field $H = \frac{\mu_0}{4\pi} \frac{q_{tip}}{r^2}$ at a distance r from the charge. During the simulation this magnetic charge moved perpendicular to the nanowire axis in 1 nm steps every 10 ps representing a velocity of 100 m/s. This is faster than the velocities of $\sim 10^{-4}$ m/s investigated experimentally. However, the simulated speed is well below those associated with exciting any precessional spin modes and is believed to be reasonable in this case. Running simulations with tip velocities matching those used in experiment was prohibitively slow and computationally expensive. To ascertain whether the increased tip velocity in simulation had a significant effect on the micromagnetic dynamics of the tip-wire interaction, initial exploratory simulations were performed with tip velocities down to 10^{-1} m/s. The same DW injection process and resultant micromagnetic states were observed across all simulated tip velocities, supporting the validity of employing $v_{tip} \sim 10^{-4}$ m/s values. To avoid a discrete jump in applied field on the nanowire when starting the simulation, the magnetic charge was initialised 300 nm away from the nanowire in the plane of the wire. Following the magnetic charge interaction with the wire, the wire was allowed to relax to an energetically stable state in zero field to obtain its final configuration.

5.15 Conclusions

In this chapter we have presented an outline and demonstration of TMW, a powerful and simple-to-use magnetic writing technique broadening the field of magnetic-SPL and allowing

unprecedented access to novel and exotic states in ASI and related nanomagnetic systems. Reconfigurable magnetic metamaterials and the associated technological benefits are now a realistic and practical prospect, setting the scene for a period of rapid development as long-proposed designs including hardware neural-networks and reconfigurable magnonic crystals may now be experimentally realised. The spin-crystal ground state and negative-temperature monopole-chain states written in this work are evidence that all possible magnetic configurations are now accessible, both previously studied yet hard-to-access states through to states exhibiting exciting new physics, considered unattainable until now.

While the ultimate throughput of any SPL technique is limited by the mechanical nature of a moving tip, the magnetic charge supplied here by an MFM tip may come from a number of sources. A DW in a nanowire of the dimensions discussed here has $q_m \approx 10^{-9} Am$, within the limits for achieving TMW. One can envisage a system comprising a three-dimensional network of nanowires whereby current-controlled DWs replace the MFM tip, greatly enhancing flexibility, throughput and integration with existing technologies.

6 | Conclusions and future outlook

The primary goal of this thesis was to advance and expand the toolbox of magnetic charge sculpting tools available to the research community. In this respect the thesis has been a success, resulting in the development of two powerful techniques for the fine control of magnetic charge landscapes and opening up previously inaccessible regions of the ASI microstate space.

This thesis has shown that it is possible to re-purpose widely available equipment to fulfil novel roles and meet long standing needs within the wider research community. It was my aim to create tools which are relatively simple to employ and require little specialist equipment or modification to equipment, while still significantly enhancing functionality relative to existing techniques. The realisation of the spin-crystal kagome ice ground state and negative effective-temperature states presented in chapter 5 are evidence of the latter, both examples of the newly accessible regions of ASI microstate space made available by the work presented in this thesis. The experimental results were all achieved with unmodified, widely-available surface-probe microscopy systems, greatly enhancing the likelihood of the methods presented here being of real use to the research community.

Looking forward, we stand at an exciting point in the development of the field. Long-promised designs for leveraging nanomagnetic systems to achieve neuromorphic computation and reconfigurable magnonic crystals now stand a real chance of experimental realisation, with the required fine micromagnetic manipulation provided by topological defect-driven magnetic writing.

In terms of neuromorphic computation, there are three essential criteria that a system must

satisfy to function as a neural network (the underlying functional element required for performing neuromorphic computations): that states may be written (1), read (2) and that the complexity of the information used to train the network must match the complexity of the network itself (3). To date, artificial spin ice satisfies conditions 1 (albeit crudely, using globally-uniform magnetic fields) and 2. Prior to this work, condition 3 was far from satisfied - the handful of global field accessible microstates are an infinitesimal fraction of the total microstate space. A failure to meet the third criteria results in the network merely memorising the simple training data, unable to usefully process additional datasets. Through topological defect-driven magnetic writing we provide a full satisfaction of condition 1, and in the process a solution to creating training datasets matching the full network complexity via intricately written microstates (or a series thereof). ASI has demonstrated the ability to exhibit ‘return-point memory’, a property whereby once placed into an arbitrary microstate, it will return to said state with high-fidelity even after numerous minor-loop (non-saturating) field cycles^{132,133}. Further, theory has shown the significance that controllable switching of even a select few macrospins may have on the system’s trajectory through microstate space, even when state evolution is driven by globally uniform fields or thermal switching^{32,95} - showing further evidence of capacity for training. These are promising early results, but represent perhaps only a hint at ASI’s full capacity for memory and training applications. We hope to now use the methods developed in this thesis to fully explore such behaviours.

Reconfigurable magnonic crystals stand in a similar embryonic state, with early proof-of-concept results demonstrating microstate-dependent spin wave frequency response in simple 1D ‘crystals’ comprising a ‘zebra-crossing’-like array of nanowires (i.e. with wires arranged with their long-axes perpendicular to the long-axis of the array)^{12,134–136} and even initial forays into ASI-based magnonic crystals^{11,137–139}. However, functionality is again limited by the lack of means to access more than a handful of micromagnetic configurations. Theoretical work has suggested that by expanding to a 2D system (such as ASI) and adding full-access to the total microstate space (for example by TMW), one may gain fine control over diverse aspects of the spin wave response, including peak position (in frequency space) and height and the ability to open and close band-gaps^{3,97,104,140–142}. The work in this thesis on integrating magnetic charge manipulation

tools with ASI, a promising 2D reconfigurable magnonic crystal candidate, paves the way for in-depth studies on exactly how much control may be exercised over magnonic band structures.

The fields of ASI and related nanomagnetic arrays now stand at a pivotal point. The techniques required to realise long-promised designs enhancing the scope of the field in both fundamental and applied directions have now been demonstrated. How the community makes use of these tools will be interesting to watch, which hypothetical designs will be proved viable, and what unexpected discoveries will the new state-writing methodologies lead to? Perhaps most interestingly, which fields outside of the more obvious candidates such as ASI will find use for these techniques?

FIN

7 | Micromagnetic time-evolution videos

Videos of the time evolution of key dynamic micromagnetic processes can be accessed online. Videos pertaining to chapters 4 and 5 have been placed in two separate YouTube playlists, detailed below. They can be accessed by their hyperlinks or by copying the chapter titles into Google:

Chapter 4: A novel method for the injection and manipulation of magnetic charge states in nanostructures

<https://www.youtube.com/playlist?list=PLtNIYiPezEZ-tFlTWojcN645PdC6biU3Q>

Chapter 5: Topological defect-driven magnetic writing: A new approach to magnetic SPL

<https://www.youtube.com/playlist?list=PLtNIYiPezEZ9i69fsTJPjOr5snyaLJZMt>

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