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Switching mechanisms and barriers in artificial spin ice systems

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Abstract

Spin Ice systems are frustrated magnetic systems, where the geometrical frustration gives rise to degeneracy of the ground-state, and enables excited states, where magnetic charges can be quantified. The cryogenic temperatures, and atomic sizes make it difficult to control these magnetic charges, also commonly referred to as "emergent magnetic monopoles". Artificial spin ice systems are magnetic meta-materials fabricated to study the magnetic charges on more accessible platforms. Magnetic moments of single atoms from spin ice materials, are replaced by the elongated magnetic islands, always magnetized along their longer axis due to the shape anisotropy. Commonly arranged on square, or hexagonal lattices, each nanoisland can reverse its magnetization state, if enough energy is introduced in the system. If the energy supplied is larger than the required switching barrier, the magnetization of one ASI element can be reversed.

In this cumulative Master thesis the switching mechanisms behind the reversals of single magnetic elements in ASI are investigated. In the first published article, different methods for the calculation of the energy barriers in a square ASI are compared, and a full micromagnetic model using the string method was developed. It is shown, that proper micromagnetic modelling can lower the mean-field barriers by 35%. Furthermore, it was shown, that the reversal process can occur via clockwise or counterclockwise rotation, leading to a redefinition of switching rates, where both barriers are encountered.

In a second publication, the developed full micromagnetic model was applied to explain the experimental findings, where the ASI lattice grown on top of a heavy metal layer (Pt), becomes thermally super-active, as the asymmetric interaction causes by the HM layer lowers the energy barriers, introducing an effective hard-axis anisotropy.

A more comprehensive study on the reversal mechanisms, temporal and spatial correlation of the switching processes in ASI were investigated in a third publication. Here, it was shown that the switching mechanisms can be magneto-statically or exchange dominated, based on the shape of the magnetic nano-islands.

In the final publication included in this Master thesis, a novel three dimensional ASI was designed, which solves all the shortcomings of the conventional ASI. Here, the switching mechanisms and barriers are investigated to prove that the tension of Dirac strings vanishes, and that the magnetic charges present in the lattice become effectively unbound, leading to uncorrelated, highly mobile, binary information carriers.

Kurzfassung

Spin Ice Systeme sind frustrierte magnetische Systeme, bei denen die geometrische Frustration zur Entartung des Grundzustands führt und angeregte Zustände ermöglicht, in denen magnetische Ladungen quantifiziert werden können. Die kryogenen Temperaturen und die atomaren Skalen erschweren die Kontrolle der magnetischen Ladungen, die auch als "magnetische Monopole" bezeichnet werden. Artificial Spin Ice Systeme sind magnetische Metamaterialien, die hergestellt werden, um die magnetischen Ladungen auf besser zugänglichen Plattformen zu untersuchen. Die magnetischen Momente einzelner Atome von Spin Ice Materialien werden durch längliche magnetische Inseln ersetzt, die aufgrund der Formanisotropie immer entlang ihrer längeren Achse magnetisiert sind. Üblicherweise auf quadratischen oder hexagonalen Gittern angeordnet, kann jede Nanoinsel ihren Magnetisierungszustand umkehren, wenn dem System genügend Energie zugeführt wird. Ist die zugeführte Energie größer als die erforderliche Schaltbarriere, kann die Magnetisierung eines ASI-Elements umgekehrt werden.

In dieser kumulativen Masterarbeit werden die Schaltmechanismen hinter den Umkehrungen einzelner magnetischer Elemente in ASI untersucht. In dem ersten veröffentlichten Artikel werden verschiedene Methoden zur Berechnung der Energiebarrieren in einem quadratischen ASI verglichen und ein vollständiges mikromagnetisches Modell unter Verwendung der String-Methode entwickelt. Es wird gezeigt, dass eine korrekte mikromagnetische Modellierung die mittleren Feldbarrieren um 35% senken kann. Darüber hinaus wurde gezeigt, dass der Umkehrprozess durch eine Drehung im oder gegen den Uhrzeigersinn erfolgen kann, was zu einer Neudefinition der Schaltraten führt, wenn beide Barrieren betrachtet werden. In einer zweiten Veröffentlichung wurde das entwickelte vollständige mikromagnetische Modell angewandt, um die experimentellen Ergebnisse zu erklären, bei denen das ASI-Gitter, das auf einer Schwermetallschicht (Pt) gewachsen ist, thermisch superaktiv wird, da die asymmetrische Austauschwechselwirkung, die durch die Pt Schicht verursacht wird, die Energiebarrieren senkt und eine effektive Hartachsenanisotropie einführt.

Eine umfassendere Studie über die Umkehrmechanismen sowie die zeitliche und räumliche Korrelation der Schaltprozesse in ASI wurde in einer dritten Veröffentlichung untersucht.

In der letzten Publikation dieser Masterarbeit wurde ein neuartiges dreidimensionales ASI entworfen, das alle Mängel des herkömmlichen ASI behebt. Hier werden die Schaltmechanismen und Barrieren untersucht, um zu beweisen, dass die Spannung der Dirac-Strings verschwindet und dass die im Gitter vorhandenen magnetischen Ladungen effektiv ungebunden werden, was zu unkorrelierten, sehr mobilen, binären Informationsträgern führt.

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Chapter 1

Introduction

The following section is intended to provide the reader the necessary background knowledge to understand the core research of the next chapters and articles. At this place it is good to mention, that the structure of this chapter will follow a path towards more and more applied physics. In the first part, we discuss the main notions of magnetism and magnetic monopoles, and continue with the main ideas behind micromagnetic theory and simulations. A general introduction into the fields of spin ice, and artificial spin ice are provided in the further sections.

1.1 Maxwell's Equations and Magnetic Monopoles

The most known theory that describes the electro-magnetic fields is summarized in Maxwell's Equations, a series of equations with which the electro-magnetic fields and waves can be fully described. One of the equations, lays the foundations of magnetic materials. Namely, the magnetic field is divergence free. What does it mean? It means that there is no particle from which the magnetic fields are sourcing, in contrast to electric field. For instance, an electron is the source of a divergent electric field. However this one particular Maxwell equation, namely, the Gauss's law for magnetism says that

$$\nabla \cdot \mathbf{B} = 0, \quad (1.1)$$

where ∇ is the Nabla operator, and $\mathbf{B}$ is the magnetic flux density. This means that the magnetic fields are source-free, as shown in Fig. 1.1; hence, there are no magnetic monopoles. If one divides a magnet bar with a south pole, and a north pole, to two equal magnets, than each of the newly obtained magnets will have a south and a north pole, respectively.

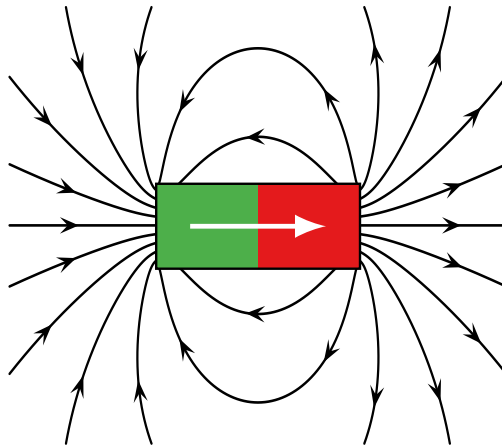


Figure 1.1: Magnetic field of a magnetic bar, where the magnetic fields build do not diverge and are closed back at the other pole. Figure adapted from [1].

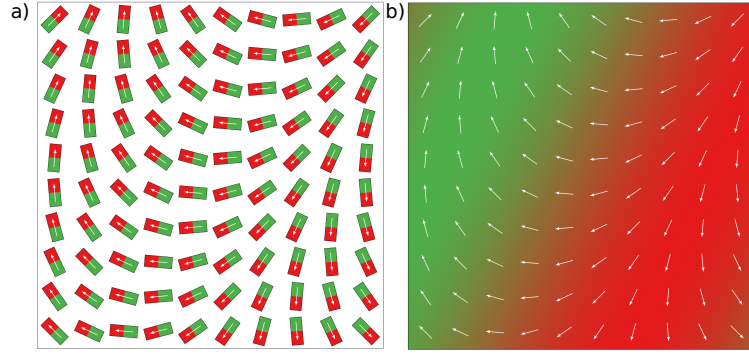


Figure 1.2: Basic principle of micromagnetic approximation, where the discrete magnetic moments from (a) are approximated to the continuous, homogeneous magnetization vector field from (b). Original figure created by Claas Abert [5, 6, 4] and adapted with his permission.

Yet, the physicists have been hunting these magnetic monopoles for a long time now. Paul Dirac succeeded to postulate their existence, which was then linked to the quantization of the electric charge [2]. Dirac said that one single magnetic monopole is enough to quantize all of the electric charges. Today, it is a well known fact that the charge of one electron is quantized and equals $e = -1.6 \times 10^{-19} \text{ C}$. However, the magnetic monopoles are still to be found.

1.2 Micromagnetics

Even though the description of magnetic fields and magnetic potentials are fully covered by the Maxwell equations, magnetism in magnetic systems is a much more complicated many body system. For the sake of simplicity, let us consider a perfect single crystal of nickel, where the unit cell is a face centered cubic cell. Each Ni atom generates its own local angular momentum; hence a magnetic moment. These magnetic moments, or as we usually call them: magnetic spins, align parallel in a ferromagnetic material. This many body system can be described theoretically in terms of density functional theory, or even an atomistic theory. In both cases single atoms, and thus, single magnetic moments have to be calculated, which limits the sizes of materials that can be studied.

A very powerful tool emerged over the past 60 years, since the initial calculations done by Brown in 1959 [3]: *Micromagnetics*. The very basic idea is to upscale the electronic picture of DFT, and the atomistic picture, to the micron scale, where everything becomes homogeneous, and continuous. This reduces the complexity of the problems significantly, while keeping the ability to qualitatively describe the most important phenomena. Figure 1.2 illustrates the transition from a discrete (a) to a continuous (b) description of magnetic systems. In the discrete picture, one can sum up all the magnetic moments, and obtain a macroscopic entity which describes the magnetic properties of the magnet in the continuous model, where

$$\int_{\Omega} \mathbf{M}(\mathbf{x}) d\mathbf{x} = \sum_i^N \mathbb{1}_{\Omega} \boldsymbol{\mu}_i, \quad (1.2)$$

with Ω being the volume of magnetic body, and $\mathbf{M}(\mathbf{x})$ the continuous magnetization, $\boldsymbol{\mu}_i$ being the magnetic moment of the atom i , and $\mathbb{1}_{\Omega}$ is the indicator function of Ω [4].

Due to the continuous character the normalization constraint emerges, where the magnetic moments are represented by the continuous vector field with normalized vectors, where the strength of the field is given by a scalar value; hence,

$$\mathbf{M}(\mathbf{x}) = M_s \mathbf{m}(\mathbf{x}), \quad (1.3)$$

where M_s is the saturation magnetization, and

$$|\mathbf{m}(\mathbf{x})| = 1, \quad (1.4)$$

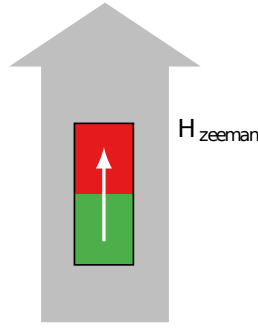


Figure 1.3: Magnetic moment under the influence of an external Zeeman field, where the magnetization aligns with the external field. Original figure created by Claas Abert [5, 6, 4] and adapted with his permission.

being the (normalized) unit magnetization. For the remaining sections of this chapter, and in the articles, we refer to $\mathbf{m}(\mathbf{x})$ simply as magnetization, where as each vector of this field is also called a *magnetic spin*, or only *spin*.

Distinct magnetization fields in different magnetic bodies will have different properties, which are differentiated by their energetic contributions. In many body systems and in a quantum mechanical picture the energetics of the systems are usually given in terms of Hamiltonian $\mathcal{H}$, which basically sums over all the energetic contributions acting on the many body system. In micromagnetics, a semi-classical theory, we describe the quantum mechanical effects in an approximated fashion by means of effective fields, and energies, rather than pure quantum-mechanical hamiltonians.

In the following, the main energetic contributions will be described, that are necessary to understand the core notions of attached papers.

1.2.1 Zeeman energy

Perhaps the easiest form of energy in magnetism in general is when an external magnetic field is applied. The magnetizations will simply try to follow the magnetic field, as seen in Fig. 1.3 The energy of the external field acting on the magnet is called the Zeeman energy and relates the magnetization to the applied external magnetic field and is defined as

$$E_{\text{zee}} = -\mu_0 \int_{\Omega} M_s \mathbf{m} \cdot \mathbf{H}_{\text{zee}} d\mathbf{x}, \quad (1.5)$$

where H_{zee} is the applied external field.

1.2.2 Exchange Energy

This energy originates from quantum mechanical effects, whereas the interaction is then described by the Heisenberg exchange interaction, which simply gives the correlation of two neighboring magnetic moments, where

$$\mathcal{H}_{\text{exch}} = -\sum_{i,j} J_{ij} \boldsymbol{\mu}_i \boldsymbol{\mu}_j, \quad (1.6)$$

where J_{ij} is the exchange coupling constant.

This interaction is best depicted by two isolated magnetization vectors. Under vanishing external perturbation, the two spins want to be magnetized in a parallel, or anti-parallel fashion based on the type of their exchange coupling. A ferromagnetic coupling ($J_{ij} > 0$), results in a parallel alignment, and an anti-ferromagnetic coupling ($J_{ij} < 0$) leads to an anti-parallel arrangement of spins, as shown in Fig. 1.4.

The continuous model of the micromagnetics and the presence of the exchange energy in the magnetic bodies are an essential aspect. Namely, the magnetization of ferromagnetic materials

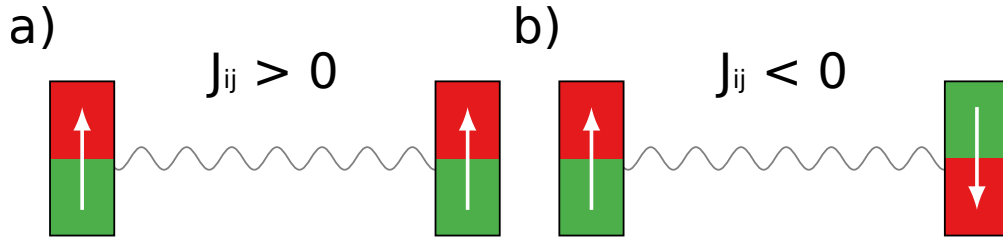


Figure 1.4: Schematics of the exchange coupling of two neighboring magnetic spins in a (a) ferromagnetic, (b) anti-ferromagnetic coupling. Original figure created by Claas Abert [5, 6, 4] and adapted with his permission.

described within the micromagnetic theory is always a smooth vector field. In other words: the ferromagnetic coupling makes the spins possible to minimize their energy only by being aligned parallel to each other. Thus, two neighboring spins cannot be aligned anti-parallel within the micromagnetic framework. This behavior exists in reality, and its called a Bloch Point, where the exchange energy becomes infinite, as, in terms of micromagnetics, it is described by a gradient of the magnetization with

$$E_{\text{exch}} = \int_{\Omega} A_{\text{ex}} (\nabla \mathbf{m})^2 d\mathbf{x}, \quad (1.7)$$

where A_{ex} is the exchange stiffness constant, and represents the micromagnetic equivalent to J_{ij} .

Nevertheless, the magnetization is not always homogeneous and smooth, as the exchange interaction requires it to be. Magnetic domains with different average magnetization directions can be formed. In this case the magnetization over-goes a continuous transition from the one direction to the other one. If the two magnetizations are oppositely oriented, this continuous transition is called a domain wall. The most common two domain walls are Bloch walls, and Néel walls. Note that the domain walls are mobile. They can be propagated by spin polarized current, or temperature gradients, or further external perturbations.

1.2.3 Dzyaloshinskii-Moriya Interaction energy

In contrast to the homogeneous and symmetric Heisenberg exchange, a different form of exchange interaction finds application in many devices and studies. The Dzyaloshinskii-Moriya Interaction (DMI) arises from the spin orbit coupling in certain magnetic materials, and it is an asymmetric exchange interaction. The most common form of DMI arises when a ferromagnetic thin film (FM) is placed on top of a heavy metal layer (HM) as platinum, or iridium. In this situation on the interface between FM and HM, the DMI arises and acts in contrary to the exchange interaction, and favors the formation of domains, chiral domain walls, and is crucial for the formation of Néel skyrmions. A schematic illustration of the spin-spin interaction is depicted in Fig. 1.5. The general, atomistic anti-symmetric energy is given by the Hamiltonian

$$\mathcal{H}_{\text{DMI}} = \sum_{i,j} D_{ij} \boldsymbol{\mu}_i \times \boldsymbol{\mu}_j, \quad (1.8)$$

where D_{ij} represents the symmetry of the system.

In the micromagnetics the energy is simplified to

$$E_{\text{DMI}} = \int_{\Omega} D_i [\mathbf{m} \cdot \nabla (\mathbf{e}_d \cdot \mathbf{m}) - (\nabla \cdot \mathbf{m})(\mathbf{e}_d \cdot \mathbf{m})] d\mathbf{x}, \quad (1.9)$$

where D_i is the scalar DMI constant, which gives the strength of the interaction.

1.2.4 Anisotropy energy

The magnetization can favor a parallel alignment to preferred axes, which are then referred to as easy-axis. The origin of the easy or hard axes can lie on the crystal lattice of the magnetic

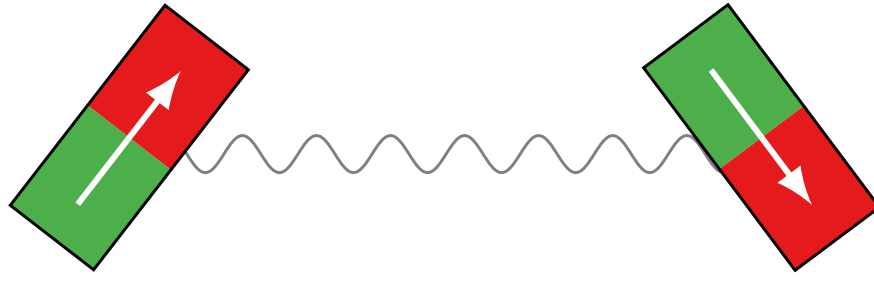


Figure 1.5: Schematic illustration of DMI, where formation of magnetic domains are favored. Original figure created by Claas Abert [5, 6, 4] and adapted with his permission.

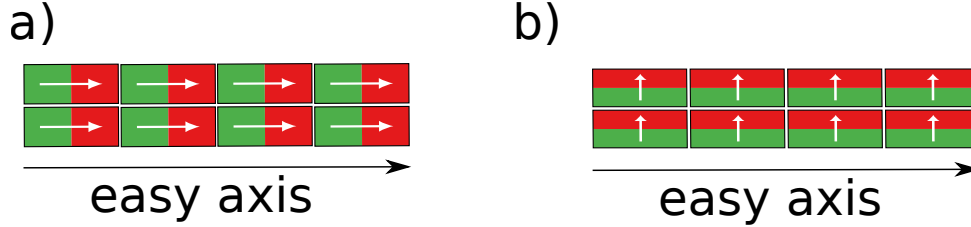


Figure 1.6: Magnetic spins being aligned with (a) the easy axis and approaching the ground state w.r.t. the anisotropy energy, and (b) the hard axis, leading to an excited state, as the magnet tends to be magnetized along the longer axis. Original figure created by Claas Abert [5, 6, 4] and adapted with his permission.

compound, or just raise from the shape of the magnet, see Fig. 1.6 Note that the anisotropy is a bidirectional energetic term: if the favored axis is x , the magnetization can either point towards $+x$ or $-x$. Both magnetizations are energetically equivalent. This can be seen in the quadratic dependency of the anisotropy energy on the magnetization, where

$$E_{\text{ani}} = - \int_{\Omega} K_{u1} (\mathbf{m} \cdot \mathbf{e}_d)^2 dx \quad (1.10)$$

gives the anisotropy energy for a uniaxial anisotropy, where K_{u1} is the anisotropy constant, and $\mathbf{e}_u$ is the easy-axis. Thus, the energy depends only on the angle between the magnetization and easy-axis.

However many materials do not have a preferred axis based on their crystallographic structure. In this case, the shape-anisotropy is the energetic contribution that decides in which direction the magnetization will (mainly) be oriented. It is very relevant for nano-magnetism, and especially in magnetic thin films, and nanowires. This energy originates from the demagnetization field of the magnet.

1.2.5 Demagnetization energy

One of the first images that everyone might recall learning about magnetism might be the response of ferrite powder when subjected to a magnetic field. The response basically depicts the magnetic field lines, where the magnetization of powder and the magnetic field align according to Eq. 1.5. The magnetic bodies generate a magnetic field based on the dipolar interactions between the magnetic moments, which are long-range interactions. The so called stray field of the magnet, acts then on the ferrite powder and it changes its location and orientation, as the acting interaction is a rather strong one.

The name demagnetization however, it comes from the fact a magnetic bodies favor demagnetized states, where the total dipole-dipole interaction is minimized. If one considers a single magnet bar as in Fig. 1.1, the magnetic field sourcing from the magnet expands over distance until it loses its strength. A neighboring magnet withing the interaction range will be demagnetized by

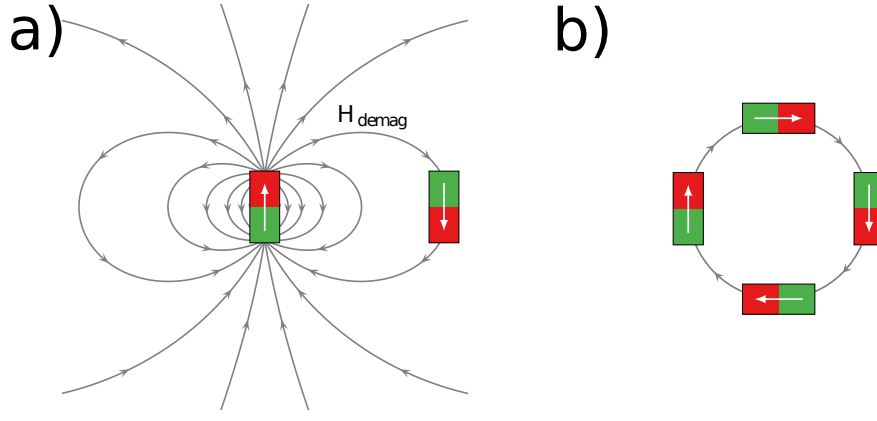


Figure 1.7: Schematic illustration of the demagnetization fields, where one magnet can demagnetize a second neighboring spin as in (a), or closed loops of magnetic fields are favored as in (b). Original figure created by Claas Abert [5, 6, 4] and adapted with his permission.

this magnetic field, see Fig. 1.7a). When one has four magnets arranged in a square manner however, as in Fig. 1.7b), the magnetic fields of the magnetic bars will join each other, minimize the density of stray-fields by building closed loops, subsequently reducing the total energy. Thus, they demagnetize each other, in order to achieve a ground state.

The demagnetizing energy is very similar to the Zeeman energy, and is given by

$$E_{\text{dem}} = -\frac{\mu_0}{2} \int_{\Omega} M_s \mathbf{m} \cdot \mathbf{H}_{\text{dem}} d\mathbf{x}, \quad (1.11)$$

where $\mathbf{H}_{\text{dem}}$ is the demagnetization field, which is one of the most costly fields in computational micromagnetics, due to its complexity, which usually can be reduced by using a combination convolution and fast Fourier transformations.

The demagnetization does also act on the magnetic body that it originates from, thus this contribution is referred to as self-demagnetizing energy, where the shape of the body is taken into account. This is nothing more than the shape-anisotropy that we have introduced earlier. Consider now a thin nano-magnet which is much longer than wider, and it has no uniaxial anisotropy axis. The demagnetization energy will favor a magnetization state where the magnet is magnetized along the longer axis, effectively making it to the easy-axis of the magnet, with an effective, (shape) anisotropy constant

$$K_{\text{shape}} = \frac{1}{2} \mu_0 M_s^2 \quad (1.12)$$

for infinitely long magnet.

1.2.6 Landau-Lifshitz-Gilbert Equation

In the field of micromagnetics, the computational methods to accurately calculate in a numerical fashion the above mentioned field and energies, such that many high-performance micromagnetic simulation software exists. Generally, a magnetic system contains many energetic contributions, where they all compete and want to achieve their own energetic minimum. Thus, the overlap of energetic states will form, such that one obtains a full energy landscape, where stable magnetization states are given by minima, while many high energy states with saddle-point character are present.

A common goal of all the simulation codes is to find the energetic minimum states of a magnetic and investigate, how the magnetization evolves with respect to time, when the body is subjected to external magnetic fields, spin-polarized currents or temperature. There exists one equation that, in a semi-classical manner, describes every single detail of this external perturbations within the theory of micromagnetics: the Landau-Lifshitz-Gilbert Equation (LLG), which says

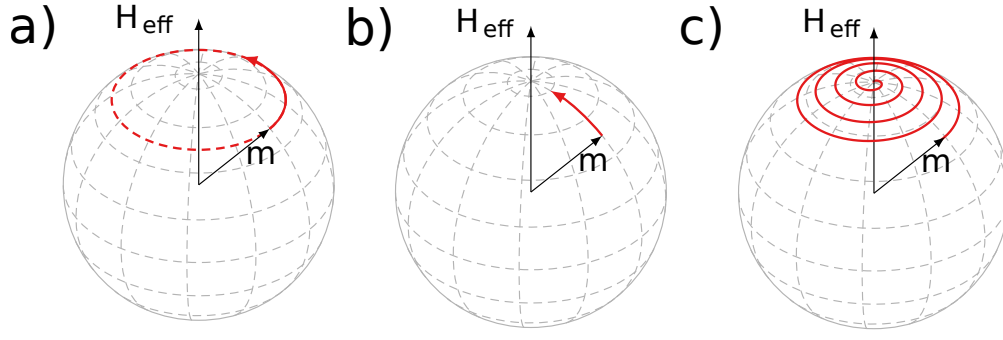


Figure 1.8: Illustration of the main mechanisms behind the LLG (Eq. (1.13)), where (a) describes the precessional evolution of the magnetization, (b) the direct relaxation due to damping term, and (c) overall evolution of the magnetization trying to align with the effective field of the system. Original figure created by Claas Abert [5, 6, 4] and adapted with his permission.

$$\partial_t \mathbf{m} = -\frac{\gamma}{1+\alpha^2} \mathbf{m} \times \mathbf{H}^{\text{eff}} - \frac{\alpha\gamma}{1+\alpha^2} \mathbf{m} \times (\mathbf{m} \times \mathbf{H}^{\text{eff}}), \quad (1.13)$$

where γ is the electron gyromagnetic ratio, α is the Gilbert damping constant, and $\mathbf{H}^{\text{eff}}$ is the effective field, that incorporates all the energetic contributions, where

$$\mathbf{H}^{\text{eff}} = -\frac{1}{\mu_0 M_s} \frac{\delta E}{\delta \mathbf{m}} \quad (1.14)$$

is the functional derivative of the energy with respect to $\mathbf{m}$.

While the first term on the right hand side of Eq. (1.13) describes the precession term, where the magnetization undergoes a precessional movement around the effective field, the second term gives the damping contribution in terms of a direct relaxation towards $\mathbf{H}^{\text{eff}}$, as illustrated in Fig. 1.8.

In easiest form it can be said that the LLG describes the temporal evolution of magnetization when a certain set of energetic contributions are considered and summarized in an effective field term, with which the magnetization tries to be aligned, in order to minimize its the total energy. A magnetic is considered to be relaxed, when the magnetization remains constant over time: hence, it represents an energetic minimum state in the total energy landscape.

1.2.7 Energy barriers and switching mechanisms

Nevertheless, the dynamical properties of the magnetic systems are also very important, where in particular cases it is of high interest to try to escape from the current energetic minimum, and reach another stable minimum configuration. In this case the two energetic states are separated by an energy barrier. The idea of energy barrier can be best understood, when one thinks of a big ball at the left base of a mountain. If one wishes to have the ball on the other side of the mountain, then one basically has to roll the ball over the mountains summit. This can be achieved only and only if one introduces a finite amount energy into the system.

Now consider two magnetic states, corresponding to two energetic minima, like the nano-island magnetized along the longer axis, as explained in Sec. 1.2.4. A magnetization along $\mathbf{e}_u$ and $-\mathbf{e}_u$ are both energetic minima due to the bidirectional uniaxial anisotropy, as shown schematically in Fig. 1.9. However, if one desires to switch from the one state to other one, than one requires to overcome a so called energy barrier: the amount of energy that one needs to introduce in the system, in order to trigger the switching mechanism and, change the direction of the magnetization. The involved switching mechanism will then provide information about how the magnetization evolves during the switching process. Most common forms of switching mechanisms are uniform rotation, or domain-wall propagation.

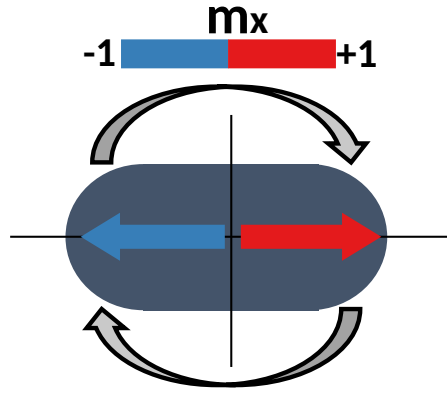


Figure 1.9: Schematic illustration of a magnetic island with two stable ground states, where $m_x = \pm 1$. If enough energy is introduced in the system to overcome the energy barrier, one can switch from one state to another.

1.3 String Method

There are different methods to calculate the energy barriers which have to be overcome in order to switch between magnetization states. One of them is, the nudged-elastic band method, where the basic idea is that one takes an elastic band and throws it well above the mountain. Then, one starts to pull at the ends of the band, until the band takes the form of the energetic landscape. A similar method is the string-method, which was later improved to a simplified and improved string method (SISM). With this method, instead of an elastic band, one uses a string, which is divided into a given number of states, called images. One starts with a random reversal path, which is usually just an interpolation between the initial and final magnetic state. With each iteration one repeats the following two steps, until the correct energy path from the landscape is reproduced.

- **Energy Minimization:** Energy of each image is minimized a small amount towards the true energy landscape, where the LLG is solved for a short time, such that the images do not fall into the initial and final state, but are minimized directly towards the energy path.
- **Interpolation:** The newly obtained images are interpolated to a new string in an equidistant manner with respect to an appropriate energy norm.

After the convergence, one obtains the energy path which is the energetically most favorable way to switch between the given magnetization states. This path is referred to as minimum energy path (MEP).

1.4 Spin Ice and emergent magnetic monopoles

If the magnetic monopoles truly exist or not, has been a debate for over a century now. And it still continues. At this place, it is good to say that the magnetic monopoles as postulated by Dirac, were not discovered yet.

But over the past two decades a specific class of magnetic materials has been investigated in much detail, both theoretically and experimentally: the antiferromagnets such as dysprosium ($\text{Dy}_2\text{O}_7\text{Ti}_2$) or holmium titanates ($\text{Ho}_2\text{O}_7\text{Ti}_2$), or as they are called today: Spin Ices[7, 8, 9, ?]. In order to understand this nomenclature, let us first have a look in the structure of these materials.

In spin ices, magnetic moments are arranged on the sites of a corner sharing tetrahedra. A tetrahedron with four magnetic *spins* builds a vertex. In each of these vertices, the magnetic

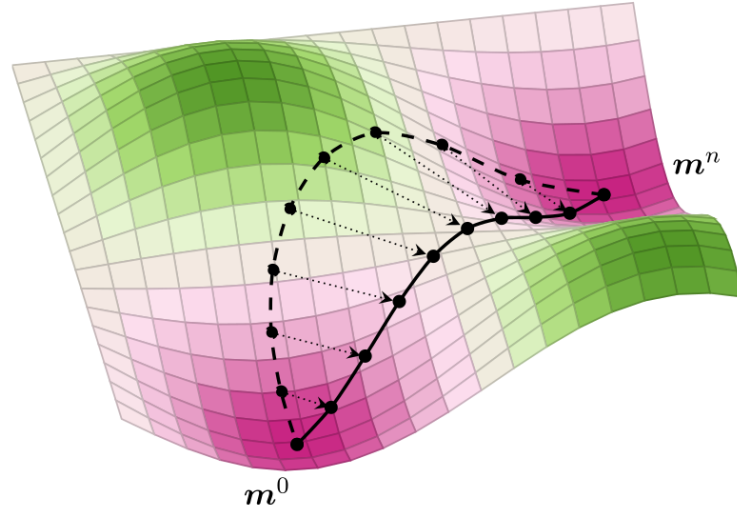


Figure 1.10: Simplified working principle of the string method where an initial string (dashed line), which transitions the magnetization from m^0 to m^n , lowers its energy for each image (dot) and reaches the minimum energy path (solid line), which is the energetically most favorable way to switch the magnetization. Figure taken from [4].

moments can point either to the tetrahedron center, or away from it. The energetic ground-state for each vertex is achieved, when the two of magnetic moments are pointing to the center, while the other two are pointing away from it. This is the so called *ice rule*. And the name originates from the ground-state configuration of the water ice. H_2O is maybe the most known chemical formula: two hydrogen atoms, and one oxygen atom. That is the chemical composition of water. When water reaches $T = 0^\circ\text{C}$, then it freezes, and it becomes ice. In the ice I_h crystal of the water ice, the oxygen atoms reside in the center of a tetrahedral unit cell. Each oxygen atom has two covalent bonds to the hydrogen atoms. Additionally, one hydrogen atom from the neighboring cells is bound over the polar hydrogen bindings to the central oxygen. In their energetic ground-state, two of the hydrogen atoms are residing inside the tetrahedron, close to the center occupied by the oxygen ion, and two are away from it.

Based on this analogy to the configuration of hydrogen atoms in the tetrahedral unit cell of the water ice crystal, the anti-ferromagnetic materials exhibiting a comparable condition for the ground-state were named spin ices. But the analogy does not end here! In the water ice, it is possible that one of the hydrogen atoms gets excited, and thus the ice rule is violated. Behind remains an ionic charge due to the dipolar interactions. Similarly, one can break the ice rule, and create an excited vertex. This can be achieved when one of the magnetic moments of the vertex reverses its magnetization direction, due to an external perturbation, e.g. external magnetic field, or temperature. Ryzhkin *et al* [10] were the first to propose and theoretically investigate the idea of magnetic charges, in analogy to the ionic charges of water. Castelnovo *et al* [11] have brought a much clearer picture with their dumbbell model, where each magnetic moment was separated to its south and north pole. Note the agglomeration of magnetic poles if a charge defect is created, as discussed above. In this case, where three magnetic moments point towards (away) and the remaining one away (towards) from the center of the tetrahedral unit-cell, then the accumulation of magnetic poles at the center of tetrahedron gives rise to an *emergent magnetic monopole*.

Multiple theoretical and experimental work have shown that these defects behave like magnetic monopoles [9, 8, 7, 12, 13], but are not true magnetic monopoles as postulated by Dirac [2]. However, his name is linked with these monopoles. If one starts from a vertex obeying the ice-rule, and thus, in the energetic ground-state, and starts to reverse the magnetization of one atom, then one basically separates two emergent magnetic monopoles. Now, two neighboring monopoles will have each a magnetic monopole due to the created charge defect. In one of the vertices, one can

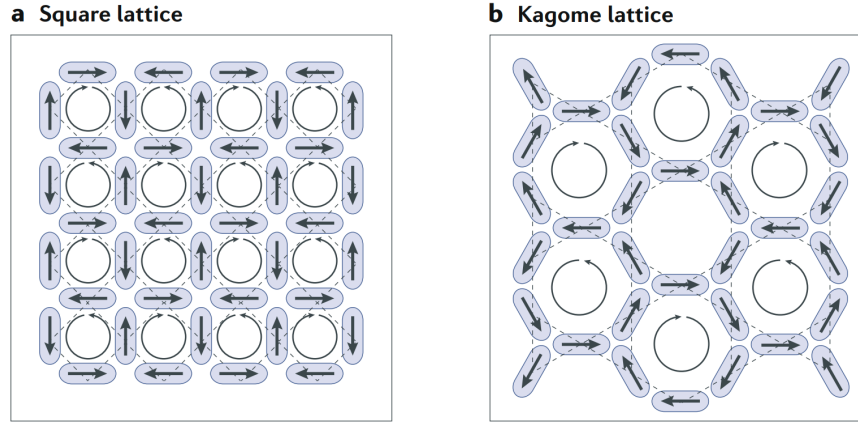


Figure 1.11: The two most common lattices of artificial spin ices (a) square, and (b) kagome ice. The arrows on the stadium shaped islands represent the macrospin and thus the average magnetization of the nano-islands, where as the circular arrows illustrate that the magnetic fields build closed loops inside the lattice, achieving long-range order. Figure taken from [15].

further propagate this quasi-particle. The trace of this propagation is called a Dirac String, and it should be an unnoticeable, energy-less and thus, tension-free entity. If these properties hold, than the magnetic monopoles separated by the Dirac strings, are effectively unbound.

Note that the ground-state in spin ices is highly degenerate. In each vertex of four spins, there exist 16 possible configurations, six of which obey the ice-rule, and thus represent the ground-state in each vertex. In the spin ice crystal with many vertices, the ground state is achieved at cryogenic temperatures where the magnetic configuration of the system consists of alternating ground-state configurations.

1.5 Artificial Spin Ices

The atomic sizes and the required cryogenic temperatures made it very difficult for the researchers to study the behavior of magnetic monopoles to a great extent. In order to understand the physics of magnetic monopoles, artificial spin ices have been designed [14, 15, 16, 17, 18, 19, 20, 21, 22, 23]. In this case, the magnetic moments of the spin ices are replaced by the macroscopic magnetizations of elongated, stadium-shaped, ultra-thin magnetic nanoislands. These islands are always magnetized along their longer axis due to the shape anisotropy. Typical nano-island sizes range from one to several hundreds of nanometers, while thicknesses are currently kept under 10 nm.

The nano-islands are arranged on different lattices, where the most important ones are the square and the kagome lattice, see Fig. 1.11-a and b. While the former is clear based on its name: the islands are arranged on square lattice. Hence, each vertex consists of four magnetic islands arranged rather like a cross. The latter is obtained when the nano-islands are arranged on a hexagonal lattice, where each vertex consists of only three islands.

1.5.1 Kagome Artificial Spin Ice

If the magnetic nano-islands are arranged on an hexagonal lattice, than the ASI lattice is called a kagome lattice [24, 25], as shown in Fig. 1.11-b. Each vertex consists of three magnetic islands, thus one only has a pseudo ice rule, and always an accumulation of magnetic charges. However, based on the dipolar interactions, it is theoretically possible to have a long range ordered well defined ground state, where the magnetic fields build closed loops within the hexagons. The experimental demonstration was realized only recently, as reported by Hofhuis *et al* in a recent

preprint [26], where a bridged system was used in order to reduce the energy barriers in the system.

1.5.2 Square Artificial Spin Ice

The first and also most common type of ASI is the square ice. Wang *et al* [14] have fabricated the first lattice by electron beam lithography. Many experimental and theoretical works have followed, where the dynamics of magnetic monopoles [27, 20, 21, 28, 29, 30, 31, 32, 33], but also the interaction of spin-waves in these materials have been investigated [34, 35].

The spin ice mimicking system does not behave perfectly like its atomic counter-part. In the square ice vertices, the degeneracy of the ground-state is lost! It was shown by Vedmedenko [36] that the magnetic charges in square ice are bound and the Dirac string carries energy: hence, its tension does not vanish. This limits the mobility of the emergent magnetic monopoles.

Chapter 2

Dependence of energy barrier reduction on collective excitations in square artificial spin ice: A comprehensive comparison of simulation techniques

The following manuscript by Sabri Koraltan, Matteo Pancaldi, Naëmi Leo, Claas Abert, Christoph Vogler, Kevin Hofhuis, Florian Slanovc, Florian Bruckner, Paul Heistracher, Matteo Menniti, Paolo Vavassori, and Dieter Suess was published in Phys. Rev. B 102, 064410 (2020).

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Personal Contributions

The personal contributions include performing the micromagnetic simulations, and post-processing of simulation results and data, preparing all figures for manuscript and writing the initial manuscript.

Bibliography reference: [37]

Dependence of energy barrier reduction on collective excitations in square artificial spin ice: A comprehensive comparison of simulation techniques

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We perform micromagnetic simulations to study the switching barriers in square artificial spin ice systems consisting of elongated single domain magnetic islands arranged on a square lattice. By considering a double vertex composed of one central island and six nearest neighbor islands, we calculate the energy barriers between two types of double vertices by applying the simplified and improved string method. We investigate by means of micromagnetic simulations the consequences of the neighboring islands, the inhomogeneities in the magnetization of the islands and the reversal mechanisms on the energy barrier by comparing three different approaches with increasing complexity. The micromagnetic models, where the string method is applied, are compared to a method commonly in use, the mean barrier approximation. Our investigations indicate that a proper micromagnetic modeling of the switching process leads to significantly lower energy barriers, by up to 35% compared to the mean-barrier approximation, so decreasing the expected average life time up to seven orders of magnitude. Hereby, we investigate the influence of parallel switching channels and the conceptual approach of using a mean-barrier to calculate the corresponding rates.

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I. INTRODUCTION

Artificial spin ice (ASI) systems are lithographically patterned lattices of elongated single domain magnetic islands [1–4]. In two-dimensional ASI systems, the magnetic islands are arranged in vertices and build a frustrated lattice due to the competing magnetostatic interactions among the islands [1,5–8]. A possible ASI system, where four islands build a vertex and are arranged on a square lattice, is called square artificial spin ice (sASI) [1,2]. Such a lattice is illustrated in Fig. 1(a). In the absence of external magnetic fields, the islands are magnetized along their long axis due to shape anisotropy, which limits the possible configurations in a vertex to $2^4 = 16$ macroscopic configurations.

Figure 1(b) shows the four types of vertex configurations in a sASI, where the energy of each vertex increases with its assigned number.

The ground state is represented by Type I vertices, since in a sASI lattice with alternating Type I vertices the stray fields between four magnetic islands arranged in a square build closed loops [9], minimizing the total energy of the system. Even though both Type I and Type II vertices obey the ice rule [2], where two magnetizations are pointing to the center

and two away from it, the latter have an increased energy due to unequal distance of the four elements forming a vertex. Type III vertices show even higher energies, where three magnetizations are pointing towards the center and one away or vice versa. This vertex configuration displays a net magnetic charge $Q_m = \pm 2q_m$, where q_m is the magnetic charge residing at the end of each magnetized island [2,4]. Type IV vertices contain magnetic islands with the magnetization pointing all to the center or away from it. This type has the highest energy level, and thus, is the most excited configuration, showing a net magnetic charge $Q_m = \pm 4q_m$.

Both experiments [10] and simulations [11,12] have been performed to analyze the thermal annealing and domain dynamics in these structures. Furthermore, it has been experimentally observed that the ground state can be achieved after thermal relaxation, where the lattice has been subject to temperatures T satisfying $T_B < T < T_{\text{Curie}}$, where T_B denotes the blocking temperature above which the magnetizations of the islands are fluctuating on the time scale of the measurement [7,13–15], and T_{Curie} denotes the Curie temperature. In this case, the islands start to switch their overall magnetic orientation along the long axis due to thermal activation, enabling the system to evolve towards the energetically favored ground state with Type I vertices. The temporal evolution of the thermally activated relaxation can be described by standard (Bortz-Kalos-Lebowitz algorithm [16]) kinetic Monte Carlo

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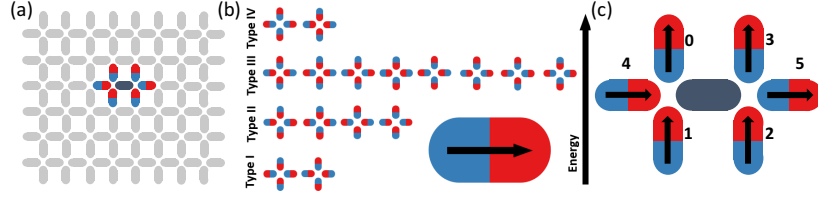


FIG. 1. Square artificial spin ice lattice. (a) Schematic illustration of a sASI lattice with a highlighted in color double vertex. (b) Vertex configurations in a sASI. Possible arrangements of the magnetizations of the islands in a square vertex, where the black arrow of the large island indicates the direction of the magnetization. All configurations that are assigned to the same vertex type have equal energies. (c) Schematic illustration of the double vertex from (a) with the central island (dark), being the island of interest in the energy barrier calculations and the neighboring islands enumerated with the island identification number j to calculate the configuration number, as given later by Eq. (2).

simulations [15,17,18] that require the energy barriers of the various switching processes as an input. Thus the ability to accurately determine the energy barriers to be overcome in order to switch the magnetization of the magnetic islands are of key importance.

By using the mean-barrier approximation to calculate the switching barriers, the experimentally observed relaxation mechanisms, and thus the dynamics and the ordering of the system, can only be artificially reproduced [15,18–21]. Namely, in order to reproduce the relaxation timescale observed experimentally, the energy barriers need to be artificially reduced. Such a reduction is usually ascribed to extrinsic factors like fabrication defects, reduction of the Curie temperature T_{Curie} [22] or saturation magnetization M_s [7].

In this work, we study the dependence of the energy barriers on collective excitations in a square lattice by performing micromagnetic simulations.

We apply the simplified and improved string method (SISM) [23] to investigate if the commonly reported barrier reduction is ascribable, and to what extent, to intrinsic fundamental physics related to the thermally induced reversal rather than a mere effect of extrinsic factors by comparing three different models.

We find that these reductions can be ascribed to the preferential switching direction of the magnetizations in a magneto-static environment, as well as nonuniform contributions to the moment reversal.

This paper is organized as follows. The simplified and improved string method is presented and the application on a double vertex is described in Sec. II. The considered geometries and sASI material parameters are introduced in Sec. III. In the Secs. III A–III C we describe the three investigated models. We discuss our findings and the results of our simulations in Sec. IV, where we analyze in detail the differences in the reversal mechanisms, rotation directions, energy barriers, corresponding switching rates and different material parameters. We summarize our conclusions in Sec. V.

II. SIMPLIFIED AND IMPROVED STRING METHOD

In order to analyze the dependence of collective excitations on the energy barriers, we use the SISM [23] to calculate the energy barrier between two magnetic states.

This method consists of three steps. In the first step, we create an initial path for the switching event. For this purpose,

we choose a coherent rotation with the uniform magnetization $\mathbf{m}_{\text{ini}}$ as our initial state. The final state of the initial path is represented by $\mathbf{m}_{\text{fin}}$. This path is discretized by 21 distinct magnetization configurations, which sample the continuous transition path in an equidistant fashion with respect to an appropriate norm [23]. In a second step, each of these 21 magnetization configurations is evolved a certain amount towards its nearest energetic minimum, and in a final step, the 21 magnetization states are rearranged along the path in order to restore the equidistant discretization of the path. The last two steps are repeated until we obtain the minimum energy path (MEP). This final path represents the lowest energy path and equally, the most probable and favorable way to switch between initial and final magnetization states with respect to the given initial path. Figure 2 is an exemplary illustration for the evolution of the energy paths to obtain the MEP. In agreement with transition-state theory [24,25] the energy barrier to switch the magnetization from $\mathbf{m}^{\text{ini}}$ to $\mathbf{m}^{\text{fin}}$ is obtained by

$$\Delta E = E^{\text{saddle}} - E^{\text{ini}}, \quad (1)$$

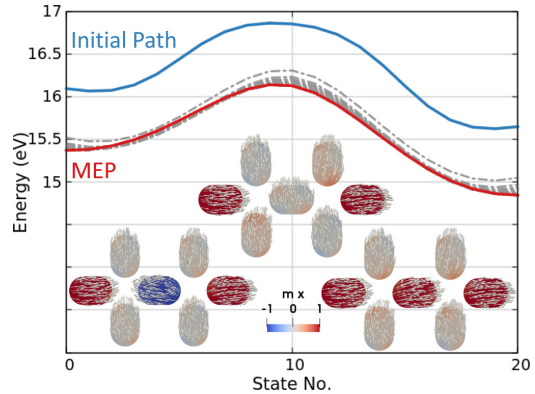


FIG. 2. Evolution of the energy paths during the SISM. For the configuration $c = 63$ illustrated in Fig. 1(c), the initial path (blue), represented by a coherent rotation, evolves towards the MEP (red) via the intermediate energy paths (gray). Inset figures represent the magnetization states for State No. = 0 (left), State No. = 10 (center) and State No. = 20 (right) of the MEP.

TABLE I. Summary of the considered models. The environment describes the magnetization of the NN islands and the central moment the magnetization of the central island during the switching event.

MODEL	MODEL 1	MODEL 2	MODEL 3
Description	Mean-barrier	Uniform environment	Full micromagnetic model
Environment	Uniform	Uniform	Dynamically relaxed
Central island	Uniform	Dynamically relaxed	Dynamically relaxed
Reversal mechanism	Coherent rotation	Minimum energy path	Minimum energy path

where E^{saddle} is the energy corresponding to the saddle point of the MEP and E^{ini} is the energy of the initial state of the MEP.

The considered energy contributions for the micromagnetic modeling are described in Appendix A.

III. MODELING

In this paper, we study the energy barriers between different vertex types of sASI. In particular, we show the influence of the nearest neighbor (NN) islands on the energy barrier by computing the MEPs with different approaches. The considered models are summarized in Table I. To obtain a direct comparison between the models, we use `magnum.fe` [26], a finite element method based micromagnetic simulation code, to calculate the minimum energy path by applying the SISM. We generate all finite element meshes using `Gmsh` [27].

We consider magnetic islands similar to those used to perform kinetic Monte Carlo simulations in literature, for which the energy barriers needed to be reduced with respect to those arising using the geometrical and physical properties of the nanoelements to reproduce the experimentally observed relaxation. Our islands have a length $L = 150$ nm, a width $W = 100$ nm, an edge-to-edge gap $g = 90$ nm, and a thickness $t = 3$ nm.

Furthermore, we use material parameters similar to bulk permalloy at $T = 300$ K, with saturation magnetization $M_s = 790$ kA/m, exchange stiffness constant $A = 13$ pJ/m and vanishing uniaxial anisotropy constant $K = 0$.

In the following, we study the energy barrier for the switching event of the magnetization of the central island in a double vertex. Figure 1(c) illustrates such a double vertex with the central island as the island of interest.

In order to effectively label the 64 possible magnetization configurations of the NN islands, we introduce the integer parameter c defined by

$$c = \sum_{j=0}^5 b_j 2^j. \quad (2)$$

The factor b_j is obtained by

$$b_j = \begin{cases} 0, & \text{if } m_k = -1 \\ 1, & \text{if } m_k = +1 \end{cases}, \quad (3)$$

where m_k is positive, if the magnetic moment points to the right (up) for horizontal (vertical) islands, as shown in Fig. 1(c). The index j denotes the NN island identification number as illustrated in Fig. 1(c). The arrows in Fig. 1(c) show the magnetization of the neighboring islands in the configuration $c = 63$.

A. Model 1. Mean-barrier approximation

For the first model, we consider the mean-barrier approximation when the NN islands are present and uniformly magnetized. Motivated by the symmetry properties of the strayfield interaction and Zeeman energies, the mean barrier [15,18–21] of the switching is described by

$$\Delta E^{\text{mean}} = \Delta E^{\text{isol}} + \frac{1}{2}(E^{\text{fin}} - E^{\text{ini}}), \quad (4)$$

where ΔE^{isol} is the energy barrier to switch the magnetization of one isolated nanostructure, E^{fin} the energy of the final state corresponding to the switched double vertex and E^{ini} the initial energy of the double vertex. A derivation of Eq. (4) is given in Appendix B. The magnetizations of the neighbors as well as of the central island are kept uniform during the switching event.

Since only the initial and final state are involved in Eq. (4), we do not need to apply the SISM in this model. In order to keep each island uniformly magnetized and the reversal mechanism a coherent rotation, we consider the energy barrier for one single nanostructure ΔE^{isol} , which is obtained from a coherent rotation of the magnetization. Thus, this yields $\Delta E^{\text{isol}} = 1.37$ eV.

B. Model 2. Uniform environment

As a step further, we apply the SISM in this model in such a fashion that the magnetic region Ω_m covers only the central island, where Ω_m denotes the magnetic region, where the SISM iteratively minimizes the energy to find the MEP, taking into account all energy contributions of the system. We continue keeping the NN islands uniformly magnetized.

Under these assumptions, the total energy of the system also includes the strayfield interactions between the central island and the NN islands, as in the previous model. The interaction energies among the NN islands are constant during the switching process due to their uniform nature. Therefore, the difference in their saddle point energy and initial energy vanishes. Note that the reversal mechanism is not a perfect coherent rotation anymore. The deviation from the rotation of a perfect uniformly magnetized configuration is associated with the application of the SISM, which allows the relaxation of the magnetization, i.e., deviation from the perfectly uniform configuration, for each step of the MEP taking into account both the shape of the island (shape anisotropy) and the interactions fields originating from the NN islands.

C. Model 3. Full micromagnetic model

For the last approach, we include all the seven islands in the magnetic region Ω_m , where the string method is applied. Thus

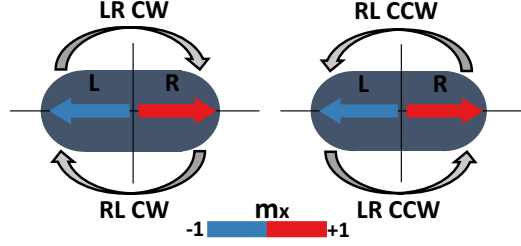


FIG. 3. Rotation directions for the switching event on the central island. We refer to a switching event with $\mathbf{m}^{\text{ini}} = (-1, 0, 0)$, $\mathbf{m}^{\text{saddle}}_{\text{CCW}} = (0, -1, 0)$, and $\mathbf{m}^{\text{fin}} = (1, 0, 0)$ as a left to right counterclockwise rotation (LR CCW). If the saddle point configuration is changed to $\mathbf{m}^{\text{saddle}}_{\text{CW}} = (0, 1, 0)$, this becomes a left to right clockwise (LR CW) rotation. Likewise, one can define a right to left counterclockwise rotation (RL CCW) and a right to left clockwise rotation (RL CW).

both the NN islands and the central island are dynamically relaxed. The interactions among the NN islands change at each state of the minimum energy path, so they cannot be neglected in the barrier calculation.

We include indirectly the effect of temperature in our system via reduced M_s and A [28–30], allowing for fluctuations to arise from the decreased coupling energy between the spins, which could even form magnetic domains. In Model 3, magnetization inhomogeneities are accounted for not only in the central island but also in the NN islands. Note that Model 2 also takes into account the fluctuations, however only in the magnetization of the central island, since the NN islands are kept uniform.

IV. RESULTS AND DISCUSSION

A. Parallel switching channels

One of the main consequences introduced by Model 2 and Model 3 and that is not predicted by Model 1, is the dependence of the energy barrier on the direction of the rotation of the central island's magnetization. For the sake of simplicity, we give to each rotation direction an acronym, as illustrated in Fig. 3.

Thereby, the switching of the central island's magnetization can occur via two parallel channels, clockwise (CW) and counterclockwise (CCW), which might have a different switching rate depending on the NN configuration. The resulting rate of switching from the Arrhenius law [25,31] is then defined as

$$f = \frac{1}{\tau} = \frac{1}{\tau_0} \cdot \left(e^{-\frac{\Delta E_{\text{CW}}}{k_B T}} + e^{-\frac{\Delta E_{\text{CCW}}}{k_B T}} \right), \quad (5)$$

with τ being the estimated lifetime of the state, τ_0 the attempt period, $k_B = 8.62 \times 10^{-5} \text{ eV/K}$ the Boltzmann constant and T is the temperature. Note that τ_0 might be different for each switching channel. Here, we assume that both channels have the same attempt frequency.

When a mean barrier approximation is used, the switching rate is given as

$$f^{\text{mean}} = \frac{2}{\tau_0} \cdot e^{-\frac{\Delta E^{\text{mean}}}{k_B T}}, \quad (6)$$

where ΔE^{mean} is the mean barrier given by Eq. (4), which is the average barrier between the CW and CCW barriers. However, in the cases where the rates via the CW and CCW channels are very different, only the fastest channel will dominate. Thereby, the mean-barrier approximation leads to significantly deviating rates underestimating the actual switching rate. A detailed discussion for these deviations is given in Appendix B. Under these assumptions, Eq. (5) can be simplified including only the exponential term corresponding to the faster channel. Hence, the estimated lifetime for a single channel is defined by

$$\tau = \tau_0 \cdot e^{\frac{\Delta E}{k_B T}}. \quad (7)$$

To better visualize the physical origin of this splitting, consider the large arrows in Fig. 4 (Model 2) in the snapshots for the saddle point configurations for both CW and CCW directions. For the former, one can see that approaching the saddle point configuration, the magnetization vectors of the central island and vertical NN islands are showing on average in the same direction, leading to an overall poles-attraction configuration. In contrast, during a CCW switching process, the saddle point configuration of the central island points to the opposite direction. Hereby the conflicting magnetic fields give rise to pole repulsion, which yields in an additional energetic level that has to be overcome.

B. Energy barriers

In order to quantify the effect of parallel switching channels, we first consider a single configuration $c = 63$, as illustrated in Fig. 1(c), and analyze the energy barriers for different rotation directions. In this case, a left to right (LR) switching of the magnetization of the central island changes the state of the double vertex from a Type III–Type III double vertex to an energetically favorable Type II–Type II. Figure 5 illustrates the MEPs for both CW and CCW rotations, where the initial energies have been shifted to zero (see Table II). The results indicate that the energy barriers for a CW rotation are lower than for a CCW rotation. As the name suggests, Model 1 is an approximation of the mean barrier (the detailed derivation is given in Appendix B) for the CW and CCW rotations, and thus, the energy path from Fig. 5 is obtained as the average energy path for these directions using uniformly magnetized NN islands and a coherent rotation. Figure 4 illustrates the initial, saddle point and final magnetization configurations for all models. According to Eq. (4), the energy barrier for Model 1 (orange) depends only on energies of the initial and final states and does not distinguish between the rotation directions. Models 2 and 3 (blue and red), however, include the spatial distribution and symmetry of the neighboring islands in the minimization process during the string method, thus breaking this degeneracy.

Namely, when considering the demagnetization field interactions from the neighboring islands, the CW rotation is energetically favorable over the CCW rotation since its

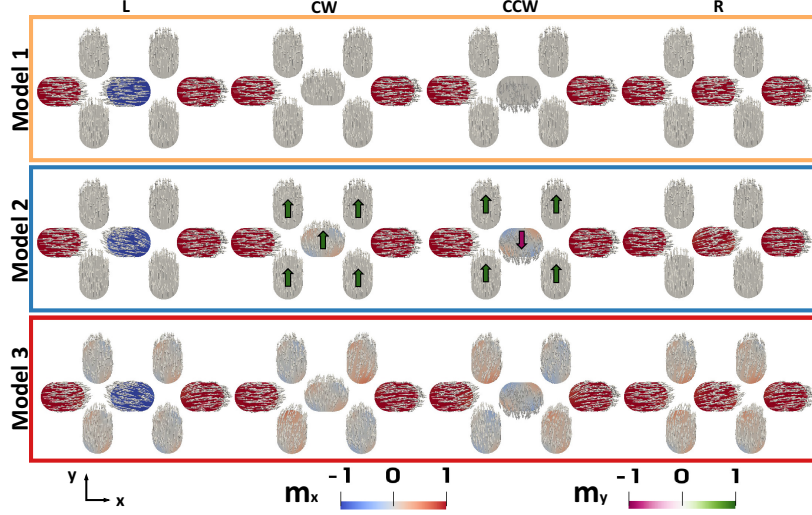


FIG. 4. Snapshots of the magnetization states during the reversal process for configuration $c = 63$. Illustration of the x component of the magnetizations using Model 1 (orange), Model 2 (blue) and Model 3 (red) for the initial state L , saddle point states for both CW and CCW rotation directions and final state R . The arrows added in the snapshots (Model 2) indicate average magnetization where the colorbar m_y indicates the average y component of the magnetization providing a visual support to understand the magnetostatic interaction between the central island and its NNs.

intermediate magnetization configurations point in positive y direction which aligns with the nonzero y component of the stray field in the configuration $c = 63$. Table II shows the values for the energy barriers of the configuration $c = 63$ and of the isolated island.

In the following, we focus on the CW rotations for the configuration $c = 63$. All models show a significant reduction of the energy barrier compared to the isolated nanostructure mainly due to the introduction of the stray-field interaction with the NN islands.

In contrast to Model 1, Model 2 introduces non-uniformities in the magnetization of the central island. The reversal mechanism obtained with the SISM, which is not a perfect coherent rotation anymore, is associated to a further reduction of the energy barrier. This discrepancy can be best seen in Fig. 4. Comparing the snapshots at the saddle point configurations, one can see that the Model 2 and Model 3 show deviations from the uniform configuration at the saddle point depicted exemplarily in Model 1 and a nonvanishing x

component of the magnetization develops at the saddle point (in Model 3 also in the vertical NN islands), at variance with the coherent rotation assumption made in Model 1.

Compared to the widely used mean-barrier approximation (Model 1), Model 2 reduces the lowest energy barrier (LR CW) about 28%.

The highest energy barrier reduction is observed for Model 3, which is the full micromagnetic model. In addition to Model 2, the neighbors are included in the magnetic region of the SISM, where the energy is minimized. Thus their magnetizations are dynamically relaxed and may change for each intermediate state in the transition path.

In this case, the stray-field interactions among the neighboring islands are not constant and the difference of their saddle point and initial states energies does not vanish. The lowest energy barrier for this model is reduced by 33% compared to Model 1 for the configuration $c = 63$.

Furthermore, only for Model 3 the magnetic system is in a true energetic equilibrium, since all islands are added in the

TABLE II. Energy barriers and estimated switching times obtained by applying the Models 1–3. The numerical results of the switching barriers ΔE and average lifetimes τ to switch the central island's magnetization from left to right (LR) for both CW and CCW rotation directions, as visualized in Fig. 3. The estimated switching times were calculated with Eq. (7) using $\tau_0 = 10^{-10}$ s and $T = 300$ K.

Direction MODEL No.	Isolated –	Configuration $c = 63$				
		CCW/CW 1	LR CCW		LR CW	
			2	3	2	3
ΔE (eV)	1.37	1.16	1.51	1.37	0.83	0.77
τ (s)	9.8×10^{12}	2.4×10^9	2.9×10^{15}	1.2×10^{13}	7.2×10^3	8.4×10^2

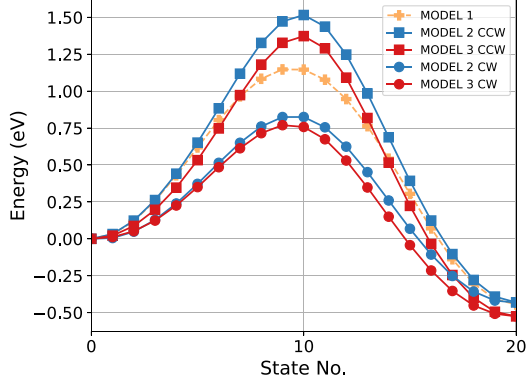


FIG. 5. Minimum energy paths for the switching of the magnetization of the central island. The MEPs for $c = 63$ considering both CCW (squares) and CW (circles) rotations, where the initial energies are shifted to zero. Here, the initial double vertex contains two Type III vertices and one obtains two Type II vertices after switching the magnetization of the central island from $\mathbf{m}^{\text{ini}} = (-1, 0, 0)$ to $\mathbf{m}^{\text{fin}} = (1, 0, 0)$, represented by State No. = 0 and State No. = 20, respectively.

magnetic region of the string method and their energies have been minimized.

Animations of the switching process for the configuration $c = 63$ using Models 2 and 3 can be found in the Supplemental Material [32].

Although the differences between the Models 2 and 3 may seem rather small, they have a major impact on the average lifetime given by Eq. (7). Since the fastest channel dominates the switching, we compare the estimated switching times for LR CW rotation directions for the configuration $c = 63$.

Here we use the attempt period $\tau_0 = 10^{-10}$ s and $T = 300$ K.

The value of the attempt period is only exemplary to show the role of the energy barrier reduction regarding the switching times. The values for τ are given in Table II.

One can see that the estimated lifetime (LR CW) calculated using Model 2 is approximately up to six orders of magnitude lower compared to Model 1, and one order of magnitude higher with respect to Model 3.

The full micromagnetic model yields a reduction of the average lifetime by about seven orders of magnitude with respect to the average barrier Model 1, often used in ASIs literature.

To show that Model 3 predicts (in nearly all cases) the lowest energy barriers, we calculate the switching barriers for all possible configurations c in a double vertex given by Eq. (2).

Figure 6 shows a direct comparison between the models for both LR CCW and LR CW rotations.

To obtain the barriers with respect to the final state (RL) one has to rotate the islands 180° around both the vertical and horizontal axis, since the rotation direction also changes. In principle, there exists an equivalent CCW barrier for each CW barrier, but with a different configuration number. If we

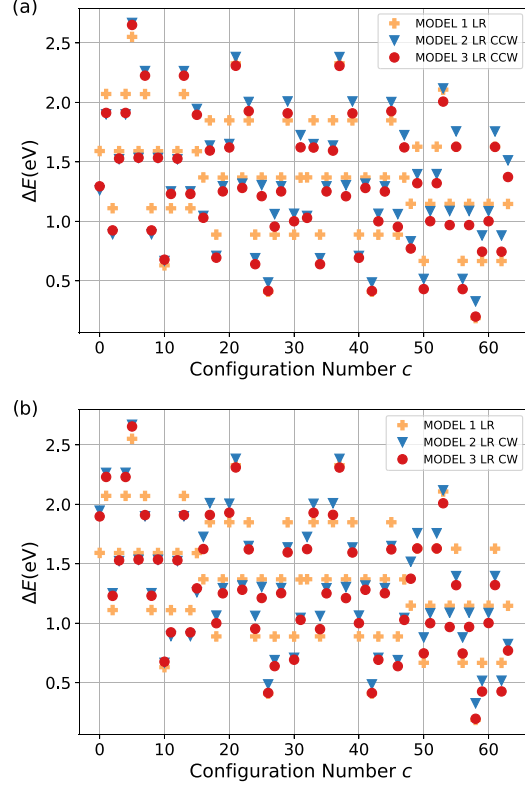


FIG. 6. Direct comparison between the Models. Energy barriers for the switching of the magnetization of the central island using all possible configurations of the NN islands in a double vertex illustrated in Fig. 1(c) via LR CCW (a) and LR CW (b) rotation directions.

consider $c = 63$ as an example, $c = 0$ LR CCW has the same barrier as $c = 63$ RL CCW.

Figure 6 shows that for all configurations at least one of the possible energy barriers is the lowest for Model 3.

Since Model 3 describes the evolution of the system through a saddle point configuration, where each NN island is dynamically relaxed to its energetic equilibrium, it yields the most realistic barrier, whereby the initial and final states are true energy minima. Note that Model 1, as well as Model 2, can result, in some cases, in lower energy barriers compared to Model 3. As it can be seen in Fig. 4, the magnetization of the NN islands are kept uniform for the first two models. Hereby, the energies of the initial and final states are not properly minimized, and thus the systems have not reached their true energetic equilibrium. This points out further limitations of these models. Consider that nonuniformities in the central island's magnetization modeled by Model 2 reduces the initial and final state energies E^{ini} and E^{fin} , respectively as the system approaches its energetic equilibrium by a certain amount $\Delta_{i,2}$ with respect to the uniform states utilized in Model 1. The

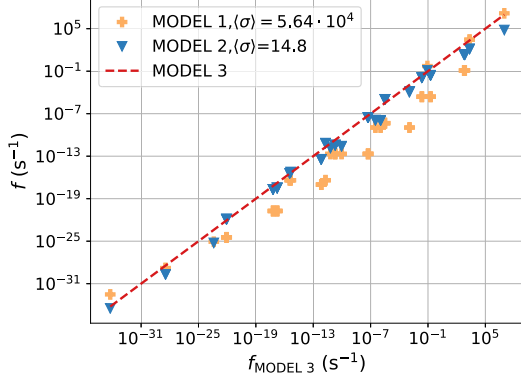


FIG. 7. Switching rates for Models 1–3. Log-log plot of the switching rates given by Eq. (5) for each model. The x axis shows the switching rates for the possible 64 configurations using Model 3, whereas the y axis shows the switching rates for the Models 1 (orange, crosses) and 2 (blue, triangles). The mean error $\langle\sigma\rangle_{\text{Model}_i}$, where $i = 1$ or $i = 2$, given by Eq. (8), is shown in the legend. The values of Model 3 are plotted as a reference line.

saddle point energy E^{saddle} is assumed to be reduced by $\Delta_{s,2}$. If $\Delta_{s,2} > \Delta_{i,2}$ the switching barrier calculated with Model 2 is lower (for a given rotation direction). However, only considering Model 3 the micromagnetic simulations can be performed to their full extent to reach the overall equilibrium states. As a consequence, the reduction of the initial and final state energies, $\Delta_{i,3}$ for the full micromagnetic model is always greater than $\Delta_{i,2}$. Hence, an equal reduction of the saddle point energies for both Models 2 and 3, and thus, $\Delta_{s,2} = \Delta_{s,3}$ can lead to a lower energy barrier for Model 2, since $\Delta_{i,3} > \Delta_{i,2}$.

C. Switching rates

Using our results from Sec. III B, the corresponding switching rates are calculated with Eqs. (5) and (6), where the correlation between the models is illustrated in Fig. 7.

The mean error

$$\langle\sigma\rangle_{\text{Model}_i} = \frac{1}{64} \sum_c \frac{f_{\text{Model}_3,c}}{f_{\text{Model}_i,c}}, \quad (8)$$

where the value $\sum_c \frac{f_{\text{Model}_3,c}}{f_{\text{Model}_i,c}}$ sums over the relative deviation factors for all configurations c , and gives an estimate of the average deviation from the full micromagnetic model. The higher the deviation from. $\langle\sigma\rangle = 1$, the more inaccurate are the calculated energy barriers compared to the full micromagnetic model, Model 3.

Even though Model 1 might be a mathematically valid approximation of the mean-barrier based on the derivation given in Appendix B, the concept of an average barrier does not necessarily imply a physically justifiable input for the kinetic Monte Carlo simulations, as the mean-barrier approximation approaches its limitations, for significantly different energy barriers for CCW and CW channels. As shown by the mean error $\langle\sigma\rangle_{\text{Model}_1} = 5.64 \times 10^4$, the mean switching rate is

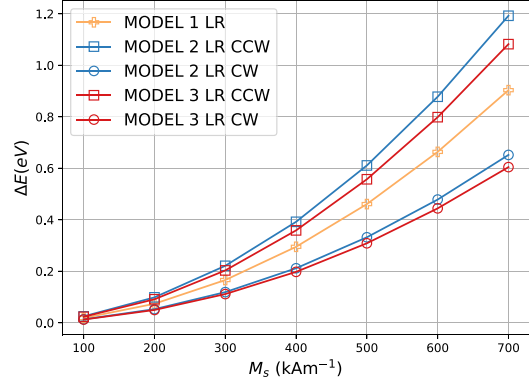


FIG. 8. Dependence of energy barriers on sASI material parameters for configuration $c = 63$. Calculated switching barriers as a function of the saturation magnetization M_s using Model 1 (orange, crosses), Model 2 (blue), and Model 3 (red) for both LR CCW (squares) and LR CW (circles) rotation directions.

underestimated by over four orders of magnitudes compared to the full micromagnetic model. With the introduction of parallel switching channels and the nonuniformities in the central island's magnetization in Model 2, the mean switching rate already improves significantly compared to Model 1, and the mean error $\langle\sigma\rangle_{\text{Model}_2} = 15.4$ decreases three orders of magnitude. Nevertheless, compared to the full micromagnetic model, the switching rates are underestimated in general and even overestimated for the cases this model reaches its limitations, as explained in Sec. IV B.

Considering once more the configuration $c = 63$, we calculate the exact average barrier using the values obtained from Model 3, hence, the average barrier is $\Delta E^{\text{avg}} = 1/2(0.77 + 1.37) \text{ eV} = 1.07 \text{ eV}$. For the switching rates of the configuration $c = 63$ calculated with Eqs. (5) and (6) we obtain $f_{\text{Model}_3} = 1.2 \times 10^{-3} \text{ s}^{-1}$ and $f^{\text{avg}} = 2.2 \times 10^{-8} \text{ s}^{-1}$. Compared to f^{mean} obtained with the mean-barrier from Model 1, which is $f_{\text{Model}_1} = 6.6 \times 10^{-10} \text{ s}^{-1}$, we see that the proper micromagnetic modeling $f_{\text{Model}_1} \rightarrow f^{\text{avg}}$ leads to an increase of the switching rate up to two orders of magnitudes, whereas differentiation between parallel switching channels $f^{\text{avg}} \rightarrow f_{\text{Model}_3}$ improves the switching rate by additional five orders of magnitude.

D. Material parameters

In order to further validate our findings, we vary the saturation magnetization M_s and calculate the energy barriers for a LR switching using the configuration $c = 63$. Here the exchange stiffness constant is scaled [29,30,33] according to $A(M_s) = A_0 \cdot (M_s/M_0)^{1.7}$, where $A_0 = 18 \text{ pJ/m}$ and $M_0 = 950 \text{ kA/m}$ are the exchange stiffness and saturation magnetization at $T = 0 \text{ K}$.

As depicted in Fig. 8, independently from the choice of the material parameters, the full micromagnetic model gives always the lowest energy barriers, and thus, the highest switching rates. Furthermore, the differences between the CW

and CCW switching channels increase with M_s , yet the CW channel yields always the lower barrier. Note that a decrease in M_s is equivalent to an increase of the temperature at which the simulations are performed. Hence, as M_s decreases the temperature of the system approaches T_{Curie} and the overall barriers are decreasing, so significantly diminishing the absolute differences between the models. That being said, in order to effectively calculate energy barriers and reproduce experiments, it is of crucial importance to accurately determine the material laws for A and M_s as a function of temperature. The scaling method can be seen as a limitation of the proposed full micromagnetic model, where the effect of thermal fluctuations is introduced only indirectly via the above mentioned micromagnetic parameters, rather than making use of stochastic methods like Langevin dynamics [34,35], Landau-Lifshitz-Bloch equation [36–38] or constrained Monte Carlo [39].

In summary, our results indicate that the values obtained with Model 1 deviate significantly from the values of Model 3. The mean barrier approximation falsely overestimates the energy barriers when an interaction field with a nonvanishing y component acts on the central island, and thus, the CW and CCW barriers are different. As a consequence, the switching rates are underestimated. Although Model 2 recovers the most important shortcomings of Model 1 by using micromagnetics and differentiating between CW and CCW channels, it is Model 3 that gives the lowest and more realistic energy barriers. Thereby, independently from the chosen material parameters, Model 3 provides the best approximation of the energy barriers and transition rates that should be utilized to model dynamical processes in sASI lattices, since the probability of switching the magnetization of an island is directly proportional to these rates.

V. CONCLUSIONS

In this paper, we use the SISIM to calculate all energy barriers to switch the magnetization of the central island of a double vertex in square artificial spin ice.

We investigate the influence of the nearest-neighbor islands on the energy barrier in a square artificial spin ice lattice by calculating the energy barrier with three different approaches.

In the first model we consider the widely used mean-barrier approximation. Besides the nonuniformities in the magnetization of the island of interest already considered in Model 2, the last model, Model 3, is a full micromagnetic model where each magnetization is dynamically relaxed depending on each other.

As a first relevant result, Model 2 yields different minimum energy paths for CCW and CW rotation directions for particular configurations, where the most probable switching occurs via the channel with the lowest energy barrier. This distinction between CW and CCW reversal is completely neglected by the average model, Model 1, often utilized in ASIs literature.

To conclude our results, the energy barrier for switching the magnetization of an island in an artificial spin ice lattice can be reduced significantly applying a full micromagnetic model on a double vertex (Model 3) compared to the energy barrier obtained with the mean-barrier approximation.

The interactions originating from the demagnetization fields from the neighboring islands and the consequent energy

difference between CW and CCW reversal paths are the first key reason for this reduction. The inhomogeneities in the magnetizations of both the central islands and of the dynamically relaxed neighbors, that arise during the reversal, introduce an additional contribution to the reduction of the energy barriers. Both effects are neglected in the mean-barrier approximation.

In most drastic cases the mean-barrier approximation can result in energy barriers 35% higher as Model 3.

While the *ad hoc* reduction of the barriers often applied in mean-barrier model can fix particular barriers, it can over- or underestimate the barriers leading to different dynamics when simulating artificial spin ice systems. The main reason resides in the fact that an average barrier reaches its limitations and has no physical significance regarding the switching rates utilized in the kinetic Monte Carlo simulations.

The presented full micromagnetic model is general and can also be applied for other artificial spin ice lattice types and magnetic materials, if both the number and spatial arrangement of the nearest neighbors are adapted.

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APPENDIX A: MICROMAGNETIC ENERGIES

According to the SISIM, we minimize the total energy of a chosen magnetic region Ω_m . For our purposes, we consider the total energy by means of micromagnetics [40] as

$$E^{\text{tot}} = E^{\text{dem}} + E^{\text{ex}}, \quad (\text{A1})$$

where E^{dem} denotes the demagnetization energy and E^{ex} represents the ferromagnetic exchange energy. We do not consider any externally applied magnetic fields. The demagnetization energy E^{dem} is described as

$$E^{\text{dem}} = -\frac{\mu_0 M_s}{2} \int_{\Omega_m} \mathbf{m} \cdot \mathbf{H}^{\text{dem}} d\mathbf{x}, \quad (\text{A2})$$

where μ_0 is the vacuum permeability, Ω_m defines the magnetic region, and $\mathbf{m}$ represents the normalized magnetization vector.

The demagnetizing field $\mathbf{H}^{\text{dem}}$ is given by

$$\mathbf{H}^{\text{dem}}(\mathbf{x}) = -\frac{M_s}{4\pi} \int_{\Omega_m} \nabla \nabla' \frac{1}{|\mathbf{x} - \mathbf{x}'|} \mathbf{m}(\mathbf{x}') d\mathbf{x}'. \quad (\text{A3})$$

Moreover, the ferromagnetic exchange energy favoring a parallel alignment of the spins is defined as

$$E^{\text{ex}} = \int_{\Omega_m} A(\nabla \mathbf{m})^2 d\mathbf{x}, \quad (\text{A4})$$

where A is the exchange stiffness constant.

APPENDIX B: MEAN-BARRIER APPROXIMATION

Especially for kinetic Monte Carlo simulations of artificial spin ice, the mean switching barrier is often expressed *solely* depending on the energies of the initial and final energy state. The initial and final state energies can be easily calculated using the point-dipole model or regular micromagnetic simulation codes, where each island is uniformly magnetized, with a single-island switching barrier E^{isol} added as an independent parameter.

The reason that the energies of the intermediate states do not need to be considered in this simple approximation is based on the symmetry properties of the stray field and Zeeman interactions, whose energies are antisymmetric under rotations of π , i.e., $E(\phi + \pi) = -E(\phi)$. Under the assumptions of a Stoner-Wohlfarth particle as central island and constantly magnetized neighboring islands, the total energy of the central island is given by an anisotropy and Zeeman term in the form

$$E(\mathbf{m}, \mathbf{H}) = E^{\text{ani}}(\mathbf{m}) + E^{\text{zee}}(\mathbf{m}, \mathbf{H}), \quad (\text{B1})$$

with

$$E^{\text{ani}}(\mathbf{m}) = -K_V (\mathbf{m} \cdot (1, 0, 0))^2, \quad (\text{B2})$$

$$E^{\text{zee}}(\mathbf{m}, \mathbf{H}) = -\mu_0 \mu_s \mathbf{m} \cdot \mathbf{H}, \quad (\text{B3})$$

where $K_V = k_{\text{eff}} \cdot V$ denotes the product of the effective anisotropy constant k_{eff} (basically due to the shape anisotropy) and the volume of the isolated nanostructure V , and $\mu_s = M_s \cdot V$ is the total magnetic moment. Furthermore, we assume that this field is sufficiently weak, so that both the initial and final states can be approximated by $\mathbf{m}^{\text{ini}} = (-1, 0, 0)$ and $\mathbf{m}^{\text{fin}} = (1, 0, 0)$. In general, the value of the activation barrier is given by the difference between the saddle point energy E^{saddle} as given by Eq. (1), i.e., central moment pointing up, $E_{\text{CW}}^{\text{saddle}}$ with $\mathbf{m}_{\text{CW}}^{\text{saddle}} = (0, 1, 0)$, or down, $E_{\text{CCW}}^{\text{saddle}}$ with $\mathbf{m}_{\text{CCW}}^{\text{saddle}} = (0, -1, 0)$ in a configuration c , and the initial energy of the configuration E^{ini} . Note, that the magnetization state with the

maximum energy involved in the energy barrier calculation can significantly deviate from $\mathbf{m}_{\text{CW/CCW}}^{\text{saddle}}$ for high interaction fields. We assume that this deviation is also negligible. In this context we obtain

$$E^{\text{ini}}(\mathbf{H}) = E(\mathbf{m}^{\text{ini}}, \mathbf{H}) = -K_V + \mu_0 \mu_s H_x, \quad (\text{B4})$$

$$E^{\text{fin}}(\mathbf{H}) = E(\mathbf{m}^{\text{fin}}, \mathbf{H}) = -K_V - \mu_0 \mu_s H_x, \quad (\text{B5})$$

$$E_{\text{CW}}^{\text{saddle}}(\mathbf{H}) = E(\mathbf{m}_{\text{CW}}^{\text{saddle}}, \mathbf{H}) = -\mu_0 \mu_s H_y, \quad (\text{B6})$$

$$E_{\text{CCW}}^{\text{saddle}}(\mathbf{H}) = E(\mathbf{m}_{\text{CCW}}^{\text{saddle}}, \mathbf{H}) = +\mu_0 \mu_s H_y. \quad (\text{B7})$$

We get for the energy barrier of an isolated nanostructure

$$\Delta E^{\text{isol}} = E_{\text{CW}}^{\text{saddle}}(\mathbf{H} = (0, 0, 0)) - E^{\text{ini}}(\mathbf{H} = (0, 0, 0)) = K_V.$$

With this result, we are finally able to express the mean-energy barrier of the CW and CCW channels by ΔE^{isol} , E^{fin} , and E^{ini} as in Eq. (4):

$$\begin{aligned} \Delta E^{\text{mean}} &= 1/2(\Delta E_{\text{CW}}^{\text{saddle}} + \Delta E_{\text{CCW}}^{\text{saddle}}) \\ &= 1/2(E_{\text{CW}}^{\text{saddle}} - E^{\text{ini}} + E_{\text{CCW}}^{\text{saddle}} - E^{\text{ini}}) \\ &= K_V - \mu_0 \mu_s H_x \\ &= \Delta E^{\text{isol}} + 1/2(E^{\text{fin}} - E^{\text{ini}}). \end{aligned}$$

Based on the aforementioned assumptions, Eq. (4) is an approximation of an average barrier between the two energy barriers for the corresponding parallel switching channels. Even when the initial and final states are involved in this equation, it might neglect the interaction fields originating from the neighboring islands. In a case, where $E^{\text{ini}} = E^{\text{fin}}$, the mean-barrier coincides with the energy barrier of one isolated nanostructure, e.g., configuration $c = 31$. But in reality there is still an effective field acting on the central island. According to the Stoner-Wohlfarth Model, the energy barrier of switching the magnetization of a single domain particle under the influence of an external field $\mathbf{H} = (0, H_y, 0)$ is $\Delta E = \Delta E^{\text{isol}}(1 - \frac{H_k}{H_y})^2$, where H_k is the strength of the anisotropy field and H_y the strength of the field acting on the single-domained particle [41]. With Eq. (4) one would obtain that $\Delta E^{\text{mean}} = \Delta E^{\text{isol}}$. This deviation points out once more, that the mean-barrier approximation method needs further corrections, to be used as input for kinetic Monte Carlo simulations.

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Chapter 3

Thermally superactive artificial kagome spin ice structures obtained with the interfacial Dzyaloshinskii-Moriya interaction

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Personal Contributions

The personal contributions include performing the micromagnetic simulations presented in the paper, writing of a code that generates finite-element meshes from AFM images, which were used as an input for further simulations, writing details about the simulations in the manuscript and in the supplemental material.

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Artificial kagome spin ice exhibits exotic magnetic correlations driven by a combination of geometric frustration and dipolar interactions that, at low-enough temperature, can result in ordered phases. This order, whether it is the ground state of several kagome rings, or the theoretically predicted long-range order of an extended array, has yet to be experimentally observed. By introducing an interfacial Dzyaloshinskii-Moriya interaction, we are able to reduce the blocking temperature of the individual nanomagnets, allowing a system of 30 kagome nanomagnets to explore its vast manifold of microstates and find its ground state. Furthermore, the extracted magnetic correlations in an extended artificial kagome spin ice are found to exhibit quantitative signatures of long-range charge order, providing evidence of the theoretically predicted continuous phase transition to the charge-ordered state. The significant lowering of the blocking temperature in nanomagnets is important for the exploitation of superparamagnetism in artificial spin systems and devices.

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Artificial spin ices are lithographically defined magnetic metamaterials that are used to explore emergent magnetic phenomena arising from geometrical frustration [1–3]. These single-domain bistable nanomagnets are arranged on two-dimensional lattices and, as a result of geometrical frustration, not all pairwise magnetic interactions can be optimized at the same time. A prominent example is the artificial kagome spin ice [4–6], where a number of phases have been theoretically predicted [7,8]. On decreasing the temperature within the ice-rule sector, a transition to a vertex-charge ordered phase is predicted to occur at $T_{\text{crit}}^{\text{charge}} = 0.35J_{\text{NN}}$, where $J_{\text{NN}} \propto m^2/a^3$ is the nearest-neighbor coupling strength between nanomagnets of magnetic moment $m = |\mathbf{m}|$ separated by a distance a (see Fig. 1). As the temperature is further decreased to $T_{\text{crit}}^{\text{spin}} \approx 0.12J_{\text{NN}}$, the kagome spin ice is predicted to transition into a long-range spin- and charge-ordered ground state.

In contrast to artificial square ice, which undergoes an Ising transition directly from the paramagnetic state at the critical temperature $T_{\text{crit}} \sim 2.5J_{\text{NN}}$ [9], the kagome system has been found to freeze at its blocking temperature before such a transition can be observed. In fact, with spatially resolved

measurements of thermally active artificial kagome spin ices, only ice-rule obedience [10] and the formation of vertex-charge crystallites [11–13] have so far been observed. Neither the low-temperature equilibrium correlations associated with the vertex-charge ordering ($T < T_{\text{crit}}^{\text{charge}}$) nor the spin ordering ($T < T_{\text{crit}}^{\text{spin}}$) of an extended kagome array have been observed. Even for a much smaller system consisting of seven hexagonal rings, as shown in Fig. 1, the probability of obtaining one of the two ground state configurations is small, since there are 10^5 competing low-energy microstates that satisfy the ice rule. Indeed, to date, the ground state of such seven-ring structures has only been achieved by direct writing [14], but not using thermally active nanomagnets [15].

To observe dynamics at the temperature scales of $T_{\text{crit}}^{\text{charge}}$ or $T_{\text{crit}}^{\text{spin}}$, the blocking temperature defined as $T_B = E_B/(k_B \ln \frac{\tau_m}{\tau_0})$ must be reduced. Here τ_m , τ_0 , and E_B are the characteristic timescale of the experimental measurement, the inverse attempt frequency of magnetic reorientation [16], and the barrier energy for magnetic reorientation, respectively, and k_B is the Boltzmann constant. For nanomagnets, $E_B = K_{\text{eff}}V$, where K_{eff} is the effective local magnetic anisotropy, and V is the volume of the nanomagnet given by its length L , width W , and thickness h (see Fig. 1). Thus T_B may be reduced by decreasing the volume of the nanomagnets. However, since the coupling constant $J_{\text{NN}} \propto m^2$ and $m = M_{\text{sat}}V$, the critical

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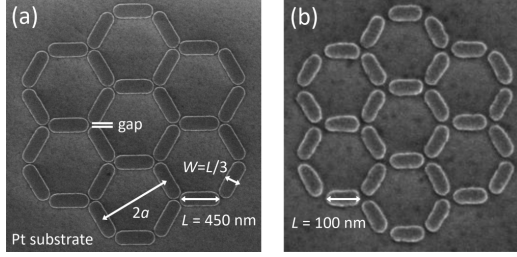


FIG. 1. Seven-ring artificial kagome spin ice structures. Scanning electron microscope images of seven-ring artificial kagome structures patterned on a 5 nm Pt film with 30 $\text{Ni}_{80}\text{Fe}_{20}$ nanomagnets of dimensions length $L \times$ width $W \times$ thickness h of (a) $450 \text{ nm} \times 150 \text{ nm} \times 2.7 \text{ nm}$ and (b) $100 \text{ nm} \times 34 \text{ nm} \times 3.3 \text{ nm}$. The lattice parameter a is defined by L and a gap of 20 nm.

temperatures will also reduce and by a far greater proportion. This latter dependency has been exploited to produce equilibrium room temperature dynamics by either decreasing the nanomagnet volume V [9,17] or the saturation magnetization M_{sat} [18,19]. However, with the ratio $T_B/T_{\text{crit}} \propto a^3/V \propto L/h$; see Secs. I and II of the Supplemental Material (SM) [20], such an approach, while useful for the spin ice regime ($T \gg T_{\text{crit}}^{\text{charge}}$), is not an appropriate strategy for observing equilibrium dynamics at $T < T_{\text{crit}}^{\text{charge}}$.

In this work, we employ an approach to decrease the nanomagnet blocking temperature T_B by exploiting the interfacial Dzyaloshinskii-Moriya interaction (DMI) [21,22]. This leads to the lowering of the effective anisotropy constant K_{eff} given by the shape anisotropy K_{xy} and a countering effective DMI anisotropy K_{DMI} [23]. The interfacial DMI therefore lowers T_B , but not T_{crit} , as the dipolar interactions between nanomagnets remain unchanged. As has been shown by Cubukcu *et al.* [23], a sufficiently strong DMI will modify the orientation of the easy axis originally dictated by shape anisotropy. We exploit this strategy, introducing interfacial DMI to produce thermally superactive nanomagnets with rapidly fluctuating moments down to temperatures of $T = 50 \text{ K}$. Arrays of such superactive nanomagnets can efficiently explore the vast network of microstates of the kagome spin ice, which we demonstrate with the observation of ground state ordering in seven-ring structures, as well as correlations close to the charge order transition in extended kagome lattices.

As indicated by the results of micromagnetic simulations, shown in Fig. 2(a) and the Supplemental Material movies [20], a quasicoherent rotation governs the magnetization reversal as determined by minimum energy path simulations [24–26] using bulk $\text{Ni}_{80}\text{Fe}_{20}$ parameters [27]. The energy associated with the intermediate state sets an energy barrier E_B with the net magnetization momentarily pointing along the short axis of the nanomagnet. The reduction in anisotropy as a result of introducing DMI originates from an out-of-plane canting of the spins at the nanomagnet edges [23,28,29]. This decreases the local spin-spin energy, as indicated by the areas highlighted in red in Fig. 2(a), and interestingly results in a more coherent reversal upon the

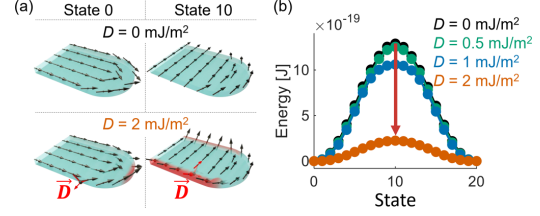


FIG. 2. Minimum energy path simulation of the moment reversal in the presence of the interfacial DMI. Simulated spin structures of a nanomagnet in its stable state (State 0, with magnetization along the long axis) and intermediate states at the energy barrier (State 10), both with and without interfacial DMI (D). With the interfacial DMI ($D = 2.0 \text{ mJ/m}^2$), the tilt of the spin at the edges, highlighted in red, reduces the total energy during magnetization reversal. (b) The minimum energy path during magnetization reversal is simulated using a full micromagnetic model with four nearest neighbors of nanomagnets and variable DMI strength D . The energy barrier E_B is the difference in energy between State 0 and State 10.

addition of the interfacial DMI. The energy barrier as a function of DMI strength D is given in Fig. 2(b), where stronger DMI leads to a larger reduction in the energy barrier as highlighted by the red arrow. We furthermore provide a direct comparison of blocking temperatures of nanomagnets fabricated on Pt (with DMI) and Si (without DMI) using temperature-dependent x-ray photoemission electron microscopy (X-PEEM) [30] (see SM Fig. 2 [20]).

The interfacial DMI effect entails a more complex dependence of the blocking temperature on the nanomagnetic volume, which in turn can be exploited to reduce $T_B/T_{\text{crit}} \propto T_B/J_{\text{NN}}$. The relation, as derived in the Supplemental Material [20],

$$\frac{T_B}{J_{\text{NN}}} \propto \left(K_{\text{xy}} \frac{L}{h} - \frac{2D_s^2}{\sqrt{A_{\text{ex}}K_{\text{xz}}}} \frac{1}{h^3} \right), \quad (1)$$

indicates that there exists a maximum $T_B/J_{\text{NN}} \propto L^{3/2}/D_s$ at $h_{\text{max}} \propto 1/\sqrt{L}$. For h lower than this value, T_B/J_{NN} decreases due to the increasing negative DMI contribution (DMI-dominant regime), whereas above this value a decrease is due to a decrease in the shape anisotropy contribution. The maximum h_{max} separating the two regimes may be decreased by either lowering the lateral dimension L , or by increasing the effective DMI D by reducing the thickness h [31]. Given that the above expression is for an ideal system, and that experimental realizations have increasing disorder as the dimensions of the nanomagnets are reduced, we investigate the predicted DMI dependence by exploring a range of seven-ring kagome systems in which both L and h are varied.

The seven-ring kagome structures are fabricated using $\text{Ni}_{80}\text{Fe}_{20}$ nanomagnets with lengths $L = 450, 300, 210, 150$, and 100 nm , and a fixed aspect ratio $L/W = 3$. The samples were fabricated with lift-off assisted electron beam lithography from a $\text{Ni}_{80}\text{Fe}_{20}$ thin-film wedge of variable thickness h between 0 and 7 nm over a distance of 6 mm deposited by thermal evaporation (base pressure 10^{-7} mbar). The gaps between neighboring nanomagnets [see Fig. 1(a)] are chosen to be as small as lithographically possible (here 20 nm) to

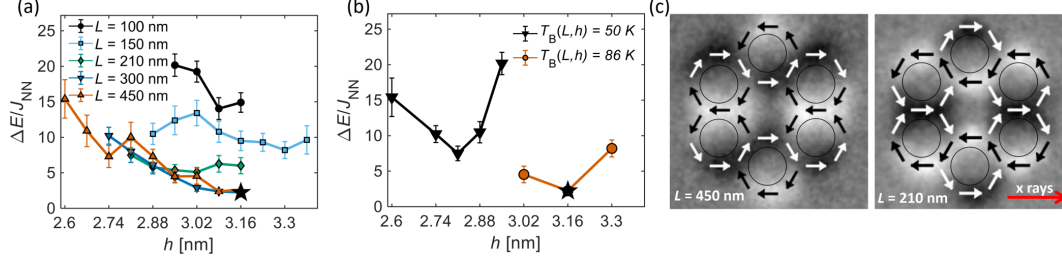


FIG. 3. Magnetic ordering in seven-ring kagome structures. (a) Normalized point-dipolar energy of magnetic configurations compared to the ground state [$\Delta E/J_{NN} = (E_{\text{config}} - E_{\text{GS}})/J_{NN}$] determined from X-PEEM images, where each point corresponds to a kagome structure incorporating nanomagnets with a particular L and h . For each data point, the configuration energies of 20 seven-ring structures are averaged, with the error bars indicating the standard deviation. The structure with $\Delta E/J_{NN}$ closest to the ground state is indicated with a black star. (b) Normalized configuration energies $\Delta E/J_{NN}$ for two experimentally determined blocking temperature values $T_B(L, h)$. (c) Example X-PEEM images of seven-ring artificial kagome spin ice structures in the ground state with nanomagnet dimensions ($L \times W \times h$) $450 \text{ nm} \times 150 \text{ nm} \times 2.7 \text{ nm}$ (top image) and $210 \text{ nm} \times 70 \text{ nm} \times 3.1 \text{ nm}$ (bottom image) fabricated on top of a 5 nm Pt film. The arrows indicate the orientation of the magnetic moments and the circles indicate closed loops of head-to-tail moments. The red arrow indicates the x-ray direction.

maximize $J_{NN} \propto 1/a^3$ by minimizing a . The interfacial DMI is introduced by fabricating the seven-ring structures on top of a 5 nm thin Pt film. From the systematic analysis described in the Supplemental Material [20], the chosen nanomagnet dimensions are expected to result in a regime that is dominated by DMI and therefore a reduction in blocking temperature.

Arrays of such seven-ring kagome structures were exposed to a quenching protocol in which the temperature was reduced from 300 down to 30 K over a period of 1 h. Imaging of the resulting magnetic configurations was performed by means of X-PEEM [32]. Magnetic contrast is obtained by exploiting x-ray magnetic circular dichroism (XMCD) and dividing the images obtained with right- and left-circularly polarized x rays at the Fe L_3 edge [33]. The resulting XMCD contrast is proportional to the projection of the magnetic moments onto the x-ray propagation vector, with moments pointing towards (away from) the incoming x rays appearing bright (dark) with a sensitivity and resolution that allows for the determination of the entire magnetic configuration. At 30 K, all configurations were no longer thermally active within the observational time frame of the experiment. To quantify how close the observed magnetic configurations are to the known ground state, the point-dipolar energy E_{config} is determined for each seven-ring structure. Specifically, we determine the average point-dipolar energy above the ground state $\Delta E = \langle E_{\text{config}} - E_{\text{GS}} \rangle$ for 20 equivalent structures, where E_{GS} is the point-dipolar energy of the ground state. In order to compare the energy of structures with different dimensions, ΔE is normalized to the nearest-neighbor dipolar coupling pre-factor J_{NN} .

A low value of ΔE suggests that the system remained thermally active at low temperatures, where it can explore the large number of low-energy states, thus reflecting a low blocking temperature T_B . In Fig. 3(a) the obtained values of ΔE as a function of h for different L are displayed for 34 unique values of h and L . The magnetic configurations closest to the ground state energy are indicated with a black star and involve $L = 300 \text{ nm}$ and $L = 450 \text{ nm}$ at $h \simeq 3.2 \text{ nm}$. In total, three seven-ring ground state configurations were found, two of which are shown in Fig. 3(c).

In Fig. 3(a), it is demonstrated that samples with a larger L generally exhibit a lower energy configuration (and therefore a correspondingly lower blocking temperature) with increasing h , a trend opposite to what is expected from Eq. (1) for the DMI-dominated regime. This observation is attributed to increasing disorder for smaller h , where it is known that such thin nanomagnets have a reduced magnetization [34]. Encouraging is that, by increasing h , ΔE is indeed reduced, indicating the emergence of the DMI effect and the observation of the ground state configuration at $h \simeq 3.2 \text{ nm}$. For the samples with smaller L , the trend is not so apparent with the magnetic configurations generally having energies well above that of the ground state as well as a broader range of ΔE . Thus systems with smaller nanomagnets tend to fall out of equilibrium at higher temperature, an observation that can be rationalized again by the increasing presence of disorder. Indeed, as the lateral dimensions of the nanomagnets are reduced, edge roughness, and therefore pinning, becomes more important. This in turn increases T_B , so that the system falls out of equilibrium at higher temperatures. This has also been observed in connected systems where lithographic irregularities for smaller sizes adversely affect the magnetic ordering [35,36] and is further discussed in the Supplemental Material [20].

The presence of disorder thus complicates the relationship between ΔE and the blocking temperature. To quantify this, we define an upper estimate to T_B as the temperature below which no more than three to five moment fluctuations were observed within 50 s, corresponding to the time of ten consecutive XMCD images. We thus obtain T_B as a function of L and h [see SM Fig. 4(a) [20]], and in Fig. 3(b) plot the data from (a) that have a T_B of $50 \pm 5 \text{ K}$ and $86 \pm 5 \text{ K}$. Inspection of the figure reveals a nonmonotonic relationship between ΔE and T_B , characterized by a minimum in ΔE . An optimal geometry exists that involves the minimization of microscopic disorder in terms of h and lithographic irregularities in terms of L , to give an artificial kagome spin ice whose low-energy configurations, and indeed the ground state, in smaller structures can be accessed—with a

characteristic blocking temperature controlled by DMI through Eq. (1).

We now turn our attention to the magnetic moment and charge ordering of an extended artificial kagome spin ice consisting of 3684 nanomagnets arranged on the sites of a kagome lattice, which has been cooled to 30 K using the same procedure described above. With the nanomagnet dimension being $450 \text{ nm} \times 150 \text{ nm} \times 2.9 \text{ nm}$, the equivalent upper estimate of the blocking temperature in the seven-ring structure would be $T_B = 80 \pm 5 \text{ K}$ [Fig. 3(b)]. These dimensions are comparable to those used in previous systems without DMI [10], which allows us to directly compare the degree of ordering in artificial kagome spin ice systems with and without the interfacial DMI. For this earlier artificial kagome system, the upper estimate of the blocking temperature using the same criterion as chosen here was found to be $T_B = 420 \text{ K}$ [10].

An alternative approach to estimating the absolute blocking temperature is to deduce the reduced blocking temperature $T_{\text{red}}^{D \neq 0} = T/J_{\text{NN}}$ at which the extended system freezes, therefore giving insight into what part of the phase diagram can be probed under equilibrium conditions. The value of T_{red} can be obtained from the spin-spin and charge-charge correlators extracted from the XMCD images that are in turn compared to those obtained from Monte Carlo simulations of the point-dipolar interacting kagome system. The temperature-dependent values of the simulated spin and charge correlators C_{ij} are shown in Figs. 4(a) and 4(b), respectively. The reduced temperature for the experimentally obtained correlator values (filled circles, with DMI) is determined with a standard-deviation correlator analysis using a spread-out function [13,37] to be $T_{\text{red}}^{D \neq 0} = 0.84$ and is indicated by the left-hand dashed line in Fig. 4.

We find our thermally superactive artificial kagome spin ice to be close to the predicted transition to the charge-ordered phase at $T_{\text{crit}}^{\text{charge}}/J_{\text{NN}} \approx 0.35$ (see SM Fig. 5 [20]). Performing the same spin and charge correlator analysis (open circles in Fig. 4 indicated by the right-hand dashed line) on the artificial kagome spin ice without interfacial DMI measured by Farhan *et al.* [10], gives $T_{\text{red}}^{D=0} = 4.49$.

The observation that the experimentally obtained temperatures T_B and T_{red} for the artificial kagome spin ice with DMI (80 K and 0.84) and without DMI (420 K and 4.49) are both decreased by a factor of 5, indicates a consistency in our approach in which the dipolar coupling strength J_{NN} is not significantly affected by interfacial DMI. We can therefore conclude that DMI is responsible for a considerable decrease in blocking temperature, thereby extending the experimentally accessible equilibrium temperature range of the artificial kagome system. The present work therefore provides a new way to control the T_B of thermally active nanomagnets, exploring the disorder-induced limits of downsizing their lateral dimensions and thickness, which remains an obstacle to observe the low-temperature phases of artificial kagome spin ice.

In conclusion, we demonstrated that the introduction of the interfacial DMI leads to a reduction in the effective anisotropy of nanomagnets, thus enhancing the superparamagnetic fluctuation rate while maintaining dipolar interactions between the nanomagnets. We demonstrated ground state ordering in seven-ring kagome structures, and obtained a thermally superactive system with a blocking temperature very close to the

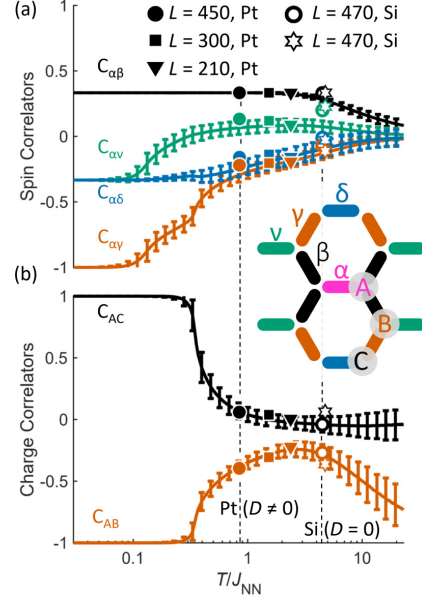


FIG. 4. Spatial correlations in artificial kagome spin ice. Monte Carlo simulations of (a) spin-spin correlations and (b) charge-charge correlations, as indicated in the schematic of the artificial kagome spin ice. The first four neighboring nanomagnets to α are indicated with β, γ, ν , and δ , while the first two neighboring vertex charges to A are indicated with B and C . The charges come from the vertex containing two-in/one-out or two-out/one-in in moments resulting in $\pm 1q$ charge [6]. The filled markers denote the correlation values determined from the X-PEEM images for an artificial kagome spin ice with interfacial DMI. The open markers show the correlators obtained for a system without the interfacial DMI, with the open circles representing the values from Farhan *et al.* [10]. The dashed lines indicate the two systems with similar J_{NN} with and without DMI.

charge-order transition in an extended artificial kagome spin ice. The implementation of these superactive nanomagnets will facilitate future studies in other highly frustrated artificial spin systems [38–43]. In addition, the ability to lower the energy barrier for switching, while maintaining a considerable coupling strength, will lead to enhanced reliability and reduced operational power for computation with nanomagnets [44].

The dataset that supports this study is available via the Zenodo repository [45].

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Supplemental Material for: Thermally superactive artificial kagome spin ice structures obtained with the interfacial Dzyaloshinskii-Moriya interaction

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Thermally active artificial spin ice provides a model system for geometric frustration in which the nanomagnetic moments take on the role of Ising spins that can be imaged in real-space and real-time. For the kagome spin ice, two long-range ordered phases have been predicted [1, 2], but these have not been experimentally verified. In order to observe these equilibrium phases, it is necessary to lower the reduced blocking temperature T_B/J_{NN} to below the critical temperatures for the phase transitions. For artificial kagome spin ice, the critical temperatures are $T_{crit}^{charge} = 0.35J_{NN}$ and $T_{crit}^{spin} \approx 0.12J_{NN}$.

I ESTIMATION OF NANOMAGNET INTERACTION STRENGTH

In order to derive the dependence of both T_B and J_{NN} on the geometrical and material parameters, we focus on the dipolar interactions between nanomagnets, which are given by the dipolar pre-factor

$$J_{NN} = \frac{\mu_0 m^2}{4\pi a^3} \propto \frac{m^2}{a^3}. \quad (1)$$

Here μ_0 is the vacuum permeability, a is the lattice constant between nearest-neighbours, and m is the magnitude of the magnetic moment given by

$$m = M_{sat} V, \quad (2)$$

where M_{sat} is the magnetisation at saturation and the volume V of a stadium-shaped nanomagnet is given by

$$V = \left(L - \frac{\pi - 4}{4} W \right) W h \propto L^2 h, \quad (3)$$

where L is the nanomagnet length, $W = L/3$ is the nanomagnet width with a fixed aspect ratio, and h is the

thickness of the nanomagnets that are spaced apart with the lattice constant

$$a \approx L + g \approx L, \quad (4)$$

where g is the gap ($g = 20 \text{ nm} \ll L$), which is the smallest separation between neighbouring nanomagnets (see Fig. 1 of the main text). Hence the dipolar interactions can be simplified to

$$J_{NN} \propto M_{sat}^2 L h^2. \quad (5)$$

II DERIVATION OF BLOCKING TEMPERATURE REDUCTION BY INTERFACIAL DMI

The energy barrier E_B that determines the blocking temperature T_B depends, to a first approximation, on the shape and volume of the nanomagnet K_{xy} , as well as the DMI anisotropy K_{DMI} resulting from the interfacial coupling with the heavy-metal layer. Following the same arguments laid out by Cubukcu *et al.* [3], the effective anisotropy due to the interfacial DMI of a simple rectangle is given by

$$K_{DMI} \approx \frac{D_s^2}{h^2 \sqrt{A_{ex} K_{xz}}} \frac{L - W}{LW}, \quad (6)$$

where A_{ex} is the exchange stiffness constant, K_{xz} is the out-of-plane shape anisotropy, and D_s is the surface DMI strength that follows from the thickness-dependent effective DMI strength D [4] given by

$$D = \frac{D_s}{h}. \quad (7)$$

The blocking temperature is then proportional to

$$T_B = \frac{E_B}{k_B \ln \frac{\tau_m}{\tau_0}} \propto (K_{xy} - K_{DMI}) V, \quad (8)$$

where k_B is the Boltzmann constant, τ_m is the characteristic measurement time, and τ_0 the inverse attempt frequency [5]. The in-plane shape anisotropy K_{xy} is given

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by

$$K_{xy} = \frac{1}{2} \mu_0 M_{\text{sat}}^2 (\mathcal{N}_x - \mathcal{N}_y), \quad (9)$$

and the out-of-plane shape anisotropy is given by

$$K_{xz} = \frac{1}{2} \mu_0 M_{\text{sat}}^2 (\mathcal{N}_x - \mathcal{N}_z) - K_u, \quad (10)$$

where $\mathcal{N}_i$ are the demagnetisation factors along the three axes ($i = [x, y, z]$) and K_u is the uniaxial anisotropy (assumed to be zero for $\text{Ni}_{80}\text{Fe}_{20}$). The blocking temperature is then given by

$$T_B \propto \left(K_{xy} - \frac{2D_s^2}{\sqrt{A_{\text{ex}} K_{xz}}} \frac{1}{Lh^2} \right) L^2 h, \quad (11)$$

and the reduced blocking temperature is given by

$$\frac{T_B}{J_{\text{NN}}} \propto \left(K_{xy} \frac{L}{h} - \frac{2D_s^2}{\sqrt{A_{\text{ex}} K_{xz}}} \frac{1}{h^3} \right). \quad (12)$$

The reduced blocking temperature Eqn. (12) is smaller for smaller lateral nanomagnet lengths L , which should result in better magnetic ordering. Furthermore, thinner nanomagnets would result in a lowering of the reduced blocking temperature due to the increased effective DMI strength.

III MINIMUM ENERGY PATH AND ENERGY BARRIER SIMULATION

In order to calculate the energy barriers between the two easy axis magnetisation states, we use the simplified and improved string method [6–8]. The string method computes the minimum energy transition path by employing an iterative approach so that the initial path evolves iteratively towards the minimum energy path (MEP). The MEP represents the energetically most favourable way to switch the magnetisation between the new, relaxed initial state and the new, relaxed final state.

The string method is implemented in magnum.fe, a micromagnetic simulation code based on the finite element method [9]. To create the geometry of the five nanomagnets and generate the finite element meshes, we use GMSH, an open-source 3D finite element grid generator [10]. For our calculations, we consider one central nanomagnet, with the easy axis along the x -direction, surrounded by its four nearest-neighbours (NN), see Supplementary Movies [11].

In the simulations, we used the magnetic material parameters of bulk $\text{Ni}_{80}\text{Fe}_{20}$, with a saturation magnetisation $M_{\text{sat}} = 790 \text{ kA/m}$ and exchange stiffness constant $A_{\text{ex}} = 13 \text{ pJ/m}$. To determine the effect of the interfacial Dzyaloshinskii-Moriya interaction (DMI), we vary the DMI strength between $D = 0.0 \text{ mJ/m}^2$ and $D = 2.0 \text{ mJ/m}^2$.

As an initial step, the reversal of the central nanomagnet is assumed to be a coherent rotation from a uniform initial state with $\hat{\mathbf{m}}_{\text{ini}} = (-1, 0, 0)$ to a uniform final state

with $\hat{\mathbf{m}}_{\text{end}} = (1, 0, 0)$. Here, $\hat{\mathbf{m}}$ denotes the unit-vector-field representation of the magnetisation configuration. This initial path is divided into 21 equidistant states with respect to the energy-weighted arc length of the rotation.

We consider the total energy of the system to be

$$E^{\text{tot}} = E^{\text{dem}} + E^{\text{ex}} + E^{\text{DMI}}, \quad (13)$$

where E^{dem} is the demagnetizing energy, E^{ex} is the ferromagnetic exchange energy, and E^{DMI} is the antisymmetric exchange energy. The demagnetizing energy is given by

$$E^{\text{dem}} = -\frac{\mu_0 M_{\text{sat}}}{2} \int_{\Omega_m} \mathbf{m} \cdot \mathbf{H}^{\text{dem}} d\mathbf{x}, \quad (14)$$

with Ω_m corresponding to the region containing magnetic material $\mathbf{m}$ i.e. the five magnetic nanomagnets, and the demagnetisation field $\mathbf{H}^{\text{dem}}$ given by

$$\mathbf{H}^{\text{dem}}(\mathbf{x}) = -\frac{M_{\text{sat}}}{4\pi} \int_{\Omega_m} \nabla \nabla' \frac{1}{|\mathbf{x} - \mathbf{x}'|} \mathbf{m}(\mathbf{x}') d\mathbf{x}'. \quad (15)$$

The ferromagnetic exchange energy is given by

$$E^{\text{ex}} = \int_{\Omega_m} A_{\text{ex}} (\nabla \mathbf{m})^2 d\mathbf{x}, \quad (16)$$

and the antisymmetric exchange energy due to the interfacial DMI is given by

$$E^{\text{DMI}} = \int_{\Omega_m} D [\mathbf{m} \cdot \nabla (\mathbf{e}_d \cdot \mathbf{m}) - (\nabla \cdot \mathbf{m})(\mathbf{e}_d \cdot \mathbf{m})] d\mathbf{x}, \quad (17)$$

with $\mathbf{e}_d$ being the surface normal to the ferromagnetic-heavy metal interface. The energy barrier E_B can be computed using

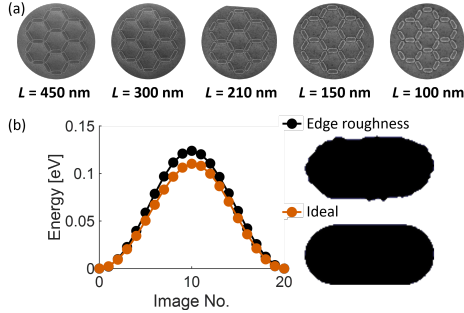
$$E_B = E^{\text{saddle}} - E^{\text{ini}}, \quad (18)$$

where E^{saddle} is the energy corresponding to the saddle point of the MEP (State 10) and E^{ini} is the energy corresponding to the initial magnetisation (State 0).

The full animation of the magnetisation reversals with and without the interfacial DMI are provided in the Supplementary Movies [11], where the colour scale represents the out-of-plane magnetisation component $\mathbf{m}_z$. It can be seen that, with the interfacial DMI, the reversal is more coherent as a result of the additional short-axis anisotropy.

IV EFFECT OF ROUGHNESS

To address the influence of lithographic edge roughness on the blocking temperature, we performed the energy barrier simulations for nanomagnets with DMI as shown in Suppl. Fig. 1. Scanning electron microscopy (SEM) images were used to define the geometry. Comparing nanomagnets with and without edge roughness, while keeping the magnetic moment constant, we find that there is a significant increase in the energy barrier by



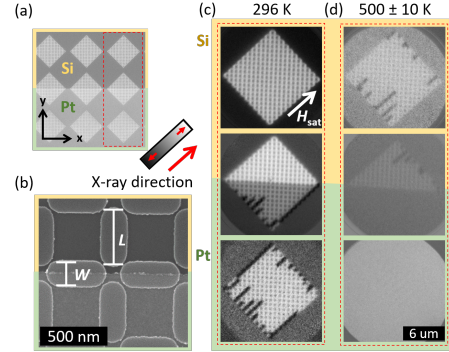
Suppl. Fig. 1. (a) Experimental scanning electron microscopy (SEM) image series of seven-ring artificial kagome spin ice structures with $L = 450$ nm, 300 nm, 210 nm, 150 nm and 100 nm. (b) Energy barrier simulation of a nanomagnet of length $L = 100$ nm and aspect ratio $L/W = 3$ with and without edge roughness. To include the edge roughness, the shape is taken from an SEM image of a nanomagnet of length $L = 100$ nm (top image). This is compared to the ideal nanomagnet (bottom image) with only small edge roughness resulting from the meshing (bottom image). In the energy barrier simulations, the total volume, and therefore the magnetic moment, of both the nanomagnet with edge roughness and the ideal nanomagnet is the same.

13.6 meV or 10% on introducing edge roughness. This corresponds to a factor of 24 in the reorientation frequency at 50 K. This is due to the fact that, with the interfacial DMI, coherent rotation is favoured and the added edge roughness gives rise to a non-uniform state that leads to an increase in the energy barrier.

V DIRECT VISUALIZATION OF THE REDUCTION OF T_B DUE TO INTERFACIAL DMI IN X-PEEM

In order to experimentally verify the origin of the reduced blocking temperature, we first measured the saturation magnetization of nanomagnets with and without the interface to the heavy-metal Pt. For this, we performed SQUID-VSM measurements and determined that identically grown $\text{Ni}_{80}\text{Fe}_{20}$ nanomagnets (with dimensions $L = 450$ nm, $W = 150$ nm, and $h = 4.2$ nm) on 5 nm Pt and on a Si substrate have the same saturation magnetization M_{sat} of 360 kA/m.

In order to experimentally verify that Pt induces an interfacial DMI, we fabricated a Pt strip on a Si substrate. We then patterned arrays of $\text{Ni}_{80}\text{Fe}_{20}$ nanomagnets, both on the Pt strip and the neighbouring Si substrate, depositing a film wedge to obtain nanomagnet arrays with different thickness. We measured the blocking temperatures using time-dependent X-PEEM [12], and the nanomagnet thickness is varied along the x-direction of the sample, see Suppl. Fig. 2(a), ensuring a blocking temperature T_B within the accessible temperature



Suppl. Fig. 2. (a) Scanning electron microscope (SEM) image of wedge sample of variable $\text{Ni}_{80}\text{Fe}_{20}$ thickness along the x-axis of the sample and (b) a zoom in of nanomagnets with their lateral dimensions L and $W = L/3$ indicated. The Si and Pt surface are indicated and can be seen in de zoom in (b) where the higher contrast surface is coming from the Pt. The red box in (a) indicates the thermally active $\text{Ni}_{80}\text{Fe}_{20}$ of thickness 4.2 nm artificial square ice on a 5 nm Pt layer at room temperature (RT) imaged with time dependent X-PEEM. In X-PEEM, nanomagnets with moments pointing towards the x-ray propagation direction, see red arrow in (a), appear bright, while moments opposing the incoming x-rays appear dark. On Si, the identically prepared array is still in a remanent magnetization state (all white) after saturating with an in-situ field applied in the direction of the x-rays. (c) On heating to 500 K, the nanomagnets on Si become thermally active on the same timescale as those on Pt at 296 K. Simultaneously, the magnetic contrast of $\text{Ni}_{80}\text{Fe}_{20}$ on Pt disappears at 500 K due to the high switching rate of the nanomagnets, resulting in an averaged grey contrast.

range of the X-PEEM. The overall size of each individual array is 22 by 22 nanomagnets, occupying an area of $10 \mu\text{m} \times 10 \mu\text{m}$. A control array was fabricated on top of both the Si and Pt interface in order to observe the effect of Pt inside a single circular field-of-view with a diameter of $15 \mu\text{m}$, see Suppl. Fig. 2.

Magnetic imaging was performed at the SIM beamline PEEM endstation at the Swiss Light Source, employing x-ray magnetic circular dichroism (XMCD) at the Fe L_3 edge [13]. XMCD contrast images are obtained by pixel-wise division of images recorded with right and left circularly polarized x-rays. The resulting XMCD contrast gives a direct measure of the projection of the magnetic moments onto the x-ray propagation vector. Moments pointing towards the incoming x-rays appear dark, while moments opposing the x-ray direction appear bright (see Suppl. Fig. 2(c-d)). For each x-ray polarization, an exposure time of two seconds is chosen, while switching polarizations regularly takes one second. This gives an overall time of roughly three seconds ($\tau_m \approx 3$ seconds) to obtain an XMCD image.

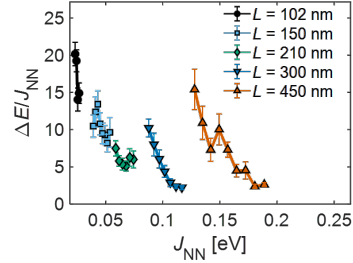
Initially, we find the thickness of $\text{Ni}_{80}\text{Fe}_{20}$ patterned on Pt where moment reorientations take place on the timescale of the acquisition time of the PEEM, see bottom panel of Suppl. Fig. 2(c). For this purpose, the sample is saturated along the x-ray direction, and subsequently, the thickness along the x-direction is found where switched magnetization appears as dark spots in an otherwise remanent state (all white). An XMCD sequence is taken to determine the reorientation rate, where 10 switches per image series is taken as the array with T_B at room temperature (296 K). As a check, the array of $\text{Ni}_{80}\text{Fe}_{20}$ nanomagnets on Si is imaged throughout and is still found to be in the magnetized state after 12 hours, see top panel of Suppl. Fig. 2(c). The sample is then heated in-situ up to 500 K, where the reorientation rate of the nanomagnets on Si is similar to the reorientation rate at room temperature for $\text{Ni}_{80}\text{Fe}_{20}$ on Pt, see top panel Suppl. Fig. 2(d). For the array with $\text{Ni}_{80}\text{Fe}_{20}$ nanomagnets on both Si and Pt (middle panel), it can be clearly seen that the reorientation rate on Si is significantly lower than that on Pt that, on average, results in a gray contrast. Hence, the energy barriers for nanomagnets on Pt are significantly lower than for those on Si for identical magnetic moments.

VI REDUCED DIPOLAR ENERGY OF SEVEN-RING STRUCTURES

In order to compare the ordering between different seven-ring artificial kagome structures, we considered the reduced dipolar energy $\Delta E/J_{\text{NN}}$, using only the spin configuration $\hat{S}$ and their distance $\hat{r}$ in fractions of the lattice parameter in the dipolar configuration energy (Eqn. 19 for the dipolar energy). The reduced dipolar energy is calculated by dividing out the dipolar pre-factor J_{NN} , that contains all the geometrical information. The energy of each seven-ring structure is computed by summing the dipolar energy over all neighbours giving E_{config} , similar to the method used by Farhan *et al.* [14], and is repeated for all 20 seven-ring structures found in a single field-of-view PEEM XMCD image. The reduced dipolar energy of the ground-state configuration E_{GS} is computed with the two possible degenerate ground-state configurations given in Fig. 3(c) of the manuscript. The obtained $\Delta E/J_{\text{NN}}$ is the average excitation above the ground-state $\langle E_{\text{config}} - E_{\text{GS}} \rangle$ and the error is given by the standard deviation of the mean. Furthermore, $\Delta E/J_{\text{NN}}$ can be compared for various dipolar pre-factors (Suppl. Fig. 3), from which we conclude that, for each L , the increased coupling (higher J_{NN} due to increased thickness) results in better magnetic ordering.

VII ORDERING AND BLOCKING TEMPERATURE IN SEVEN-RING KAGOME STRUCTURES

In order to compare the magnetic ordering of kagome structures with differently sized nanomagnets, we plot



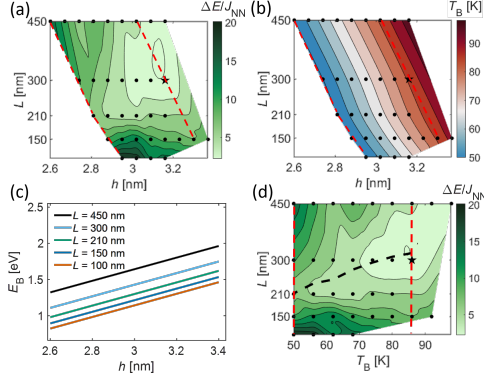
Suppl. Fig. 3. Point-dipolar energy of the magnetic configurations of kagome structures compared to the ground state energy normalized to the dipolar pre-factor ($\Delta E/J_{\text{NN}} = \langle E_{\text{config}} - E_{\text{GS}} \rangle / J_{\text{NN}}$) determined from X-PEEM images where each point corresponds to an average of 20 seven-ring structures for various combinations of L and h . Each colour series corresponds to a particular L and is plotted against J_{NN} , the dipolar pre-factor given by Eqn. (1). The error bars indicate the standard deviation. The lower value of J_{NN} for thinner nanomagnets results in poorer ordering, i.e. lower $\Delta E/J_{\text{NN}}$. Thus each line for a given L shows a downward trend from left (thin nanomagnets) to right (thicker nanomagnets).

the average configuration energies $\Delta E/J_{\text{NN}}$ with an experimentally determined blocking temperature $T_B(L, h)$ at two temperatures of 50 K and 86 K indicated by the dashed red lines in Suppl. Fig. 4(a,c). We have defined T_B as the temperature below which only 3 to 5 moment fluctuations were observed within 50 sec, corresponding to the time of 10 consecutive XMCD images. To determine the blocking temperature T_B as a function of L and h , we identified using X-PEEM the thickness h of kagome structures with $L = 450$ nm, 300 nm, 210 nm, 150 nm and 100 nm, for which $T = 50$ K and at $T = 86$ K are the blocking temperatures.

Since $T_B \propto h$, we can linearly interpolate the blocking temperatures for all remaining thicknesses, as given in Suppl. Fig. 4(b). The resulting average configuration energies $\Delta E(L, T_B)$ are plotted in Suppl. Fig. 4(c) for different nanomagnet lengths L and blocking temperatures $T_B(L, h)$. In Suppl. Fig. 4(c), we have included the location at each lateral dimension where an optimum magnetic ordering is obtained, i.e. a low value of $\Delta E(L, T_B)$. From the dashed black line through these low values of $\Delta E(L, T_B)$ it can be seen that larger lateral dimensions result in better magnetic ordering, with $\Delta E/J_{\text{NN}}$ closer to that of the ground state.

VIII MONTE CARLO SIMULATIONS

We used Monte Carlo (MC) simulations to compute the equilibrium configurations by modelling the individual nanomagnets as point-dipole macrospins and with Ising degrees of freedom along the length of each nanomagnet, arranged on a finite kagome lattice [1, 2, 15, 16]. The



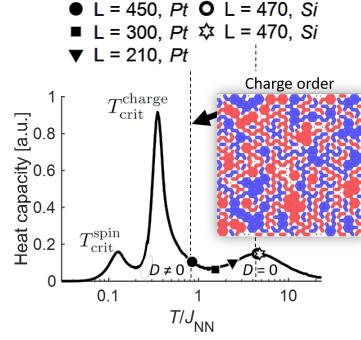
Suppl. Fig. 4. (a) The point-dipolar energy of magnetic configurations compared to the ground state normalized to the dipolar pre-factor ($\Delta E/J_{NN} = (E_{\text{config}} - E_{\text{GS}})/J_{NN}$). These values are determined from X-PEEM images where each point corresponds to nanomagnets with a particular L and h . Each data point corresponds to the average of $\Delta E/J_{NN}$ for 20 seven-ring structures. The location of the observed global minimum in ΔE is indicated with a black star. (b) The experimentally obtained blocking temperatures $T_B(L, h)$ at two temperatures of 50 K and 86 K (red dashed lines) with the remaining $T_B(L, h) \propto h$ linearly interpolated at each L . The two blocking temperatures are also indicated with red dashed lines in (a) and (d). (c) Energy barrier simulations of nanomagnets for various L and h , with the resulting linear relationship between E_B and h . (d) The reduced configuration energy $\Delta E/J_{NN}$ as a function of the interpolated blocking temperatures. Indicated with a dashed black line is the average minimum energy configuration at each blocking temperature, with a trend towards larger L and $T_B(L, h)$ resulting in better magnetic ordering.

Hamiltonian is as follows:

$$H = \frac{1}{2} \sum_{\langle i,j \rangle} \frac{J_{NN}}{(r_{ij}/a)^3} [\hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j - 3(\hat{\mathbf{S}}_i \cdot \hat{\mathbf{r}}_{ij})(\hat{\mathbf{S}}_j \cdot \hat{\mathbf{r}}_{ij})], \quad (19)$$

where $\hat{\mathbf{S}}_i$ is the macrospin unit vector for the macrospin i , $\hat{\mathbf{r}}_{ij}$ is the unit vector connecting the macrospins i and j , and r_{ij} is the distance between the neighbouring macrospins with, a , the nearest-neighbour distance. Interactions up to fifth nearest-neighbours ($r_{ij} = 3.55$) are required to obtain converging results. The simulations were carried out on an array containing $N = 2646$ macrospins with periodic boundary conditions. The simulations are performed with a dipolar pre-factor $J_{NN} = 379$ K to which the temperatures are scaled.

We then perform the simulation in three regions with varying temperature stepsizes and MC sweeps. Here an MC sweep is defined as N single site MC steps followed by $N/2$ hexagonal loop MC steps, where the latter loop algorithm is the same as that used in Ref. [1]. The high-



Suppl. Fig. 5. The heat capacity from MC simulations, showing three peaks corresponding to the paramagnetic-ice cross-over, and the two phase transitions $T_{\text{crit}}^{\text{charge}}$ and $T_{\text{crit}}^{\text{spin}}$, to the charge and spin-ordered phases, respectively. The filled markers correspond to the reduced temperatures determined from the X-PEEM images for an artificial kagome spin ice with interfacial DMI. The open markers correspond to the reduced temperatures obtained experimentally for systems without the interfacial DMI, with the open circle corresponding to the values from Farhan *et al.* [17]. The dashed lines indicate two systems with and without DMI (D), and similar J_{NN} . The extent of the twofold degenerate charge-ordered domains obtained from X-PEEM images is indicated in red and blue in the inset for $L = 450$ nm on Pt with $T/J_{NN} = 0.84$.

temperature paramagnetic to Spin Ice I crossover is simulated by varying the temperature from 9000 K down to 600 K in steps of 100 K with 10000 MC sweeps at each temperature to obtain a well-converged heat capacity. The Spin Ice I to Spin Ice II transition ($T_{\text{crit}}^{\text{charge}}$) is simulated by varying the temperature from 600 K down to 100 K in steps of 5 K and with 10000 MC sweeps. The Spin Ice II to Long-Range Order transition ($T_{\text{crit}}^{\text{spin}}$) is simulated by varying the temperature from 100 K down to 1 K in steps of 1 K with 100000 MC sweeps, eventually resulting in a system-spanning long-range ordered kagome spin ice. The resulting temperature-dependent heat capacity is given in Suppl. Fig. 5. Furthermore, the reduced temperatures (dashed lines) of the experimental systems with and without DMI are indicated. For $L = 450$ nm on Pt (with DMI) it is found that $T/J_{NN} = 0.84$ and for $L = 450$ nm on Si (without DMI) the reduced temperature $T/J_{NN} = 4.49$. Hence, the obtained reduced temperature of the nanomagnets with DMI is closer to the charge-order phase transition.

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Chapter 4

Chiral switching and dynamic barrier reductions in artificial square ice

The following manuscript by Naëmi Leo, Matteo Pancaldi, Sabri Koraltan, Pedro Villalba González, Claas Abert, Christoph Vogler, Florian Slanovc, Florian Bruckner, Paul Heistracher, Kevin Hofhuis, Matteo Menniti, Dieter Suess, Paolo Vavassori was published in New J. Phys. 23 033024.

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Personal Contributions

The personal contributions include performing all the micromagnetic simulations presented in the paper, and preparing figures for supplemental material and videos, and writing the manuscript together with the other authors.

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PAPER

Chiral switching and dynamic barrier reductions in artificial square ice

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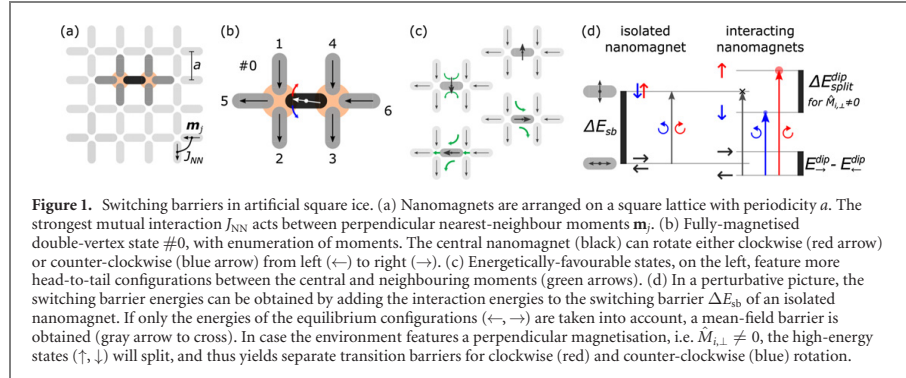
Supplementary material for this article is available [online](#)

Abstract

Collective dynamics in lithographically-defined artificial spin ices offer profound insights into emergent correlations and phase transitions of geometrically-frustrated Ising spin systems. Their temporal and spatial evolution are often simulated using kinetic Monte Carlo (kMC) simulations, which rely on the precise knowledge of the switching barriers to obtain predictive results in agreement with experimental observations. In many cases, however, the barriers are derived from simplified assumptions only, and do not take into account the full physical picture of nanomagnetic switching. Here we describe how the immediate magnetic square- or kagome-ice environment of a nanomagnet reversing via quasi-coherent rotation can induce clockwise and counter-clockwise switching channels with different barrier energies. This energy splitting for chiral reversal channels can be sizeable and, as string-method micromagnetic simulations show, is relevant for artificial spin ice systems made of both exchange- as well as magnetostatically-dominated units. Due to the barrier splitting and further reductions due to non-uniform reversal, transition rates can be exponentially enhanced by several orders of magnitude compared to mean-field predictions, especially in the limit of rare switching events where thermal excitation is less likely. This leads to significantly faster relaxation time scales and modified spatial correlations. Our findings are thus of integral importance to achieve realistic kMC simulations of emergent correlations in artificial spin systems, magnonic crystals, or the evolution of nanomagnetic logic circuits.

Artificial spin ice systems are lithographically-created lattices of elongated single-domain nanomagnets, and have been designed to investigate the effect of correlations and the onset of long-range order in frustrated two-dimensional magnetic lattices [1–4]. Of particular interests is the evolution of extended spin ice lattices from a field-saturated state towards an energetically favourable (ground) state, driven by thermally-activated reversal of individual nanomagnets. Such experiments have been performed mainly using photoelectron emission microscopy, and gave valuable insight on the relaxation process and the formation of spatial correlations [5–9].

The understanding of experimentally-observed temporal evolution of extended spin ices are often guided and supported by model predictions, for example from kinetic Monte Carlo (kMC) simulations.



Their major advantage over full micromagnetic simulations is that they are less computationally costly, and thus can be extended to larger systems and longer time scales. This coarse-grained approach, which disregards microscopic details of the moment reversal, allows to simulate systems with a large number of moments evolving over long time scales, which otherwise would be too computationally-costly to be implemented in full micromagnetic simulations.

To obtain correct relaxation time scales and spatial correlations, kMC simulations rely instead on the precise knowledge of the rates for individual moment reversal. To match the measured experimental time scales the two main parameters determining the switching barriers used in the kMC simulations—the single-particle barrier and the interaction strength—are often adjusted [7, 9–11]. These changes, however, are usually derived from simplified or approximative assumptions only or associated with extrinsic influences, which do not take into account the full physical picture of nanomagnetic switching. Furthermore, an often-used mean-field approach does not consider the intrinsic freedom for clockwise or counter-clockwise rotations [7, 8, 12–14], which can lead to distinct switching barriers, as we previously showed [15].

In this work, we derive that a net perpendicular field from a defect-free double-vertex environment acting on the switching nanomagnet enables favourable chiral reversal pathways in artificial square and kagome ice. Using artificial square ice as an instructive example, we compare switching barriers obtained from micromagnetic string-method simulations for exchange- and magnetostatic-dominated geometries to those derived from simplified point-dipole predictions. We find that the latter consistently overestimates the barriers and underestimates the chiral splitting of the former, and are not applicable even with renormalised parameters in the case where non-coherent reversal modes are possible.

Reductions and splitting of the switching barriers lead to exponentially enhanced transition rates especially in the limit of rare events, as we show with a modified Arrhenius law. Using the rates for the chiral transition channels as input for kMC simulations, we find that the evolution of an extended square ice proceeds much faster, and involves different spatial correlations when compared to a mean-field model.

The influence of the immediate environment on the nanomagnetic switching thus is a key ingredient to correctly model the relaxation dynamics of artificial spin ices, as well as of functional magnonic materials and small-scale circuits for computation. We therefore expect our results to be relevant to different communities making use of thermally-driven relaxation of interacting nanomagnets.

This work is structured into three sections: in section 1 a basic understanding is derived on how the magnetic environment can lead to chiral switching channels in artificial square ice. Section 2 compares point-dipole model predictions to micromagnetic simulations of nanomagnets of different dimensions and material parameters. In section 3, ramifications of the modified switching barriers on the switching rates of single nanomagnets and relaxation kinetics of extended artificial square ice are discussed.

1. Chiral moment reversal

Artificial spin ices are lithographically-designed magnetic metamaterials with identical nanomagnets arranged on lattices with different geometries [1–4]. In the following, we focus our discussion on the example of artificial square ice, in which nanomagnets are arranged on a square lattice with periodicity a , see figure 1(a).

Due to their shape anisotropy, each stadium-shaped nanomagnet with length l , width w , and thickness t is quasi-uniformly magnetised, and thus behaves like an Ising macrospin. Without applied field or interactions with neighbours, the magnetic moment will align with the long axis, i.e. to the left ($\leftarrow$) or right ($\rightarrow$) if considering the black nanomagnet in figure 1(a). To spontaneous switch between these energetically-degenerate configurations (without loss of generality from $\leftarrow$ to $\rightarrow$), the moment rotates to overcome a metastable state for which the net moment points along the nanomagnets' short axis, i.e. $\uparrow$ or $\downarrow$. The difference between the metastable and equilibrium micromagnetic energies E^{mm} gives the single-nanomagnet switching barrier ΔE_{sb} :

$$\Delta E_{\text{sb}} = E_{\downarrow}^{\text{mm}} - E_{\leftarrow}^{\text{mm}}. \quad (1)$$

The value of ΔE_{sb} depends on the size, shape and material of the individual elements [16–19]. For the assumption of uniform magnetisation, the barrier is given by the shape anisotropy $\Delta E_{\text{sb}}^{\text{shape}} = K^{\text{shape}}V$, with V being the volume of the nanomagnet. Values of K^{shape} are either tabulated for ellipsoidal geometries [20] or can be calculated via magnetostatic simulations [21, 22].

The mutual coupling between nanomagnets is given by magnetostatic interactions, which takes the following form between point-dipole moments $\mathbf{m}_i$ and $\mathbf{m}_j$ separated by a distance vector $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$:

$$E^{\text{dip}} = \frac{\mu_0}{4\pi|\mathbf{r}_{ij}|^3} \left[\mathbf{m}_i \cdot \mathbf{m}_j - 3 \frac{(\mathbf{m}_i \cdot \mathbf{r}_{ij})(\mathbf{m}_j \cdot \mathbf{r}_{ij})}{|\mathbf{r}_{ij}|^2} \right]. \quad (2)$$

In artificial square ice the strongest coupling, denoted by J_{NN} , occurs between nearest-neighbour nanomagnets at a 90° angle, see figure 1(a). Using the lattice periodicity a and the net moment $m = |\mathbf{m}| = M_{\text{sat}}V$ of a nanomagnet with volume V and material saturation magnetisation M_{sat} , we define a convenient energy scale $J_{\text{NN}}^{\text{dip}}$:

$$J_{\text{NN}}^{\text{dip}} = \frac{3}{2\sqrt{2}} \frac{\mu_0}{\pi} \frac{m^2}{a^3}. \quad (3)$$

Due to the pronounced distance dependence, $E^{\text{dip}} \propto r^{-3}$, the coupling between nanomagnets meeting at the vertex points highlighted in figure 1(a), is dominant over further-range interactions [23, 24]. Therefore, we investigate the switching barriers for moment reversal of a central nanomagnet under the influence of its closest neighbours only.

1.1. Switching environments

For an infinite artificial square ice the environment that influences the reversal of a nanomagnet forms a double-vertex configuration, as depicted in figure 1(b). Here, each tip of the central nanomagnet (black) is in close interaction to three other nanomagnet (gray), whose magnetisation remains largely unchanged during the reversal of the central nanomagnet. The extended square lattice then can be considered as an infinite tiling of these motifs.

We denote each magnetic equilibrium configuration with a state number $\#i$ determined by the arrangement of the surrounding nanomagnets, and the orientation of the central (switching) nanomagnet, which can point to the left ($\leftarrow$) or to the right ($\rightarrow$). The state number $\#i$ can be obtained from the binary representation of the relative configuration of the six surrounding nanomagnet numbered 1 to 6 according to the scheme shown in figure 1(b):

$$i = \sum_{j=1}^6 b_j 2^{j-1} \quad \text{with} \quad b_j = \begin{cases} 0 & \text{if moment points down or to left} \\ 1 & \text{if moment points up or to right.} \end{cases} \quad (4)$$

Half of the $2^6 = 64$ environment configurations of the double vertex are depicted in figure 2. The remaining states $\#i$ can be derived by applying a time reversal operation on the configurations $\#(2^6 - 1 - i)$.

1.2. Switching barriers from a point-dipole model

In the following, we discuss a perturbative approach to switching barriers in artificial square ice. Here, the single-nanomagnet barrier ΔE_{sb} , figure 1(d), is modified due to energy contributions arising from the interactions with the immediate magnetic environment.

To discuss a specific example, we focus on the fully-magnetised double-vertex environment $\#0$ depicted in figure 1(b). The energy of the configuration where the central moment points (exactly) to the left, $\leftarrow$, is lower than when it points to the right, $\rightarrow$, where three magnetic charges meet at each vertex point. Using simplified assumptions and symmetry arguments, one can derive a mean-field switching barrier $\langle \Delta E_i \rangle_{\leftarrow \rightarrow}^{\text{dip}}$

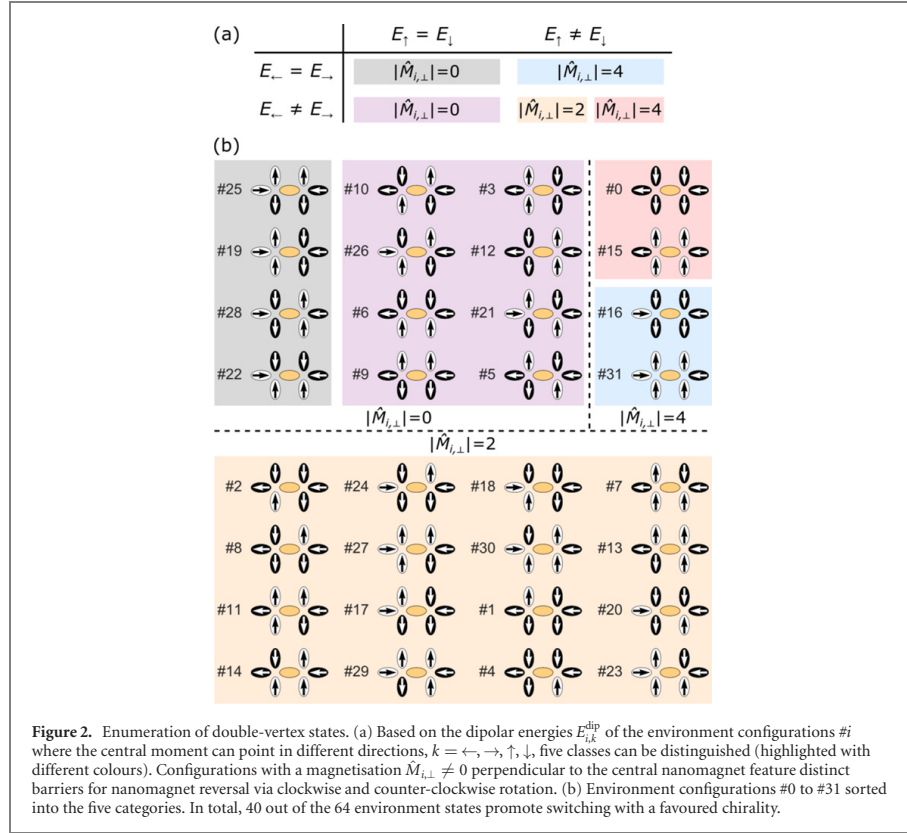


Figure 2. Enumeration of double-vertex states. (a) Based on the dipolar energies E_{ik}^{dip} of the environment configurations i where the central moment can point in different directions, $k = \leftarrow, \rightarrow, \uparrow, \downarrow$, five classes can be distinguished (highlighted with different colours). Configurations with a magnetisation $\hat{M}_{i,\perp} \neq 0$ perpendicular to the central nanomagnet feature distinct barriers for nanomagnet reversal via clockwise and counter-clockwise rotation. (b) Environment configurations #0 to #31 sorted into the five categories. In total, 40 out of the 64 environment states promote switching with a favoured chirality.

(as the average of the barriers for clockwise and counter-clockwise rotation, see appendix A in reference [15]). Its value depends solely on the energies of the equilibrium configurations, as indicated by the gray arrow in figure 1(d):

$$\langle \Delta E_i \rangle_{\leftarrow \rightarrow}^{\text{dip}} = \Delta E_{\text{sb}}^{\text{shape}} + \frac{1}{2} (E_{i,\rightarrow}^{\text{dip}} - E_{i,\leftarrow}^{\text{dip}}). \quad (5)$$

This mean-field barrier, however, is missing a crucial point, as independent relaxation pathways via clockwise and counter-clockwise rotation of the central nanomagnet need to be considered, i.e.

$$\Delta E_{i,\leftarrow \rightarrow}^{\text{dip}} = \Delta E_{\text{sb}}^{\text{shape}} + (E_{i,\uparrow}^{\text{dip}} - E_{i,\downarrow}^{\text{dip}}), \quad (6)$$

and

$$\Delta E_{i,\leftarrow \rightarrow}^{\text{dip}} = \Delta E_{\text{sb}}^{\text{shape}} + (E_{i,\downarrow}^{\text{dip}} - E_{i,\uparrow}^{\text{dip}}). \quad (7)$$

These barriers are not necessarily equivalent, as shown in figure 1(c): due to their staggered spatial arrangement, the central moment in a fully magnetised environment #0 will preferably align ferromagnetically with its neighbours, thus forming a head-to-tail flux-closure configuration which reduces the dipolar energy term in equation (2). Therefore, transitions via counter-clockwise rotations (blue) of the central nanomagnet will be largely favoured over those via clockwise rotations (red).

From the dipolar energies for all environmental states and orientation of the central moment, figure 3(a), one can obtain the respective clockwise and counter-clockwise switching barriers, figure 3(b). Under the assumption that $\uparrow$ and $\downarrow$ align perfectly along the short axis of the nanomagnets, the energy splitting between the configurations is symmetric around the mean-field barrier $\langle \Delta E_i \rangle_{\leftarrow \rightarrow}^{\text{dip}}$ (marked by crosses), and equals the three distinct values shown in figure 3(c).

Barrier splitting occurs for all environments which feature a perpendicular effective field $\hat{M}_{i,\perp}$ generated by the neighbouring nanomagnets, that acts on the central nanomagnet (indices b_j are as defined in

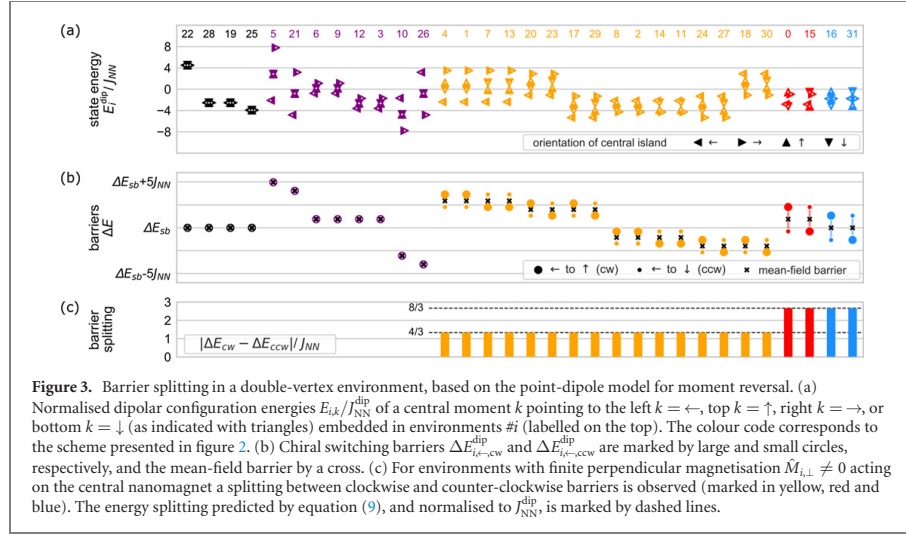


Figure 3. Barrier splitting in a double-vertex environment, based on the point-dipole model for moment reversal. (a) Normalised dipolar configuration energies $E_{i,k}^{dip}/J_{NN}^{dip}$ of a central moment k pointing to the left $k = \leftarrow$, top $k = \uparrow$, right $k = \rightarrow$, or bottom $k = \downarrow$ (as indicated with triangles) embedded in environments $\#i$ (labelled on the top). The colour code corresponds to the scheme presented in figure 2. (b) Chiral switching barriers $\Delta E_{i,←-cw}^{dip}$ and $\Delta E_{i,←-ccw}^{dip}$ are marked by large and small circles, respectively, and the mean-field barrier by a cross. (c) For environments with finite perpendicular magnetisation $\hat{M}_{i,\perp} \neq 0$ acting on the central nanomagnet a splitting between clockwise and counter-clockwise barriers is observed (marked in yellow, red and blue). The energy splitting predicted by equation (9), and normalised to J_{NN}^{dip} , is marked by dashed lines.

equation (4)):

$$\hat{M}_{i,\perp} = \sum_{j=1}^4 \frac{\mathbf{m}_j}{|\mathbf{m}_j|} = -\sum_{j=1}^4 (-1)^{b_j}. \quad (8)$$

The normalised perpendicular magnetisation $\hat{M}_{i,\perp}$ can take the values of 0 (black and purple in figures 2 and 3), ± 2 (orange), and ± 4 (red and blue) only. Thus, we can modify equation (5) to include an additional energy term:

$$\Delta E_{i,←-cw/ccw}^{dip} = \langle \Delta E_i \rangle_{←-to-→}^{dip} \mp \frac{J_{NN}^{dip}}{3} \hat{M}_{i,\perp}. \quad (9)$$

For a central moment initially pointing to the left ($\leftarrow$), the second term (derived in appendix A) is subtracted for clockwise, and added for counter-clockwise reversal.

In conclusion, with a simplified point-dipole model the switching barriers are modified by the choice of clockwise vs counter-clockwise rotation, if the moment interacts with an effective perpendicular stray field generated by its environment. As shown in figure 2, 40 out of the 64 double-vertex configurations of artificial square ice feature a finite perpendicular magnetisation $\hat{M}_{i,\perp} \neq 0$ acting on the central nanomagnet. In particular, we expect a maximum chiral barrier splitting for the fully-magnetised environments (marked in red), which are commonly-used initial states for thermal relaxation studies of artificial spin ice [7].

The conclusions of this section are of general validity for arrays of interacting nanomagnets. The calculation of switching barriers for clockwise and counter-clockwise transitions can be easily adapted to other moment configurations. As an example, in supplementary material (<https://stacks.iop.org/NJP/23/033024/mmedia>) figure 1 we show that chiral barrier splitting occurs in over 60% of the moment configurations of artificial kagome spin as well.

The existence of separate chiral switching channels is by no means a curiosity, since nanomagnetic switching will occur predominantly via the more favourable pathway. We thus expect profound consequences on the switching rates and transition kinetics when taking into account the chiral splitting.

2. Micromagnetic switching

The point-dipole switching barriers represent a perturbative approach parametrised by two parameters only: first, ΔE_{sb} describes the switching barrier of an isolated nanomagnet, and implicitly depends on its shape and size [20]. Second, J_{NN}^{dip} quantifies the energies of equilibrium configurations due to the interactions between nanomagnets placed on the square lattice, and modify the switching barrier of individual nanomagnets. The mean-field energy barrier in equation (9), however, does not take into account possible non-coherent moment reversal. In particular, it does not describe the influence of material parameters such as the saturation magnetisation M_{sat} and the exchange strength A_{ex} , thermal fluctuations, and the magnetic environment. Due to these effects the net moment can be dynamically reduced during reversal, e.g. via non-uniform buckling modes, vortex creation, or domain formation [25–28].

To have a nuanced look on how the energy barrier depends on (1) the material parameters, (2) the nanomagnet shape and size, and (3) the interactions with neighbouring nanomagnets, we now turn to a full micromagnetic simulation of the reversal barriers.

2.1. Implementation of string-method simulations

Contrary to simulations employing the Landau–Lifschitz–Gilbert equations to explicitly solve the time-dependent evolution of the nanomagnetic reversal, we determined the associated energy barrier using a time-independent string method. In this approach, starting from a coherent moment reversal, the moment configurations are iteratively optimised to a minimum-energy path through configuration space, and thus yield the lowest energy barrier associated to that reversal process. As in our previous work [15], which also discusses further simulation details, we implement here the *simplified and improved string method* [29] using the finite-element micromagnetic code *magnum.fe* [30]. We consider two artificial square ice geometries with distinct choices for M_{sat} and A_{ex} , representing different regimes. Meshes discretising the considered geometries, i.e. an individual nanomagnet and the double-vertex configurations, were created with the software *gmsh* [31].

First, we consider a geometry largely dominated by exchange interactions, which favour a coherent reversal, and thus may resemble the macrospin model derived in section 1: nanomagnets with dimensions $l \times w \times t = 150 \text{ nm} \times 100 \text{ nm} \times 3 \text{ nm}$ are placed on a square lattice with periodicity $a = 240 \text{ nm}$. The material parameters $M_{\text{sat}} = 790 \text{ kA m}^{-1}$ and $K = 0$ correspond to bulk permalloy ($\text{Fe}_{0.2}\text{Ni}_{0.8}$) values at 300 K. The exchange stiffness $A_{\text{ex}} = 13 \text{ pJ m}^{-1}$ was obtained from a temperature-dependent scaling [32–35]

$$A_{\text{ex}} = A_{\text{ex}}(0) \left(\frac{M_{\text{sat}}}{M_{\text{sat}}(0)} \right)^{1.7}, \quad (10)$$

where $M_{\text{sat}}(0) = 950 \text{ kA m}^{-1}$ and $A_{\text{ex}}(0) = 18 \text{ pJ m}^{-1}$ denote the respective permalloy bulk parameters at 0 K.

Second, we consider a system for which we expect sizable magnetostatic effects leading to non-uniform magnetic configurations during reversal: Nanomagnets with dimensions $l \times w \times t = 470 \text{ nm} \times 170 \text{ nm} \times 3 \text{ nm}$ are placed on a square lattice with periodicity $a = 600 \text{ nm}$. This geometry, or choices close to it, have been used in several experimental studies [7, 13, 36, 37]. The saturation magnetisation $M_{\text{sat}} = 350 \text{ kA m}^{-1}$ corresponds to the value given in [7], and from the scaling in equation (10) we obtain $A_{\text{ex}} = 3.25 \text{ pJ m}^{-1}$. Although the saturation magnetisation is lowered significantly, the exchange stiffness is even more reduced, and thus we expect the energetics dominated by magnetostatic interactions. We again assume a vanishing magnetocrystalline anisotropy, $K = 0$.

2.2. Influence of environment on reversal

We select representative environment states for each of the five classes introduced figure 2(b), for which we expect no (black and purple), intermediate (yellow), and high barrier splitting (red and blue), respectively. For these states, shown at the top of figure 4, we compare the chiral switching barriers obtained from a macrospin approximation (left) and micromagnetic string-method simulations (right) for the two different square ice geometries (schematics on the left are shown to scale). Clockwise and counter-clockwise barriers are marked in red and blue, respectively, and the difference (barrier splitting) is plotted as black bars.

The macrospin model considers variation of the single-moment barrier ΔE_{sb} (calculated from the shape anisotropy of a uniformly-magnetised nanomagnet $\Delta E_{\text{sb}}^{\text{shape}} = K^{\text{shape}} V$) due to point-dipole-like interactions quantified by $J_{\text{NN}}^{\text{dip}} \propto (M_{\text{sat}} V)^2 / a^3$, as derived in section 1.2. We also plot the mean-field barrier from equation (5) (black horizontal line).

The chiral barriers from the string-method simulations are calculated from the energy difference between the micromagnetic net energies of the metastable barrier configuration ($\uparrow, \downarrow$) and the initial configuration ($\leftarrow$):

$$\Delta E_{i \leftarrow, \text{cw}}^{\text{mm}} = E_{i \uparrow}^{\text{mm}} - E_{i \leftarrow}^{\text{mm}}, \quad (11)$$

$$\Delta E_{i \leftarrow, \text{ccw}}^{\text{mm}} = E_{i \downarrow}^{\text{mm}} - E_{i \leftarrow}^{\text{mm}}. \quad (12)$$

For the exchange-dominated square-ice geometry, figures 4(a)–(e), the mean-field barrier always overestimates the lower of the two micromagnetic barriers (for the chosen cases corresponding to counter-clockwise reversal marked in blue), as already discussed in our previous work [15]. Compared to the point-dipole model, micromagnetic simulations consistently give lower chiral switching barriers. The difference of the barrier energies, i.e. the chiral splitting, is enhanced, and remains proportional to the

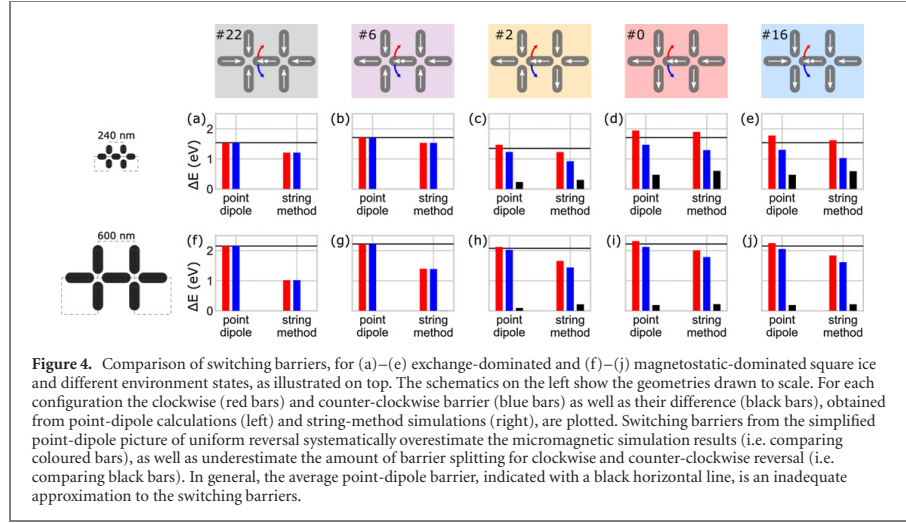


Figure 4. Comparison of switching barriers, for (a)–(e) exchange-dominated and (f)–(j) magnetostatic-dominated square ice and different environment states, as illustrated on top. The schematics on the left show the geometries drawn to scale. For each configuration the clockwise (red bars) and counter-clockwise barrier (blue bars) as well as their difference (black bars), obtained from point-dipole calculations (left) and string-method simulations (right), are plotted. Switching barriers from the simplified point-dipole picture of uniform reversal systematically overestimate the micromagnetic simulation results (i.e. comparing coloured bars), as well as underestimate the amount of barrier splitting for clockwise and counter-clockwise reversal (i.e. comparing black bars). In general, the average point-dipole barrier, indicated with a black horizontal line, is an inadequate approximation to the switching barriers.

perpendicular moment $|\hat{M}_{i,\perp}|$ generated by the environment. The reversal process is still governed by an almost-coherent rotation of the central moment (appendix C.1). Therefore, one can obtain reasonable switching barriers from the perturbative decomposition of equation (9) by using a lower single-moment barrier $\Delta E_{\text{sb}}^{\text{string}}$ and stronger interactions $J_{\text{NN}}^{\text{mm}}$ (appendix C.2).

For the magnetostatic-dominated square-ice geometry, figures 4(f)–(j), the differences are even more pronounced. This is because the reversal behaviour is no longer uniform, as discussed in appendices C.1 and C.2. In particular, environments with vanishing perpendicular magnetisation $\hat{M}_{i,\perp} = 0$ (black and purple) non-coherent reversal modes are promoted by *C*– or *onion*–like magnetic microstates in the central switching nanomagnet [25–28]. This leads to large reductions of the switching barrier, e.g. more than -50% in the case of state #22 in figure 4(f), when compared to the point-dipole model. In contrast, environments with $M_{\perp} \neq 0$ favour S-like micromagnetic configurations associated with quasi-coherent reversal [38–40]. Further details about the micromagnetic switching are discussed in appendix C and supplementary figure 2.

In general, the point-dipole barriers overestimate all micromagnetic barriers by a considerate margin, and underestimate the chiral barrier splitting, as is evident when comparing the black bars shown in figures 4(h)–(j). For environments with a finite perpendicular magnetisation the barrier splitting is of similar magnitude, figures 4(h)–(j), and thus does not follow the proportionality $|E_{i,\uparrow} - E_{i,\downarrow}| \propto |\hat{M}_{i,\perp}|$. Therefore, the simplified decomposition of switching barriers according to equation (9) is no longer valid when the moments can switch via non-coherent reversal mechanisms.

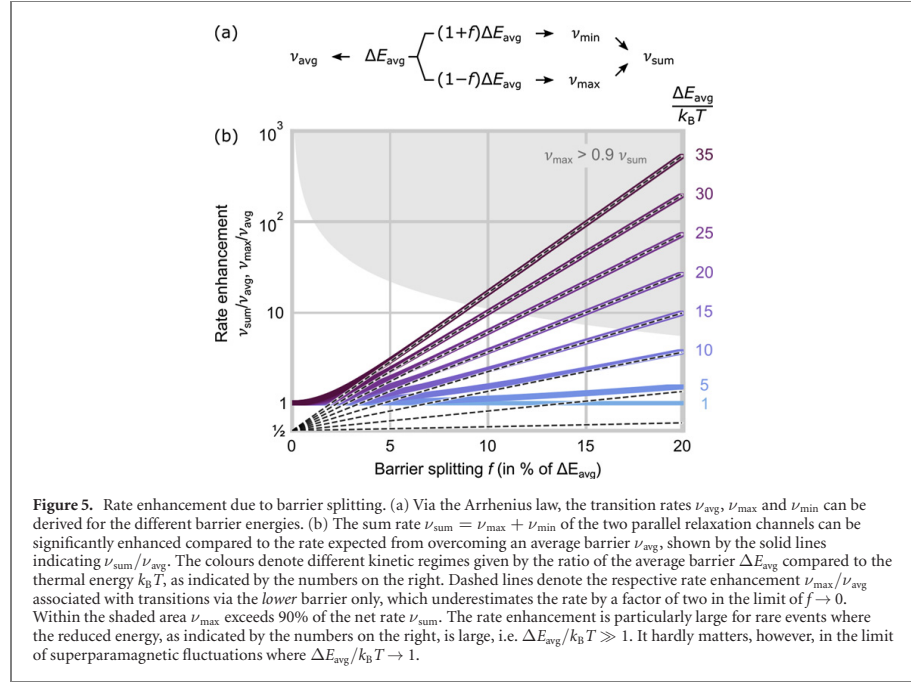
In general, by taking into account the micromagnetic nature of the moment reversal, we observe both a reduction of the switching barriers as well as an enhanced separation of the chiral barriers for environments with $\hat{M}_{i,\perp} \neq 0$. These results hint to the fact that barrier reductions often utilised in simplified models to simulate the evolution of artificial spin ices [7, 8], and which are often attributed to *extrinsic* causes such as nanofabrication imperfections, could be—at least partially—associated with *intrinsic* barrier splitting and incoherent reversal modes.

As lower barriers are easier to overcome for thermally-induced reversal, we therefore expect significant enhancement of the kinetics of artificial square ice, which may yield different relaxation time scales and emergent correlations.

3. Transition kinetics

The effect of barrier splitting on the net transition rate can be generalised by using an average barrier

$$\Delta E_{\text{avg}} = \frac{1}{2} |\Delta E_{i,\uparrow,\text{cw}} + \Delta E_{i,\uparrow,\text{ccw}}|, \quad (13)$$



and a factor f , which describes the symmetric splitting of the clockwise and counter-clockwise barriers around the average barrier, as depicted in figure 5(a),

$$f = \frac{1}{2} \frac{|\Delta E_{i \leftarrow \rightarrow, \text{cw}} - \Delta E_{i \leftarrow \rightarrow, \text{ccw}}|}{\Delta E_{\text{avg}}}. \quad (14)$$

For the barriers derived from the point-dipole picture in section 1.2, the average barrier corresponds to the mean-field barrier of equation (5), i.e. $\Delta E_{\text{avg}}^{\text{dip}} = \langle \Delta E_i \rangle_{\leftarrow \rightarrow}^{\text{dip}}$. The splitting factor vanishes, $f = 0$, for environmental states without a perpendicular magnetisation $\hat{M}_{i,\perp} = 0$ acting on the switching moment. From the results of micromagnetic simulations discussed above we obtain non-zero values of f between a few percent up to about 20%.

3.1. Modified Arrhenius law for barrier splitting

The temperature-dependent transition rate for spontaneous switching over an average energy barrier ΔE_{avg} can be obtained via the Arrhenius law [41, 42], with the attempt frequency ν_0 and the Boltzmann constant $k_B = 8.62 \times 10^{-5} \text{ eV K}^{-1}$:

$$\nu_{\text{avg}}(\Delta E_{\text{avg}}, T) = 2\nu_0 \exp\left(-\frac{\Delta E_{\text{avg}}}{k_B T}\right). \quad (15)$$

The attempt frequency ν_0 depends on the shape, size, and material of the nanomagnets. Typical values of ν_0 are in the order of $10^9 \dots 10^{12} \text{ Hz}$ [42, 43], and even faster time scales have been discussed [44, 45].

We need to consider the clockwise and counter-clockwise reversal as parallel and independent channels of relaxation, and the rates associated to each of the two barrier energies, i.e. $\Delta E_{\text{avg}}(1-f)$ and $\Delta E_{\text{avg}}(1+f)$, need to be added to obtain an effective transition rate. Therefore, for the definition of the rate in equation (15), we explicitly included a pre-factor of two, to account for degenerate clockwise and counter-clockwise relaxation channels over the average barrier.

Due to the pronounced non-linearity of the Arrhenius law, the summation of rates leads to an effective increase of the net transition rate ν_{sum} of thermally-activated switching when compared to the rate ν_{avg} associated with the average barrier (derivation in appendix B):

$$\nu_{\text{sum}} = \nu_{\text{avg}}(\Delta E_{\text{avg}}, T) \cosh\left(f \frac{\Delta E_{\text{avg}}}{k_B T}\right). \quad (16)$$

The ratio of the joint rate compared to the average-barrier rate, i.e. $\nu_{\text{sum}}/\nu_{\text{avg}}$, are compared in figure 5(b) for different splitting ratios f and reduced energies $\Delta E_{\text{avg}}/k_{\text{B}}T$. The exponential rate enhancement is particularly pronounced in the limit of rare events with $\Delta E_{\text{avg}}/k_{\text{B}}T \gg 1$ where a splitting of barriers can increase the (albeit low) transition rates by several orders of magnitude (purple lines). In contrast, in the limit of superparamagnetic fluctuations, i.e. $\Delta E_{\text{avg}}/k_{\text{B}}T \rightarrow 1$, barrier splitting increases the net rate only moderately (blue lines).

For the assumption that transitions occur predominantly via the lower barrier *only*, we have to consider the transition associated to the smaller barrier, i.e. $(1-f)\Delta E_{\text{avg}}$, see dashed lines in figure 5 giving the ratio $\nu_{\text{max}}/\nu_{\text{avg}}$. For high splitting ratios f and $\Delta E_{\text{a}}/(k_{\text{B}}T) \gg 1$ the rate ν_{max} will approach ν_{sum} , as transitions by the higher-lying barrier become irrelevant. The shaded area of figure 5 marks where ν_{max} exceeds 90% of the value ν_{sum} . Within this regime, transitions via the lower-lying barrier might be a good approximation of the net reversal rate. In the limit of $f \rightarrow 0$, however, where both barriers are equal, using the rate ν_{max} will underestimate the net rate by a factor of two, i.e. $(\nu_{\text{max}}/\nu_{\text{sum}})_{f \rightarrow 0} = 1/2$.

In the case of artificial square ice, and depending on the kinetic regime given by the relation between the energies ΔE_{sb} , J_{NN} , and $k_{\text{B}}T$, approximating the transition rates via the mean-field barrier [7, 8, 13, 37] or the minimum barrier [46, 47] thus may significantly underestimate the speed of evolution.

3.2. Temporal evolution of extended square ice

To illustrate the consequences of barrier splitting, we now turn to the evolution of extended square-ice arrays. In many thermal relaxation studies of artificial spin ice, the main interest lies in the onset of phase transitions and formation of emergent correlations. In many experiments, the system evolves from a field-set fully magnetised state, for which we predict a particularly strong barrier splitting. We therefore expect that the initial demagnetisation of a magnetic-field-saturated artificial square ice array will be particularly affected by the modified transition kinetics.

To model the relaxation, kMC simulations are often employed [48–50]. The kMC algorithm provides a numerical solution to the master equation, which is a system of linear differential equations describing the evolution of the probabilities for Markov processes in systems that jump from one state to another in continuous time [51]. Using this method, both the equilibrium expectation values of populations and their dynamical evolution during a thermalization process can be retrieved.

In this work, kMC simulations are performed using a custom-written code [52], with a system of 50×50 moments and periodic boundary conditions. The initial configuration is uniformly-magnetised, with the net magnetisation being parallel to a diagonal direction of the array. The demagnetisation due to spontaneous moment reversals is tracked over time for 125×10^3 kMC steps, and averaged over 20 individual simulation runs. As an illustrative example, we here consider the effect of symmetric barrier splitting only, and discount further barrier reductions associated to non-uniform reversal. Thus, we use the point-dipole energy barriers from figure 3(b) with parameters $J_{\text{NN}}^{\text{dip}} = 0.178$ eV and $\Delta E_{\text{sb}} = 1.327$ eV (i.e. using values for the exchange-dominated square-ice geometry), and calculate the environment-dependent transition rates ν_{avg} and ν_{sum} at a temperature of $T = 300$ K as input parameters for the kMC simulations.

Figure 6(a) compares the time evolution of the net magnetisation of square ice for rates from the mean-field barriers (dashed black line) to the model taking into account chiral barrier splitting (solid red line). The time is measured in multiples of the inverse attempt frequency, $\tau = \nu_0^{-1}$. We find that the onset of demagnetisation for the split-barrier model (red) happens two orders of magnitude earlier than for the average-barrier model (black). In the case of the average-barrier model, the demagnetisation involves bouts of rapid evolution interrupted by phases with little change, indicating avalanche-like dynamics [53–56]. In contrast, the split-barrier model shows a smooth demagnetisation, with a rate (solid thick lines indicate evolution from 90% to 50%) which is about three orders of magnitude faster compared to the mean-barrier model.

When assessing the emergent spatial correlations, we find that the evolution for the mean-field model is governed by the propagation of strings of ground-state vertices (in blue) wrapping the system (due to the periodic boundary conditions), as shown in figure 6(b). The final state has a magnetisation of about 16% of its initial value. The snapshots of spatial configuration of the split-barrier model at $M = 0.9$ and $M = 0.5$ (with M normalised to the initial field-set magnetisation) in figure 6(c) appear somewhat similar to that of the mean-field case. There are more possible transitions for the system to explore, and we would like to point out that this model recovers the separation of strings observed in experiments, but does not require to invoke external disorder e.g. via variation of the switching barriers as done in [8]. The final state of the evolution corresponds to a multi-domain state with almost vanishing magnetisation, $M \approx 0$.

Thus, our kMC results show that the modified hierarchy of transition barriers due to the chiral barrier splitting may have subtle, but relevant, consequences: in certain cases, the kinetic relaxation pathways are

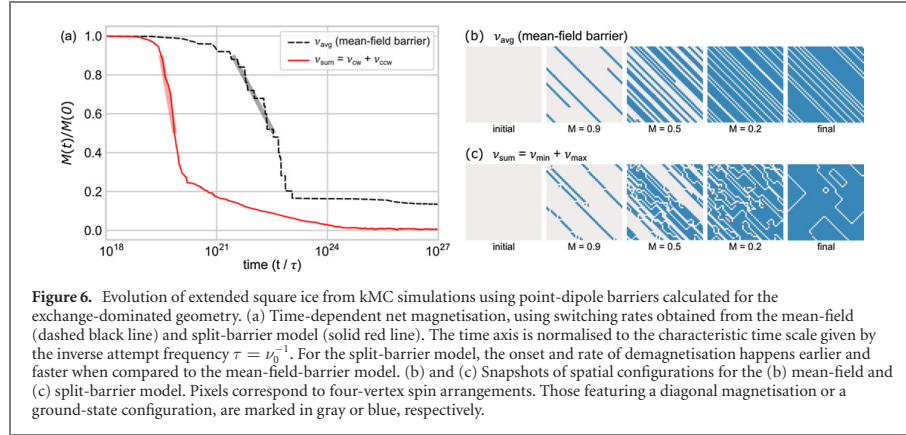


Figure 6. Evolution of extended square ice from kMC simulations using point-dipole barriers calculated for the exchange-dominated geometry. (a) Time-dependent net magnetisation, using switching rates obtained from the mean-field (dashed black line) and split-barrier model (solid red line). The time axis is normalised to the characteristic time scale given by the inverse attempt frequency $\tau = \nu_0^{-1}$. For the split-barrier model, the onset and rate of demagnetisation happens earlier and faster when compared to the mean-field-barrier model. (b) and (c) Snapshots of spatial configurations for the (b) mean-field and (c) split-barrier model. Pixels correspond to four-vertex spin arrangements. Those featuring a diagonal magnetisation or a ground-state configuration, are marked in gray or blue, respectively.

not simply dictated by equilibrium-energy arguments. This will modify the emergence of spatial correlations, which needs to be explored in a systematic study and compared to experimental results [1, 9, 10, 53, 57–64].

4. Conclusions

To realistically model the temporal evolution of artificial spin ices or small-scale nanomagnetic circuits it is necessary to know the switching barriers for the single-moment reversal. In this work, we quantified how magnetostatic interactions with neighbouring nanomagnets modify the switching barriers in artificial square and kagome ice in absence of *extrinsic* effects such as defects or spurious fields. We found that for environments which feature a finite perpendicular magnetic field acting on the switching nanomagnet clockwise and counter-clockwise moment reversals need to be considered independently. The resulting barrier splitting can be sizeable. In the case of exchange-dominated nanomagnets supporting coherent rotation modes, the splitting can be predicted from a modified point-dipole model. Taking into account the finite size of the nanomagnets and the influence of material parameters, further barrier reductions were obtained from micromagnetic simulations. These reductions are particularly strong for magnetostatically-dominated nanomagnets embedded in environments that do not promote reversal via a distinct chiral switching channel.

The splitting and reduction of transition barriers exponentially increase the transition rates when compared to a mean-field average barrier. Depending on the dynamical regime, which depends on the relationship between the average energy barrier, barrier splitting and temperature, we found that transition rates are especially enhanced in the limit of rare events. We modelled the evolution of extended artificial square ice with kMC simulations, and compared a mean-field model with the model that takes into account the barrier splitting. We found that the onset and speed of evolution is largely enhanced in the latter case. Furthermore, while mean-field barriers are solely dictated by equilibrium-energy arguments, the chiral switching barriers depend on the kinetics of reversal. Thus, more and different relaxation pathways are accessible, which modifies the emergent spatial correlations and routes towards the ground state.

Our results are a step towards a deeper understanding of the single-moment switching of nanomagnetic systems, highlighting how faster time scales of relaxation can be caused via *intrinsic* interactions with the magnetic environment. These findings are relevant to the field of artificial spin systems, and can be extended from square ice to other moment configurations, such as kagome ice [6, 53, 54, 65], and square-ice-like tetris, shakti, and brickwork lattices featuring asymmetric moment coordinations [66–68]. We also expect that these concepts are relevant for the utilisation of magnetic metamaterials for magnonics [39, 69–74] and nanomagnetic computation [13, 37, 47, 75, 76].

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

Appendix A. Derivation of point-dipole barriers

We assume that all moments are strictly parallel ($\leftarrow$, $\rightarrow$) to the nanomagnet long axis, and that the environment nanomagnets remain static during the reversal of the central moment. We furthermore assume that the configuration of highest energy correspond to those with perpendicular central moment ($\uparrow$, $\downarrow$). These approximations are valid for weak interactions only, and in general are a gross oversimplification, as due to the pairwise couplings the macrospins may rotate away from the local symmetry axis. This would result e.g. in non-symmetric splitting for clockwise and counter-clockwise transitions (i.e. $\Delta\phi \neq \pi$). Nevertheless, the strict limitation of moment direction allows to employ the anti-symmetry of the dipolar interaction energy under moment rotations of π ,

$$E^{\text{dip}}(\phi + \pi) = -E^{\text{dip}}(\phi). \quad (\text{A.1})$$

The mean-field barrier $\langle \Delta E_i \rangle_{\leftarrow \rightarrow}^{\text{dip}}$ is the average of clockwise and counter-clockwise energy barriers. Under the above assumptions, and as derived in appendix B of reference [15], it is determined by ΔE_{sb} and the energy difference between the equilibrium states before and after switching. It does not depend, however, on the energies of the intermediate high-energy configuration.

The barrier splitting $\Delta E_{\text{split}}^{\text{dip}}$ between clockwise and counter-clockwise rotation, see equation (9), can be calculated from the energy difference of the high-energy states. Using the anti-symmetry argument of equation (A.1), i.e. $E_{i,\uparrow}^{\text{dip}} = -E_{i,\downarrow}^{\text{dip}}$, one obtains

$$\Delta E_{\text{split}}^{\text{dip}} = \left| E_{i,\uparrow}^{\text{dip}} - E_{i,\downarrow}^{\text{dip}} \right| = 2 \left| E_{i,\uparrow}^{\text{dip},*} \right|. \quad (\text{A.2})$$

Here, $E_{i,\uparrow}^{\text{dip},*}$ takes into account the dipolar interaction terms with the central moment only as couplings between other moments remain unchanged by the reversal, and thus fall out of the energy difference.

As shown in figure A.1(b), in the high-energy state interactions with the horizontal nanomagnets, i.e. moments 5 and 6 in figure 1(b), will vanish as $J_{\parallel}^* = 0$. Due to the staggered arrangement of the moments, a ferromagnetic head-to-tail alignment of the central nanomagnet to its perpendicular neighbours is favourable, whereas the opposite orientation is penalised, see figure 1(c). Thus, only the net perpendicular magnetisation $\hat{M}_{i,\perp} = \sum_{j=1}^4 (-1)^{b_j}$ is relevant to the splitting. With a modified pair-wise interaction $J_{\text{NN}}^{\text{dip},*} = J_{\text{NN}}^{\text{dip}}/3$ one obtains

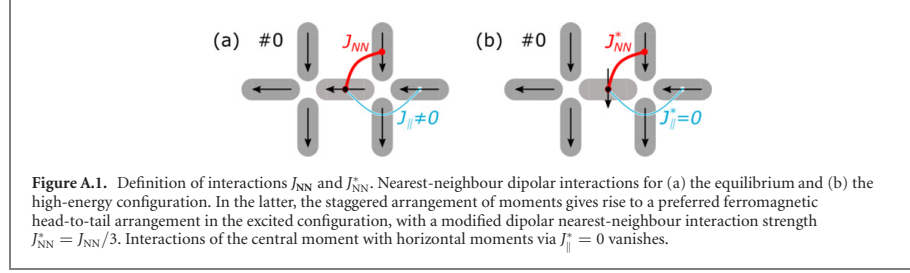
$$\Delta E_{\text{split}}^{\text{dip}} = \frac{2}{3} J_{\text{NN}}^{\text{dip}} \hat{M}_{i,\perp}. \quad (\text{A.3})$$

Appendix B. Arrhenius law for barrier splitting

If the transition barriers split symmetrically by a fraction f around the average barrier ΔE_{avg} to values $\Delta E_{\text{cw/ccw}} = (1 \pm f)\Delta E_{\text{avg}}$, the joint effective rate $\nu_{\text{sum}} = \nu_{\text{cw}} + \nu_{\text{ccw}}$ can be expressed as follows:

$$\nu_{\text{sum}} = \nu_{\text{cw}} + \nu_{\text{ccw}} \quad (\text{B.1})$$

$$= \nu_0 \left[e^{-(1+f)C} + e^{-(1-f)C} \right] \quad (\text{B.2})$$



$$= \nu_0 e^{-C} (e^{-fC} + e^{+fC}) \quad (\text{B.3})$$

$$= 2\nu_0 e^{-C} \cosh(fC) \quad (\text{B.4})$$

$$\nu_{\text{sum}} = \nu_{\text{avg}}(\Delta E_{\text{avg}}, T) \cosh\left(f \frac{\Delta E_{\text{avg}}}{k_B T}\right). \quad (\text{B.5})$$

Here, $C = \Delta E_{\text{avg}}/(k_B T)$ denotes the reduced average switching energy barrier. We assume that the attempt frequencies ν_0 are independent of the energy of the saddle point, i.e. $\nu_0^{\text{cw}} = \nu_0^{\text{ccw}} = \nu_0$. The transition rate $\nu_{\text{avg}}(\Delta E_{\text{avg}}, T)$ is defined in equation (15).

The maximum of the clockwise and counter-clockwise switching rates is associated to the lower-lying energy barrier $(1-f)\Delta E_{\text{avg}}$. In the limit of $f \rightarrow 0$, ν_{max} is a factor of two smaller than the rate $\nu_{\text{sum}}(f=0)$, and approaches the value of ν_{sum} for large splitting f or large reduced energy $C \gg 1$:

$$\nu_{\text{max}} = \max(\nu_{\text{cw}}, \nu_{\text{ccw}}) \quad (\text{B.6})$$

$$= \nu_0 e^{-C(1-f)} = \nu_0 e^{-C} e^{fC} \quad (\text{B.7})$$

$$= \nu_{\text{sum}} (1 + e^{-2fC})^{-1} \quad (\text{B.8})$$

$$= \begin{cases} \frac{1}{2} \nu_{\text{sum}} & \text{for } f \rightarrow 0 \\ \nu_{\text{sum}} & \text{for } fC \gg 1 \end{cases}. \quad (\text{B.9})$$

Appendix C. Additional simulation results

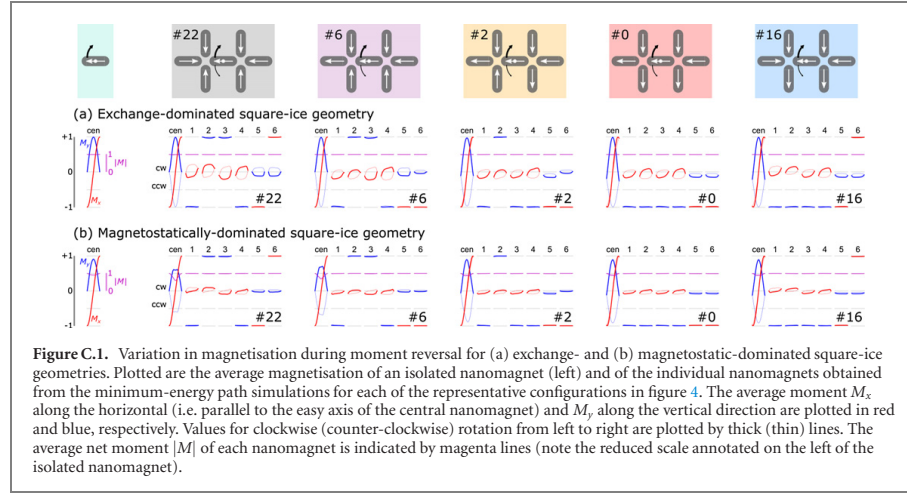
C.1. Magnetisation during reversal

To quantify the uniformity of the magnetic reversal, figure C.1 shows the averaged moments for a single (non-interacting) nanomagnet and each of the representative double-vertex configurations presented in figure 4. Here, the average magnetisation of each nanomagnet is plotted for every step of the string-method minimum-energy path. The horizontal coordinate roughly corresponds to the rotation angle ϕ of the central moment, with end points denoting initial and final equilibrium states.

In general, we find that in equilibrium the net moment $|\mathbf{M}|$ (magenta lines) is very close to one. Therefore, the static nanomagnets assume an almost saturated configuration, with the magnetisation largely aligned with the long axis of the nanomagnet and limited edge bending (M_x , red lines). Interactions with neighbouring moments, however, can induce sizeable perpendicular moment contributions ($M_x \perp M_y$, blue lines) in environments that feature a finite perpendicular magnetisation $\hat{M}_{i,\perp} \neq 0$, i.e. configurations #0, #2, and #16.

In the case of the geometry with small nanomagnets dominated by exchange interactions the magnitude $|\mathbf{M}|$ remains largely constant, figure C.1(a). The reversal thus represents a quasi-uniform rotation of the central moment. During reversal, the magnetisation components of the neighbouring nanomagnet can vary, allowing the system to evolve via the most efficient pathway.

For the geometry with large nanomagnets dominated by magnetostatic energy, shown in figure C.1(b), the switching of the non-interacting nanomagnet (left) involves a reduction of the net moment to 91%, and thus does not conform to a uniform moment rotation. For environment states #0, #16, and #2 with $\hat{M}_{i,\perp} \neq 0$ the reduction of net magnetisation is similar to that of an individual nanomagnet. In contrast, for environment states #6 and #22, with $\hat{M}_{i,\perp} = 0$, we observe a pronounced reduction of magnetisation in the high-energy configuration to less than 70% of the net moment. This indicates reversal via non-coherent modes, which are favoured to occur in nanomagnets that conform to C- or onion-like magnetic



microstates promoted by the order of the surrounding nanomagnets [38–40]. This leads to a reduction of the switching barrier as well, as discussed in section 2. The magnetic configuration of neighbouring nanomagnets varies less during reversal when compared to the small-island geometry. This is because the relative volume fraction of the large magnets meeting at the vertex point, where the spin structure varies the most, is smaller.

C.2. Micromagnetic barriers

The switching barriers obtained from micromagnetic string-method simulations for the environment states #0–#31 are summarised in figure C.2 with (a)–(c) showing the results for the exchange-dominated, and (d)–(f) the magnetostatic-dominated geometry.

We compare the clockwise and counter-clockwise barriers, large and small circles in figures C.2(a) and (d), to a *modified* mean-field model. The predictions are based on equations (5) and (9), but instead of energies derived from point-dipole calculations we use those obtained from micromagnetic simulations, as follows:

First, the switching barrier ΔE_{sb} of an isolated nanomagnet simulated with the string-method is used, as opposed to the shape anisotropy calculated for a uniformly-magnetised nanomagnet. For the exchange-dominated nanomagnet geometry we obtain $\Delta E_{sb}^{\text{shape}} = 1.540$ eV compared to $\Delta E_{sb}^{\text{string}} = 1.327$ eV, which is a reduction of -14%. For the magnetostatic-dominated nanomagnet geometry, the barrier reduction is even bigger, with $\Delta E_{sb}^{\text{shape}} = 2.153$ eV and $\Delta E_{sb}^{\text{string}} = 1.691$ eV (-22%), as the reversal is no longer coherent (see figure C.1(b)).

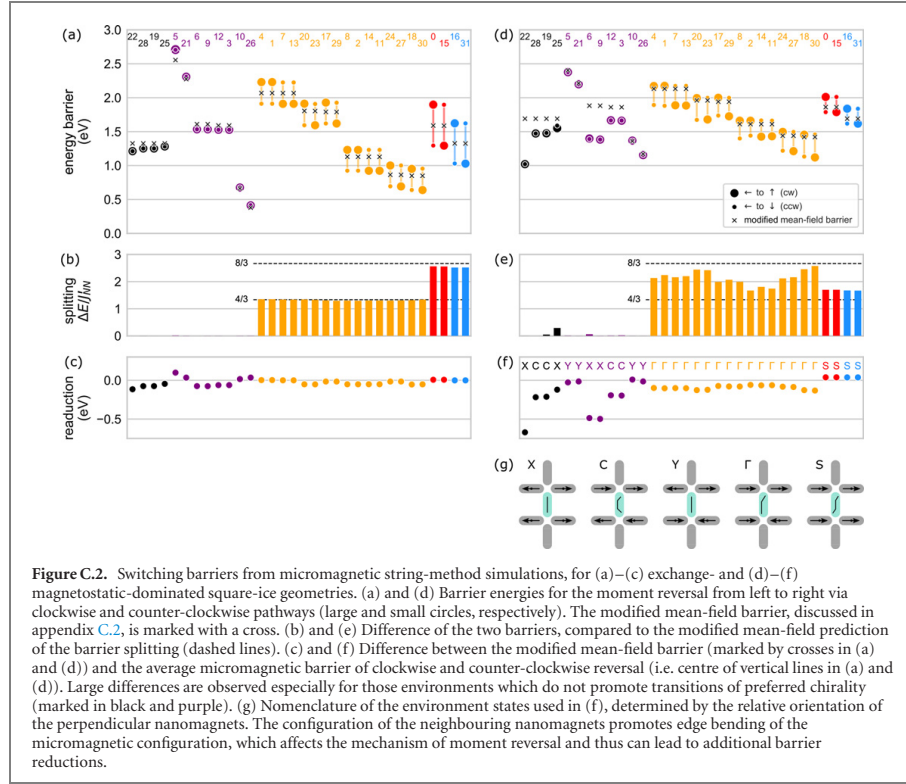
Second, $E_{i \leftarrow}^{\text{mm}}$ and $E_{i \rightarrow}^{\text{mm}}$ correspond to the micromagnetic equilibrium energies of the static configurations. Together with $\Delta E_{sb}^{\text{string}}$, a modified mean-field barrier can be calculated from equation (5) (crosses in figures C.2(a) and (d)).

Third, to estimate the barrier splitting, the nearest-neighbour interaction J_{NN}^{mm} is rescaled. The rescaling is motivated by the point-dipole model, which predicts an energy difference of $(16 - \frac{24\sqrt{5}}{125})J_{NN}^{\text{dip}}$ between the lowest-lying ground state and highest monopole state:

$$J_{NN}^{\text{mm}} = \frac{E_{\text{max}}^{\text{mm}} - E_{\text{min}}^{\text{mm}}}{16 - \frac{24\sqrt{5}}{125}}. \quad (\text{C.1})$$

For the exchange-dominated small-island geometry we find that the modified mean-field barrier gives a passable estimate for the average switching barrier, as the small differences in figure C.2(c) show. The chiral barrier splitting is well-described by the rescaled energy J_{NN}^{mm} , albeit with a small reduction for fully-magnetised environments marked red and blue in figure C.2(c).

For the magnetostatic-dominated reversal in the large-island geometry, figures C.2(d)–(f), the mean-field approach fails: the barrier splitting is both overestimated for environments with $|\hat{M}_{i,\perp}| = 4$ (red and blue) as well as underestimated in the case of $|\hat{M}_{i,\perp}| = 2$ (yellow). In particular, the mean-field



barrier predictions fails for environments with $|\hat{M}_{i,\perp}| = 0$ (black and purple), where reversal via non-uniform modes are favoured, as discussed before. The reductions compared to the mean-field barrier seem to be particularly strong for environments which feature ‘X’ and ‘C’ configurations of the perpendicular moments, see figures C.2(f) and (g). The symmetry breaking by the environment promotes so-called *onion* or *C*— microstates in the central nanomagnet, respectively [38–40]. These states promote non-coherent reversal, as illustrated in supplementary material figure 2, and a reduction of the switching barrier, as discussed in figure C.1 and appendix C.1.

With the exception of $\Delta E_{\text{sb}}^{\text{string}}$, which is a result of the string-method simulation, the energies $E_{i,\leftrightarrow}^{\text{mm}}$ and $J_{\text{NN}}^{\text{mm}}$ can be obtained from static equilibrium micromagnetic simulations, e.g. using OOMMF [21] or MuMax3 [22]. This makes this approach attractive to estimate more realistic switching barriers based on a perturbative decomposition of a single-nanomagnet behaviour plus a correction term due to interactions with the neighbouring moments. This approach seems valid for relatively small nanomagnets favouring reversal via uniform modes, but fails if more complex reversal mechanisms are accessible.

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Chiral switching and dynamic barrier reductions in artificial square ice

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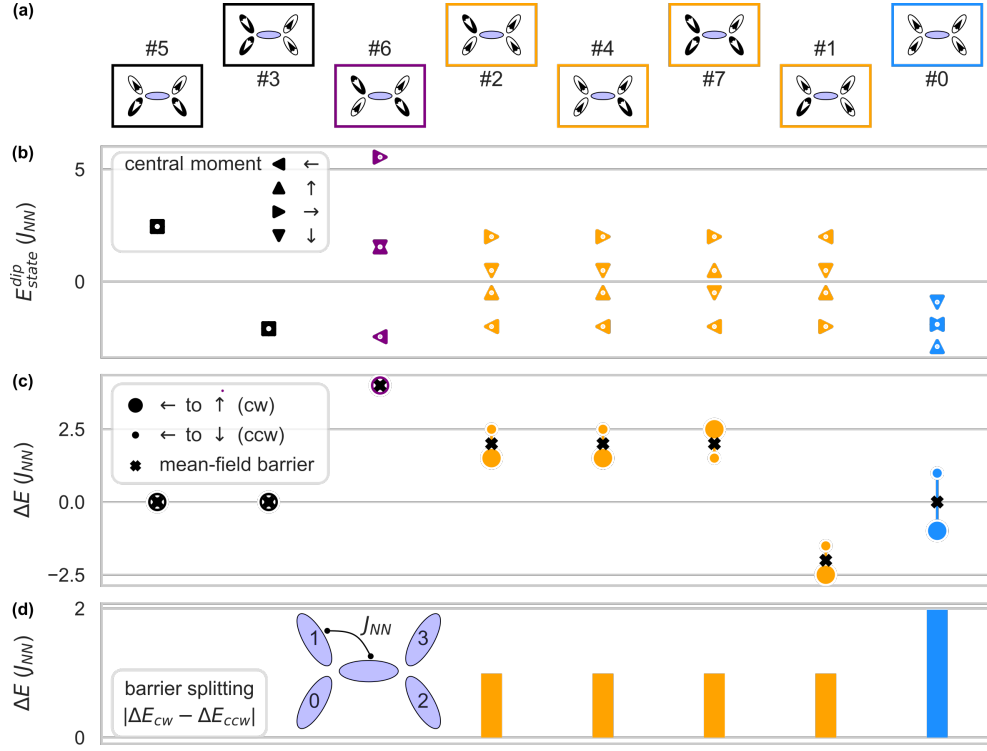


Figure 1. Barrier splitting for artificial kagome spin ice. (a) Depiction of half of the moment configurations that surround the central nanomagnet. State numbers are obtained with the enumeration of moments shown in the inset of (d) and Eq. (4) in the main manuscript. (b) Dipolar energies of configurations with the central moment pointing to the left, up, right, or down. (c) Energy corrections for clockwise and counter-clockwise reversal to the single-island switching barrier ΔE_{sb}^{shape} . (d) Net amount of chiral barrier splitting, with three cases to be distinguished. Energies in (b-d) are given as multiples as the nearest-neighbour dipolar coupling strength J_{NN} .

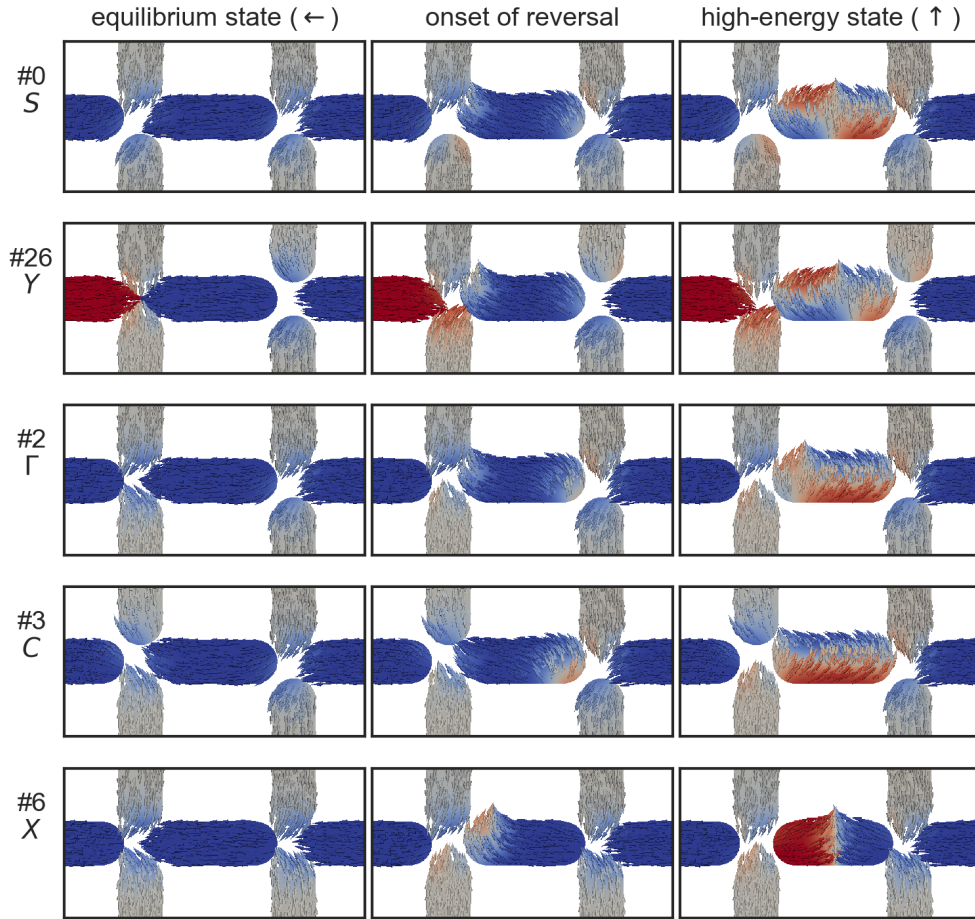


Figure 2. Edge bending in magneto-statically dominated large-island geometry. Micromagnetic configuration of environment states with different coordination of the perpendicular nanomagnets, with the notation as introduced in Fig. C2(g). Environments with *S*, *Y*, Γ configuration promote quasi-coherent reversal modes. In contrast, environments with *C*, and *X* configuration are susceptible to non-coherent reversal, which leads to further reductions of the barrier energies.

Chapter 5

Tension-free Dirac strings and steered magnetic charges in 3D artificial spin ice

The following manuscript by Sabri Koraltan, Florian Slanovc, Florian Bruckner, Cristiano Nisoli, Andrii V. Chumak, Oleksandr V. Dobrovolskiy, Claas Abert, and Dieter Suess was published in Npj Comput. Mater. (2021) 7:125; DOI: 10.1038/s41524-021-00593-7.

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Personal Contributions

The personal contributions include conceiving the original concept of 3DASI with F.Slanovc, performing all the micromagnetic simulations, and post-processing of simulation results and data, preparing all figures for manuscript and writing the initial manuscript with contributions from all the authors.

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Tension-free Dirac strings and steered magnetic charges in 3D artificial spin ice

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3D nano-architectures presents a new paradigm in modern condensed matter physics with numerous applications in photonics, biomedicine, and spintronics. They are promising for the realization of 3D magnetic nano-networks for ultra-fast and low-energy data storage. Frustration in these systems can lead to magnetic charges or magnetic monopoles, which can function as mobile, binary information carriers. However, Dirac strings in 2D artificial spin ices bind magnetic charges, while 3D dipolar counterparts require cryogenic temperatures for their stability. Here, we present a micromagnetic study of a highly frustrated 3D artificial spin ice harboring tension-free Dirac strings with unbound magnetic charges at room temperature. We use micromagnetic simulations to demonstrate that the mobility threshold for magnetic charges is by 2 eV lower than their unbinding energy. By applying global magnetic fields, we steer magnetic charges in a given direction omitting unintended switchings. The introduced system paves the way toward 3D magnetic networks for data transport and storage.

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INTRODUCTION

Data storage and transport devices ranging from hard disk drives to flash memories, from CMOS to spintronic technologies are of crucial importance in today's technological world. Usually, these devices are based on 2D structures approaching their limitations every day. 3D structures started to emerge over the past years, leading to significant improvements in both reducing the dimensions and increasing their efficiency, e.g., flash memories^{1,2}. In these data storage devices, the third dimension is used by the simple stacking of the same 2D structures. Thus, the whole power of the additional third dimension is not utilized.

Over the past years, spin ices, a class of 3D materials, have been investigated in detail. Spin ices are frustrated systems where the magnetic moments are residing on the sites of a pyrochlore lattice, a lattice with corner-sharing tetrahedra, and commonly referred to as dipolar spin ices (DSI)^{3–9}. Its degenerate ground state obeys the *ice-rule*, where two magnetic moments point to the center of the tetrahedra and two away from it. Switching a magnetic moment breaks the ice rule and creates a pair of magnetic charges in the centers of the vertices^{5,6}. These magnetic charges, commonly referred to as *emergent magnetic monopoles*, can be separated with a finite energy cost and propagated through the lattice. In a classical analog to Dirac's theory of magnetic monopoles¹⁰, monopole motion in spin ice leaves a trace called a Dirac String (DS), which is simply the chain of flipped magnetic moments connecting the two separated positive and negative magnetic poles. Theoretical and experimental studies have shown that the energetic ground state is degenerate, constrained by the ice rule and that the magnetic monopoles are connected via DS at low temperatures^{3–8,11,12}.

In order to study the geometrical frustrations and the magnetic charges on the more controllable platform, 2D artificial spin ices (2DASIs) have been designed and investigated in detail^{13–18}. There, lithographically patterned nanomagnets are arranged on different

lattices. The most common ASI lattices are the square^{19–22} and kagome ices^{23–26}. The dynamics in these systems has been extensively researched ranging from re-configurable wave band structures to tailored spin-wave channels^{27–30}.

Further frustrated systems show similar properties including artificial colloidal ice, and particle spin ice systems, where the topological defects can be controlled and steered^{31–34}.

In contrast to DSI, the reduced dimensionality and geometric frustration in square ice lift the degeneracy of the ice rule^{14–18,35,36}, thus limiting monopole mobility. Note that a unique ordered ground state was theoretically predicted for DSI with zero total magnetization per cubic unit cell, being induced by long-range dipolar interactions between the magnetic moments³⁷. However, the lift of degeneracy of the ground state is much higher for 2D systems. Various methods have been explored to regain spin ice degeneracy, including quasi three-dimensional lattices^{14,16,38–43} and interaction modifiers⁴⁴. After early designs³⁹ and realizations⁴⁵ had envisioned a 3D artificial spin ice, the three-dimensional frustrated nanowire-lattice⁴⁶ was manufactured by two-photon lithography⁴⁷, in which charge propagation was later demonstrated⁴⁸. However, in this lattice, the degeneracy of the ground state is still lifted, as the 3D structure consists of an interconnected nanowire lattice. In addition, the magnetic charges are connected via DS's, which store energy due to the presence of domain walls at the vertex centers.

In this work, we combine the advantages of both DSI and ASI and present a 3DASI lattice, where numerical investigations are performed. Our lattice is a 3D nano-magnetic network of vertices consisting of four disconnected 3D magnetic ellipsoids with perfect Ising behavior enabling tension-free DS's and unbound magnetic charges by recovering the lost degeneracy of the ice rule obeying states. Within this work, we refer to the vertices obeying the ice rule as the ground state. Vertices where three Ising spins point towards or away from the center are referred to

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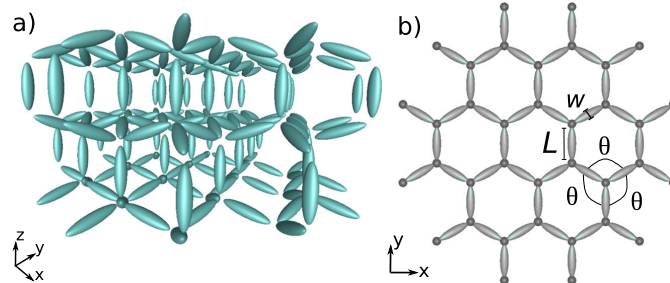


Fig. 1 3DASI lattice. **a** 3D illustration of the 3DASI lattice, and **b** the top view. The length of the ellipsoids L and maximal width w as well as the angle between the ellipsoids θ are depicted in **(b)**. The circles between the ellipsoids are the cross-section of ellipsoids pointing in or out of the plane, as a consequence of the chosen top view.

as Type III, and where all four elements point in or out of the center are called Type IV vertices. Here, magnetic monopoles are at the centers of the vertices which show a net magnetic charge. Namely, a Type III vertex hosts one magnetic monopole with charge $Q = \pm 2q_m$, and a Type IV vertex hosts a magnetic monopole with charge $Q = \pm 4q_m$ ⁴⁹.

In the 3DASI lattice, we conserve the full accessibility of 2DASI and investigate numerically the propagation of emergent magnetic monopoles. We demonstrate that the difference in energy, required to create magnetic charges and their transport, is around 2 eV, which enables the controlled propagation of unbound charges. Considering the emergent magnetic monopoles as binary, mobile information carriers, the presented lattice demonstrates the steered motion of charge carriers in a 3D magnetic nano-network at room temperatures. The controllability of these charges paves the way toward a 3D magnetic network for data transport and storage.

RESULTS

Modeling

In recent years, the direct-write technique of focused electron beam induced deposition (FEBID) has reached a high level of maturity for 3D nanofabrication. Many complex-shaped nano-architectures have become available, providing access to experimental investigations of curvature-, geometry-, and topology-induced effects in various disciplines, including magnetism, superconductivity, photonics, and plasmonics^{50–54}.

Inspired by the recent developments in 3D optical lithography and focused particle 3D nano-printing by FEBID, we present a 3DASI lattice, where magnetic rotational ellipsoids are arranged along the main axis of a tetrahedron, resulting in an angle $\theta = \arccos(-1/3) \approx 109.5^\circ$ between the elements, reproducing the Ice I_h crystal of the water ice^{3,5,9}. Figure 1 illustrates the designed 3DASI lattice.

Note that in Fig. 1, we illustrate only the magnetic ellipsoids forming the lattice. In reality, the ellipsoids can be fabricated by direct-writing, i.e., FEBID, and interconnected with a magnetic insulator, e.g., platinum⁵¹ or niobium-based⁵⁵ compounds, as exemplarily illustrated in Supplementary Fig. 1. With FEBID as a suitable nanofabrication technique of the 3DASI lattice, the maximal width of the ellipsoids w can be reduced down to few tens of nanometers, while the length L can be chosen up to a few micrometers^{50–53}.

Note that optical lithography techniques such as two-photon lithography have already been employed to fabricate three-dimensional magnetic structures such as nanowire lattices and magnetic buckyballs^{46,48,56,57}.

We choose rotational magnetic ellipsoids as ASI elements because the self-demagnetizing field of the ellipsoids is homogeneous thus keeping the magnetic elements uniformly magnetized in the direction of the longer axis with no edge inhomogeneities^{58–61}. Hence, our model shows nearly perfect Ising behavior and is suitable to separate and host magnetic charges. The maximal average deviation from a perfect uniform magnetization for one ellipsoid in our model is found to be $\Delta\phi \approx 4 \times 10^{-4}$ deg, where the interactions with nearest and next-nearest neighbors were encountered.

In this work, we restrict our model to three layers to conduct numerical experiments in feasible times. However, the region of interest is only the middle layer, meaning that we analyze the 3DASI lattice as a bulk system. Furthermore, the lateral dimensions are kept finite being confined only for the dynamic simulations of monopole propagation. Note that in finite-sized systems boundaries can be engineered to control the bulk behavior or introduce and confine defects in spin ice systems^{62,63}.

Dirac strings and magnetic monopoles

Because of the symmetry of the tetrahedra, the degeneracy of the ice rule is not lifted in 3DASI. Consequently, the tension of the DS should vanish. To obtain an energy scale, we approximate the ellipsoids by magnetic dipoles with the dipole moment $\mu = M_s V$, where M_s is the saturation magnetization and V is the volume of the nanostructure. In this formulation, the dipolar interaction energy for each pair of magnetic dipole moments $\mathbf{m}_i, \mathbf{m}_j$ at positions $\mathbf{r}_i, \mathbf{r}_j$ in the lattice is given by the dipolar interaction energy

$$E_{\text{dip}} = \frac{\mu_0}{4\pi|\mathbf{r}_{ij}|^3} \left[\mathbf{m}_i \cdot \mathbf{m}_j - 3 \frac{(\mathbf{m}_i \cdot \mathbf{r}_{ij})(\mathbf{m}_j \cdot \mathbf{r}_{ij})}{|\mathbf{r}_{ij}|^2} \right] \quad (1)$$

with $\mathbf{r}_{ij} := \mathbf{r}_j - \mathbf{r}_i$ and vacuum permeability μ_0 . Then from Eq. (1), we obtain the energy scale

$$J_{NN} = \frac{3}{2\sqrt{2}} \frac{\mu_0 \mu^2}{\pi a^3} \quad (2)$$

as introduced in ref. ⁶¹, where a is the lattice constant.

From it we obtain the energy levels for the different vertex types, shown in Table 1. Note the degeneracy in the ice rule obeying vertices.

Considering the monopole picture⁶, the magnetic ellipsoids can be approximated by the dumbbell model, which allows one to separate two magnetic monopoles in each magnetic ellipsoid. This model brings further corrections, as the nearest-neighbor exchange interactions within the vertices are also accounted. However, here we consistently compare the structural difference

between the 2D and 3D artificial spin ice vertices, where the point dipole approximation covers the relevant interactions.

To consider a more complete model including further corrections to Eq. (1), we employ calculations via micromagnetic simulations with *magnum.fe*⁶⁴, using material properties similar to the cobalt-iron (CoFe) alloys used in FEBID. Note that with this technique, the material parameters are already tunable^{52,65} for 2D structures, and advances are expected in the near future. We choose a saturation magnetization $M_s = 800 \text{ kA m}^{-1}$ and exchange stiffness constant $A_{\text{ex}} = 15.5 \text{ pJ m}^{-1}$. The ellipsoids have the dimensions $L = 100 \text{ nm}$, $w = 20 \text{ nm}$, and a vertex-to-vertex distance of $a = 120 \text{ nm}$, see Supplementary Fig. 2 for a detailed comparison between 2DASI and 3DASI elements. It is noteworthy that in our studies we especially focused on the 3D structures of sizes that have been demonstrated for platinum-based nanofabrications. Magnetic CoFe/CoFeB structures are larger, but the down-scaling is expected within the next years.

Figure 2a shows the calculated energies of all the possible vertices in 2DsASI and 3DASI. The latter show clearly the sixfold degeneracy of the ground state Type I/II. This degeneracy allows for magnetic charges to be separated because a tension-free DS is created, making them *truly* unbound. In Fig. 2a, we chose to plot all $2^4 = 16$ possible configurations within a vertex. In reality, for vertices in the 3DASI lattice, there exists only one unique ground

state configuration, the ice-rule configuration, up to symmetry operations, i.e., rotations and mirroring, always obeying the *2in-2out* ice rule.

According to the original definition of emergent magnetic monopoles, the energy required to create a monopole-antimonopole pair (charge separation), should be larger than the energy required to propagate it^{6,10,16,66–68}. We verify these properties by applying a full-micromagnetic model to calculate the energy barriers to separate and propagate the magnetic charges^{60,69,70}. A detailed description of the full micromagnetic model and its application on 2DASIs is given in ref. ⁶⁰.

Figure 2b illustrates the minimum energy paths (MEPs) for the switching processes within a DS. Starting from a lattice in the ground state, as illustrated in Fig. 2d), we separate two magnetic charges by switching the magnetization of one ellipsoid, and thus creating two Type III defects, which are depicted in Fig. 2e). The positive charge is then propagated to increase the length of the DS, shown in Fig. 2f–h). It can be seen, that the separation MEP (magenta) yields a separation barrier ΔE_s approximately 2 eV higher than the propagation barriers ΔE_p (green MEPs), which are nearly constant.

Note that the simplified and improved string method is a static method. Here, the energy landscape is reproduced in such a fashion, that the magenta curve in Fig. 2b is the energetically most favorable way to switch from state 0 to state 20, where two Type III defects are created. With these calculations, we are able to obtain direct visualization of the switching mechanism. Each state with energy E corresponds to a unique magnetic configuration that is reached within this transition. The reversal mechanisms are highly non-coherent and complex, being dictated by domain wall movements. Note that the reversal mode of magnetic elements can be an exchange or magneto-statically dominated⁶¹ and depend strongly on the chosen magnetic parameters. Impurities during the fabrication process, can lead to changes in the magnetic properties, and impact the reversal mode and transition kinetics of the system. With FEBID as a fabrication method, the

Table 1. Dipolar interaction energies in each vertex type in 2D and 3D ASI in units of J_{NN} , which is $J_{\text{NN}} \approx 0.43 \text{ eV}$ for the chosen material and geometry parameters.

Vertex type	$E_{\text{dip}}^{2\text{D}} (J_{\text{NN}})$	$E_{\text{dip}}^{3\text{D}} (J_{\text{NN}})$
Type I	$2(\sqrt{2}/3 - 2)$	$-5/(2\sqrt{3})$
Type II	$-2\sqrt{2}/3$	
Type III	0	0
Type IV	$2(\sqrt{2}/3 + 2)$	$15/(2\sqrt{3})$

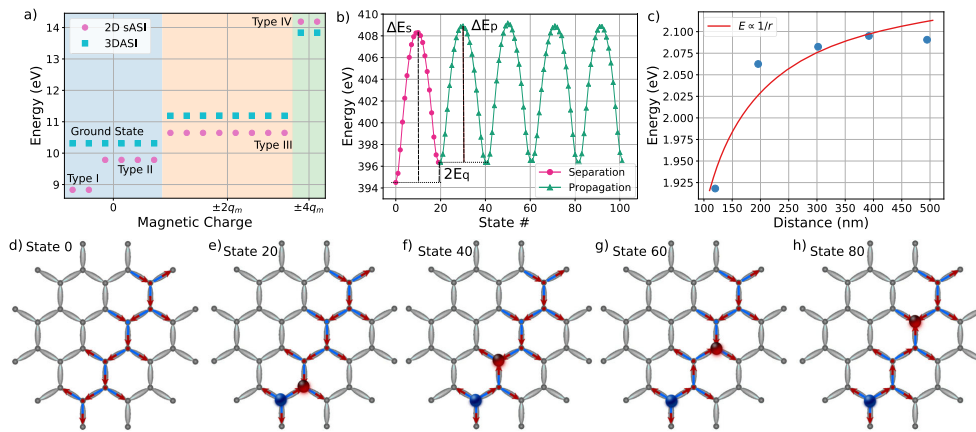


Fig. 2 Magnetostatic energies and minimum energy paths for propagation and separation of charges. **a** Total energies per vertex calculated with a micromagnetic treatment by taking into account energetic contributions of the exchange and demagnetization fields, where every possible magnetic configuration in a 2DsASI (circles) and in our 3DASI (squares) was considered, as illustrated in Supplementary Figs. 3, and 4, respectively. **b** Minimum energy paths to separate (magenta) the two magnetic charges by switching one ellipsoids magnetization in the ground state, and propagating it further through the lattice in a constant direction (green). The difference of saddle points and the corresponding minima yields the separation barrier ΔE_s and propagation barrier ΔE_p , while the energy of the lattice increases by $2E_q \approx 2 \text{ eV}$ due to the two additional charge defects. **c** Interaction energy between the magnetic charges after monopole separation and during propagation, which follows a $1/r$ Coulomb Law. **d–h** Schematic illustrations of the magnetization configurations for the ellipsoids in the DS with the positive (dark red) and negative (dark blue) charged magnetic monopoles. Snapshots of magnetizations are depicted in Supplementary Fig. 5, and animations for switching processes are available online.

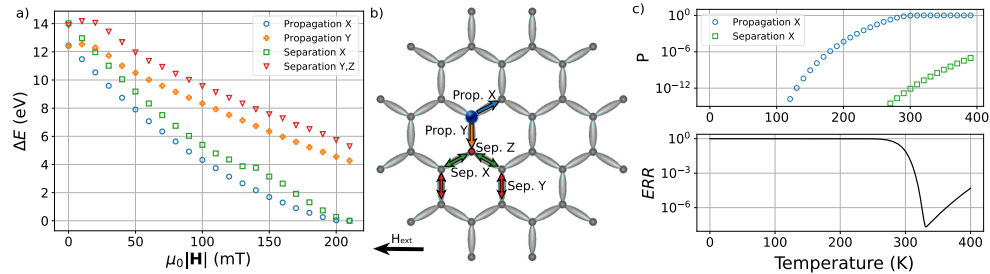


Fig. 3 Arbitrary field finite temperature micromagnetic analysis. **a** Dependence of the energy barrier on the applied external fields at $T = 0$ K to switch the magnetizations of different ellipsoids as depicted in **(b)**. **c** Upper panel: the switching probability P at different temperatures for the relevant transitions from **(b)** calculated with FTM analysis, Eq. (3), where a Zeeman field with $\mu_0|\mathbf{H}| = 180$ mT, attempt frequency $f_0 = 50$ GHz and pulse time $t_{\text{final}} = 0.15$ ns were chosen. Lower panel: mean error rate of successful propagation correlated with the unintended separation, as given by Eq. (4).

parameters are freely tunable^{52,65}, and thus the reversal modes can be tuned as well.

By separating the emergent monopoles, the energy of the system increases about $2E_g = 2$ eV, which represents the energy increase due to Type III defects. Since we propagate only one charge, while the environment does not change, the energy of the system should stay constant during the propagation. This is possible in our 3DASI lattice due to the sixfold degeneracy of the ice-rule. Thus, propagating one magnetic monopole to the next vertex does not lead to tension in the DS. Note that, in non-degenerate 2DsASI, DS's necessarily carry a tension. However, we need to consider the possible Coulomb interactions between magnetic charges^{5,6}. In contrast to the classical DSI, this interaction can be neglected in comparison to the energy barriers in our 3DASI lattice⁶. Nevertheless, this energy is present in our lattice, as shown in Fig. 2c where we illustrate the gain in energy after the monopoles are separated and propagated as a function of the direct distance between the monopoles. After separation, the monopoles are at the centers of two neighbored vertices, whereas the distance between them is $a = 120$ nm. To obtain the interaction energies we calculate the demagnetization energies within the bulk layer for all energetic minima from Fig. 2b and subtract the energy where the system is in an equilibrium configuration with no magnetic monopoles (State 0). The obtained interaction energies follow a Coulomb law with shifted energy $E \propto 1/r$, where r is the shortest distance between the monopoles⁶.

Note that these results are obtained at $T = 0$ K. In order to analyze the dependence of the stability and the propagation properties of a single monopole on both externally applied fields and temperature, we use the arbitrary field finite temperature micromagnetic analysis (FTM)⁷¹.

Arbitrary field FTM analysis

Starting from an initial magnetization state with one magnetic monopole of charge $Q = -2q_m$, we calculate the energy barriers to switch the magnetization of different 3DASI elements under a uniform external magnetic field with $\mu_0\mathbf{H} = -\mu_0|\mathbf{H}|\mathbf{e}_x$. Here, $\mu_0|\mathbf{H}|$ denotes the strength of the applied field, and $\mathbf{e}_x$ is the unit vector along x . $\mu_0|\mathbf{H}|$ is varied between 0 and 200 mT.

Commonly, magnetic monopoles are found in pairs. However, finite-sized systems allow trapping unpaired magnetic monopole at boundaries⁶². Similarly, the finite sizes of our lattice allow particular boundary ellipsoids to act as monopole injectors, and thus, single monopoles can be present in the lattice. Depending on the initial magnetic configuration of the middle layer, multiple monopoles with equal or opposite charges can be injected. For this particular initial state, we inject solely one monopole and are

interested in its propagation in the desired direction without creating additional charges.

As illustrated in Fig. 3b, we consider different transitions, where the magnetic charge is propagated along X (blue arrow) and Y (orange arrow), or additional charges are separated in a neighbored vertex, where the separation in three different directions is considered, X (green arrows), Y and Z (red). By doing so, we cover all possible transitions in our lattice, where the existing charges with $Q = -2q_m$ are either propagated or additional monopole-antimonopole pairs are created at different locations. The initial magnetization configuration is the same as in Fig. 4e. Snapshots of magnetizations during switching processes for the relevant transitions are illustrated in Supplementary Fig. 6.

Figure 3a shows the calculated energy barriers as a function of the applied field strength for the transitions depicted in Fig. 3b. Our results indicate that the propagation barrier along X (blue, circles) is always lower than any other barrier. At vanishing fields, the barriers for propagation X and Y are equal, as demonstrated above. With the increasing field and approaching the coercive field, all barriers are lowering, ultimately, vanishing once the critical field is reached.

In the FTM analysis, described by Suess et al.⁷¹, a field-driven transition occurs with the switching probability

$$P = 1 - \exp\left(-f_0 t_{\text{final}} e^{\left(\frac{-\Delta E(H)}{k_B T}\right)}\right), \quad (3)$$

where f_0 denotes the attempt frequency, $\Delta E(H)$ the energy barrier at a given field magnitude, k_B the Boltzmann constant, t_{final} the time of applied field pulse and T is the temperature.

To analyze the stability of the propagation at room temperature, we consider $T = 300$ K and calculate the associated probabilities where we choose $\mu_0|\mathbf{H}| = 180$ mT, $f_0 = 50$ GHz, and $t_{\text{final}} = 0.15$ s. f_0 is chosen according to values considered in ASI literature⁷² and t_{final} based on experimental expectations. Note that these two values appear only as a common factor in Eq. (3) and therefore other values for f_0 can always be compensated by an appropriate choice of t_{final} . The field strength $\mu_0|\mathbf{H}| = 180$ mT is chosen such that the switching barrier of the propagation is below $300k_B T$.

The upper panel of Fig. 3c illustrates the relevant probabilities for the switching possibilities depicted in Fig. 3b. Our results indicate that only the element of interest will change its magnetization, as its switching probability increases with the temperature, where $P_{\text{prop}}(300 \text{ K}) > 0.9$. All the other probabilities are nearly zero at temperatures around $T = 300$ K. We chose to depict only the switching probabilities for the propagation and separation along x , as the other probabilities are lower than 1×10^{-15} .

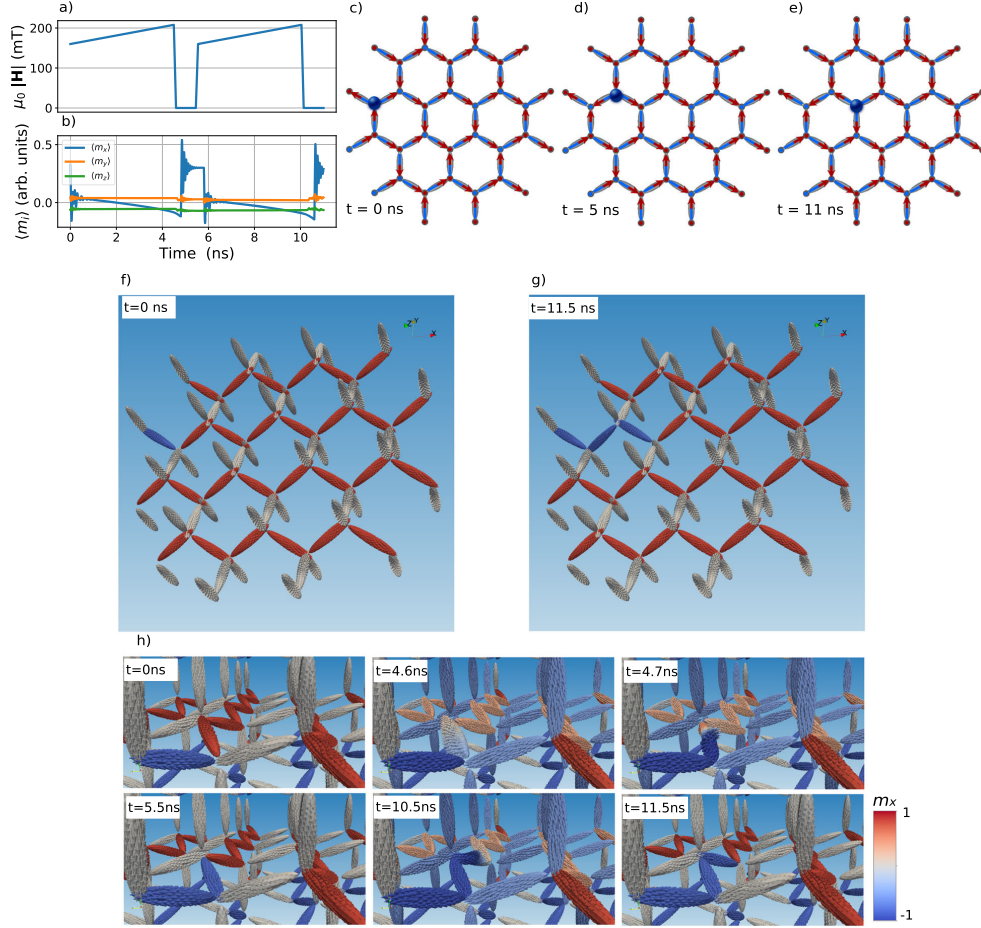


Fig. 4 Field-driven monopole propagation. Temporal evolution of the magnitude of the applied field (a) and of the averaged magnetization components (b) within the bulk layer during the x -propagation. The field is applied along $-x$ in order to maximize the Zeeman energy acting on the ellipsoid whose magnetization should be switched. The field is turned off after $t = 4.8$ ns in order to transfer the magnetic charge to the next vertex controlling the propagation. c–e Schematic illustration of the monopole (dark blue) propagation. f–h Snapshots of the magnetization states from the output of the conducted micromagnetic simulations, where f illustrates the initial and g the final magnetic configurations. The magnetization evolution during the simulations is depicted at different times (white boxes) in (h). Color in f–h shows the x -component of the magnetization, as given by the colorbar.

However, we still need to define the mean switching error rate

$$\text{ERR}(T) = 1 - P_{\text{prop}}(1 - P_{\text{sep}}(T))^N, \quad (4)$$

which describes the probability for an unsuccessful propagation, i.e., falsely switch of one element leading to separation (or propagation failure due to insufficient thermal energy) in a lattice with N elements, where P_{prob} describes the propagation probabilities and P_{sep} the separation probabilities.

In the lower panel of Fig. 3c, we see that the error rate significantly decreases after $T = 290$ K, as the X propagation probability increases to nearly 1. For $T > 330$ K the propagation is guaranteed, however the separation probability increases leading to an increment in the mean error rate. This indicates that the monopoles can be propagated in a controlled manner, in the desired direction, and that they can be accessed at temperatures

around room temperature, improving the scalability and thermal stability of pyrochlore DSI.

Field-driven controlled monopole propagation

The controlled propagation can be achieved at any temperature, including $T = 0$ K if one adjusts the strength, direction, and duration of the globally applied magnetic fields. As illustrated in Fig. 3a, the energy barrier vanishes for large enough fields. To investigate this in a rather direct manner, we conduct numerical experiments, where we continue with our micromagnetic treatment and investigate the propagation of monopoles by applying pulses of external magnetic fields at $T = 0$ K.

We start from an initial system where the magnetic configuration is chosen in such a fashion that every vertex obeys the ice-rule, and thus it is in an energetic minimum. Reversing the

magnetization in one boundary ellipsoid injects exactly one magnetic monopole with $Q = -2q_m$, see Fig. 4c and f. After the injection, we confine the boundary ellipsoids as well as top and bottom layers and investigate the dynamics of the magnetic monopole propagation in a bulk lattice, see Supplementary Fig. 7.

We continue by applying a magnetic field along $-x$ direction, as in the FTM analysis above. The field profile can be seen in Fig. 4a, where the magnitude of the applied field increases linearly over 4.8 ns from 160 to 208 mT and is then turned off abruptly after the propagation is successful.

Figure 4b shows the temporal evolution of the averaged magnetization components within the middle layer. Our results indicate that the magnetic charge is propagated to the next vertex via domain wall movements as predicted by our string method calculations and FTM analysis. Magnetization snapshots at chosen times from Fig. 4h show that until the critical field is reached, small deviations from the ising state are observed for the ellipsoids in the ground state, explaining the slow decrease in the average x -component of the magnetization from Fig. 4b. Once the applied field is large enough, the propagation takes place keeping the other vertices in the ground state, avoiding additional charge separations. By turning off the external field, we relax all the ellipsoids back to their ising state and control the propagation by demand. One can see in Fig. 4b that the average magnetization drops rather abruptly when the magnetic charge is propagated one vertex further with each step, demonstrating the free and controlled propagation of an unbound emergent charge in the presented lattice. In the Supplementary notes, we also discuss the magnetic charge being further propagated to the next x -row by applying the same protocol with a magnetic field along y and illustrate this propagation in Supplementary Figs. 8 and 9.

DISCUSSION

We investigated micromagnetically a 3DASI lattice, which combines the advantages of classical 2DASI and pyrochlore DSI lattices, recovering the lost frustration and degeneracy of the ground state of the 2DsASI by enabling tension-free DS and thermally stable unbound magnetic monopoles.

In conclusion, string method simulations and FTM analysis show that the finite-sized 3DASI lattice allows the creation and propagation of magnetic charges, which can exist freely or in pairs being connected via tension-free DS at arbitrary temperatures. The interaction between two unbound magnetic monopoles follows a $1/r$ dependency showing further evidence for the presence of Coulomb interaction in our 3DASI lattice. We showed by numerical experiments, that a single unbound charge can be propagated by an external magnetic field, without creating additional magnetic charges.

Low-energy and ultra-fast switching in the 3DASI lattice, as well as the stability of emergent magnetic monopoles at room temperature and above, paves a way towards functional 3D magnetic nano-networks for data transport and storage. The controlled propagation of the magnetic charges is of high interest regarding the idea of *magnetricity*^{66,73}, which might lead to further devices¹⁸.

Even though we focused only on the center layer and therefore regarded our model as a bulk lattice, the limitations of the z layers might show additional and equally interesting phenomena which will be investigated in detail in further work. By *shaving off* at a given z dimension, the lattice would have magnetic vertices, containing only three ellipsoids, which would *always* host a magnetic charge. The top and bottom layers could act as charge plasma layers, which might *decharge* in the middle layer, creating a magnetic capacitor.

METHODS

String method

For each switching process, we start from an initial state and a final state interconnected via 19–20 magnetization states, corresponding to a coherent rotation of the magnetization of the element of interest. In a second step, the total energy of each image is driven a small step towards its energetic minimum. The energy contributions are the demagnetization energy, the exchange energy, and Zeeman energy (FTM analysis). The third step consists of cubic interpolation of the path, such that the magnetization states are equidistant according to an appropriate energy norm^{60,64,69}. The last two steps are repeated until convergence is reached.

Micromagnetic simulations

By using the finite and boundary element method based simulation code `magnum.fe`⁶⁴, we solve the Landau–Lifshitz–Gilbert (LLG) equation⁷⁰:

$$\frac{\partial \mathbf{m}}{\partial t} = -\frac{\gamma}{1 + \alpha^2} \mathbf{m} \times \mathbf{H}^{\text{eff}} - \frac{\alpha \gamma}{1 + \alpha^2} \mathbf{m} \times (\mathbf{m} \times \mathbf{H}^{\text{eff}}), \quad (5)$$

where α is the Gilbert damping constant, γ is the reduced gyromagnetic ratio, $\mathbf{m}$ is the magnetization unit vector, and $\mathbf{H}^{\text{eff}}$ is the effective field term, which includes the considered energy contributions from demagnetizing, exchange, external and anisotropy fields (magnetic confinement). Vertex state energies are computed by calculating the demagnetizing and exchange energies from the relaxed structures (high damping $\alpha = 1$). For the field-driven dynamic simulations, we also include global external fields and solve the LLG for a given time. In addition, the magnetization components are averaged within the middle (bulk) layer for each time step, as depicted in Fig. 4b.

DATA AVAILABILITY

The data that support the findings of this study are available on request from the corresponding author S.K. The data are not publicly available due to closed-source simulation software `magnum.fe`.

CODE AVAILABILITY

The micromagnetic code used to obtain the findings of this study is not publicly available due to closed-source simulation software `magnum.fe`. The codes to create the finite element meshes for the used geometry of the 3DASI lattice are available on request from the corresponding author S.K.

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AUTHOR CONTRIBUTIONS

S.K. and F.S. conceived the concept. C.A. and F.B. wrote and improved the micromagnetic codes. S.K. performed the micromagnetic simulations. S.K., F.S., F.B.,

C.N., A.V.C., O.V.D., C.A., and D.S. further improved and developed the concept and discussed the results. All authors contributed to the paper.

COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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Tension-free Dirac strings and steered magnetic charges in 3D artificial spin ice

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SUPPLEMENTARY NOTES

In order to provide a better understanding about the performed micromagnetic simulations, we show with the additional material below snapshots of the magnetization states during the different methods applied in the main paper, as well as additional and a deepened description of the methods.

FEBID Nanofabrication. The presented 3DASI lattice can be fabricated by making use of Focused Electron Beam Induced Deposition nanofabrication technique[1], [2], [3], [4], where Cobalt-Iron in different compositions can be used. Regarding our specific case, it is possible to fabricate a magnetic vertex of 3DASI lattice, where the magnetic ellipsoids are interconnected by a platinum-based compound as shown in Ref. [2], or niobium-based as in Ref. [5]. A rather realistic illustration for the 3DASI vertex is given in Supplementary Fig. 1.

Vertex Energies. Following the structure of the article, we provide in addition to Fig. 2a) of the main text, the detailed description of the followed micromagnetic approach to calculate the energies associated with all the possible magnetization states in a 2D square ice vertex and in our 3DASI lattice, depicted in Supplementary Fig. 3, and Supplementary Fig.4 respectively. For the former we use magnetic islands having an ellipsoidal form instead of the state-of-the-art stadium-shaped islands, to facilitate the comparison to our model.

Here, the planar islands have a length $L = 100\text{nm}$, a width $w = 20\text{nm}$ and a height $h = 13.33\text{nm}$ to match the volume of the 3D ellipsoid used in the 3DASI. By doing so we obtain the equal total magnetic moment $\mu = M_s V$ in both models. Figure 2 illustrates a direct comparison of the magnetic elements used to build the vertices, where the dimensions of the magnetic bodies are also depicted.

After setting the initial magnetization states according to each magnetic configuration corresponding to a particular vertex type, we solve the Landau-Lifshitz-Gilbert equation[6] at high damping $\alpha = 1$ for 5 ns to achieve the energetic ground state. The calculated total energies reflect contributions of the exchange and demagnetization energies, where the latter includes both the shape anisotropy and the interaction fields among the elements. For this purpose we use material parameters as given by Ref.[4], [7], where $M_s = 800\text{kAm}^{-1}$ and $A_{\text{ex}} = 15.5\text{pJm}^{-1}$ were used.

String Method Simulations. To support the minimum energy paths given in Fig. 3 in the main text, we provide snapshots of the evolution of the magnetization states during the string method simulations in Supplementary Fig. 5 showing the separation (magenta box) and propagation (green box) of the magnetic charge. Our results show that it is energetically most favorable when the reversal mechanism is dictated by nucleating and propagating complex and rather elliptical domain-walls. Note that we considered a not well ordered ground state to show that propagation barrier remains nearly constant, being independent of the switching direction as it is in the case of the 2DsASI[8].

Finite temperature micromagnetics. In addition to the results presented in the main paper, we provide here snapshots of the magnetization states during the MEP's, where the path were discretized in 20 equidistant images, and #0 depicts the initial state, #10 the magnetization state, with the highest energy, and #20 illustrates the final magnetization state.

The presented snapshots were taken from the simulations where an external field along $-x$ was applied and $\mu_0|\mathbf{H}| = 180\text{mT}$. One can see that the switching mechanisms are again highly non-coherent and complex and are dictated by domain-wall propagation.

Field driven dynamics. To show that the unbound magnetic charges can be moved within the lattice, we globally apply external magnetic fields. In the main text, we chose to illustrate and discuss the propagation along x -direction.

For this purpose we are confining our lattice for two different reasons. The first, and the most important reason, is that we want to characterize the dynamics when we consider our 3DASI lattice as a bulk lattice. Hence, we are confining the top and

bottom layers, use them rather as boundary conditions, to have rather exact solutions for the demagnetization fields within the bulk layer.

The second reason is that we are interested in the dynamics of one magnetic charge which is already present in the lattice. Therefore, an initial magnetization state is chosen, which includes one magnetic charge with $Q = -2q_m$, while every other vertex is a well ordered ground state. Furthermore during our investigations we observed that the magnetic charges can be nucleated from the boundaries into the lattice, when the edge ellipsoids are not confined. Thus, we are specifically confining certain magnetic ellipsoids at the edges to avoid finite size and boundary effects, as much as possible within a micromagnetic framework.

Regarding the approach we are using to confine the above mentioned magnetic elements, we set a very high uniaxial anisotropy along the main axis of each magnetic ellipsoid, where the anisotropy constant $K_u = 1 \text{ MJm}^{-3}$ allows this confinement. The confined ellipsoids can be seen in Supplementary Fig.7, where the red ellipsoids are confined, and blue ellipsoids do not have an additional uniaxial anisotropy, being allowed to change their magnetization during the magnetization processes.

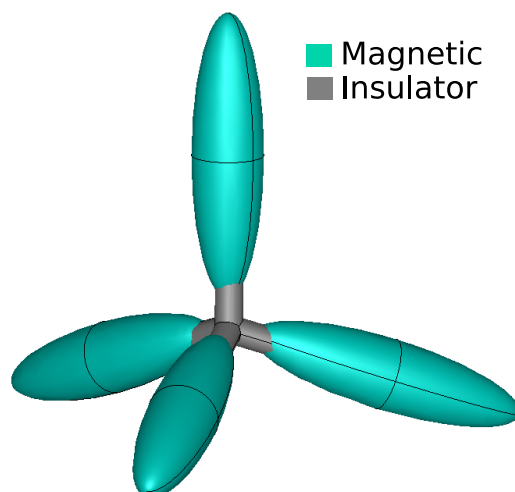
Using the same material parameters, we apply external field pulses to transfer the magnetic charge present in the system. The magnitude of the field is increasing from $\mu_0 H(0) = 160 \text{ mT}$ to $\mu_0 H(0) = 208 \text{ mT}$ in $\Delta t = 4.8 \text{ ns}$. The magnitude, and thus, the pulse profile is depicted in Fig. 4a) of the main text.

The micromagnetic simulations are conducted by solving the LLG using low damping $\alpha = 0.1$, and considering the energetic contributions from the exchange, demagnetization and uniaxial anisotropy (confinement) fields. We support the temporal evolution of the averaged magnetization components of the not-confined magnetic ellipsoids depicted in Fig. 4b) in the main text, with the animation given in "Stepwise_X_Propagation_0_to_11_5ns.gif", to better understand the switching behavior and the dynamics of the system.

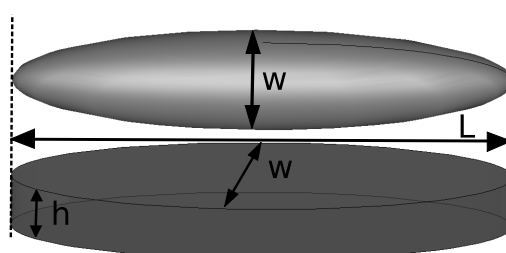
As mentioned in the main text, the magnetic charges can be freely propagated through the lattice based on the degeneracy of the ground state and the ability of our lattice to allow unbound charges. For this purpose, we performed additional micromagnetic simulations continuing from the simulations, where the magnetic charge was propagated a distance of $2a = 240 \text{ nm}$ along the x -direction (of the lattice), and then $a = 120 \text{ nm}$ along the y -direction. For the latter, the external magnetic field was applied along the y -axis being rotated by an angle $\phi = 20^\circ$ around the z -axis. This slightly tilting was done to maximize the Zeeman energy acting on the ellipsoid of interest, and simultaneously minimize the Zeeman energy acting on the other islands facilitating the switching process.

Supplementary Fig. 8a) provides the magnitude of the applied field as a function of time, while Supplementary Fig.8b) shows the temporal evolution of the averaged magnetization components within the middle layer. Our results indicate clearly that the switching process leads again to an abrupt increase in the y -component once the critical field is reached and the domain-wall starts to nucleate, which ultimately switches only the magnetization of the ellipsoid capable to propagate the magnetic charge along y -direction. This reversal basically transfers the magnetic charge from one x -row to another. The animation of this switching is provided in "Y_Propagation_with_fields.gif".

SUPPLEMENTARY FIGURES



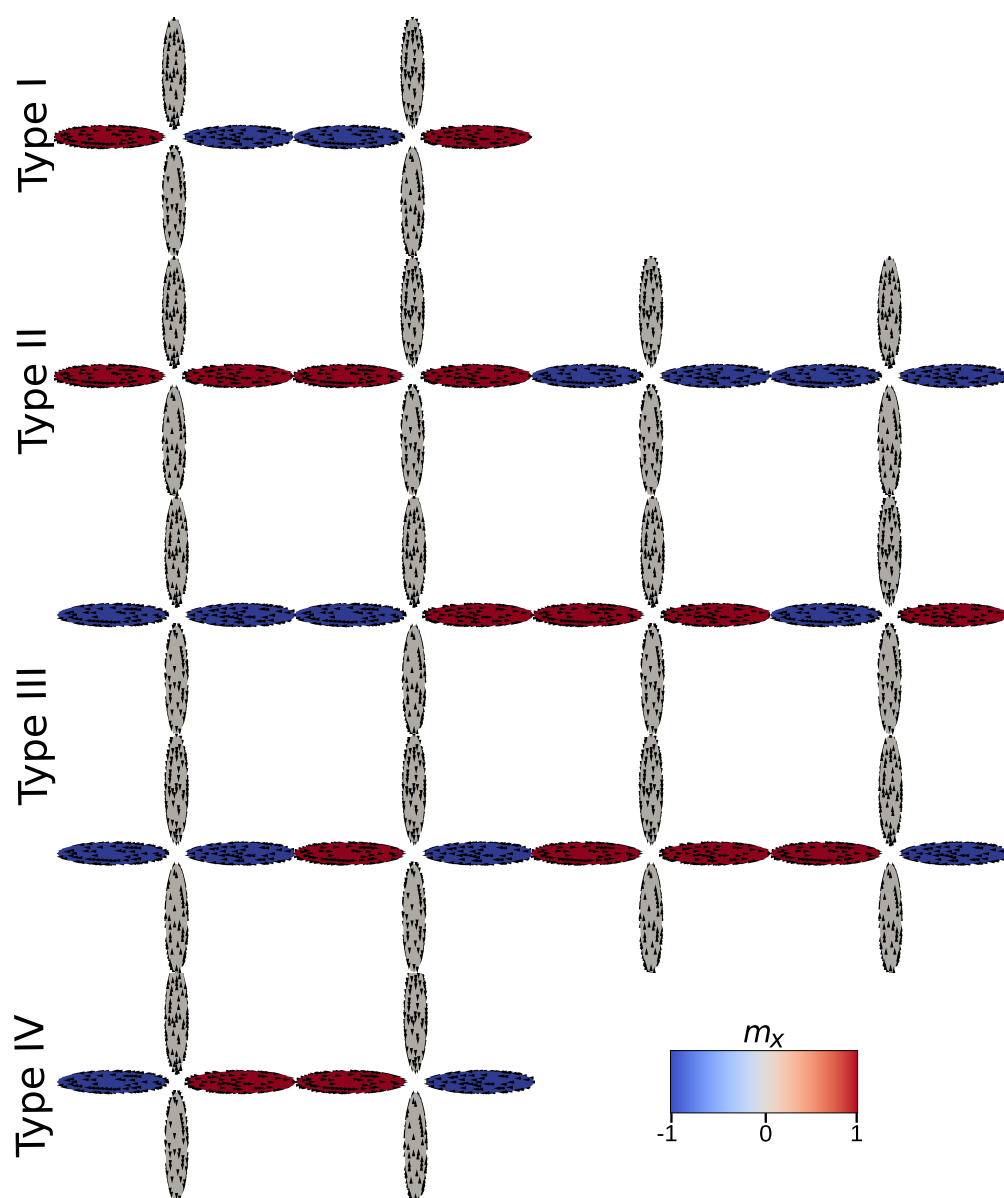
Supplementary Fig. 1: Realistic illustration of a 3DASI vertex which could be fabricated by FEBID, where the insulator material based on Platinum is connecting the magnetic ellipsoid fabricated using Cobalt-Iron alloys.



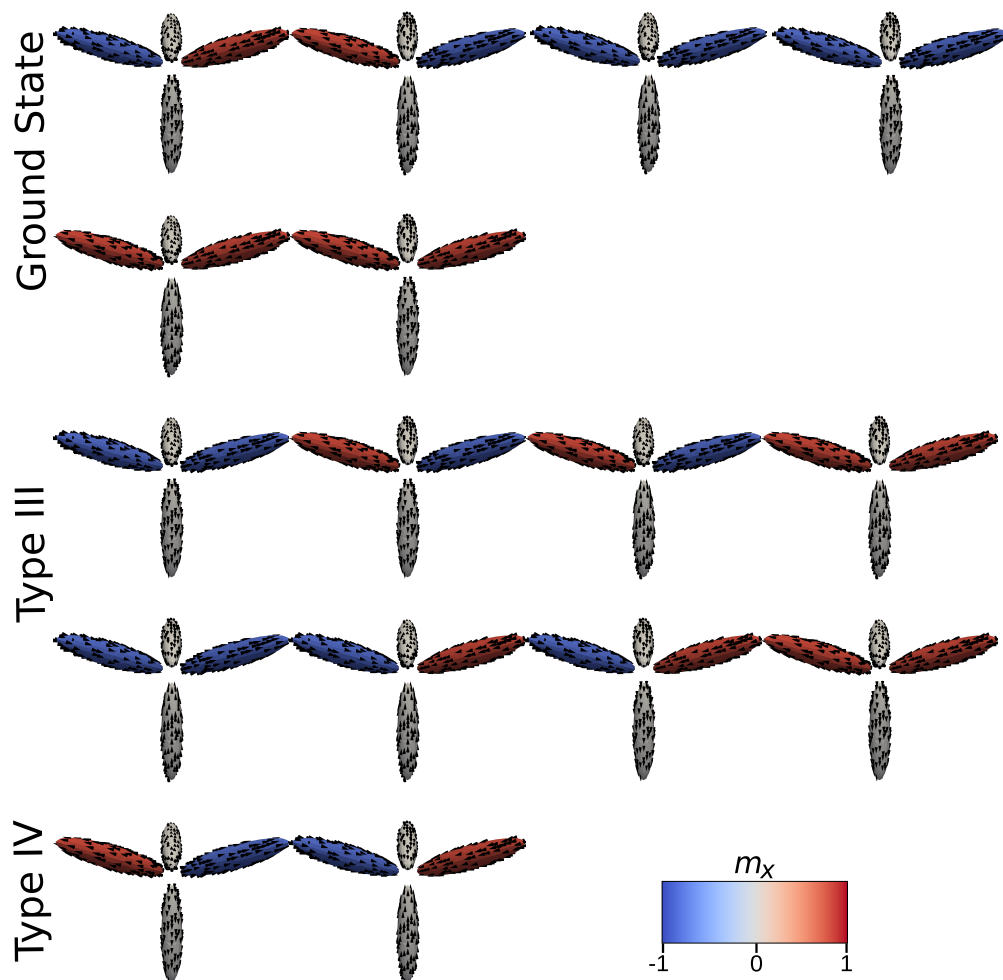
Supplementary Fig. 2: Comparison of the used 2D and 3D ASI magnetic elements, with equal volumes, and thus equal total net magnetic moments.

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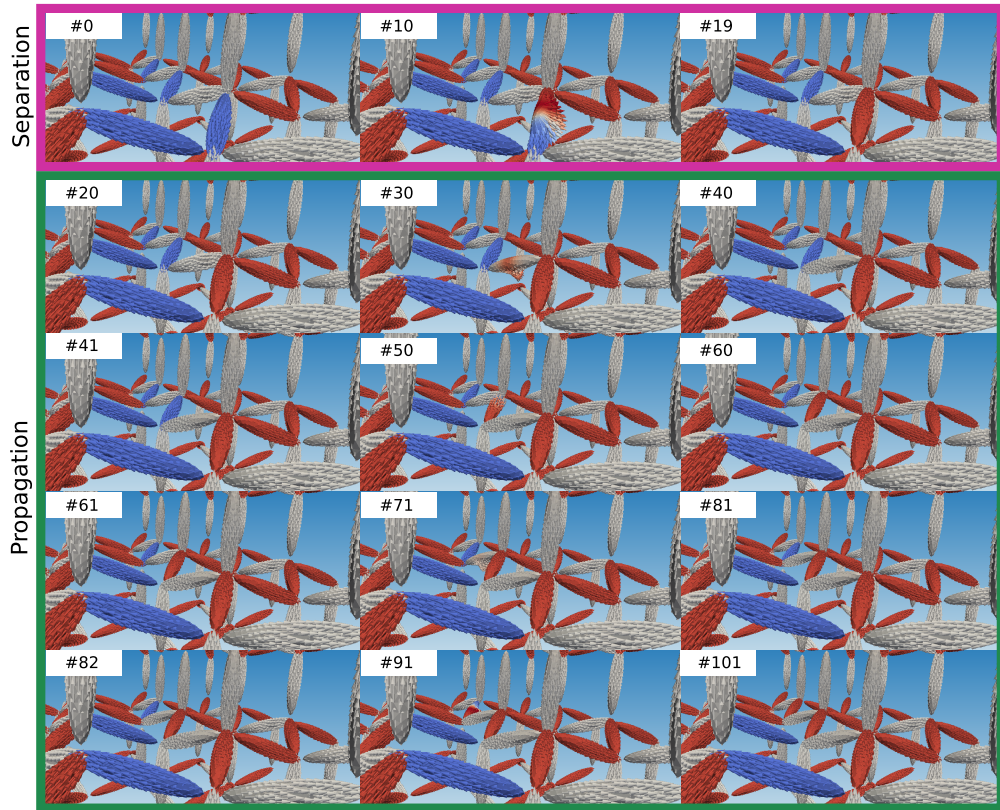
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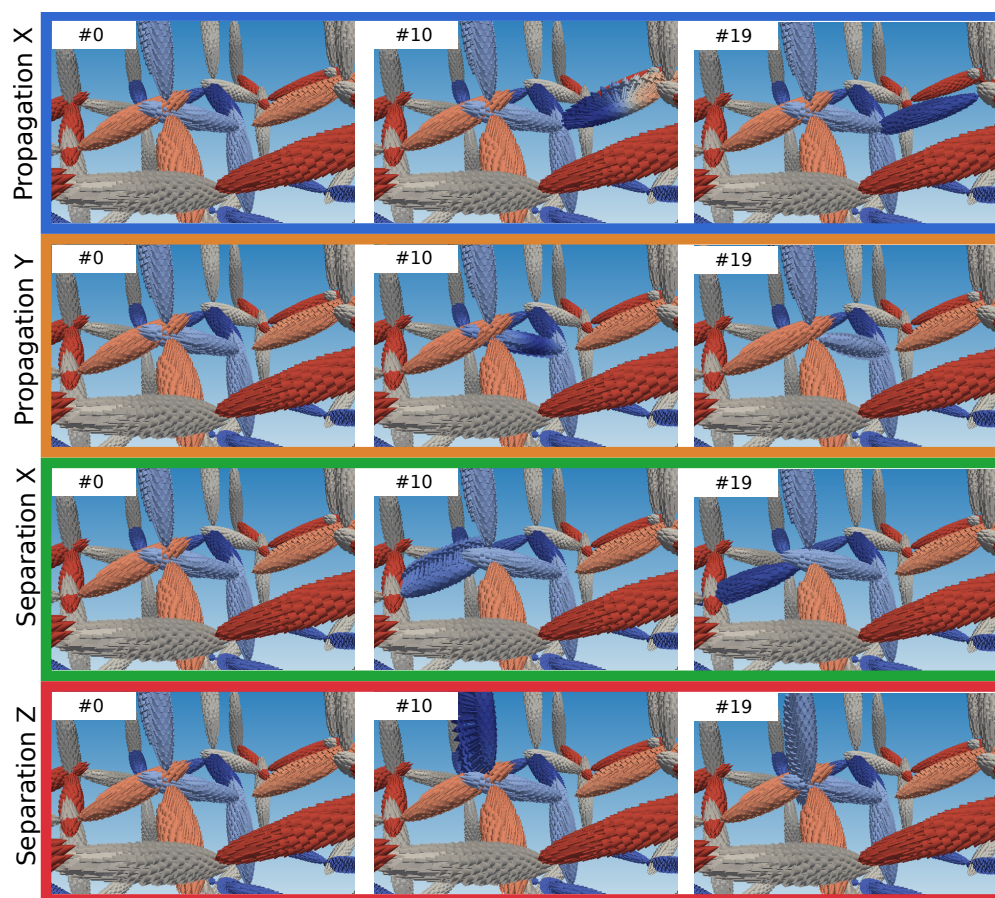
Supplementary Fig. 3: Possible orientations of the magnetizations states in the state-of-the-art square artificial spin ice. Colors illustrated the x -component of the magnetization, as given by the colorbar.



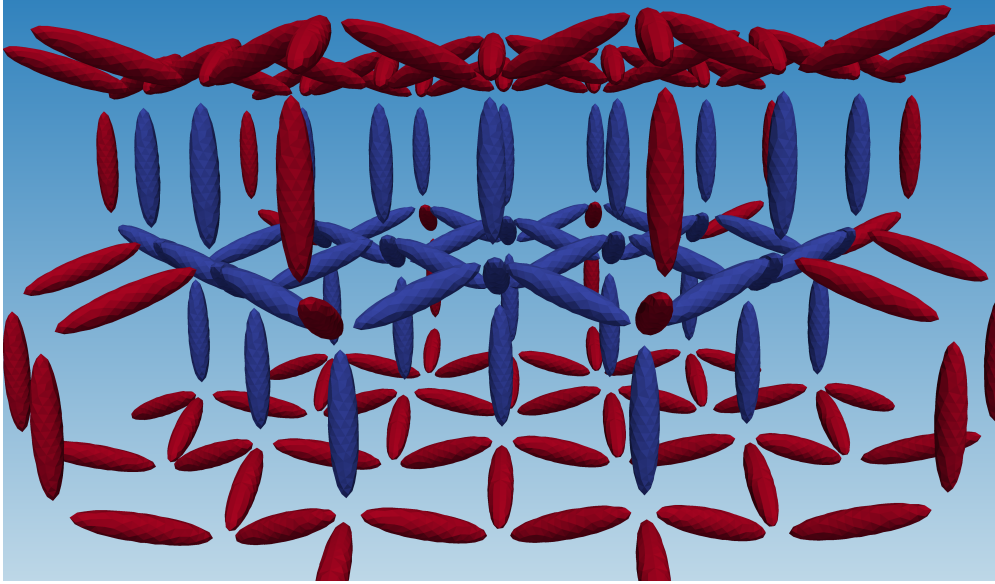
Supplementary Fig. 4: Same as 3 but for 3DASI lattice, where the classical Type I and Type II represent the ground state obeying the ice rule and are rotational symmetric, and one cannot distinguish between them.



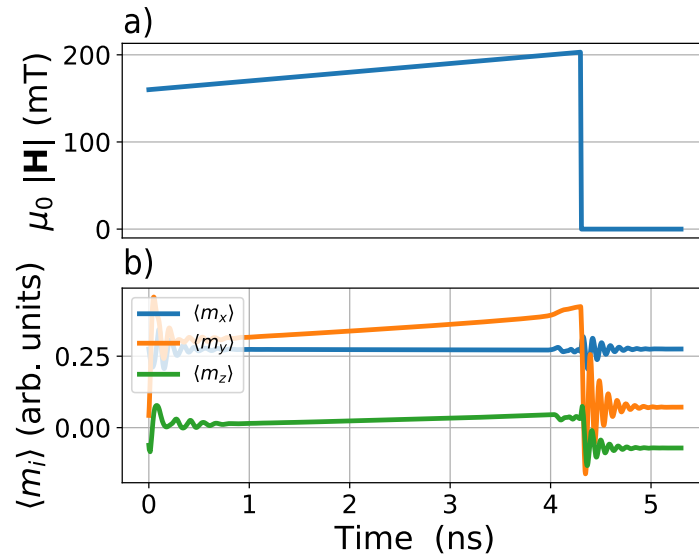
Supplementary Fig. 5: Snapshots of the magnetization states during the string methods simulations taken at different state numbers illustrated in Fig. 3 in the main text. For each row, the initial magnetization state before the switching (left), at the saddle point of the MEP (center) and after the switching (right). The switching occurs over domain-wall propagation, which transfers the Type III defect energy to the next possible vertex.



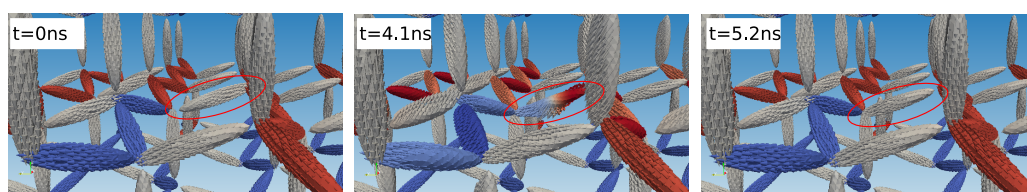
Supplementary Fig. 6: Snapshots of the magnetization states during the string method simulations used for the FTM analysis, and taken at different state numbers, #0 (initial state), #10 (maximal energy state) and #19 (final state) for the relevant transitions: propagation X (blue), propagation Y (orange), separation X (green) and separation Z (red). Colorbar as in Supplementary Fig. 3.



Supplementary Fig. 7: The confinement of the 3DASI lattice to investigate the dynamic properties of magnetic charges by applying external (global) magnetic fields. Red ellipsoids have an artificially set uniaxial anisotropy with $K = 1 \text{ MJ/m}^3$ to confine them, while the blue ellipsoids are used to resolve the magnetization dynamics in a bulk system.



Supplementary Fig. 8: The temporal evolution a) of the magnitude of the applied magnetic field and b) averaged magnetization components within the bulk layer of the 3DASI lattice, where the switching of the ellipsoid of interest takes place over a reversal mechanism dictated by a domain wall nucleation and propagation starting after $t = 4$ ns. At $t = 4.3$ ns the field is cut off in order to relax the magnetic elements back to their ising state.



Supplementary Fig. 9: Snapshots of the magnetization states from the output of the conducted micromagnetic simulations taken at $t = 0\text{ns}$, $t = 4.1\text{ns}$ and $t = 5.2\text{ns}$ where the switching of the ellipsoid of interest (highlighted by the red ellipse) via a domain wall is observed, as it was predicted by the string methods simulations. Colorbar as in Supplementary Fig. 3.

Conclusions

This cumulative thesis includes four publications in peer-reviewed journals. The general topic of all papers revolves around artificial spin ices and the concept of magnetic monopoles, which are created as magnetic charges if energetic ground states are broken within this magnetic meta-materials.

In the first publication, Koraltan *et al.* [37], it was shown that the energy barriers required to switch the magnetizations of nano-islands cannot be calculated by means of simple approximations such as a mean-field approximation. Furthermore, a full micromagnetic model was introduced within the concept of double-vertex approximations, where the collective excitations lead to the lowering of energy barriers, but where also the direction of the reversal starts to play a key role. Subsequently, the notion of direction dependent switching rate is introduced, where the probability of the switching does not take into consideration only the rate with the lowest barrier, but rather a sum of switching rates.

In a second publication, Hofhuis *et al.* [18], the full-micromagnetic from [37] model was used to calculate the energy barriers in a kagome artificial spin ice lattice grown on top of a heavy metal layer: platinum. It was shown in the experiments and confirmed by the full-micromagnetic modeled string method barrier calculations, that the the interface Dzyaloshinskii-Moriya interaction on the interface between Pt and nano-islands induces an anisotropy along the hard axis, which lowers the energy barriers, and lowers the blocking temperature, making it possible to come step closer towards the long-range ordered energetic ground-state in a seven ring kagome lattice.

The third publication, Leo *et al.* [38], the switching mechanisms in double vertices were investigated in more detail, where two main switching mechanisms were found to be the magneto-statically and exchange dominated reversals. Kinetic Monte Carlo simulations have shown that the switching rates calculated as in [37] lead to different spatial and temporal evolution within the square ice lattice, and confirms the necessity of the full-micromagnetic double vertex approximation and chiral switching barriers.

The last publication, Koraltan *et al.* [39], concentrates on a solution to solve the shortcomings of 2D square ice, where the degeneracy of the ground state is lifted and the magnetic monopoles become effectively correlated and bound. In the newly proposed 3DASI lattice, the magnetic ellipsoids replace the planar stadium shaped islands, and have perfect ising behavior. Due to the perfect symmetry of the new lattice, the degeneracy of the ground state is recovered, and it is shown by micromagnetic simulations that all ice rule obeying vertices have equal energies. Furthermore, the full-micromagnetic energy barrier calculations show that the conditions for a tension-free Dirac strings and unbound magnetic monopoles are full-filled by demonstrating the energy barriers for creating the monopole pair are large than the barriers required to propagate them. The latter remains constant independent of propagation length, which is a further proof of unbound magnetic monopoles. The highly mobile and unbound monopoles were then propagated in different directions by applying external magnetic fields, demonstrating the first propagation on-demand of one single magnetic monopole.

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