Temperature-induced collapse of spin dimensionality in magnetic metamaterials

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Spin and spatial dimensionalities are universal concepts, essential for describing both phase transitions and dynamics in magnetic materials. Lately, these ideas have been adopted to describe magnetic properties of metamaterials, replicating the properties of their atomic counterparts as well as exploring properties of ensembles of mesospins belonging to different universality classes. Here, we take the next step when investigating magnetic metamaterials not conforming to the conventional framework of continuous phase transitions. Instead of a continuous decrease in the moment with temperature, discrete steps are possible, resulting in a binary transition in the interactions of the elements. The transition is enabled by nucleation and annihilation of vortex cores, shifting topological charges between the interior and the edges of the elements. Consequently, the mesospins can be viewed as shifting their spin dimensionality, from 2 (XY-like) to 0 (vortices), at the transition. The results provide insight into how dynamics at different length scales couple, which can lead to thermally driven topological transitions in magnetic metamaterials.

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I. INTRODUCTION

Magnetic metamaterials [1] composed of mesospins as building blocks [2] offer the possibility of tailoring magnetic interactions and dynamics in almost arbitrary ways. Previous investigations have mainly been focused on the collective magnetic order, dynamics, and more exotic aspects such as frustration of artificial spin systems [1,3–5], while the internal magnetic order, dynamics, and more exotic aspects such as investigations have mainly been focused on the collective magnetic order, dynamics, and more exotic aspects such as frustration of artificial spin systems [1,3–5], while the internal magnetization and dynamics of the mesospins are less explored [6–8]. For instance, extensive efforts have been made to mimic magnetic systems of various spatial and spin dimensionalities, where the shapes of the elements have been used to enforce mesospins to be Ising- or XY-like [2,9–13]. Even systems consisting of both Ising and XY mesospins have been fabricated and investigated, which is a testament to the versatility of metamaterials [14,15]. The common denominator in all these investigations is that the spin dimensionality is treated as a static property. Yet, the mesospins offer access to continuous degrees of freedom and rich internal magnetic states that go well beyond their atomic analogues. This attribute is the focal point of the present study, as we turn our attention to arrays of mesospins that can exhibit two distinct magnetization states: collinear and vortex spin textures [16,17]. The mesospins can be thought of as having variable spin dimensionality, where the interaction strength depends on the inner magnetic textures. It is therefore possible to couple the changes in spin dimensionality to changes in the collective properties of the mesospins. The coupling of these internal and external degrees of freedom, in combination with the exploration of the role of topological effects on the observed transitions [18,19], is therefore the main motivation behind this work.

II. MATERIALS AND METHODS

A. Sample manufacturing

Two sets of samples were prepared: one set for photoemission electron microscopy (PEEM) studies employing x-ray magnetic circular dichroism (XMCD) and the other set for magneto-optical Kerr effect (MOKE) measurements. The samples were prepared depositing elemental Fe (99.95 at. %) and Pd (99.95 at. %) in an ultrahigh-vacuum (base pressure below $\sim 2 \times 10^{-7}$ Pa) DC magnetron sputtering system, operating using high-purity argon gas (99.995%). The following sample structure was used: fused silica/Pd [40 nm]/Fe$_{13}$Pd$_{87}$ [10 nm]/Pd [2 nm] [20]. By using FePd, the ordering temperature and the spin dimensionality of the parent material can be chosen by the distribution and concentration of Fe, independently of film thickness [21,22]. After growth, electron beam lithography (EBL) was used to pattern the Fe$_{13}$Pd$_{87}$ layers into circular islands arranged on square lattices (see Fig. 1). The Ar$^+$-ion milling process, following development of the EBL resist and subsequent mask deposition, was stopped prior to penetration through the Pd seed layer, providing this way electrical continuity across the whole sample surface. The PEEM-XMCD samples with a disk diameter of $D = [75, 150, 350]$ nm were fabricated having two interdisk distances: one with the nearest-neighbor distance set to

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The synchrotron beam was tuned to the Fe absorption without any external magnetic field. The energy of the sample was cooled from room temperature to roughly 100 K, in a temperature range of 80 K. To understand the role of the interactions, we chose to discuss the size dependence of internal spin textures and spontaneous magnetic order in circular islands. We begin by discussing the role of the interactions, and the interplay between the involved length scales, we need to have a look at the energy landscape of the magnetic textures.

The magnetic energy of the metamaterial can be expressed as \( E = E_{\text{sat}} + E_{\text{j}} + E_{\text{s}} \), where \( E_{\text{i}} \) is the energy cost of the magnetic texture arising from exchange interactions within the islands, \( E_{\text{j}} \) is the magnetostatic energy, and \( E_{\text{s}} \) is the energy associated with the magnetostatic coupling between the islands. By calculating the energy as a function of the vortex core displacement \( d \) for a disk with radius \( R \), a single path—out of many—in the energy landscape separating the collinear and the vortex state can be obtained. Ding et al. used the same reasoning when calculating the energy barrier separating the two states in a single Co dot analytically [28]. Here, we chose to do it numerically as the calculations can be generalized to include interactions of elements in an arbitrary array. The results obtained from calculations of interacting \( (G = 20 \text{ nm}) \) and noninteracting \( (G = 40 \text{ nm}) \) disks with \( D = [75, 150, 350] \text{ nm} \) are shown in Fig. 3. When a vortex state is obtained when the distance between the islands is large, while a substantial number of mesospins have a collinear component, when the distance between the islands is 20 nm. Thus, for this particular diameter of islands, the interaction appears to be strong enough to influence the inner magnetic states. To understand the role of the interactions and the interplay between the involved length scales, we need to have a look at the energy landscape of the magnetic textures.

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The p-polarized incident laser beam with \( \lambda = 659 \text{ nm} \) had a Gaussian profile with a spot diameter of roughly 2 mm. The number of islands contributing to the signal are therefore on the order of \( 10^7 \). The reflected laser beam was passed through an analyzer (extinction ratio of \( 1 : 10^5 \)) and then captured using a Si biased detector, connected to a pre-amplifier and lock-in amplifier. In addition, the lock-in amplifier was used to modulate the incident laser beam using a Faraday cell. The sinusoidal external magnetic field applied along [10] of the square lattice (see Fig. 1) had an amplitude of 40 mT and a frequency of 0.11 Hz unless otherwise stated. The magnetization data in all figures have been binned (3 : 1) for aesthetic purposes.

### III. RESULTS AND DISCUSSION

To determine the magnetic state of the noninteracting and strongly coupled disks, photoemission electron microscopy, employing x-ray magnetic circular dichroism (PEEM-XMCD), was performed at the HERMES beaml ine at the SOLEIL synchrotron [24], and the 11.0.1 beamline at the Advanced Light Source [25]. Prior to imaging, the samples were cooled from room temperature to roughly 100 K, in absence of any external magnetic field. The energy of the synchrotron beam was tuned to the Fe \( L_2 \) edge (708.4 eV), and magnetic contrast was obtained from the asymmetry ratio of intensities between right- and left-handed circularly polarized synchrotron radiation.

### C. Micromagnetic simulations and topological considerations

To understand the role of the interactions, we chose to explore the energies of the vortex and collinear state for different disk sizes, using the MuMax3 micromagnetic simulation software [26]. The simulations, used to mimic the experimental conditions, were performed using one single disk, as well as square arrays of disks \( (G = 20 \text{ nm}) \), with periodic boundary conditions. The saturation magnetization, \( M_{\text{sat}} = 3.5 \times 10^5 \text{ A/M} \), and exchange stiffness, \( A_{\text{ex}} = 3.36 \times 10^{-12} \text{ J/m} \), were chosen based on previous work by Östman et al. [20] and Ciuciulkaite et al. [27]. The in-plane cell size was set to 0.50(1)\( l_{\text{ex}} \), where \( l_{\text{ex}} \) is the exchange length as defined by \( M_{\text{sat}} \) and \( A_{\text{ex}} \) [26].

### D. Hysteresis protocols

The magnetization data were collected using a MOKE system in longitudinal configuration with the sample mounted on a cryostat, in a temperature range of 80 K < \( T < 400 \text{ K} \).
D = 75 nm

D = 150 nm

D = 350 nm

G = 20 nm

G = D + 40 nm

FIG. 2. Diameter and interaction dependence of the disk magnetic texture. PEEM-XMCD images of interacting islands (left column), and noninteracting islands (right column), recorded at approximately 100 K after cooling from room temperature in absence of external fields. Disks with a diameter of 350 nm have a preferred vortex texture for both interacting and noninteracting disks, while disks with 75 nm diameter end up in the collinear state in both cases. Disks with a diameter of 150 nm, display a stabilization of the collinear state when the distance between the islands is small, while otherwise exhibiting vortex textures. The white circles indicate the disk sizes and positions, obtained by overlapping the PEEM–x-ray absorption spectroscopy images.

FIG. 3. The energy landscape of the transition between the vortex and the collinear state. The landscape was obtained numerically by gradually moving the vortex core outward from the center of the disks. Filled symbols represent the energy of interacting disks (G = 20 nm), while empty symbols for noninteracting disks (G = ∞). The shaded areas represent the magnetostatic coupling $E_j$. The energies plotted are normalized to the total energy of the vortex state $E_v$. The energy barrier between the two states can be chosen by the selection of the diameter and the distance between the islands. The top part shows a schematic of the states for different values of $d$, where the red dot indicates the position of the vortex core; when $d > R$, a C state is obtained.

Shifting the core from the center gives rise to a collinear component, and consequently a stray field with a corresponding magnetostatic energy. For the purpose of the calculation, the (virtual) vortex core can even be moved outside the disk ($d > R$), corresponding to a C state (see Fig. 3), with varying degree of gradients in the magnetic texture. The energy maxima obtained at $d \approx 0.9R$ can be viewed as activation barriers separating the vortex and the collinear states. The energy difference between the interacting and noninteracting islands increases with increasing $d$, highlighted by the shaded areas in the figure. The activation barrier separating the collinear from the vortex state is also affected by changes in $G$.

In these simulations we use the energies obtained by MuMax3’s built-in functions for the vortex and the collinear state, without allowing for relaxation of the magnetization with respect to the total energy. Using this approach, the intermediate states can be calculated without the systems collapsing into either the vortex or the collinear state, enabling us to estimate the height of the activation barrier. Qualitatively, the same results can be obtained in relaxed systems by moving the core by means of an applied magnetic field. This approach, however, does not provide any information on the potential in the vicinity of the maximum of the activation barrier. In the simulations with interacting islands, all vortex cores were displaced such that the collinear component of each island was along [10], mimicking the influence of an applied field along [10] as in the MOKE experiments. The choice of either uniform or alternating vortex chirality had a negligible impact on the outcome of the simulations.

The size dependence in the energy of interacting and noninteracting mesospins is summarized in Fig. 4. For noninteracting islands the collinear state has the lowest energy when $D \lesssim 140$ nm, while a vortex state is favored when the diameter is larger. With an inter-island distance of 20 nm, a collinear state is favored up to about $D \approx 190$ nm. Consequently the critical size at which the vortex state is favored is shifted to larger diameters when interactions become prominent. Hence, a region of bistability (marked by a gray shading in the figure) is obtained. We have thereby rationalized the results displayed in Fig. 2, i.e., why the 150 nm islands form vortices in absence of interactions, while a significant amount of the mesospins show a collinear component when
The energy of the collinear state is lowered (blue squares) by inter-island interactions. As a result, it is possible to stabilize collinear states for a range of diameters which otherwise favor formation of vortices (gray shaded area). The energies plotted are normalized to the total energy of the vortex state $E_v$.

While it is trivial to annihilate vortices by applying an external field, the opposite is certainly not true. For this reason we chose to focus on the field and temperature dependence of islands with a diameter of 250 nm and larger (see Appendix A) [29,30]. The states can thereby be defined as having topological charge of +1, determined by $1 - g$, with $g$ being the genus of the structure [19]. The boundary of the islands can host magnetic charges with fractional winding numbers, $w$, which together with the winding number of the bulk charge, $q$, must add up to 1 (see Appendix A) [19,31]. Consequently, the winding of the vortex can be seen as being transferred to the edge of the island when the magnetization changes from a vortex to a collinear state ($d > R$). The transition from a vortex to a collinear state therefore involves no change in the total winding number, while the magnetic cores and their polarity annihilate at the edges of the islands.

While it is trivial to annihilate vortices by applying an external field, the opposite is certainly not true. For this reason we chose to focus on the field and temperature dependence of islands with a diameter of 250 nm and larger ($D = [250, 350, 450] \text{ nm}$), with gaps $G = 20 \text{ nm}$. Disks in this size range spontaneously form vortices when cooled, while the application of an external magnetic field results in a collinear state, allowing us to control the magnetic texture of the mesospins. The stability of the dressed collinear state can thus be investigated by removing the external field while monitoring the magnetization of the samples. Representative magnetization loops for the 450 nm islands, recorded at four different temperatures, are provided in the top half of Fig. 5. The bottom part illustrates the temperature dependence of the magnetization.

At $T = 275 \text{ K}$ the hysteresis loops display a typical vortex nucleation ($H_a$) and annihilation ($H_n$) signature with zero remanence [32], in line with both the PEEM-XMCD results as well as the simulations. At temperatures below $\approx 250 \text{ K}$, the sample exhibits clear remanence, consistent with the presence of a ferromagnetic state. Ferromagnetic response requires alignment of mesospins with a net moment, in stark contrast to the zero-field PEEM-XMCD results. The reduction of the remanent magnetization ($M_r$) with temperature contains both the decrease of the moment of the material (intrinsic material properties) as well as changes in the texture and orientation of the mesospins. To disentangle these contributions we need to identify their signatures. Changes in the material-related magnetization can be described by a power law up to the ordering temperature of the materials ($T_C$), while the temperature de-
temperature of the texture and orientation of the mesospins is unknown. However, separation of the two contributions can be obtained by identifying the difference in their field dependence. For instance, it is sufficient to apply a relatively weak field to remove most of the magnetic texture within a disk, while weak fields only marginally affect the thermally induced excitations of the magnetization in the material. This is seen in the field dependence displayed in Fig. 5: at $T = 275$ K, a field of approximately 6 mT is sufficient for obtaining a transition from a vortex to a collinear state. Nevertheless, this field does little to alter the thermally induced reduction of the magnetization. We can therefore consider the magnetization at a field of 40 mT as predominantly representative for the material; i.e., we define this magnetization as the saturation of the mesoscopic texture ($M_s$). Consequently, the temperature dependence of $M_m = M_s/M_i$, the inferred remanence of the mesospins. A plateau with $M_m = 3/4$ is observed, consistent with constant magnetic texture and thereby a fixed topology of the mesospins, below 220 K. At the same time, a substantial fraction of the moment (1/4) is perpendicular and/or antiparallel with respect to the direction of the net magnetization. At 220 K there is an abrupt change in $M_m$, which vanishes at 270 K. From 270 K and up to the intrinsic Curie temperature of the material, the obtained hysteresis loops are consistent with the presence of vortex states. Thus, we observe a sharp transition from a collective state of interacting mesospins to noninteracting vortex states of the elements with zero net magnetization. To this observation we assign a change from a state with a spin dimensionality of 2 to a state with zero spin dimensionality. This result should be compared with previous findings of Östman et al. where no remanence was observed even at temperatures as low as 5 K, for islands of the same size and similar material composition, but with $G = 50$ nm [20]. This highlights the impact of size and other material properties and complexity that may extend beyond the immediate field of physics [34,35].

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The temperature dependence of the topologically homeomorphic transition is not only defined by the material properties and size of the mesospins but also by their mutual interactions and internal degrees of freedom. In particular, the distance between the elements alters the inter-island interactions and thereby the internal magnetic states, while at the same time, the internal magnetic states affect the interactions and thereby the magnetic order. The interplay between these two length scales leads to an exotic transition, which is to say that it is not based on thermally induced randomization. Rather, the transition pertains to a thermally activated change within the elements, resulting in a collapse of the effective interaction strength and the mesospin dimensionality. The transition discussed here does not have any trivial classical counterpart, calling for a new conceptual framework involving mutual dependence of energy and length scales [33]. The results represent stumbling steps toward understanding emergent properties and complexity that may extend beyond the immediate field of physics [34,35].

The data that support the findings of this study are available from the authors upon reasonable request.
FIG. 6. Magnetic texture and topology of a vortex (left) and a collinear state (right). There is substantial texture in the magnetic state referred to as collinear; however, the net transverse moment is zero. The interior topological charge $q = +1$ (vortex) can be transferred to two topological charges, with $w = +\frac{1}{2}$, at the edges (collinear) [19]. The schematics next to the textures depict the changes in the direction of the magnetization with respect to the edge and the resulting winding number.

and can be driven by applying external fields or thermally induced fluctuations [20]. The amount of topological charge that can be residing in an element is determined by the genus $g$ (number of holes) of the structure, providing an additional knob for the design of texture topology in magnetic metamaterials. This transition is followed by a change in the effective spin dimensionality of the mesospins, which is 0 for the vortex and 2 for the collinear state (see also Table I). Note that in the interior of the mesospins, noninteger winding numbers cannot exist [19,29]. This is not the case though for the edges where half-integer winding numbers are possible and thus topological charges, $w$, which further correlate to stray field emanating from or into the mesospin [19]. These appear in pairs, resulting in integer numbers for the total charge on the edge.

The spin dimensionality $d$ relates to the behavior of the effective mesospin moment, which for planar mesospins can be 0, 1, or 2 dimensional [2,14,16], as illustrated in Table I. The sense of rotation $c$ describes the circulation (clockwise and anticlockwise) of the magnetic texture and acquires nonzero values in the vortex case (0-dimensional mesospin) [36]. The polarity $p$ relates to the direction of the out-of-plane component (up or down) of the magnetization for the core in a vortex. Finally, when considering the planar topology, the bulk topological charge $q$ defines the magnetic texture of the mesospins,

TABLE I. Classification scheme for the planar mesospins.

| Mesospin property | Vortex | Ising | XY-rotor |
|-------------------|--------|-------|----------|
| Dimensionality, $d$ | 0 | 1 | 2 |
| Sense of rotation, $c$ | $\pm 1$ | 0 | 0 |
| Polarity, $p$ | $\pm 1$ | 0 | 0 |
| Bulk topological charge, $q$ | +1 | 0 | 0 |
| Edge topological charge, $w$ | 0 | $\frac{1}{2}$, $\frac{1}{2}$ | $\frac{1}{2}$, $\frac{1}{2}$ |

FIG. 7. Size and frequency dependence of the dimensionality transition. Top: Temperature dependence of the remanent versus the in-field (40 mT) magnetization ratio, for all available disk diameters and for a gap $G = 20$ nm. The values of $T_m$ determined from the inflection points of the fitted curves, versus the reciprocal of the disk diameter $D$, is found in the inset of Fig. 5. Bottom: The frequency dependence of the transition for a disk diameter $D = 450$ nm and gap $G = 20$ nm. A monotonic shift to higher temperature with increasing frequency is observed, compatible with thermally activated relaxation processes of magnetic metamaterial. The orange filled circles in each panel mark the extracted temperatures presented in the respective insets.

APPENDIX B: KINETICS

The observed size dependence (upper panel of Fig. 7) can be compared with results of Ding et al., where the relaxation time of noninteracting Co dots was found to rapidly decrease with increasing dot size at a given temperature [28]. This is quite the opposite of our findings presented here,
leading us to conclude that it is the interactions of larger islands that lead to an increased collinear stability, rather than a direct dependence on $D$. With this in mind, we can understand our observed size dependence. For larger islands, more thermal energy is required to break the bonds between the collinear islands; however, as soon as a few islands do transition into the vortex state, their neighbors suddenly have fewer collinear neighbors with which to interact. This initiates an avalanche effect, where the entire lattice collapses into the vortex state over a narrow range of temperatures. Since larger islands without collinear neighbors have faster relaxation times, the transition becomes sharper for larger islands. This is also seen in our simulations (Fig. 3), where the difference in activation barrier height (from the collinear to the vortex state) between islands with fewer collinear neighbors with which to interact. This further emphasizes the drastic to the vortex state) between islands with the different curves is therefore the temperature at which the transition is activated (when $M_r/M_s$ has reached zero only varies marginally between the samples.

To obtain a better understanding on the interplay between kinetics and thermodynamics, we explored the frequency dependence, $f$, of the applied field on the remanent magnetization. The dwell time at zero field provides a time window for a relaxation, which is proportional to the inverse scanning frequency. The findings are illustrated in the lower panel of Fig. 7, depicting a clear frequency dependence of the response from the 450 nm disks. Increasing the scanning frequency results in an increase of the transition temperatures (inset in Fig. 7, lower panel), illustrating the impact of the kinetic limitations on the transition [8,37]. Consequently, we conclude that the ferromagnetic component observed at low temperatures corresponds to a metastable arrested state. The frequency dependence cannot be captured by a single exponential, highlighting the non-Arrhenius behavior of the homeomorphic transition. We are not aware of any models that can capture the observed behavior.

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