Exquisitely balanced thermal sensitivity of the stochastic switching process in macroscopic ferromagnetic ring elements

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We have studied the repeatability of the magnetic switching behavior of macroscopic ferromagnetic permalloy ring elements using focused magneto-optic Kerr effect magnetometry. In 5 μ m diameter permalloy rings, the circulation of the stable vortex state is found to alternate stochastically from one applied field cycle to the next due to the influence of thermal fluctuations. Micromagnetic simulations are used to demonstrate that the symmetry of the structure creates an exquisitely balanced system where the configuration of a nanoscopic region of magnetic moments dictates the switching mechanism of the entire macroscopic structure, resulting in a degree of sensitivity to thermal agitation usually only observed in structures three orders of magnitude smaller.

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I. INTRODUCTION

Thermal fluctuations play a key role in the switching dynamics of thin ferromagnetic films and elements. It is well known, for example, that nanoscale ferromagnetic particles smaller than a characteristic size may spontaneously flip their magnetization back and forth over a single energy barrier solely through the influence of thermal agitation.^{1–3} The size of this energy barrier scales with the volume and hence, in larger structures with micrometer-scale dimensions, such as have been suggested for use in magnetic biosensor^{4,5} and magnetic random access memory (MRAM)^{6,7} applications, the energy barriers between magnetically saturated states are much greater than thermal energy and spontaneous reversals may no longer occur. Furthermore, to minimize their energy, larger elements tend to exhibit multi-domain structure and reverse via complex domain-wall nucleation and propagation mechanisms involving many small energy barriers.⁸ In such systems, thermal effects generally manifest only through changes in average properties of the system such as the temperature and sweep rate dependence of the field-induced reversal dynamics.⁹ The question therefore arises as to whether the switching of a macroscopic element can be controlled by a single energy barrier corresponding to a very small number of spins, allowing thermal perturbations to have a more fundamental effect on the switching process. To study such effects, it is necessary to design a system with a high degree of symmetry, where changes in the configurations of small regions of spins are more likely to be able to affect the macroscopic switching. Highly symmetric structures may offer various switching routes which are energetically similar, allowing the configuration of small regions of spins to perturb the system into one macroscopic switching route or another. Ferromagnetic ring structures^{10,11} have recently received much interest due to the fact that they support two welldefined stable configurations: the "vortex" state, in which the magnetization circulates around the ring, and the bidomain "onion" state, which is characterized by two head-to-head domain walls. Both states can generally be observed in a

single reversal between saturated magnetic configurations, the onion-to-vortex state transition receiving particular attention in this work due to its unusual nucleation-free domainwall motion mechanism. The symmetry of the ring and in particular the symmetry of the onion-to-vortex transition manifested in such structures makes the ferromagnetic ring system an ideal candidate for studying thermal effects in macroscopic elements. Here we present a study of the repeatability of the magnetic switching mechanism in 5 μ m diameter polycrystalline permalloy ring structures using a focused magneto-optic Kerr effect (MOKE) magnetometer. It is shown that the circulation direction of the vortex state achieved during a reversal alternates stochastically from one field cycle to the next due to the thermal excitation of a small group of spins over a single localized energy barrier. The measurements show that even in these macroscopic structures, thermal effects can profoundly alter the switching mechanism and determine the magnetic states reached during a reversal.

II. METHOD

Polycrystalline permalloy (Ni₈₀Fe₂₀) rings were fabricated on thermally oxidized Si(100) using electron beam lithography and lift-off processing. The magnetic layer was 45 nm thick and capped with 3 nm Au to prevent oxidation. The rings had outer and inner diameters of 5 and 2.5 μ m, respectively, and were positioned with edge-to-edge separations greater than one ring diameter to prevent interactions between elements. Isolated ferromagnetic rings of these dimensions are expected to exhibit two-step magnetization switching behavior which has been well characterized,¹⁰ and is also shown in Fig. 1. As the ring magnetization relaxes from saturation, strong demagnetizing fields produce the onion state. with two head-to-head domain walls forming along the axis of the applied field. At a small reverse field, the Zeeman energy gradient depins one of the domain walls, causing it to sweep through one side of the ring and annihilate with the other domain wall, producing a vortex state in which the



FIG. 1. Hysteresis loop from a single 5 μ m diameter permalloy ring. Schematic diagrams indicating the magnetic states of the ring are also shown.

magnetization circulates around the ring. The magnetization circulation direction (clockwise or anticlockwise) of this state is dictated by which side of the ring is reversed in this initial transition, and therefore depends on whether the depinned domain wall sweeps clockwise or anticlockwise around the structure. The second part of the magnetization reversal occurs at a higher reverse field when a domain nucleates in the unswitched half of the ring and expands to produce the opposite onion state.

The magnetization reversal of the rings at room temperature was measured using a focused MOKE magnetometer in which the probing laser spot could be focused to a diameter of $\sim 2 \ \mu$ m, allowing the measurement of magnetic hysteresis loops from either entire elements or highly localized regions within an element. A sinusoidal magnetic field was applied to the rings at a frequency ~ 2 Hz with amplitude sufficient to switch the ring into the onion state on both the upward and downward field sweeps.

To measure the circulation of the vortex state via which the ring magnetization reversed, the magnetometer laser spot was focused onto the side of the ring, halfway between the idealized positions of the head-to-head domain walls (see Fig. 2). As the circulation direction of the vortex state was determined by which side of the ring was reversed in the initial onion-to-vortex transition, by measuring whether the observed side underwent the low field onion-to-vortex transition or high field vortex-to-reverse onion transition, the circulation of the vortex state could be identified. A more detailed explanation of the method of determining the circulation of vortex states in ring structures may be found elsewhere.¹² The process was automated so that the vortex circulation could be measured for several hundred consecutive reversals, allowing a systematic study of the repeatability of the magnetic switching.

In order to understand the switching of the rings on a nanoscopic level, two-dimensional (2D) micromagnetic simulations were carried out using the freely available OOMMF software package.¹³ Although the computational requirements of simulating a system of the size of the experi-



FIG. 2. Distributions showing the number of reversals over which a vortex circulation was maintained for the (a) clockwise (V+) and (b) anticlockwise (V-) vortex states. The line shows a geometric probability distribution fit to the data. The position of the laser spot relative to the applied field is also shown on a scanning electron microscope image of the ring.

mentally measured ring were beyond what was available, the switching mechanism in ring elements is only weakly dependent on the outer diameter,¹⁴ and hence two smaller rings with outer diameters of 1 and 2 μ m were simulated to understand the factors that determine the vortex state circulation. The thickness of the rings was 45 nm and both had aspect ratios similar to that of the experimental system. The intrinsic parameters used in the simulation were M_s $=860 \times 10^3 \text{ Am}^{-1}$, $A=13 \times 10^{-12} \text{ Jm}^{-1}$, and $K_1=0 \text{ Jm}^{-3}$. The damping constant was $\alpha = 0.5$. A cell size of 4 nm was used, which was smaller than the characteristic exchange length relevant to the type of domain walls formed in rings of these dimensions (Néel), and also represented the edge roughness of the element. As the thickness of the ring was larger than the exchange length of permalloy, a 3D treatment would be expected to be required, however previous works have shown extremely good agreement between experimental results and 2D simulations for elements of similar thicknesses,^{10,12} most likely due to the fact that the geometry of the system confines spin directions to dominantly lie within the sample plane. A 2D simulation was therefore used to reduce computational time.

III. RESULTS

Hysteresis loop measurements with an entire 5 μ m ring focused in the laser spot showed the expected two-step reversal process, with the onion-to-vortex transitions and vortex-to-reverse onion transitions occurring at ±10 and ± 115 Oe, respectively. A representative hysteresis loop is shown in Fig. 1. To study the repeatability of the switching, the circulation of the vortex state was measured from a ring for each reversal over 100 consecutive field cycles. This measurement was performed six times and the data combined, giving a total dataset of 1200 reversals. It was found that, while in general the ring switched its magnetization back and forth using the same vortex circulation on both the upward and downward field sweeps, after a random number of field cycles of switching via one vortex state the ring would unexpectedly begin to switch via the other. Several other rings in the array were observed in this manner, and it was found that all showed similar behavior. To characterize this behavior, distributions were plotted showing the number of consecutive field reversals a ring switched by one vortex circulation before changing to the other (Fig. 2). For both the clockwise (V+) and anticlockwise (V-) vortex states, the distributions are skewed toward low numbers of consecutive reversals, but with a significant number of occasions where a single circulation direction is sustained for 20 reversals or more. To attempt to explain the form of the data, a simple geometric probability distribution was fitted, characterized by a probability, P, of the ring switching to the other vortex state in a subsequent reversal. This distribution took the form $NP(P-1)^{(n-1)}$, where N was the total number of reversals and *n* was the number of reversals over which a given vortex state was stable. The model produced extremely good agreement with experimental data, and fit parameters P=0.17 and 0.12 were obtained for the V+ and V- vortex states, respectively. This indicated that the vortex switching process was stochastic in nature and suggested that, despite the size of the structures, thermal activation was a likely cause of the behavior.

In order to understand how thermal effects could have such an unusually strong influence on the behavior of a macroscopic element, a micromagnetic simulation of the fieldinduced reversal of a 45-nm-thick permalloy ring with a 1 μ m diameter and an aspect ratio similar to the experimental ring was performed, and the factors controlling vortex circulation examined. The main spin configurations observed in the simulation are shown in Fig. 3. As the applied field was reduced from saturation, two approximately symmetric transverse domain walls formed at the top and bottom of the ring [Fig. 3(a)]. When the field was further reduced, the highlighted spins at the top and bottom edges of the ring relaxed from a radial to a circumferential configuration to reduce the stray-field energy of the system. It is noticeable that these two sets of spins point in the same direction around the ring (clockwise) so that they already possess some character of one of the vortex states [Fig. 3(b)]. The relaxation of these edge spins caused the head-to-head domain walls to curve toward one side of the ring. At a lower field still, vortex walls nucleated at the edge of the ring and moved into a central position within the width [Fig. 3(c)].



FIG. 3. Magnetic configurations from a micromagnetic simulation of a 1 μ m diameter permalloy ring with a width of 0.25 μ m and a thickness of 45 nm.

This was expected for rings with thicknesses of the order of the experimental ring as the vortex wall has far lower strayfield energy than the transverse wall, which is seen in thinner structures.^{15,16} It is important to note that the vortex walls both nucleated in the half of the ring into which the transverse walls were curved, suggesting that it was the direction of the edge spins prior to the vortex wall nucleation that dictated the half of the ring in which the vortex walls formed. Just before the applied field reached zero, the side of the ring containing the two vortex walls reversed, producing the vortex state which began to deform asymmetrically as the applied field reversed [Fig. 3(d)]. The transition was initiated by the movement of each domain wall toward the other, which created a "vortex nucleation avalanche" as has been described elsewhere.¹⁷ The circulation of the vortex state was clockwise, as had been suggested by the direction of the edge spins prior to the transition. Eventually the applied field became strong enough to nucleate a reverse domain in the lefthand side of the ring which propagated through the element leaving two vortices pinned asymmetrically at the outer edge as the reverse onion state was reached [Fig. 3(e)]. At a sufficiently high reverse field, the vortices were eliminated producing a transverse wall reverse onion state similar to the initial configuration [Fig. 3(f)]. It is noticeable here that the slight asymmetries in the configuration of the edge spins suggest that if the field had been reversed again, the vortex walls would have nucleated in the left side of the ring, rather than the right side as occurred on the downward field sweep. This would have then caused the right side to reverse first to form the clockwise vortex state and maintain the vortex circulation seen in the first reversal, as observed in the shortterm behavior of the experimental system.



FIG. 4. Simulated variation of Zeeman energy with wall displacement in a 1 μ m diameter permalloy ring.

In the case of the simulated system, the ring spontaneously switched into the vortex state before the applied field was reversed, however in the experimentally studied ring a small reverse field was required to depin the head-to-head walls and initiate the reversal. It is therefore interesting to investigate how a configuration similar to Fig. 3(c) would react to a field applied in opposition to its net magnetization. To study this, one of the vortex walls in the simulated ring was artificially rotated around the circumference of the ring and the Zeeman energy under reverse field calculated at each point (Fig. 4). It was found that a Zeeman energy gradient was created such that rotating the vortex walls back toward a symmetric position at the top of the ring was energetically unfavorable, and hence the applied field would push the vortex domain wall through the side of the ring in which it nucleated, in the same manner as the spontaneous reversal that occurred in the simulation. The implication of this is that, in both a spontaneous and a field-assisted reversal, the initial position of the vortex wall nucleation controls which vortex state is obtained.

The simulation shows that a series of events occurs which uniquely determines which vortex state is formed. First, a relaxation of edge spins causes a curving of the domain wall. This in turn makes it preferable for the vortex walls to nucleate in one side of the ring. The vortex walls are then restricted to reverse the side in which they nucleate, thereby defining which vortex state is created. The simulation also showed that the ring magnetization is expected to reverse back and forth using the same vortex state, as was observed in the short term behavior of the experimentally measured rings, due to the vortex walls nucleating in opposite sides of the ring on the upward and downward field sweeps.

For the cycle to be broken and the opposite vortex circulation to be used, as was observed in the experimental system, it is necessary for at least one of the vortex walls to nucleate in the other side of the ring to that expected, and hence sweep the other way around the ring, creating the opposing vortex state. As the position in which the vortex walls nucleate is controlled uniquely by the direction of the edge spins, the only way that this is possible is for the edge spins to reverse their direction prior to the wall nucleation. Such a flipping of the spins would involve passing through a configuration similar to that in Fig. 3(a), where the edge spins are radially aligned, which would create an energy barrier due to its high magnetostatic energy. This offers an explanation of how thermal agitation could cause the stochastic behavior seen experimentally: although the energy required to affect the states of the entire ring is much greater than thermal energies, the sensitivity of the ring to the configuration of the edge spins creates an exquisitely balanced system of two equivalent reversal mechanisms separated by a highly localized energy barrier.

As the geometry of the focused MOKE apparatus did not allow the experimental temperature to be altered, an alternate set of measurements was taken to validate the model. Switching distributions were measured from a 5 μ m ring at a range of applied field amplitudes. As the deviation angle of the edge spins from a symmetric, radial alignment is dependent on the magnitude of the applied field supporting them, increasing the field amplitude should lower the energy barrier between the two edge spin configurations and increase the rate of switching between vortex states. With an applied field amplitude of $\sim \pm 190$ Oe, it was found that 600 consecutive reversals of the ring proceeded via the anticlockwise vortex state only. It is likely that this was because at the maximum applied field the ring was in a configuration similar to that shown in Fig. 3(e), where two vortex walls are pinned at the outer edge of the ring, making it impossible for the edge spins to reverse and new walls to nucleate in the other side of the ring. After increasing the field amplitude to $\sim \pm 290$ Oe, a total of 12 transitions were measured between vortex states over the same number of field cycles. A further increase to $\sim \pm 400$ Oe produced a total of 33 transitions which allowed approximate values of P of 0.03 for V+ and 0.07 for V- to be obtained. Finally at $\sim \pm 480$ Oe a total of 51 transitions occurred and P was measured to be 0.08 and 0.07 for V+ and V-, respectively. Diagrams indicating how the frequency of the vortex switching increased with field amplitude are shown in Fig. 5. The results show the expected increase in vortex circulation switching as the applied field amplitude was increased, lowering the energy barrier for the edge spins.

To further validate the assumption that thermal activation is the cause of the stochastic behavior, the energy barriers involved in the experimental system were quantified. From the Néel-Brown law,¹⁸ the probability of switching a group of spins over an energy barrier is given by $P = Ne^{-\Delta E/k_B T}$, where N is the number of attempts being made to switch the spins over the barrier, ΔE is the magnitude of the energy barrier, k_B is Boltzmann's constant, and T is the temperature. N is found by multiplying the thermal attempt frequency, which lies in the range 10 MHz to 1 GHz for ferromagnetic thin films, by the time over which the switch is possible, which in this case was assumed to be one-quarter of the applied field period. Using the probabilities measured from Fig. 2, the energy barriers between the two reversal mechanisms were estimated to be 0.4-0.5 eV, depending on the value of attempt frequency used.

A second micromagnetic simulation was carried out for a 2 μ m diameter permalloy ring with a width of 0.75 μ m and a thickness of 45 nm. Although the ring was not as large as the one measured experimentally, the ring width was only 0.5 μ m smaller and it exhibited similarly structured vortex walls, and hence was expected to be a relatively good system to enable a quantitative estimate of the size of the energy barriers involved in the stochastic switching process. The ring switching behavior was observed for several simulated field cycles and it was found that the ring switched via a



FIG. 5. Graphs showing the vortex switching behavior of the ring over 600 reversals, at a variety of applied field amplitudes. (+1) signifies a switch by the clockwise vortex state and (-1) a switch by the anticlockwise vortex state. The applied field amplitudes where (a) ± 190 Oe, (b) ± 290 Oe, (c) ± 400 Oe, and (d) ± 480 Oe.

single vortex circulation during all reversals as expected. The edge spins once again dictated the direction of the vortex state, providing evidence that this is a general phenomenon of vortex wall rings. In the first reversal from saturation, symmetric spin configurations similar to Fig. 3(a) were produced prior to the nucleation of the vortex walls. In the third reversal, it was found that configurations similar to Fig. 3(b), with tilted edge spins, were produced at the same applied fields. This occurred because the ring was initially saturated and hence the configurations reached in the first applied field cycle showed a higher degree of symmetry than those in later reversals, where complete saturation was never obtained. The energy differences between these two types of configurations were analogous to the energy barrier between the two equivalent switching mechanisms and could therefore be

used to estimate its magnitude. It was found that, while at high field the symmetric configurations were energetically favorable, once the applied field decreased, their energy rapidly became greater than that of a configuration with tilted edge spins. Over a limited range of applied fields, similar to those applied to the experimental ring, the energy barrier fell within the 0.4–0.5 eV range measured from the experimental distributions. For example, at 485 Oe the total energies (the sum of the Zeeman, exchange, and demagnetizing energies) were -21610.95 and -21611.32 eV for the symmetric and tilted configurations, giving a calculated energy barrier of 0.37 eV.

The good quantitative agreement between the measured and simulated energy barriers provides evidence that the thermal activation model of the stochastic switching is essentially accurate. However, for a given ring system it is likely that the switching frequency is influenced by other properties of the ring such as edge roughness and shape defects which could also affect the switching of the edge spins, and perhaps bias the ring toward certain configurations.

IV. CONCLUSION

In conclusion, we have demonstrated that macroscopic ferromagnetic ring elements can show an unusually high sensitivity to thermal agitation due to the influence of a highly localized region of edge spins on the switching mechanism of the entire element. The sensitivity of the switching mechanism to the configuration of this region allows thermal fluctuations to disproportionately influence the magnetization reversal, causing stochastic switching. The system can be considered analogous to a rod balanced on one end: the rod can fall one of two ways, and the energy barrier separating the two final states is large, however while the rod is balanced, even a relatively small perturbation may alter its final state. In the system studied, the symmetry of the ring means that the effect of the edge spins is particularly pronounced, however the relaxation of edge spins is observed in most macroscopic element systems, for example in the formation of S and C states in rectangular elements.¹⁹ Liu *et al.* have previously noted that the configurations of edge spins in elongated elements can change the remanent states produced in hard axis reversals,²⁰ providing support for the view that such effects can be important even in lower symmetry elements. In view of this, it seems unlikely that the stochastic behavior we have observed will be confined to ring elements and therefore the action of thermal effects should be considered in the design of macroscopic devices of sizes orders of magnitude larger than those corresponding to the superparamagnetic limit.

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