Coercivity of Ultrathin Films With In-Plane Magnetization

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Abstract

We report numerical and analytic results for a model of coercivity and magnetization reversal in an array of square monolayer-height magnetic islands on a monolayer of magnetic material with in-plane magnetization. Reversal nucleates at step edges where local two-fold anisotropy is present in addition to the intrinsic four-fold anisotropy of the (001) flat surface of a cubic crystal. Simple analytic formulae for the coercive field are derived that agree well with numerical simulations.

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Ultrathin films always have step edges. This is significant because the magnetic anisotropy at sites of reduced crystallographic symmetry can compete successfully with the intrinsic anisotropy of the flat surface and thereby control coercivity and magnetization reversal [1]. In this paper, we study magnetization reversal at T = 0 for a model ultrathin ferromagnetic film with simple cubic crystal structure and monolayer-scale surface roughness. We develop formulae for the coercive field that predict behaviors that compare favorably with results from numerical simulations.

We focus on the case of in-plane magnetization and choose a simple, high symmetry, surface morphology. The model film is composed of one complete magnetic layer on a nonmagnetic substrate with a periodic array of square monolayer-height magnetic islands with side length L and center-to-center separation D placed on top (Figure 1). Since exchange coupling guarantees that atomic moments remain aligned over microscopic distances, a *two-dimensional* classical XY model with spin lengths S_i proportional to the film thickness at lateral atomic site i will be sufficient for our purposes. The magnetic energy is

$$\begin{aligned} E = &- \sum_{\langle i,j \rangle} J_{ij} \cos(\vartheta_i - \vartheta_j) - a^2 \sum_i K_2^i \, S_i \cos^2 \vartheta_i \\ &- a^2 \sum_i K_4^i \, S_i \cos^2 2\vartheta_i - \mu H \sum_i S_i \cos \vartheta_i \end{aligned} \tag{1}$$

where the angles ϑ_i denote the directions of the vector spins $\mathbf{S}_i$ relative to [100], $J_{ij} = J(\min[S_i, S_j])^2$ is the exchange energy between nearest neighbor sites i and j, K_2^i and K_4^i specify the strength of two-fold and four-fold magnetic surface anisotropies at site i, a is the lattice constant, and $\mu = \mu_0 m$, where m is the atomic magnetic moment. We choose the material parameters as $J \sim 10^{-21}$ J, $a = 0.3$ nm, and $m \sim 10^{-23}$ J/T. All sites are assigned a small four-fold anisotropy $K_4 \sim 10^{-2}$ mJ/m^2 and, as suggested by the phenomenological Néel model [2], step edges are assigned a uniaxial anisotropy. The latter is chosen here to lay perpendicular to the local step edge with strength $K_2 \sim 1$ mJ/m^2. These numerical values are consistent with recent experiments [3,4].

The energy expression Eq. (1) includes magnetostatics only through constraining the

magnetization to lie in plane and through the numerical values of the anisotropies which, since they are taken from experiment, include some of the effects of magnetostatics. We do this because it is useful to analyze the effect of competing anisotropies alone so that the consequences of re-introducing the dipolar interactions can be appreciated more readily.

We study magnetization reversal by numerically following the spin configuration at the local energy minimum as a function of applied field by using a combination of the conjugate gradient method and spin relaxation dynamics [5]. Since the domain wall width, $W \simeq 8\sqrt{J/2K_4} \simeq 200a$, is large on the atomic scale, atomic spins are grouped together in 20×20 blocks. This blocking allows study of much larger systems than would otherwise be feasible.

Our calculations support the view that monoatomic steps of single crystal ultrathin films both nucleate rotated domains and impede the motion of domain walls. The combination of these processes and coherent rotation within domains determines the changes in magnetization as the applied field is reversed. For surfaces with no steps, or when the island separation D is small, our model reproduces the Stoner-Wohlfarth result that magnetization reversal occurs by coherent rotation with a coercive field H_C equal to $H_{SW} = 8a^2K_4/\mu$. The magnitude of H_{SW} ($\sim 5 \times 10^5$ A/m $\sim 2\pi 10^3$ Oe) is much larger than typical measured coercivities for ultrathin films. Such a discrepancy between experiment and theory for the coercive field is known as Brown's paradox [6]. For the geometry studied here, we suggest that the paradox is resolved by the nucleation of domains at step edges. The competition between this nucleation, domain expansion through constrictions due to steps, and coherent rotation of domains leads to many types of complex hysteresis loops as a function of island size and separation. Concentrating on the coercive field, we find that the numerical results accord surprisingly well with simple energy balance arguments described below. In particular, we find four regimes for the coercive field where H_C/H_{SW} varies successively as W/L, LW/D^2, $W/(D - L + 2W)$, and $(D - L + 4\sqrt{2}W)/D$ as L/D increases.

In the absence of magnetic fields and island edges, a wall between two domains with orientations $\vartheta = 0$ and $\vartheta = \pi/2$ has energy per unit length $\sigma \propto \sqrt{J\,K_4}$, and wall width

$W \propto \sqrt{J/K_4}$. The wall energy can be written in terms of the wall width as $\sigma \propto H_{SW}\mu W/a^2$. The change in energy when a domain with spins at angle $\pi/2$ (that is, pointing in the +y direction) is introduced in an otherwise uniform system with spins at angle 0 (pointing in the +x direction) is

$$E = \sigma P - \mu H\, A \tag{2}$$

where P and A are the perimeter and area of the $\pi/2$ domain and the applied field is in the -x direction. Eq. 2 can be minimized with the constraints imposed by island edges to determine stable spin configurations. The critical circular domain radius in the absence of island edges is $r_{crit} \propto W/h$. Here and below, we use $h = H/H_{SW}$ to denote fields scaled by H_{SW}. Domains with radii shorter than r_{crit} shrink and domains with radii larger than r_{crit} expand. The critical radius is quite large for small fields. The resolution of Brown's paradox requires a nucleation mechanism that can create domains with linear dimension larger than r_{crit}. We suggest that the uniaxial anisotropy found at island edges can create such domains.

In the model studied here, the island edges running parallel to the applied field have uniaxial anisotropy perpendicular to the field. This uniaxial anisotropy is strong enough that, even before remanence, rotated domains are nucleated at these island edges. After remanence, a domain nucleated at an island edge of length L remains pinned by perpendicular island edges with uniaxial anisotropy parallel to the applied field direction. As the applied field is increased in the negative direction the pinned domain expands and has a lens shape with the same curvature as a circle with radius r_{crit} for $r_{crit} > L/2$. While isolated domains are unstable toward expansion or contraction depending on their radius, a domain pinned at a step of length, L, is stable with a radius of curvature given by r_{crit} for $r_{crit} > L/2$. If the radius of curvature were smaller, the domain would contract, increasing the radius until it reaches r_{crit}. If it were larger, the domain would expand, decreasing the radius until that point. For $r_{crit} < L/2$ the domain expands forever or until a barrier is reached. The four coercivity regimes follow from this result and Stoner-Wohlfarth coherent rotation within domains.

When the island channels are larger than the island edges ($L < D - L$) the domain covers nearly the entire terrace after it bursts at the field defined by $r_{crit} = L/2$. This field is

$$h_L \propto W/L. \tag{3}$$

This is the scaled coercive field if h_L is large enough. Figure 2A shows the numerically calculated [5] coercive field as a function of W/L for three system sizes as labeled. The darkened region of each curve is in this scaling regime and would collapse onto a single straight line if the formula was exact. The scaling form holds quite well for large islands that are far apart. When the islands are small, a more general scaling form holds in which the coercive field is given by some universal function of W/L, independent of D (for D large enough).

If $h_L > 1/(3\sqrt{6})$, the $\pi/2$ state is unstable and the magnetization in the expanding domain rotates from 0 to π when the domain bursts. For smaller values of h_L it only rotates to slightly past $\pi/2$. In this case, h_L is still the coercive field if the spins rotate far enough past $\pi/2$ to compensate the remaining domain walls surrounding island edges with anisotropy parallel to the field direction. These walls have magnetization in the +x direction $M_0 m_2 LW$ and area $a_2 LW$ where M_0 is the magnetization of a unit cell and m_2 and a_2 are dimensionless constants of proportionality. The magnetization for $h > h_L$ is $M_2 = (M_0/D^2)[m_2 LW - (D^2 + L^2 - a_2 LW)h]$. Here, we have used the result that the component of the magnetization parallel to the applied field for a system with four fold anisotropy is equal to $M_0 H/H_{SW}$ for small fields. When the island edges are large enough that the partial rotation past $\pi/2$ does not compensate the magnetization remaining around the island edges, the coercive field is greater than h_L. The magnetization, M_2, is zero at the field

$$h_2 \propto \frac{WL}{D^2 + L^2 - a_2 LW}, \tag{4}$$

so that h_2 is the coercive field when $h_2 > h_L$. The crossover always occurs at a small value of L/D, so that near the crossover we can approximate h_2 by

$$h_2 \propto \frac{WL}{D^2}. \tag{5}$$

Figure 2B shows the scaled coercive field in this second scaling regime. Again, this scaling form works quite well.

For $L > D/2$ the $\pi/2$ domain can't squeeze through the terraces between islands until the field

$$h_3 \propto \frac{W}{D - L - a_3 W} \tag{6}$$

is reached. The parameter $a_3 = 2$ if each island edge is surrounded by a rectangular domain wall of thickness W. For $h_3 < h_2$, h_2 is the coercive field. For $h_2 < h_3$, h_3 is the coercive field. Figure 2C shows the scaled coercive field in the third scaling regime. This regime is quite small. We do not know the exact value of the constant, a_3, so this scaling form cannot be expected to be completely satisfied. We have made Fig. 2C with $a_3 = 2$.

For still larger islands, the net magnetization can be zero even when $h < h_3$ and the $\pi/2$ domains haven't burst through all of the channels. (However, domains can fuse across some of the channels.) The magnetization per spin is $M_4 = (M_0/D^2)[L(D - L + a_4 W)m_4 - (D^2 + L^2 - L(D - L + a_4 W))h]$ so the coercive field is

$$h_4 \propto \frac{L(D - L + a_4 W)}{D^2 + L^2 - L(D - L + a_4 W)}. \tag{7}$$

For $D >> W$ and L close to D this is

$$h_4 \propto \frac{D - L + a_4 W}{D}. \tag{8}$$

When $h_3 < h_4$ then h_3 is the coercive field. When $h_4 < h_3$ then h_4 is the coercive field. It is possible that h_4 is always less than h_3 in which case there are only three regimes and h_3 is skipped. Figure 2D shows the scaled coercive field in the last regime. The data for $D = 32$ does not scale because the field at which the 90° state becomes unstable is less than h_3 and h_4 for small D. We use $a_4 = 4\sqrt{2}$ which assumes rectangular domain walls of length $\sqrt{2}W$ on the islands abutting the island edges.

There are several important considerations relevant to experiments designed to test our analytic predictions. Our numerical results exhibit the best scaling when D is large, i.e., large island separations. This regime requires very slow growth (compared to diffusion times) on very flat substrates. In addition, we have assumed that the reversal is not determined by sample edge effects. Different behavior is expected if reversal is caused by the expansion of domains nucleated at such edges. In this case, the coercive field will be determined by the motion of domains through the constrictions caused by the steps. Finally, since this calculation has not included magnetostatic effects, the films should be as thin as possible. The relative importance of magnetostatics increases in proportion to the thickness of the film.

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FIGURES

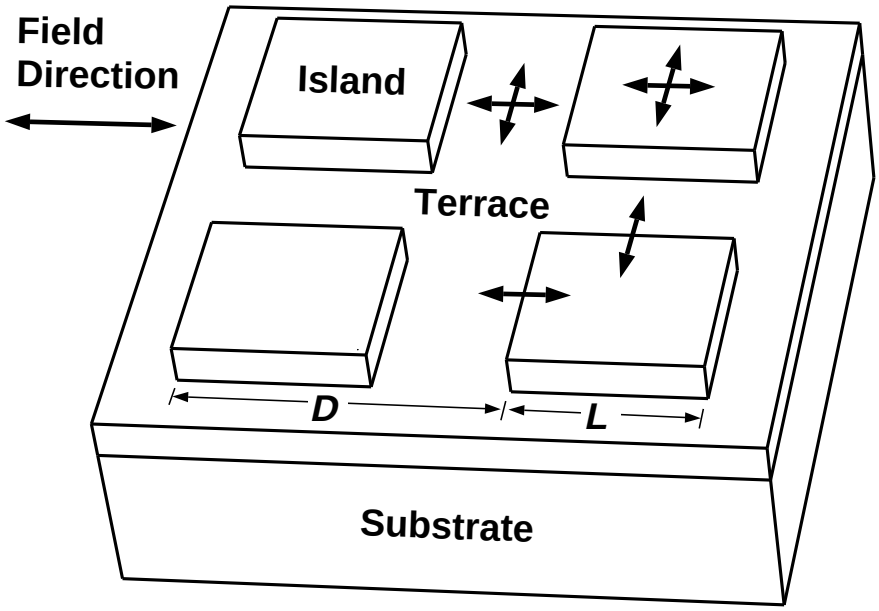

FIG. 1. Schematic view of the rough ultrathin film morphology used in this work. The indicated island geometry is repeated periodically. Arrows indicate local anisotropy axes. In the text, the regions two layers thick are referred to as islands, and the surrounding area is referred to as the terrace. In addition, the parts of the terrace between two islands is sometimes referred to as a channel.

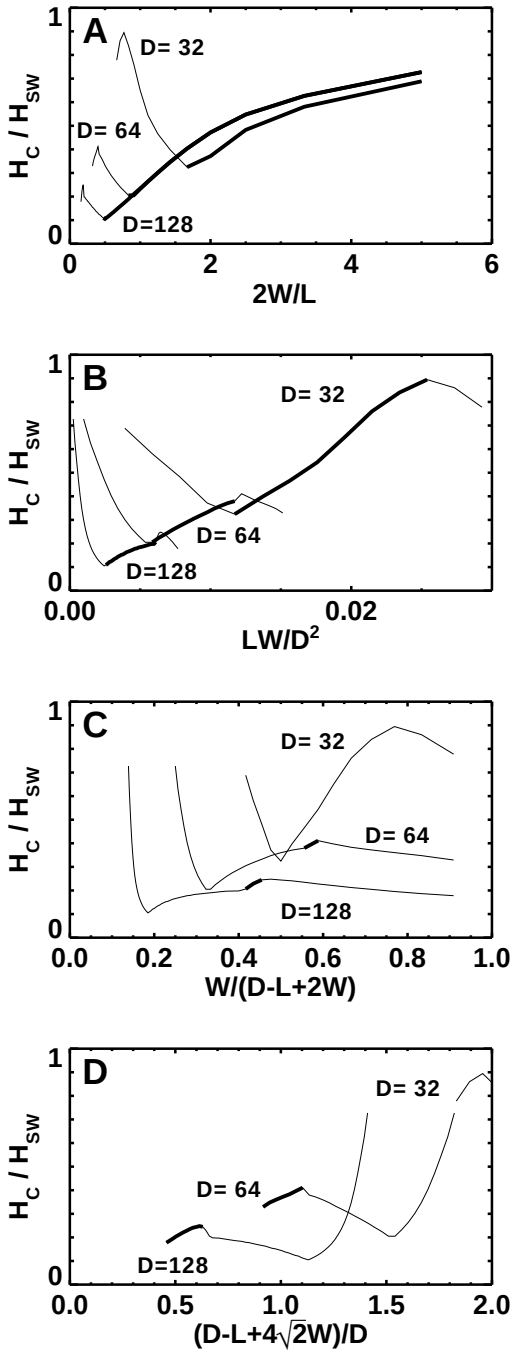

FIG. 2. Coercive field H_C/H_{SW} as a function of several scaling forms. H_C is obtained upon quasistatic reversal of the external field in (1) for the film geometry of Figure 1 for different system sizes as labeled. The four panels show the different scaling forms discussed in the text. If each scaling was exact the darkened portion of each curve would collapse on a straight line.