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Hysteresis loops of Co–Pt perpendicular magnetic multilayers

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Abstract

We develop a phenomenological model to study magnetic hysteresis in two samples designed as possible perpendicular recording media. A stochastic cellular automaton model captures cooperative behaviour in the nucleation of magnetic domains. We show how this simple model turns broad hysteresis loops into loops with sharp drops like those observed in these samples, and explains their unusual features. We also present, and experimentally verify, predictions of this model, such as the temperature dependence of the hysteresis loop shape, and the existence and time dependence of drops in first-order reversal curves. We suggest how insights from this model may apply more generally.

1. Introduction

Magnetic thin films with magnetization oriented perpendicular to the plane of the film have become attractive for recording as magnetic bits become smaller. Cobalt–platinum multilayers are thin films with desirable characteristics for future recording media, such as large ratio of coercive field (field on the major hysteresis loop where $M = 0$) to saturation magnetization and sharp initial drops in their hysteresis loops. Although grain sizes are currently too large for applications, they are interesting model systems, both as high coercivity thin films (Weller *et al* 2001), and as a testing ground for Ising model based simulations of hysteresis in real materials.

These films are also of interest as interacting dynamical systems. In particular, the precise onset of magnetization reversal, which has been observed before in Co–Pt multilayers (Weller *et al* 2001, Phillips *et al* 2001, Hatwar and Blanton 1997, Bennett *et al* 2001, Della Torre *et al* 2000), suggests the shape of the hysteresis loop may be dominated by nucleation. This is the case for some magnetic thin films of interest in recording (Phillips *et al* 2001, Hatwar

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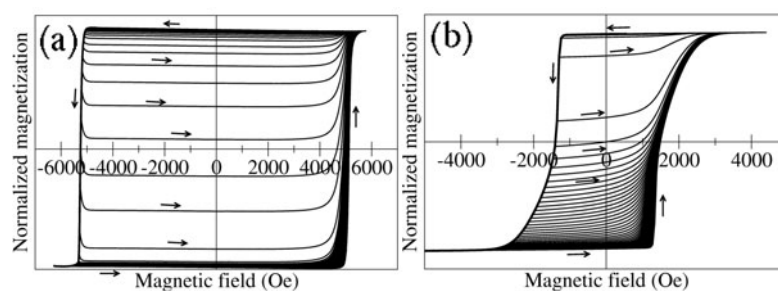


Figure 1. Major hysteresis loops (envelope) and first-order reversal curves for two samples provided by Terris at IBM. Sample (a) shows a sudden sharp drop in the major hysteresis loop, while sample (b) shows a similar drop as the magnetization begins to fall, but then has a broad tail.

and Blanton 1997, Mansuripur 1995). Nucleation phenomena are of broad interest, e.g. in phase transitions (Langer 1968), materials synthesis (Fan *et al* 2003), and protein aggregation (Slepoy *et al* 2001). Unresolved issues for these Co–Pt multilayers include the origin of the broad tail in the hysteresis loop near magnetic saturation and the drop in first-order reversal curves following reversal of the magnetic field (figure 1). Additionally, differences between nominally similar samples are not well understood.

Here we obtain the major hysteresis loop and first-order reversal curves (measured by decreasing the magnetic field along the major hysteresis loop until a predefined reversal field, then tracing out a reversal curve as the field is again increased to saturation) for two Co–Pt multilayer samples, then develop a lattice-based model to describe key features of these measurements. The model makes testable predictions concerning time dependence of the reversal curves and temperature dependence of the hysteresis loop, which we test. Our model is remarkable not in terms of quantitative agreement with experiment, but in the physical insight it provides.

2. Materials and data collection

Two Co–Pt multilayer samples were provided by Terris at IBM. These samples are similar in composition to those described by Weller *et al* (2001) and consist of a 20 nm Pt buffer layer, followed by 10 Co(0.6 nm)/Pt(1 nm) bilayers. Growth temperatures for the multilayers were 157 and 320 °C for the samples of figures 1(a) and (b), respectively. Reversal curves (figure 1) were taken with an alternating gradient magnetometer (Princeton Measurements Corporation). The procedure for each reversal curve, unless otherwise noted, was to decrease the magnetic field every 0.1 s until reaching a predefined reversal field. Then, the magnetic field was gradually increased again to trace out a first-order reversal curve. Along this reversal curve, the magnetization was collected and averaged over 0.13 s at each data point.

For temperature-dependence data, we used a Quantum Design superconducting quantum interference device (SQUID) magnetometer in the standard (DC) operating configuration with the sample perpendicular to the applied field. We measured full hysteresis loops at 5 K and then just the descending half loops at 300 K.

3. Experimental results

Experimentally, we find that the hysteresis loops of these two samples exhibit several unusual features. To our knowledge, a simple explanation of these has not yet been provided. Particularly, we note the broad tail (as the magnetization approaches saturation) shown in

figure 1(b). We believe this is especially remarkable. Modelling in terms of exchange coupling can produce sharp drops due to an energy barrier associated with nucleation of reverse domains, but the rounding at the tail is typically comparable to the rounding at the onset (as the magnetization begins to fall) (Miyashita *et al* 2002, Zhu and Bertram 1991). Here, we find that the tail is much broader than the onset. This has been observed previously (Phillips *et al* 2001, Bennett *et al* 2001, Della Torre *et al* 2000). Additionally, we notice a drop in the reversal curves in figure 1(a) following reversal of the magnetic field. That is, the magnetization continues to fall even after the magnetic field begins to increase. This phenomenon has been observed without comment in a similar material (Della Torre *et al* 2000). Here, we explain these features. Additionally, we find that despite the fact that the materials involved in these two samples are quite similar there are striking differences in the hysteresis loops.

Our results using the SQUID magnetometer will be discussed below after presenting initial results from our model, as the SQUID was used to test predictions of our model.

4. Model and computational results

4.1. The model

Detailed microscopic behaviour of magnetic samples like these is admittedly complex. But Sethna *et al* have argued that one should be able to trade realistic microscopic degrees of freedom for a set of rules as part of a renormalization procedure to study different classes of hysteretic behaviour at long length and timescales (Sethna *et al* 2001). While some multiparameter micromagnetic (time-dependent Landau–Lifshitz–Gilbert equation) models can produce hysteresis loops similar to those observed here (Lyberatos 1998), we present a simpler model which describes these features in a simple, physically transparent manner and, we believe, may capture much of the essential physics of these samples.

Stochastic cellular automata models, such as the one we use here, have been used successfully before to model nucleation barriers (Slepoy *et al* 2001). Ours involves spins—representing individual magnetic bits that are either up or down—on a 2D triangular lattice. The key model ingredients are as follows.

- (1) *Anisotropy field.* Each spin is initially assigned an anisotropy field, defined as $H_k = 2K/(\mu_0 M_s)$ where K is the anisotropy energy. $-H_k$ can be thought of as the field at which an isolated spin would flip from up to down, and conversely for $+H_k$. Thus each spin has a coercivity and is inherently hysteretic. We assign different H_k values to each spin, typically according to a lognormal distribution. All of our results presented here use a lognormal distribution, but we have tried Gaussian distributions and get similar results with slightly less broad tails (see below for discussion of tail breadth). The H_k values can also be thought of as pinning fields for individual spins. Since our goal is to develop a minimal model which can reproduce the observed hysteresis loops for these samples, we assume that the distribution of anisotropy fields is fixed as a function of temperature.
- (2) *Energy barrier.* At each step, we first treat each spin independently and compute an energy barrier for it to flip. For an up-spin, in the case of magnetic field $H > -H_k$, we use $\Delta E = \frac{AH_k}{2} \left(1 + \frac{H}{H_k}\right)^2$, where A is a constant, and for a down-spin in the case of $H < H_k$ there is simply a sign change for the second term inside the quadratic (Bertotti 1998). The barrier is zero otherwise.
- (3) *Attempt probability.* We evaluate the probability of the individual spin attempting to flip, $P = \exp(-\Delta E/k_B T)$, where T is an input parameter, and determine whether the spin attempts to flip. We do this for every spin on the lattice and keep a list of those that attempt.

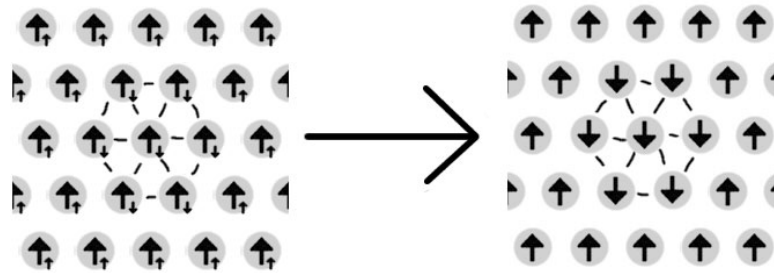


Figure 2. Left, a group of seven spins, where each of the seven attempts (small arrows) to flip down in the same step is the smallest group of spins that can stably flip down (right) in our rules. Thus our rules provide a nucleation barrier to formation of a domain of down spins near $M = 1$, and similarly when near $M = -1$ for formation of a domain of up spins.

- (4) *Cooperative rules.* We then apply rules which require cooperativity between spins and establish a nucleation barrier. The model we use is as follows: for a spin to flip from up to down, it must have at least three neighbours that are already down or will flip down in the same step (and similarly going from down to up). Thus the smallest size group of spins that can flip down is 7, as in figure 2. This effectively builds in a nucleation barrier and allows us to explore what effect this has on the hysteresis loop. These rules play a diminishing role after initial nucleation.
- (5) *Magnetization and field.* After updating every site on the lattice, we calculate and output the magnetization and step the magnetic field. This represents one simulation step. We then repeat this procedure until reaching saturation.

It is important to note that we do not wait until the sample equilibrates before stepping the field. We thus link a simulation step with some small amount of real time, τ , which has been done successfully before to model kinetic effects in hysteresis loops (Rao *et al* 1990). The relevant timescale for this may be the ‘attempt frequency’ for crossing an energy barrier, which is generally considered to be on the order of a nanosecond (He *et al* 1996). In our simulation, it is the ratio of the size of the magnetic field step to the attempt frequency, τ , which sets the shape of the hysteresis loop and the magnitude of the drop in the first-order reversal curves, which will be discussed below. In all our results presented here this ratio is kept fixed at unity, except when we mention the effects of increasing or decreasing field sweep rate.

In our model, we end up with essentially one adjustable parameter, the temperature, which also serves as a measurement of the importance of disorder. Thus, although we can also adjust the width of the lognormal distribution we use, this has essentially the same effect as decreasing the temperature, and therefore we have not adjusted this width in the results we present here. We can also shift the mean of the distribution, but this seems to affect only the coercivity or width of the hysteresis loop, not its qualitative shape, so again, we have not done this in the results we present here.

4.2. Computational results

For our results, we simply run the model we described above, on a 200×200 lattice (we have tried larger and smaller lattices (data not shown) to ensure that results do not depend strongly on size). Our results are shown in figure 3. First, we examine the behaviour of the model without cooperativity (4, above), and find that, in that case, our model yields hysteresis loops that are quite broad. These loops acquire a sudden drop in magnetization due to the nucleation barrier

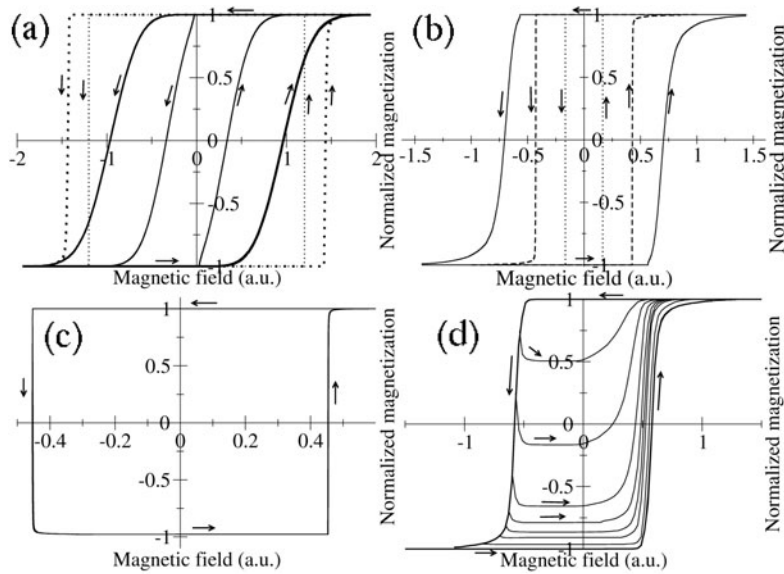


Figure 3. Some hysteresis loops from the model. Arrows indicate direction of hysteresis. (a) Hysteresis loops with and without cooperative rules for two different temperatures, $T = 0.04$ (thicker curves) and $T = 0.1$ (thinner curves). The broad curves (solid curves) are the results when all the spins act independently; the sharp curves (dotted curves) show the effect of the nucleation barrier established by our cooperative rules. (b) Hysteresis loops at different temperatures ($T = 0.03, 0.1$, and 0.3 ; solid, broad dashes, and dots, respectively). Notice the overall sharpening at higher T and the much broader tail at lower T . (c) Calculated first-order reversal curves at $T = 0.1$. Shown here are reversals at $M = 0.9, 0.5$ and 0 , but the curves essentially all overlap because in all three, even following reversal, magnetization continues to drop nearly to $M = -1$ before stabilizing. (d) First-order reversal curves and envelope at $T = 0.05$. Here, first-order reversal curves are more reasonable. Note that the magnetization does drop following reversal. We find that the amount of this drop decreases at lower temperatures or for more broad distributions of H_k .

when we apply the cooperative rules (figure 3(a)). The temperature dependence of this effect is somewhat unusual (figure 3(b)), in that the drop is actually sharper at higher temperature and both the drop and the tail are broader at low temperature.

The explanation for these effects follows: the sudden drop in the hysteresis loop is due to the relief of the constraints of the cooperative rules following nucleation of domains. That is, once the nucleation barrier is crossed, domains can grow, and it is much easier to add additional spins to a domain (a spin that is still up neighbouring a down domain already has some down neighbours and so it is easier for it to have sufficient down neighbours to flip down) than it was to nucleate the domain in the first place. This is similar to a previous description of reversal in terms of two coercivities, a nucleation field H_n necessary to nucleate reversed domains and a field H_p necessary to overcome domain wall pinning. If $H_n > H_p$, reversal can occur very rapidly following nucleation (Weller *et al* 2001, Phillips *et al* 2001). However, our model is different in that we do not have a single pinning coercivity. We refer to the large cascade of spin-flips following nucleation as ‘avalanching,’ as described in random-field Ising models (Perkovič *et al* 1995), although in the Ising model reversed domains tend to pull their neighbours along with them, while here the only driving force is the external field.

This avalanching applies to the temperature dependence of the drop in that at high temperature more thermal energy is available to allow the avalanches to proceed. More thermal

energy means bigger avalanches and sharper hysteresis loops, because it helps to overcome outlying H_k values that would tend to pin domain walls. Higher temperature means the disorder is less important and so avalanches are bigger. Low internal disorder has been shown to produce sharp hysteresis loops with large avalanches before (Perković *et al* 1995, Berger *et al* 2001), but our model includes both internal disorder and the effects of temperature, and shows how temperature affects this sharpness.

The broadening of the tail at low temperatures arises from a freezing in of the hysteresis loop to follow the actual distribution of H_k at low temperatures. That is, in the tail of the hysteresis loop, our rules play little role (most remaining up spins have enough down neighbours they can flip as soon as it is favourable for them individually to do so) and spins act independently, so the tail samples the distribution of H_k and must follow it more closely at low temperatures. This can be seen in figure 3 by the rejoining of the $T = 0.04$ curves in the tail region. This tail behaviour emerges naturally from our model. It has been observed before and its origin hinted at in phenomenological Preisach modelling (Della Torre *et al* 2000) but we have provided a simple physical explanation.

We also find (figures 3(c), (d)) that our model produces a drop in the first-order reversal curves. At reasonably high temperatures (i.e. $T = 0.1$ in our model) it greatly exaggerates this effect (figure 3(c)). Lower temperatures or broader distributions of H_k make this effect more reasonable (figure 3(d)). Additionally, our model predicts increasing field sweep rate will shrink this effect. Taking bigger steps in magnetic field gets back to the field necessary to stop the reversal curves from dropping sooner, so they do not fall as far. The drop simply due to avalanching—once a domain nucleates, it can grow easily and a significantly less negative field is required to stop the avalanche of domain growth. Consider, for example, a spin with a low H_k which, if isolated, would be easy to flip. If it has several higher H_k neighbours it will not flip until long after it would be favourable for the spin, if isolated, to flip. But once its neighbours flip, it is favourable for it to flip, even if the magnetic field reversal begins. This also means that, here, the asymmetric tail is present even when the distribution of anisotropy fields is symmetric (i.e. Gaussian).

It is interesting to note that even though our simulation lattice is 200×200 , which is fairly small, this still allows multiple sites of spin reversal. One might naively expect that, since our model establishes such a strong nucleation barrier, immediately after nucleation a single domain would propagate through the whole sample. While we have not exhaustively examined domain structures, we do see nucleation of multiple domains in many cases. Thus our nucleation barrier seems to be helping to produce a precise onset of magnetization reversal by allowing averaging (over clusters of spins) to play a large role. That is, it is not single-spin anisotropy fields that are important to set the onset of magnetization reversal, but rather the average anisotropy fields of the weakest clusters of spins.

4.3. Experimental predictions and tests

Based on these results, we would predict that experimentally one should be able to allow the sample to equilibrate before taking a reversal curve and the effect should disappear, since it is a kinetic effect. On the other hand, slowing the field sweep rate along the reversal curve should allow avalanches to proceed further and the effect should get bigger. This was tested experimentally and confirmed for sample (a) (figure 4) and the drop also appears for sample (b) if the field sweep rate is slowed enough (Pike 2002).

If we assume our theoretical model is capturing this effect accurately, τ provides the conversion between simulation steps and experimental time. Thus a crude estimate of the effective τ for these samples is on the order of a few milliseconds. This is obviously much

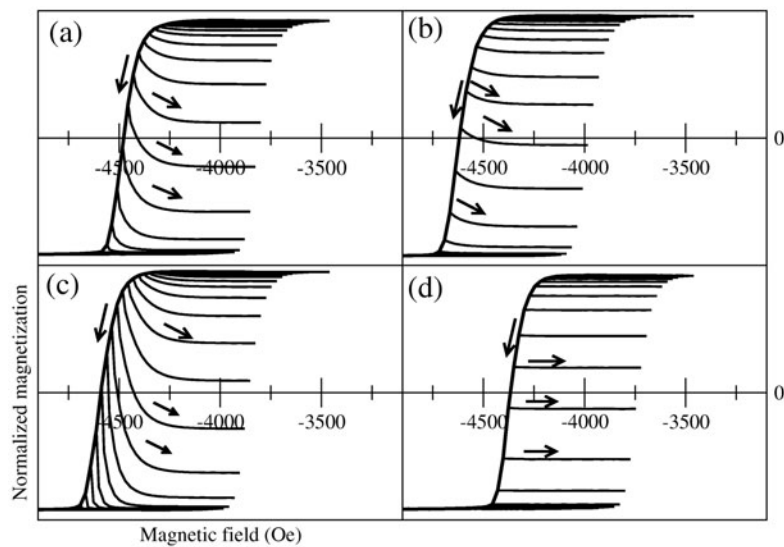


Figure 4. Close-ups of the region where the reversal curves drop show the time dependence of the effect for the sample of figure 1(a). The major hysteresis loop (descending half) is shown in broader lines; partial first-order reversal curves (truncated on the right) are in narrower lines. Essentially, when there is more pausing before or during measurement, the magnetization drops farther. (a) The normal reversal curves are taken by decreasing the field in 0.1 s steps, then increasing and averaging for 0.13 s at each field point. (b) Averaging for 3 s at each field point gives avalanches more time to proceed and the magnetization drops further. (c) Equilibrating for 60 s prior to taking data on the reversal curve eliminates the effect but puts the magnetization lower at a given field. (d) As (b) but the field sweep time prior to reversal is also slowed to 3 s.

larger than the typical value of around a nanosecond mentioned above, and would suggest that these samples are unusual. This is consistent with the observation that this drop is not typically observed in experimental reversal curves for other materials. On the other hand, our simulation dynamics are artificial, so they need not be directly related to the timescales for the real system. One would expect some kind of scaling of the simulation times with the timescales for the real system, but this remains to be explored.

To check our prediction of tail broadening at low temperature, we used a SQUID magnetometer as described in the data collection section to compare hysteresis loops at 5 K with those at 300 K. Results are shown in figure 5; both samples show a broadening of the tail at lower temperatures, in qualitative agreement with the predictions of figure 3(b). However, our model's hysteresis loops have a coercivity that is strongly temperature dependent, but only one of these samples does.

5. Discussion and conclusions

Overall, our results are especially significant in that we qualitatively explain the experimentally observed drop in the first-order reversal curves as a kinetic effect due to avalanching and provide experimentally verified suggestions for reducing or increasing this effect. This drop has been observed in another material and attributed to thermal relaxation (Basso *et al* (2000) also called creep by Nattermann *et al* (2001))—thermal growth of domains at fields below that necessary to induce domain depinning. While thermal energy allows this drop to be larger in our model, we would argue both on theoretical and experimental grounds that our mechanism is distinct.

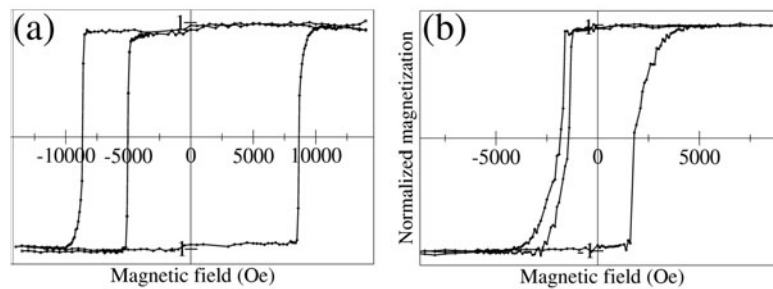


Figure 5. Descending half of the major hysteresis loops for our samples as measured at two different temperatures with a SQUID magnetometer. Curves with the most negative coercivity are 5 K; the others are at 300 K. The 5 K curves also show the ascending half of the major loop. (a) The same sample as figure 1(a) shows a significantly larger coercivity at lower temperature (leftmost curve) and a broader tail, while sample (b) shows a slightly broader tail (leftmost curve) but little change in coercivity.

Experimentally, we find that kinetics strongly influence this effect. And in our model, even at $T = 0$, there is still a nucleation barrier which can result in avalanches and produce a (smaller) drop in reversal curves (unpublished data).

Additionally, our cooperative rules produce an asymmetric hysteresis loop tail. This is due to nucleation, because the nucleation barrier has a profound effect on the initial drop in magnetization. The effects of our cooperative rules disappear by the tail, allowing the tail to still be broad and individual spins to act independently. This effect has only been captured before in vastly more complicated models (Lyberatos 1998) or by parameter-tuning (Della Torre *et al* 2000).

Within our model, there are two possible origins of the experimentally observed broad tail. The first is the mechanism we describe here, which does not depend on having an asymmetric distribution of H_k . To our knowledge, this effect is absent in models dealing in terms of exchange coupling, as exchange coupling strong enough to produce a nucleation barrier will also tend to prevent a broad tail. However, strongly asymmetric distributions of H_k can produce an asymmetry as well. Independent measurements of the distribution of anisotropy fields will be very useful to distinguish between these two cases.

Our model suggests the importance of inherently hysteretic spins, or hysterons, rather than Ising spins, where hysteresis comes solely from interactions between spins. In samples like these, with strong uniaxial anisotropy, there may be two separate energy scales, one associated with the energy barrier to flip individual grains, and one governing the exchange coupling between neighbouring grains which determines a cooperative nucleation barrier. This may suggest that the single parameter of the exchange coupling in Ising models is not sufficient to describe hysteresis in materials like these.

The fact that nucleation is an irreversible process, as described above, helps to create a broad tail in our model despite the absence of any significant rounding at the top, and without invoking asymmetric distributions. On the other hand, models based on Ising spins may not capture this behaviour, as the deviation from fully polarized state at the top and bottom of the hysteresis loop would be due to reversible flips of outliers and thus would be symmetric. Hence, we suggest that interacting hysteron models, or cooperative nucleation models of the type discussed here, will be better for describing the hysteresis in materials with broad asymmetric tails.

Experimentally, it would be useful to attempt to measure the distribution of anisotropy fields for these materials, as this could be used to distinguish between the two potential

explanations of the broad tail mentioned above. This distribution can be evaluated experimentally (Bottoni *et al* 2000), and if it is found to be symmetric, this would highlight the importance of the nucleation barrier in these materials.

A further place in which future work on modelling in this area could make contact with experiment is that of domain structure. There have been suggestions that the domain structure of thin films like these can give insight into the magnetization reversal process—in particular, the relative strength of the domain nucleation field (H_n) and domain wall pinning field (H_p) discussed above (Phillips *et al* 1996, Miyashita *et al* 2002, Zhu and Bertram 1991, Hellwig 2004, Zhang *et al* 2001). Domain size appears to be strongly influenced by the number of layers in Co–Pt multilayers (Hellwig 2004, Phillips *et al* 1996). Examining the domain structure of samples like those examined here could provide insight into why the shapes of the hysteresis loops are so different for these two samples. However, there is a difficulty in comparing theoretical domain structures from our model with experimental ones. Our simulations are done on a 200×200 lattice, but how big is one lattice site? A lattice site should probably represent a grain, but we lack good data on grain sizes, so it is difficult to know whether domain sizes in our simulation would match with those from experiment or not. If this question can be resolved, such a comparison will likely prove valuable.

In conclusion, our model qualitatively explains important hysteresis features of these samples and predicts experimental kinetic effects. We explain the drop in the reversal curves and the broad tail. Our results suggest that exploring Ising-type models with exchange coupling and inherently hysteretic spins may give new insight into some materials.

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