Recently experimental techniques, such as magnetic force microscopy (MFM), have enabled the magnetic state of individual sub-micron particles to be resolved. Motivated by these experimental developments, we use Monte Carlo simulations of two-dimensional kinetic Ising ferromagnets to study the magnetic relaxation in a negative applied field of a grain with an initial magnetization $m_0 = +1$. The magnetostatic dipole-dipole interactions are treated to lowest order by adding to the Hamiltonian a term proportional to the square of the magnetization. We use droplet theory to predict the functional forms for some quantities which can be observed by MFM. One such quantity is the probability that the magnetization reversal may occur in individual single-domain ferromagnets indicates that the switching mechanism in such particles may involve local nucleation and subsequent growth of droplets of the stable phase.

I. INTRODUCTION

The processes by which magnetization reversal occurs in the nanoscale ferromagnets that will make up the next-generation recording media are the subject of active research. One quantity for which theory and experiment often disagree is the lifetime $\tau$, which is the time required for a particle with initial magnetization $m_0 = +1$ to reach $m = 0$ when a magnetic field in the $-\hat{z}$ direction is applied. Micromagnetics, a theoretic technique in which differential equations are numerically solved on a coarse-grained lattice, predicts that the lifetime is given by the Arrhenius equation

$$\tau \propto \exp(\beta \Delta F)$$

(1)

with $\Delta F \propto L^d$. Here $\beta^{-1}$ is the temperature in units of energy, $\Delta F$ is the free-energy barrier between the stable and metastable phases, and $L$ is the linear system size. This same prediction is made by the standard Néel-Brown theory of single-domain ferromagnets. The evident failure of Eq. (1) with $\Delta F \propto L^d$ for somewhat larger grains is ascribed to the existence of more than one domain in larger particles, as is a corresponding peak in plots vs. $L$ of the switching field $H_{sw}$, which is the field required to yield a given lifetime.

Recently experimental techniques such as MFM have been used to resolve the magnetic properties of isolated, well-characterized single-domain particles (see, e.g., Ref. [3]). This is an important advance, since previous experiments on ferromagnetic powders left uncertainties due to the range of grain sizes and orientations and the local magnetic environments. Observations of individual particles by MFM have made it clear that even for many single-domain particles, Néel-Brown theory is inadequate.

We have applied the statistical-mechanical droplet theory of metastable decay to nanoscale ferromagnets with large uniaxial anisotropy, and compared Monte Carlo simulations of square-lattice Ising systems with droplet-theory predictions. (For a review of droplet theory, see Ref. [4].) The agreement between theory and simulation is quite good, and despite the crudeness of the Ising model as a model for real magnets, it shows good qualitative agreement with the MFM experiments. We find rich $L$-dependent behavior in the standard Ising model, even though its equilibrium structure is a single domain for all $L$. This suggests that for some strongly anisotropic magnetic materials, magnetization reversal may occur through the nucleation and growth of non-equilibrium droplets. Details of our work are given in Refs. [5] and [6], including formulae for general dimensionality. For simplicity, we only discuss the two-dimensional case here.

II. APPLIED DROPLET THEORY

To be concrete, consider a two-dimensional kinetic Ising ferromagnet ($s_i = \pm 1$) with Hamiltonian

$$\mathcal{H} = -J \sum_{n.n.} s_i s_j - H \sum_i s_i + L^{-2} D \left( \sum_i s_i \right)^2$$

(2)

and Metropolis single-spin-flip dynamics on a square lattice with periodic boundary conditions. The last term represents a mean-field approximation for the dipole-dipole interaction energy and is taken to be zero except as noted. The critical radius of a “droplet” of $s_i = -1$ spins surrounded by $s_i = +1$ spins occurs when the free energy of the droplet $(2\pi R_0 - \pi R_0^2 |H|)$ is maximum: $R_c \approx \sigma / 2 |H|$, where $\sigma$ is the surface tension per unit length. Droplets smaller than this will very probably
shrink and vanish; larger droplets will very probably grow and reverse the magnetization of the system. In a sufficiently large system, the probability per unit time that a critical droplet forms, centered at a given site, is given by droplet theory as

$$I \propto |H|^3 \exp \left( -\beta \pi \sigma^2 / 2 |H| \right).$$  \hspace{1cm} (3)

The details of the magnetization reversal depends on the number of critical droplets the system forms.

For weak fields or small systems \((L < R_c)\), no critical droplet can form. This is called the Coexistence (CE) Region, and since two interfaces (remember: periodic boundary conditions) must form to reverse the magnetization,

$$\tau \propto \exp \left\{ \beta \left[ 2\sigma L - O \left( H L^2 \right) \right] \right\}.$$ \hspace{1cm} (4)

For slightly larger \(L\), the first supercritical droplet will grow to the size of the system before another one can form. The lifetime in this Single Droplet (SD) Region is

$$\tau \approx \left[ t/4 J \right]^{-1}.$$ \hspace{1cm} (5)

In both the CE and SD regions, switching is a Poisson process, so the standard deviation of the lifetime is comparable to \(\tau\). Both Eq. (4) and Eq. (5) are actually special cases of the Arrhenius equation, Eq. (1), but in neither case is \(\Delta F\) proportional to \(L^d\). Note that if \(\tau\) is held constant and the system size is increased, Eq. (4) implies that the magnetic field must increase, whereas Eq. (5) implies that the magnetic field must decrease. This shows that the peak in \(H_{sw}\) occurs near the crossover between the CE and SD regions (see Fig. 1).

FIG. 1. The relation between the switching field \(H_{sw}\) and the system width \(L\) for two different fixed lifetimes (solid curves), calculated by kinetic Ising model simulations at \(k_B T = 0.8k_B T_c \approx 1.815 J\). The dotted curve is near the crossover between the CE and SD regions; the dash-dotted curve is near the crossover between the SD and MD regions.

For a system in the SD region, \(P(m > 0) = \exp \left( -t/\tau \right)\) is shown as a function of field in Fig. 2 for \(L = 30, 100, 300\). The solid curves are fits of droplet theory predictions to the MC data. The dashed curve is the fit of the droplet-theory prediction for \(L = 100\) extrapolated to \(L = 300\).

FIG. 2. The probability that \(m > 0\) for a kinetic Ising system in the SD region. \(T = 0.8 T_c, t = 914\) Monte Carlo steps per spin (MCSS) and \(L = 10\). The solid curve is a fit of \(\exp(-t/\tau)\) to the MC data, where \(\tau\) is given by Eq. (4). The inset figure shows the fitted curve over a wider range in \(H\).

FIG. 3. The probability that \(m > 0\) for a kinetic Ising system in the MD region. \(T = 0.8 T_c, \tau = 40.7\) MCSS and \(L = 30, 100, 300\). The solid curves are fits of droplet theory predictions to the MC data. The dashed curve is the fit of the droplet-theory prediction for \(L = 100\) extrapolated to \(L = 300\).

The probability that the magnetization is greater than zero \(P(m > 0)\) is shown as a function of field in Fig. 3 for a system in the SD region. This probability is what is most easily observed in MFM experiments, and it decays exponentially with time in both the CE and SD regions. In the SD region, the system is very unlikely to return to the metastable state from the stable state, so

$$P(m > 0) = \exp \left( -t/\tau \right).$$ \hspace{1cm} (6)

In the CE region such backwards switching takes place
on a timescale comparable with the initial decay, so the situation is more complicated.

For sufficiently large \( L \) or \( H \), several supercritical droplets may form before any one of them has grown to the size of the system. This is the Multi-Droplet Region (MD). Such systems were first studied by Kolmogorov, Johnson and Mehl, and Avrami, and have a lifetime

\[
\tau \approx \left[ \frac{I\pi v^2}{3\ln 2} \right]^{-1/3},
\]

where the radial growth velocity \( v \) is assumed to be proportional to \( |H| \). Although \( \tau \) is independent of \( L \), the variance of the lifetime is proportional to \( (v/L)^2 \). Measuring \( P(m > 0) \) as a function of \( H \) or \( t \) thus provides a means of estimating the proportionality constant between \( v \) and \( H \) (see Fig. 3). Details are given in Ref. \([5]\).

The addition of the dipole-dipole interaction energy in Eq. (2) makes the forms for the lifetime somewhat more complicated than we have presented here. In the CE and SD regions, a form of the Arrhenius equation [Eq. (1)] still applies, and switching is still a Poisson process, so Eq. (6) still applies for the SD region. In the MD region, the system evolves in a time-dependent effective field, \( H_{\text{eff}} = H - 2Dm(t) \). The time-dependent magnetization can be found to \( O(D^2) \) fairly easily, and we find numerically that the \( O(D^2) \) correction is relatively small. We can then solve analytically for \( \tau \) to \( O(D^2) \) and find good agreement with simulation results (Fig. 4). A detailed treatment of the \( D > 0 \) case is given in Ref. \([7]\).

\[\text{FIG. 4. The lifetime } \tau \text{ for a kinetic Ising system in the MD region, normalized by the lifetime in a similar system with } D = 0. \; T = 0.8T_c. \; \text{The solid curves are droplet-theory predictions.}\]

III. CONCLUSION

We have used Monte Carlo methods to simulate magnetization switching in two-dimensional kinetic Ising ferromagnets. The results of the simulations can be well explained by droplet theory and show good qualitative agreement with experiments, despite the comparative simplicity of the Ising model. This simplicity, in turn, allows us to develop an understanding of the underlying statistical mechanics. Particular features to make the model more realistic, such as appropriate boundary conditions, quenched randomness, and less rigorous anisotropy will be added in later studies.

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