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Disorder and coercivity in magnetic particle systems

Yang Zhao and H. Neal Bertram

Center for Magnetic Recording Research, University of California, San Diego, La Jolla, CA 92093, USA

Correspondence to: Dr. Y. Zhao, Center for Magnetic Recording Research, University of California, San Diego, La Jolla, CA 92093, USA.

Computer simulation has been utilized to understand the hysteretic behavior of magnetic particle systems. Particles are assumed to be single domain, spherical in shape, and possess no intrinsic anisotropy. Neighboring spins are not exchange-coupled. Particles are either randomly packed or displaced from cubic lattice positions in random directions. The gyromagnetic equation of motion with Landau—Lifshitz damping is solved for each spin during a dynamic process. Regular spin arrangements yield no hysteresis or coercivity. For spin configurations randomly displaced from a cubic lattice, hysteretic behavior is observed. Coercivity increases with randomness of particle position. Detailed examination of the magnetization reversals reveals chain formations and transient vortex states in randomly packed arrays. Coercivity versus packing fraction inferred for finite sized particles shows a maximum.

1. Introduction

New media with high coercivities, along with fine chemical and mechanical properties, have great potential for high density recording. Such materials include granular metal films consisting of ultrafine ferromagnetic particles (such as Fe, Ni, Co, etc) embedded in an insulating matrix (such as SiO₂, Al₂O₃). Composite thin films of Fe—SiO₂ have drawn special attention for their outstanding magnetic properties [1,2]. Microstructural studies reveal that at iron volume fractions below the percolation threshold, the single-domain iron particles are roughly spherical in shape, randomly positioned, and generally isolated by the amorphous silica matrix [3,4].

In this work, a dynamical micromagnetic model has been utilized to understand the effect of particle position randomness on array coercivity [5]. Unlike a lattice having cubic symmetry, where at least for saturation the net dipole field is identically zero at all spin sites, long range magnetostatic interactions in random granular films are randomly distributed both in magnitude and direction. It is therefore interesting to see how the disorder of magnetostatic interactions introduced by random particle position yields hysteresis and a finite coercivity.

2. Simulation

In this model, particles are assumed to be single domain, spherical in shape, and possess no intrinsic anisotropy. Furthermore, particles are not exchange-coupled. For later comparisons with random arrays of particles, hysteresis curves are initially calculated for ordered systems, in which the spins are positioned on cubic or hexagonal lattice points and confined in a spherical or rectangular space with free boundaries. Random particle arrays can be generated by directly packing finite sized spheres provided that the particles do not overlap. An alternate approach will be to

displace spins from their cubic lattice positions, randomly, a finite distance in all directions. This latter method actually gives a convenient measurement of particle position randomness: the maximum distance a certain spin can deviate from its ordered state, which can be called the amplitude of random position perturbation. Thus, by varying this perturbation amplitude, we can gradually go from an ordered system to a disordered one.

The system energy is comprised of magneto-static interactions and Zeeman energy due to a uniform external field:

$$\begin{aligned} E_{\text{tot}} &= E_{\text{mag}} + E_{\text{ext}} \\ &= -\frac{1}{2}\nu \sum_{i \neq j} \frac{3(\hat{\mathbf{r}}_{ij} \cdot \mathbf{M}_i)(\hat{\mathbf{r}}_{ij} \cdot \mathbf{M}_j) - (\mathbf{M}_i \cdot \mathbf{M}_j)}{r_{ij}^3} \\ &\quad - \mathbf{H}_{\text{ext}} \cdot \sum_i \mathbf{M}_i \end{aligned} \quad (1)$$

where ν is particle volume, M_i the magnetization of i th spin and r_{ij} the spatial separation between i th and j th particles. Interactions between spin pairs are of simple dipole-dipole form. The effective field on each spin is then calculated from

$$\mathbf{H}_{\text{eff}} = -\frac{\partial E_{\text{tot}}}{\partial \mathbf{M}}. \quad (2)$$

The gyromagnetic equation of motion with Landau-Lifshitz damping is solved numerically during a dynamic process with an adiabatically decreasing external field [3]:

$$\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} - \frac{\lambda}{M} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{\text{eff}}). \quad (3)$$

It is found that the precession term causes a significant increase in computation time, and calculation results are insensitive to the damping constant for large damping. Here, the first term which represents the gyromagnetic rotation was neglected. Computations are performed for 250-300 particles within aspherical sample shape.

The energy density can be rewritten in reduced form:

$$\begin{aligned} \frac{E_{\text{tot}}}{pM_s^2} &= -\frac{1}{2} \sum_{i \neq j} \frac{3(\hat{\mathbf{r}}_{ij} \cdot \hat{\mathbf{m}}_i)(\hat{\mathbf{r}}_{ij} \cdot \hat{\mathbf{m}}_j) - (\hat{\mathbf{m}}_i \cdot \hat{\mathbf{m}}_j)}{\frac{r_{ij}^3}{l^3}} \\ &\quad - \frac{\mathbf{H}_{\text{ext}}}{pM_s} \cdot \sum_i \hat{\mathbf{m}}_i, \end{aligned} \quad (4)$$

where M_s is the saturation magnetization, $l^3 = V/N$ with V the sample volume and N the number of spins. The volume packing fraction p is defined as $N\nu/V$. Given a particle position distribution, the external field is scaled with the particle volume packing fraction and saturation magnetization. Therefore, magnetization curves may be plotted in a scaled form as M/M_s vs H_{ext}/pM_s for a given configuration. A linear increase in the switching field is therefore expected with increasing

packing fraction. This scaling law holds as long as the increase in packing fraction causes no change in the distribution of relative particle locations, or r_{ij}/l .

3. Results and discussion

3.1. Ordered systems

For arrays of particles positioned on a cubic or hexagonal lattice, no hysteretic behavior is observed. Fig. 1 shows the magnetization curve for an array of particles positioned on cubic lattice and confined in a spherical space. Initially all spins are pointing vertically due to a sufficiently large external field in that direction. As the applied field is reduced, the magnetization pattern remains unchanged until a nucleation field is reached. If the applied field is lowered below the nucleation field, spins start to rotate away from the field direction. The magnitude of this nucleation field depends on the sample shape. Arrays with a larger dimension along the external field direction nucleate at smaller positive field due to reduced sample shape demagnetization. In figs. 2a and b the reversal configuration after nucleation is illustrated. In fig. 2a, a vertical plane is shown at $H_{ext}/pM_s = 1.6$, which reveals columnar buckling along vertical lines. This behavior is similar to the reversal of a single column of spins. Since vertical particle pairs experience twice as much magnetostatic interaction as the horizontal pairs in a saturated state, relatively weak coupling between vertical columns is expected at the beginning of reversal. In fig. 2b, a horizontal plane is shown at the same external field. Coherent vortex correlation between groups of four vector chains is observed.

As the external field is further lowered, particle magnetizations gradually rotate into horizontal planes. Fig. 3a shows the same vertical plane as in fig. 2a near zero external field, and fig. 3b, the corresponding horizontal plane. At $H_{ext}/pM_s = 0.0$, spins lie in horizontal planes, where vortex structures are clearly present. In some planes parallel to the external field, anti-ferromagnetic patterns are found. The same calculation has been repeated for hexagonal structured arrays with an overall rectangular sample shape, and zero coercivity is again found regardless of external field orientation with respect to the hexagonal lattice.

3.2. Disordered systems

When the spins are randomly displaced from their cubic lattice positions, magnetization curves exhibit hysteresis and a finite coercivity. As the random perturbation amplitude, defined as the maximum distance to which particles are allowed to deviate from their cubic lattice positions, is increased, a greater array coercivity is observed. Fig. 4 shows the reduced coercivity $H_c I^3/m_s$ vs random perturbation amplitude A_r/l , where l is the lattice spacing and m_s the magnetic moment of each spin. The curve would be smooth and show little fluctuation if significantly larger arrays had been utilized.

Finite coercivity and increase in the reduced coercivity with increasing disorder may be attributed to the clustering of magnetic dipoles due to their positional disorder. As the array is randomly perturbed, the pair correlation function of spin position broadens. Some particles get close enough to each other that they can be treated approximately as an isolated system from the rest of the array. Dipole interactions within the cluster significantly exceed those from the rest of particles. As pointed out by Lybertos and Wohlfarth [6], the effective interaction fields within a particle system can be either predominantly positive or negative. Some clusters have an overall positive interaction. A simple example is a pair of spins positioned along the external field direction. Since a negative external field is required for nucleation, these spin clusters contribute to

the coercivity of the entire array. On the other hand, some clusters have an overall negative interaction, such as two spins in a plane perpendicular to the applied field, and nucleation of these clusters occur before the external field is reduced to zero. Hence, the latter clusters do not contribute to the coercivity. As the amplitude of random perturbation increases, more clusters are formed, and stronger interparticle interactions arise within clusters. Consequently, the array coercivity increases.

3.3. Coercivity and packing fraction

The packing fraction of randomly perturbed arrays is related to the amplitude of position perturbation in a way to keep dipole spheres from overlapping. The closest distance between two spins should be no less than the particle diameter when calculating the packing fraction for a particular array generated by perturbation:

$$d = l - 2A_r, \quad (5)$$

where d is the particle diameter, A_r the random perturbation amplitude, and l again as defined by $l^3 = V/N$. It follows that the higher the positional disorder, the smaller the array packing fraction. An extreme case is a closely-packed hexagonal array in which the highest packing fraction is achieved for identical spheres. From fig. 4, a relationship can be obtained between the coercive force and the percentage packing by recognizing that

$$p = \frac{\pi d^3}{6l^3} = \frac{\pi}{6} \left(1 - \frac{2A_r}{l} \right)^3. \quad (6)$$

in fig. 5, H_c/M_s is plotted against p . For small p , the scaling law of magnetostatic interaction dominates, and coercivity increases with p ; as p gets large, particle position randomness decreases, and eventually this causes a decrease in coercivity.

Assuming a finite particle size, a random array of single-domain particles can be directly generated for a specific packing fraction. Simulations are then repeated for randomly packed arrays with different packing fractions. Results show that coercivity plateaus at high packing fractions (fig. 6). This is again explainable by the fact that since each particle has a finite size, array randomness decreases as more particles are packed.

3.4. Reversal process of a randomly packed array

Hysteresis has been explored for systems with a small number of grains in order to investigate cluster formation. Dynamical equations are solved for forty single domain particles randomly packed in a cubic space. Examination of detailed reversal processes reveals chain reversals and transient spin vortices (figs. 7a-f). The spin configuration at $H_{ext}/M_s = 0.8$ is shown in fig. 7a. Spin chains aligned vertically remain in that direction, while spins with close horizontal neighbors rotate away from the initial saturation direction to form horizontal chains reducing the magnetostatic energy. At $H_{ext}/M_s = 0.0$, a vortex of four spins forms in the upper-right corner after the reversal of the marked spin (fig. 7b). At $H_{ext}/M_s = -0.8$ (in fig. 7c), a two-spin chain in the center-front corner (as indicated by short arrows) reverses; near the top-left corner, another two-spin chain (as indicated by the long arrow at top of the figure) reverses its direction from left to right, weakly joining the curved four-spin chain on the rear-left side of the cube (indicated by the long arrow on the left side of fig. 6c). At $H_{ext}/M_s = -1.4$, the vortex

formed earlier in the upper-right corner ceases to exist due to the reversal of its two spins, and this causes the three spins just below it to reverse downward as well (fig. 7d). As H_{ext}/M_s reaches -1.6, a three-spin chain in the front-left (marked by 3 arrows) reverses abruptly (fig. 7e). Finally at $H_{ext}/M_s = -2.0$, the four-spin chain on the rear-left side breaks up and reverses (fig. 7f), almost leading the loop to closure. As the external field is further increased, spins rotate reversibly towards the field direction to reach saturation.

4. Conclusion

Dynamic reversal processes of magnetic particle assemblies have been studied by numerically solving the gyromagnetic equation of motion with Landau-Lifshitz damping. M - H curves show little hysteresis behavior for arrays of particles positioned on cubic or hexagonal lattice. Disordered systems reveal hysteresis and coercivity increases with array randomness. Chain reversals and transient vortex states are observed during the reversal process of randomly packed arrays. Coercivity versus packing fraction inferred for finite sized particles shows a maximum.

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Fig. 1.

Fig. 2.

Fig. 3.

Fig. 4.

Fig. 5.

Fig. 6.

Fig. 7.

Fig. 7. (continued)