June 2001

Cooperative magnetism and the Preisach model

Ralph Skomski
University of Nebraska-Lincoln, rskomski2@unl.edu

David J. Sellmyer
University of Nebraska-Lincoln, dsellmyer@unl.edu

JOURNAL OF APPLIED PHYSICS VOLUME 89, NUMBER 11 1 JUNE 2001

Cooperative magnetism and the Preisach model

R. Skomski[a] and D. J. Sellmyer
Department of Physics and Astronomy and Center for Materials Research and Analysis, University of Nebraska, Lincoln, Nebraska 68588

Cooperative and noncooperative magnetization processes in magnetic nanostructures are investigated. Using model calculations it is shown that the Preisach model and related approaches, such as Henkel, ΔM, and ΔH plots, describe magnetism on a mean-field level and cannot account for intra- and inter-granular cooperative effects. For example, the ΔM plot of a nucleation-controlled two-domain particle gives the false impression of a positive intergranular interaction. A simple but nontrivial cooperative model, consisting of two interacting but nonequivalent particles, is used to show that cooperative effects are most pronounced for narrow switching-field distributions, i.e., for nearly rectangular loops. This is unfavorable from the point of magnetic recording, where one aims at combining narrow loops with a noncooperative local switching behavior. A general rule is that the neglect of cooperative effects leads to an overestimation of the coercivity.
[DOI: 10.1063/1.1355344]

I. INTRODUCTION

The distinction between cooperative and noncooperative phenomena is a key feature of modern physics. The term cooperative refers to the simultaneous involvement of two or more particles, as opposed to an ensemble of noninteracting particles or particles whose interaction is mapped onto a mean field. Atomic-scale ferromagnetism is essentially cooperative, because interatomic exchange keeps neighboring atomic spins well aligned on a local scale, and low-lying excitations are spin-wave excitations rather than single-spin flips. However, most magnetic materials encountered in practice are nano- or microstructured, and the behavior of a crystallite in a magnetic material may well be noncooperative or ''single-grain'' like.

In a sense, important approaches such as the Preisach model,[1] Wohlfarth's remanence relation, and Henkel plots[2,3] rely on the existence of well-defined magnetic particles or grains embedded in a magnetic environment. From a practical point of view, it is necessary to distinguish between *intra*-granular cooperative phenomena inside a grain or particle and *inter*-granular cooperativity caused by interactions between particles or crystallites (Fig. 1). Nanomagnetic cooperativity gives rise to a variety of phenomena. For example, delocalized nucleation modes such as curling[4,5] are intra-granular cooperative effects, the activation volume in magnetic-viscosity and sweep-rate experiments is determined by the degree of cooperativity,[6] and random-anisotropy scaling laws[7,8] are a direct consequence of intergranular exchange. Cooperative effects are undesired in magnetic-recording media, where they tend to reduce the storage density,[6] but desired in permanent magnets and soft-magnetic amorphous alloys, where they suppress the effect of anisotropy-field fluctuations.[5,9]

This work investigates analytically how cooperative phenomena affect the reliability of mean-field-type approaches and how they manifest themselves in properties such as the coercivity.

II. REMANENCE ANALYSIS

The simplest approach is to treat the interaction of a given grain or particle (index i) with the environment on a mean-field level. This is achieved by introducing an interaction field of the type

$$\mathbf{H}_{i,\mathrm{MF}} = \Sigma_i \mathscr{T}_{ik} \mathbf{M}_k , \quad (1)$$

where $\mathbf{M}_k$ is the magnetization of the kth grain and $\mathscr{T}_{ik}$ is, in general, an interaction tensor. This interaction field is then added to the external field $\mathbf{H}_i$ in order to trace the magnetization $\mathbf{M}_i$ as a function of the local field $\mathbf{H}_i = \mathbf{H} + \mathbf{H}_{i,\mathrm{MF}}$.

This mean-field approach is implied by a variety of approaches. First, it is the basis of the Preisach model,[1] which has found applications in various areas of magnetism.[10–13] Second, it is exploited by remanence-analysis methods based on Wohlfarth's remanence relation,[14] such as Henkel plots,[2,3] ΔM plots,[15,16] and ΔH plots.[17,18] Wohlfarth's remanence relation

$$M_D(H) = M_R(\infty) - 2M_R(H) \quad (2)$$

predicts the dc demagnetizing remanence $M_D(H)$ as a function of the isothermal remanence $M_R(H)$.

As emphasized by Wohlfarth,[14] the applicability of Eq. (2) is limited to noninteracting fine-particle ensembles. In the case of interactions, it is suitable to use the Henkel plot,[2,3] where M_D is plotted as a function of M_R and Eq. (2) yields a straight line. Alternatively, one can plot the deviation from the ideal value M_D in Eq. (2) as a function of M_R

$$\Delta M = M_D(H) + 2M_R(H) - M_R(\infty). \quad (3)$$

[a]Electronic mail: rskomski@unlserve.unl.edu

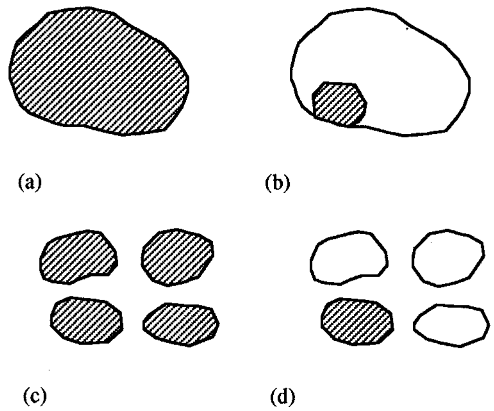

FIG. 1. Cooperativity in ferromagnetic nanostructures: (a) intra-granular and cooperative, (b) intra-granular and noncooperative, (c) inter-granular and cooperative, and (d) inter-granular and noncooperative. All processes are cooperative from an atomic point of view.

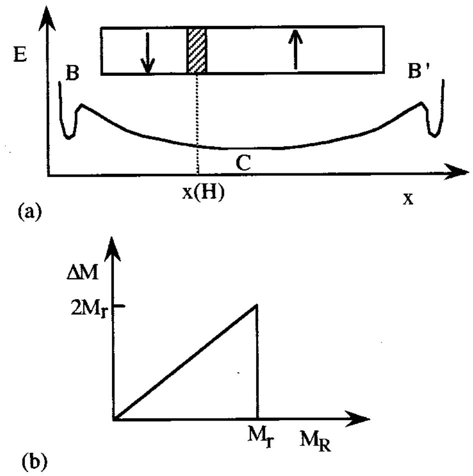

FIG. 2. ΔM interpretation of a two-domain magnet.

Usually, this ΔM curve is normalized by the ''ordinary'' remanence $M_r = M_R(\infty)$. Another way of plotting the remanence curves is to subtract the fields at which M_R and the ''transformed'' dc remanence $M_D^* = (M_r - M_D)/2$ reach the magnetization value M

$$\Delta H = H(M_R = M) - H(M_D^* = M). \tag{4}$$

As elaborated by Veitch,[17] plotting ΔH as a function of M can be used to make quantitative predictions, as compared to the qualitative Henkel and ΔM plots. When the interaction is assumed to be linear, $H_{\mathrm{MF}} = JM$, then the slope $\partial \Delta H / \partial M$ of the ΔH curve is equal to $-J$.

A popular interpretation is that positive ΔM curves (negative ΔH slopes) indicate positive or ''ferromagnetic'' interparticle interactions, whereas negative ΔM values indicate negative interactions. However, this interpretation is not able to account for cooperative effects. As emphasized by Wohlfarth, deviations from Eq. (1) are not necessarily due to interparticle interactions but may also be due to ''multidomain and incoherent rotation effects''.[14] Since coherent rotation is limited to particles smaller than about 20 nm,[5] most magnetization processes are incoherent.

Consider, for example, the motion of domain walls in a nucleation-controlled particle. For simplicity, we restrict ourselves to the one-dimensional model shown in Fig. 2(a). After thermal demagnetization, the magnet is in a global two-domain minimum (wall position C). A small field moves the wall to the left or right (virgin curve) until the domain wall is pinned (wall position B or B′). Figure 2(b) shows the ΔM plot for this magnetization process: ΔM is positive, but there is no point in interpreting the fairly complicated reversal mechanism Fig. 2(a) as a ferromagnetic interaction between neighboring grains.

III. TWO-PARTICLE INTERACTIONS

A key question is how cooperative processes affect the hysteresis loop. On a mean-field level, the magnet's internal interactions are mapped onto a correction of the external field Eq. (1). This means that positive and negative interactions yield increasing and decreasing slopes $\partial M / \partial H$ at H_c, respectively. This basic feature remains valid for weak interactions, but in the case of strong interactions there appear qualitatively new features. Furthermore, we will see that strong interactions in the sense of this work may be quite weak.

The simplest interaction model is two particles coupled by some magnetostatic or exchange coupling J. Going beyond earlier work,[19] we consider two nonidentical particles A and B characterized by the respective switching fields $H = -H_A$ and $H = -H_B$, where $H_B \geqslant H_A$. This makes it possible to explore the effect of the *switching-field distribution* $H_B - H_A$. The energy of the two-particle magnet is

$$\frac{E}{V_O} = -J\cos(\phi_A - \phi_B) - K_A\cos^2(\phi_A) - K_B\cos^2(\phi_B) - \mu_O M_S H(\cos\phi_A + \cos\phi_B), \tag{5}$$

where K_A and $K_B \geqslant K_A$ are the anisotropies of the respective particles, V_O is the volume of the particles, $H = H_Z$ is the external field, and J is an effective coupling constant incorporating both magnetostatic interactions and exchange. Figure 3 shows the real-space meaning of Eq. (5); the only function of the oblate shape of the particles is to reduce the number of degrees of freedom to two (ϕ_A and ϕ_B). After saturation in a large positive field, both moments are aligned along $\mathbf{e}_Z$ ($\phi_A = \phi_B = 0$), but on reducing (reversing) the field magnetization reversal occurs at the switching field H_S.

The nucleation behavior of the two-particle system is obtained by normal-mode stability analysis of Eq. (5), very similar to the determination of the nucleation field in nanoparticles and bulk magnets.[5,20] There are two limits of interest: broad switching-field distributions ($K_B \gg K_A$) and narrow switching-field distributions ($K_B \approx K_A$). In the case of a broad switching field distribution, $K_B - K_A \gg J$, the magnetic reversal is noncooperative, and after some straightforward calculation we find that the particles A and B switch at different reversed fields $(2K_A + J)/\mu_O M_S$ and $(2K_B - J)/\mu_O M_S$, respectively. Figure 3(b) illustrates the beginning of the switching of the first particle. By contrast, for $K_B = K_A$ the magnetization reversal is cooperative. When J is

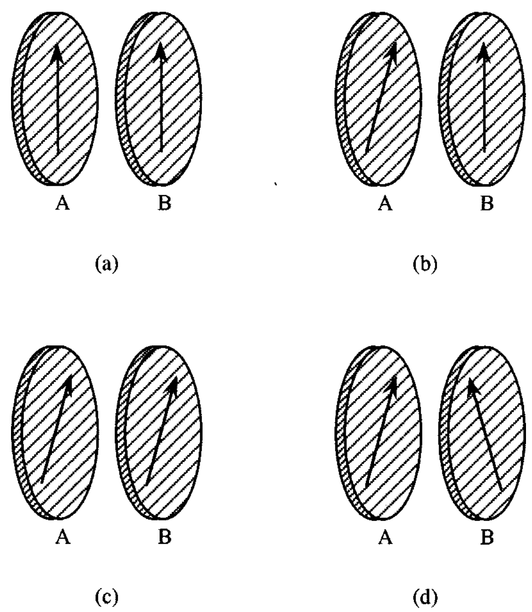

FIG. 3. A two-particle cooperative model.

positive or ferromagnetic, then the reversal is coherent [Fig. 3(c)] and the common switching field H_S is equal to $2K_A/\mu_O M_S$, whereas for $J<0$, that is for ''antiferromagnetic'' interaction [Fig. 3(d)] the common switching field H_S is $2(K_A+J)/\mu_O M_S$.

It is important to note that the cooperative $J>0$ switching field, $2K_A/\mu_O M_S$, is smaller than the corresponding Preisach prediction $(2K_A+J)/\mu_O M_S$. In other words, the Preisach model tends to overestimate the coercivity when microscopically well-defined parameters are used. This is a typical mean-field effect, similar to the overestimation of the Curie temperature by the statistical mean-field approximation. Physically, the overestimation of the coercivity is due to the neglect of the cooperative mode Fig. 3(c) by the Preisach model.

Comparing the two individual switching fields yields the estimate that cooperativity becomes important when $2K_A+J=2K_B-J$, that is, for $J\geqslant K_B-K_A$. For a given interaction strength, cooperative effects are therefore most pronounced for narrow switching-field distributions. Alternatively, the larger the micromagnetic susceptibility $\chi=\partial M/\partial H$, the more important are collective phenomena.[21] The corresponding dimensionless expansion parameter, which must be very small to ensure the applicability of the Preisach model, can be written as $\chi\Delta H/M_S$, where $\Delta H\sim J$ is the field equivalent of the interaction strength.

IV. DISCUSSION AND CONCLUSIONS

The problem of cooperative nanomagnetism is closely related to the problem of micromagnetic localization.[20,22] In the sense of this study, the term cooperative is equivalent to the delocalization of nucleation modes. A good example of an intra-granular cooperative phenomenon is the curling mode in homogeneous ellipsoids of revolution,[4,5,20] which are extended throughout the particle (delocalized from the point of view of a single particle). However, when the grain is larger than the Bloch-wall width, then morphological inhomogeneities at the surface or in the grain may cause localization. Essentially, magnetization reversal starts in a small part of the particle, the remainder of the particle giving rise to some effective interaction field, and then proceeds by domain-wall motion.[5]

In conclusion, we have investigated the role of cooperative magnetization reversal in ferromagnetic structures. The Preisach model and its extensions as well as related experimental techniques such as the Henkel, ΔM, and ΔH plots fail to account for both intra- and inter-granular cooperative phenomena. In particular, the Preisach model overestimates the coercivity, because it maps cooperative processes onto successive noncooperative magnetization jumps. As a rule, cooperative effects are strongest for narrow switching-field distribution.

ACKNOWLEDGMENTS

The authors have benefited from numerous discussions with J. P. Liu, C.-P. Luo, R. D. Kirby, M. Zheng, and H. Zeng. This research is supported by DOE, NSF, CMRA, and DOD.

[1] F. Preisach, Z. Phys. **94**, 277 (1935).
[2] O. Henkel, Phys. Status Solidi **2**, 78 (1962).
[3] O. Henkel, Phys. Status Solidi **7**, 919 (1964).
[4] A. Aharoni, Rev. Mod. Phys. **34**, 227 (1962).
[5] R. Skomski and J. M. D. Coey, *Permanent Magnetism* (Institute of Physics, Bristol, 1999).
[6] D. J. Sellmyer, M. Yu, R. A. Thomas, Y. Liu, and R. D. Kirby, Phys. Low-Dimens. Semicond. Struct. **1-2**, 155 (1998).
[7] E. Callen, Y.-J. Liu, and J. R. Cullen, Phys. Rev. B **16**, 263 (1977).
[8] K.-H. Fischer and A. J. Hertz, *Spin Glasses* (University Press, Cambridge, 1991).
[9] J. P. Liu, R. Skomski, Y. Liu, and D. J. Sellmyer, J. Appl. Phys. **87**, 6740 (1999).
[10] T. Song and R. M. Roshko, IEEE Trans. Magn. **36**, 223 (2000).
[11] V. Christoph and K. Elk, J. Magn. Magn. Mater. **74**, 143 (1988).
[12] M. LoBue, D. R. Cornejo, V. Basso, F. P. Missell, and G. Bertotti, IEEE Trans. Magn. **35**, 2194 (1999).
[13] C. Michelakis, D. Samaras, and G. Litsardakis, J. Magn. Magn. Mater. **207**, 188 (1999).
[14] E. P. Wohlfarth, J. Appl. Phys. **29**, 595 (1958).
[15] G. Lauhoff and T. Suzuki, J. Appl. Phys. **87**, 5702 (2000).
[16] K. O'Grady and H. Laidler, J. Magn. Magn. Mater. **200**, 616 (1999).
[17] R. J. Veitch, IEEE Trans. Magn. **26**, 1876 (1990).
[18] J. W. Harrell, D. Richards, and M. R. Parker, J. Appl. Phys. **73**, 6722 (1993).
[19] W. Chen, S.-F. Zhang, and H. N. Bertram, J. Appl. Phys. **71**, 5579 (1992).
[20] R. Skomski, J. P. Liu, and D. J. Sellmyer, Phys. Rev. B **60**, 7359 (2000).
[21] R. Skomski, J. P. Liu, J. M. Meldrim, and D. J. Sellmyer, in *Magnetic Anisotropy and Coercivity in Rare-Earth Transition Metal Alloys*, edited by L. Schultz and K.-H. Müller (Werkstoffinformationsgesellschaft, Frankfurt, 1998), p. 277.
[22] R. Skomski, J. Appl. Phys. **83**, 6503 (1998).