Influence of dipolar collective effects on coercivity and demagnetizing factors in hard magnetic materials

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Magnetization reversal in systems made of high coercivity exchange-decoupled grains is modeled. Dipolar collective effects, negligible for a sample in the form of a foil with out-of-plane magnetization, become more and more significant as the sample shape evolves toward three-dimensional geometry, and reach their maximum in case of a foil with in-plane magnetization. These collective effects are manifested by coercivity variations, which allow differences in coercivity for samples of the same material but having different shapes to be understood. Taking collective effects into account requires the introduction of a correction term to the demagnetizing factor. The numerical results obtained are experimentally confirmed for NdFeB permanent magnets.

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A variety of modern magnetic materials are constituted of an assembly of exchange decoupled single-domain grains. This includes perpendicular recording media and high-performance sintered magnets, such as NdFeB and $SmCo_5$. In such systems, whether magnetization reversal under field occurs by coherent rotation[1] or by nucleation/propagation,[2–4] it can be described as discrete, i.e., the moment of any grain switches from $+m$ to $-m$ ($m = M_s V_g$, where M_s is the spontaneous magnetization and V_g is the grain volume). This applies because the moment variation δm characterizing the reversal process, until the coercive barrier is reached (stage 1 of reversal), can be neglected with respect to the magnetization variation occurring after the coercive barrier has been overcome, which leads to complete reversal of the considered moment (stage 2 of reversal). In other words, when scanning the applied field from positive to negative values, magnetic configurations such that the moment of any considered grain differs significantly from $+m$ or $-m$ are never local energy minimum states of the system. There is a large amount of experimental evidence that magnetization reversal in such systems, as well as in many other heterogeneous magnetic materials occurs by such discrete switching rather than by continuous rotation/domain-wall motion.[5–14] Magnetization reversal of the whole material occurs by successive irreversible discrete jumps corresponding to individual grain moments reversal and the material magnetization M may take intermediate values from $+M_s$ to $-M_s$.

In a recent publication, we have shown that usual demagnetizing field corrections do not apply to the above category of hard magnetic materials.[15] The demagnetizing field in such heterogeneous magnetic materials may be conveniently separated into three terms: (i) the usual term $H_{DM} = -NM$, where M is the sample average magnetization, (ii) the cavity term due to magnetic poles at the surface of the individual grains, H_{Dcav}, and (iii) the term representing the self-demagnetizing field within the grains, H_{Dself}. The self-demagnetizing field of a particle is proportional to M_s, not M, because all considered grains are in the $+m$ non reversed state just before reversal. Note that an additional self-demagnetizing field-energy term exists, resulting from the magnetization variation occurring at stage 1 of reversal. This term does not depend on M and it essentially characterizes the reversal process itself. In the rest of the manuscript, it is implicitly included into the so-called intrinsic coercive field, H_c, of which the main source is the magnetocrystalline anisotropy. The demagnetizing field slope thus derived is compared to the "usual" demagnetizing field slope, which assumes homogeneous magnetization, in Fig. 1. Neglecting collective effects in magnetization reversal, one shows that the M-dependent contribution to the demagnetizing field amounts to $-(N - N_g)M$ instead of the usual value $-NM$.[15] In Ref. 15 we defined $N' = N - N_g$ as the Hard Demag factor. The contribution of the demagnetizing field to the slope of the magnetization variation is equal to $1/N'$ instead of the usual $1/N$ value. In the case of coherent rotation, reversal begins at $H_{app} = -H_c + (N - N_g)M_s$ (here $H_c > 0$ by definition). The nucleation volume, from which reversal begins, is equal to the grain volume and the contribution to coercivity due to the self-demagnetizing field disappears. In the case of nucleation/propagation, the nucleation volume may be con-

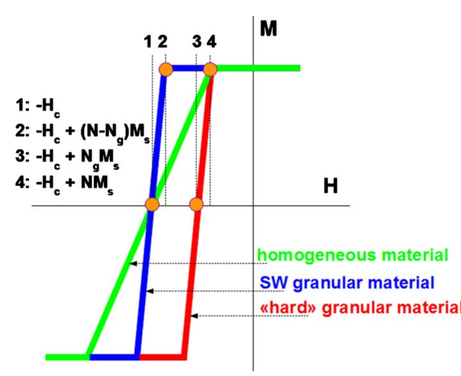

FIG. 1. (Color online) Schematic demagnetization curves in systems characterized by the same intrinsic coercive field H_c: (i) material with homogeneous magnetization; (ii) SW granular material with discrete switching; and (iii) "hard" particle assembly with discrete switching by nucleation/propagation.

sidered as negligible with respect to the grain volume. The self-demagnetizing field $N_g M_S$, acting on the nucleus, is equivalent to an external applied field and reversal begins at the same point as for homogeneous material: $H_{app}=-H_c+NM_s$.

In the above discussion, possible collective effects in magnetization reversal were neglected. Accordingly, in numerical modeling of an out-of-plane (oop) magnetized foil, described in Ref. 15 collective effects did not appear. Such effects were minimized by the fact that a certain coercive field distribution, on the order of the experimental H_c distribution, was considered. In the present Brief Report we demonstrate that, even when a certain H_c distribution exists, collective effects appear for other sample shapes [three-dimensional (3D) samples or foils with in-plane (ip) magnetization]. These affect the slope of the magnetization variation determined in the absence of collective effects. In the discussion, we refer specifically to so-called "hard" particle assemblies in which reversal occurs by nucleation/propagation. Apart from the coercive field shift discussed above, the results obtained apply equivalently to Stoner-Wohlfarth (SW) particles.

A perfectly textured assembly of hard particles was assumed. The particles were modeled as cubes, arranged on a cubic lattice with both the anisotropy axes of the grains and the applied external magnetic field H_{app} oriented along the z axis and the particle moments being either $+m$ or $-m$. At every field step the demagnetizing field H_D at the center of each cube was calculated, using well-known analytical formulas.[16] The conditions for magnetization reversal in a given particle was $H_{app}+H_{D_z}<-h_c$, where h_c is the intrinsic grain's coercive field and H_{D_z} is the projection of the total demagnetizing field on the z axis. The absolute value of the sample's coercive field, taken at $M=0$, is denoted as H_c. In the calculation, $\mu_0 M_s=1.2$ T and a normal distribution of h_c values was assumed with a mean value $\langle \mu_0 H_c \rangle=2$ T. The exact choice of these values, which are close to those characterizing NdFeB magnets, is of no importance for the present discussion. However, it is important that the width of the h_c distribution is on the order of the coercive field value, in order to minimize the collective effects related to the finite geometry of the system.

A quasi-two-dimensional (2D) system was considered first, made of 40×40 cubes, arranged in either XY (oop magnetization) or in XZ (ip magnetization) planes. The associated magnetization reversal curve, obtained by neglecting all demagnetizing field effects, is shown in Fig. 3 (curve 1). Taking demagnetizing fields into account, different distributions of switching field are now obtained depending whether texture is oop [Fig. 2(b) or ip Fig. 2(c)]. The corresponding magnetization curves are shown in Fig. 3, curve 2 (oop) and Fig. 3, curve 3 (ip). The usual demagnetizing field correction ($N=0.035$) does not apply for the ip magnetization curve (not shown in Fig. 3). However, the Hard-Demag correction $N'=N-N_g$, which works for the oop case,[15] does not work perfectly either (compare curves 1 and 4 in Fig. 3), and the calculated coercive field for in-plane texture is larger than the coercive field for the out-of-plane case, by a value $\mu_0 \delta H_c=0.11$ T. Note that coercivity differences between samples of different forms made of the same material, have

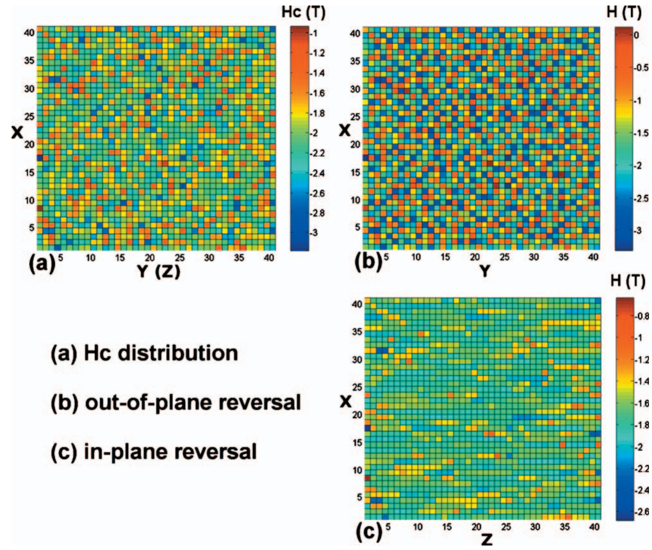

FIG. 2. (Color) Distribution of switching fields in a quasi-2D system of 40×40 exchange-decoupled hard magnetic cubes: (a) no dipolar interactions (coercivity distribution); (b) out-of-plane magnetization switching; and (c) in-plane magnetization switching.

been observed experimentally before, but no interpretation was proposed for this phenomenon.[17]

The imperfect applicability of the Hard-Demag corrections for the in-plane texture case may be qualitatively understood by considering Fig. 2(c). Numerical simulation reveals that elongated stripes are formed along the magnetization direction, representing particles in a given region having almost identical switching field value (in spite of the significant H_c distribution). The stripes indicate the occurrence of collective effects in magnetization reversal, leading to long-distance magnetic heterogeneities which invalidate the simple expression used to evaluate the cavity field.

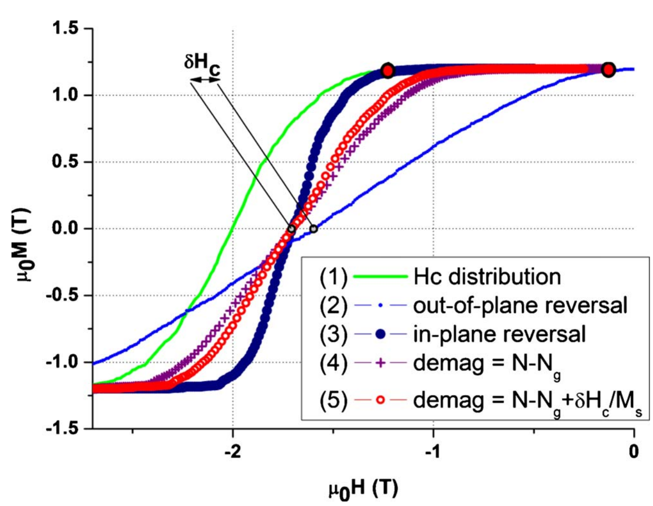

FIG. 3. (Color online) Magnetization curves for a quasi-2D system of 40×40 hard magnetic cubes with ip and oop magnetization. (1) Intrinsic coercivity distribution; (2) magnetization reversal in the oop case; (3) magnetization reversal in the ip case; (4) Hard-Demag correction N' applied to curve (3): does not reproduce the H_c distribution; and (5) Hard-Demag correction N'' applied to curve (3): reproduces the H_c distribution.

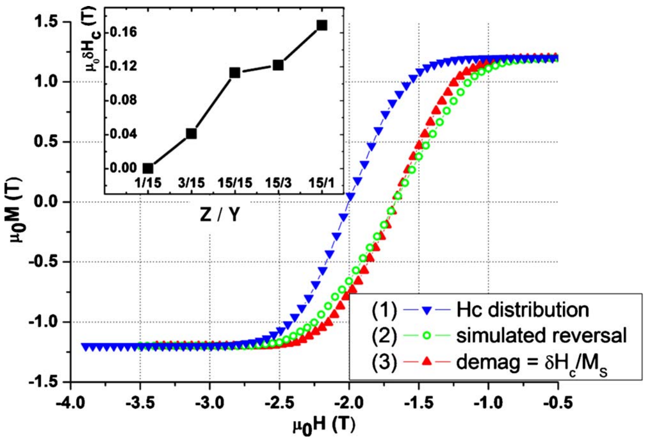

FIG. 4. (Color online) Magnetization curves for a 3D system of $15\times15\times15$ hard magnetic cubes. (1) Intrinsic coercivity distribution; (2) simulated magnetization reversal; and (3) Hard-Demag correction N'' applied to curve (2): reproduces the H_c distribution. Inset: increase in the coercive field offset δH_c, in a $(15\times Y\times Z)$ systems, manifesting the collective effects, as the system evolves from the out-of-plane configuration $(15\times15\times1:Z/Y=1/15)$ to the in-plane $(15\times1\times15:Z/Y=15/1)$ one.

The same type of calculations was performed for different 3D stacking of cubes $(15\times15\times15)$, $(15\times15\times3)$, and $(15\times3\times15)$. Note that for a sample having the shape of a cube $(15\times15\times15)$ the Hard-Demag correction gives $N'=N-N_g=0$. The calculated curve 2 in Fig. 4 should directly follow curve 1 representing the coercive field distribution, which is not the case. Figure 4 inset, shows how the coercive field difference δH_c defined for each given system with respect to the out-of-plane coercive field, increases progressively as one goes from the out-of-plane to the in-plane configuration. At the same time, examination of the switching field maps for each system (not shown, of the same type as shown in Fig. 2 for the 2D extreme cases) reveals that collective effects become more and more significant.

The occurrence of collective effects, revealed by numerical simulation for non out-of-plane magnetization configurations, may be understood qualitatively by considering the contributions involved in the demagnetizing field energy. This includes a first term representing the usual sample-shape-dependent demagnetizing field, which, in the case of homogeneous magnetization (evaluated here at the scale of the environment of each grain and not to be confused with the uniform magnetization characterizing soft magnetic materials), is equal to $\frac{1}{2}\mu_0 NM^2$ and a second term representing the cavity field and which for the case of homogeneous magnetization amounts to $-\frac{1}{2}\mu_0 NM^2$. In the case of flat oop samples, the positive sample-shape-dependent term is dominant (N is large). This term being proportional to M^2, is minimized, for a given M value, when the magnetization is uniform (and the above "uniform magnetization" relation applies). Under such conditions collective effects are unfavored. In contrast, for foils with ip magnetization, N is small and the cavity field term dominates. The associated energy term is negative, it is minimized for heterogeneous configurations of the grain moments. Collective effects are favored.

The existence of the coercive field shift δH_c may be considered as an experimental evidence for the occurrence of collective effects. It can be quantified by introducing an additional demagnetizing field correction term amounting to $\delta H_c/M_s$. A general Hard-Demag correction for a sample of any shape is thus derived: $N''=N'+\delta H_c/M_s=N-N_g+\delta H_c/M_s$, where δH_c is the coercive field difference between, on one side, a hypothetic system in which there are no collective effects in magnetization reversal, and, on the other side, the considered sample. From the above discussion, the former system may be assumed to be represented by a sample in the form of a foil with oop magnetization. Thus $\delta H_c=H_c(\text{oop})-H_c(\text{sample})$. Figure 3 (curve 5) and Fig. 4 (curve 3) demonstrate the applicability of the general Hard-Demag correction.

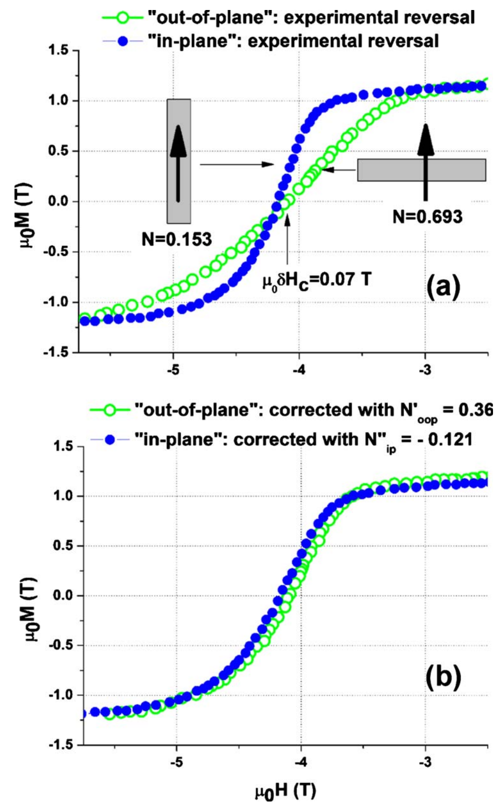

FIG. 5. (Color online) (a) Experimental magnetization curves of NdFeB platelets with "oop" (having $N=0.693$) and "ip" ($N=0.153$) geometries. (b) Applying the general Hard-Demag corrections N'' ($N'\equiv N''$ for the oop case) yields two parallel curves, thus revealing the applicability of the model.

In order to test experimentally the above described effects, two platelets were cut from a commercial NdFeB magnet. The platelet demagnetizing factor along the preferred magnetization directions were 0.693 and 0.153, respectively.[18] The hysteresis cycles measured at $T=175$ K in fields up to 7 T, are shown in Fig. 5(a). The coercive field of the ip platelet is higher than that of the oop one, by $\mu_0\delta H_c=0.07$ T. When applying usual demagnetizing field corrections to the ip and the oop magnetization curves, the corrected curves obtained do not follow each other, which confirm the inapplicability of these corrections. The Hard-Demag corrections, as described above, were then applied. The N'_{oop} demagnetizing factor was applied for correcting the oop magnetization curves $N'_{oop}(\equiv N''_{oop})=0.693-0.333=0.36$, see Fig. 5(b), open circles. Similarly, the N''_{ip} demagnetizing factor was derived: $N''_{ip}=0.153-0.333+0.059=-0.121$, where the last term $\delta H_c/M_s$ is equal to 0.07/1.2 [Fig. 5(b), filled circles]. The obtained curves are parallel to each other thus illustrating the applicability of the present model.

In this Brief Report we demonstrated that collective effects may affect magnetization reversal in an assembly of exchange-decoupled hard magnetic grains. A coercivity shift characterizes a given sample compared to a 2D out-of-plane textured film. This shift is a manifestation of collective effects, it requires the application of an additional demagnetizing field correction. A general expression for the demagnetizing field correction was obtained. The experimental results, obtained from samples of different geometry, cut from a commercial NdFeB magnet, confirmed the model's conclusions.

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