Stray field fluctuations in soft-hard nanostructured materials: Its influence on the shift of minor hysteresis loops

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Magnetostatic interactions play a relevant role in the macroscopic magnetic behavior of multiphase and nanostructured samples. In particular, interactions between nanometric hard particles and a soft magnetic matrix in which the particles are embedded lead to a shift of the minor hysteresis loops, as observed in coupled layers by the effect of the exchange biasing. The experimentally observed dependence of the magnetostatic interactions with the particle concentration is explained here as a consequence of the random arrangement of the particles and the characteristics of their associated magnetostatic-field fluctuations. The predictions of the proposed model show a remarkable agreement with the experimental data.

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The hysteretic behavior of the multiphase nanostructured systems depends, on the one side, on the intrinsic properties, morphology, and defect distribution of each one of the involved phases, and, on the other side, it depends crucially, on the magnitude and characteristics of the exchange and magnetostatic interphase coupling.[1] In some magnetoelectronic devices, such as the spin valves, the global system behavior is ruled by the coupling between ferromagnetic (soft) and antiferromagnetic (hard) thin layers that is known as interfacial exchange biasing[2] or exchange-induced anisotropy.[3] Due to the technological relevance of the spin valves, this effect has been the subject of many recent reports.[4–6] This coupling leads to a shift of the minor hysteresis loops corresponding to the reversal, only, of the soft phase magnetization. We should, nevertheless, remember the fact that magnetostatic interactions coexist in any nanostructured systems with the exchange ones and that it is generally rather difficult to separate their contributions to any given hysteretic feature.[7]

Magnetostatic interactions play an important role in patterned structures for future magnetic recording media and magnetic random access memories (MRAM) influencing the switching field distribution and the achievable device density.[8] In three dimensional systems with statistically distributed components the exact treatment and prediction of the effects of magnetostatic interactions is a problem still to be solved.

In this paper we will report on the particle concentration dependence of the hysteresis shift, *hs*, observed in nanostructured two-phase systems and originated by magnetostatic interactions. Experiments previously carried out[9] in a composite system formed by Fe particles (average optical microscope size 45 μm, saturation magnetization $\mu_0 M_1 = 2.1\,\mathrm{T}$) and Ba hexaferrite particles (average transmission electron microscope particle diameter 30 nm, saturation magnetization $\mu_0 M_2 = 0.6\,\mathrm{T}$) exhibit the compositional dependence of the shift of the minor loop, as shown in Fig. 1. The shift corresponds to minor loops obtained by reversing the Fe magnetization after previously saturating the sample and is not observed if the applied demagnetizing field is large enough so as to fully reverse the Ba hexaferrite particles' magnetization. We will show that such compositional dependence of *hs* is a typical signature of the magnetostatic interactions since it reflects the evolution of the local fluctuations of the stray field with the volume percentage of the hard phase.

Let us consider initially that n magnetically hard single particles, with volume v_2 and previously magnetized along the positive $+$Oz direction having a spontaneous magnetization M_2 are embedded in a soft magnetic matrix with volume $v_1 = v - nv_2$ and spontaneous magnetization $M_1 > M_2$. We shall consider that under the effect of moderate applied fields, M_1 can be oriented along either the positive z axis (M_1^+ configuration) or along the negative z axis (M_1^- configuration), while the hard particles' magnetization remains in both cases rigidly saturated along the positive z axis (which restricts our discussion to minor loops). The difference in magnetostatic energy between both configuration ΔF can be expressed by using an effective field *hs* as

$$\Delta F = 2\mu_0 v_1 M_1 hs, \tag{1}$$

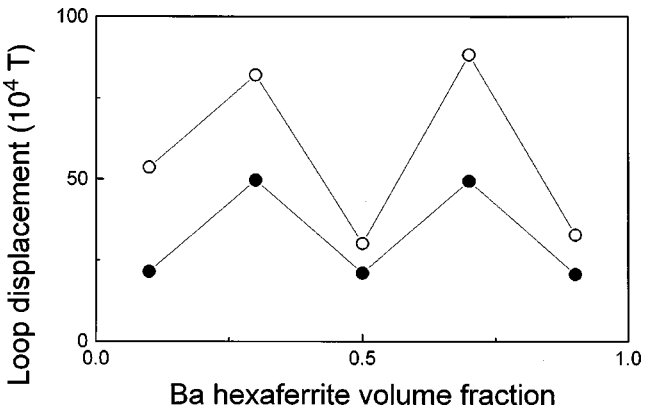

FIG. 1. Experimental hysteresis shift as a function of composition for two series of samples milled during 15 (empty dots) and 120 min (filled dots), respectively (Ref. 9).

where *hs* should coincide with the shift of the minor loop corresponding to M_1 reversal provided that the reversal takes place coherently.

Note that the M_1^+ configuration can be envisioned as a system formed by the matrix with volume (v_1+nv_2) and magnetization M_1, and n particles of volume v_2 with magnetization M_2-M_1 embedded in the matrix. Analogously, the M_1^- configuration can be thought of as the combination of the matrix with magnetization $-M_1$ and n particles embedded in it with magnetization M_2+M_1. If we denote by N and N' the demagnetizing factor of the matrix and the average demagnetizing factor of the particles along the z axis, respectively, it is easy to find the difference in magnetostatic energy $\Delta F=F(M_1^+)-F(M_1^-)$ as

$$\Delta F=2\mu_0 M_1 M_2 v_2\left\{(N-N')n+\sum_i \sum_{j>i} D_{i,j}\right\}, \qquad (2)$$

where $D_{i,j}$ are the linear coefficients describing the field at the ith particle as a function of the pole density located at the interface between the jth particle and the matrix. This last term, describing interactions between poles at the matrix-particles interfaces, becomes negligible in the limit of high dilution, i.e., for very small n. Under these conditions the hysteresis shift, according to Eqs. (1) and (2), is given by

$$hs=M_2 n v_2\{(N-N')/(v-nv_2)\}. \qquad (3)$$

This hysteresis shift (3) calculated from Eq. (1), corresponds to the difference between the average field acting on M_1 at the M_1^+ and M_1^- configurations, respectively, and as stated above it would correspond to the experimental *hs* if the reversal of M_1 took place coherently. However, if the magnetization reversal is induced locally by nucleation, the nonuniform character of the magnetostatic field acting on M_1 will be of maximum relevance. Suppose that the average over v_1 of the field $H_z(r)$, acting on M_1 due to the poles at the matrix surface as well as those at the interfaces, is zero in both M_1^+ and M_1^- configurations. In other words, the spatial average of the field intensity acting along the positive z axis, $\langle H_{z,+}\rangle$, is equal to the average of the negative ones, $\langle H_{z,-}\rangle$. This is, for instance, the case of a spherical particle embedded in a spherical matrix for which $N=N'$ and according to Eq. (3) $hs=0$. However, for the M_1^+ configuration the average $\langle H_{z,-}\rangle^+$ is smaller than the average $\langle H_{z,+}\rangle^-$ corresponding to the M_1^- configuration. Note that for the M_1^+ configuration the field acting on M_1 at the position r is the sum of a uniform (or slow-fluctuating) field $-NM_1$ and another spatially fluctuating component, due to the poles induced at the particle-matrix interfaces, $-\mu_0 v_2 \Sigma_j D_{ir}$ (M_2-M_1). In the M_1^- configuration, the uniform field is NM_1, whereas the fluctuating component changes to $\mu_0 v_2 \Sigma_j D_{ir}$ (M_2+M_1). The coefficients D_{ir} fluctuate spatially in sign with a correlation length of the order of the particle size. In a randomly distributed and oriented set of ellipsoidal particles embedded in a spherical matrix the average $\langle D_{ir}\rangle$ over v_1 is zero. Related to the magnetization reversal process, the fluctuating stray field should be averaged over volumes with dimensions of the order of the exchange-correlation length of the soft phase. Within this volume the effect of the magnetostatic-field fluctuations on the magnetization is smoothed by exchange interactions. In particular, for the case of nanometric particles having a size comparable to the domain wall width of the soft phase, the suitable averaging region is that of the volume of the particles. If within this volume the average field is opposite to M_1 and overcomes in strength a threshold field (i.e., the nucleation field H_n), the reversal of M_1 should locally begin. Therefore, the difference in the applied field required to reach locally H_n for the M_1^+ and M_1^- configurations is given by the difference in the fluctuation of the field corresponding to these two configurations, that is, related to the average of the positive D_{ir},$\langle D_{ir+}\rangle$, according to the relation $2\mu_0 v_2\langle D_{ir+}\rangle M_2$. This difference corresponds exactly to *hs* when the reversal mechanism is a single nucleation event and, in general, it should be related to *hs*. The local fluctuations depend on the spatial symmetry of the particle distribution and implicitly on the volume fraction x of the particles $[x=nv_2/(v_1+nv_2)=nv_2/v]$.

In order to estimate some particular stray field fluctuations and for the sake of simplicity we consider spherical particles, having diameter d, with a first-neighbor coordination number equal to 6. In this case D_{ir} becomes $(1/4\pi)(3\cos^2\theta_{ir}-1)/d_{ir}$,[3] where θ_{ir} and d_{ir} are the polar angle and the distance of the center of the ith particle with respect to the point r. Every averaging volume (having a dimension equal to that of the particles) has six first neighbors, corresponding to the $\langle 100\rangle$ directions and all of them at $d_{ir}=d$. Two of them, labeled z and $-z$, respectively, are centered along the Oz axis (polar neighbors) and four, labeled xy cells, occupy equatorial positions. For the first coordination shell, and considering only the field at the center of the averaging volume, it is verified that

$$\sum_j (3\cos^2\theta_{1j}-1)=(2n_z-n_{xy})=(3n_z-n), \qquad (4)$$

where n_z and n_{xy} are the number of particles present at the polar and equatorial positions, respectively, and n is the total number of neighboring particles $0\leqslant n\leqslant 6$ [the standard deviation of n corresponding to a random distribution of first neighborhoods is $(1/6)^{1/2}$]. For any fixed composition x, the number of possible configurations at each first coordination shell is given by $6!/6x!(6-6x)!$ (where we have replaced n by $6x$). The average of the positive field values at the center of the averaging volume over the first neighborhood can be obtained from Eq. (4) with the condition $n_z>n/3$, as

$$\left\langle \sum_{\{nz,\ n/3,\ 2\}} (3\cos^2\theta_i-1)\right\rangle=(3n_z-n)P(n_z) \qquad (5)$$

where $P(n_z)$ is the probability corresponding to the presence of n_z first neighbors at the polar positions, given by $[2!/n_z!(2-n_z)!][4!/(n-n_z)!(4-n+n_z)]$.

It is easy to show that if we assume that the field at the center of the averaging cube is given by $(v/d_2^3)\langle\Sigma(3\cos^2\theta_{2i}-1)\rangle$, the particles at the four second-neighboring cells located at the $\langle 110\rangle$ sites contribute to the

absolute value of the field at the central cell approximately proportional to $-1/2\sqrt{2}$ (per cell), whereas those present at the $\langle 101 \rangle$ positions (eight cells) contribute to that field in an approximate amount of $1/4\sqrt{2}$ (per cell). The contribution of this second neighborhood to positive values of $\Sigma(3\cos^2\theta_{2i}-1)$ can be calculated analogously to that of the first neighborhood. Figure 2 shows $(\langle\Sigma(3\cos^2\theta_{1i}-1)\rangle/d_1^3+\langle\Sigma(3\cos^2\theta_{2i}-1)\rangle/d_2^3)$ as a function of x.

It is worth noting that the compositional dependence of the average positive field, shown in Fig. 2, is remarkably similar to that exhibited by the experimentally observed *hs* in Fig. 1. Also, the experimental magnitude of the loop shift (close, for the compositional maxima, to 10^{-2} T) is very similar to the field created by a Ba hexaferrite particle having the dimension of those present in our samples at a distance from its center coinciding with the particle diameter (1.3×10^{-2} T).

In summary, we have found that the shift of the minor loops observed in soft-hard nanostructured systems, due to interphase magnetostatic interactions, is related to the spatial fluctuation of the local stray field. This dependence can be understood when the reversal of the M_1 phase takes place through a single nucleation event. By using reasonable approximations that allow us the estimation of these fluctuations, a noticeable qualitative good agreement with the experimental compositional dependence of the shift has been obtained.

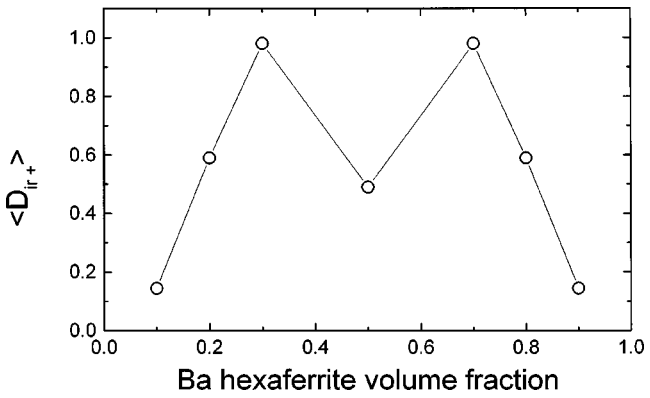

FIG. 2. Fluctuation of the stray field acting on the M_1 phase as a function of composition.

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