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Rate-dependence of the switching field distribution in nanoscale granular magnetic materials

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The external field sweep-rate dependence of the intrinsic switching field distribution in perpendicular recording media is investigated. We derive a scaling relationship for switching field distributions at different sweep-rates, which we then validate by means of large-scale kinetic Monte Carlo simulations based on interacting Stoner–Wohlfarth particles. After demonstrating the possible occurrence of large differences between switching field distributions at slow time scales of conventional magnetometry and very fast processes relevant in magnetic recording, we propose a technique for extrapolating between these very different sweep-rates. © 2010 American Institute of Physics. [doi:10.1063/1.3477956]

Magnetic information storage relies on recording and preserving information in nanoscale magnetic grains that are generally coupled by magnetostatic and exchange interactions. The presence of these interactions considerably complicates magnetization processes as well as the extraction of local materials properties from macroscopic measurements. An important example of this problem is the identification of intrinsic switching field distributions $D(H_S)$ from macroscopic hysteresis loops in highly anisotropic granular magnetic materials, which are being used for magnetic recording. $D(H_S)$ is defined as the distribution of switching thresholds of individual particles in the interaction-free case and it is a crucial materials property for magnetic recording as it directly determines the local precision of bit transitions.

 $D(H_S)$ is related to the anisotropy field distribution $D(H_K)$ of grains, from which it mostly differs due to thermal activation, which introduces additional relaxational time scales and leads to a dependence of the particle switching field on the external magnetic field sweep rate $R.^3$ It also leads to a dependence from the volume distribution D(V) of the grains because the activation barrier for the thermal relaxation phenomena is volume dependent. A general relationship between $D(H_S)$ measured at different sweep rates and between $D(H_S)$ and $D(H_K)$ is not known and will be a subject of the present study.

A recent experimental study on perpendicular recording materials (PMRs) (Refs. 4 and 5) demonstrated a 50% difference between the relative widths of $D(H_S)$ measured at time scales of a typical laboratory magnetometry (R $\sim 10^2~{\rm Oe/s})$ and time scales more closely related to magnetic recording (R $\sim 10^8~{\rm Oe/s})$. In these experiments the relative width of $D(H_S)$ was quantified by the ratio of the standard deviation to the coercive field, σ_S/H_C , as is conventional. Similar experimental observations in terms of the squareness S^* of the hysteresis loops have also been reported very recently. 6

The problem of identifying the rate dependence of $D(H_S)$ partly arises from the fact that extracting $D(H_S)$ from hysteresis loops in interacting particle assemblies is typically

a very difficult problem and techniques developed for this purpose^{7–15} rely on various degrees of approximations. Importantly, they essentially ignore thermal relaxation effects and it is unclear how accurately the methods relate to the fast subnanosecond time scale relevant for write processes in hard disk drives. In the present work, we investigate this issue by using a kinetic Monte Carlo model of PMR materials. We find that the recently developed version of the $\Delta H(M, \Delta M)$ -methodology ^{13,14} allows accurate extraction of $\sigma_{\rm S}$ of D(H_S) from the computed hysteresis loops even in the presence of exchange and dipolar interactions. Having demonstrated the accurate determination of $\sigma_{\rm S}$, we then study theoretically the rate-dependence of σ_S/H_C showing that slow time scale measurements can overestimate the relative width σ_K/H_K of $D(H_K)$ by as much as 300%. We also derive a scaling relationship, which relates σ_S/H_C and σ_K/H_K for different R.

Following Ref. 16, the well established kinetic Monte Carlo modeling framework assumes a planar assembly of N Stoner-Wohlfarth (SW) particles i of volume V distributed on a hexagonal lattice of spacing a. The magnetization of every particle normalized by the saturation value M_S is described by a dimensionless unit vector free to rotate. The uniaxial anisotropy axes of particles are oriented perpendicular to the film plane collinear with the external field of strength H_a. It is assumed that the anisotropy constants K_i and correspondingly the anisotropy fields $H_{K,i}=2K_i/M_S$ (CGS units) vary randomly from particle to particle according to the lognormal probability distribution having mean $\langle H_{K,i} \rangle$, median H_K , and standard deviation $\sigma_K = (\langle H_{K,i}^2 \rangle)$ $-\langle H_K \rangle^2$)^{1/2}. For reasons of transparency of the present work we will ignore distributions of anisotropy axis orientations $D(\alpha)$ and volumes D(V) of the particles.

The nearest neighbor exchange interactions are quantified by an exchange field constant H_{EX} , which is the total exchange field on a particle in the saturation state. The long-range magnetostatic coupling between the grains is approximated by dipolar interactions. Thermal switching involves the evaluation of particle-dependent energy barriers ΔE separating positive and negative magnetization states of the particles using the effective field concept. We assume thermal transitions to be governed by the Arrhenius–Néel relaxation law w^{\pm} = f_0 exp($-\Delta E^{\pm}/k_B T$), where w^+ and w^- are the transitions to be given by the Arrhenius–Néel relaxation law w^- are the transitions to be given by the Arrhenius–Néel relaxation law w^- are the transitions to be given by the Arrhenius–Néel relaxation law w^- are the transitions are the transitions of the particle w^+ and w^- are the transitions of the particle w^- and w^- are the transitions of the particle w^- and w^- are the transitions of the particle w^- and w^- are the transitions of the particle w^- and w^- are the transitions of the particle w^- and w^- are the transitions of the particle w^- and w^- are the transitions of the particle w^- and w^- are the transitions of the particle w^- and w^- are the transitions of the particle w^- and w^- are the transitions of the particle w^- and w^- are the transitions of the particle w^- and w^- are the transitions of the particle w^- and w^- are the particle w^- and w^- and w^- are the particle w^- and w^- are the particle

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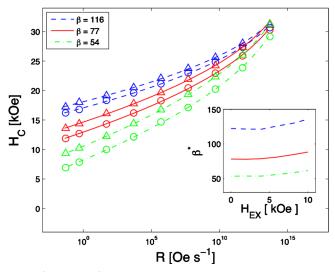


FIG. 1. (Color online) Dependence of the coercive field H_C on the external field sweep rate R for different thermal stability factors for the non-interacting case (\bigcirc) and the case with dipolar interactions and exchange $H_{EX}=10~\mathrm{kOe}(\Delta)$. Lines represent the least-square fits of Eq. (3). Inset: Dependence of β^* obtained from fitting Eq. (3) for $0 \le H_{EX} \le 10~\mathrm{kOe}$.

sition rates for escaping from the positive and negative states, respectively, f_0 is the attempt frequency factor, k_B is the Boltzmann constant, and T is the temperature. The particle-dependent characteristic relaxation time depends on w^{\pm} and equals to $\tau = 1/(w^+ + w^-)$. The actual probabilities for switching between the particle states are given by the following:

$$P(t_{m}) = P^{0} \exp(-t_{m}/\tau) + w^{+} \tau [1 - \exp(-t_{m}/\tau)], \qquad (1)$$

which results as a solution of the corresponding single-particle master equation. ¹⁶ In Eq. (1), P^0 is the probability of finding a particle in the negative state initially at H_a and $P(t_m)$ is the probability of preserving that state after the time step t_m . For an external field changing in constant steps ΔH_a the field sweep-rate is defined as $R = \Delta H_a/t_m$.

In simulations, we compute hysteresis loops, including recoil curves which are required below for the identification of D(H_S) using the $\Delta H(M,\Delta M)$ -method. 13,14 We assume a system of N=160×160 particles with lattice spacing $a\!=\!8$ nm and grain heights L=5.5, 8, and 12 nm. Setting $M_S\!=\!400$ emu/cc, T=300 K, and $\langle H_{K,i}\rangle\!=\!40$ kOe (H_K=38 kOe), this gives thermal stability ratios $\beta\!=\!KV/k_BT$ =54, 77, and 116. We set $\sigma_K/H_K\!=\!0.2$. This choice of parameter sets is guided by the boundary of what is relevant for typical recording media design or might be so in the future, where thermal relaxation effects will be especially important. However, our results are general within the range of validity of the assumed Arrhenius–Néel relaxation picture, i.e., they are not reliant on any specific parameter set.

We first analyze the sweep-rate dependence of the dynamic coercive field H_C obtained from the computed hysteresis loops. Figure 1 shows $H_C(R)$ for different H_{EX} and β . Exchange interactions cause a squaring up of the hysteresis loops and an increase in H_C , which becomes more pronounced for reduced β .

The dependence of H_C on the measurement time scale is typically quantified by Sharrock's equation in case of a relaxational experiment 18 or by similar equations relevant for swept-field experiments, 19 which is the case here. We briefly

summarize the derivation, as it will be used later for our generalization toward switching field distributions.

Considering the physical picture leading here to Eq. (1), the switching threshold $H_{S,i}$ of a single SW particle with anisotropy $H_{K,i}$ and the thermal stability ratio β_i corresponds to its coercivity; $H_{S,i}\!=\!H_{C,i}.$ Then the $\sigma_S\!=\!(\langle H_{C,i}^2\rangle\!-\!\langle H_{C,i}\rangle^2)^{1/2}.$ Due to the thermal relaxation $H_{S,i}\!<\!H_{K,i}$ but they coincide in the nonrelaxational case for $t_m\!\to\!0$ (or equivalently $R\!\to\!\infty$). The dependence of $H_{C,i}$ on R can be evaluated by integrating Eq. (1) over the hysteresis process generated by a magnetic field decreasing at the rate R and starting from saturation. This yields the following: 19

$$H_{C,i} = H_{K,i} \{ 1 - [\beta_i^{-1} \ln(t_i f_0)]^{1/2} \}$$
 (2)

with $t_i = (2\beta_i R)^{-1} H_{K,i} (1 - H_{C,i} / H_{K,i})^{-1}$, being the effective time. Integrating this equation over the anisotropy distribution $D(H_K)$ in the noninteracting particle case gives a macroscopic version of Eq. (2) applicable in the noninteracting case: $H_C = H_K \{1 - [\beta^{-1} \ln(tf_0)]^{1/2}\}$, with $t = (2\beta R)^{-1} H_K (1 - H_C / H_K)^{-1}$ and H_C and H_K representing, respectively, the median of $H_{C,i}$ and $H_{K,i}$. This equation is normally fitted also to interacting particle assemblies in which case H_K and β are interaction-dependent fit parameters and will be distinguished using the standard notation as H_0 and β^* , respectively. Thus the macroscopic analogy of Eq. (2) is as follows:

$$H_C = H_0 \{ 1 - [\beta^{*-1} \ln(tf_0)]^{1/2} \}$$
 (3)

and $t=(2\beta^*R)^{-1}H_0(1-H_C/H_0)^{-1}$, defines the effective time as previously.²¹

According to Fig. 1, Eq. (3) fits the H_C versus R data very well for all interactions despite the fact that it was derived using the noninteracting particle picture. The inset shows that the fit parameter β^* increases with H_{EX} , which relates to the increase in the effective activation volume due to the stabilizing exchange interaction. Nonetheless we note that the resulting fit values correspond to the preset values of β rather well. In addition, H_0 is found to vary only slightly with H_{EX} .

We now address the issue of R-dependence of the standard deviation σ_S of $D(H_S)$. Figure 2(a) shows σ_S obtained from hysteresis loops using the $\Delta H(M,\Delta M)$ -method. 13,14 Although other available methods can be used for extracting σ_S as well, we found here that the $\Delta H(M,\Delta M)$ -method accurately represents the intrinsic σ_S for all R and in the entire range of exchange interactions considered. For instance, this can be seen from the figure where practically all data points for $H_{EX}{=}\,10\,$ kOe fall on top of the noninteracting case line corresponding to a given β . We note that in the noninteracting case σ_S was confirmed to agree with the differentiated major loop which directly corresponds to $D(H_S)$.

In the noninteracting case the dependence σ_S versus R can also be calculated analytically. Squaring both sides of Eq. (2), expressing the mean and the second moment over the distribution $D(H_K)$, and using the same assumptions as above 20 gives the following:

$$\sigma_{S} = \sigma_{K} \{ 1 - S[\beta^{-1} \ln(tf_{0})]^{1/2} \}$$
 (4a)

with
$$S = \left(\langle H_{K,i} \rangle \frac{\langle H_{K,i} \rangle - \langle H_{K,i}^{-1/2} \rangle^2}{\langle H_{K,i}^2 \rangle - \langle H_{K,i} \rangle^2} \right)^{1/2}$$
. (4b)

The factor S is dependent only on the properties of the underlying anisotropy distribution $D(H_K)$ and is independent of

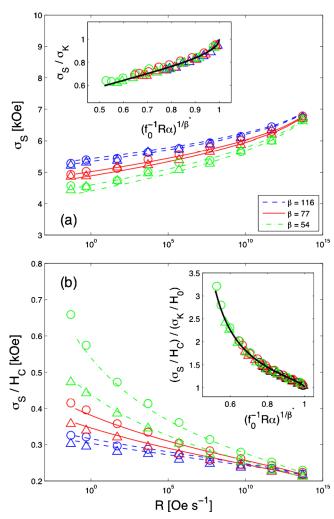


FIG. 2. (Color online) (a) Dependence of the switching field distribution width σ_S on the external field sweep rate R for different thermal stability factors for the non-interacting case (\bigcirc) and the case with dipolar interactions and exchange $H_{EX}=10\,$ kOe(Δ). Lines represent plots of Eq. (4) using H_0 and β^* obtained from fitting Eq. (3) to $H_C(R)$. Inset: A collapse of all the fitted curves onto a universal function $y=1-2^{-1}[\ln(1/x)]^{1/2}$ after rescaling the axes. $\alpha=2\beta^*(1-H_C/H_0)/H_0$. (b) The same as (a) but for the ratio σ_S/H_C given by Eq. (5). Inset: the universal function has now the following form: $y=\{1-2^{-1}[\ln(1/x)]^{1/2}\}/\{1-[\ln(1/x)]^{1/2}\}$.

R or any quantities related to thermal relaxation. S can be calculated exactly for a lognormal distribution which gives $S \sim 1/2$ for our numerical examples (S=1/2 for a symmetric distribution).

Although Eq. (4) has been derived assuming the noninteracting particle picture, Fig. 2(a) shows that it also reproduces well the data for the interacting case if the interaction-dependent parameters β^* and H₀ obtained from fitting Eq. (3) are used. Similarly, using Eqs. (3) and (4) a general relationship between σ_S/H_C and σ_K/H_K can be expressed in the following form:

$$\frac{\sigma_{\rm K}}{\rm H_0} = \frac{\sigma_{\rm S}}{\rm H_C} \times \frac{1 - [\beta^{*-1} \ln(\rm tf_0)]^{1/2}}{1 - 2^{-1} [\beta^{*-1} \ln(\rm tf_0)]^{1/2}},\tag{5}$$

with $t=(2\beta^*R)^{-1}H_0(1-H_C/H_0)^{-1}$ as before and after using S=1/2 for the lognormal distribution. Equation (5) allows determining the intrinsic σ_K/H_0 based on the β^* and H_0 obtained from fitting Eq. (3) and σ_S obtained by the $\Delta H(M,\Delta M)$ -method. Figure 2(b) shows excellent agreement between the computed data and Eq. (5). Insets in Figs. 2(a)

and 2(b) show that after rescaling the axis all data collapse well onto a single line, further validating the approach in the presence of interactions.

In summary, numerical calculations based on the kinetic Monte Carlo SW-model show that the laboratory time scale measured σ_S/H_C can substantially overestimate the intrinsic σ_K/H_K ; by as much as 300% within the R range considered. Techniques to obtain σ_S , such as the $\Delta H(M,\Delta M)$ -method used in this work, have only been demonstrated at slow rates R. We propose a powerful and precise experimental procedure for extrapolating to the σ_S/H_C at recording frequencies, which is the truly relevant characteristic related to magnetic recording. The procedure is based on a combination of rate-dependent magnetometry, specifically H_C versus R measurements and a single R measurement of hysteresis loop data needed for applying the $\Delta H(M,\Delta M)$ -method.

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¹⁷These distributions will generally result in an additional broadening of the magnetization transition and a decorrelation between the ratios $\sigma_{\rm S}/{\rm H_C}$ and $\sigma_{\rm K}/{\rm H_K}$. A quantification, in how far D(α) and D(V) modify the result and the overall scaling relations found here, will be a subject of future studies. ¹⁸M. P. Sharrock, J. Appl. Phys. **76**, 6413 (1994).

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²⁰Derivation assumes (1) $\langle [\ln(t_if_0)]^{1/2} \rangle \sim [\ln(\langle t_i\rangle f_0)]^{1/2} = [\ln(tf_0)]^{1/2}$, with the notation $\langle t_i \rangle = t$, and that t_i is only weakly correlated with $H_{K,i}$. This is reasonable because β_i is proportional to $H_{K,i}$ and because of the anticorrelation nature the ratio $H_{C,i}/H_{K,i}$, which results in a sharply peaked probability distribution for t_i centered around the mean t. (2) That the coercive field characterizes switching of the most typical particles in the medium, which is the case for magnetic recording materials, to allow neglecting higher order covariance terms. We also note, that equivalent equation holds for mean values $\langle H_{C,i} \rangle$, $\langle \beta_i \rangle$, and $\langle H_{K,i} \rangle$ as long as the differences $\langle H_{C,i} \rangle - H_C$ and $\langle H_{K,i} \rangle - H_K$ are not too large and follow similar trends as a function of R.

²¹Note that t does not appear in the case of the Sharrock equation, which applies to different experimental procedures based on measuring the remanent coercivity.^{3,18} Note also, that it is easier to fit the inverted Eq. (3), i.e., after expressing R as a function of H_C.