Simultaneous determination of intergranular interactions and intrinsic switching field distributions in magnetic materials

O. Hovorka, Y. Liu, K. A. Dahmen, and A. Berger

Citation: Appl. Phys. Lett. **95**, 192504 (2009); doi: 10.1063/1.3263732
View online: http://dx.doi.org/10.1063/1.3263732
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v95/i19
Published by the American Institute of Physics.

Simultaneous determination of intergranular interactions and intrinsic switching field distributions in magnetic materials

O. Hovorka,[1,a)] Y. Liu,[2] K. A. Dahmen,[2] and A. Berger[1]
[1]*CIC nanoGUNE Consolider, E-20018 Donostia-San Sebastián, Spain*
[2]*Department of Physics, University of Illinois at Urbana Champaign, Urbana, Illinois 61801, USA*

(Received 31 July 2009; accepted 23 October 2009; published online 12 November 2009)

We develop a generally applicable method for the accurate measurement of intrinsic switching field distributions and the determination of exchange and dipolar interactions in granular magnetic materials. The method is based on the simultaneous analysis of hysteresis loop and recoil curve data. Its validity and practical implementation are demonstrated by means of computational modeling using a reference function identification scheme. We find the methodology to be numerically accurate in a wide parameter range, far exceeding the previously utilized mean-field interaction regime used in other methodologies. © *2009 American Institute of Physics*.
[doi:10.1063/1.3263732]

Nanoscale granular magnetic materials are at the heart of modern nanotechnology and have been utilized in applications ranging from biological sensors to information storage technology, such as the hard disk drives.[1] Further progress in the development of these technologies relies on devising new materials and new processing procedures to enable appropriate tailoring of physical properties, as well as on the availability of efficient and accurate characterization tools applicable at the nanoscale.

The present paper is concerned with the challenge of characterizing granular materials, such as the ones used for state-of-the-art perpendicular magnetic recording media.[2] An important issue with respect to these materials is the extraction of the intrinsic switching field distributions (SFDs) from macroscopic hysteresis loop measurements. A number of methods have been developed for this purpose in the past few years, which include the First Order Reversal Curve (FORC) method,[3,4] the Veerdonk method,[5] and the $\Delta H(M, \Delta M)$-method based on the mean field approximation,[6–8] as well as the complex transverse ac susceptibility technique.[9–11] In general, the precise knowledge of the SFD is essential because it is intimately related to the grains' volume and anisotropy dispersions[12] and, moreover, it is an important quality-defining characteristic of magnetic recording materials.[13] However, all the above methods have substantial reliability deficiencies and, furthermore, do not allow for an accurate and separate measurement of exchange and magnetostatic interactions in-between individual grains. Such complete information is most desirable, as it would allow for an independent optimization of all recording media properties. For instance, it was predicted that the intergranular exchange coupling has to be optimized to achieve the best possible noise characteristics of recording materials.[14] Similarly, magnetostatic couplings must be tuned to prevent media stability deterioration. Clearly, the ability to quantify intergranular interactions efficiently and accurately in addition to improved reliability of SFD measurements is an outstanding problem in the field of magnetic materials characterization. Its solution is the main goal of the present paper.

In this letter, we will first introduce our approach in general terms. Subsequently, we will use the interacting hysteron model as one specific implementation to demonstrate the feasibility and practicability of our approach. This will also enable us to show that for a unique identification of model parameters it is necessary to base the data analysis on the measurement set $H_{\oplus}(M, \Delta M)$, which is obtained by combining $\Delta H(M, \Delta M)$-data, calculated as field differences between the recoil curves and the major loop (Fig. 1), with the inverted hysteresis loop H(M), i.e.,

$$H_{\oplus}(M, \Delta M) = \Delta H(M, \Delta M) \oplus H(M). \quad (1)$$

The symbol $\oplus$ is used to emphasize that $H_{\oplus}(M, \Delta M)$ is obtained by merely joining the $\Delta H(M, \Delta M)$ and H(M) data sets and not as their sum.

The method is based on comparing the $H_{\oplus}(M, \Delta M)$ set with similar "reference" functions obtained from modeling,

$$R_{\oplus}(M, \Delta M; \{p^i\}) = \Delta R(M, \Delta M; \{p^i\}) \oplus R(M; \{p^i\}), \quad (2)$$

where $\Delta R(M, \Delta M; \{p^i\})$ and $R(M; \{p^i\})$ denote, respectively, field differences and major loop parts of the computed data. $\{p^i\}$ is a set of model specific parameters, such as exchange and dipolar couplings, for instance, as well as the intrinsic SFD. The comparison between $H_{\oplus}(M, \Delta M)$ and $R_{\oplus}(M, \Delta M; \{p^i\})$ is carried out using least-squares fitting to identify the specific model parameter set $\{p^i\}$ corresponding to that reference function which fits the input $H_{\oplus}(M, \Delta M)$ data best. As discussed in detail below, the identification pro-

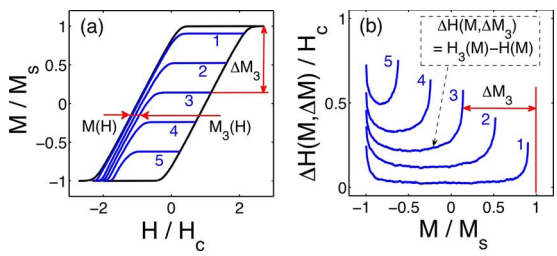

FIG. 1. (Color online) (a) Hysteresis loop M(H) and a set of five recoil curves obtained by reversing the applied field at distances ΔM_l, $l=1,\ldots,5$ from saturation. (b) $\Delta H(M, \Delta M)$-data calculated as field differences between the recoil curves 1–5 and the decreasing major hysteresis loop branch.

[a)]Author to whom correspondence should be addressed. Tel.: +34 943 574 000. FAX: +34 943 574 001. Electronic mail: o.hovorka@nanogune.eu.

cedure we use here is based on a grid-search method,[15] which allows employing microscopic models with no analytical solution available, such as coupled spin models, micromagnetics, or models including thermal relaxation, for instance. Also, with this approach, one does not have to rely on simplifications such as the mean-field approximation.

The model of choice is first used to compute $R_{\oplus,k}(M,\ \Delta M;\{p^i\}_k)$ for different sets of parameters $\{p^i\}_k$, $k=1,2,\ldots$. Then, the optimum $\{p^i\}_k$ is found for which the sum of squares

$$\chi_k^2 = \sum_j \{H_\oplus^j - R_{\oplus,k}[M^j,\ \Delta M;(p^i)_k]\}^2 \tag{3}$$

is minimum with respect to all k. In Eq. (3), the summation index j runs through every individual data point within the measurement $H_\oplus(M,\ \Delta M)$. The reference functions are precomputed for all physically relevant parameter sets $\{p^i\}_k$ and tabulated in a way that allows for direct comparison to arbitrarily sized and spaced measurement data sets $H_\oplus(M,\ \Delta M)$.[16]

Here, we demonstrate one specific implementation of the method using the interacting hysteron model,[8] which is computationally efficient enough to allow for the calculation of thousands of reference functions while it is still a sufficiently realistic model for describing the magnetization reversal in perpendicular recording materials. The model Hamiltonian is

$$H = -J_{EX}\sum_{\langle ij\rangle} S_iS_j + J_{DP}\sum_{ij,i\neq j} S_iS_jd_{ij}^{-3} - \sum_i [H + f_i(S_i)]S_i, \tag{4}$$

where S_i denotes the hysteron (i.e., hysteretic spin variable), and d_{ij} is the distance between two hysterons i and j. The first and second sums represent the exchange and dipolar interactions quantified by constants J_{EX} and J_{DP}, respectively, and the function f_i defines the SFD, which we assume here to be a Gaussian of variance σ. Within the framework of this model, hysteresis behavior depends only on the ratios J_{EX}/σ, J_{DP}/σ, and H/σ.[8]

To demonstrate that $H_\oplus$, i.e., joint hysteresis loop and recoil curve data set, is necessary for the analysis, we start by considering the case where Eqs. (1) and (2) contain only the $\Delta H(M,\ \Delta M)$ part and the major loop is not included. Reference functions $\Delta R_k(M,\ \Delta M;J_{EX}^k/\sigma,J_{DP}^k/\sigma)$ are generated from model calculations using 40 000 hysterons on a hexagonal lattice for a mesh of 41×21 interaction pairs varying between $0<J_{EX}^k/\sigma<1$ and $0<J_{DP}^k/\sigma<1$ and fixed σ. This interaction range covers the range for realistic materials used in magnetic recording.[17] Due to the fact that the model behavior depends only on the ratio H/σ rather than on H and σ separately, σ can be reintroduced for every k as a simple linear fit-coefficient σ_{fit}. By doing so, one avoids introducing the σ-axis as an additional dimension in the J_{EX}^k and J_{DP}^k parameter space, which would be required otherwise.

To test the restricted method, we pick one specific reference function as input data and ask if it can be distinguished from all others. As quantitative measure, we use the 95%-confidence level, which is a standard for statistical evaluation of the fitting quality.[15] Clearly, we expect at least one (and ideally only one) match, namely, the one for which J_{EX} and J_{DP} values are identical to those for the reference function taken as an input (denoted as J_{EX}^{in} and J_{DP}^{in}), and also $\sigma_{fit}=\sigma$ as a result of the fit.

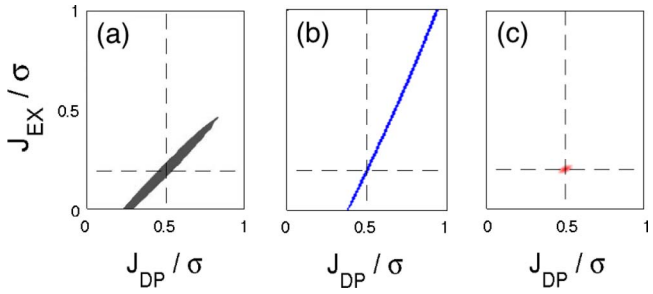

FIG. 2. (Color online) (a) The elliptical region corresponds to reference functions $\Delta R_k(M,\ \Delta M)$ indistinguishable in the 95%-confidence-level statistics sense from input $\Delta H(M,\ \Delta M)$ data corresponding to $J_{EX}^{in}=0.2$ and $J_{DP}^{in}=0.5$. (b) The stripe region is obtained when only major loop data $R_k(M)$ and $H(M)$ are used as the reference and input functions. (c) The dot corresponds to the unique reference function $R_{\oplus,u}(M,\ \Delta M)$ agreeing with the input data $H_\oplus(M,\ \Delta M)$. The crossing point of the dashed lines shows location of the reference function chosen as the input data set. The slope of the pattern in (a) is $(J_{EX}^{in}-J_{EX}^k)/(J_{DP}^{in}-J_{DP}^k)\sim1$ and in (b) is $(J_{EX}^{in}-J_{EX}^k)/(J_{DP}^{in}-J_{DP}^k)\sim2$.

Results from the analysis are shown in Fig. 2. The gray region in Fig. 2(a) corresponds to those ΔR_k, which are indistinguishable from $\Delta H(M,\ \Delta M)$ corresponding to $J_{EX}^{in}=0.2$ and $J_{DP}^{in}=0.5$ within the assumed 95%-confidence level. Thus, we see that $\Delta H(M,\ \Delta M)$ is statistically indistinguishable from ΔR_k for numerous k and we have no way of knowing which ΔR_k is the true one. However, despite the fact that we cannot uniquely identify the exact reference function, we found that σ_{fit}-values obtained from comparing $\Delta H(M,\ \Delta M)$ with all ΔR_k located inside the region in Fig. 2(a) always correspond to the correct variance σ of the SFD. This demonstrates uniqueness of the identification with respect to the SFD parameters, a fact that is further confirmed in Fig. 3(a). Here, we show results for $\Delta H(M,\ \Delta M)$ with

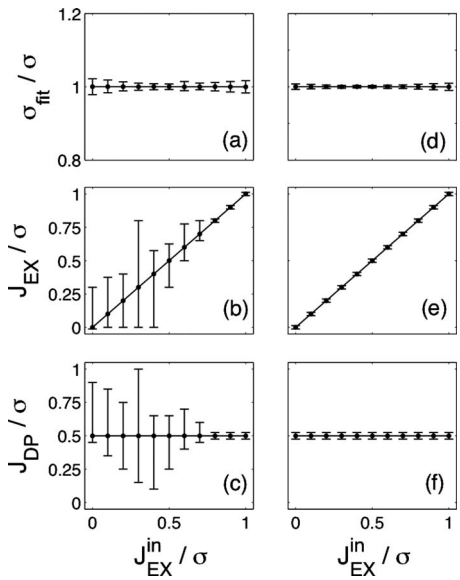

FIG. 3. [(a)–(c)] σ_{fit}, J_{DP}, and J_{EX} parameters estimated from the $\Delta H(M,\ \Delta M)$ data obtained from the hysteron model. Large error bars in (b) and (c) mean that interactions cannot be determined within any reasonable confidence. Small error bars in (a) show uniqueness with respect to σ_{fit}. [(d)–(f)] The same analysis for the combined $H_\oplus(M,\ \Delta M)$-method demonstrating unique identification of all model parameters.

$J_{DP}^{in}=0.5$ and varying J_{EX}^{in} and find $\sigma_{fit}/\sigma \sim 1$ and only small error bars in all cases. Our numerical studies show that identical conclusions apply for all choices of J_{DP}^{in} and J_{EX}^{in}, except for the strong exchange coupling limit, at which point magnetization reversal becomes fully correlated and recoil curves are experimentally not accessible anymore. When compared with the previously developed mean-field $\Delta H(M, \Delta M)$-technique for determining the SFD,[6–8] the present method, if based only on the $\Delta H(M, \Delta M)$ parts of Eqs. (1) and (2), already gives accurate results in a much broader materials parameter range [as shown in Fig. 3(a) and Ref. 17]. The significant improvement results from the fact that the present method is not relying on the mean field assumption.

On the other hand, the fact that reference functions for different interactions are not distinguishable yields large error bars in Figs. 3(b) and 3(c), and exact identification of interactions is not possible. To find a suitable pathway to solve this problem, we have repeated the above approach with Eqs. (1) and (2) restricted to the major hysteresis loop term only. The corresponding analysis produces again an extended region of reference functions R_k that exhibit a good match to the input H(M) data, as shown in Fig. 2(b). Therefore, also in this case, individual interactions cannot be identified. However, the patterns in Figs. 2(a) and 2(b) are tilted along different directions, and this difference turns out to be a characteristic and fundamental feature caused by the underlying physics of magnetization reversal. A detailed analysis of our simulation results shows that local correlations of magnetic reversal are determined predominantly by the net interaction field produced by its immediate neighbors, which is proportional to the difference $J_{EX}-J_{DP}$ (for a more detailed discussion of this correlation-compensation effect, see Ref. 8). Therefore, we expect a similar correlation behavior to occur for two systems if $J_{EX}^{in}-J_{DP}^{in} \sim J_{EX}^{k}-J_{DP}^{k}$, which is approximately the slope of the pattern seen for the $\Delta H(M, \Delta M)$-data in Fig. 2(a). Such correlations are dominating the behavior of $\Delta H(M, \Delta M)$-data sets because the mean-field interaction effects are already removed from them by definition.[6] On the other hand, the slopes of hysteresis loops H(M) for two systems are expected to be the same if their mean interaction fields are the same. In the present model, the total interaction field on a given moment due to dipolar coupling is about double of the exchange fields (exchange and dipolar fields are, respectively, 6σ and 11.56σ in saturation), so that two systems will have the same mean interaction field if $J_{EX}^{in}-2J_{DP}^{in} \sim J_{EX}^{k}-2J_{DP}^{k}$, which agrees well with the pattern in Fig. 2(b).

These observations suggest that using the set $H_{\oplus}(M, \Delta M)$ obtained by combining the $\Delta H(M, \Delta M)$ and H(M) data is a pathway to identifying a unique solution for the SFD as well as the intergranular interactions. Indeed, repeating the above analysis using full Eqs. (1) and (2) confirms uniqueness properties. As demonstrated in Fig. 2(c), the region of reference functions agreeing with the input set is narrow, given the precision of our statistical analysis and centered on the reference function chosen for the input data. The corresponding analysis for all J_{EX}^{in}, J_{DP}^{in} pairs (outside of the fully correlated regime) corroborated the general suitability of this method for an exact identification of all model parameters. Examples illustrating this ability are shown in Figs. 3(d)–3(f), where one observes strongly reduced error bars.

Identical conclusions apply when a more realistic lognormal SFD is used instead of the Gaussian SFD shown here. In this case, an additional fit parameter corresponding to the skewness of the lognormal distribution is required, which increases the dimension of the precomputed parameter space $\{p^i\}$. An analysis analogous to Fig. 3 showed the same level of precision for the identification of all model parameters including the lognormal SFD itself. Furthermore, it is important to mention that the present approach is not restricted to using the hysteron model or any specific class of models in general. Any suitable microscopic model can be utilized to generate the set of reference functions according to Eq. (2). Uniqueness and reliability properties for such models have to be verified independently. One can also ask the more general question of how the SFD and other parameters that are identified by means of long-time scale measurements relate to the nanosecond properties that are relevant for magnetic recording. The extent of thermally activated processes is very different in either case. In principle, the characterizing measurements could be performed on recording time scales, in which case the identification based on "nonthermal" modeling is expected to be accurate. Alternatively, one would have to attempt developing thermal activation models that are also capable of representing correlated magnetization reversal, which is a subject of active research.

Work at nanoGUNE acknowledges the EROTEK Program, Project No. IE06-172, and the Spanish Ministry of Science and Education under the Consolider-Ingenio 2010 Program, Project No. CSD2006-53. O.H. acknowledges the Marie Curie International Reintegration Grant, Project No. 224924.

[1] S. D. Bader, Rev. Mod. Phys. **78**, 1 (2006).
[2] S. N. Piramanayagam and K. Srinivasan, J. Magn. Magn. Mater. **321**, 485 (2009).
[3] C. R. Pike, A. P. Roberts, and K. L. Verosub, J. Appl. Phys. **85**, 6660 (1999).
[4] M. Winklhofer and G. T. Zimanyi, J. Appl. Phys. **99**, 08E710 (2006).
[5] R. J. M. van de Veerdonk, X. Wu, and D. Weller, IEEE Trans. Magn. **39**, 590 (2003).
[6] A. Berger, Y. H. Xu, B. Lengsfield, Y. Ikeda, and E. E. Fullerton, IEEE Trans. Magn. **41**, 3178 (2005).
[7] A. Berger, B. Lengsfield, and Y. Ikeda, J. Appl. Phys. **99**, 08E705 (2006).
[8] Y. Liu, K. A. Dahmen, and A. Berger, Appl. Phys. Lett. **92**, 222503 (2008).
[9] C. Papusoi, Jr., Phys. Lett. A **265**, 391 (2000).
[10] G. Ju, H. Zhou, R. Chantrell, B. Lu, and D. Weller, J. Appl. Phys. **99**, 083902 (2006).
[11] G. Ju, B. Lu, R. J. M. van de Veerdong, X. W. Wu, T. J. Klemmer, H. Zhou, R. Chantrell, A. Sunder, P. Asselin, Y. Kubota, and D. Weller, IEEE Trans. Magn. **43**, 627 (2007).
[12] G. Bertotti, *Hysteresis in Magnetism: For Physicists, Materials Scientists, and Engineers* (Academic, New York, 1998), p. 228.
[13] Y. Shimizu and H. N. Bertram, IEEE Trans. Magn. **39**, 1846 (2003).
[14] H. J. Richter and S. D. Harkness, MRS Bull. **31**, 384 (2006).
[15] P. R. Bevington and D. K. Robinson, *Data Reduction and Error Analysis* (McGraw-Hill, New York, 2003), p. 194.
[16] For the general evaluation of Eq. (3), one requires an interpolation scheme to match the individual numerical reference function points $R_{\oplus,k}$ to randomly spaced $H_{\oplus}$ data.
[17] This range covers perpendicular recording materials with saturation magnetization M_s in the range of 0–1500 emu/cm^3, anisotropy fields $H_k \sim 5$–40 kOe, SFD spread $\sigma_h \sim 0.05$–$0.5H_k$, and exchange field H_{ex} (assuming six-neighbor interactions) in the range of 0–$2.5H_k$ such that $H_{ex}/\sigma_h < 5$.