Interaction Fields Evaluation in Fine Particle Systems

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Abstract—A computer simulation model for estimation of the interaction fields in fine particle systems has been developed. The method uses the experimental $\delta M_a(H)$ plot, constructed from the Fourier description of the initial magnetization curve and the hysteresis loop, and the $\delta M(H)$ plot, obtained from the remanence curves. The dependencies of the interaction fields on the external magnetic field have been obtained from the experimental data for a sample of a Ba–ferrite fine powder.

Index Terms—Magnetic analysis, magnetic domains, magnetic interactions, magnetic recording noise, magnetization processes, remanence, single-domain particles.

I. INTRODUCTION

THERE HAS been a remarkable upsurge in the use of remanent magnetization measurements in recent years in the search of the interpretation of interaction effects in particulate and thin-film media. First, Henkel [1] plotted the dc demagnetization remanence curve $M_d(H)$, versus the isothermal remanent magnetization curve $M_r(H)$ which, in accordance with the fundamental Wohlfarth relation [2] in the case of no interactions, should give a linear plot with a slope of -2. Any deviation from the idealized straight line is attributed to interactions. A variation of the Henkel technique is the use of the first derivatives of the remanent magnetization curves, i.e., the switching field distribution, for investigation of the interaction effects [3], [4]. Kelly *et al.* [5] defined the interaction-based deviation, δM, as

$$\delta M(H) = M_d(H) - (1 - 2M_r(H)) \qquad (1)$$

which gives the sign and the relative strength of the interactions as well as their field dependencies. Here $M_r(H)$ and $M_d(H)$ are normalized to the saturation remanence value $M_r(\infty)$.

Several other attempts were made to construct plots that lead to the evaluation of the interaction fields. An alternative to the δM plot is the δH plot [6] which gives information about the interaction field strength as a function of the magnetization state.

In most phenomenological models [7]–[11], it is, in general, well accepted that during the initial "*i*" magnetization process and the demagnetization "*d*" process, there are different interaction fields acting (H_{int}^i and H_{int}^d, respectively) as a function of the magnetic state of the material. Thus, the effective fields

present in the sample, in each case, can be written as

$$H_i = H + H_{\text{int}}^i \qquad (2)$$

for $0 \leq H \leq +H_{\max}$ and

$$H_d = H + H_{\text{int}}^d \qquad (3)$$

for $-H_{\max} \leq H \leq +H_{\max}$.

However, neither of the procedures mentioned above can give a direct evaluation of H_{int}^i and H_{int}^d separately, essentially because there is only one relation, $\delta M(H)$, connecting them.

In all of the works cited above, one particular form for the interaction field was assumed

$$H_{\text{int}}(M_{i,d}) = \alpha M_{i,d} + f(M_{i,d}). \qquad (4)$$

The first term in (4) is a mean interaction field which depends on the global magnetization and the local exchange interactions. The second term is related to fluctuations of the mean field and has been assumed to be $f(M_d) = 0$ [7], [8], $f(M_d) = \beta(1 - M_d^2)$ [9], [10], or $f(M_d) = \beta(1 - M_d^2)M_d$ [11]. The parameters α and β were then determined by numerical simulations of δM curves using (4) and choosing parameters that would produce agreement with the measured δM curves.

Recently, a Fourier description of magnetization curves for a Stoner–Wohlfarth model [12] system has been reported [13]. The normalized magnetization functions $m^d(H)$ and $m^i(H)$ (the exponents d and i refer to the loop and to the initial curve, respectively) have been expanded in Fourier series

$$m^d(x) = \sum_{n=1}^{N} a_n^d \cos(nx) + \sum_{n=1}^{N} b_n^d \sin(nx) \qquad (5)$$

$$m^i(x) = \sum_{n=1}^{N} a_n^i \cos(nx). \qquad (6)$$

Here $x = (\pi/2)(1 - H/H_{\max})$ for H changing between $+H_{\max}$ and $-H_{\max}$, and $x = (\pi/2)(3 + H/H_{\max})$ for H changing between $-H_{\max}$ and $+H_{\max}$, and N is the number of harmonics. It has been found that for the model curves, the coefficients a_n^i coincide with a_n^d, and the difference

$$\delta M_a(H) = \sum_{n=1}^{N} a_n^d \cos(nH) - \sum_{n=1}^{N} a_n^i \cos(nH) \qquad (7)$$

is equal to zero for a noninteracting system. This coincidence has been explained using simple symmetry relations of the curves considered.

Experimentally, it has been shown [13], [14] that the $\delta M_a(H)$ plot is not zero in certain field intervals, and the shape of this plot is similar to the shape of the corresponding
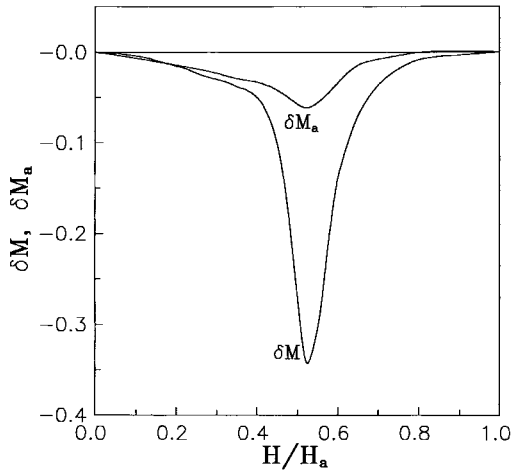
Fig. 1. Experimental δM and δM_a plots for a Ba–ferrite powder [14].

Fig. 2. Calculated 3-D plot of δM versus H_i/H_a and H_d/H_a.

$\delta M(H)$ plot. In Fig. 1, experimental $\delta M(H/H_a)$ and $\delta M_a(H/H_a)$ for a M-type Ba–ferrite powder [14] are represented (H_a is the anisotropy field for a Stoner–Wohlfarth system). A detailed description of the sample preparation and the microstructural characterization has already been reported [15]. A simple model [14] has been proposed demonstrating that the use of both $\delta M(H)$ and $\delta M_a(H)$ plots could give additional information about the mean internal fields.

Usually, the δM plots for barium ferrite particulate tapes are positive because the hexagonal platelet shape of the particles, combined with their perpendicular anisotropy, promotes the formation of stacks of particles, which exhibit positive interactions (see, for example, [16]). However, Patel *et al.* [17] showed the δM plots for powder samples are negative for all fields, which suggest that the interactions are always demagnetizing, forming a closed flux cluster configurations. This is the case of the sample, whose δM plot is shown in Fig. 1.

The aim of the present work was to develop a model allowing the estimation of the interaction fields (H_{int}^i and H_{int}^d) directly, by using both the experimental $\delta M_a(H)$ and $\delta M(H)$ plots.

II. MODEL

The phenomenological model assumes that a magnetic material consists of a collection of single domain uniaxial particles of random orientation. Each particle follows Stoner–Wohlfarth coherent rotation during reversal. It is assumed that the effective fields during initial magnetization and demagnetization processes, H_i and H_d, are vector sums of the applied field H and mean interaction fields, H_{int}^i and H_{int}^d, respectively, as defined by (2) and (3). It is assumed that there are no particles that switch back their magnetizations from their saturation remanence directions to the opposite ones, during the return of the applied field to zero (recoil). It is also accepted that the fields $H_{\mathrm{int}}^{i,d}$ do not change the remanence after recoil, so the remanence is achieved by applying and switching off the field H_i (or H_d) in the framework of the Stoner–Wohlfarth model.
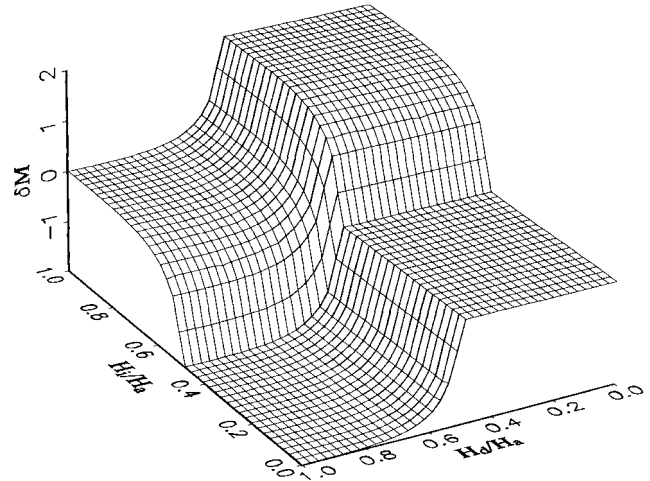
For a couple of H_i and H_d, the values of the initial magnetization, M^i, demagnetization, M^d, and remanent magnetizations, M_r and M_d, can be calculated, then

$$\delta M(H_i, H_d) = M_d(H_d) - (1 - 2M_r(H_i)) \qquad (8)$$

can be obtained, and Fig. 2 shows its three–dimensional (3-D) plot versus H_i/H_a and H_d/H_a. The cases of positive ($\delta M > 0$ for $H_i > H_d$) and negative ($\delta M < 0$ for $H_i < H_d$) interactions are well expressed in this plot. The plateau of zero δM in the plot corresponds to H_i and H_d less than $0.5H_a$, when no irreversible rotation of the particle's magnetization occurs.

By definition, $\delta M_a(H)$ (7) is the difference between the initial magnetization curve, $M^i(H)$, and another function, which is an "artificial" initial magnetization curve. In the case of no interactions, this curve coincides with $M^i(H)$.

We introduce a relation, δM_c, as the difference between the real and the "artificial" initial magnetization curves, $M^i(H_i)$ and $M^i(H_d)$, respectively, calculated for different internal field dependencies

$$\delta M_c(H_i, H_d) = M^i(H_i) - M^i(H_d). \qquad (9)$$

The reason for the introduction of this new plot will become clearer in the following paragraphs.

In Fig. 3, the 3-D plot of δM_c as a function of H_i/H_a and H_d/H_a is represented. It can be seen that when H_i and H_d are less than $0.5H_a$ (and are not equal), δM_c is not zero, as it is for δM in Fig. 2. The reason is that δM_c, in contrast to δM, is determined by both reversible and irreversible rotations.

The projections of a set of equi-δM curves (curves with the same δM and different H_i/H_a and H_d/H_a), taken from Fig. 2, on the (H_i, H_d) base are shown in Fig. 4 (solid curves). As can be seen, by looking at one specific equi-δM projection, the δM data alone are not enough to estimate the interaction fields, because a δM value may be obtained for different couples of H_i and H_d.

In the same figure, several equi-δM_c curves (broken lines) are also represented. As can be seen, their shape is not the same as that of the solid curves, and there are crossings between the two types of curves. From a given couple of δM and δM_c for a
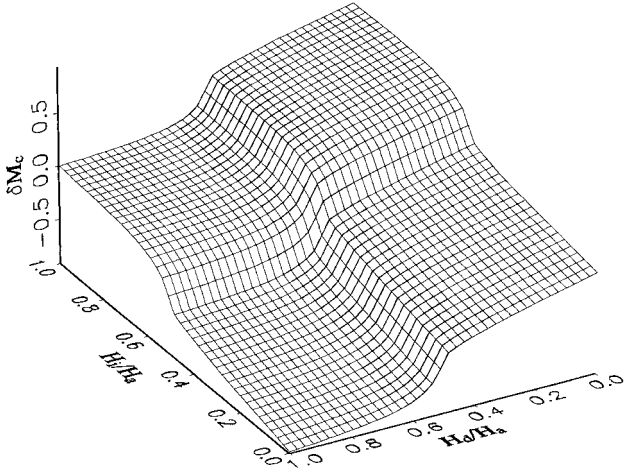
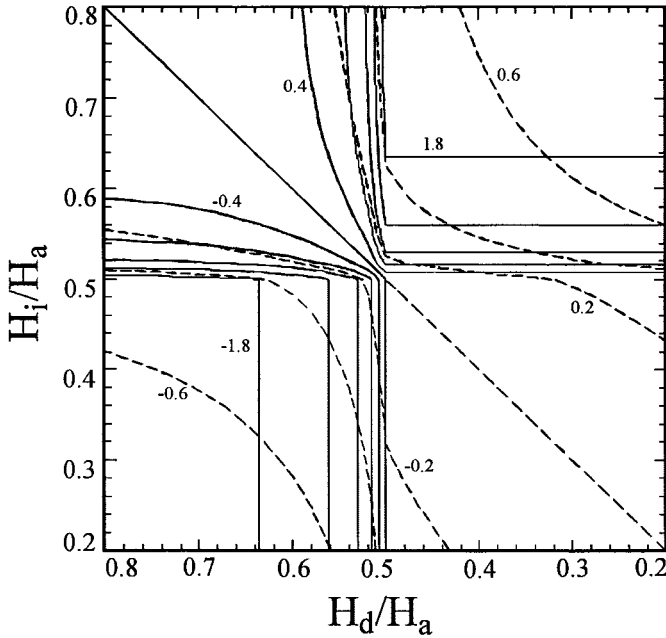Fig. 3. 3-D plot of δM_c versus H_i/H_a and H_d/H_a, calculated by using (9).

Fig. 4. Projections of several equi-δM curves (the solid ones) and equi-δM_c (dashed) curves on the (H_i, H_d) base. The attached numbers show the values of the corresponding equi-δM and equi-δM_c curves.

particular value of the external field H, one can determine H^i_{int} and H^d_{int} by the intersection of the corresponding equi-δM and δM_c curves in Fig. 4.

At this point we assume that, at zero external field, the interaction field H^i_{int} is equal to zero, due to the random particle magnetization distribution in the demagnetized state. If one also assumes that H^d_{int} is not zero at the remanence state after saturation, then $\delta M_c(H = 0)$ will be equal to $-M^i(H^d_{\mathrm{int}})$. However, as a result of the Fourier description of a couple of initial magnetization-hysteresis curves, one can obtain a δM_a plot as defined in (7), which for zero external field corresponds to $\pi/2$ in the Fourier expansion [18]. Obviously, δM_a, defined as a difference between two even functions (each of them is a sum of cosines), will always be zero at this field.
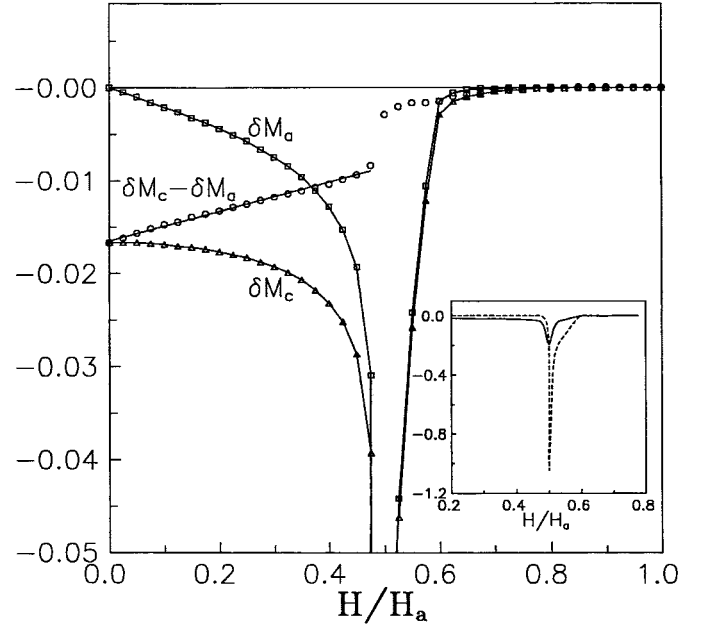

Fig. 5. δM_a (squares) and δM_c (triangles) plots, calculated for the case of $H^i_{\mathrm{int}}(H) = 0.1 M^{\mathrm{SW}}_i(H)$ and $H^d_{\mathrm{int}}(H) = 0.1 M^{\mathrm{SW}}_d(H)$, as well as the difference between them, $\delta M_c - \delta M_a$ (circles). The inset shows the δM (dashed curve) and δM_c plots in full scale.

Thus, there is a difference between the δM_a and δM_c plots. In order to use our model, as developed above, the experimental δM_a plot must be transformed somehow to a δM_c plot.

To do this transformation, we investigated the behavior of δM_c and δM_a by calculating the initial magnetization curves, $M^i(H_i)$, the hysteresis loops, $M^d(H_d)$, the "artificial" initial magnetization curves, $M^i(H_d)$, as well as the remanence curves assuming several different interaction field dependencies, $H^i_{\mathrm{int}}(H)$ and $H^d_{\mathrm{int}}(H)$. For example, the δM_a and δM_c plots, calculated for the particular case of $H^i_{\mathrm{int}}(H) = -0.1 M^{\mathrm{SW}}_i(H)$ and $H^d_{\mathrm{int}}(H) = 0.1 M^{\mathrm{SW}}_d(H)$ are presented in Fig. 5 (the inset in the same figure shows the δM and δM_c plots in full scale). The $M^{\mathrm{SW}}_i(H)$ and $M^{\mathrm{SW}}_d(H)$ are the remanence curves versus the external field for a Stoner–Wolfarth (SW) system. The difference between the two plots, $\delta M_c - \delta M_a$ is also shown. Here it is accepted that in the Fourier expansion, this difference is distributed between the two functions used to represent the hysteresis curve (5), as at $H = 0$, it is taken by the sine part only. Up to the minimum in the plots, this difference can be fitted with a sufficient accuracy by a correction function, $f_{\mathrm{corr}}(H)$ in the form of a straight line

$$f_{\mathrm{corr}}(H) = -M^i(H^d_{\mathrm{int}|_{H=0}}) - kH. \qquad (10)$$

The value of $M^i(H^d_{\mathrm{int}|_{H=0}})$ can be calculated as follows. If H^d_{int} is not zero at $H = 0$, there will be a deviation of the saturation remanence $M_r(\infty)$ from the SW value of 0.5, and then H^d_{int} can be estimated as the field causing this deviation. After that $M^i(H^d_{\mathrm{int}|_{H=0}})$ can be calculated.

The slope of the correction function is taken to be $-k$, where k is determined by the initial slope of the δM_a plot. It is reasonable to assume that, as $H^i_{\mathrm{int}} = 0$ in the close vicinity

of $H = 0$, the change of δM_a in this region is determined by the H_{int}^d only.

After the point of the minimum in the plots, the difference $\delta M_c - \delta M_a$ is very small, and the correction function can be regarded as equal to zero.

For all the interaction field dependencies used in calculations of the magnetization curves, the shape of the $\delta M_c - \delta M_a$ curve was approximately the same, and the way of estimation of $f_{\mathrm{corr}}(H)$ holds for all of them.

Therefore, the sequence of steps one should follow to estimate the interaction fields from experimental magnetization curves are:

1) the construction of δM and δM_a plots using (1) and (7);
2) the estimation of $M^i(H_{\mathrm{int}|_{H=0}}^d)$ from the saturation remanence value and the slope k from the initial part of the δM_a plot;
3) the transformation of the δM_a plot to a δM_c plot by adding f_{corr} (10);
4) the determination of H_i and H_d for a given couple of $\delta M(H)$ and $\delta M_c(H)$, by the intersection of the corresponding equi-δM and δM_c curves in Fig. 4 and subsequently, evaluation of H_{int}^i and H_{int}^d from (2) and (3).

The method has been applied on a sample of disordered Ba–ferrite fine particles, using the experimental plots of Fig. 1, from which in can be seen that $\delta M(H)$ decreases monotonically as H increases in the initial part of the plot. This behavior can not be explained in the framework of the SW model unless one assumes a very sharp increase of H_{int}^d/H_a from the $H_{\mathrm{int}|_{H=0}}^d$ value, which does not seem to be very reasonable. The initial decrease of the $\delta M(H)$ is probably caused by the presence of interaction fields after recoil, leading to an increase of M_r, a decrease of M_d, and thus a decrease of δM. This effect will be introduced in the calculations in a following work.

The thermal activation effects have been taken into account in the calculations, using the particle size distribution from [15] as the sample under consideration here is the same one. For a uniaxial particle, the lifetime τ of a given metastable state separated by an energy barrier ΔE from the other metastable state is given by

$$\tau^{-1} = f_0 \exp(-\Delta E/kT) \qquad (11)$$

first proposed by Néel [19]. The pre-exponential factor f_0 for a measurement time of 1 s is typically taken to be 10^{11} s. Here, it is assumed that whenever $\tau > 1$ s, the magnetic moment stays at the first metastable state. Otherwise, it jumps to the other metastable state.

The obtained H_{int}^i/H_a and H_{int}^d/H_a dependencies versus the external field are shown in Fig. 6. The initial parts of the curves (dashed lines) represent the interaction fields obtained by using the experimental δM_a plot only and assuming a monotonous decrease of these fields when H increases from 0 to $H_a/2$. The other parts of H_{int}^i/H_a and H_{int}^d/H_a curves (solid curves) have been obtained by using both δM and δM_a plots. It should be noted that the H_{int}^d curve can not be fitted by a function of a form given by (4). The latter yields $H_{\mathrm{int}|_{H=0}}^d =$
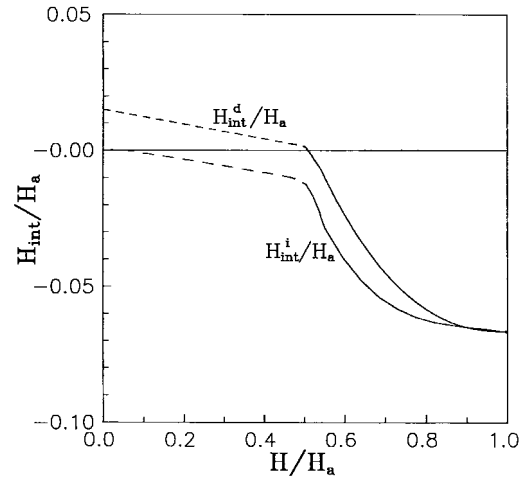

Fig. 6. H_{int}^i/H_a and H_{int}^d/H_a for Ba–ferrite fine powder, obtained from the data in Fig. 1.

$-H_{\mathrm{int}|_{H=\infty}}^d$ as $M_d(H = 0) = 1 = -M_d(H = \infty)$. In the present model, the interaction fields act during the application of the external magnetic field and depend on the system's magnetization and interparticle configuration. As our system consists of a collection of particles of random orientation, $H_{\mathrm{int}|_{H=0}}^d$ can not be equal to $H_{\mathrm{int}|_{H=\infty}}^d$, as it is for aligned particles, because the magnetization states are different at these external fields. These are the saturation remanence state for the case of zero external field (the particles' magnetizations distributed in a hemisphere) and magnetizations aligned along the external field direction for the case of $H = \infty$.

III. CONCLUSION

A method for estimation of the interaction fields H_{int}^i and H_{int}^d, acting during the initial magnetization and demagnetization processes versus the external magnetic field in disordered fine particle systems, has been developed. The method uses both experimental $\delta M_a(H)$ and $\delta M(H)$ plots, constructed from the initial magnetization curve and hysteresis loop and the remanence curves, respectively. The experimental dependencies of the interaction fields versus the external magnetic field for a Ba–ferrite fine powder have been estimated.

REFERENCES

[1] O. Henkel, "Remanenzverhalten und wechselwirkungen in hartmagnetischen teilchenkolleltiven," *Phys. Stat. Sol.*, vol. 7, pp. 919–929, 1964.
[2] E. P. Wohlfarth, "Relations between different modes of acquisition of the remanent magnetization of ferromagnetic particles," *J. Appl. Phys.*, vol. 29, pp. 595–596, 1958.
[3] M. Fearon, R. W. Chantrell, and E. P. Wohlfarth, "A theoretical study of interaction effects on the remanence curves of particulate dispersions," *J. Magn. Magn. Mat.*, vol. 86, pp. 197–206, 1990.
[4] R. Proksch and B. Moskowitz, "Interactions between single domain particles," *J. Appl. Phys.*, vol. 75, no. 10, pp. 5894–5896, 1994.
[5] P. E. Kelly, K. O'Grady, P. I. Mayo, and R. W. Chantrell, "Switching mechanisms in cobalt-phosphorus thin films," *IEEE Trans. Magn.*, vol. 25, no. 5, pp. 3881–3883, 1989.
[6] R. J. Veitch, "Anhysteretic susceptibility and static magnetic properties of interacting small particles," *IEEE Trans. Magn.*, vol. 26, no. 5, pp. 1876–1878, 1990.

[7] X. He, C. Alexander, Jr., and M. R. Parker, "Measurement of interaction fields in particulate tapes by linearization of the Henkel plot," *IEEE Trans. Magn.*, vol. 28, no. 5, pp. 2683–2685, 1992.

[8] Y. Kato and M. Tsutsumi, "An analysis of the magnetization reversal process," *IEEE Trans. Magn.*, vol. 28, no. 5, pp. 2686–2688, 1992.

[9] X. Che and H. N. Bertram, "Phenomenology of δM curves and magnetic interactions," *J. Magn. Magn. Mat.*, vol. 116, pp. 121–127, 1992.

[10] J. W. Harrell, D. Richards, and M. R. Parker, "Delta-H plot evaluation of remanence behavior in barium ferrite tapes and disks," *J. Appl. Phys.*, vol. 73, no. 10, pp. 6722–6724, 1993.

[11] T. R. Hoffend and M. J. Vos, "Phenomenological models of interactions and fluctuations in magnetic recording media," *IEEE Trans. Magn.*, vol. 31, no. 6, pp. 2901–2903, 1995.

[12] E. C. Stoner and E. P. Wohlfarth, "A mechanism of magnetic hysteresis in heterogeneous alloys," *Philosoph. Trans. R. Soc. London*, vol. A240, pp. 559–642, 1948.

[13] V. Masheva, J. Geshev, and M. Mikhov, "Fourier analysis of hysteresis loops and initial magnetization curves: Application to the singular-point-detection method," *J. Magn. Magn. Mat.*, vol. 137, pp. 350–357, 1994.

[14] J. Geshev, V. Masheva, and M. Mikhov, "Interaction effects detected by Fourier analysis of hysteresis loops and initial magnetization curves," *IEEE Trans. Magn.*, vol. 30, no. 2, pp. 863–865, 1994.

[15] O. Popov, P. Rachev, M. Mikhov, F. Calderon, J. L. Sanchez Ll., and F. Leccabue, "Experimental study of the Hopkinson effect in fine $BaFe_{12}O_{19}$ particles," *J. Magn. Magn. Mat.*, vol. 99, pp. 119–122, 1991.

[16] S. J. Greaves, K. O'Grady and A. M. Lane, "Interaction effects in barium ferrite dispersions using low temperature magnetometry," *IEEE Trans. Magn.*, vol. 32, no. 5, pp. 4046–4048, 1996.

[17] V. Patel, M. El-Hilo, K. O'Grady, and R. W. Chantrell, "Fluctuation fields of reversal in oriented and nonoriented barium ferrite media," *IEEE Trans. Magn.*, vol. 29, no. 6, pp. 3622–3624, 1993.

[18] R. M. Josephs, D. S. Crompton, and C. S. Crafft, "Characterization of magnetic oxide recording media using Fourier analysis of static hysteresis loops," *IEEE Trans. Magn.*, vol. 22, no. 5, pp. 653–655, 1986.

[19] L. Néel, "Influence des fluctuations thérmiques sur l'aimantation de grains ferromagnétiques très fins," *Comptes Rendus*, vol. 228, pp. 664–666, 1949.