Interaction Fields Evaluation in Fine Particle Systems

J. Geshev and J. E. Schmidt

Abstract—A computer simulation model for estimation of the interaction fields in fine particle systems has been developed. The method uses the experimental $\delta M(H)$ 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, which gives the sign and the relative strength of the interactions as well as their field dependencies. Here $H_i$ and $H_d$ 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 $\delta M$ plot is the $\delta H$ plot [6] which gives information about the interaction field strength as a function of the magnetic 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 ($H_{ix}$ and $H_{id}$, 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

$H_i = H + H_{ix}$

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

$H_d = H + H_{id}$

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

However, neither of the procedures mentioned above can give a direct evaluation of $H_{ix}$ and $H_{id}$ 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

$H_{\text{int}}(M_{ix}) = \alpha M_{ix} + f(M_{ix})$.  \hspace{1cm} (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 $\alpha$ and $\beta$ were then determined by numerical simulations of $\delta M$ curves using (4) and choosing parameters that would produce agreement with the measured $\delta M$ curves.

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

$\tilde{m}_i(x) = \sum_{n=1}^{N} a_{i,n} \cos(nx) + \sum_{n=1}^{N} b_{i,n} \sin(nx)$

$\tilde{m}_d(x) = \sum_{n=1}^{N} a_{d,n} \cos(nx)$. \hspace{1cm} (6)

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

$\delta M_0(H) = \sum_{n=1}^{N} a_{i,n} \cos(nH) - \sum_{n=1}^{N} a_{d,n} \cos(nH)$

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 $\delta M_0(H)$ plot is not zero in certain field intervals, and the shape of this plot is similar to the shape of the corresponding...
δM(H) plot. In Fig. 1, experimental δM(H/\Ha) and δM(0)/H/\Ha) for a M-type Ba–ferrite powder [14] are represented (\Ha 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 δM(H) and δM(0)/H/\Ha) 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 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 \(H_{int}\) and \(H_{int}^{\perp}\) directly, by using both the experimental δM(H) and δM(0)/H/\Ha) 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, \(H_{int}\) and \(H_{int}^{\perp}\), 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 \(H_{int}^{\perp}\) 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

\[
\delta M(H_i, H_d) = M_d(H_d) - (1 - 2M_i(H_i))
\]  

(8)
can be obtained, and Fig. 2 shows its three–dimensional (3-D) plot versus \(H_i/\Ha\) and \(H_d/\Ha\). 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 \(\delta M\) in the plot corresponds to \(H_i\) and \(H_d\) less than \(0.5\Ha\), when no irreversible rotation of the particle’s magnetization occurs.

By definition, \(\delta M(0)/H/\Ha\) (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 \(M_i(0)/H/\Ha\).

We introduce a relation, \(\delta M_c\), as the difference between the real and the “artificial” initial magnetization curves, \(M_i(0)/H/\Ha\) and \(M_i(H)\), respectively, calculated for different internal field dependencies

\[
\delta M_c(0)/H/\Ha = M_i(0)/H/\Ha - M_i(H/\Ha).
\]  

(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 \(\delta M_c\) as a function of \(H_i/\Ha\) and \(H_d/\Ha\) is represented. It can be seen that when \(H_i\) and \(H_d\) are less than \(0.5\Ha\) (and are not equal), \(\delta M_c\) is not zero, as it is for \(\delta M\) in Fig. 2. The reason is that \(\delta M_c\) in contrast to \(\delta M\) is determined by both reversible and irreversible rotations.

The projections of a set of equi-\(\delta M\) curves (curves with the same \(\delta M\) and different \(H_i/\Ha\) and \(H_d/\Ha\)), 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-\(\delta M\) projection, the \(\delta M\) data alone are not enough to estimate the interaction fields, because a \(\delta M\) value may be obtained for different couples of \(H_i\) and \(H_d\).

In the same figure, several equi-\(\delta M_c\) 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 \(\delta M\) and \(\delta M_c\) for a...
Fig. 3. 3-D plot of $\delta M_c$ versus $H_d/H_a$ and $H_d/H_a$, calculated by using (9).

Fig. 4. Projections of several equi-$\delta M$ curves (the solid ones) and equi-$\delta M_c$ curves (dashed) curves on the $(H_d, H_a)$ base. The attached numbers show the values of the corresponding equi-$\delta M$ and equi-$\delta M_c$ curves.

Fig. 5. $\delta M_a$ (squares) and $\delta M_c$ (triangles) plots, calculated for the case of $H_{1}(H) = 0.1M_{SW}(H)$ and $H_{d}(H) = 0.1M_{SW}(H)$, as well as the difference between them, $\delta M _{a} - \delta M _{c}$. The inset shows the $\delta M$ (dashed curve) and $\delta M _{c}$ plots in full scale.

Thus, there is a difference between the $\delta M_a$ and $\delta M_c$ plots. In order to use our model, as developed above, the experimental $\delta M_a$ plot must be transformed somehow to a $\delta M_c$ plot.

To do this transformation, we investigated the behavior of $\delta M_a$ and $\delta M_c$ by calculating the initial magnetization curves, $M_i(H_d)$, the hysteresis loops, $M_d(H_d)$, the "artificial" initial magnetization curves, $M_i^a(H_d)$, as well as the remanence curves assuming several different interaction field dependencies, $H_{a}^R(H)$ and $H_{d}^R(H)$. For example, the $\delta M_a$ and $\delta M_c$ plots, calculated for the particular case of $H_{a}^R(H) = -0.1M_{SW}(H)$ and $H_{d}^R(H) = 0.1M_{SW}(H)$ are presented in Fig. 5 (the inset in the same figure shows the $\delta M$ and $\delta M_c$ plots in full scale). The $M_{SW}^R(H)$ and $M_{SW}^d(H)$ are the remanence curves versus the external field for a Stoner–Wolffarth (SW) system. The difference between the two plots, $\delta M_a - \delta M_c$ 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, $f_{\text{corr}}(H)$ in the form of a straight line

$$f_{\text{corr}}(H) = -M_i(H_{\text{int}}|H=0) - kH.$$  

The value of $M_i(H_{\text{int}}|H=0)\) can be calculated as follows. If $H_{\text{int}}^d$ is not zero at $H = 0$, there will be a deviation of the saturation remanence $M_s(\infty)$ from the SW value of 0.5, and then $H_{\text{int}}^d$ can be estimated as the field causing this deviation. After that $M_i(H_{\text{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 $\delta M_a$ plot. It is reasonable to assume that, as $H_{\text{int}}^d = 0$ in the close vicinity
of $H = 0$, the change of $\delta M_a$ in this region is determined by the $H^d_{\text{int}}$ only.

After the point of the minimum in the plots, the difference $\delta M - \delta M_a$ 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 $\delta M - \delta M_a$ curve was approximately the same, and the way of estimation of $f_{\text{corr}}(H)$ 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 $\delta M$ and $\delta M_a$ plots using (1) and (7);
2) the estimation of $M^d(H_{\text{int},a})$ from the saturation remanence value and the slope $k$ from the initial part of the $\delta M_a$ plot;
3) the transformation of the $\delta M_a$ plot to a $\delta M_c$ plot by adding $f_{\text{corr}}$ (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-$\delta M$ and $\delta M_c$ curves in Fig. 4 and subsequently, evaluation of $H^d_{\text{int}}$ and $H^d_{\text{int},a}$ 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 $H^d_{\text{int},a}/H_a$, from the $H^d_{\text{int},a}|_{H=0}$ 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 $M_i$, a decrease of $M_d$, and thus a decrease of $\delta M$. 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 $\tau$ of a given metastable state separated by an energy barrier $\Delta E$ from the other metastable state is given by

$$\tau^{-1} = f_0 \exp(-\Delta E/kT)$$  \hspace{1cm} (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 $H^d_{\text{int}}/H_a$ and $H^d_{\text{int},a}/H_a$ 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 $\delta M_a$ plot only and assuming a monotonous decrease of these fields when $H$ increases from 0 to $H_a/2$. The other parts of $H^d_{\text{int}}/H_a$ and $H^d_{\text{int},a}/H_a$ curves (solid curves) have been obtained by using both $\delta M_a$ and $\delta M_a$ plots. It should be noted that the $H^d_{\text{int}}$ curve can not be fitted by a function of a form given by (4). The latter yields $H^d_{\text{int}}|_{H=0} = -H^d_{\text{int}}|_{H\to\infty}$ as $M_a(H = 0) = 1 = -M_a(H = \infty)$. 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, $H^d_{\text{int}}|_{H=\infty}$ can not be equal to $H^d_{\text{int},a}|_{H=\infty}$, 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 $H^d_{\text{int}}$ and $H^d_{\text{int},a}$ 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 $\delta M_c(H)$ 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
