Assessing interface coupling in exchange-biased systems via in-field interaction plots

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An in-field interaction plot, δMR, has been recently introduced, presenting important advantages over the classical remanence plots. Here a general δMR is proposed, allowing to assess interactions even in systems with shifted and asymmetric major loops. To construct such a plot, a recoil loop (which incorporates a first-order reversal curve, FORC) and the position of the center of the major loop are only needed. Applying the method on exchange-biased Co/IrMn bilayer gives two types of δMR obtained for measuring field either parallel or antiparallel to the exchange-bias direction. This provides valuable information on the reversal mechanism and allows distinguishing between effects coming from coupling into the ferromagnet (Co) and those stemming from interactions in its interface with the antiferromagnet (IrMn). The essentially nonzero general δMR plot obtained from the major loop revealed to be a precise measure of the reversal asymmetry. The technique can readily be adjusted for use in other scientific fields where hysteresis is observed. We provide free software which generates such δMR plot(s).

I. INTRODUCTION

Wohlfarth pointed out a simple relation1 between remanence curves of systems with symmetric major magnetization M, versus magnetic field H, hysteresis loops. These are the isothermal remanent magnetization curve MR(H), which represents the remanence obtained by the application and removal of a positive field on an initially demagnetized sample, and the DC demagnetization curve MR(H), i.e., the remanence resultant from the application of a negative field to a sample initially at saturation remanence. The Wohlfarth’s relation

\[ M_d(H) = 2M_r(H) - M_r(\infty) \]  

should be valid for non-interacting uniaxial-anisotropy systems no matter whether the magnetization reversal occurs via domain nucleation followed by domain-wall motion or coherent rotation.

Nonzero δM(H) = 2M_r(H) − M_d(H) − M_r(∞) values of, e.g., initially thermally or AC demagnetized samples, are ascribed to magnetic interactions; positive values are normally attributed to exchange-like coupling favoring a ferromagnetic state and negative values are associated with dipolar-like interactions stabilizing the demagnetized state.2–5 Non-interacting cubic-anisotropy systems present intrinsically positive δM plots.6–8

In exchange bias (EB) systems with shifted (by the so-called EB-field, H_{eb}) and often asymmetric major hysteresis loops, δM plots cannot be used in their classical forms. Even though this technique has been adapted to biased systems, it still requires demagnetization. Interaction plots based on initial magnetization and hysteresis curves are easy to obtain and present characteristics very similar to those of the remanence ones. Nevertheless, these still require an initially demagnetized state.

Generalized and/or integral δM plots and functions, obtained with the help of first-order reversal curves (FORCs) have also been proposed.11–14 Methods based on FORCs and also on second-order reversal curves (SORCs) and remanent SORCs have been used to study magnetization reversal mechanisms and interactions as well. Pike et al.15 have claimed that FORC diagrams give more precise information on magnetic interactions than the δM plots. FORCs have also been used in studies of magnetic interactions in EB systems.18–20 However, the greatly-increased amount of FORC and SORC data as compared to those of the remanence plots, together with the complexity of their analyzes, could make their interpretation rather difficult, particularly true when magnetic interactions are present and the Preisach-like interpretation is not applicable. The technique is often considered as a magnetic fingerprint and not a method that provides quantitative information.21–23

Recently, a relation analogous to that of Wohlfarth but between in-field magnetization curves has been derived for systems with symmetric major hysteresis loops,

\[ M_{rec}(H) = 2M_{bys}(H) - M_{sym}(H). \]  

Here \( M_{bys} = \frac{1}{2}(M_{dsc} + M_{asc}) \), being \( M_{dsc}(H) \) and \( M_{asc}(H) \) the descending and ascending branches of the major loop, and \( M_{sym}(H) \) the curve symmetric, in respect to the origin of the coordinate system, to the extended recoil curve \( M_R(H) \) note that the latter also represents a FORC with reversal field \( H_R \). Based on Eq (2) an in-field interaction plot has been introduced,

\[ \delta M_R(H) = M_R(H) + M_{sym}(H) - 2M_{bys}(H). \]  

It is acquired in an easier and faster manner than δM and does not demand demagnetization, significantly simplify-
First, a relation interconnecting four parts of a recoil loop allowing the definition of the general $\delta M_R$ plot is derived. A $M_R(H)$ loop, measured after positive saturation for $\phi_H = 0^\circ$ of the Co/IrMn film, is given in Fig. I(a) where $M - M_{\text{shift}}$ is plotted as a function of $H = H_{\text{ext}} - H_{eb}$, being $M_{\text{shift}}$ the shift of the major loop along the magnetization axis (a non-zero $M_{\text{shift}}$ could be observed in a system for which, e.g., the maximum positively-saturated field ($-H_{\text{max}}$) is unable to reverse some positively-saturated magnetization components). Up to $H$ equal to the recoil field $H_R$, the ascending parts with positive and negative $H$ values of the respective recoil loop coincide with those of the major loop, $M^+_d(H)$ and $M^-_d(H)$. The ascending parts of the recoil loop, with negative and positive $H$ are denoted here as $M^+_R(H)$ and $M^-_R(H)$. A recoil curve is traced after some soft magnetization components have rotated irreversibly along $M^-_d(H)$. Since for ideal systems only reversible changes occur along $M^-_R(H)$, the magnetization varies by $M^+_R(H) - M^-_R(H) = M^-_d(H) - M^-_R(H)$ for $H < 0$. Along $M^-_R(H)$, the soft components reverse back their magnetizations and the respective variation equals $M^-_d(H) - M^-_R(H)$. The two variations differ only in sign, so

$$M^+_R(H) - M^+_d(H) = M^-_R(H) - M^-_d(H).$$

One can further extend the recoil curve by assuming that, in the $-H_{\text{max}} \leq H \leq H_R$ field region, $M^-_R(H) \equiv M^-_d(H)$, making Eq. 4 valid for all $H$. Let refer to $M^-_d(-H)$ as the curve symmetric of $M^-_d(H)$ through the center of the major loop, and to $M^-_R(-H)$ as the curve symmetric of $M^-_R(H)$, i.e., $M^-_d(-H) = -M^-_d(H)$ and $M^-_R(-H) = -M^-_R(H)$. Utilizing these curves (see Fig. I), we define

$$\delta M_R(H) = M^+_R(H) + M^-_R(-H) - M^+_d(H) - M^-_d(-H).$$

Note that the ascending part of the major loop does not take the part of the above equations. Evidently, the plot introduced for symmetric loops is a special case of the general $\delta M_R(H)$, where $M_R(H) \equiv M_{\text{asc}}(H)$. Besides a recoil loop, the only parameter needed for the construction of a general $\delta M_R(H)$ is the position of the center of the major loop ($H_{eb}, M_{\text{shift}}$).

### III. RESULTS AND DISCUSSIONS

For the case of uniaxial anisotropy, nonzero deviations of $\delta M_R(H)$ are ascribed to magnetic interactions. The shape of $\delta M_R$ shown in Fig. 1(a) is similar to that of the plot obtained for the unbiased Co film with symmetric major loop. For thin films, an initial increase of $\delta M(H)$ and $\delta M_R(H)$ is attributed to parallel (ferromagnetic) exchange coupling and a negative dip to antiparallel (dipolar-like) interactions. However, as it will be demonstrated below, at least part of the negative

**FIG. 1.** Magnetization curves measured for the IrMn/Co film with $H_{eb} = -242$ Oe for $\phi_H = 0^\circ$. (a) Representative recoil loop $M_R(H)$ and the corresponding $\delta M_R$ plot. Here $M^+_d(-H)$, dash line, and $M^-_R(-H)$, dot line, are the curves symmetric through the origin of $M^-_d(H)$ and $M^-_R(H)$. The ascending branch of the major loop (grey line) is also plotted; (b) using the latter as $M_R(H)$ gives rise to a nonzero $\delta M_{\text{major}}$.}

**II. GENERAL $\delta M_R$ PLOT**

Here a $\delta M_R$ plot more general than that introduced for symmetric hysteresis loops is introduced, allowing to assess interactions even in EB systems. The technique is applied to analyze data obtained at 300 K via EZ9 MicroSense vibrating sample magnetometer on a magnetron-sputtered, onto a Si(100) substrate, Ta(5 nm)/Ru(15 nm)/Co(5 nm)/IrMn(7 nm)/Ta(3 nm) film, where IrMn refers to a (111)-textured Ir$_{20}$Mn$_{80}$ layer, and on a film with the same composition except for the structural and magnetic properties of these films are reported in Ref. [7]. The EB direction of the Co/IrMn film was set by an in-plane magnetic field applied during deposition. Its direction is given by $\phi_H$, where $\phi_H = 0^\circ$ and $180^\circ$ refer to $H_{\text{ext}}$ parallel and antiparallel to the EB direction.

The determination of $H_{eb}$ is correlated to that of the coercivity ($H_c$) which, normally, is considered as the half-width at half-height of a hysteresis loop. In EB systems, however, due to the characteristic loop’s asymmetry, a more general definition is used. It employs $H_{sw1}$ and $H_{sw2}$, i.e., the respective switching fields of $M_{\text{asc}}$ and $M_{\text{dsc}}$, resulting in $H_c = 1/2(H_{sw2} - H_{sw1})$ and $H_{eb} = 1/2(H_{sw1} + H_{sw2})$. Here, $H_{sw1}$ and $H_{sw2}$ are the positions of the peaks of the first-order field derivatives of $M_{\text{asc}}$ and $M_{\text{dsc}}$, respectively.
\[ \delta M_{R}(H) \] could result from the asymmetry of the magnetization reversal typical for EB systems.

The technique can also be applied to major loops by taking \( M_{\text{asc}}(H) \) as recoil curve in Eq. 5. The asymmetry of an EB major loop, with one of its branches steeper than the other, results in an essentially nonzero \( \delta M_{R}^{\text{major}}(H) \) as seen in Fig. 2(b). Such a plot of the unbiased Co film with symmetric major loop equals zero for any \( H_{\text{ext}} \). Thus, the \( \delta M_{R}^{\text{major}} \) plot in Fig. 2(b) is a footprint of FM/AF interface coupling.

The information concerning interactions estimated from the \( \delta M_{R}^{c} \) plot from Fig. 2(a) could be compared to that obtained from the remanence \( \delta M \) plots displayed in Fig. 2. Since our Co/IrMn film presents EB, these plots [where the states of \( M_{t}(H_{\text{ext}}) = 0 \) were attained by dc demagnetization] are obtained following the method introduced in Ref. 7. The shapes of the two such plots displayed in Fig. 2 are characteristics for ferromagnetic coupling with no indication for presence of demagnetizing interactions, differently from \( \delta M_{R}(H) \).

This dissimilarity should be attributed to the distinct routines used by the remanence and in-field magnetization techniques. Here, the \( \delta M_{R}^{\text{major}} \) plot is obtained from remanence curves measured for \( -H_{\text{max}} \leq H_{\text{ext}} \leq H_{\text{eb}} \), and \( \delta M_{R}^{+} \) derives from curves traced for \( H_{\text{eb}} \leq H_{\text{ext}} \leq H_{\text{max}} \). Each plot reflects magnetization reversals that occur along either the descending or the ascending branches of the major hysteresis loop. In contrast, a \( \delta M_{R} \) is generated from a recoil loop with \( H_{\text{ext}} \) cycled following the \( H_{\text{max}} \rightarrow H_{\text{eb}} \rightarrow H_{\text{R}} \rightarrow H_{\text{eb}} \rightarrow H_{\text{max}} \) path. According to its definition, \( \delta M_{R} \), correlates processes taking place along the descending loop’s branch with processes occurring along the ascending branch. Thus, \( \delta M_{R} \), differently from \( \delta M \), evidences effects stemming from the asymmetry of the reversal, indicating that the negative part of the \( \delta M_{R} \) from Fig. 2(a) might originate from this asymmetry.

Major and recoil loops with \( H_{R} \approx H_{c} \), measured for \( \phi_{H} = 0^\circ \) and \( 180^\circ \) for descending fields of the Co/IrMn bilayer, are shown in Figs. 3(a) and (b); the resultant \( \delta M_{R} \) and \( \delta M_{R}^{\text{major}} \) plots are given in Figs. 3(c) and (d). Due to the greater slope of the descending branch as compared to the ascending one of the major loop traced for \( \phi_{H} = 0^\circ \), its \( \delta M_{R}^{\text{major}} \) is virtually negative. Given the reversed asymmetry of the \( 180^\circ \) major loop (with descending branch with lesser slope than the ascending one), the \( \delta M_{R}^{\text{major}} \) is mainly positive. It is identical to \( -\delta M_{R,0^\circ}^{\text{major}} \) through shifted in field by \( 2|H_{\text{eb}}| \).

On the other hand, the asymmetry of the magnetization reversal results in rather different \( \delta M_{R} \) plots for \( \phi_{H} = 0^\circ \) and \( 180^\circ \). While the former presents an initial increase followed by a negative dip, the latter is essentially positive. Its deviations from the zero line are almost negligible in the field region where the respective \( \delta M_{R}^{\text{major}} \) initiates its growth. Noteworthy, \( \delta M_{R} \) in Fig. 3(c) changes from positive to negative at virtually the same field at which the negative growth of \( \delta M_{R}^{\text{major}} \) begins. These features strongly support the suggestion that the effects of the reversal’s asymmetry determining the shape of \( \delta M_{R}^{\text{major}} \) are also evidenced in \( \delta M_{R} \).

It is instructive to further elucidate the effects on the interaction plots caused exclusively by changes of the FM/AF interface coupling. Our Co/IrMn film presents very stable EB properties given that its \( H_{\text{eb}} \) and \( H_{c} \) values have not practically changed over the six-year period after its deposition. Nevertheless, it is possible to induce significant variations of \( H_{\text{eb}} \), and even reverse the EB direction, conducting the following experiment. A piece of the bilayer was kept at 300 K in 4 kOe static magnetic field, sufficient to saturate the FM along the direction antiparallel to the EB one. After certain time intervals, hysteresis and recoil loops were measured and immediately after that the sample was placed back at the con-
with sample kept with saturating H film measured after progressively increasing time intervals above treatment). This has led to a gradual decrease of some biasing uncompensated spins (UCSs) located at the interface with the adjacent FM. This has led to a gradual decrease of the so-called thermal EB field drift, $\phi_H = 0^\circ$ equals $H_{eb}$, and the respective recoil loops for $H_{eb} \approx H_c$ (dashed lines) of the Co/IrMn film measured after progressively-increasing time intervals with sample kept with saturating $H_{ext}$ antiparallel to the EB direction; each magnetization is normalized to the respective saturation value $M_s$. (b) Temporal change of $H_{eb}$. The respective $\delta M^\text{major}_R$ and $\delta M_R$ are shown in (c) and (d); the plots obtained for $\phi_H = 180^\circ$, dash lines in (d), are shifted by $2H_{eb}$.

The positive parts of all $\delta M_R$ plots in Fig. 4(d) are not essentially altered by the superposition since $\delta M^\text{major}_R$ is roughly nil in the respective field regions. The negative $\delta M_R$ plots for $\phi_H = 0^\circ$ become deeper and the respective field region extends as compared to that of the unbiased film, resembling the characteristics of the (negative) $\delta M^\text{major}_R$ plots. The $\delta M_R$ plots obtained for $\phi_H = 180^\circ$, on the other hand, do not practically retain negative values, eradicated by the superimposed curves proportional to the (positive) $\delta M^\text{major}_{R,180^\circ}$ plots.

Thus, at least for our bilayer, the cross-examination of the major and the pair of recoil-loop plots allowed distinguishing effects coming from magnetic coupling into the FM layer (the initial, positive part of $\delta M^\text{major}_R$) from those stemming from interactions at its interface with the AF (the negative $\delta M_R$ for $\phi_H = 0^\circ$); the proper existence of a non-zero $\delta M^\text{major}_R$ plot is a signature of FM/AI coupling.

Recoil loops and $\delta M_R$ plots might provide valuable information on the magnetization reversal mechanism associated to differences in the nucleation process, e.g., these can indicate whether the reversal occurs via either domain-wall motion or coherent rotation. Figure 5
shows a pair of experimental major and recoil loops measured for the Co/IrMn film together with fitting curves calculated through the polycrystalline model for EB. It considers that the FM consists of small-sized domains calculated through the polycrystalline model for EB using $t_{\text{set}} = t_{\text{rot}} = 0.5 \text{ nm}$, $m_{\text{set}} = m_{\text{rot}} = 0.64M_{\text{FM}} = 900 \text{ emu/cm}^3$, $J_{\text{set}}t_{\text{set}} = 24J_{\text{rot}}t_{\text{rot}} = 1.67 \times 10^{-1} \text{ erg/cm}^3$. Gaussian FM easy-axis distribution with 30° standard deviation, equally distributed easy axes of the rot-type UCSs, and uniaxial anisotropies: $K_{\text{FM}} = 7.35 \times 10^6 \text{ erg/cm}^3$. $K_{\text{set}} = 68K_{\text{rot}} = 9 \times 10^6 \text{ erg/cm}^3$. (b) The respective $\delta M_{\text{R}}^\text{major}$ and $\delta M_{\text{R}}$ plots.

FIG. 5. (a) Symbols: major hysteresis loop and a recoil loop of the Co/IrMn film measured at $\phi_H = 0^\circ$ after it was kept for 1 hour at $\phi_H = 180^\circ$ in $H_{\text{ext}} = 4 \text{ kOe}$. Lines: curves calculated via the polycrystalline model for EB using $t_{\text{set}} = t_{\text{rot}} = 0.5 \text{ nm}$, $m_{\text{set}} = m_{\text{rot}} = 0.64M_{\text{FM}} = 900 \text{ emu/cm}^3$, $J_{\text{set}}t_{\text{set}} = 24J_{\text{rot}}t_{\text{rot}} = 1.67 \times 10^{-1} \text{ erg/cm}^3$. Gaussian FM easy-axis distribution with 30° standard deviation, equally distributed easy axes of the rot-type UCSs, and uniaxial anisotropies: $K_{\text{FM}} = 7.35 \times 10^6 \text{ erg/cm}^3$. $K_{\text{set}} = 68K_{\text{rot}} = 9 \times 10^6 \text{ erg/cm}^3$. (b) The respective $\delta M_{\text{R}}^\text{major}$ and $\delta M_{\text{R}}$ plots.

FIG. 6. FORCs (a) and $\delta M_{\text{R}}$ plots (b) of the Co/IrMn bilayer. Map of the $\delta M_{\text{R}}(H_{\text{FM}}, H_{\text{ext}})$ values plotted inside the major hysteresis loop (c). The right half of the map is obtained using the data from panels (a) and (b), and the left half from data attained for $\phi_H = 180^\circ$ (not shown). The $\delta M_{\text{R}}$ plot denoted by the dashed line in (b) corresponds to the path shown by a dashed line in (c). The variations of the interaction parameter $\alpha$ with $H_{\text{FM}}$ for $\phi_H = 0^\circ$ and $180^\circ$ are given in (d).
major loops, $\delta M_R^{\text{major}} = 0$ and so does $\alpha$. Such a parameter should certainly be employed in phenomenological models (yet to be developed) for quantitative assessment of interactions through $\delta M_R$ plots. The variations of $\alpha$ with $H_R$ for both measurement configurations, $\phi_R = 0^\circ$ and $180^\circ$, are given in Fig. 4(d). Whereas due to the reversal asymmetry the two curves are not identical, these present practically one and the same amplitude, though attained at different $H_R$ values.

Certainly, more systematic studies on the method should be conducted in a variety of systems to clarify its full potentiality for interaction effects estimations.

IV. SUMMARY AND CONCLUSIONS

The general $\delta M_R$ plot introduced here and applied to Co/IrMn gives a simple, yet efficient, way to assess interaction effects even in systems with shifted and asymmetric major hysteresis loops. The essentially nonzero $\delta M_R^{\text{major}}$ plot revealed to be a precise measure of the reversal asymmetry. Also, the two distinct $\delta M_R$ plots, obtained for the same recoil field but for measuring field parallel or antiparallel to the exchange-bias direction, together with the two-dimensional $\delta M_R$ diagram, provide valuable information on the magnetization reversal mechanism and allow distinguishing effects coming from magnetic coupling into the ferromagnet from those stemming from interactions in its interface with the antiferromagnet. Also, the here-defined interaction parameter, i.e., the area enclosed by a $\delta M_R$ curve, could be used to quantitative measure the interaction effects and the major loop’s asymmetry. This technique can readily be adjusted for assessing effects caused by deviations from theoretical behavior of other hysteretic quantities.

Free software which generates the introduced here general $\delta M_R$ plots and also yields $\alpha$ is available for download at http://www.if.ufrgs.br/pes/lam/dMr.html

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