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Asymmetric magnetization reversal in exchange-biased polycrystalline F/AF bilayers

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Abstract

This paper describes a model for magnetization reversal in polycrystalline ferromagnetic (F)/antiferromagnetic (AF) exchange-biased bilayers. We assume that the exchange energy can be expanded into cosine power series. We show that it is possible to fit experimental asymmetric shape of hysteresis loops in exchange-biased bilayer for any direction of the applied field. The hysteresis asymmetry is discussed in terms of energy considerations. An angle β is introduced to quantify the easy axis dispersion of AF grains.

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Keywords: Exchange bias; Thin magnetic films; Magnetization reversal

1. Introduction

Exchange bias is a phenomenon known for a long time [1] and characterized by a displacement (of shift value H_s), often called "bias field", of the hysteresis loop along the field axis as a result of exchange coupling between ferromagnetic (F) and antiferromagnetic (AF) layers. The original model proposed by Meiklejohn and Bean [1] is based on a perfect uncompensated monolayer of spin at the surface of the AF layer. This model predicts a shift field H_s that is two orders of magnitude larger than that observed experimentally. Several recent theories give improved predictions of the magnitude of H_s, but they interpret in a different way the physical origin of the effect. For example, Mauri et al. [2] have shown that realistic values for H_s can be obtained if one assumes a domain wall formation in the AF layer during the reversal of the F magnetization. Using localized atomic spins, Koon [3] suggested 90° spin–flop coupling between the AF and F layers and predicted a correct magnitude of H_s. A different approach was undertaken by Malozemoff [4] who assumed that the interface roughness greatly reduces the number of uncompensated spins at the AF surface giving rise to a smaller bias field. Stiles and McMichael have proposed a model for polycrystalline F/AF bilayers [5] consisting of an F layer interacting with independent AF grains using three contributions for the energy of each grain, a direct coupling, a spin–flop coupling and a partial

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domain wall in the AF grains. They showed that the unidirectional anisotropy comes from stable grains as the magnetization is rotated, while the hysteretic effects come from grains where the AF order is switchable. In all cases the spin–flop coupling does not contribute to the unidirectional anisotropy. Despite many theoretical studies of the origin and the magnitude of the bias field, a complete description of the exchange anisotropy should incorporate explanations for the other features of the exchange-biased systems such as the directional properties of the exchange bias and the strong asymmetry of the hysteresis loop that occurs in these systems. The angular dependence of H_s and the coercivity H_c was first explored experimentally in NiFe/CoO bilayers [6]. The variation of H_s and H_c versus the field angle θ_H was described with cosine Fourier series, with odd and even terms for H_s and H_c, respectively, instead of simple sinusoidal functions as suggested initially [1]. The angular dependence of the exchange anisotropy in F/AF bilayers has been studied both theoretically and experimentally by several authors. For some of them [6,7], the complicated angular behavior comes from the larger contribution of higher-order terms $a_n \cos(n\theta_H)$ $(n \geqslant 3)$ in H_s and H_c. Kim et al. [8] have modeled the F/AF bilayer as a cubic structure and used an Heinsenberg Hamiltonian to examine the angular dependence of perfect interfaces. For compensated interface, their numerical results of angular dependence of the exchange field were fit following the work of Ambrose et al. [6], who have shown that the angular dependence has a simple cosine function. For a rough interface, the behavior is more complicated and simple cosine form is not sufficient and higher-order terms appear. Kim et al. [8] suggested that the possible origin of the higher-order terms may come from the nonrigid spin structure of the AF layer. Geshev et al. [9] assumed a planar domain wall formation at the AF side of the interface with the reversal of F orientation [9]. Xi and White [10] have shown that the complex angular dependence of the exchange bias in NiFe/CrMnPt bilayers can be understood by a simple Stoner–Wohlfath model involving only a uniaxial and a unidirectional exchange coupling. Very recently Krivorotov et al. [11] have shown that the exchange anisotropy, induced by the F/AF exchange coupling, consists of a biaxial component, which gives rise to the enhanced coercivity. In the latter model, the unidirectional component is responsible for the shift of the hysteresis loop and the threefold symmetry component for the symmetry breaking in the magnetization reversal process. Tang et al. [12] have recently shown that the exchange bias effect, described by a unidirectional anisotropy, is also accompanied by induced uniaxial and fourfold in-plane contribution in Fe/MnPd exchange-biased bilayers. Most of these authors obtain good agreement between the theoretical and experimental angular variation of H_s and H_c.

Many authors have only fit the angular dependence [6,8] of H_s and H_c. Xi et al. [10] have used the fit parameters for the angular dependence of the coercivity and the exchange field to calculate the hysteresis loops for different directions of the applied field. For the easy axis hysteresis loop, the coercivity is about two times larger than the measured one and the shifted hysteresis loop is symmetric. Our approach is different, first we fit the easy axis hysteresis loop and then, by using the fit parameters, calculate the angular dependence of the coercivity and the shift field and also the hysteresis loop for any angle of the applied field.

As to the asymmetry of the hysteresis loops in exchange-biased bilayers, there is a large variety of asymmetric shapes. The modeling of hysteresis loops and especially its asymmetry was achieved only for some particular shapes [2,13,14]. However, none of these above methods describe satisfactorily the shape of the measured easy axis hysteresis loop shown in Fig. 2(a). This is why we propose a theoretical model based on the development of the energy functional for the exchange field in cosine powers series. We show that the experimental asymmetric hysteresis loop measured along the easy axis can be fit with our theoretical model. In the present work, we study the magnetization reversal process using conventional (VSM) magnetometry in NiFe/MnNi bilayers. The theoretical magnetization curves, calculated for different orientations of the applied field, are in good agreement with the experimental data. We show that the asymmetry in the magnetization

reversal may be understood in terms of energetic considerations.

2. Sample preparation and analysis

Substrate\$Ni_{81}Fe_{19}$(520 Å)\$Mn_xNi_{100-x}$ (800 Å) bilayers were grown on Corning glass substrate by RF diode sputtering using a standard Z 550 Leybold equipment with a magnetic field of 24 kA/m Oe applied during deposition to induce an uniaxial anisotropy. The background pressure was lower than 4×10^{-7} mbar. Ni chips were homogeneously added to a 4 in diameter Mn target in order to get films in the Mn composition range 5–80 percent. The chemical homogeneity was verified by electron probe micro-analysis (EPMA) on several points of the sample. The Mn composition variation is about one percent on the entire sample. As the as-deposited samples did not exhibit exchange bias, after deposition, they were annealed in a magnetic field of 80 kA/m, aligned with the easy axis of the film, at 300°C for 5 h to induce the exchange field. The crystallographic structure was examined by X-ray diffractometry with Cu Kα radiation. A (1 1 1) texture for the $Ni_{81}Fe_{19}$ was favored. Annealed Mn_xNi_{100-x} films deposited on the underlying $Ni_{81}Fe_{19}$ film were found to have a (1 1 1) texture with an FCT structure in the composition range $40 < x < 80$ that exhibits exchange bias [15]. The magnetic properties, such as the saturation magnetization M_s, were obtained from magnetization loops (M–H loops) measured at room temperature using a VSM. It is well established that the shift of the hysteresis loop of exchange-biased bilayers depends on intrinsic properties such as the AF layer thickness, the F layer thickness and the deposition conditions (for a review see Ref. [16]). All these parameters induce hysteresis loop with a more or less pronounced asymmetry. For our study we have chosen samples with a well-defined magnetization reversal asymmetry observed by easy axis hysteresis loop. In our samples the forward loop shows a slope, as if the full magnetization reversal takes place slowly, while the recoil loop is very square which may be the consequence of either coherent magnetization flip (such as in monodomain particles) or the presence of high-mobility domain walls.

3. Model

The magnetization reversal that occurs in exchange-biased bilayers cannot be explained with a simple algebraic addition of an exchange field to the applied magnetic field H_a because of the asymmetric shape of the hysteresis loop. In order to take into account the exchange bias, Meiklejohn and Bean [1] have proposed a linear term that accounts for the hysteresis curve shift, but which does not explain the asymmetry of the hysteresis loops for exchange-biased F/AF systems. Owing to the large variety of hysteresis asymmetric curves, one should admit that one-parameter exchange energy is no longer sufficient to interpret the form of these curves. In order to take into account the complex physical phenomena giving rise to the asymmetry of hysteresis curve, other terms should be added to the exchange energy expression describing the F/AF exchanged-biased samples. Several authors added a nonlinear biquadratic term proposed by Slonczewski [17]. However, this term is not sufficient to account for a large variety of asymmetric hysteresis curves known for various systems of F/AF samples. Based on symmetry considerations, Ambrose et al. [6] proposed a Fourier odd and even series development of the shift field and the coercivity, respectively, that accounts for the angular variation of H_s and H_c. More recently, using the Stoner–Wohlfath model, involving only a uniaxial and a unidirectional exchange coupling, Xi et al. [10] fitted the hard axis hysteresis loop to experimental data. Nevertheless, the easy axis hysteresis loop shape fitted poorly with experimental curve. Different mechanisms of magnetization reversal, such as the AF domain wall formation, the different direction of easy axis in AF and F layers, the grain formation, etc., contribute to the hysteresis curve shape. Due to the complexity of magnetization reversal processes, which are strongly influenced by the physical state of F/AF interface and therefore very sensitive to the physical processes of sample preparation, a large

number of Fourier series terms should be taken into account in order to achieve an agreement between the experimental data and theoretical calculations. In general, Fourier series convergence is rather slow. This is why we propose a cosine power series expansion of the exchange energy. Moreover, the grain formation and the easy axis distribution of different grains with respect to the magnetization direction of F layer lead us to introduce a mean easy axis direction of different AF grains. The exchange coupling between different grains, and the way they are influenced by F layer, suggests that statistical mean easy axis of AF grains may deviate from that of F layer. The exchange coupling with the AF layer is assumed to result in a single domain in the F film as recently observed in $NiFe/Fe_{50}Mn_{50}$ by magneto-optic Kerr effect [18]. Therefore, we admit a uniform switching of the total magnetization of the F layer. The magnetic configuration of the F/AF bilayer is shown in Fig. 1. The magnetic energy per unit area of exchange-coupled F layer and AF layers can be written as follows:

$$E = K_F t_F \sin^2\varphi - H_a M_F t_F \cos(\theta - \varphi) - E_e(\varphi - \beta), \quad (1)$$

where K_F is the anisotropy constant of the F layer with its magnetization M_F making an angle φ with

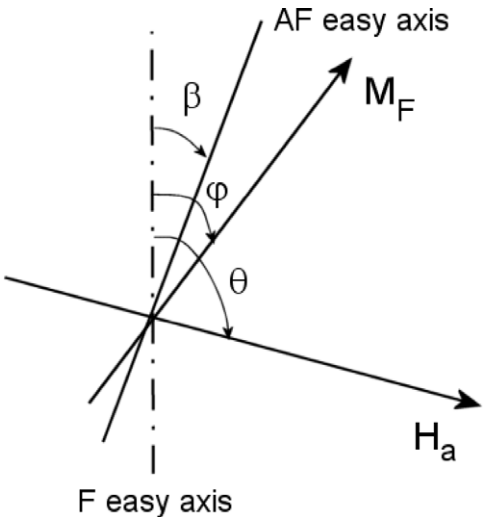

Fig. 1. Vector diagram for an exchange-coupled F/AF bilayer submitted to an applied field H_a.

respect to the F easy axis. The first term represents the anisotropy energy of the F layer. The second term corresponds to the Zeeman energy of the F layer submitted to an external static field H_a applied at an angle θ relative to the anisotropy easy axis of the F layer. The last term represents the exchange anisotropy energy, where β is the angle of the AF mean statistical easy axis with respect to the F easy axis. $E_e(\varphi\text{-}\beta)$ may be written as

$$E_e(\varphi - \beta) = \sum_n J_n \cos^n(\varphi - \beta). \quad (2)$$

If one admits, a priori, that $E_e(\varphi - \beta)$ is linear, $E_e(\varphi - \beta) = J_1 \cos(\varphi - \beta)$, then that represents the simple unidirectional energy originally proposed by Meiklejohn and Bean [1] (with $\beta = 0$) and used recently by Xi et al. [10] (with $\beta \neq 0$). The biquadratic term of $E_e(\varphi - \beta)$ was already used by Slonczewski [17] and later by Stamps [13] to describe the coupling of the F layer to both sublattices of two-sublattice antiferromagnet. Sometimes, based on crystal symmetry considerations, threefold and fourfold components were used in $E_e(\varphi - \beta)$ energy functional. The threefold component was recently found to be responsible for the symmetry breaking in the magnetization reversal process in Fe/MnF_2 [11]. Tang et al. [12] have shown that the exchange bias effect, described by a unidirectional anisotropy, is also accompanied by induced uniaxial and fourfold in-plane contribution in Fe/MnPd exchange-biased bilayers. Because of the complexity of the exchange bias phenomenon, we cannot limit our development to any finite number of terms. So the total energy in terms of effective field can be written as

$$\frac{E}{t_F M_F} = \frac{H_K}{2}\sin^2\varphi - H_a \cos(\theta - \varphi) - \sum_n H_n \cos^n(\varphi - \beta), \quad (3)$$

where H_K is the anisotropy field and the H_n are the coefficients of the power series development. Based on the above model, the magnetization curves of the F/AF bilayer are obtained by numerical calculations.

The coefficients H_1, H_2, H_3 etc, being the power expansion coefficients of energy functional, do not reflect the overall energy symmetry. We have truncated cosine power series at $n = 5$ for which the fit to the shape of easy and hard axis hysteresis curves was the best. The fitting to the easy axis hysteresis loop improves when one takes into account more terms. However, in that case, the hysteresis loop calculated for $\theta \neq 0$ by using fit parameters of easy axis hysteresis loop shows parasitic minor loops. One may obtain better fit by using 10 or 20 series terms. But that will not mean anyhow that the crystal has a 10th- or 20th-order symmetry since only the sum of all terms should provide such symmetry and not the each term. When we truncated the expansion to $n = 4$ the best fit to easy axis hysteresis curve becomes square.

Providing a physical signification to the 5th-order coefficient is difficult since our samples were prepared with RF sputtering and none of the textures were well pronounced. After a thermal annealing, an interdiffusion have been induced [19]. Moreover, as reported in a previous work [15], a ternary NiMnFe alloy seems to be formed in the F/AF interface region. Chen et al. [19] showed that the interdiffusion alters dramatically the exchange bias and that the hysteresis loop shape becomes asymmetrical. To the best of our knowledge, there is no systematic study concerning the influence of the alloy formed at the interdifusion layer at the F/AF interface on the exchange energy functional. So it is a difficult task to associate a physical meaning with the exchange energy expansion coefficients.

4. Results and discussions

In the first step we have fitted the hysteresis loop to the theoretical model given by Eq. (3), when the field is applied along the easy axis. The comparison between theoretical and experimental loops is shown in Fig. 2(a). One can observe a strong asymmetry between the left and right side of the measured hysteresis loop. The magnetization reversal on the left side of the loop exhibits a hard-axis-type slope, while the reversal for the right-hand side of the loop occurs with a vertical

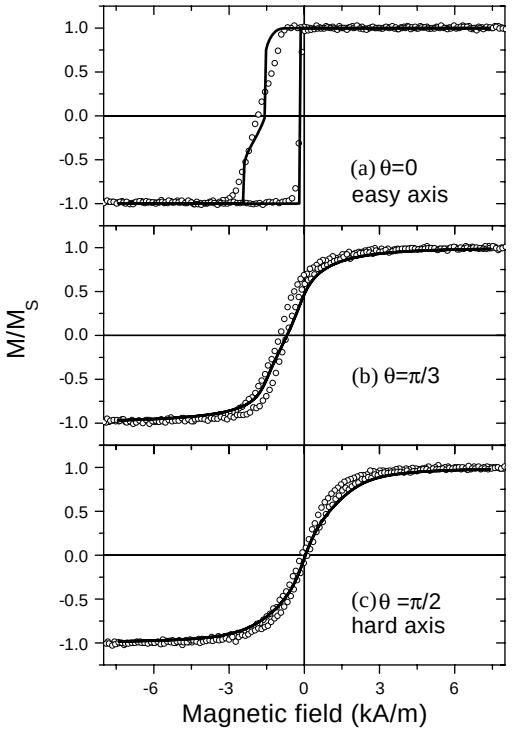

Fig. 2. Representative hysteresis loops for a $Ni_{81}Fe_{19}$(520 Å)/$Mn_{46}Ni_{54}$(800 Å) bilayer at (a) 0, (b) $\pi/3$, (c) $\pi/2$ of the applied field referred to the easy axis. The experimental data are shown by scattered empty dots. The solid lines are the theoretical magnetization curves.

branch of the loop. It can be observed that the theoretical magnetization curve fits well the measured hysteresis loop. The model gives values expected either for the hysteresis loop shift ($H_s = 950$ A/m) and coercivity ($H_c = 900$ A/m) or for the strong asymmetry in the magnetization reversal. The values of the fit parameters are $\beta = -0.064$, $H_K = 533$ A/m, $H_1 = 1661$ A/m, $H_2 = -903$ A/m, $H_3 = 616$ A/m, $H_4 = 501$ A/m, $H_5 = -599$ A/m. The first term H_K is about two times larger than the anisotropy field of the as-deposited NiFe layer (i.e. 360 A/m). The second term H_1, that may be related to the exchange-biased field, is about 1.5 larger than the hysteresis loop shift.

One can observe a distinct kink on the left side of the calculated magnetization loop, whereas the experimental data show a smooth variation. Leigthon et al. [20] attributed that kind of kink

to a two-stage magnetization reversal. The defect concentration is of high importance for exchange bias characterization as indicated by several authors (see the review by Stamps [13]). The higher-order Fourier coefficients in the exchange energy functional of single-crystal samples are generally much larger than those for the polycrystalline samples. In polycrystalline materials the situation is complicated due to the spread in AF grain concentration. It is further complicated in the presence of an interdiffusion layer that may be a ternary NiMnFe alloy. According to Mewes et al. [21] the Stoner–Wolfarth model shows sharp edges which are rounded in the experiment for polycrystalline samples. This may explain the smooth variation of our experimental results.

Energy diagrams are often useful to explain the magnetization reversal in magnetic heterostructures. Fig. 3 represents the left (Fig. 3(a)) and the right (Fig. 3(b)) side of the measured hysteresis loop. In our calculations, for each value of the applied field, the system is in a local energy minimum which is plotted for the forward (left side) and reverse (right side) loops in the same figure. The energy curve presents an hysteretic behavior as a function of applied field. In the same figure, one can observe a strong asymmetry in the magnetization reversal due to the slow variation of energy local minimum as a function of applied field. On the right side of the figure (Fig. 3(b)), there is an abrupt lowering of local energy minimum at about $H_a = 0$, which corresponds to an abrupt change of the energy minimum angle (φ). At the left side of the loop, the magnetization must overcome an energy barrier that vanishes after an increase ΔH of the field (Fig. 3(a)). This reversal may occur with a number of discontinuous jumps giving rise to "kinks" in the hysteresis loop. For the reverse loop, the magnetization vector flips rapidly from $\varphi = \pi$ to 0 at a critical field about $H_a \simeq 0$.

We have used the same fit parameters to calculate the magnetization curves for an applied field making different angles ($\pi/3$ and $\pi/2$) with the F easy axis. The results are shown in Figs. 2(b) and (c). As observed in the Fig. 2(b) and (c), the small deviation of experimental and theoretical curves is attributed to the initial dispersion of the as-deposited NiFe anisotropy revealed by a small coercivity measured along the hard axis. This may come from the aligning field in the sputtering chamber which lacks uniformity and may give rise to the F anisotropy dispersion. Indeed, we did not include in our model such a dispersion term which

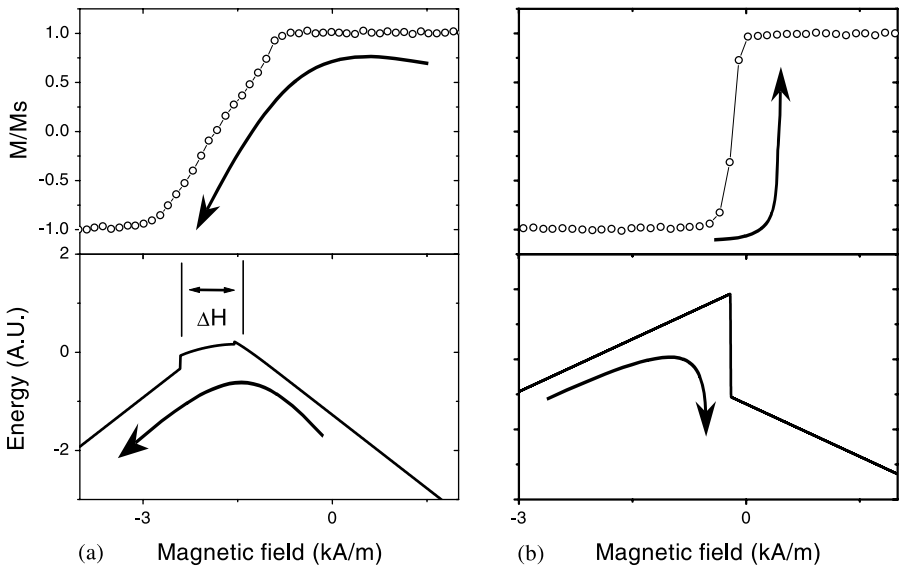

Fig. 3. (a) Forward measured magnetization hysteresis loop (upper) and corresponding calculated energy hysteresis loop (lower). (b) Reverse measured magnetization hysteresis loop (upper) and corresponding calculated energy hysteresis loop (lower).

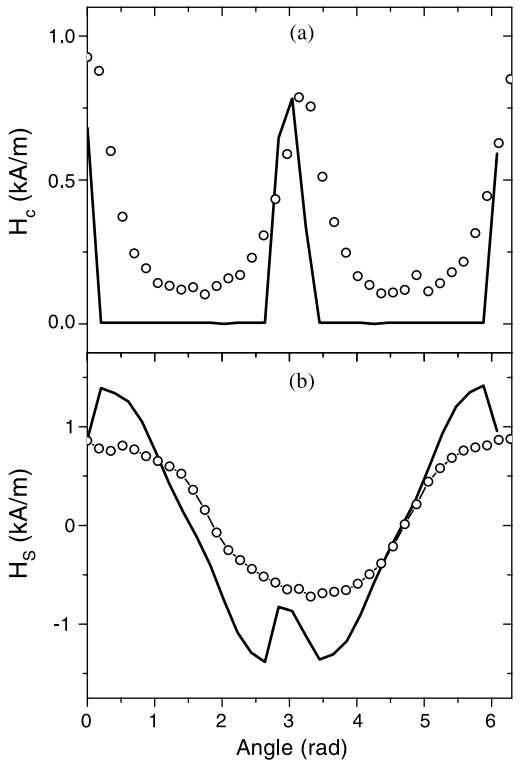

Fig. 4. Angular dependence of (a) shift field H_s and (b) coercivity of an exchange coupled NiFe/MnNi bilayer. The open dots represent the experiments and the solid line the theory.

is the main cause of that difference. Fig. 4 describes the comparison of theoretical and experimental angular variation of the coercivity H_c (Fig. 4(a)) and the hysteresis loop shift H_s (Fig. 4(b)). The fit parameters correspond to the easy axis hysteresis loop. The behavior of the coercivity is very similar to that found by Ambrose et al. [6]. The measured coercivity does not vanish in the directions different from that of the easy axis ($\theta \neq 0$). For some angles there is discrepancies between the computed hysteresis shift and the measured one. First, the model predicts maxima of H_s located at about $\theta = \pm\pi/15$ and $\pi \pm 2\pi/3$. This behavior is different from the believed one, according to which the largest values of H_s are expected at $\theta = 0$ and π as mentioned by several authors [6,10,4]. We observed experimentally the largest values of H_s for $\theta = 0$ and π. Our angular dependence of the coercivity and hysteresis loop shift differs from the published results [6]. In our calculations the theoretical angular variation of H_c and H_s is obtained from a unique set of parameters deduced by the fit to the easy axis hysteresis loop instead of two different fits for the coercivity and the exchange field as usually presented [6,7].

We think that the discrepancy between theoretical and experimental results in Fig. 4 is due to the fact that our samples are polycrystalline. Riedling et al. [22] show that a complex $H_s(\theta)$ behavior may be due to the epitaxially grown samples. Liu et al. [23] realised epitaxially grown NiFe/FeMn exchange-biased bilayers on Si(1 0 0) films buffered by Cu. LEED experiments supported the existence of a six fold symmetry. The obtained $H_s(\theta)$ and $H_c(\theta)$ curves were strongly dependent on the growth technique. Polycrystalline samples show perfect $\cos(\theta)$ behavior even in the presence of a well-pronounced induced (1 1 1) texture. This work supports our experimental data where also a well-defined $\cos(\theta)$ behavior was observed. The epitaxial samples in the work of Liu et al. show that $H_s(\theta)$ and $H_c(\theta)$ curves have several minima for different Cu buffer layers. The behavior of $H_s(\theta)$ and $H_c(\theta)$ curves shapes may also depend on the existence of an interdiffusion layer, but we do not have sufficient experimental data to confirm or exclude such an influence.

An important feature of the present model is the nonzero value of $\beta = -0.064$ which is the angle between the F and AF easy axis directions. We suppose that it is due to the AF grains formation and the local exchange energy variation at the interface of F/AF layers. Owing to the interface imperfections, the easy axis of AF grains will suffer some dispersion [24,5]. A statistical mean easy axis direction can be a useful parameter to quantify that dispersion. The physics of such dispersion is somewhat complicated due to the lack of information about the exchange energy local variation, the roughness characteristics and its influence on the energetic balance of concurrent energetic processes for energy minimization. This is why the value of β, a priori, may be slightly different from zero. So we think that it can be an additional pertinent parameter to describe the exchange bias.

5. Conclusions

In summary, we have presented a model based on the representation of the exchange energy by cosine power series. We have shown that it is possible to fit the asymmetric shape of the easy axis hysteresis loop. The asymmetric hysteresis loop shape is discussed on the basis of energy considerations. An angle β is introduced to quantify the easy axis dispersion of AF grains. The easy axis fit parameters describe satisfactorily the hysteresis curves in other directions.

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