Experimental method to determine the misalignment of the easy axis of ferromagnetic and antiferromagnetic films in exchange biased bilayers

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A hard axis hysteresis loop shift from the origin has been observed in the exchange biased MnPt/NiFe. A model that accounts for the hard axis hysteresis loop shift is proposed. For the correct description of the experimental results, a misalignment between the easy axis of ferromagnetic and antiferromagnetic films in the exchange biased MnPt/NiFe bilayers has to be taken into account. An experimental method to measure the misalignment in noncollinear exchange biased systems is developed. The method is based on the measurements of hard axis hysteresis loops at angles $\pi/2$ and $(-\pi/2)$ with respect to the easy axis of ferromagnetic film. A study of the misalignment as a function of antiferromagnetic layer thickness is carried out. Our approach is generalized for complex systems with high-order anisotropies.

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I. INTRODUCTION

Exchange biased ferromagnetic (F) antiferromagnetic (AF) systems were the subject of intensive studies in the past decade due to their applications in spin valves and other devices.[1] In a F-AF bilayer, the magnetic moment of F film is sometimes strongly coupled at the interface to the magnetic structure of AF film. That gives rise to an enhanced coercive field, to a shift of the hysteresis curve, and sometimes to an asymmetry of the latter.[2–5]

Several models were proposed to describe the exchange-biased systems. A nonexhaustive list of past studies concerning these systems can be found in reviews Refs. 2–5. We will mention only some of them, especially those closely related to our work. The model of Meiklejohn and Bean[6] is based on the exchange coupling between the uncompensated monolayer spin at the surface of the AF layer with the magnetic moment of F film. Their model predicts a shifted square hysteresis curve along the easy axis of the F film; however, the magnitude of the shift seems to be different from that predicted theoretically. In order to correctly account for the observed shift (H_e) and the enhancement of the coercive field (H_c), domain wall formation in the AF or in the F films was proposed.[2,7] In most models, the influence of the misalignment of the easy axis of F and AF films was neglected. It is now well established that in some F/AF exchange coupled systems, such a misalignment exists.[8–15] The misalignment observed by Haas *et al.*[8] was greater than 7°. Xi and White described the angular dependence of the exchange anisotropy of noncollinear systems.[10] Layadi showed that the ferromagnetic resonance technique can be used to study the exchange anisotropy of misaligned systems.[11] The misalignment influence on the torque curve was investigated in Ref. 12. Jungblut *et al.*[13] has seen some experimental evidence for the existence of misalignment NiFe/MnFe exchange biased bilayers.

In a recent paper, we proposed a phenomenological model based on the Fourier power series representation of exchange energy in order to account for the trapeze-like shape of the easy axis hysteresis curve[14,15] of NiFe-NiMn exchange coupled bilayers. We have shown that the asymmetrical form of a hysteresis curve may be related to the misalignment of the easy axis of F and AF films.

In a recent paper, Camarero *et al.*[16] treated the asymmetry in collinear exchange biased bilayers and underlined the importance of noncollinear systems in exhibiting a wide variety of asymmetrical hysteresis curves.

In the present work, we have observed a hard axis hysteresis loop shift from the origin in the exchange biased MnPt/NiFe bilayer. A model that accounts for the hard axis hysteresis loop shift is proposed. For the correct description of the experimental results, we consider a misalignment between the easy axis of ferromagnetic and antiferromagnetic films. The misalignment is determined from the measurements of hard axis hysteresis loops at angles $\pi/2$ and $(-\pi/2)$ with respect to the easy axis of ferromagnetic film. We study the misalignment as a function of AF layer thickness in exchange biased MnPt/NiFe systems. Then we generalize our method for complex systems with high-order anisotropies.

II. THEORY

We consider that the magnetization has a uniform distribution along the axis perpendicular to the F film and there is no helical structure along that direction.[17] In our model we treat the AF layer as an assembly of very small noninteracting grains with each grain having a randomly distributed easy axis with respect to the easy axis of the F film.[18–24] In each AF layer the magnetic structure is considered to be frozen into a rigid spin configuration exchange coupled with the magnetic moment of the F film.

The easy axis direction of the jth AF grain makes an angle θ_j with the easy axis of the F film. The exchange coupling strength of the jth AF grain to the F film is noted by J_j and the anisotropy constant of AF grains by K_{AF}. The jth AF grain has an area S_j and thickness t_{AF}. If we consider that the $K_{\mathrm{AF}}t_{\mathrm{AF}}S_j$ (anisotropy energy of the jth grain) is very large compared to the interfacial exchange term J_jS_j, then in each grain the AF spins are assumed to be aligned along their easy

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axis and therefore the AF anisotropy energy is neglected.[25] Thus only terms related to the F film will contribute to the energy of the system.

Under an external magnetic field with the strength H applied in the film plane and making an angle α with the F easy axis, the energy E of the system can be written in the following form:

$$E = K_f S_f t_f \sin^2(\varphi) - \mu_0 HMS_f t_f \cos(\alpha - \varphi) - \sum_j J_j S_j \cos(\theta_j - \varphi); \quad (1)$$

all in-plane spins in F film are oriented along the same angle φ with respect to the easy axis of the F film.[17] S_f is the area of the F film. K_f is the anisotropy constant in the F film. M is the F saturation magnetization.

Our model is different from that of Fulcomer and Charap,[18,19] in spite of some similarities. For instance, the AF layer is treated as an assembly of very small noninteracting grains exchange coupled to the magnetic moment of the F monodomain film. In Fulcomer and Charap's model, it is assumed that the easy axis of all AF grains is oriented along the same direction. Usually, the direction of F easy axis is privileged for calculations. The direction AF spins may be different from that of AF easy axis.

In our model we assume that the easy axes of AF grains are randomly distributed with respect to the F easy axis. The AF spins in each grain are supposed to be frozen along the grain's easy axis. Our model reflects well the AF/F bilayer spin configuration in which the AF film is deposited first. The easy axes of AF grains are defined by deposition process, the surface state, the texture, etc. The deposition of F film on AF does not change considerably the easy axis directions of AF grains. The easy axis of F film is well defined since it is grown under applied field. Therefore, our model is basically different from the model of Fulcomer and Charap, and predicts a different behavior for AF-F exchange coupled bilayers.

The sum in expression (1) can be written in the following form:

$$\sum_j J_j S_j \cos(\theta_j - \varphi) = \cos(\varphi) \sum_j J_j S_j \cos(\theta_j) + \sin(\varphi) \sum_j J_j S_j \sin(\theta_j). \quad (2)$$

In general, J_i is the same for all AF grains. However, in the case of stoichiometric (texture) inhomogeneities, a possible dispersion of J_i may appear for different grains. In the most general case we assume that J_i may be different from grain to grain.

If we introduce two new statistical parameters J and β defined as

$$\tan(\beta) = \frac{\sum_j J_j S_j \sin(\theta_j)}{\sum_j J_j S_j \cos(\theta_j)}, \quad (3a)$$

$$J = \left\{ \sqrt{\left[\sum_j J_j S_j \sin(\theta_j) \right]^2 + \left[\sum_j J_j S_j \cos(\theta_j) \right]^2} \right\} \Big/ S_f, \quad (3b)$$

then

$$\sum_j J_j S_j \cos(\theta_j - \varphi) = J S_f \cos(\varphi - \beta) \quad (3c)$$

and finally the total energy can be written in the following form:

$$E = K_f S_f t_f \sin^2(\varphi) - \mu_0 HMS_f t_f \cos(\alpha - \varphi) - JS_f \cos(\varphi - \beta). \quad (4)$$

As can be seen from Eq. (3a) (see also Refs. 14 and 15), β is the statistical mean angular deviation of the AF easy axis from that of F film. The hysteresis curve is obtained in the Stoner-Wolfarth coherent rotation approximation by minimizing the energy with respect to the angle φ. In the energy expression β has a fixed value since it describes the mean deviation of the easy axis of individual AF grains with respect to the easy axis of F film and we suppose that a small external field cannot alter the AF ordering. Thus Eq. (4) can be written in the following form:

$$E_r(\varphi) = \frac{H_f}{2} \sin^2(\varphi) - H \cos(\alpha - \varphi) - H_e \cos(\beta - \varphi). \quad (5)$$

Here we have noted $E_r = E/\mu_0 M t_f S_f$, $H_f = 2K_f/\mu_0 M$, and $H_e = J/\mu_0 M t_f$.

In the case of exchange-biased systems having asymmetrical hysteresis curves along the easy axis of F film (NiFe/MnNi, for example[14,15]) the exchange energy cannot be described by a simple energy term: $JS_f \cos(\varphi - \beta)$ [Eq. (4)]. This is due to the complexity of magnetization reversal processes. In a previous paper we introduced a phenomenological model based on exchange energy cosine power development. As shown in the Appendix, even in this more general case, a statistical angle β can be introduced to describe the mean angular deviation of the F and AF easy axis.

The angular dependence of hysteresis curves of F-AF bilayers with perfectly aligned easy axis has specific features. For example, the maximum value of the coercivity appears for the easy axis hysteresis loop ($\alpha = 0$). The in-plane hard axis hysteresis loops ($\alpha = \pi/2$ and $\alpha = -\pi/2$) are not shifted with respect to the origin. They pass through the origin ($H = 0$, $M = 0$) and generally it is believed that one should obtain the same curve when it is measured either at $\alpha = \pi/2$ or at $\alpha = -\pi/2$.

The angular dependence of the hysteresis curve of F-AF bilayers with misaligned easy axis is different. The maximum value of coercivity does not appear along the easy axis of the F film.[10] The hard axis hysteresis loop is shifted with respect to the origin. It can be shown with the help of Eq. (5) that for misaligned F/AF exchange coupled bilayers, the hysteresis curves are different when measured at angles $\alpha = \pi/2$ and $\alpha = -\pi/2$ with respect to the F easy axis. Thus, the misalignment can be described indirectly with the help of the difference of these curves (at $\alpha = \pi/2$ and $\alpha = -\pi/2$).

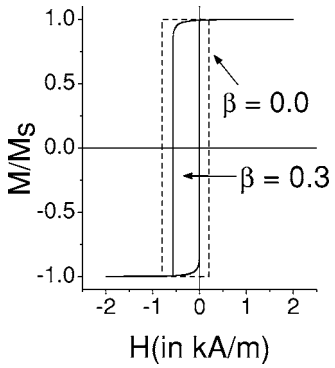

FIG. 1. Easy axis ($\alpha=0$) hysteresis loop obtained by minimization of Eq. (5). The values of H_f and H_e are supposed to be 0.5 and 0.3 kA/m, respectively. The dashed line corresponds to $\beta=0$ and the solid line to $\beta=0.3$.

In Figs. 1 and 2, one can see the influence of the misalignment angle on easy and hard axis hysteresis loops, respectively. These curves are calculated by numerical minimization of Eq. (5). As can be seen in Fig. 1, the magnetization rotation along the easy axis for $\beta=0.3$ rad is not abrupt, as in the case of $\beta=0.0$ rad. The magnetization switching does not occur at $H=H_e\pm H_f$. In Fig. 2, one can see that the hard axis hysteresis loop for $\beta=0.3$ rad does not pass through the origin ($H=0$, $M=0$).

In order to describe experimentally the misalignment angle β, let us call the beta indicator (BI) the normalized modulus of the difference of the hysteresis curves measured at angles $\alpha=\pi/2$ and $\alpha=-\pi/2$ with respect to the F easy axis at $H=0$,

$$BI=\left(\frac{M(H)_{\pi/2}-M(H)_{-\pi/2}}{M_s}\right)_{H=0}=\frac{M(0)_{\pi/2}-M(0)_{-\pi/2}}{M_s} \tag{6}$$

For a perfectly aligned system, BI is zero. For misaligned systems, BI is different from zero. We have calculated in Fig. 3 the BI as a function of β for different values of $\gamma=H_f/H_e$. As can be seen from this figure, the dependence of BI on $\sin\beta$ is practically linear. The minimization of the

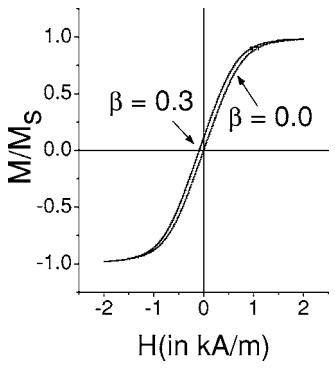

FIG. 2. Hard axis ($\alpha=\pi/2$) hysteresis loop is obtained by minimization of Eq. (5). The values of H_f and H_e are supposed to be 0.5 and 0.3 kA/m, respectively. The dashed line correspond to $\beta=0$ and the solid line to $\beta=0.3$.

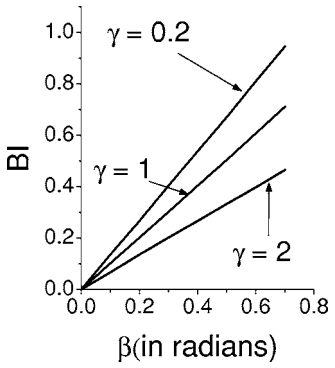

FIG. 3. Dependence of BI defined by Eq. (6) on β calculated with the help of hard axis hysteresis curves obtained by the minimization of the energy given by Eq. (5).

energy given by Eq. (5) for $\alpha=\pm\pi/2$ together with the definition of BI by Eq. (6) yields $BI=2\sin(\varphi)$. The latter can be transformed into

$$\sin\beta=\frac{BI}{2}\left[\sqrt{1-\gamma^2\frac{BI^2}{4}\left(1-\frac{BI^2}{4}\right)}+\gamma\left(1-\frac{BI^2}{4}\right)\right]. \tag{7}$$

For small values of BI and moderate values of γ, the dependence of β on BI is linear,

$$\beta\approx\frac{BI}{2}(1+\gamma). \tag{8}$$

If the value of γ is known, the value of BI can be measured from the in-plane hard axis hysteresis loops at $\alpha=\pi/2$ and $\alpha=-\pi/2$ with respect to the easy axis. However, as can be seen on Fig. 1, in the case misaligned F-AF bilayers, the value of H_f cannot be determined or estimated from the easy axis hysteresis loop. This is why we introduce another parameter Δ, which is the separation of the hysteresis loops at $\alpha=\pi/2$ and $\alpha=-\pi/2$ at $M=0$. The minimization of the energy given by Eq. (5) leads to the following expression for the separation parameter (SP) Δ (note that for $\alpha=\pm\pi/2$, $M=0$ if $\varphi=0$):

$$\Delta=2H_e\sin(\beta). \tag{9}$$

So the value of β can be determined experimentally, once the value of H_e is determined from the easy axis hysteresis loop shift. Thus BI and SP are pertinent parameters to determine the misalignment between the easy axis of F and AF films. In the following section, the misalignment angle between the easy axis of F and AF films in exchange coupled MnPt/NiFe bilayers is determined.

III. EXPERIMENTAL RESULTS

Glass / $Mn_{60}Pt_{40}(t_{AF})$ / $Ni_{81}Fe_{19}$(18 nm) / Ta(5 nm) / Al(5 nm) and glass / $Ni_{81}Fe_{19}$(18 nm) / $Mn_{60}Pt_{40}(t_{AF})$ / Ta(5 nm) / Al(5 nm) were grown at room temperature with a magnetic field of 24 kA/m, applied during the deposition to induce uniaxial anisotropy in the F film, by using a rf diode sputtering system by standard Z550 Leybold equipment under Ar pressure of 1×10^{-2} mbar. The background

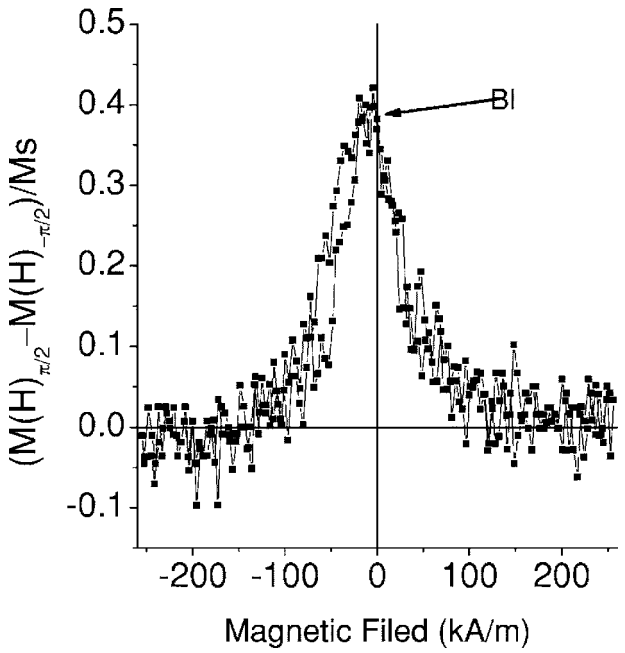

FIG. 4. Normalized difference of in-plane hard axis hysteresis loops at $\alpha=\pi/2$ and $\alpha=-\pi/2$ with respect to the F easy axis. BI is defined as the value of this curve at $H=0$.

pressure was better than 2×10^{-7} mbar. Pt(99.99%) chips were homogeneously added to a 4 in. diam Mn(99.99%) target in order to insure a Mn composition in which the MnPt/NiFe exhibits a shifted hysteresis loop after annealing.[27,28] The chemical composition was checked by XRF. After the deposition, the samples were annealed at 300 °C for 3 h without magnetic field to induce the exchange coupling. A (111) texture for the $Ni_{81}Fe_{19}$ was favored and the $Mn_{60}Pt_{60}$ films were found to have a (111) texture with a fct structure after annealing. The NiFe/MnPt samples do not show any bias.

The magnetization hysteresis loops were measured at room temperature with a vibrating sample magnetometer (VSM). The coercive field of the biased NiFe layer was defined as half of the shifted *M-H* loop width.

To obtain the BI or the SP, we started by saturating at $\alpha=+\pi/2$ (with respect to the F easy axis) and then obtained the first ($\alpha=+\pi/2$) hard axis hysteresis loop. The sample position was changed to $\alpha=-\pi/2$ position at $H=0$, and the second hard axis loop was measured starting from ($\alpha=-\pi/2$) saturated state. Figure 4 shows a typical experimental $\alpha=\pm\pi/2$ hard axis hysteresis loop difference curve. BI corresponds to the maximum value of the peak in Fig. 4. To measure the transverse and longitudinal magnetization components simultaneously, two additional coils were added to a standard VSM. The measurements of both components of magnetization—transverse and longitudinal—are drawn in Fig. 5. The measured values of the magnetization are normalized with respect to the value of the parallel saturation magnetization M_S. It can be observed in Fig. 5 that in the case of a magnetic field applied along the easy axis, the transverse magnetization may reach 60% of the parallel saturation magnetization. The two maxima of the transverse magnetization correspond to the coercive fields. A strong

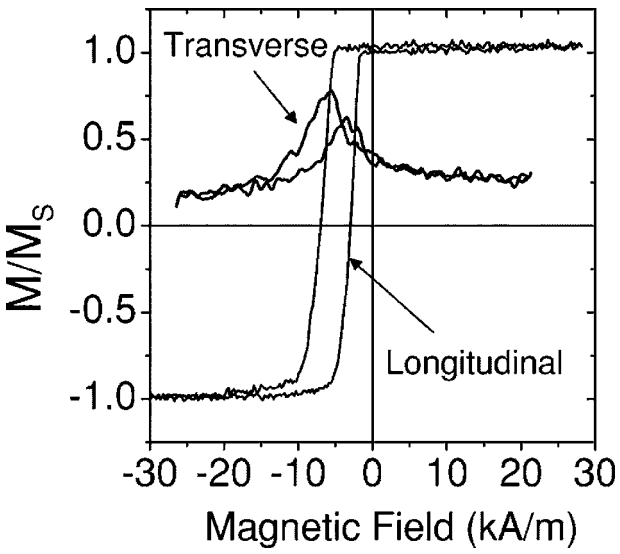

FIG. 5. Longitudinal and transverse magnetization hysteresis curves in MnPt/NiFe bilayer along the easy axis of the F film.

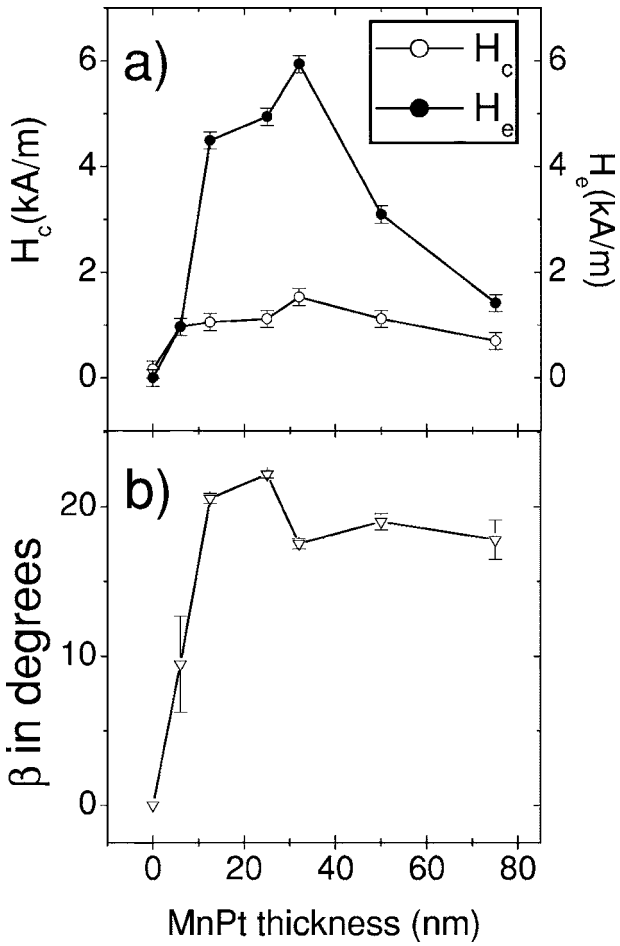

FIG. 6. (a) Dependence of H_e and H_c as a function of MnPt thickness in MnPt/NiFe exchange coupled bilayer. The critical thickness is estimated to be 8 nm. (b) Misalignment angle calculated Eq. (9) by measuring the separation parameter Δ from in-plane hard axis hysteresis loops.

asymmetry is observed in the transverse magnetization loop. That suggests that, in our samples, the magnetization reversal along the easy axis is complicated and should be considered as a mixture of magnetization rotation and domain-wall propagation. However, 60% of magnetization reversal by coherent rotation in the transverse magnetization curve of MnPt/NiFe bilayer (Fig. 5) justifies the use of the coherent rotational approximation in this case. In Fig. 6(a), we presented the behavior of H_c and H_e as a function of AF layer thickness.

The exchange biasing appears at a critical AF layer thickness t_{crit} of about 8 nm. At $t_{AF}=t_{crit}=8$ nm, H_e increases by passing through a maximum at $t_{AF}=25$ nm ($H_e^{max}=5$ kA/m) and decreases down to $H_e=1.5$ kA/m at $t_{AF}=78$ nm. Such behavior is generally observed at low temperatures.[29,30] At room temperatures, the shift field H_e has a constant value for t_{AF} greater than the critical thickness.[31]

In Fig. 6(b), the misalignment angle β is drawn as a function of t_{AF} determined by formula (9). For $t_{AF}<t_{crit}$, $\beta=0$ and for $t_{AF}>t_{crit}$, β increases by passing through a maximum at $t_{AF}=20$ nm and then saturates ($\beta_{sat}=20°$) at $t_{AF}=25$ nm. The curve of β as a function of t_{AF} behaves like H_c and H_e.

In MnPt/NiFe, the AF film is deposited first, therefore the easy axis directions of AF grains are already defined depending on the 1 deposition process, the surface state, the texture, etc. The deposition of F film on AF does not change considerably the easy axis directions of AF grains. The easy axis of F film is well defined since it is grown under applied field. Therefore, this may be the reason for the misalignment observed in MnPt/NiFe exchange coupled bilayers.

IV. CONCLUSION

We observed a strong asymmetry in the exchange biased MnPt/NiFe systems and a shift of hard axis hysteresis loop from the origin. Our proposed model predicts the hard axis hysteresis loop shift and is coherent with our polycrystalline MnPt/NiFe exchange biased bilayers obtained by sputtering. An experimental method is proposed to measure the misalignment in noncollinear exchange biased systems. A misalignment angle of the order of 20° is experimentally observed in exchange biased MnPt/NiFe bilayers by measuring the in plane hard axis hysteresis loops at angles $\alpha=\pi/2$ or $\alpha=-\pi/2$ with respect to the F easy axis. The behavior of the misalignment as a function of antiferromagnetic layer thickness has the same features as that of H_e and H_c. Our approach is generalized for complex systems with high-order anisotropies.

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APPENDIX

In order to describe the asymmetry of hysteresis curves in some F-AF exchange coupled systems, we have considered in a previous work a rather general expression for the exchange energy.[14,15] This was justified by the complexity of magnetization reversal processes. The exchange bias is an interfacial phenomenon and therefore is strongly influenced by the physical state of the F/AF interface. The exchange energy local variations are due to the roughness, the interdiffusion, etc. and they depend strongly on the sample preparation method, F-AF material choice, etc. This is why the exchange energy for the jth AF grain should be, in general, an unknown function of the angle $\theta_j-\varphi$. It depends on the spin structure of the interface and on the exchange interactions among the moments in atomic scale. So in the most general case it should be developed into a Fourier series.[26] Then for the total energy the following expression can be written:

$$E = K_f S_f t_f \sin^2(\varphi) - \mu_0 H M S_f t_f \cos(\alpha-\varphi) - \sum_j S_j \left[\sum_{n=1}^{N} J_{jn} \cos\{n(\theta_j-\varphi)\} \right]. \tag{A1}$$

$N=1$ and 2 may be related to classical and biquadratic exchange energy terms.[32] The same way it can be shown that if we introduce $(J_1, J_2, J_3, \ldots J_N)$, $(\beta_1, \beta_2, \beta_3, \ldots \beta_N)$ and $(h_1=J_1/\mu_0 M t_f,\ h_2=J_2/\mu_0 M t_f,\ h_3=J_3/\mu_0 M t_f, \ldots, h_N=J_N/\mu_0 M t_f)$, then the reduced exchange energy can be presented by

$$E_r = \frac{H_f}{2}\sin^2(\varphi) - H\cos(\alpha-\varphi) - \sum_{n=1}^{N} h_n \cos[n(\beta_n-\varphi)] \tag{A2}$$

where

$$\tan(n\beta_n) = \frac{\sum_j J_{jn} S_j \sin(n\theta_j)}{\sum_j J_{jn} S_j \cos(n\theta_j)} \text{ and}$$

$$J_n S_f = \sqrt{\left[\sum_j J_{jn} S_j \sin(n\theta_j)\right]^2 + \left[\sum_j J_{jn} S_j \cos(n\theta_j)\right]^2}. \tag{A3}$$

To enhance the convergence, we considered in previous papers[14,15] a cosine power series instead of a Fourier series. Equation (A2) can be presented in the following form:

$$E_r = \frac{H_f}{2}\sin^2(\varphi) - H\cos(\alpha-\varphi) - \sum_{n=1}^{N} u_n \cos^n[\beta_n-\varphi]. \tag{A4}$$

Here u_n are unknown parameters. The physical meaning of u_n is less explicit than those of h_n, however u_n can always be related to h_n. For example, if the Fourier power series in Eq. (A4) is truncated at $N=5$, then $h_1=u_1+3u_3/4+5u_5/8$, $h_2=(u_2+u_4)/2$, $h_3=u_3/4+5u_5/16$, $h_4=u_4/8$, and $h_5=u_5/16$. This can be written also as $u_1=h_1-3h_3+5h_5$, $u_2=2h_2-8h_4$, $u_3=4h_3-20h_5$, $u_4=8h_4$, and $u_5=16h_5$.

In our previous work,[14,15] for simplicity, it was considered that $\beta_j=\beta(j=1,2,3,\ldots,N)$. Without any simplifying assumption, the exchange energy cosine power series expansion can be given as follows:

$$E_{\mathrm{ex}} = -\sum_{n=1}^{N} u_n \cos^n[\beta_n - \varphi]. \tag{A5}$$

As a measure of mean angular deviation of the F and AF easy axis, we consider the value of β_1 with

$$\tan(\beta_1) = \frac{\sum_j J_{j1} S_j \sin(\theta_j)}{\sum_j J_{j1} S_j \cos(\theta_j)}. \tag{A6}$$

β_1 as well as the other parameters $[\beta_j(j \neq 1)$, u_j, etc.] can be obtained by a nonlinear fit of experimental data to our model as described in our previous work.[14,15]

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