Irreversible and reversible measurements of exchange anisotropy

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Recently, several different experimental techniques have been used to measure the unidirectional exchange coupling, or exchange anisotropy, in ferromagnetic/antiferromagnetic bilayers. In particular, it has been found that reversible ac susceptibility measurements show significantly larger exchange anisotropy than the conventional irreversible hysteresis loop measurements. We have theoretically investigated the irreversible and reversible measurements of exchange anisotropy and our analysis shows that when the magnetic degrees of freedom of the antiferromagnet are taken into account, different measurement techniques may indeed give different results for the exchange coupling. [S0163-1829(99)02545-X]

The exchange-bias effect, which arises from the interfacial exchange coupling between a ferromagnet (FM) and an antiferromagnet (AF), was discovered more than 40 years ago.¹ It is so named because the phenomena manifests itself in a shifted hysteresis loop for the bilayer film. Recently, considerable interest in the FM/AF exchange coupling has been revived because of its application to giant magnetoresistive spin-valve heads for high-density recording systems.² However, a satisfactory understanding of this phenomena has not yet been developed. Initially, the exchange-bias effect was assumed³ to arise from the exchange coupling at an uncompensated interface between the FM and AF layers. This argument leads to an exchange field two orders of magnitude too large. Two alternative models, a random-field model by Malozemoff⁴ and a planar-domain-wall model by Mauri and co-workers,⁵ were proposed to explain this difference. This discrepancy between the theoretical predictions and experimental observations and even among the different models stimulated attempts to study this effect with measurement techniques other than the hysteresis loop measurement.

For hysteresis loop measurements, such as B-H looper measurements, vibrating sample measurements, and superconducting quantum interference device (SQUID) measurements, the unidirectional exchange anisotropy J_E is given by $H_e M_s t_F$, where the exchange field H_e is the displacement of the hysteresis loop, and M_s and t_F are the saturation magnetization and thickness of the FM film, respectively. Hysteresis loop measurements involve the irreversible switching of the magnetization of the FM film that could introduce complications of the magnetic structure of the FM films on the exchange-bias energy measurement. Other techniques in which the magnetization is only perturbed by a small amount have recently been employed. The first reversible measurement of the exchange anisotropy was based on the anisotropic magnetoresistance (AMR) of Co/CoO bilayers as a function of the angle between an in-plane applied magnetic field and the exchange-bias direction.⁶ The second reversible technique was the ac susceptibility measurement, in which only small rotations of the magnetization are involved in the presence of a small in-plane applied field.⁷ In the case of AMR measurements, the exchange anisotropy J_E is obtained indirectly by fitting the angular-dependent resistivity curve for the exchange field H_e . In the ac susceptibility measurement, J_E is obtained directly in a similar way as the susceptibility of an antiferromagnet. These two measurements found an exchange anisotropy energy several times larger than that obtained from hysteresis loop measurements.^{6,7} Two other reversible measurement techniques, ferromagnetic resonance (FMR) and Brillouin light scattering (BLS), have also been used to determine the exchange anisotropy of the Ni₈₀Fe₂₀/NiO and Fe/FeF₂ bilayers, respectively.^{8,9} The unidirectional exchange anisotropy values measured by in-plane FMR were about 20% less than the loop shift measured via magnetoresistance. The difference was explained by the hysteresis loop asymmetry and a "rotatable anisotropy" related to a domain configuration in the AF layer.⁸ The values obtained from BLS for Fe/FeF2 bilayers were 25% larger than those obtained from SQUID magnetometry and the difference here was explained by higher-order terms in the unidirectional anisotropy.⁹ Among these reversible measurements, the ac susceptibility measurement distinguishes itself by its large difference from the hysteresis loop result. Thus, an analysis of the ac susceptibility measurement and a comparison between the ac susceptibility measurement, hysteresis loop measurement, and FMR measurement are the focus of this paper.

The magnetization of an exchange-biased FM layer in an ac susceptometer with a small ac field applied at an angle θ with respect to the unidirectional axis induced by the exchange coupling is shown in Fig. 1. For a given value of the ac field $h_{\rm ac}$ the magnetic energy per unit area of an exchange-coupled FM can be written as follows:

$$E = K_u t_F \sin^2 \alpha - J_E \cos \alpha - h_{\rm ac} M_s t_F \cos(\theta - \alpha).$$
(1)

The first term is the uniaxial anisotropy of the ferromagnet. The easy axis of the exchange-biased FM layer is assumed to lie along the exchange coupling direction in our discussion. Such a *uniaxial anisotropy* was not taken into account in the analysis of Ström, Jönsson, and Dahlberg.⁷ The second term

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FIG. 1. Schematic representation of an exchange biased ferromagnetic layer in a presence of a small ac field h_{ac} .

is the exchange anisotropy whose value is the subject of these experimental measurements. The small applied ac field, given in the third term, causes the magnetization to deviate towards θ by a small angle α . The value of α can be found as the equilibrium point at which the derivative of the energy *E* with respect to α is equal to zero. The result is

$$\alpha = \frac{M_s h_{\rm ac} t_F \sin \theta}{J_E + 2K_u t_F}.$$
 (2)

The ac susceptibility is defined in terms of the oscillating component of the magnetization along the applied field. Hence, $\chi = \partial (M_s \cos(\theta - \alpha)) / \partial h_{\rm ac}$, which gives

$$\chi = \frac{(M_s \sin \theta)^2 t_F}{J_F + 2K_u t_F}.$$
(3)

Comparing this result to that of Ström, Jönsson, and Dahlberg,⁷ we see that there is still a sine-square dependence on the exchange-biasing direction θ , but the denominator now shows a combination of the interfacial exchange coupling J_E and the uniaxial anisotropy $2K_u t_F$ instead of just J_E . This difference cannot be ignored for Co/CoO bilayers, since for bulk Co, the uniaxial anisotropy¹⁰ at low temperature is about 5×10^6 ergs/cm³. The uniaxial anisotropy associated with the AF film. Using Eq. (3) with the measured value of χ gives a value for J_E , which is lower than the value of 2.06 ergs/cm² inferred by Ref. 7 by an amount depending upon the uniaxial anisotropy.

Although the incorporation of the ferromagnetic anisotropy may resolve the difference between J_E obtained from a loop measurement and the ac susceptibility measurement, this value is still much smaller than what would be predicted from the Meiklejohn-Bean model.¹ There are two concerns about using a hysteresis loop measurement to determine the exchange anisotropy. One has to do with knowing the magnetic structures of the FM and AF layers and the other has to do with the dynamics of the irreversible switching of the two layers.¹¹ The magnetization of a thin-soft ferromagnetic layer in a presence of an external field is generally a single domain. This may not be the case for exchange bias at the interface. However, a theoretical study has shown that there is no helical structure, for example, along the thickness di-



FIG. 2. Schematic representation of a ferromagnetic/ antiferromagnetic bilayer for the planar domain wall model. The moments of only one sublattice of the antiferromagnetic layer are shown.

rection in a thin ferromagnetic layer, e.g., $\mathrm{Ni}_{\mathrm{80}}\mathrm{Fe}_{\mathrm{20}}$ with thickness up to 500 Å, exchange coupled with a ferromagnetic, ferrimagnetic, or antiferromagnetic film at the interface.¹² Furthermore, Parkin and co-workers et al.¹³ have observed a uniform magnetization distribution throughout the thickness of a 400 Å Ni₈₀Fe₂₀ layer coupled with a Fe50Mn50 layer. The possibility of planar domains has also been ruled out in the Ni₈₀Fe₂₀ layer in the presence of a sufficiently large applied field. However, there might exist a complicated magnetic moment arrangement inside the AF layer due to the exchange coupling with the FM layer. Recently, we found in Ni₈₁Fe₁₉/Cr_{45.5}Mn_{45.5}Pt₉ bilayers that the shifted hysteresis loops of the Ni₈₁Fe₁₉ had the shapes consistent with the existence of a planar domain wall.¹⁴ This suggests that we should incorporate the possibility of an AF domain in the analysis of these measurement techniques for J_E . Figure 2 illustrates the existence of a β -degree domain wall in one of the sublattices of the AF layer. In the following discussion, we assume that the AF thickness is infinite and that the transition is reversible.¹⁵ The total magnetic energy per unit area is^{5,16}

$$E = K_u t_F \sin^2 \alpha - J_E \cos(\alpha - \beta) + \sigma_W (1 - \cos \beta) - h_{ac} M_s t_F \cos(\theta - \alpha), \qquad (4)$$

where α is the angle of the FM magnetization with respect to the easy axis of the FM layer, β the angle of the AF moment at the interface with respect to the easy axis z of the AF layer, and θ the direction of the ac applied field. Here we again assume that the FM and AF layers have the same easy axis. For a given value of the small ac field $h_{\rm ac}$, the equilibrium points of α and β can be from

$$\frac{\partial E}{\partial \alpha} = K_u t_F \sin 2\alpha + J_E \sin(\alpha - \beta) - h_{\rm ac} M_s t_F \sin(\theta - \alpha) = 0,$$
$$\frac{\partial E}{\partial E}$$

$$\frac{\partial L}{\partial \beta} = -J_E \sin(\alpha - \beta) + \sigma_W \sin \beta = 0.$$
 (5)

Because the angles α and β are very small, sin $\alpha \approx \alpha$, and a direct relationship between α and h_{dc} can be obtained by eliminating β from Eq. (5). Then following the same procedure in deriving Eq. (3), the ac susceptibility is given as

$$\chi = \frac{(M_s \sin \theta)^2 t_F}{\frac{J_E \sigma_W}{J_E + \sigma_W} + 2K_u t_F}.$$
(6)

Notice that the exchange anisotropy J_E is "renormalized" by the domain-wall energy σ_W . Now the exchange anisotropy J_{ac} obtained from ac susceptibility measurements is $J_E \sigma_W / (J_E + \sigma_W)$. Mauri and co-workers⁵ have previously pointed out that the formation of a planar domain wall in the AF layer also limits the exchange field H_e obtained from a hysteresis loop measurement, no matter how large the interfacial exchange coupling. Therefore, the measured exchange anisotropy values from either irreversible or reversible measurements are combinations of the FM/AF interfacial exchange coupling and the AF domain-wall energy.

For the hysteresis loop measurements, the exchange field H_e can be determined from the numerical calculations based on Eq. (4) by a Stoner-Wohlfarth approach described in the paper of Mauri *et al.*⁵ In the simple case that the uniaxial anisotropy of the FM layer is zero, the FM magnetization rotates with no hysteresis in the presence of an applied field along the FM easy axis. In this case we can obtain an analytic expression for J_{hl} the effective exchange anisotropy governing the shift of the hysteresis loop. The result is

$$J_{hl} = \frac{J_E \sigma_W}{\sqrt{J_F^2 + \sigma_W^2}}.$$
(7)

Our analysis of the exchange anisotropy is based on the assumption that the magnetization rotation in the presence of an external field, whether irreversible or reversible, is a quasiequilibrium process in which the magnetization follows the applied field.

Theoretical calculations for the FMR frequency may also be developed based on the planar-domain-wall model. The total energy for the exchange-coupled FM layer is written in a three-dimensional form with the magnetostatic shape anisotropy energy $2\pi M_s^2 t_F$ included. The exchange coupling and the planar-domain-wall motion are restricted to the film plane in our calculations. The resonance frequencies in the presence of the domain wall are (a) for $J_E > \sigma_W$

$$\begin{split} \left(\frac{\omega}{\gamma}\right)^2 = & (H_a + H_u + 4 \pi M_s + H_E) \\ & \times \left(H_a + H_u + \frac{H_E H_W}{H_E + H_W}\right) \quad (\varphi_H = 0), \end{split}$$

$$\begin{split} \left(\frac{\omega}{\gamma}\right)^2 &= (H_a + H_u + 4 \,\pi M_s + H_E) \\ &\times \left(H_a + H_u - \frac{H_E H_W}{H_E - H_W}\right) \quad (\varphi_H = \pi), \end{split}$$

and (b) for $J_E < \sigma_W$



FIG. 3. Exchange anisotropy $J_{\rm ac}$ obtained from ac susceptibility measurement, J_{nl} from hysteresis loop measurement, and $J_{\rm FMR}$ from ferromagnetic resonance measurement. The results are for infinite AF thickness.

$$\left(\frac{\omega}{\gamma}\right)^{2} = (H_{a} + H_{u} + 4\pi M_{s} + H_{E})$$

$$\times \left(H_{a} + H_{u} + \frac{H_{E}H_{W}}{H_{E} + H_{W}}\right) \quad (\varphi_{H} = 0),$$

$$\left(\frac{\omega}{\gamma}\right)^{2} = (H_{a} + H_{u} + 4\pi M_{s} - H_{E})$$

$$\times \left(H_{a} + H_{u} + \frac{H_{E}H_{W}}{H_{E} - H_{W}}\right) \quad (\varphi_{H} = \pi). \quad (8)$$

In the above equations, the angle φ_H shows the direction of the applied field referred to the exchange coupling direction. The effective field parameters represent the anisotropy energy terms, i.e., $H_u = 2K_u/M_s$, $H_E = J_E/M_s t_F$, and H_W $= \sigma_W/M_s$. The exchange field H_{FMR} is obtained from the FMR measurement as half of the difference in the resonance field^{8,17} with the applied field of magnitude opposite to the exchange anisotropy direction. The shape anisotropy $2\pi M_s^2 t_F$ is orders larger than $K_u t_F$, J_E , and σ_W . Assuming $4\pi M_s$ is much larger than the resonance field, the exchange anisotropy J_{FMR} obtained by FMR as $H_{\text{FMR}} M_s t_F$ is

$$J_{\rm FMR} = \begin{cases} \sigma_W \frac{J_E^2}{J_E^2 - \sigma_W^2} & (J_E > \sigma_W) \\ J_E \frac{\sigma_W^2}{\sigma_W^2 - J_E^2} & (J_E < \sigma_W). \end{cases}$$
(9)

The resulting J_{ac} , J_{hl} , and J_{FMR} are shown in Fig. 3 with the interfacial exchange coupling J_E normalized to σ_W . We see that if a domain wall forms in the AF layer, the values from the three kinds of measurements will be different. Both J_{ac} and J_{hl} are smaller than the interface exchange coupling J_E and domain-wall energy σ_W , while J_{FMR} is larger than J_E or σ_W . The formation of the AF planar domain wall "loosens" the pinning of the FM magnetization along the exchange-bias direction and makes its rotation easier in a small applied field. This makes J_{ac} smaller than J_{hl} . When $J_E = \sigma_W$, the domain wall is no longer stable and results in the divergence of J_{FMR} . In this case, the assumption of reversible behavior of the AF cannot be applied. In the extreme cases, i.e., $J_E \ll \sigma_W$ and $J_E \gg \sigma_W$, the measured exchange anisotropy J_{ac} , J_{hl} , and J_{FMR} approach J_E and σ_W , respectively, and the difference between them goes to zero.

Experimental data for Ni₈₀Fe₂₀/NiO (Ref. 8) and Ni₈₁Fe₁₉/Pt₁₀Mn₉₀ (Ref. 18) show that $J_{FMR} < J_{hl}$, whereas our result shows that J_{FMR} should always be greater that J_{hl} . One reason for this difference may be due to the fact that the model presented in this paper does not allow for irreversible motion of the AF. Nor does it explicitly take into account the

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granular nature of the films.¹⁹ In particular, if J_E is larger than σ_W , the uncompensated moments of the AF will be pulled along with the FM magnetization. If the resulting twist in the AF structure becomes too large, the AF irreversibly jumps to a new angle.²⁰ The experimental results therefore represent an averaging over grains with different values of J_E and σ_W . For comparison with the results of this paper, measurements should be carried out on a single crystal bilayer.

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