Angular dependence of exchange coupling in ferromagnet/antiferromagnet bilayers

T. Ambrose, R. L. Sommer, and C. L. Chien
Department of Physics and Astronomy, The Johns Hopkins University, Baltimore, Maryland 21218
(Received 28 March 1997)

Angular dependence of the exchange coupling in a NiFe/CoO bilayer film has been measured. Different angular dependence and symmetry have been observed for the exchange field H_E and the coercivity H_C. The values of H_E and H_C are due to unidirectional and uniaxial components of the magnetic anisotropy energy, in which the odd and the even terms lead to H_E and H_C, respectively. The proposed free energy accounts for the experimental results. [S0163-1829(97)04226-4]

A ferromagnetic (FM) material generally displays a symmetric hysteresis loop centered at zero magnetic field ($H=0$) as demanded by time-reversal symmetry.[1] As first discovered by Meiklejohn and Bean, the hysteresis loops of slightly oxidized Co particles are distinctively displaced from $H=0$.[2,3] They showed that this unique behavior is caused by an exchange coupling between the surface antiferromagnetic (AF) Co oxides and the FM Co core.[2,3] This exchange coupling can be better revealed in a much more controlled manner in FM/AF bilayers (e.g., NiFe/FeMn, NiFe/NiO).[4–7] When an FM/AF bilayer is cooled in a magnetic field from temperatures above the Néel temperature T_N of the AF to $T<T_N$, an exchange coupling is locked in. The hysteresis loop is now shifted from $H=0$ by an amount termed the exchange field H_E, which can be as much as 1 kOe.[8] The shifted hysteresis loop is also accompanied by a substantial coercivity H_C, which is much larger than the intrinsic value of the FM layer.[4–10] The AF layer plays the essential role of providing the exchange coupling, which disappears at $T \geqslant T_N$, and the FM layer facilitates the observation of the resultant exchange coupling through the shifted hysteresis loop.

The phenomenon of exchange coupling at the FM/AF interface is very rich indeed. For a given FM/AF system, not only the values of H_E and H_C depend on temperature in their distinctive ways, they also depend on the thicknesses t_{FM} and t_{AF} of the FM and the AF layers.[4–10] For example, H_E has been found to be proportional to $1/t_{\mathrm{FM}}$, a signature consistent with the notion that the exchange coupling is probably an interfacial effect.[4] The robust exchange coupling has been exploited for manipulating the spin structures in FM/AF multilayers.[10] Technologically, the FM/AF exchange coupling is also of prime importance; it is at the heart of the spin-valve magnetoresistance devices.[11]

Yet, despite extensive investigations and technological importance, the nature of the exchange coupling in FM/AF bilayers remains poorly understood. Meiklejohn and Bean have originally suggested an *ad hoc* anisotropy energy term of $K_E \cos\phi$, which would account for the shifted hysteresis loops, where $K_E = H_E / M_F$ and ϕ is the angle between the direction of the FM magnetization M_F and the exchange anisotropy axis.[3] However, to date, the actual spin arrangement among the magnetic constituents (directions of the FM magnetization and the sublattice magnetizations of the AF) remains unknown. It has been assumed in most theoretical models and experimental analyses that at the FM/AF interface the first atomic layer of the AF is ferromagnetically aligned *parallel* to the FM layer, and the subsequent atomic layers of the AF are ferromagnetically ordered with alternating spin directions.[3,12–14] However, the validity of this simple model has been questioned. Among other difficulties, the expected exchange field for such a spin structure is about two orders of magnitude larger than the experimentally observed values. Recently, using micromagnetics calculations, Koon shows that the interfacial AF layer has a compensated AF structure with spin directions nearly *perpendicular* to the anisotropy axis,[15] reminiscent of the spin-flop transition in a uniaxial AF.[16] The small net magnetization of the canted sublattice magnetizations would alleviate the discrepancy in the values of the exchange field. Such a spin structure has also been suggested by some recent experimental studies.[17] In addition to the unsettled spin structure, many other aspects, including the relevant Hamiltonian for the exchange coupling, the angular dependence of H_E, and the large enhancement of H_C, have not been addressed. Theoretical models for exchange coupling thus far have attempted to account only for H_E and ignoring H_C completely.[12–14]

Since the exchange coupling at the FM/AF interface depends on the various directions of the constituent magnetic layers, revealing the angular dependence of the exchange coupling is of crucial importance in unraveling the nature of the exchange coupling. In this work, we have measured the angular dependence of H_E and H_C of exchange-coupled FM/AF bilayers. We show that the exchange field H_E is *unidirectional*, whereas the coercivity H_C is *uniaxial*, and more importantly, both H_E and H_C are integral parts of the exchange coupling, in which the *odd* and *even* terms of the total anisotropy energy lead to H_E and H_C, respectively. The commonly held belief of a simple cosine term for the exchange coupling has been demonstrated to be inadequate. We also propose a model-independent free energy, which accounts for the experimental results.

For this study, permalloy ($\mathrm{NiFe}=\mathrm{Ni_{81}Fe_{19}}$) with a small intrinsic H_C was chosen as the FM layer and CoO with a $T_N=292$ K was used as the AF layer. In bulk CoO, as has been shown by neutron diffraction, the moments align ferromagnetically in each (111) plane and alternate along the [111] direction.[18,19] A bilayer film consisting of 300 Å of NiFe on 100 Å of CoO have been grown on a 300-Å buffer layer of Cu on Si(100) by magnetron sputtering. The Cu

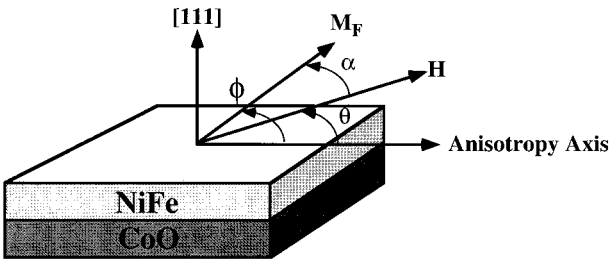

FIG. 1. Structure of an exchange-coupled (111) NiFe(300 Å)/CoO(100 Å) bilayer. The anisotropy axis denotes the direction of the field cool and that of the dc magnetic field during deposition. By physically rotating the sample about an axis perpendicular to the film plane, the magnetic field (**H**) is in the film plane and at an angle θ with respect to the anisotropy axis. The magnetization $\mathbf{M}_F$ of the ferromagnet is at an angle α with the magnetic field, and at an angle $\phi=\alpha+\theta$ with the anisotropy axis.

buffer layer provides a clean starting surface and promotes growth of oriented [111] CoO in the bilayer. For comparison, a single-layer film of 300 Å of NiFe was also deposited onto 300 Å Cu on Si(100). In both cases, the NiFe layer was deposited in a dc field of approximately 200 Oe, applied in the plane of the film, to induce a uniaxial anisotropy.

The bilayer sample was first field cooled from 320 to 80 K in a 10-kOe field, applied in the same direction as that of the deposition field, in order to induce the exchange coupling. This unique direction (field cooled and deposition field) is the anisotropy axis, denoted as $\theta=0$. Hysteresis loops were measured at 80 K in a vibrating sample magnetometer by physically rotating the sample about an axis perpendicular to the sample plane so that the applied field (**H**) is in the sample plane and at various angles θ with respect to the anisotropy axis, as shown in Fig. 1. It may be noted that most previous measurements of the exchange coupling in FM/AF bilayers have utilized only the measuring geometry of either $\theta=0$ or $\theta=\pi$.[4–10]

Representative hysteresis loops of NiFe/CoO bilayer with different angle θ (indicated in the upper left-hand corner in each case) are shown in Fig. 2. To quantify the exchange coupling in each case, the exchange field H_E and the coercivity H_C are defined in terms of the two field values H_{left} and H_{right} at which $M=0$: $H_E=-(H_{\text{right}}+H_{\text{left}})/2$ and $H_C=(H_{\text{right}}-H_{\text{left}})/2$. In this convention, the hysteresis loop shifted to negative H values has a positive H_E value. As shown in Fig. 2, large exchange coupling occurs near $\theta=0$ and π, whereas the smallest ($H_E=0$) occurs at $\theta=\pi/2$and $3\pi/2$. In Fig. 2, the results for various θ values are arranged to illustrate the underlying symmetry. The two columns exhibit complementary hysteresis curves that differ in θ by π. It is clear that under the operation of $+M\rightarrow-M$ and $+H\rightarrow-H$, the results in one column yield the results in the other column. One further notes that the results with one value of θ is the same as that with $-\theta$, or $2\pi-\theta$, as shown in Fig. 2 (e.g., $\pi/4$ and $7\pi/4$, $3\pi/4$, and $5\pi/4$).

The angular dependence of H_E and H_C as a function of θ are shown in Fig. 3(a) and 3(b), which contain the most essential features of the exchange coupling. It is evident that the basic symmetry properties, shared by both H_E and H_C, are $H_E(2\pi+\theta)=H_E(\theta)=H_E(-\theta)$ and $H_C(2\pi+\theta)$

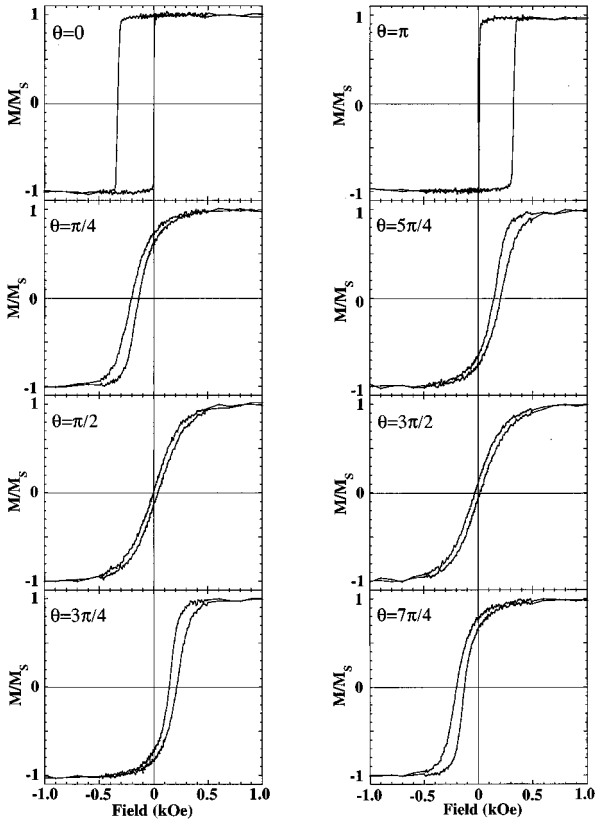

FIG. 2. Hysteresis loops of an exchange-coupled (111) NiFe(300 Å)/CoO(100 Å) bilayer at various angle θ (referred to Fig. 1).

$=H_C(\theta)=H_C(-\theta)$. Thus, the most general form for either H_E or H_C is $\Sigma b_n\cos n\theta$, where n is a positive integer, and b_n is a constant. However, H_E and H_C separately possesses *additional* symmetry of $H_E(\pi+\theta)=-H_E(\theta)$ and $H_C(\pi+\theta)=H_C(\theta)$. In the case of H_E, the symmetry of $H_E(\pi+\theta)=-H_E(\theta)$ dictates that the most general form of $H_E(\theta)$ contains only the odd n terms,

$$H_E(\theta)=\sum_{n=\text{odd}} b_n\cos n\theta, \tag{1}$$

which conclusively demonstrates that the exchange field H_E is *unidirectional*. The results in Fig. 3(a) can be adequately described by $H_E(\theta)=206$ Oe $[\cos\theta-0.21\cos3\theta+0.01\cos5\theta+\cdots]$ shown by the solid curve. It has been widely believed that the largest values of H_E are expected at $\theta=0$ and π, which would be the case if H_E should have a simple $\cos\theta$ dependence. Instead, as shown in Fig. 3(a), the largest values of H_E are located near $\theta\approx\pm\pi/4$ and $\pi\pm\pi/4$, due to the fact that H_E is described by Eq. (1) and not just a simple cosine term.

Turning now to H_C, the additional symmetry is $H_C(\pi+\theta)=H_C(\theta)$, which by the same token leads to the most general form for H_C of having only the even n terms

$$H_C(\theta)=\sum_{n=\text{even}} b_n\cos n\theta, \tag{2}$$

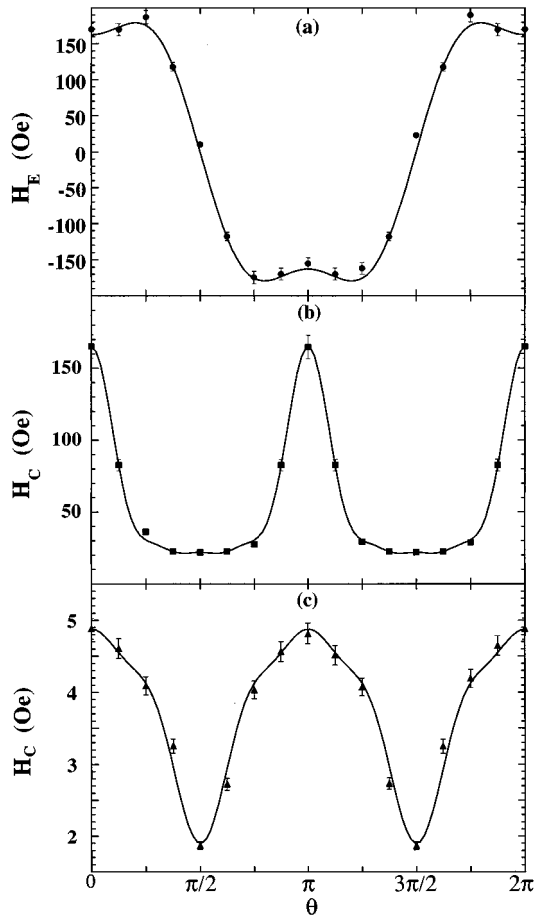

FIG. 3. Angular dependence of (a) exchange field H_E and (b) coercivity H_C of an exchange-coupled (111) NiFe(300 Å)/CoO(100 Å) bilayer. Shown in (c) is the angular dependence of H_C for a single layer NiFe film.

which shows that H_C is *uniaxial*. The solid curve in Fig. 3(b) is $H_C(\theta)=57$ Oe $[1+0.98\cos2\theta+0.54\cos4\theta+0.23\cos6\theta+\cdots]$. These analyses clearly show that to account for the exchange coupling, anisotropy terms beyond $\cos\theta$ must be included.

Before further discussion, it is useful to compare the results of the exchange-coupled NiFe/CoO bilayer with those of single NiFe layer also measured at 80 K. The hysteresis loops of a single NiFe layer are of course completely symmetrical (i.e., $H_E=0$). The angular dependence of its coercivity is shown in Fig. 3(c). First of all, the value of H_C for the exchange-coupled NiFe [Fig. 3(b)] is *not* those of a single uncoupled NiFe layer [Fig. 3(c)]; the former is as much as 30 times larger. Second, while the results shown in Fig. 3(c) contain the same general symmetry of $H_C(\theta)$ described by Eq. (2), the actual angular dependence of $H_C(\theta)=3.7$ Oe $(1+0.35\cos2\theta-0.11\cos4\theta+0.05\cos6\theta+\cdots]$, shown as the solid curve in Fig. 3(c), is very much different from that shown in Fig. 3(b). This comparison underscores the fact that in exchange-coupled FM/AF bilayers, both H_E *and* H_C are the consequences of the exchange coupling. Any theoretical model of exchange coupling must address not only H_E but H_C as well.

In the following, using symmetry considerations, we provide a model-independent description of the exchange coupling that accounts for all the experimental results. Regardless of the actual spin structure of the FM and the AF in the exchange coupling, which remains unknown at present, the anisotropy axis (the field-cooled direction for the AF and the uniaxial anisotropy axis of the FM) is uniquely defined in the exchange coupling in an FM/AF bilayer. At the FM/AF interface, the sublattice magnetizations of the AF has a net magnetization $\Delta\mathbf{M}$ along the anisotropy axis, irrespective of the actual spin structure. For example, in the simple two-sublattice case, $\Delta\mathbf{M}=\mathbf{M}_1+\mathbf{M}_2$ is along the anisotropy axis, and $|\Delta\mathbf{M}|\ll|\mathbf{M}_1|,|\mathbf{M}_2|$, regardless of whether $\mathbf{M}_1$ and $\mathbf{M}_2$ are parallel or nearly perpendicular to the anisotropy axis. Once the FM/AF bilayer has been field-cooled across T_N to low temperatures, the relatively small external field $\mathbf{H}$ cannot alter the AF ordering. Thus, the relevant free energy with $\mathbf{H}$ in the layer plane is

$$F=-U(\phi)-\mathbf{M}_F\cdot\mathbf{H}=-U(\phi)-M_FH\cos\alpha, \qquad (3)$$

where the angles have been defined in Fig. 1, in which α is the angle between the magnetization $\mathbf{M}_F$ of the FM and the external field $\mathbf{H}$, θ is the angle between $\mathbf{H}$ and the anisotropy axis, and $\phi=\alpha+\theta$ is the angle between $\mathbf{M}_F$ and the anisotropy axis. In Eq. (3), the first term $U(\phi)$ is the all important magnetic anisotropy energy for exchange coupling, and the second term is the Zeeman term.

The detailed form of $U(\phi)$ would depend on the actual spin structure of the constituent layers and their interactions. However, from symmetry considerations, the anisotropy energy $U(\phi)$ must satisfy the general symmetry of $U(2\pi+\phi)=U(\phi)$ and $U(\phi)=U(-\phi)$. Thus, the most general form is $U(\phi)=\Sigma a_n\cos n\phi$, where n is a positive integer. Without loss of generality, the anisotropy energy $U(\phi)$ can be partitioned into odd-n terms and the even-n terms, which are, respectively, the unidirectional energy $UD(\phi)=a_1\cos\phi+a_3\cos3\phi+\cdots$ and the uniaxial energy $UA(\alpha)=a_2\cos2\phi+a_4\cos4\phi+\cdots$, with the inherent symmetry of $UD(\pi+\phi)=-UD(\phi)$ and $UA(\pi+\phi)=UA(\phi)$. It is useful to examine first the simplest case of $F=-a_1\cos(\alpha+\theta)-M_FH\cos\alpha$ including only the $n=1$ term of $U(\phi)$. In this case, there is no coercivity nor the complexity associated with irreversibility. Minimizing F readily leads to the solution of a reversible hysteresis loop with zero H_C and $H_E(\theta)=(1/M_F)a_1\cos\theta$. It is straightforward to show that if only the odd-n terms of $U(\phi)$ are included, i.e., the unidirectional $UD(\phi)=a_1\cos\phi+a_3\cos3\phi+\cdots$, there is still no coercivity and the exchange field is

$$H_E(\theta)=\frac{1}{M_F}[a_1\cos\theta-3a_3\cos3\theta+5a_5\cos5\theta-7a_7\cos7\theta+\cdots]. \qquad (4)$$

One recognizes that this is precisely the experimentally observed angular dependence of Eq. (1).

Hysteresis loops with coercivity are realized when even-n terms of $U(\phi)$ are included. The simplest of the even-n terms is the $n=2$ term, with which we have $F=-a_2\cos2(\alpha+\theta)-M_FH\cos\alpha$. This expression of free energy is the same as that in the Stoner-Wohlfarth model for single-domain particles with uniaxial anisotropy leading to hysteresis loops with coercivity.[20] It is well known that the uniaxial

anisotropy, described by the even-n terms, introduces irreversibility and hysteresis loops with coercivity. As such, no closed form solutions are generally available, and one resorts to the Preisach method and other methods to obtain the hysteresis loops numerically.[21] However, the angular dependence of H_C of FM/AF exchange coupling must involve the even-n terms as described in Eq. (2). As made clear in these analyses, both H_E and H_C are both consequences of the anisotropy energy $U(\phi)$ for the exchange coupling.

The above analyses using the free energy of Eq. (3) and a model-independent anisotropy energy of $U(\phi) = \Sigma a_n \cos n\phi$ accounts for the observed angular dependence of the exchange field and the coercivity. The coefficients a_n depend on the actual spin structure of the magnetic constituent layers and the various $\cos n\phi$ terms are related to the microscopic interactions among the moments. For example, assuming a simple AF with two sublattice magnetizations $\mathbf{M}_1$ and $\mathbf{M}_2$, with $\Delta\mathbf{M}=\mathbf{M}_1+\mathbf{M}_2$ along the anisotropy axis, the interaction of the form $(\mathbf{M}_1+\mathbf{M}_2)\cdot\mathbf{M}_F$ leads to the $\cos\phi$ term, $(\mathbf{M}_1\cdot\mathbf{M}_F)^2+(\mathbf{M}_2\cdot\mathbf{M}_F)^2$ leads to the $\cos 2\phi$ term, and $(\mathbf{M}_1\cdot\mathbf{M}_F)^3+(\mathbf{M}_2\cdot\mathbf{M}_F)^3$ leads to the $\cos 3\phi$ and the $\cos\phi$ terms, etc. The original model of Meiklejohn and Bean[3] amounts to including only the $(\mathbf{M}_1+\mathbf{M}_2)\cdot\mathbf{M}_F$ term, which is inadequate for the observed angular dependence.

In summary, we have determined the angular dependence of the exchange coupling in FM/AF bilayer films. Both the exchange field H_E and the coercivity H_C, with unidirectional and uniaxial characteristics, respectively, are integral parts of the exchange coupling. The proposed free energy, which is independent of the detailed spin structures and microscopic interactions, accounts for all the experimental results. The values of H_E and H_C, expressed by a cosine series with odd and even terms, respectively, are due to unidirectional and uniaxial parts of the anisotropy energy. The experimental results cannot be reproduced by a simple cosine term as originally suggested.

This work was supported by ONR Grant No. N00014-91-J-1633 and NSF MRSEC Program No. DMR-96-32526.

[1] See, e.g., A. H. Morrish, *The Physical Principles of Magnetism* (Wiley, New York, 1965), p. 333.
[2] W. H. Meiklejohn and C. P. Bean, Phys. Rev. **102**, 1413 (1956).
[3] W. H. Meiklejohn and C. P. Bean, Phys. Rev. **105**, 904 (1957).
[4] C. Tsang, N. Heiman, and K. Lee, J. Appl. Phys. **52**, 2471 (1981).
[5] C. Tsang and K. Lee, J. Appl. Phys. **53**, 2606 (1982).
[6] M. J. Carey and A. E. Berkowitz, Appl. Phys. Lett. **60**, 3061 (1992).
[7] R. Jungblut, R. Coehoorn, M. T. Johnson, J. aan de Stegge, and A. Reinders, J. Appl. Phys. **75**, 6659 (1994).
[8] T. Ambrose and C. L. Chien (unpublished).
[9] O. Allegranza and M. Chen, J. Appl. Phys. **73**, 6218 (1993).
[10] T. Ambrose and C. L. Chien, Appl. Phys. Lett. **65**, 1967 (1994).
[11] B. Dieny, V. S. Speriosu, S. S. P. Parkin, B. A. Gurney, D. R. Wilhoit, and D. Mauri, Phys. Rev. B **43**, 1297 (1991).
[12] A. P. Malozemoff, Phys. Rev. B **35**, 3679 (1987).
[13] D. Mauri, H. C. Siegmann, P. S. Bagus, and E. Kay, J. Appl. Phys. **62**, 3047 (1987).
[14] A. P. Malozemoff, Phys. Rev. B **37**, 7673 (1988).
[15] N. Koon, J. Appl. Phys. **81,** 4982 (1997).
[16] See, e.g., R. M. White, *Quantum Theory of Magnetism* (Springer-Verlag, Berlin, 1983), p. 114.
[17] T. J. Moran and I. K. Schuller, J. Appl. Phys. **79**, 5109 (1996).
[18] C. G. Shull, W. A. Strauser, and E. O. Wollan, Phys. Rev. **83**, 333 (1951).
[19] D. Herrmann-Ronzaud, P. Burlet, and J. Rossat-Mignod, J. Phys. C **11**, 2123 (1978).
[20] E. C. Stoner and E. P. Wohlfarth, Trans. R. Soc. London **240**, 599 (1948).
[21] See, e.g., I. D. Mayergoyz, *Mathematical Models of Hysteresis* (Springer-Verlag, Berlin, 1991).