Long-Range Exchange Bias across a Spacer Layer

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Ferromagnet (FM)/antiferromagnet (AF) exchange bias, heretofore considered due to nearest-neighbor exchange coupling at the FM/AF interface, has been found to be long range. Using trilayers of FM/spacer/AF with Ag, Au, and Cu as spacer layer materials, the strength of the observed exchange coupling decays exponentially and extends to as much as 50 Å. [S0031-9007(97)04594-8]

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Among the most important advances in magnetic layers in recent years is the discovery of coupling between magnetic layers. Interlayer coupling was first observed between Fe layers across Cr [1], and subsequently in multilayers with various ferromagnetic and spacer layers [2,3]. The interlayer coupling can be either ferromagnetic (FM) or antiferromagnetic (AF) depending on the thickness of the spacer layer. Thus, as the thickness of the spacer layer is varied, one often observes an oscillatory behavior of the interlayer coupling, which alternates between FM and AF, extending to more than 100 Å [4]. Various theoretical models have been proposed to account for the phenomenon [5]. The prevailing idea is that the interlayer coupling is RKKY-like and that the oscillations are determined by the extremal spanning vectors of the Fermi surface of the spacer material.

Another type of coupling exists between FM and AF materials [6], unlike the interlayer coupling between two FM materials mentioned above. When an FM/AF bilayer is field cooled below the Néel temperature T_N of the AF layer, the FM/AF exchange coupling is established [7–10]. The hysteresis loop of the FM layer, instead of being centered at zero magnetic field, is now displaced from $H = 0$ by an amount noted as the exchange field H_E, as if the FM layer is under a biased magnetic field. Hence, this phenomenon is also known as exchange bias. Technologically, exchange bias is of crucial importance in spin-valve field-sensing devices [11]. However the physics of FM/AF exchange coupling, including the Hamiltonian of the coupling, remains poorly understood. The temperature dependence of H_E, as well as the dependence upon the thickness of the FM and AF layers, has not been satisfactorily accounted for [7–10]. Even the spin arrangement of the FM and AF layers, upon which all microscopic models of coupling rely, remains controversial. Most models have assumed that the first magnetic layer of the AF orders ferromagnetically and parallel to the magnetization of the FM [6,12,13]. This assertion has been contradicted by recent micromagnetic [14] and experimental [15] studies.

A key issue, central to the physics of exchange bias, is the nature of the coupling. All experimental studies of FM/AF exchange coupling have been conducted where the FM layer is always in direct contact with the AF layer. Furthermore, all theoretical models for FM/AF exchange coupling to date have assumed a nearest-neighbor coupling at the interface. In these models, the exchange field is the result of coupling between the first magnetic layers of the two materials. For example, one well-known expression for the magnitude of the exchange field is [12]

$$H_E = n|J|S_F S_{AF}/t_F M_F\,, \tag{1}$$

where S_F and S_{AF} are the spins of the magnetic moments, respectively, in the FM and the AF materials at the interface, t_F and M_F are, respectively, the thickness and magnetization of the FM layer, J is the spin-spin interaction strength between S_F and S_{AF}, and n is the number of nearest-neighbor interactions per unit area at the FM/AF interface. One long-standing difficulty of Eq. (1) is that the value of J is too large to account for the observed value of H_E [12,13].

In this Letter, we have probed the assertion that exchange bias is a nearest-neighbor exchange coupling by purposely inserting a nonmagnetic spacer layer between the FM and the AF layers. The observation of FM/AF exchange coupling across a nonmagnetic layer demonstrates that the exchange bias is a long-range interaction extending to several tens of Å. This coupling is not oscillatory but decays exponentially. The range of FM/AF exchange coupling is specific to the spacer material, and thus most likely electronic in nature.

Noble metal (Cu, Au, and Ag) spacer layers of varying thickness were inserted between FM permalloy (Py = $Ni_{81}Fe_{19}$) and AF CoO. The multilayer structure of Cu/Py/noble metal/CoO/Cu/Si was fabricated in a magnetron sputtering system, which can accommodate layers with a uniform thickness and wedge-shaped layers. A copper layer of 100 Å was used as an underlayer to promote the growth of [111] oriented CoO and as a capping layer for protective purposes. All layers had uniform thickness except the spacer layer, which was wedged. The CoO layer, nominally 300 Å, was deposited by rf sputtering from a CoO target. A thickness of 300 Å of CoO was chosen for all samples, because this is well above the thickness for appreciable finite size effects [16]. The Py layer, approximately 300 Å thick, was deposited by dc sputtering in a magnetic field to induce in-plane uniaxial anisotropy. The multilayer structure and the wedge nature of the spacer layer were confirmed by small-angle x-ray diffraction and by cross-sectional transmission electron microscopy (XTEM) [17]. For the magnetic measurements, the layer structure was diced along the wedge direction yielding many samples in which the thickness of the spacer layer was the only variable.

Magnetic hysteresis loops were measured using a vibrating sample magnetometer. Before measurement, each sample was first field cooled in a 10 kOe field from 300 K, which is above $T_N \approx 290$ K of CoO, to 80 K with the magnetic field parallel to the easy axis of the Py. The field-cool direction established the anisotropy axis of the FM/AF exchange bias. Representative hysteresis loops at 80 K for the 300 Å Py/Au/300 Å CoO samples with various Au thicknesses t_{Au} are shown in Fig. 1. It is evident that these hysteresis loops are shifted from $H = 0$, a telltale sign of exchange coupling, which persists for t_{Au} in excess of 30 Å. Exchange coupling also causes a much increased coercivity (H_C), which is defined as one half of the loop width at $M = 0$. As shown in Fig. 1, the values of H_E and H_C decrease monotonically for increasing values of t_{Au}. Only at $t_{\mathrm{Au}} \geq 36$ Å have we observed $H_E = 0$ and $H_C = 2$ Oe, the characteristics of Py with no exchange coupling. These results are repeatable with a minimal "training" effect, which in the present case, amounts to a slight decrease in the exchange field of 2% to 4% [18]. Similar results have been obtained in samples with Cu and Ag spacer layers, but the thickness ranges that show exchange coupling are different as described below. These shifted hysteresis loops clearly demonstrate exchange bias in structures, in which the AF and FM layers are *separated* by a spacer layer. They provide the first evidence that the FM/AF exchange bias, which has always been assumed to be a nearest-neighbor coupling, can in fact be a long-range interaction.

The temperature dependence of H_E and H_C of the Py/spacer/CoO trilayers is similar to those of the Py/CoO bilayers [17]. The exchange field H_E is nearly constant at low temperatures but decreases as T_N of CoO is approached, and vanishes above T_N. The coercivity H_C decreases steadily with increasing temperature and retains the value of about 2 Oe at $T \geq T_N$, the value for uncoupled Py [10].

To further establish exchange coupling across a spacer layer, the underlying symmetry of the exchange coupling must be established, beyond the shifted loops shown in Fig. 1. It has recently been shown in exchange-coupled FM/AF bilayers that the exchange field H_E and the coercivity H_C have specific symmetry where the unidirectional and the uniaxial parts of the FM/AF exchange coupling anisotropy energy give rise to H_E and H_C, respectively [19]. For such angular dependence measurements, we first field cooled each FM/spacer/AF sample to 80 K, with the field along the anisotropy axis. Then hysteresis loops were measured at various angles θ with respect to the anisotropy axis, by physically rotating the sample about an axis perpendicular to the sample plane so that the applied field H remains in the sample plane. In terms of angular dependence, the hysteresis loops shown in Fig. 1 correspond to the case with $\theta = 0$. The angular dependence of the measured values of H_E and H_C are shown in Fig. 2 for representative samples of Py/Au/CoO with $t_{\mathrm{Au}} = 7.3$ and 13.5 Å. The exchange field indeed exhibits the unidirectional symmetry $[H_E(\theta) = H_E(-\theta) = -H_E(\pi \pm \theta)]$, whereas the coercivity shows the uniaxial

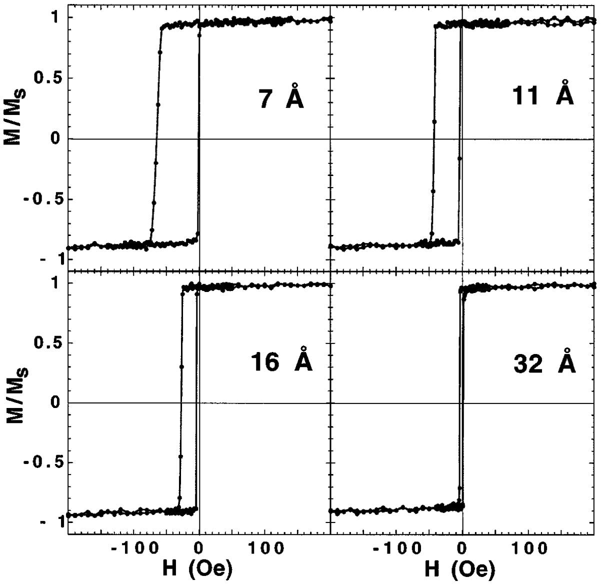

FIG. 1. Hysteresis loops for 300 Å Py/Au/300 Å CoO. The thickness of the Au spacer layer is indicated in each case.

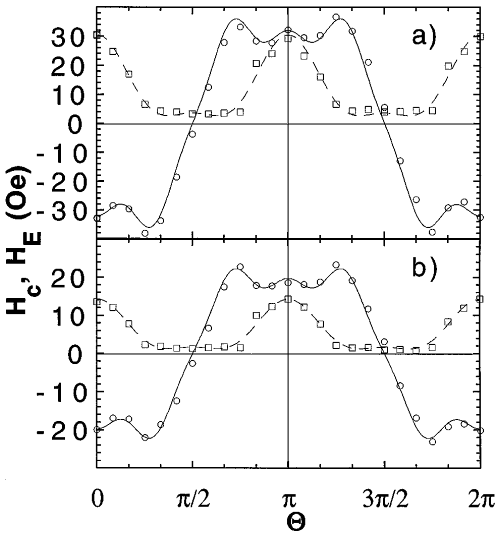

FIG. 2. Angular dependence of the exchange field H_E (solid line) and coercivity H_C (dashed line) for Py/Au/CoO trilayers with Au spacer layer thickness of (a) 7.3 Å and (b) 13.5 Å.

symmetry $[H_C(\theta) = H_C(-\theta) = H_C(\pi \pm \theta)]$. This confirms that the inherent symmetry afforded by the bias exchange has been preserved.

Because of unidirectional symmetry, the angular dependence of H_E can generally be expressed as a cosine series with odd θ terms. The H_E results shown in Fig. 2 can be well described, as shown by the solid curves, using

$$H_E(\theta) = H_{E_o}[\cos\theta - 0.27\cos 3\theta + 0.03\cos 5\theta + 0.08\cos 7\theta], \quad (2)$$

with $H_{E_o} = -38.1$ Oe in Fig. 2(a) and -23.5 Oe in Fig. 2(b). It is interesting to note that because of significant higher order terms beyond the $\cos\theta$ term, the largest values of H_E are not at $\theta = 0$ or π as commonly believed, but near $\pm\pi/4$. This feature has also been observed in FM/AF bilayers without the spacer layer [19]. The uniaxial symmetry of coercivity dictates that its angular dependence is expressible as a cosine series with even θ terms. The H_C results shown in Fig. 2 can be expressed by

$$H_C(\theta) = H_{C_o}[1 + \cos 2\theta + 1.1\cos 4\theta + 0.4\cos 6\theta], \quad (3)$$

as shown by the dashed curves, using $H_{C_o} = 12.2$ Oe in Fig. 2(a) and 5.7 Oe in Fig. 2(b). As the thickness of the spacer layer increases, the angular dependence of H_E and H_C do not vary significantly, except that the values of H_{E_o} and H_{C_o} decrease. These results firmly establish the transmission of exchange coupling across a spacer layer.

In Fig. 3(a), the values of H_E at 80 K of 300 Å Py/Ag/300 Å CoO are shown as a function of the spacer layer thickness. Excluded are the data for extremely small spacer layer thickness (≤ 4 Å), where the values of H_E vary significantly due to pinholes or discontinuous spacer layers. For increasing spacer layer thickness, the exchange field H_E decreases monotonically and smoothly, up to a maximum thickness t_c, beyond which exchange coupling vanishes. Similar dependence of H_E on the spacer layer thickness has been observed for Au and Cu spacers as shown in Fig. 3(b). However, the observed values of t_c of 65 ± 4 Å for Ag, 36 ± 4 Å for Au, and 20 ± 4 Å for Cu are different. In these samples, the absence of pinholes or discontinuities in the spacer layer were verified by XTEM [17].

In FM/AF exchange coupling, the FM layer is coupled to the AF layer with a coupling strength A, which is proportional to J [Eq. (1)] and can be expressed as $A = t_F M_F H_E$. In the present case, with $t_F = 300$ Å and $M_F = 800$ emu/cm^3 for the Py layer, one obtains A (in erg/cm^2) $= 2.4 \times 10^{-3} H_E$ (in Oe), which is also shown in Fig. 3(a). Of particular interest is the manner with which J depends on spacer layer thickness t. It is clear that the dependence of J is nonoscillatory and monotonically decreasing. We have found that a power dependence (t^{-n}), regardless of the value of n, gives an unsatisfactory description of the data. The dependence of

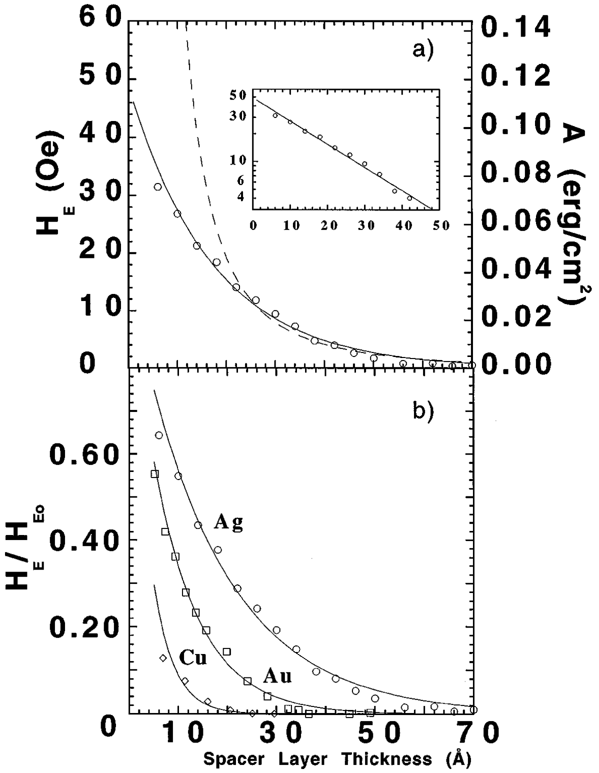

FIG. 3. (a) The dependence of exchange field H_E and coupling strength A on Ag spacer layer thickness t. The dashed curve shows the dependence of $\exp(-t/\lambda)/t^2$. In the inset, semilog plot of H_E vs Ag spacer layer thickness. (b) H_E/H_{E0} vs thickness of Ag, Au, and Cu spacer layer. The solid curves in (a) and (b) are best-fit results using $H_E = H_{E0}\exp(-t/L)$.

$$J \propto \exp(-t/\lambda)/t^2 \quad (4)$$

also does not describe the overall results, unless one excludes data with smaller t, as shown by the dashed line in Fig. 3(a). However, the experimental results can be well described by a simple exponential expression of

$$J \propto \exp(-t/L) \quad (5)$$

over the entire thickness range, as shown by the solid line in Fig. 3(a). The exponential behavior is further illustrated in the inset in Fig. 3(a). The value of L in Eq. (5) provides a measure of the range of the coupling. For Ag, Au, and Cu, the values $L = 17.3$, 9.2, and 4.1 Å have been found, respectively. The value of L for Ag is almost twice as large as that for Au. These results indicate that the range of the exchange coupling across a spacer layer is specific to the spacer material, and the long-range exchange coupling is probably electronic in nature.

Thus far, theoretical and experimental investigations of coupling of FM layers have been focused mostly on the oscillatory interlayer coupling observed in the FM/spacer/FM structure. Oscillatory coupling using Cu, Ag, and Au as a spacer layer has been well established in these structures. However, the characteristics of the new layer structure of FM/spacer/AF are very different from those of FM/spacer/FM. This new FM/spacer/AF layer structure, in which we have observed long-range exponentially decaying coupling, has not been previously addressed theoretically nor experimentally.

Under certain conditions, the rare occurrence of nonoscillatory but decaying coupling may be observed in FM/spacer/FM systems if the spacer layer possesses special features. Several theoretical studies [20,21] have shown that in the case of FM/insulator/FM, due to the exponentially decaying overlap of the FM wave function extending into the insulating barrier, the coupling strength is not oscillatory but varies as that shown in Eq. (4). Some of these features have been observed experimentally using an insulating spacer layer [22,23]. In the case of FM/metal/FM structure, if the density of states of the metallic spacer layer has a peak near and above the Fermi level (e.g., in Cr and SiFe), the oscillatory contribution may be suppressed [24]. The resulting coupling may be exponentially decaying at large distances. The recent observation of exponentially decaying coupling in Fe/SiFe/Fe structure by de Vries *et al.* is consistent with this expectation [25]. In these cases, the special features of the spacer layer, such as an energy gap and an unusual density of states, give rise to the exponential distance dependence of the interlayer coupling. In the case of FM/spacer/AF, the noble metals do not have these special features, and have previously exhibited only oscillatory coupling in the FM/spacer/FM structure. Yet, as we have demonstrated here, the long-range bias exchange coupling has a distinctive exponential behavior across noble metal spacer layers. This suggests that these are characteristics unique to the FM/AF exchange bias and the FM/spacer/AF structure.

The long-range nature of FM/AF exchange coupling should be a key ingredient, but has thus far been overlooked in the microscopic models of exchange bias. For example, because of the long-range nature of exchange coupling, the expression for H_E may need to be revised to

$$H_E = \frac{S_F S_{AF} \sum J_{ij} \cos\theta_{ij}}{t_F M_F}, \tag{6}$$

where $\cos\theta_{ij}$ is the angle between S_{Fi} and S_{AFj}. The summation not only includes the interactions at the FM/AF interface, as in Eq. (1), but rather all interactions within the range of the exchange coupling. The quantity J_{ij} denotes the strength of the interaction between any given S_{Fi} and S_{AFj} and its value decreases exponentially with their separation. More importantly, because the two sublattices of the AF will give contributions of opposite sign, the effective $J = \sum J_{ij} \cos\theta_{ij}$ is much smaller than the J_{ij} inside the summation. In this manner, the long-standing discrepancy of a single large expected J and a small measured H_E in Eq. (1) is alleviated.

In summary, we have observed long-range coupling between FM and AF materials *across* a nonmagnetic spacer layer, extending to as much as 50 Å. The coupling strength was found to decay exponentially, and was material dependent. Our results demonstrate the long-range nature of exchange bias, contrary to the prevailing assumption that exchange bias is a short-range interaction at the FM/AF interface.

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