Exchange Coupling in Ferromagnet/Antiferromagnet Bilayers with Comparable T_C and T_N

X. W. Wu and C. L. Chien
Department of Physics and Astronomy, The Johns Hopkins University, Baltimore, Maryland 21218
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Exchange coupling has been observed in FM/AF bilayers with T_C close to, and less than, T_N. The exchange field H_E and coercivity H_c show an unusual temperature dependence due to the $1/M_F$ dependence and the influence of the FM ordering by the AF layer at $T > T_C$. The exchange coupling energy vanishes at T_N, whereas the intrinsic H_c vanishes at T_C. The FM/AF exchange coupling preserves some degree of ordering in the FM layer even at $T > T_C$. [S0031-9007(98)07210-X]

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A great deal of attention has recently been focused on the elusive mechanism of FM/AF exchange coupling and the technological applications of the resultant exchange bias in spin-valve field-sensing devices and magnetic random access memories [1–8]. When a bilayer of a ferromagnet (FM) and an antiferromagnet (AF) is cooled in a magnetic field through the Néel temperature T_N of the AF to lower temperatures, an exchange bias is locked in. The hysteresis loop of the FM, instead of centering at zero magnetic field (H) as is usually the case, is shifted from $H = 0$ by an amount known as the exchange field H_E and accompanied by an enhanced coercivity H_c.

To date, although exchange coupling has been observed in a number of FM/AF systems (e.g., NiFe/FeMn, NiFe/CoO, Fe/FeF_2, etc.), the Curie temperature T_C of the FM has always been much higher than T_N of the AF. This is due in part to the conjecture that during field cooling across T_N, the FM layer should be ferromagnetically ordered, while the AF order and the exchange coupling are being established. Consequently, our understanding of exchange coupling has been limited only to the case of $T_C \gg T_N$ in which exchange bias has been realized in the temperature range of $T < T_N$ and that both H_E and H_c terminate at T_N. In this work, key issues and new features of exchange coupling have been addressed by studying FM/AF bilayers, where the T_C of the FM is *close to and less than* the T_N of the AF.

For an FM layer of thickness t_F and magnetization $\mathbf{M}_\mathrm{F}$, exchange coupled to an AF layer, the relevant free energy per unit area can be generally expressed as

$$F_A = U(\mathbf{S}_\mathrm{F}, \mathbf{S}_\mathrm{AF}) - t_F \mathbf{M}_\mathrm{F} \cdot \mathbf{H}\,, \tag{1}$$

where the first term is the all important coupling term, which includes the interactions among the spins $\mathbf{S}_\mathrm{F}$ and $\mathbf{S}_\mathrm{AF}$ of the FM and AF moments, respectively, and the second term is the Zeeman energy of the FM layer in an external field $\mathbf{H}$. Intense interest has been directed at the form of $U(\mathbf{S}_\mathrm{F}, \mathbf{S}_\mathrm{AF})$, the spin structures of the FM and the AF layers, and the microscopic mechanisms with which the exchange coupling is established [1,2,9–13]. The exchange field H_E is the magnetic field at which the direction of the magnetization of the FM is reversed. For a unidirectional U, the exchange field H_E can be obtained from Eq. (1) as

$$H_E = \frac{|U(\mathbf{S}_\mathrm{F}, \mathbf{S}_\mathrm{AF})|}{t_F M_F}\,. \tag{2}$$

The temperature dependence of H_E is due to those of M_F and $|U(\mathbf{S}_\mathrm{F}, \mathbf{S}_\mathrm{AF})|$, and the latter depends on the order parameters $\langle \mathbf{S}_\mathrm{F} \rangle$ and $\langle \mathbf{S}_\mathrm{AF} \rangle$ of the FM and the AF layers. In the simplest model with interactions among the interfacial moments only, $|U(\mathbf{S}_\mathrm{F}, \mathbf{S}_\mathrm{AF})|$ reduces to $n|J\mathbf{S}_\mathrm{F} \cdot \mathbf{S}_\mathrm{AF}|$, where n is the number of interactions per unit area with strength J. Assuming a collinear spin structure, one obtains the well-known but oversimplistic form of $H_E = n|J|\mathbf{S}_\mathrm{F}\mathbf{S}_\mathrm{AF}/t_F M_F$ [1,2], which generally does not predict the right order of magnitude for the observed values of H_E [9–13].

In addition to H_E, it has also been well established experimentally that the coercivity H_c of an FM layer, exchange coupled to an AF layer, is much larger than that for an uncoupled FM layer [1–8]. Thus far, most of the theoretical efforts have been devoted to addressing only H_E [9,12,13]; the enhanced coercivity has been an unresolved theoretical issue. Recently, however, Zhang *et al.* have proposed a theoretical model, which is based on the random field at the FM/AF interface [12], to account for the enhanced H_c in exchange-coupled FM/AF bilayers [14]. The theory predicts that at low temperatures H_c varies as

$$H_c(T) = (\alpha/t_F^{3/2} - \beta T/t_F^2)/M_F\,, \tag{3}$$

where the factors α and β involve the exchange coupling strengths among the magnetic moments in the layers and at the interface. The theory predicts that the value of H_c at low temperatures depends on t_F with a $1/t_F^{3/2}$ dependence, which has recently been experimentally observed [14]. At elevated temperatures, according to Eq. (3), H_c acquires a linear T dependence, also in agreement with the experimental data [3–8].

Since the situation of $T_C \gg T_N$ has hitherto been the case for exchange-coupled FM/AF bilayers, where M_F and $\langle \mathbf{S}_\mathrm{F} \rangle$ of the FM layer are essentially constant at $T < T_N$, only the $1/t_F$ dependence of H_E and the $1/t_F^{3/2}$ dependence of H_c can be experimentally established. In this work, by designing FM/AF bilayers with T_C of the FM close to the T_N of the AF, we have observed the strong $1/M_F$ dependence predicted for the exchange field H_E and the coercivity H_c. Furthermore, we have determined the temperature dependence of the interaction energy $|U(\mathbf{S}_\mathrm{F}, \mathbf{S}_\mathrm{AF})|$ in which both $\langle \mathbf{S}_\mathrm{F} \rangle$ and $\langle \mathbf{S}_\mathrm{AF} \rangle$ vary with temperature. We also show that exchange bias not only can be established in bilayers with $T_C < T_N$, exchange coupling exists also at $T > T_C$. These results provide new insight into the elusive exchange coupling in FM/AF bilayers.

To experimentally explore these issues, the values of the magnetic ordering temperatures (T_C and T_N) of the constituent layers must be tailored to be close to each other. For this purpose, we have used a well-known crystalline AF CoO ($T_N = 290$ K) but resorted to amorphous FMs in which the composition of the alloys can be varied as facilitated by the noncrystalline structure [15]. It has been well established that the value of T_C of amorphous $(\mathrm{Fe}_x\mathrm{Ni}_{1-x})_{80}\mathrm{B}_{20}$, among many other amorphous alloy systems, can be tailored to any value between 40 and 700 K by controlling the relative composition of Fe and Ni [15–17]. In particular, we have chosen the composition of a-$(\mathrm{Fe}_{0.1}\mathrm{Ni}_{0.9})_{80}\mathrm{B}_{20}$ with a T_C close to and below T_N of CoO in order to reveal certain characteristics of the exchange coupling. Single layers of a-$(\mathrm{Fe}_{0.1}\mathrm{Ni}_{0.9})_{80}\mathrm{B}_{20}$ and bilayers of a-$(\mathrm{Fe}_{0.1}\mathrm{Ni}_{0.9})_{80}\mathrm{B}_{20}$/CoO have been fabricated at room temperature by magnetron sputtering at 5 mTorr Ar in a chamber with a base pressure of 8×10^{-8} Torr. The single-layer FM a-$(\mathrm{Fe}_{0.1}\mathrm{Ni}_{0.9})_{80}\mathrm{B}_{20}$ films exhibit $T_C = 240$ K, which is about 50 K below the $T_N = 290$ K of CoO, as shown in Fig. 1. These samples are magnetically soft with square hysteresis loops and small coercivity ($H_c < 2$ Oe) as shown in the inset of Fig. 1. Bilayers of 900 Å of a-$(\mathrm{Fe}_{0.1}\mathrm{Ni}_{0.9})_{80}\mathrm{B}_{20}$ and 300 Å of CoO have been measured in a vibrating sample magnetometer. A thicker FM layer has been used here because of the low Fe content in a-$(\mathrm{Fe}_{0.1}\mathrm{Ni}_{0.9})_{80}\mathrm{B}_{20}$, in which the magnetization is primarily due to the Fe moments [14–16]. The magnetization value at 80 K for a-$(\mathrm{Fe}_{0.1}\mathrm{Ni}_{0.9})_{80}\mathrm{B}_{20}$ is about 4 times and 7 times smaller than those of permalloy (NiFe) and Fe, respectively.

After field cooling with a 10 kOe field from $T > T_N$ to 80 K, the a-$(\mathrm{Fe}_{0.1}\mathrm{Ni}_{0.9})_{80}\mathrm{B}_{20}$/CoO bilayers display shifted hysteresis loops at various temperatures as shown in Fig. 2. These are telltale signs of exchange bias, despite the fact that $T_C = 240$ K is considerably *lower* than $T_N = 290$ K, demonstrating that $T_C > T_N$ is not a prerequisite for establishing exchange coupling. Evidently, the induced magnetization in the FM layer at 290 K by the cooling field is sufficient for establishing exchange coupling. It is further noted by the hysteresis loops at 255 and 270 K that exchange bias and ferromagnetic ordering of the FM layer persist to temperatures *above* the T_C of the FM layer.

Before discussing the unusual temperature dependence of H_E and H_c, it is useful to mention the behavior of H_E and H_c in other FM/AF bilayers with $T_C \gg T_N$. In NiFe/CoO bilayers having the same AF layer, the value of H_E shows a plateau at low temperatures, decreases more rapidly as T_N is approached, and vanishes at T_N [5,8]. The value of H_c decreases quasilinearly with increasing temperature and acquires the value of the uncoupled NiFe layer at $T \geq T_N$. These well-known behaviors are shown in Fig. 3(a). Very different results have been observed

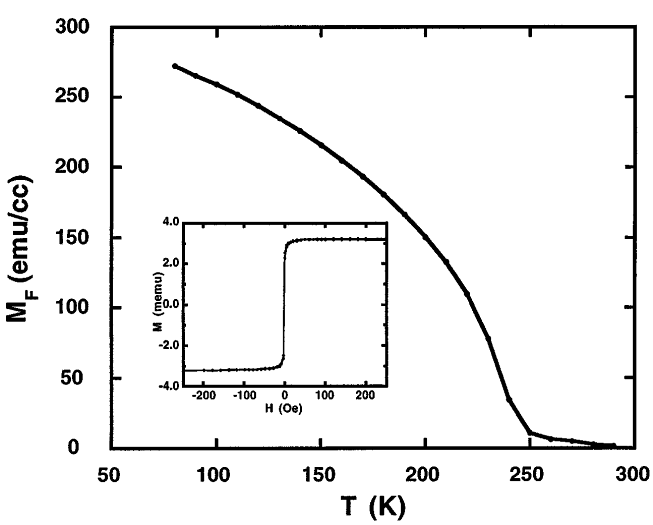

FIG. 1. Temperature dependence of the magnetization at $H = 200$ Oe of a single layer amorphous $(\mathrm{Fe}_{0.1}\mathrm{Ni}_{0.9})_{80}\mathrm{B}_{20}$ film. The inset shows the hysteresis loop of a-$(\mathrm{Fe}_{0.1}\mathrm{Ni}_{0.9})_{80}\mathrm{B}_{20}$ film at 100 K.

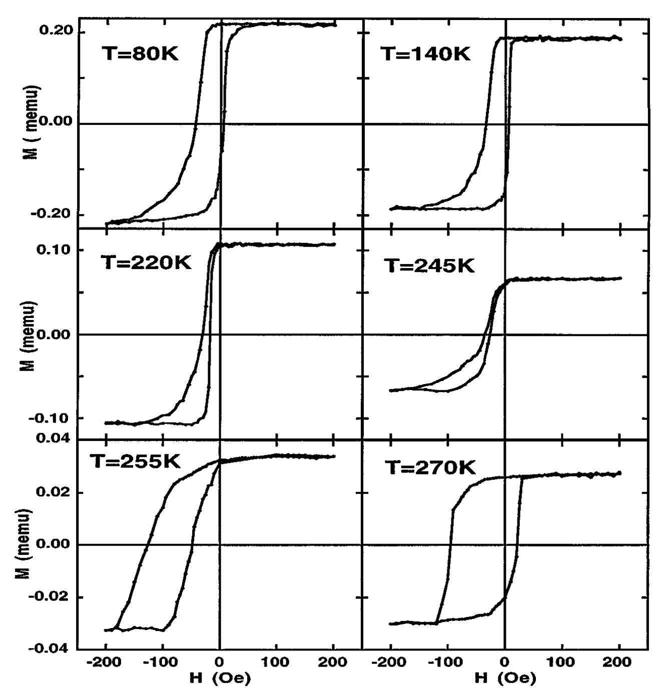

FIG. 2. Hysteresis loop of a-$(\mathrm{Fe}_{0.1}\mathrm{Ni}_{0.9})_{80}\mathrm{B}_{20}$/CoO bilayer at various temperatures from 80 to 270 K.

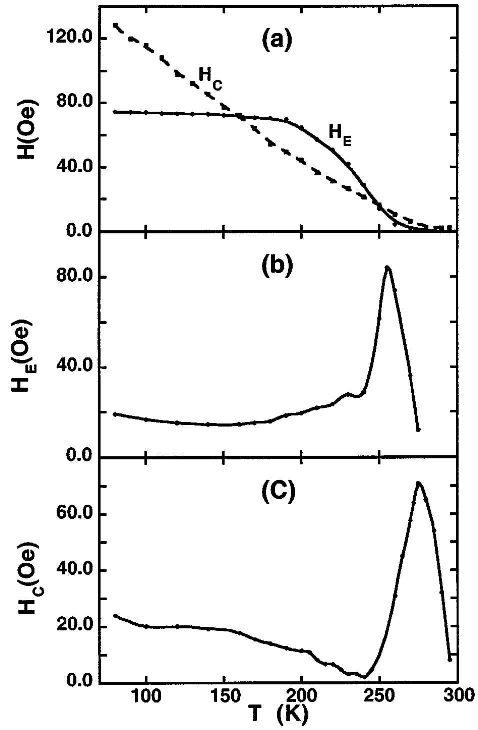

FIG. 3. Temperature dependence of (a) exchange field H_E and coercivity H_c of NiFe/CoO bilayers, (b) exchange field, and (c) coercivity of a-$(Fe_{0.1}Ni_{0.9})_{80}B_{20}$/CoO.

for a-$(Fe_{0.1}Ni_{0.9})_{80}B_{20}$/CoO bilayer as shown in Figs. 3(b) and 3(c). At low temperatures ($T \leq 160$ K), the values of H_E are roughly unchanged. However, at $T > 160$ K, the value of H_E begins to increase and sharply so near 250 K. At $T > 260$ K, H_E decreases precipitously and vanishes at about 280 K. In the case of H_c, its values decrease steadily at low temperatures but rise sharply at $T > 240$ K, displaying a maximum at about 275 K, before decreasing to the single-layer value at 290 K. Both H_E and H_c show a sharp peak feature at, respectively, 260 and 275 K, at which their values are about 4 times higher than those at low temperatures.

The rapid rise of H_E and H_c at $T < T_C$ is due to the $1/M_F$ dependence of H_E and H_c as shown in Eq. (2) and Eq. (3), respectively, as $M_F(T)$ of a-$(Fe_{0.1}Ni_{0.9})_{80}B_{20}$ decreases with temperature as shown in Fig. 1. At $T > T_C$, within the temperature range of $T_C < T < T_N$, the values of H_E and H_c must eventually decrease precipitously because of the diminishing AF order, in addition to the disappearing magnetization of the FM layer. Thus, both the $1/M_F$ dependence of H_E and H_c, and the diminishing magnetic ordering, give rise to a pronounced peak in H_E and H_c at 260 and 275 K. The actual locations of the peaks at 260 and 275 K are the consequences of the competing influences and are thus not significant. These unusual temperature dependences for H_E and H_c could not be observed in previous FM/AF bilayers with $T_C \gg T_N$, because $M_F(T)$ of the FM layer hardly varies.

With the values of the exchange field $H_E(T)$, the magnetization $M_F(T)$, and the thickness t_F of the FM layer determined, we can address the temperature dependence of the exchange coupling energy $|U(\mathbf{S}_\mathrm{F}, \mathbf{S}_\mathrm{AF})|$, which, according to Eq. (2), is $t_F M_F(T) H_E(T)$. As shown in Fig. 4(a), $|U(\mathbf{S}_\mathrm{F}, \mathbf{S}_\mathrm{AF})|$ decreases monotonically with T for the entire temperature range and vanishes near T_N, despite a sharp peak feature in H_E. It should be noted that in all previously studied FM/AF bilayers with $T_C \gg T_N$, $|U(\mathbf{S}_\mathrm{F}, \mathbf{S}_\mathrm{AF})|$ exhibits essentially the same temperature dependence as that of H_E because $M_F \approx$ constant. Since $\langle \mathbf{S}_\mathrm{F} \rangle$ is nearly constant in these cases, the temperature dependence of $|U(\mathbf{S}_\mathrm{F}, \mathbf{S}_\mathrm{AF})|$ reflects only the variation of $\langle \mathbf{S}_\mathrm{AF} \rangle$ with temperature. In contrast, in the present FM/AF bilayers, H_E and $|U(\mathbf{S}_\mathrm{F}, \mathbf{S}_\mathrm{AF})|$ have very different temperature dependences, as shown in Figs. 3(b) and 4(a), because both $\langle \mathbf{S}_\mathrm{AF} \rangle$ and $\langle \mathbf{S}_\mathrm{F} \rangle$ vary strongly with temperature. It may also be noted, while both $|U(\mathbf{S}_\mathrm{F}, \mathbf{S}_\mathrm{AF})|$ and $M_F(T)$ decrease monotonically with temperature, $M_F(T)$ decreases faster than that of $|U(\mathbf{S}_\mathrm{F}, \mathbf{S}_\mathrm{AF})|$. Consequently, H_E rises sharply with temperature near T_C, as described by Eq. (1).

Because the apparent temperature dependence of H_c is dominated by the $1/M_F$ dependence, it is useful to investigate $H_c M_F$, which reveals the intrinsic temperature dependence of H_c. As shown in Fig. 4(b), the intrinsic temperature dependence of H_c is *linear*, in accordance to Eq. (3), whereas the apparent temperature dependence of H_c, as shown in Fig. 3(b), is strongly influenced by the diminishing M_F. It is particularly noteworthy, as shown in Fig. 4(b), that the intrinsic coercivity extrapolates to

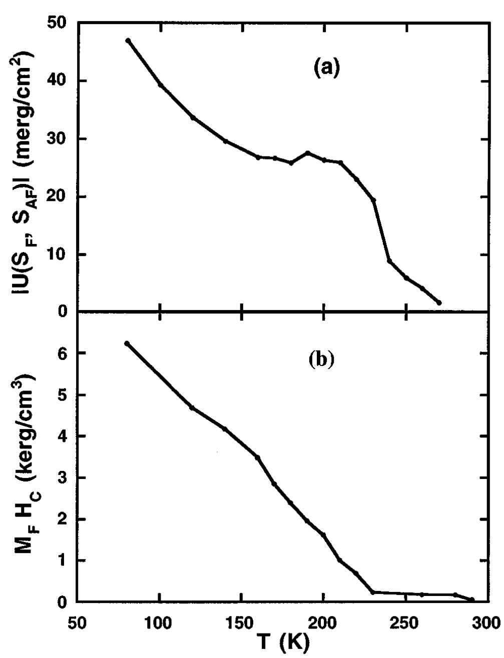

FIG. 4. Temperature dependence of (a) exchange coupling energy $|U(\mathbf{S}_\mathrm{F}, \mathbf{S}_\mathrm{AF})|$ and (b) product of coercivity H_c and magnetization M_F of a-$(Fe_{0.1}Ni_{0.9})_{80}B_{20}$/CoO.

240 K, the T_C of the FM layer, whereas the coupling energy $|U(\mathbf{S}_F, \mathbf{S}_{AF})|$ extrapolates to T_N as shown in Fig. 4(a).

If the FM layer in the FM/AF bilayers should behave the same as that of an isolated FM layer, for which $M_F = 0$ and $\langle \mathbf{S}_F \rangle = 0$ at $T > T_C$, then $|U(\mathbf{S}_F, \mathbf{S}_{AF})|$, H_E, and H_c would all vanish at T_C. No shifted hysteresis loops with a finite width would have been observed. Instead, finite values of H_E and H_c [Figs. 3(b) and 3(c)] and $|U(\mathbf{S}_F, \mathbf{S}_{AF})|$ [Fig. 4(a)] have been observed at T *above* T_C, to nearly T_N. Evidently, once the exchange coupling has been established, $\langle \mathbf{S}_F \rangle$ remains finite in the temperature range of $T_C < T < T_N$. These are evidences that, because of the exchange coupling between the two layers, the AF ordering with a slightly higher T_N *preserves* some degree of ordering in the FM layer even at T *above* T_C. Mutual influence of two AF layers has been previously reported in $(CoO)_m(NiO)_n$ superlattices, where m and n are the numbers of monolayers of CoO and NiO [18]. The $(CoO)_m(NiO)_n$ superlattice does not exhibit two, but one unique $T_N = [mT_N(\text{CoO}) + nT_N(\text{NiO})]/(m + n)$, where $T_N(\text{CoO}) = 290$ K and $T_N(\text{NiO}) = 525$ K. In the present case, we show evidence of FM ordering influenced by an adjacent AF layer.

In summary, with a tailored value of T_C close to and less than T_N in the a-$(Fe_{0.1}Ni_{0.9})_{80}B_{20}$/CoO bilayers, we have observed new features of exchange coupling which could not have been observed in previous FM/AF bilayers with $T_C \gg T_N$. We have observed the $1/M_F$ dependence of both H_E and H_c, and we have also determined the exchange coupling energy $|U(\mathbf{S}_F, \mathbf{S}_{AF})|$ and its temperature dependence in which both $\langle \mathbf{S}_{AF} \rangle$ and $\langle \mathbf{S}_F \rangle$ vary strongly with temperature. This intrinsic temperature dependence of H_c is linear and extrapolates to T_C, whereas the exchange coupling energy $|U(\mathbf{S}_F, \mathbf{S}_{AF})|$ vanishes at T_N. We have provided evidence that exchange coupling exists even in the temperature range of $T_C < T < T_N$, indicating the influence on the FM ordering by the adjacent AF layer. Finally, in spite of the long-held belief and the common practice of $T_C > T_N$ as the prerequisite for establishing exchange bias in FM/AF bilayers, we have demonstrated exchange coupling in bilayers in which $T_C < T_N$.

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