Memory effects of exchange coupling in ferromagnet/antiferromagnet bilayers

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The magnetization state of the ferromagnet is crucial in the cooling process for establishing exchange coupling in ferromagnet/antiferromagnet bilayers. Using special cooling procedures, the value and even the sign of the exchange bias field in several bilayers can be greatly altered. While the coercivity only depends on temperature, the exchange bias field shows an accumulative memory of the thermal and field history of the bilayer. We propose that this is due to the formation of a domain wall with a temperature dependent width in the antiferromagnet. [S0163-1829(99)02130-X]

A great deal of experimental and theoretical attention has been focused on the intriguing physics of the exchange coupling between a ferromagnet (FM) and an antiferromagnet (AF), and the central role of exchange bias in spin-valve devices. To establish the exchange coupling experimentally, it is a common practice to cool the FM/AF bilayer in a dc magnetic field from $T>T_N$ to lower temperatures, where T_N is the Néel temperature of the AF.[1] In the cooling process, the AF order is established while the FM layer is in the single-domain state. The resultant exchange coupling causes the hysteresis loop of the FM layer to shift by the amount of the exchange bias field (H_E), accompanied by a larger coercivity (H_c) than that of the uncoupled FM layer. After this common cooling process, the values of both H_E and H_c decrease with increasing temperature[1–7] until the so-called blocking temperature (T_B) at which H_E vanishes and H_c retains its uncoupled FM value. For some AF (e.g., CoO), the values of T_B and T_N are essentially the same, whereas in others (e.g., NiO) T_B can be noticeably lower than T_N.[2]

It has often been taken that once cooling across T_N has been accomplished, a unique exchange coupling has been established. We show in this work that both the value and the sign of H_E depend on the cooling process, in which the state of the FM layer is of key importance. More importantly, we show that the resultant exchange coupling retains an accumulative memory effect of the *entire* cooling procedure. Not only the value and sign of H_E can be tailored, but the so-called blocking temperature T_B can also be manipulated to have virtually any value less than T_N. On the other hand, the value of H_c is uniquely defined at each temperature, independent of the cooling history and the resultant values of H_E and T_B. The unidirectional anisotropy, which gives rise to H_E, can thus be altered and manipulated, while the uniaxial anisotropy associated with H_c remains unchanged.

These results are relevant to the microscopic origin of the exchange coupling. It has been generally accepted that the FM/AF coupling is due to the interactions among the FM and the AF moments across the FM/AF interface. Most micromagnetic models, with or without interfacial roughness and defects, assume certain spin structures for the FM and the AF layers or allow the FM and the AF moments to arrive at a spin structure through their interactions.[8–12] The emerging picture is that the exchange coupling is the result of an uncompensated magnetization $\mathbf{\Delta M_{AF}}$ at the FM/AF interface, and a domain wall that forms in the AF material involving several layers of AF moments.[9,10] The domain wall forms when the magnetization of the FM is reversed, whether the coupling at the interface is ferromagnetic ($J>0$), antiferromagnetic ($J<0$),[1] or of a spin-flop type.[11] The observed memory effect is a manifestation of the AF domain wall manipulated by magnetic field and temperature.

The features presented in this work have been observed in FM/AF bilayers of Py/CoO, Py/FeMn, a-FeNiB/CoO, and Py/(111)CoO, where Py$=Ni_{81}Fe_{19}$, a-FeNiB$=$amorphous $Fe_4Ni_{76}B_{20}$, and (111)CoO is a single crystal film epitaxially grown on a (0001) sapphire substrate. The constituent layers have the ordering temperatures of $T_C(\mathrm{Py})=850$ K, $T_C(a\text{-FeNiB})=150$ K, $T_N(\mathrm{CoO})=290$ K, and $T_N(\mathrm{FeMn})=458$ K. In the Py/CoO and the Py/FeMn bilayers we have the well-known case of $T_C \gg T_N$, whereas in a-FeNiB/CoO we have the unusual situation of $T_C \ll T_N$.[13] These bilayer samples were made in a magnetron sputtering system with a base pressure of 8×10^{-8} Torr. The FM layer was deposited in a magnetic field to induce an easy axis. Magnetic hysteresis measurements were made in a vibrating sample magnetometer. The uncoupled FM layer displays a square loop with a coercivity of a few Oe [Fig. 1(a)].

It is essential to specify the conditions, particularly the state of the FM layer, during the cooling process in establishing the exchange coupling. Only the usual field-cooling (FC) procedure with a sufficiently large dc magnetic field assures a saturation magnetization (M_s) of the FM layer, and subsequently the usual exchange bias. In addition to the usual FC procedure, we have used two new cooling procedures to achieve a different magnetization (M) of the FM layer, where $M\neq M_s$. The three cooling procedures are (1) the usual FC from $T>T_N$ in a dc field of 200 Oe with $M=M_s$, (2) demagnetize the FM layer at $T>T_N$ using an oscillating magnetic field of decreasing magnitude until $H=0$ and $M=0$, and then ZFC, and (3) cool from $T>T_N$ in an ac magnetic field (denoted as ACFC), either of 200 Oe oscillating at 1/4 Hz (for Py/CoO) or 50 Oe at 1 Hz (for Py/FeMn), with a time-varying M averaged to $0(\langle M\rangle=0)$. The duration of the cooling procedure is approximately 10 minutes for the Py/FeMn, and 40 minutes for the Py/CoO. The frequency of the oscillating field is therefore much higher than the cooling rate.

The results of the Py/FeMn bilayer at 300 K are shown in

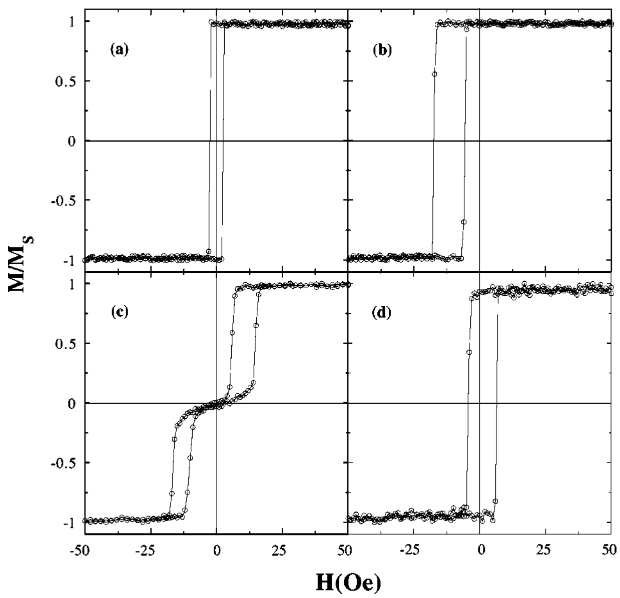

FIG. 1. Magnetic hysteresis loops of Py(150 Å)/FeMn(300 Å)/Cu(300 Å) (a) at 400 K, (b) at 300 K after field cooling from 400 K in a 200 Oe field, (c) at 300 K after demagnetizing at 400 K, and zero field cooling, and (d) at 300 K, after cooling from 400 K in a oscillating field of 50 Oe at 1 Hz.

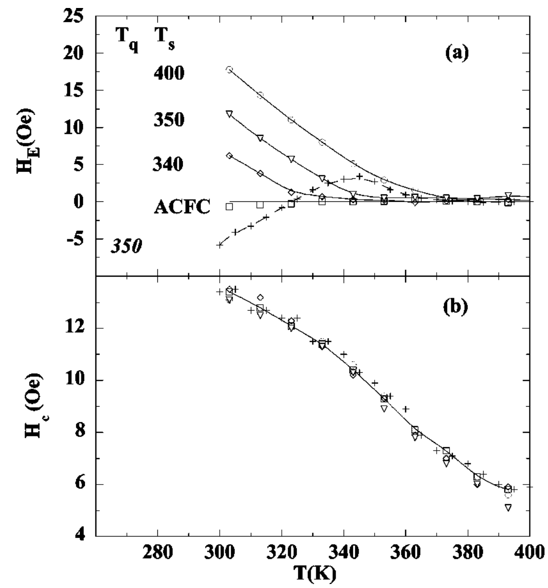

FIG. 2. Temperature dependence of (a) H_E and (b) H_c for Py(150 Å)/FeMn(300 Å)/Cu(300 Å), after cooling in a 50 Oe oscillating field at 1 Hz from 400 K to T_s and cooling in a 200 Oe field from T_s to 300 K. Also shown are the results after the sample has been cooled in a $+200$ Oe field from 400 K to $T_q = 350$ K, and in a -200 Oe field from $T_q = 350$ K to 300 K.

Fig. 1 for the three procedures mentioned above. The usual FC procedure (1) from 400 K results in a shifted hysteresis loop at 300 K, as shown in Fig. 1(b). Under the demagnetizing and ZFC procedure (2), there are two loops shifted to opposite sides as shown in Fig. 1(c). This is because the FM layer has formed a striped domain structure. Due to the uniaxial anisotropy of the FM, the magnetization in each domain is aligned in either of two equally preferred directions, resulting in *two* different unidirectional anisotropies after cooling. Although the sample was ZFC, it is as if two samples were FC with opposite magnetic fields. Under the ACFC procedure (3), the resultant loop displayed *no* exchange field ($H_E = 0$) and only an enhanced H_c, as shown in Fig. 1(d). Because the magnetization in the FM layer was changing during ACFC, there was no preferred direction with which to induce an FM/AF exchange bias. The oscillating field method can completely suppress the exchange bias. Qualitatively, the same results as those in Fig. 1 have been observed in the Py/CoO bilayers, except that the measurements were made at 200 K instead of 300 K, because CoO has a T_N of only 290 K.[14] These results demonstrate that the state of the magnetization of the FM during cooling dictates the resulting exchange bias in FeMn/Py and CoO/Py bilayers. However, in FeF_2/Fe bilayers, the magnitude of the cooling field has been shown to be of great importance; for cooling fields of different magnitudes, both positive and negative exchange bias fields have been observed.[4]

To further explore the establishment of the exchange bias, the Py/FeMn bilayer was first ACFC from 400 K in an oscillating field to a temperature T_s, then FC in a dc field of 200 Oe from T_s to 300 K. The first part of the process was designed to suppress, and the second part to induce, exchange coupling. A series of measurements was then made at increasing temperatures from 300 K. The results at various temperatures with different T_s are shown in Fig. 2. For example, when the sample was ACFC to $T_s = 340$ K, and then FC in a dc field of 200 Oe from $T_s = 340$ K to 300 K, the exchange bias at 300 K is no longer zero. For increasing temperature, the exchange bias field decreases and vanishes at 340 K, the same as that of T_s, and remains zero at $T > T_s$. Similar results for other values of T_s are shown in Fig. 2. The bilayer exhibits exchange bias for $T < T_s$, but no exchange bias for $T > T_s$. These results demonstrate that the bilayer sample has the *memory* of the temperature T_s at which the dc cooling-field was switched on; *and* that at $T > T_s$, there was no exchange bias. To erase the memory of the entire cooling procedure, the sample must be heated to $T > T_N$. Memory effects similar to those observed in Py/FeMn have also been observed in Py/CoO, as shown in Fig. 3.

To further demonstrate the memory effect, we have studied the consequence of reversing the direction of the dc field during field-cooling. The Py/CoO sample was FC in a $+200$ Oe field to a temperature T_q, at which the field was reversed to -200 Oe. The sample was then FC from T_q to 200 K, i.e., FC in a positive field in $T > T_q$, and FC in a negative field from T_q to 200 K. Measurements were then made at increasing temperature from 200 K to T_q and extending to 300 K. The results are shown in Fig. 3 for the Py/CoO bilayer. Consider $T_q = 265$ K in Fig. 3 for example. The value of H_E in the negative FC range ($200\,\mathrm{K} < T < T_q$) now increases with increasing temperature, before reverting to decreasing with temperature in the positive FC range ($T > T_q$). For a sufficiently high T_q (e.g., 270 K) even the sign of H_E can be reversed. These results again demonstrate clearly that the bilayer has the accumulative memory of a positive FC in $T > T_q$, followed by a negative FC in 200 K $< T < T_q$. The same memory effects of the reversing cooling field have also been observed in Py/FeMn, shown by

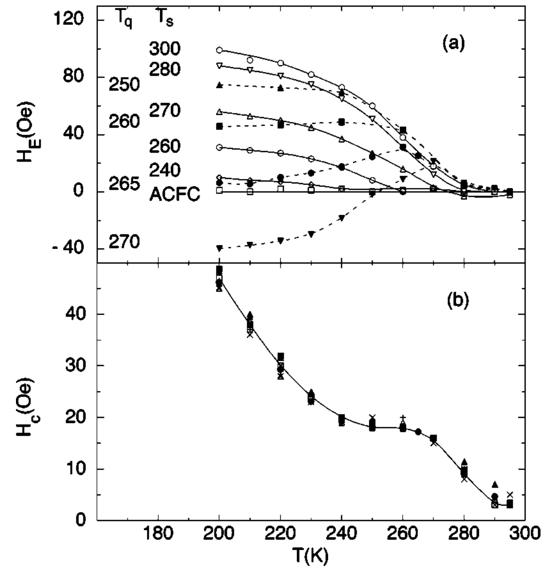

FIG. 3. Temperature dependence of (a) H_E and (b) H_c for Py(180 Å)/CoO(470 Å), after cooling in a 200 Oe oscillating field at 1/4 Hz from 300 K to T_s and in a 200 Oe dc field from T_s to 200 K. Also shown are the results after the sample has been cooled in a $+200$ Oe field from 300 K to T_q, and in a -200 Oe field from T_q to 200 K.

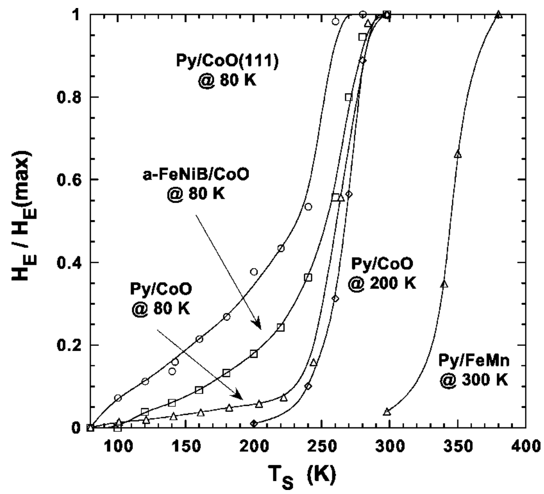

FIG. 4. Dependence of exchange bias field H_E on T_s at which field cooling was initiated of Py(150 Å)/FeMn(300 Å) at 300 K, Py(180 Å)/CoO(470 Å) at 200 K and at 80 K, Py(180 Å)/(111)CoO(470 Å) at 80 K, and a-$Fe_4Ni_{76}B_{20}$(300 Å)/CoO(218 Å) at 80 K. H_E(max) is the value obtained at these temperatures under normal field cooling.

the results of $T_q = 350$ K in Fig. 2. Furthermore, results similar to those in Fig. 3 have been observed in Py/(111)CoO, where (111)CoO is epitaxially grown on a single crystal (0001) sapphire substrate.

We have thus demonstrated that the exchange bias field can be locked in or suppressed at *any* temperature below T_N, and furthermore, the value of exchange field can acquire *any* value less than the maximum allowed at that temperature by full field-cooling, i.e., $-H_E(\text{max}) \leqslant H_E \leqslant +H_E(\text{max})$. The full strength of the exchange field H_E(max) requires field-cooling throughout the entire temperature range of $T \leqslant T_N$. However, using ACFC from T_N to T_S, and FC from T_s, the value of H_E at a given temperature can be altered to any value less than H_E(max). The results of H_E/H_E(max) for Py/FeMn measured at 300 K (taken from Fig. 2), Py/CoO measured at 200 K (taken from Fig. 3), and Py/(111)CoO at 80 K are shown in Fig. 4 as a function of T_s. It is interesting to note that when H_E measured at a low temperature (e.g., 80 K) equals its maximum value H_E(max), T_s equals T_N. A measurement at a low temperature is thus capable of determining T_N. For example, the results of Py/CoO show that T_N of CoO is 292 K. However, Py/(111)CoO shows that T_N of (111)CoO is slightly lower at 280 K, because of a tetragonal distortion due to epitaxy.[15] This indicates that both the FM and the AF are essential in the establishment of the exchange bias. A change in either the AF structure (polycrystalline vs single crystal) or the FM magnetization during cooling modifies the domain wall formation in the AF.

Also shown in Fig. 4 are the results for a-$Fe_4Ni_{76}B_{20}$/CoO, for which $T_C = 150$ K is much less than T_N, instead of $T_C \gg T_N$ as in all the other cases. We have recently shown that in a FM/AF bilayer where $T_C \ll T_N$, exchange coupling can still be established, and persists even to $T > T_C$ where there is no FM ordering.[13,16] In the present context, as shown in Fig. 4, the value of H_E of a-FeNiB/CoO at 80 K depends on T_s in a manner qualitatively the same as those of the traditional Py/FeMn and Py/CoO bilayers.

These results observed in various FM/AF bilayers illustrate that the accumulative memory effect is a general phenomenon. Under normal FC, T_B at which H_E vanishes is the same as T_N for the CoO/Py bilayer. It is noted in Figs. 2 and 3 that since H_E can now be made to vanish at T_s, T_s effectively becomes T_B, and its value can be manipulated to be any value below T_N. The single crystal (111)CoO layer shows the same general behavior and memory effect as those of polycrystalline CoO. Thus the accumulative memory effect is intrinsic to FM/AF exchange coupling.

Most remarkably, while the exchange bias field H_E can be altered to such a great extent, the coercivity H_c is *uniquely* defined at each temperature regardless of the different thermal and field cycles, as shown in Fig. 2(b) and Fig. 3(b). It is important to note that H_c is temperature specific for all different cooling procedures. In addition, the unique coercivity decreases to the value of the uncoupled FM at T_N, irrespective of both the temperature at which H_E vanishes, and the cooling procedure. These results indicate that the coercivity is unaffected by the cooling procedure which only shifts the location of the loop as signified by H_E, but not the loop width, which is H_c. Because H_E can be altered greatly by different cooling procedures and H_c remains intact, the observed memory effect is unlikely to be due to lateral inhomogeneity, roughness, and other imperfections at the FM/AF interface as has been suggested.[5–7]

We propose that the key to the observed memory effect is the AF domain wall, which arises from the exchange coupling between the AF and the FM. Experimental observation of such an AF domain wall has been proven to be challenging, far more so than the FM domain wall. However, the existance of the AF domain wall in exchange coupled systems has been indicated by several micromagnetic calculations.[8–10] The magnetic anisotropy of the AF (K_{AF}) not only affects the *energy* of the domain wall $E \approx 4\sqrt{A_{AF}K_{AF}}$, but also the *width* of the domain wall

$\Delta \approx \pi \sqrt{A_{AF}/K_{AF}}$.[9] With decreasing temperature, K_{AF} increases,[17] hence the energy of the domain wall increases, resulting in an increase in H_E. At the same time, an increasing K_{AF} will result in a *decreasing* domain wall thickness. It is the decreasing domain wall thickness with decreasing temperature that results in the memory effect.

Consider the example of cooling in a positive field from $T > T_N$ to T_q, and then cooling in a negative field from T_q to lower temperatures. Just below T_N, the width of a domain wall would be large, because of the small value of K_{AF}. Thus, an applied field will affect a large part of the AF, and establish a unidirectional anisotropy. As T is decreased from T_N, the wall width becomes smaller. Suppose at T_q, the width of a potential domain wall has been reduced to Δ_0. Reversing the field at T_q will introduce a domain wall of such a width, while AF spins farther from the interface remain unaltered. During further cooling from T_q with the negative field, these AF spins farther than Δ_0 away from the interface are *not* effected, and retain the memory of the cooling at $T > T_q$. If one now measures H_E at a temperature below T_q, the hysteresis loop reveals the spin structure frozen in when reversing the field at T_q. The spin structure due to the positive cooling field appears only above T_q. In short, the accumulative memory effect in exchange coupling is the consequence of a *changing* AF domain wall. The magnitude of H_c does not depend on the creation and/or alteration of partial domain walls in the AF, hence it is independent of the cooling procedure.

The accumulative memory effect also has technological implications. We show that exchange bias can be established by FC from *any* temperature $T_s < T_N$, below which exchange coupling would occur for both $T_C > T_N$ and $T_C < T_N$. A larger H_E at a given operating temperature can be obtained by using a higher T_s. Another important consequence of the memory effect is that the established exchange bias in a device (e.g., spin-valve GMR head) can be compromised by inadvertent temperature fluctuation in the presence of a magnetic field.

In summary, we have shown that the state of the magnetization of the FM is the crucial parameter in establishing exchange coupling. The exchange bias can be modified greatly in its value and sign by changing the cooling procedure. We show that the resultant exchange coupling depends on the entire thermal and field history from T_N to the measurement temperature, not merely on crossing the Néel temperature. A model including the effects of domain wall formation in the AF accounts well for the accumulative memory effect. While the exchange bias can be manipulated, the coercivity always maintains a unique value at a given temperature.

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