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Exchange coupling in the paramagnetic state

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Exchange coupling has been observed in a ferromagnet (FM)/antiferromagnet (AF) bilayer with Curie temperature (T_C)$\ll$Néel temperature (T_N) in the regime of $T<T_N$, even when the FM layer is in the paramagnetic state. The exchange field H_E and coercivity H_C of the coupled bilayer show completely different temperature dependence from conventional FM/AF bilayers with $T_C>T_N$. With increasing temperature, H_C vanishes at T_C, whereas H_E persists to T_N. The results show that the exchange coupling can be established in FM/AF bilayers regardless of the relative values of T_C and T_N. [S0163-1829(99)08525-2]

Exchange coupling between a ferromagnet (FM) and an antiferromagnet (AF) has attracted a great deal of attention recently due to the elusive mechanism for the FM/AF coupling[1–10] and the prominent role that exchange bias plays in spin-valve field sensing devices.[11] When a FM/AF bilayer, with the Curie temperature (T_C) of the FM higher than the Néel temperature (T_N) of the AF, has been field-cooled across T_N, an exchange bias is set in. The resultant hysteresis loop of the FM is now shifted by an amount termed the exchange field (H_E), accompanied by an enhanced coercivity (H_C). In almost all cases thus far reported, T_C has always been much higher than T_N. During field cooling across T_N, the FM layer is in the single-domain state while the exchange coupling is being locked in. In these exchange-coupled FM/AF bilayers, the resultant values of H_E and H_C always decrease with increasing temperature until T_N, at which $H_E=0$ and H_C reaches the value for a single uncoupled FM layer. These well-established experimental facts indicate that the AF ordering and the value of T_N, which is the lower of T_N and T_C, dictate the characteristics of the exchange coupling. It has been generally accepted that $T_C>T_N$ is a prerequisite for establishing FM/AF exchange coupling. Indeed, virtually all theoretical models of exchange bias have assumed the condition of $T_C>T_N$ and have focused on the AF ordering, the AF domains, and their interactions with the FM layer.[1,3,4,9]

Recently, we have demonstrated exchange coupling in an FM/AF bilayer with T_C near but *less* than T_N.[12] The inverse dependence of H_E and H_C on the FM layer magnetization M_F, previously predicted theoretically, has been experimentally observed. The exchange coupling has been shown to persist to $T>T_C$ until T_N. However, because of the proximity of T_C and T_N, induced FM ordering by the AF ordering also occurs at $T>T_C$, complicating the observation of the intrinsic temperature dependence of H_E and H_C. In this work, we have studied an FM/AF bilayer with T_C *much lower* than T_N, a hitherto unexplored regime where the FM ordering is absent when the exchange coupling is being established. We have observed exchange coupling in this system, which persists well into the paramagnetic (PM) state ($T>T_C$). The behaviors of H_E and H_C in such a system are very different from those in the traditional systems with $T_C>T_N$, as well as that with T_C near T_N. These results provide new insight into the elusive mechanisms of exchange coupling and are relevant to applications of exchange coupling in spin-valve devices.

Most of the AF materials with which FM/AF exchange coupling has been established, have relatively low values of T_N, such as FeMn ($T_N=493$ K), CoO ($T_N=291$ K), and NiO ($T_N=525$ K). The values of T_C for common crystalline FM's such as Fe, Co, and permalloy are several hundred degrees higher than those values of T_N. On the other hand, the values of T_C of many amorphous FM alloys can be tuned within a wide range of values by altering the composition as facilitated by the noncrystalline structure. For example, T_C of amorphous $(\text{Fe-Ni})_{80}B_{20}$ can be tailored to any value from 40 K to over 600 K by changing the relative composition of Fe and Ni.[13,14] Previously, we have chosen the FM a-$Fe_8Ni_{72}B_{20}$ ($T_C\approx240$ K)/AF CoO ($T_N=291$ K) system, with the T_C slightly below the T_N.[12] In this work, we employed a-$Fe_4Ni_{76}B_{20}$ ($T_C\approx150$ K) and CoO to examine features in exchange coupling with $T_C\ll T_N$.

Amorphous $Fe_4Ni_{76}B_{20}$, a soft FM with square hysteresis loops, can be readily fabricated by sputtering. Antiferromagnetic CoO has a convenient T_N near room temperature. In addition, the so-called blocking temperature (T_B), at which $H_E=0$, is virtually the same as T_N in CoO, whereas in NiO and FeMn one has the complication that T_B is much lower than T_N. Single layers of a-$Fe_4Ni_{76}B_{20}$ and bilayers of a-$Fe_4Ni_{76}B_{20}$ (300 Å)/CoO (250 Å) were fabricated in a multi-source sputtering system with a base pressure of 5×10^{-8} Torr onto room temperature Si substrates. Magnetic measurements were made by a vibrating sample magnetometer (VSM) with a field of 12 kOe and a SQUID magnetometer with a field of 50 kOe.

The temperature dependence of magnetization of a-$Fe_4Ni_{76}B_{20}$, illustrating $T_C\approx150$ K, is shown in Fig. 1 for external fields of 5 and 500 Oe. The hysteresis loop of a single 300 Å a-$Fe_4Ni_{76}B_{20}$ layer at 80 K is shown in Fig. 2(a), exhibiting a square loop with a small coercivity of only 0.4 Oe, which are characteristics of a soft FM. However, a bilayer of a-$Fe_4Ni_{76}B_{20}$ (300 Å)/CoO (250 Å), field-cooled in a field of 10 kOe to 80 K, shows a shifted hysteresis loop with large values of H_E and H_C, which are clear signatures of exchange coupling. The hysteresis loops measured at successively higher temperature from 80 to 290 K are shown in

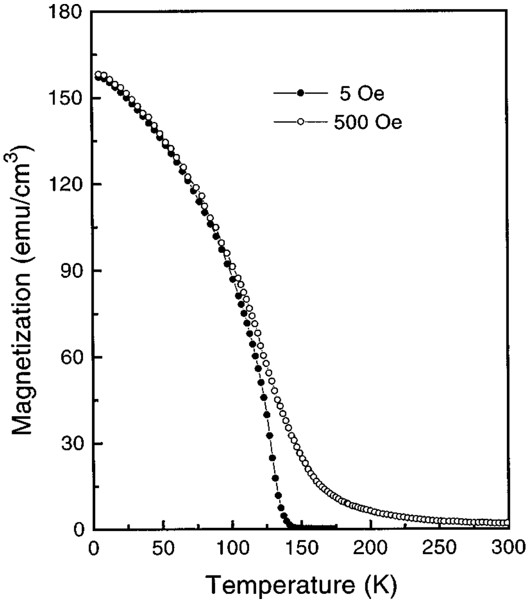

FIG. 1. Temperature dependence of magnetization of a single a-$Fe_4Ni_{76}B_{20}$ layer at 5 and 500 Oe.

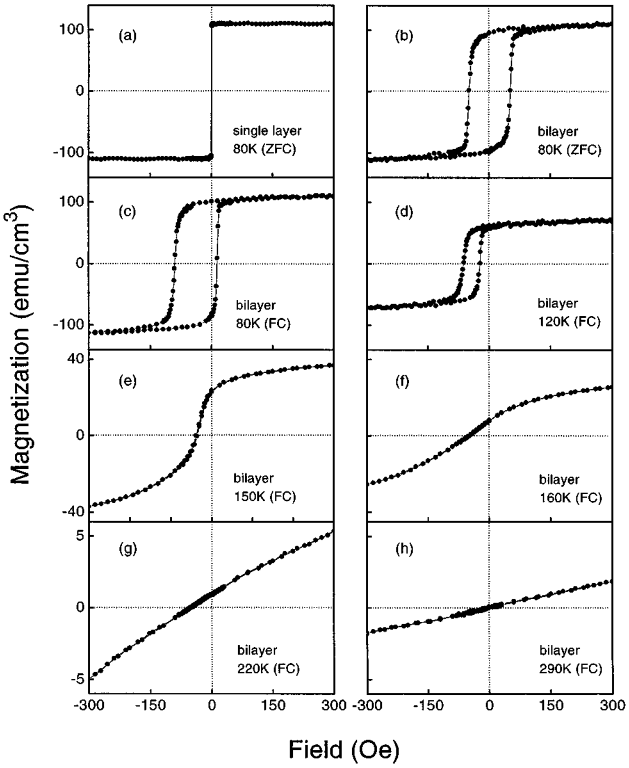

FIG. 2. Hysteresis loops of a single layer a-$Fe_4Ni_{76}B_{20}$ at 80 K (a) and a bilayer of a-$Fe_4Ni_{76}B_{20}$ (300 Å)/CoO (250 Å) at 80 K after zero-field cooling to 80 K (b), after field cooling in 10 kOe to 80 K and measured at 80 K (c), 120 K (d), 150 K (e), 160 K (f), 220 K (g), and 290 K (h).

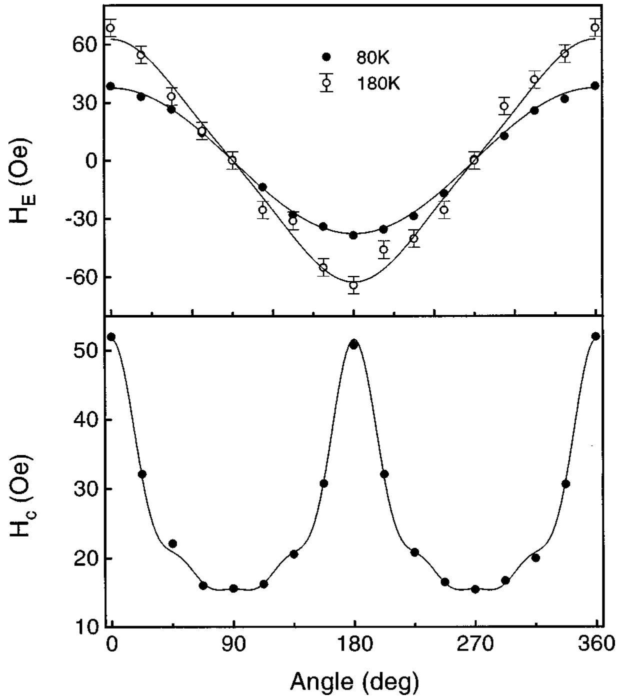

FIG. 3. Angular dependence of exchange field H_E and coercivity H_C of a-$Fe_4Ni_{76}B_{20}$ (300 Å)/CoO (250 Å) at 80 K (solid symbols) and 180 K (open symbols).

Figs. 2(c)–2(h). At higher temperatures, the coercivity progressively decreases and vanishes near T_C. Most strikingly, the *collapsed* loop at $T>T_C$ continues to be shifted with an exchange field H_E, which first increases to a maximum before decreasing progressively to zero at 290 K, the T_N of CoO. Thus, we not only have observed exchange coupling at $T<T_C$ in a bilayer where T_C is much less than T_N, but also at $T>T_C$, when the FM layer is in the PM state. Note that this is different from the previous a-$Fe_8Ni_{72}B_{20}$/CoO system with T_C slightly below T_N, where the persistence of the exchange coupling above T_C is manifested by a shifted hysteresis loop with substantial coercivity due to the short range FM ordering.

To more firmly establish exchange coupling in this unusual situation, we have measured the angular dependence of the exchange coupling in a procedure described elsewhere.[15] By directing the external field at various angles with respect to the anisotropy axis imposed by field cooling, one can determine the angular dependence of H_E and H_C. We have performed these measurements at both $T<T_C$ and $T>T_C$ for the exchange-coupled bilayer. As shown in Fig. 3, at 80 K, H_E and H_C exhibit the expected unidirectional and uniaxial symmetry, respectively. At 180 K, while H_C already vanishes to zero, H_E retains the unidirectional symmetry. These results unambiguously demonstrate exchange coupling at $T<T_C$ and $T>T_C$.

The temperature dependence of H_E and H_C, obtained from the hysteresis loops shown in Figs. 2(c)–2(h), are presented in Fig. 4. A number of striking features are evident. First of all, H_E and the enhanced H_C do not both vanish at T_N, completely different from what has been universally observed in bilayers with $T_C>T_N$. Instead, while H_E vanishes at T_N, H_C vanishes at a lower temperature near T_C. This indicates vividly that in exchange-coupled FM/AF bilayers, the exchange field is dictated by the AF ordering, but the coercivity, although significantly enhanced by the exchange coupling, is intrinsic to the FM ordering. Most importantly, the collapsed loop continues to be shifted from $H=0$ at $T>T_C$, i.e., the exchange coupling persists when the FM layer is already in the PM state.

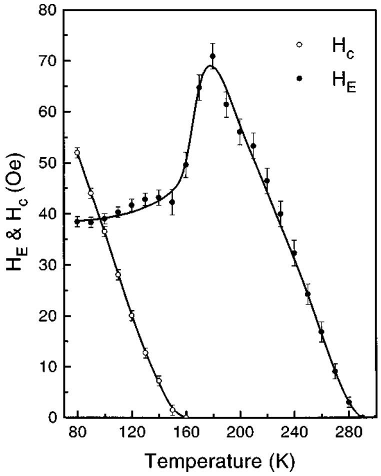

FIG. 4. Temperature dependence of exchange field H_E and coercivity H_C of a-$Fe_4Ni_{76}B_{20}$ (300 Å)/CoO (250 Å) after field cooling in 10 kOe to 80 K.

It is noted in Fig. 4 that, while H_C decreases monotonically with temperature and reaches the terminal value at T_C, H_E shows a sharp rise near T_C before decreasing towards zero at T_N. These results are different from those of a-$Fe_8Ni_{72}B_{20}$/CoO system where both H_E and H_C show a peak near T_C.[12] The peak feature of H_E in both systems is due to the inverse dependence of H_E on the FM layer magnetization M_F at $T<T_C$. As temperature is increased towards T_C, the diminishing M_F causes an increase of H_E. At $T>T_C$, as T_N is approached, H_E must decrease towards zero due to the diminishing AF ordering. The behavior of H_C in a-$Fe_8Ni_{72}B_{20}$/CoO and a-$Fe_4Ni_{76}B_{20}$/CoO are very different. In a-$Fe_8Ni_{72}B_{20}$/CoO system, H_C is complicated by the induced short range FM ordering at $T>T_C$ because T_C is close to T_N.[12] In the present a-$Fe_4Ni_{76}B_{20}$/CoO system with a much lower T_C, H_C vanishes at T_C, demonstrating clearly that H_C is intrinsic to the FM ordering.

Additional measurements have been made to elucidate the establishment of the exchange coupling in the PM state. When the bilayer was zero-field cooled (ZFC) to 80 K, the hysteresis loop exhibits a larger H_C but no exchange field, as shown in Fig. 2(b). The large value of H_C indicates that exchange coupling has already been set in, but without the unidirectional anisotropy, which has not yet been defined without field cooling. In another measurement, when the bilayer was field-cooled from $T>T_N$ under a 10 kOe field to 220 K, a shifted loop with no H_C, same as that shown in Fig. 2(g), was observed. We have also field-cooled the bilayer at 10 kOe to 220 K, turned off the field, and cooled the bilayer in zero field from 220 to 80 K. At 80 K, we obtained the same hysteresis loop as the one shown in Fig. 2(c), which is for a sample field-cooled at 10 kOe to 80 K without interruption. These results indicate that the exchange coupling has been locked in at T_N *before* the FM ordering has been established. Once established, subsequent cooling of the sample, with or without a field, does not alter the exchange coupling.

The microscopic origin of exchange coupling in general is the unidirectional anisotropy frozen in the AF spin structure, once the FM/AF bilayer is field-cooled across T_N.[1,3,4,9] In traditional FM/AF bilayers with $T_C>T_N$, the FM ordering, which already exists at T_N, has been featured in all theoretical models as a necessity for establishing the unidirectional anisotropy in the AF. The magnitude of the cooling field (H_{cool}) is of little consequence on H_E once the FM is aligned in the single-domain state using a modest H_{cool}.[16] In the previous case of T_C near but less than T_N, the short range FM ordering induced by the applied magnetic field facilitates the establishment of the exchange coupling. Once established, the exchange coupling persists into $T>T_C$ as certain FM ordering is still preserved by the neighboring AF layer, until T approaches T_N. In the present case of $T_C \ll T_N$, the key question is the establishment of the exchange coupling when the FM layer is still in the PM state. It is natural to expect that the induced magnetization (M_{PM}) in the PM layer to play a similar role as that of a FM layer in pinning the AF layer. The value of H_{cool} is now expected to play a crucial role at T_N because any induced magnetization can only result from a nonzero H_{cool}. It is therefore of interest to investigate the dependence of the resultant exchange coupling on the magnitude of H_{cool} during field-cooling across T_N. Experimental results show that, at 290 K, which is much higher than T_C, the induced M_{PM} is small and increases with H_{cool} unabated without saturation. For small values of H_{cool} (e.g., less than 10 kOe), the induced M_{PM} is proportional to H_{cool}. If this induced M_{PM} were to establish the AF spin structure that gives the strength of the exchange coupling, one might expect H_E to scale with M_{PM} and hence follow roughly with H_{cool}. Moreover, since M_{PM} cannot be saturated with H_{cool}, neither would H_E. Instead, the observed H_E depends much more strongly on H_{cool}, as shown in Fig. 5. It initially increases rather sharply with H_{cool}, then levels off to a constant for H_{cool} larger than 10 kOe. This indicates that a PM with a modest induced M_{PM} could achieve the full strength of the exchange coupling without the necessity of a fully aligned FM state as assumed in all theoretical models.

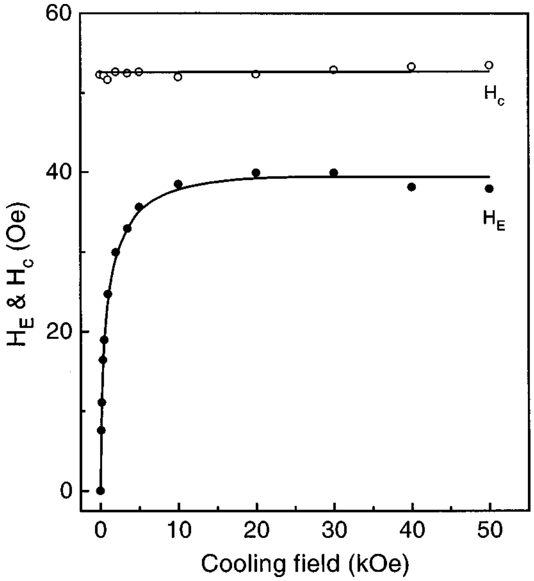

FIG. 5. The values of exchange field H_E and coercivity H_C at 80 K of a-$Fe_4Ni_{76}B_{20}$ (300 Å)/CoO (250 Å) after cooling from 300 K under various cooling field H_{cool}.

The emerging mechanism for the exchange coupling is as follows. As the bilayer is field cooled across T_N, the induced

M_{PM} in the PM causes the AF spin structure that gives rise to the exchange coupling. In addition to the applied field, the establishment of the AF ordering imposes an effective field on the then PM a-$\mathrm{Fe_4Ni_{76}B_{20}}$. This effective field assists in aligning the PM moments adjacent to the FM/AF interface to be strongly coupled to the AF spin structure, resulting in the unidirectional exchange coupling. It should be noted that the *entire* a-$\mathrm{Fe_4Ni_{76}B_{20}}$ layer contributes to the measured induced M_{PM} in an external field, whereas only the *interfacial* moments in the vicinity of the FM/AF interface are of importance for establishing the exchange coupling. The aforementioned dependence of H_E on H_{cool} indicates that the interfacial moments are aligned more readily than the remaining PM moments because of the proximity with the AF ordering, which provides, in addition to H_{cool}, a strong effective field. These interfacial moments play a crucial role in establishing the exchange coupling when the FM is in the PM state. Once established, the pinned AF spin structure provides an exchange field to the a-$\mathrm{Fe_4Ni_{76}B_{20}}$ layer in both the FM state and the PM state until the AF ordering vanishes at T_N.

As mentioned earlier, H_C acquires much enhanced values in exchange-coupled bilayers.[1–8] While most theoretical efforts have been devoted to addressing H_E, some models of H_C have recently been advanced.[17] As shown in Fig. 5, despite the wide range of values of H_E as a result of different values of H_{cool}, H_C does not depend on H_{cool} at all, hence it is an intrinsic property of the FM layer. However, for the exchange-coupled FM layer, its value (52 Oe) is more than two orders of magnitude larger than that (0.4 Oe) of a single uncoupled FM layer.

Finally, the realization of exchange coupling in bilayers with $T_C \ll T_N$ also has important implications in technological application of exchange coupling in spin-valve devices. For FM/AF bilayers with optimized performance, one can broaden the search to a greater variety of FM and AF materials to realize suitable values of H_E and H_C near room temperature without regard to the condition of $T_C > T_N$.

In summary, contrary to the common perception of $T_C > T_N$ as a prerequisite for exchange coupling between a FM and an AF layers, we have demonstrated exchange coupling where $T_C \ll T_N$. The exchange coupling exists not only in $T < T_C$, but also in $T_C < T < T_N$, where the bulk of the FM layer is in the PM state. The characteristic H_E and H_C vanish at T_N and T_C, respectively. We show that, instead of the requirement of a FM layer, even a modest induced magnetization in the PM layer can cause the AF spin structure that gives rise to exchange coupling. The interfacial moments adjacent to the AF layer are likely to be responsible for establishing the exchange coupling in the paramagnetic state.

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