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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 \( T_C \) < 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.\(^1\) 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 \( (T>C) \). 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 \( (T_N=493 \text{ K}) \), CoO \( (T_N=291 \text{ K}) \), and NiO \( (T_N=525 \text{ K}) \). 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 \( \text{Fe-Ni}_{52}\text{B}_{20} \) 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 \( a-\text{Fe}_{8}\text{Ni}_{72}\text{B}_{20} \) \((T_C \approx 240 \text{ K})\)/AF CoO \((T_N=291 \text{ K})\) system, with the \( T_C \) slightly below the \( T_N \).\(^12\) In this work, we employed \( a-\text{Fe}_{78}\text{Ni}_{12}\text{B}_{20} \) \((T_C \approx 150 \text{ K})\) and CoO to examine features in exchange coupling with \( T_C \approx T_N \).

Amorphous \( \text{Fe}_{72}\text{Ni}_{28}\text{B}_{20} \), 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 that \( T_N \) in CoO, whereas in NiO and FeMn one has the complication that \( T_B \) is much lower than \( T_N \). Single layers of \( a-\text{Fe}_{5}\text{Ni}_{92}\text{B}_{20} \) and bilayers of \( a-\text{Fe}_{9}\text{Ni}_{91}\text{B}_{20} \) \((300 \text{ Å})/\text{CoO} \((250 \text{ Å})\) were fabricated in a multi-source sputtering system with a base pressure of \( 5 \times 10^{-8} \text{ Torr} \) 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 \( a-\text{Fe}_{5}\text{Ni}_{92}\text{B}_{20} \), illustrating \( T_C \approx 150 \text{ K} \), is shown in Fig. 1 for external fields of 5 and 500 Oe. The hysteresis loop of a single \( 300 \text{ Å} \) \( a-\text{Fe}_{5}\text{Ni}_{92}\text{B}_{20} \) 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 \( a-\text{Fe}_{5}\text{Ni}_{92}\text{B}_{20} \) \((300 \text{ Å})/\text{CoO} \((250 \text{ Å})\) 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
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 $\alpha$-Fe$_3$Ni$_{72}$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. 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. 1. Temperature dependence of magnetization of a single $\alpha$-Fe$_3$Ni$_{72}$B$_{20}$ layer at 5 and 500 Oe.

FIG. 2. Hysteresis loops of a single layer $\alpha$-Fe$_3$Ni$_{72}$B$_{20}$ at 80 K (a) and a bilayer of $\alpha$-Fe$_3$Ni$_{72}$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 $\alpha$-Fe$_3$Ni$_{72}$B$_{20}$ (300 Å)/CoO (250 Å) at 80 K (solid symbols) and 180 K (open symbols).

$T<T_C$ 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 $\alpha$-Fe$_3$Ni$_{72}$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.
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$_{9}$Ni$_{72}$B$_{20}$/CoO system where both $H_E$ and $H_C$ show a peak near $T_C$. 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$_{9}$Ni$_{72}$B$_{20}$/CoO and $a$-Fe$_{9}$Ni$_{72}$B$_{20}$/CoO are very different. In $a$-Fe$_{9}$Ni$_{72}$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 $T_N$. In the present $a$-Fe$_{9}$Ni$_{72}$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 $T_N$. 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 $H_{cool}$. 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 $T_C<T_N$, 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. 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 $a$-Fe$_x$Ni$_{76}$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 $a$-Fe$_x$Ni$_{76}$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 $a$-Fe$_x$Ni$_{76}$B$_{20}$ layer in both the FM state and the PM state until the AF ordering vanishes at $T_N$.

As mentioned earlier, $H_E$ acquires much enhanced values in exchange-coupled bilayers. While most theoretical efforts have been devoted to addressing $H_E$, some models of $H_E$ have recently been advanced. As shown in Fig. 5, despite the wide range of values of $H_E$ as a result of different values of $H_{cool}$, $H_E$ 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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$^1$W. H. Meiklejohn and C. P. Bean, Phys. Rev. 102, 1413 (1956); 105, 904 (1957).