

Antiferromagnetic spin flop and exchange bias

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(Received 20 September 1999)

The effect of the antiferromagnetic spin flop on exchange bias has been investigated in antiferromagnetic (MnF₂)-ferromagnetic (Fe) bilayers. Cooling and measuring in fields larger than the antiferromagnetic spin-flop field, H_{SF} , causes an irreversible reduction of the magnitude of the exchange bias field, H_E . This indicates that, contrary to what is normally assumed, the interface spin structure does not remain “frozen in” below T_N if large enough fields are applied.

Exchange bias, H_E , is the shift of the hysteresis loop along the field axis in systems with ferromagnetic (FM)-antiferromagnetic (AFM) interfaces.¹ This shift is induced by a unidirectional exchange anisotropy created if the FM/AFM system is cooled (or grown) in a static magnetic field below the Néel temperature of the AFM.¹⁻⁵ Despite the technological interest in these structures for spin valve devices⁶ there is little basic understanding of the phenomenon. From the theoretical point of view, theories based on AFM domains,⁷⁻¹¹ spin waves,¹² FM domains,¹³⁻¹⁵ or uncompensated interface spins¹⁶ have been proposed to explain exchange bias quantitatively. Also, recent experimental studies aimed at understanding basic phenomena, such as 90° FM-AFM coupling,^{17,18} the role of the FM transition temperature, T_C ,¹⁹ memory effects,^{20,21} reversible techniques to measure H_E ,^{15,22,23} or artificial AFM-FM systems,²⁴ have been reported. Of special interest, both theoretically and experimentally, is the interface spin structure.¹ However, although it probably controls exchange bias, little is known about it. For example, it is commonly assumed that the AFM spin structure at the interface, due to the AFM/FM exchange coupling, is “frozen in” when crossing the AFM Néel temperature, T_N .¹ Therefore, the exchange bias field, for AFM samples with large grains, usually remains independent of the number of flux reversals, i.e., no *training effect* is observed.¹ Moreover, it has been shown that large cooling fields can affect the exchange bias field.²⁵⁻²⁹ For example, FeF₂-Fe and MnF₂-Fe bilayers cooled in large fields exhibit hysteresis loops that shift in the direction of the applied field (i.e., *positive* exchange bias),²⁵⁻²⁷ contrary to what is observed in most systems. Thus, the interface spin structure can be modified with the cooling field in some systems. However, this new

spin structure also remains “frozen” below T_N , i.e., positive exchange bias systems do not exhibit training effects.^{25,27}

If an AFM bulk system is subject to a very large magnetic field applied along its anisotropy axis there is a threshold field above which having the spin sublattices parallel and antiparallel to the applied field is energetically unfavorable. Thus, the spins “flop” to a configuration where both sublattices are perpendicular to the applied field.³⁰ However, if the field is applied away from the AFM anisotropy axis the effect is drastically reduced.³¹ This transition, denoted *spin flop* (SF), has been extensively studied in MnF₂ single crystals^{32,33} amongst other AFM's.³⁰

In this paper we discuss the effect of the AFM spin flop in exchange biased MnF₂-Fe bilayers. We observe that crossing the AFM spin-flop field, H_{SF} , both in cooling and measuring below T_N has a strong, irreversible, effect in exchange bias, mainly reducing the magnitude of H_E . The results indicate that contrary to what is commonly assumed, the “frozen in” interface spin structure can be changed *irreversibly* below T_N when crossing the SF-AFM phase boundary.

The spin-flop phase diagram of MnF₂,³² is shown schematically in Fig. 1. As can be observed in the figure, when lowering the temperature from above T_N , MnF₂ exhibits a paramagnetic (PM)-AFM transition in the field range $0 < H \lesssim 120$ kOe. For larger fields, the transition becomes PM-SF. Well below T_N , MnF₂ undergoes two transitions with increasing field, from AFM to SF and at larger fields from SF to PM. For example, at $T=10$ K the AFM-SF transition occurs at $H_{AFM-SF} \approx 90$ kOe, while $H_{AFM-SF} \approx 110$ kOe and $H_{AFM-SF} \approx 120$ kOe for $T=50$ K and $T=61$ K, respectively. The SF-PM transitions take place at much larger fields (not shown in Fig. 1). In this study we performed two types of experiments: (i) field cooling experiments, consist-

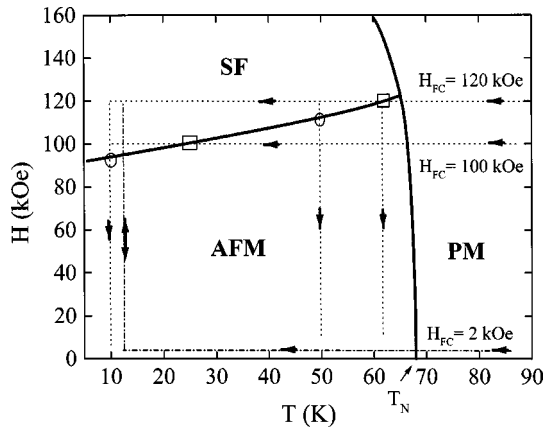


FIG. 1. Schematic field versus temperature phase diagram for MnF_2 single crystals (Ref. 32), where PM, AFM and SF correspond to the paramagnetic, antiferromagnetic and spin-flop phases, respectively. The dotted lines show different cooling paths ($H_{FC} = 100$ kOe and $H_{FC} = 120$ kOe), where the squares indicate the AFM-SF transition, while the circles show the SF-AFM transition in the field cooling procedure.

ing of cooling in different fields, H_{FC} , from above T_N , to the measuring temperature ($T = 10, 50$, and 61 K). When the measuring temperature is reached, the field is reduced to $H = 0.6$ kOe and a hysteresis loop with maximum applied field of $H_{max} = \pm 0.6$ kOe is carried out; (ii) maximum applied field experiments, in which the sample is cooled in a small field, $H_{FC} = 2$ kOe, to $T = 10$ K, where consecutive hysteresis loops of increasing maximum applied field H_{max} are carried out. In the field cooling experiment we follow different horizontal dotted lines (cooling paths) in Fig. 1 until the measuring temperature is reached, after which we follow the vertical dotted line to $H = 0.6$ kOe. Thus, depending on H_{FC} and T we cross different phase transition lines. For example, for $H_{FC} = 100$ kOe we cross the PM-AFM line at $T \approx 67$ K while at $T \approx 25$ K we cross the AFM-SF line (shown with a square in Fig. 1). Finally, at certain temperatures, when reducing the field we cross again the SF-AFM line (shown with a circle in Fig. 1). Note that depending on H_{FC} and T we either cross only the PM-AFM phase line (e.g., $H_{FC} = 100$ kOe for $T = 50$ and 61 K and $H_{FC} = 120$ kOe for $T = 61$ K in Fig. 1) or the PM-AFM, AFM-SF, and SF-AFM lines (e.g., $H_{FC} = 120$ kOe and 10 kOe for $T = 10$ K and $H_{FC} = 120$ kOe for $T = 50$ K in Fig. 1). In the maximum applied field experiment we follow the horizontal dash-dot line in the cooling procedure, crossing the PM-AFM line. At $T = 10$ K we follow the vertical dash-dot line. Note that depending on the maximum applied field, we either remain in the AFM phase or cross the AFM-SF phase line.

The growth of the MnF_2 -Fe bilayers on $\text{MgO}(100)$ has been described elsewhere.²⁷ Briefly, the different layers were grown by sequential *e*-beam evaporation, MnF_2 (65 nm at a rate of 0.2 nm/s) at $T_S = 300^\circ\text{C}$ and Fe (14 nm at a rate of 0.1 nm/s) grown at $T_S = 150^\circ\text{C}$. A buffer layer of ZnF_2 (25 nm at a rate of 0.2 nm/s) was grown at $T_S = 200^\circ\text{C}$ to improve the crystallinity of the AFM layer. Finally, the bilayers were capped by 3 nm of Al at a rate of 0.05 nm/s at $T_S = 150^\circ\text{C}$, to prevent oxidation. The MnF_2 layer grows ‘‘quasiepitaxially’’ (twinned) in the (110) direction with a rocking curve width of about 2° , while the Fe layer is poly-

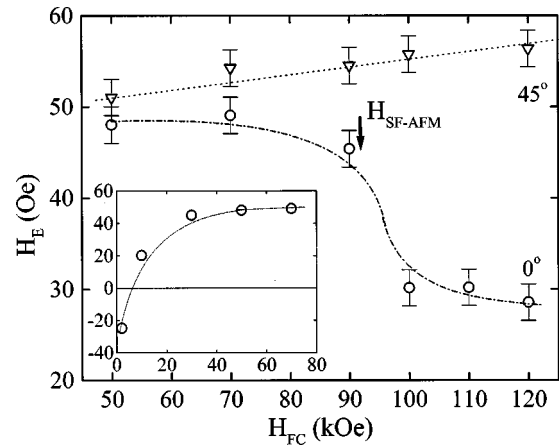


FIG. 2. Dependence of the exchange bias field, H_E , on the cooling field, H_{FC} , at $T = 10$ K when cooling along 0° (\circ) and 45° (∇) for large cooling fields. The inset shows the dependence of H_E on H_{FC} at $T = 10$ K when cooling along 0° for small fields. The lines are guides to the eye.

crystalline. Note that we will refer to the 0° direction as the AFM anisotropy axis ($\langle 001 \rangle$) and its corresponding twin, while the 45° direction is the one at 45° of the AFM anisotropy axis and its corresponding twin.

The magnetic measurements were carried out using vibrating sample VSM ($H_{max} = 120$ kOe) and superconducting quantum interference device (SQUID) ($H_{max} = 70$ kOe) magnetometers. The samples are cooled from 150 K [i.e., above $T_N(\text{MnF}_2) = 67.3$ K] to the measuring temperature in the presence of different cooling fields ($H_{FC} = 0.10$ – 120 kOe) along the 0° or 45° directions. Hysteresis loops were measured at several temperatures with different maximum applied fields in the range $H_{max} = 0.6$ – 120 kOe. Note that the remanent fields of both apparatus were carefully measured and corrected *a posteriori*.

We should point out that in some systems magnetization measurements only give a lower limit of the interfacial coupling.¹⁵ The strong anisotropy in the AFM and the absence of training effects imply this is not the case here.

For the field cooling experiment, at $T = 10$ K, H_E exhibits a strong dependence on the cooling field, H_{FC} , for moderate cooling fields ($H_{FC} < 70$ kOe) applied along the 0° direction, as can be seen in the inset of Fig. 2. H_E changes monotonically from negative exchange bias to positive exchange bias (for $H_{FC} > 10$ kOe), similar to what is observed when cooling along 45° .²⁷ However, if H_{FC} along 0° , exceeds 90 kOe, the magnitude of H_E exhibits a sharp reduction (Fig. 2). Moreover, if the cooling field is applied along 45° , going beyond cooling fields of $H_{FC} = 90$ kOe has no evident effect (Fig. 2). As shown in Fig. 3, if the sample is cooled along 0° to $T = 50$ K (instead of to $T = 10$ K) in different H_{FC} , H_E increases steadily up to $H_{FC} = 110$ kOe, while showing a decrease in H_E for $H_{FC} = 120$ kOe. In turn, if the sample is cooled along 0° to $T = 61$ K in different H_{FC} , H_E exhibits no anomaly (Fig. 3) up to $H_{FC} = 120$ kOe. Looking at the spin-flop phase diagram (Fig. 1), at $T = 10$ K the spin-flop field is about $H_{SF} \approx 90$ kOe, coinciding with the onset of the downturn of H_E at $T = 10$ K (see Fig. 2). Moreover, $H_{SF}(T = 50$ K) ≈ 110 kOe, consequently the step in $H_E(H_{FC})$ occurs for

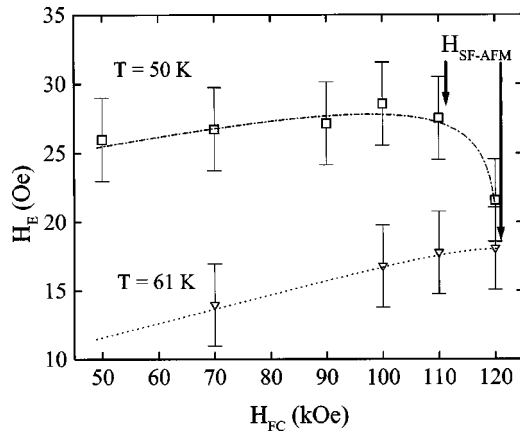


FIG. 3. Dependence of the exchange bias field, H_E , on the cooling field, H_{FC} , along 0° to $T=50$ K (\square) or $T=61$ K (∇). The lines are guides to the eye.

$H_{FC} > 110$ kOe (see Fig. 3). Similarly, $H_{SF}(T=61$ K) ≈ 120 kOe, thus no anomaly in $H_E(H_{FC})$ is observed when the sample is cooled to $T=61$ K (see Fig. 3). If a new spin structure was “frozen in” for large H_{FC} , one would expect changes in H_E at each measuring temperature. However, as shown in Figs. 2 and 3 for $H_{FC}=100$ or 110 kOe there is only a reduction in H_E for $T=10$ K. This indicates that the step in $H_E(H_{FC})$ is *not* induced at T_N , but when crossing the SF-AFM line at the measuring temperature (see circles in Fig. 1). The results also indicate that contrary to the AFM spin-flop transition for single crystals, the AFM spin flop probed by exchange bias is *not* reversible. In summary, the AFM (or FM) spin structure at the interface does *not* remain frozen below T_N , but changes irreversibly when crossing the AFM-SF phase boundary.

It could be argued that crossing the AFM-SF line at high temperatures (square in Fig. 1) and then crossing the SF-AFM line at low temperatures (circle in Fig. 1) are not equivalent, and this could induce irreversibility. Thus, we carried out the maximum applied field experiment.

Figure 4 shows that after cooling in a small field ($H_{FC}=2$ kOe) to $T=10$ K the exchange bias, H_E , is independent of the maximum field reached during the measurements of the hysteresis loops up to $H_{max}=80$ kOe. For larger maximum applied fields the magnitude of H_E decreases to about half. If after a $H_{max}=70$ kOe ($H < H_{SF}$) hysteresis loop a $H_{max}=0.6$ kOe is sequentially measured, H_E displays no change. However, if one carries out a $H_{max}=0.6$ kOe hysteresis loop after a $H_{max}=120$ kOe ($H > H_{SF}$) one, H_E is almost zero, i.e., drastically different from both the original $H_{max}=0.6$ kOe ($H_E \approx -37$ Oe) and $H_{max}=120$ kOe ($H_E \approx -20$ Oe) loops. Hence, as shown in Fig. 4, *irreversibility* is also found when carrying out hysteresis loops of increasing maximum field. In this case, the SF-AFM (AFM-SF) line is crossed at the same temperature. It is noteworthy that the coercivity, H_C , shows only small anomalies (usually within the experimental error) where H_E shows discontinuities. Some of these experiments were also carried out for FeF_2/Fe bilayers [at $T=10$ K $H_{SF}(\text{FeF}_2)=400$ kOe]. None of the experiments showed any anomaly for cooling or measuring fields in the range 50 kOe $< H_{FC} < 120$ kOe.

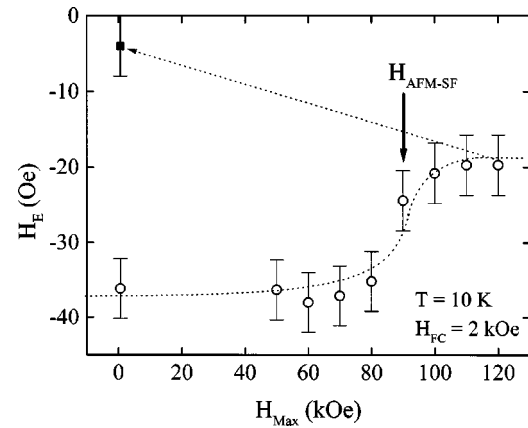


FIG. 4. Dependence of the exchange bias field, H_E , on the maximum field of the hysteresis loops, H_{max} , after cooling to $T=10$ K in $H_{FC}=2$ kOe along 0° . The arrow shows the change in exchange bias field, H_E , for a $H_{max}=0.6$ kOe (solid symbol) hysteresis loop measured after the $H_{max}=120$ kOe hysteresis loop. The lines are guides to the eye.

The lack of discontinuities in $H_E(H_{FC})$ or $H_E(H_{max})$ when cooling or measuring along 45° for $\text{MnF}_2\text{-Fe}$ or when cooling along 0° for $\text{FeF}_2\text{-Fe}$ confirms once more the spin flop to be the origin of the $\text{MnF}_2\text{-Fe}$ results. If the cooling or measuring field is applied away from the AFM anisotropy axis one would not expect to have a spin-flop transition, consequently there should be no anomaly in H_E . Moreover, due to its large anisotropy, FeF_2 has its spin-flop field, $H_{SF}=400$ kOe, thus $H=120$ kOe should not affect H_E , as observed.

These results indicate that spin-flop transitions can be easily studied in AFM thin films using exchange bias. The sensitivity of most other techniques would become inadequate when the AFM layers become very thin. However, exchange bias relies on the magnetization of the FM layer in a FM/AFM couple, thus, in principle, this technique should work independently of the AFM layer thickness.

The origin of the irreversibilities in H_E is more puzzling because the AFM-SF-AFM transitions are reversible in AFM single crystals.³¹ However, although the SF transition is *macroscopically* reversible, it may be *microscopically* irreversible. This microscopic irreversibility may give us a clue on possible mechanisms for the behavior of H_E . Following Malozemoff’s model,⁷ if the AFM layer breaks up into domains and the size of these domains is larger when crossing the SF-AFM boundary than when crossing the paramagnetic-AFM boundary, this could lead to a reduction of H_E . Also, if crossing the SF-AFM phase boundary changes the orientation of the AFM interface spins due to the different interaction between the AFM and FM spins before crossing the different phase boundaries, this could lead to (a) a change in the interface coupling, which based on Koon’s⁹ model could reduce H_E , or (b) a reduction in the number of uncompensated AFM interface spins (e.g., by a change in AFM domain size) which according to the model of Takano *et al.*¹⁶ should reduce H_E . Finally, following the model of Kiwi *et al.*,^{13,15} the changes in the AFM spin structure in the SF phase would modify the FM spin structure at the interface which in turn

would affect H_E . Note that the last case does not really assume an irreversible change of the AFM spin structure but an irreversible change of the FM interface structure.

In conclusion, we have observed that the effect of cooling and measuring in fields larger than the AFM spin-flop field in exchange biased AFM(MnF_2)-FM(Fe) bilayers is to reduce H_E irreversibly. The results can be qualitatively explained using some exchange bias models if an irreversible

change of the interface spin structure is assumed. Note that these results can be interpreted as exchange bias being a “minor” loop effect, i.e., exchange bias systems should only exhibit reversible loop shifts, H_E , for fields smaller than the AFM spin-flop field. This could be particularly relevant for epitaxial or single-crystal AFM's with low anisotropies.

This work was supported by the U.S. DOE. J.N. thanks the Spanish Government for its financial support.

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