Simple Mechanism for a Positive Exchange Bias

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Abstract

We argue that the interface coupling, responsible for the positive exchange bias ($H_E$) observed in ferromagnetic/compensated antiferromagnetic (FM/AF) bilayers, favors an antiferromagnetic alignment. At low cooling field this coupling polarizes the AF spins close to the interface, which spin configuration persists after the sample is cooled below the Néel temperature. This pins the FM spins as in Bean’s model and gives rise to a negative $H_E$. When the cooling field increases, it eventually dominates and polarizes the AF spins in an opposite direction to the low field one. This results in a positive $H_E$. The size of $H_E$ and the crossover cooling field are estimated. We explain why $H_E$ is mostly positive for an AF single crystal, and discuss the role of interface roughness on the magnitude of $H_E$, and the quantum aspect of the interface coupling.

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In recent experiments by Schuller et al., a ferromagnetic (FM) film is grown on a compensated antiferromagnetic (AF) surface under large cooling field, and the hysteresis loop is observed to shift along the positive side of the field axis. This phenomena belongs to the general category of exchange anisotropy, first discovered more than 40 years ago by Meiklejohn and Bean. However, different from the original observation and later theories, a compensated (i.e., no net magnetization) surface was used and the sign of the bias was unexpected. The compensated part is resolved by a recent micromagnetic calculation by Koon, but the sign remains only speculations. Without knowledge of the detail structure at the interface or confirmation of the existence of AF domains, we try to build a simple intuitive theory. It not only can explain the main features of the phenomena, but also gives reasonable quantitative estimations. Quantum aspect of the interface coupling is analyzed in the second half of the paper.

Experimentally the exchange bias, $H_E$, decreases with increasing temperature and vanishes at the Néel temperature. This shows that the coupling of the FM spins to the ordered AF spins is crucial for the exchange bias. Furthermore, the plot of $\ln H_E$ v.s. $\ln t_F$ is found to fit nicely by a straight line with slope $= -1$ where $t_F$ denotes the thickness of the FM film. This can be viewed as another support to concentrate on the interface coupling for the source of exchange bias. The interpretation is based on, if the F spins at the interface (of number $N$) are stabilized each by an energy, $E$, due to their coupling to the AF spins, we need to divide the total change, $NE$, by the total number of FM spins in the film, $\approx Nt_F$, when converting to the shift in the hysteresis loop. This gives $H_E \approx E/t_F$ which explains the linearity and its slope in the ln-ln plot. Interface roughness will increase the interface area (while the total spin number remains unchanged) and introduce an extra factor $\alpha > 1$ into $H_E \approx \alpha E/t_F$. However, when the easy axes of FM and AF are parallel, surface roughness may also introduce frustrations (see Fig. 1) which will diminish the coupling. This does not happen when the easy axes are perpendicular since there is no preferred direction for any FM spin from its neighboring AF spins. We shall distinguish these two orientations, parallel/perpendicular easy axes, and assign them separately to the negative/positive $H_E$.
cases. Such a 90 degree rotation of the FM easy axis for Fe/(110)FeF$_2$ single crystal due to the AF ordering was indeed observed by examining the hysteresis loops. That is, the easy axis of FM spins, originally in the (001) direction at $T = 300$ K, rotates to (1 $\bar{1}$0) at $T = 10$ K for which a positive $H_E$ was measured.

For our theory, the interface coupling, $\sum J_c \vec{S}_F \cdot \vec{S}_{AF}$, is assumed to favor an antiferromagnetic alignment with $J_c \approx J_{AF}$ (the coupling constant between AF spins). This will be justified if, take Fe/FeF$_2$ for instance, the fluoric ions happen to lie at the interface and mediate the coupling between neighboring irons from either sides (the superexchange mechanism). However, if the irons across the interface build a direct chemical bond, presumably their coupling will of the same order and sign as $J_F$ in the bulk Fe. But since $J_F$ is twenty times stronger than $J_{AF}$, this $\vec{S}_{AF}$ will be locked rigidly parallel to $\vec{S}_F$ and can be treated as an extension of the ferromagnetic film. The relevant interface will now be between this first layer and the next layer of the antiferromagnetic, which of course favors an antiferromagnetic alignment and agrees with our assumption.

At low cooling field for which $H_E$ is negative, the easy axes of FM and AF spins are assumed to be parallel. Using the mean field analysis, we estimate the deviation from the positive $z$-axis (which results in a nonzero magnetization for AF) of each spin-up $\vec{S}_{AF}$ at the interface due to its antiferromagnetic coupling with a spin-up $\vec{S}_F$ neighbor is of the order of $1 - \tanh [(J_{AF} \cdot q - J_c)/k_B T]$ where $q$ is the number of nearest neighbors for each $\vec{S}_{AF}$ at the surface. At the usual operating temperature, say $T = 10$ K, the magnetization is approximately $-2 \cdot \exp [- (J_{AF} \cdot q - J_c)/k_B T]$. To obtain the total energy change for the system, we need to multiply it by $J_c$ and $N/2$ (number of up-spin $\vec{S}_{AF}$ at the interface). Note that this magnetization points antiparallel to the FM spins.

When the cooling field is large enough to cause a positive $H_E$, we assume that the easy axis of FM spins rotates and becomes perpendicular to the AF easy axis. Different from the previous case, polarization of the AF spins is now mainly due to the cooling field and, not just those $\vec{S}_{AF}$ at the interface but, all spins are involved. A physical justification for making such a rotation may lie in the fact that the perpendicular magnetic susceptibility
of AF spins \((\approx 1/J_{AF})\) is much larger than the parallel one at low temperatures. By canting the AF spins more effectively towards the field direction the system can gain more energy from the Zeeman effect. Note that the polarization here points parallel to the external field, i.e., the easy axis of FM spins, and is opposite to that caused by the interface coupling.

We can estimate the minimum strength of cooling field, \(H_{\text{cool}}\), required to obtain a positive magnetization by comparing these two energy changes:

\[
\frac{H_{\text{cool}}^2}{J_{AF}} \cdot t_{AF} N = J_c e^{-(J_{AF} \cdot q - J_c)/k_B T} \cdot N
\]

This gives \(H_{\text{cool}} \approx 0.2\ T\) for \(t_{AF} = 90\ nm\), the right magnitude to cause the sign change of \(H_E\) experimentally. Had the easy axes been perpendicular at low cooling field, RHS of the above equation would become \(J_c^2 N/J_{AF}\) and give too high a threshold field \(H_{\text{cool}} \approx 5\ T\). The thickness \(t_{AF}\) becomes very large for a single crystal, which implies an easier entrance into the positive-\(H_E\) scenario. This is again consistent with observations that \(H_E\) is mostly positive when an AF single crystal is used (the fact that its surface is much rougher than in films also contributes). The appearance of an exchange bias due to the locking of FM spins by the polarization is the same as in Bean’s original model, except that an uncompensated AF surface is not required here and \(H_E\) can become positive when the cooling field is strong. We do not know how the polarization survives below the Néel temperature. This could be the place where possible AF domains or impurities need to be introduced. Experimental evidence for this “memory” is found when putting samples, field cooled in 2 kOe, under 70 kOe magnetic field at low temperatures (10K). \(H_E\) is found to remain unchanged to within 5% of the \(H_{\text{cool}} = 2\ kOe\) value.

Aside from possible instability due to finite temperature fluctuations, the main conclusion of Koon that FM orders perpendicular to the AF easy magnetization axis was checked to be correct by Kiwi using a Monte-Carlo calculation. We shall examine the validity of this conclusion against a full quantum mechanical treatment, i.e., we analyze the change of vacuum energy, \(E_{\text{vac}}\), due to the virtual process of FM spins emitting and reabsorbing AF spin waves via the interface coupling. Suhl and Schuller have considered the special case
when the FM and AF easy axes are parallel, and found a negative $E_{\text{vac}}$. We extend their calculations to a general angle, $\phi$, between these two easy axes (see Fig.2) in order to find the most stable spin orientation.

Quantum mechanically the interface coupling, $\sum J_c \vec{S}_F \cdot \vec{S}_{AF}$, where the summation runs over all sites at the interface, can be decomposed into raising and lowering operators as $[S_F^+ S_{AF}^- + S_F^- S_{AF}^+]/2 + S_F^z S_{AF}^z$. Since the easy axis of $\vec{S}_F$ is now in the $(0, \sin \phi, \cos \phi)$ direction, we need to reexpress $S_F^z$ and $S_F^\pm$ in terms of the new projection and raising/lowering operators:

$$P^z \equiv S_F^u \sin \phi + S_F^u \cos \phi$$

$$P^\pm \equiv S_F^\pm \pm i(S_F^u \cos \phi - S_F^u \sin \phi).$$

(2)

In the mean time follow the standard spin wave derivation in rewriting the AF spin operators in terms of boson operators $a^+$ and $a$ which create and destroy spin deviations,

$$S_i^x \approx \sqrt{s^2/2} (a_i + a_i^+),$$

$$S_i^y \approx -i \sigma_i \sqrt{s^2/2} (a_i - a_i^+),$$

$$S_i^z = \sigma_i (s - a_i^+ a_i),$$

(3)

where $l$ is the site label and $\sigma_i = 1/-1$ at the spin-up/down $\vec{S}_{AF}$ site, and $s/S$ is the size of the AF/FM spin. Since there is no confusion now between the different spin notations, we shall drop the subscripts $F$ and $AF$ from now on.

The interface coupling becomes $J_c$ times

$$\sum_u \left[ P_u^z \cos \phi - \frac{P_u^+ - P_u^-}{2i} \sin \phi \right] (s - a_u^+ a_u) - \sum_d \left[ P_d^z \cos \phi - \frac{P_d^+ - P_d^-}{2i} \sin \phi \right] (s - a_d^+ a_d)$$

$$+ \sqrt{s^2/2} \sum_u \left\{ \left[ \frac{P_u^+ + P_u^-}{2} + i P_u^z \sin \phi + \frac{P_u^+ - P_u^-}{2} \cos \phi \right] a_u^+ 
+ \left[ \frac{P_u^+ + P_u^-}{2} - i P_u^z \sin \phi - \frac{P_u^+ - P_u^-}{2} \cos \phi \right] a_u \right\}$$

$$+ \sqrt{s^2/2} \sum_d \left\{ \left[ \frac{P_d^+ + P_d^-}{2} + i P_d^z \sin \phi + \frac{P_d^+ - P_d^-}{2} \cos \phi \right] a_d^+ 
+ \left[ \frac{P_d^+ + P_d^-}{2} - i P_d^z \sin \phi - \frac{P_d^+ - P_d^-}{2} \cos \phi \right] a_d \right\}$$

(4)
where the subscript \( u/d \) denotes the site with or neighboring a down/up AF spin (see Fig. 2). The fact that the ordering temperature of FM (\( \approx 770 \) K) is much higher than that of AF (\( \approx 78 \) K) allows us to assume that the FM spins are very much rigid while the AF spins deviate. In the ground state, the brackets \( s - a_u^+ a_u \) and \( s - a_d^+ a_d \) have the same values, and the first and the second terms of Eq. (1) cancel. Now expressing \( a_{u,d}^+ \) and \( a_{u,d} \) in terms of their Fourier conjugates \( a_k^\pm \) and \( a_k \), and then Bogoliubov transforming \( \mathbf{A} \) to the AF spin wave operators \( b_k \) and \( b_k^\pm \):

\[
\begin{align*}
  b_k &= a_k \cosh \theta_k + c_{-k}^\pm \sinh \theta_k, \\
  b_k^\pm &= a_{k,\pm} \cosh \theta_k + c_{-k}^\pm \sinh \theta_k, \\
  b_{-k} &= a_{k,\pm} \sinh \theta_k + c_{-k}^\pm \cosh \theta_k, \\
  b_{-k}^\pm &= a_k \sinh \theta_k + c_{-k}^\pm \cosh \theta_k.
\end{align*}
\]

(5)

(6)

Eq. (4) becomes

\[
J_c \sqrt{\frac{s}{2N}} \sum_k \left( q_k b_k^\pm + q_k^\dagger b_k \right)
\]

(7)

where

\[
q_k \equiv \sum_u \left\{ \frac{P_u^+ + P_u^-}{2} + iP_u^z \sin \phi + \frac{P_u^+ - P_u^-}{2} \cos \phi \right\} \cosh \theta_k \cdot e^{iku} \\
+ \left\{ \frac{P_u^+ + P_u^-}{2} - iP_u^z \sin \phi - \frac{P_u^+ - P_u^-}{2} \cos \phi \right\} \sinh \theta_k \cdot e^{-iku} \\
+ \sum_d \left\{ \frac{P_d^+ + P_d^-}{2} + iP_d^z \sin \phi + \frac{P_d^+ - P_d^-}{2} \cos \phi \right\} \sinh \theta_k \cdot e^{-ikd} \\
+ \left\{ \frac{P_d^+ + P_d^-}{2} - iP_d^z \sin \phi - \frac{P_d^+ - P_d^-}{2} \cos \phi \right\} \cosh \theta_k \cdot e^{ikd} \right\}. 
\]

(8)

We can redefine \( b_k \) and \( b_k^\pm \) to eliminate the linear terms in Eq. (7). This shifts the vacuum energy of the antiferromagnetic Hamiltonian, \( \sum_k \omega_k b_k^\dagger b_k \), by \( E_{\text{vac}} = -(sJ_c^2/2N) \sum_k \frac{q_k^2}{\omega_k} \). The summation can be written out as

\[
\begin{align*}
  \sum_k \frac{1}{\omega_k} \left\{ \sum_u \frac{P_u^+ P_u^-}{4} \left[ (1 + \cos \phi)^2 \cosh^2 \theta_k + (1 - \cos \phi)^2 \sinh^2 \theta_k + \sin^2 \phi \sinh 2\theta_k \cos 2ku \right] \\
  + \sum_d \frac{P_d^+ P_d^-}{4} \left[ (1 + \cos \phi)^2 \sinh^2 \theta_k + (1 - \cos \phi)^2 \cosh^2 \theta_k + \sin^2 \phi \sinh 2\theta_k \cos 2kd \right] \\
  + \sin^2 \phi \left[ \sum_u \left( \frac{P_u^z}{2} \right)^2 \left( \cosh 2\theta_k - \sinh 2\theta_k \cos 2ku \right) + \sum_d \left( \frac{P_d^z}{2} \right)^2 \left( \cosh 2\theta_k - \sinh 2\theta_k \cos 2kd \right) \right] \right\}
\end{align*}
\]

(9)
where cross terms $P^+_u P^-_d$ and products of two raising or lowering operators have been neglected since we are averaging over the ground state. According to the completeness relation, $\sum_u \cos 2ku + \sum_d \cos 2kd$ is only nonzero when $k = 0$ or $\pi$ (setting the lattice constant to be unity). $k = 0$ mode is neglected on physical ground since it involves translationally moving the whole sample and is not what we expect. Substituting the groundstate expectation values of $\langle P^+_u P^-_d \rangle = 2S$ and $\langle (P^z_{u/d})^2 \rangle = S^2$ into Eq. (9) gives

$$E_{\text{vac}} = -\frac{sJ^2}{2} \left\{ \left[ S \left( 1 + \cos^2 \phi \right) + S^2 \sin^2 \phi \right] \sum_k \frac{\cosh 2\theta_k}{\omega_k} + \left( \frac{S}{2} - S^2 \right) \sin^2 \phi \frac{\sinh 2\theta_{\pi}}{\omega_{\pi}} \right\}$$

(10)

The summation of $\cosh 2\theta/\omega$ is of the order of $\ln (J_{AF}/H_A)/J_{AF}$, while $\sinh 2\theta_{\pi}/\omega_{\pi} \approx 1/H_A$ where $H_A$ is the anisotropic field for the AF spins. Normally, $H_A$ is of the order of a few hundred gauss and much smaller than $J_{AF}$, and so we expect the second term in Eq. (10) to dominate as long as $\phi \neq 0$ or $\pi$. When $S$ is bigger than $1/2$ (experimentally $S = 2$), $E_{\text{vac}}$ becomes positive. This means that the system is more stable when the FM/AF easy axes are parallel, compared to being perpendicular, which is opposite to the conclusion from micromagnetic calculations. Of course, the inclusion of anisotropy field on both sides, finite temperature, and couplings in further layers are necessary to determine the final preference. But at least the above calculations show that the quantum nature of the interface coupling, neglected in former classical treatments, may reverse their conclusions and should be properly taken into account.

In conclusion, we have presented a simple mechanism to explain why the sign of exchange bias, $H_E$, changes from negative to positive at high cooling field for the FM/compensated AF bilayers. We believe that the negative/positive bias is realized when the easy axes of FM and AF are parallel/perpendicular. And the time reversal symmetry is broken by the polarization induced by the interface coupling/cooling field respectively. We can explain why the surface roughness enhances $H_E$ when it is positive, while diminishes its magnitude when negative. We also estimate the right size of $H_E$ and the crossover cooling field. Quantum nature of the interface coupling is shown to give an opposite preference of spin alignments to former classical treatments.
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FIGURES

FIG. 1. Roughness introduces frustration to the ferromagnetic spin when its neighbouring spins are in opposite directions.

FIG. 2. Interface between the ferromagnetic and fully compensated antiferromagnetic layers modeled in the text. The cross/dot symbol denotes down/up spins. The $z$-axis is defined to be along the AF easy axis, and FM spins point in $(0, \sin \phi, \cos \phi)$ direction.
Fig. 1, Hong
Fig. 2, Hong