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Correlation between antiferromagnetic interface coupling and positive exchange bias

J. Nogués
Departament de Física, Universitat Autònoma de Barcelona, 08193 Bellaterra, Spain
and Department of Physics 0319, University of California–San Diego, La Jolla, California 92093-0319

C. Leighton and Ivan K. Schuller
Department of Physics 0319, University of California–San Diego, La Jolla, California 92093-0319

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The induced moment in antiferromagnetic (AFM)–ferromagnetic (FM) (FeF_2-Fe and MnF_2-Fe) bilayers has been studied from the shift along the *magnetization axis* of the exchange-biased hysteresis loops. The magnetization shift depends strongly on the cooling field and microstructure of the AFM layer. The shift for small cooling fields can be opposite to the cooling field, indicating that, in some cases, the presence of the FM layer induces an *antiferromagnetic* coupling at the interface. Samples with negative magnetization shifts (*antiferromagnetic* coupling) exhibit large changes in exchange bias H_E as a function of cooling field and *positive* exchange bias. Samples with positive magnetization shifts (*ferromagnetic* coupling) show almost no change in H_E with cooling field and the exchange bias field remains always negative. These results confirm the theoretical assumption that an *antiferromagnetic* interface coupling is necessary to observe *positive* exchange bias.

Exchange bias 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] As shown earlier, large cooling fields affect the exchange bias field in unexpected and interesting ways.[6–11] For example, FeF_2-Fe and MnF_2-Fe bilayers cooled in a large field exhibit hysteresis loop shifts in the direction of the applied field (i.e., *positive* exchange bias),[6,10,11] contrary to what is observed in most systems. It was proposed that a necessary condition for *positive* H_E is an antiferromagnetic coupling at the FM–AFM interface.[6,10–15] Unfortunately, a direct experimental observation of this coupling is difficult. Different exchange bias theories predict the formation of AFM domains,[16–18] canting of the AFM spins,[12] uncompensated spins in the AFM,[19] or the formation of domains in the FM.[20,21] In addition, FeF_2 and MnF_2 are piezomagnetic.[10,22,23] Most of these effects should result in a ''net'' moment in the AFM or FM layers. In this paper, we discuss the relationship between this induced moment and interface coupling. We show that if the coupling is *antiferromagnetic*, the sample exhibits positive exchange bias for large cooling fields, as proposed theoretically.[6,12,14,15,20]

To obtain a range of interface couplings, a number of FeF_2-Fe and MnF_2-Fe bilayers were grown at different substrate temperatures ($T_S=200$–$375\,°C$) and different AFM thicknesses ($t_{AFM}=50$-200 nm).[6,11,24] The growth of the FeF_2-Fe and MnF_2-Fe bilayers on MgO(100) has been described elsewhere.[11,24] Briefly, the different layers were grown by sequential *e*-beam evaporation of FeF_2 or MnF_2 (t_{AFM} at a rate of 0.2 nm/s) and Fe (15 nm at a rate of 0.1 nm/s). In the case of MnF_2 a buffer layer of ZnF_2 was grown at $T_S=200\,°C$ to improve the crystallinity of the AFM layer. The AFM layers were grown at a substrate temperature in the range $T_S=200$–$300\,°C$ (FeF_2) or $T_S=275$–$375\,°C$ (MnF_2), while the Fe layers were always grown at $T_S=150\,°C$. Finally, the bilayers were capped by 5 nm of Al at a rate of 0.05 nm/s at $T_S=150\,°C$, to prevent oxidation.

The magnetic measurements were carried out using a superconducting quantum interference device magnetometer. The samples are cooled from 120 K [above $T_N(FeF_2)=78.4$ K and $T_N(MnF_2)=67.3$ K] to 10 K in the presence of various cooling fields ($H_{FC}=0.1$–70 kOe). Hysteresis loops between ±10 kOe were measured at $T=10$ K after each field cooling. To compare with other systems the exchange bias is given as an interface energy, $H_E t_{FM} M_{FM}$, where t_{FM} and M_{FM} are the thickness and the saturation magnetization of the FM layer, respectively. The net induced moment is obtained from high accuracy (±10 kOe) magnetization measurements with the FM layer fully saturated [$H_C(T=10\ K)\sim0.1$ kOe].

Generally, the absolute values of the magnetic moments at $H=-10$ kOe and $H=+10$ kOe, for the same cooling field, are different, i.e., the loop is shifted in the magnetization axis. Note that with the present data we cannot determine if the origin of the induced moment is a surface (interface) or a bulk effect in the AFM or FM layers, i.e., it is not clear what normalization to use m/A (moment per area), m/V_{AFM} (moment per AFM volume) or m/V_{FM} (moment per FM volume). For illustrative purposes we have assumed that the induced moment is an AFM volume effect, thus $M_{Shift}\equiv m/V_{AFM}$. This ''upwards'' or ''downwards'' shift of the hysteresis loop (M_{Shift}) has a maximum of about 1% of the total magnetization for FeF_2-Fe.

It is noteworthy that uncompensated moments have been found in CoO layers.[19,25] However, for sufficiently high fields, these spins are found to align with the applied field,[25] thus they should not, in principle, induce a vertical shift in AFM-FM exchange biased films.

As shown in Figs. 1(b) and 2(b), M_{Shift} depends strongly on the cooling field. For small cooling fields it is upwards or

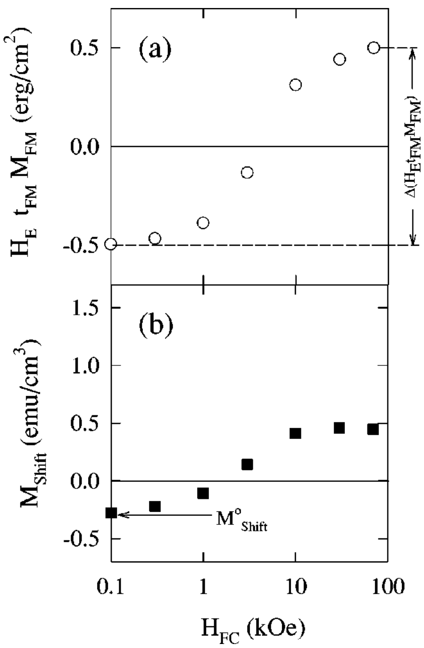

FIG. 1. Dependence of (a) the exchange bias interface energy, $H_E t_{FM} M_{FM}$, and (b) the shift in the magnetization axis, M_{Shift} on the cooling field, H_{FC}, for a FeF_2(200 nm)–Fe (15 nm)–Al (5 nm), where the FeF_2 layer was grown at $T_S = 300\,°C$.

downwards depending on the microstructure of the sample [see Figs. 1(b) and 2(b)]. For large cooling fields M_{Shift} is always upwards. It is noteworthy that single FeF_2 films also show a shift along the magnetization axis, which is *always* upwards, independently of cooling field.[26]

The correlation between the low ($H_{FC} = 0.1$ kOe) field vertical shift (M^o_{Shift}) and exchange bias is striking. A large upward M^o_{Shift} [Fig. 2(b)] is correlated with a slight cooling field dependence of H_E [Fig. 2(a)]. On the other hand, a small *downwards* M^o_{Shift} [Fig. 1(b)] is correlated with a large cooling field dependence of H_E together with positive exchange bias [Fig. 1(a)].

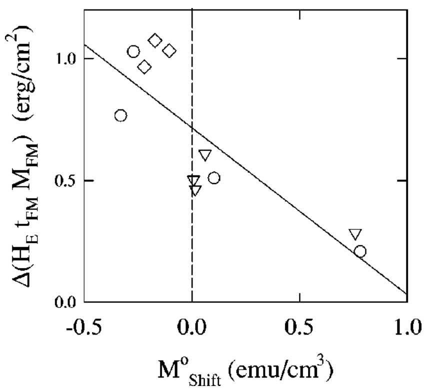

FIG. 3. Dependence of the total change in exchange bias between small and large cooling fields, $\Delta(H_E t_{FM} M_{FM})$, on the shift in the magnetization axis for low cooling fields, M^o_{Shift}, for $FeF_2(t_{AFM})$–Fe (15 nm)–Al (5 nm) samples of different AFM thickness: $t_{AFM} = 50$ nm (▽), $t_{AFM} = 100$ nm (○) and $t_{AFM} = 200$ nm (◇). The solid line is a guide to the eye.

The dependence of the total change in exchange bias with cooling field $\Delta(H_E t_{FM} M_{FM})$ on the low cooling field vertical shift, M^o_{Shift}, is shown in Fig. 3. $\Delta(H_E t_{FM} M_{FM})$ is largest for the samples that exhibit negative M^o_{Shift}. Whereas, samples with $M^o_{Shift} = 0$ have only a moderate change in $\Delta(H_E t_{FM} M_{FM})$. On the other hand, samples with positive M^o_{Shift} show almost no change in H_E for any cooling field [i.e., $\Delta(H_E t_{FM} M_{FM}) \sim 0$]. As can be seen in Fig. 4, similar trends were observed in MnF_2-Fe. However, the small vertical shifts in this system (in the range of 10^{-8}–10^{-7} emu) make the analysis more difficult.

A shift upwards indicates that the induced moment is in

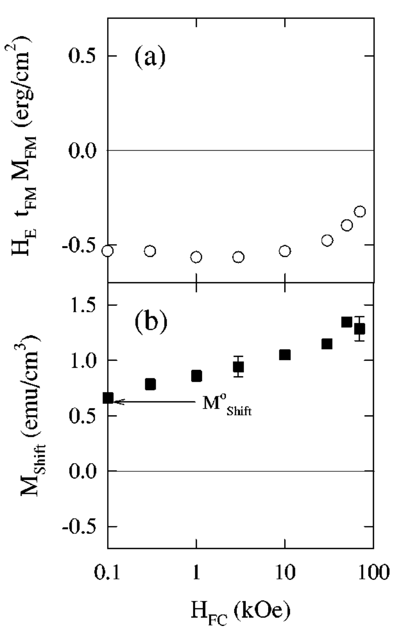

FIG. 2. Dependence of (a) the exchange bias interface energy, $H_E t_{FM} M_{FM}$, and (b) the shift in the magnetization axis, M_{Shift} on the cooling field, H_{FC}, for a FeF_2(100 nm)–Fe (15 nm)–Al (5 nm), where the FeF_2 layer was grown at $T_S = 200\,°C$.

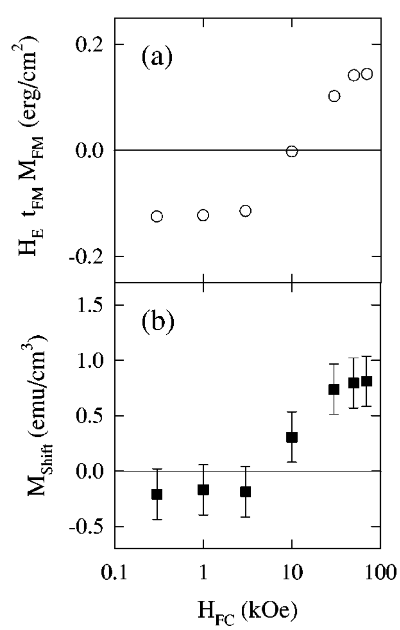

FIG. 4. Dependence of (a) the exchange bias interface energy, $H_E t_{FM} M_{FM}$, and (b) the shift in the magnetization axis, M_{Shift} on the cooling field, H_{FC}, for a MnF_2(85 nm)–Fe (14 nm)–Al (5 nm), where the MnF_2 layer was grown at $T_S = 275\,°C$.

the direction of the cooling field, whereas a downwards shift implies that the induced moment is in the opposite direction. In this second case, for small cooling fields, the Fe layer forces the net magnetization of the AFM/FM bilayer to be opposite to the cooling field. At large fields, the overall magnetization of the FM layer is always in the direction of the applied field, therefore a downward shift indicates that the coupling between the FM and AFM layers has to be *antiferromagnetic*.

Two cases must be considered; large and small cooling fields. For large cooling fields the interface coupling is overcome, thus the AFM layer behaves independently from the FM layer and the shift is always upwards, i.e. a moment is induced in the direction of the cooling field (similar to what is observed for single FeF_2 films[26]). It is noteworthy that the negative to positive H_E and the upwards to downwards M_{Shift} transitions occur at different fields. This is due to the intrinsic *upwards* shift of the FeF_2 layer, which added to the interfacial effect causes the crossover in M_{Shift} to move to lower fields. The sign of the shift for small cooling fields depends on microstructure (i.e., growth conditions) of the AFM layer. Smoother[6,11,24] samples tend to have negative shifts, whereas rough samples exhibit positive shifts. For small cooling fields, H_E (i.e., standard exchange bias) and the sign of the shift (upwards or downwards) are uncorrelated. This is not surprising, since under normal circumstances H_E depends on the strength of the interface coupling but not its sign.

Due to growth inhomogeneities the coupling is probably a mixture of *antiferromagnetic* or *ferromagnetic* interactions. Whereas the magnetization shift is given by the average of both interactions, H_E has a more complex relation with the sign and magnitude of the coupling. Both types of couplings induce H_E at low cooling fields, however, only antiferromagnetic coupling changes the sign of H_E at large cooling fields (''positive'' H_E). Hence, at present, a more quantitative understanding of the correlation between the shift along the magnetization and field axes in exchange bias systems is difficult.

Most exchange bias models ignore the possibility of vertical loop shifts,[16–18,12,19] except a model that postulates the formation of FM domains.[20] However, as discussed in the introduction, most models could indirectly account for a vertical shift. Unfortunately, due to the similar behavior displayed by the vertical shift in terms of m/A (moment per area), m/V_{AFM} (moment per AFM volume) and m/V_{FM} (moment per FM volume) we are unable to discriminate between the different models.

In conclusion, we have measured the induced moments in exchange biased FeF_2/Fe and MnF_2/Fe by a ''vertical'' shift of the hysteresis loop. This magnetization shift is related to the coupling at the interface, with downwards shifts indicating *antiferromagnetic* couplings at the interface. The type of coupling at the interface (*ferromagnetic* or *antiferromagnetic*) determines the large cooling-field behavior of the exchange bias, however, H_E for small cooling fields has a complex dependence with the coupling sign.

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[1] For a recent review see, J. Nogués and Ivan K. Schuller, J. Magn. Magn. Mater. **192**, 203 (1999).

[2] W.H. Meiklejohn and C.P. Bean, Phys. Rev. B **102**, 1413 (1956).

[3] R. Jungblut, R. Coehoorn, M.T. Johnson, J. aan de Stegge, and A. Reinders, J. Appl. Phys. **75**, 6659 (1994).

[4] S. Soeya, H. Hoshiya, R. Arai, and M. Fuyama, J. Appl. Phys. **81**, 6488 (1997).

[5] T. Lin, C. Tsang, R.E. Fontana, and J.K. Howard, IEEE Trans. Magn. **31**, 2585 (1995).

[6] J. Nogués, D. Lederman, T.J. Moran, and Ivan K. Schuller, Phys. Rev. Lett. **76**, 4624 (1996).

[7] T.J. Moran and Ivan K. Schuller, J. Appl. Phys. **79**, 5109 (1996).

[8] T. Ambrose and C.L. Chien, J. Appl. Phys. **83**, 7222 (1998).

[9] J. Nogués, D. Lederman, B.A. Gurney, and Ivan K. Schuller (unpublished).

[10] J. Nogués, T.J. Moran, D. Lederman, Ivan K. Schuller, and K.V. Rao, Phys. Rev. B **59**, 6984 (1998).

[11] C. Leighton, J. Nogués, H. Suhl, and Ivan K. Schuller, Phys. Rev. B **60**, 12 837 (1999).

[12] N.C. Koon, Phys. Rev. Lett. **78**, 4865 (1997).

[13] H. Suhl and Ivan K. Schuller, Phys. Rev. B **58**, 258 (1998).

[14] T.M. Hong, Phys. Rev. B **58**, 97 (1998).

[15] R. Ramírez, M. Kiwi, and D. Lederman, Bull. Am. Phys. Soc. **42**, 445 (1997).

[16] A.P. Malozemoff, J. Appl. Phys. **63**, 3874 (1988).

[17] D. Mauri, H.C. Siegmann, and P.S. Bagus, J. Appl. Phys. **62**, 3047 (1987).

[18] *Selected Works of Louis Néel*, edited by N. Kurti (Gordon and Breach, New York, 1988), p. 469.

[19] K. Takano, R.H. Kodama, and A.E. Berkowitz, W. Cao, and G. Thomas, Phys. Rev. Lett. **79**, 1130 (1997).

[20] M. Kiwi, J. Mejía-López, R. Portugal, and R. Ramírez, Europhys. Lett. **48**, 573 (1999).

[21] N.M. Salansky, B.P. Khrustalev, A.S. Melnik, and Z.I. Sinegubova, Thin Solid Films **4**, 105 (1969); B.H. Miller and E.D. Dahlberg, Appl. Phys. Lett. **69**, 3932 (1996).

[22] M. Chirwa, L. Lundgren, P. Nordblad, and O. Beckman, J. Magn. Magn. Mater. **15-18**, 457 (1980).

[23] A.S. Borovik-Romanov, Zh. Éksp. Teor. Fiz. **38**, 1088 (1960) [Sov. Phys. JETP **11**, 786 (1960)].

[24] J. Nogués, D. Lederman, T.J. Moran, Ivan K. Schuller, and K.V. Rao, Appl. Phys. Lett. **68**, 3186 (1996).

[25] T. Ambrose and C.L. Chien, Phys. Rev. Lett. **76**, 1743 (1996).

[26] S. Adenwalla, G. Felcher, J. Nogués, and Ivan K. Schuller, J. Appl. Phys. **81**, 5307 (1998).