Exchange coupling in epitaxial CoO/NiFe bilayers with compensated and uncompensated interfacial spin structures

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An uncompensated antiferromagnetic surface of CoO is essential for the establishment of exchange bias. Exchange coupling for compensated surfaces is manifested by a large coercivity but no exchange bias field. Degradation of a compensated surface leads to an exchange bias field. These results clarify a key outstanding issue in exchange bias.

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The nature of the exchange bias phenomenon in bilayers of a ferromagnet (FM) and an antiferromagnet (AF), first observed by Meiklejohn and Bean in 1956,[1] has attracted a great deal of attention in recent years due to the intriguing physics[2] and its central role in spin-valve field sensing devices.[3] After the exchange coupling has been established, most commonly by field cooling, the hysteresis loop of the FM layer is shifted by the amount of the exchange bias field H_E accompanied by an enhanced coercivity H_C. Central to exchange bias is the spin structures of the AF and the FM layers. Early models, assuming rigid AF spin structures throughout the reversal of the FM layer are incompatible with many experimental results.[1] The emerging picture is that the AF moments near the interface rotate with the FM moments due to the strong interfacial coupling. The anisotropy of the AF, which prevents rotation of the AF moments at a distance from the interface, causes the formation of a domain wall in the AF.[4–8] An additional magnetic field must overcome the energy required to establish the domain wall, as manifested by a shifted hysteresis loop.

One of the central issues that has perhaps attracted the most attention is whether exchange bias is contingent upon an uncompensated AF spin structure at the interface.[4–8] The unidirectional nature of the exchange bias necessitates the existence of an uncompensated spin structure at or near the FM/AF interface, either inherent to the spin structure of the AF or due to roughness of the interface.[7] Most surprisingly, in contradiction to the expectation, exchange bias has been observed for AF surfaces that are supposedly compensated (e.g., Fe/[110]FeF_2).[9,10] To account for exchange bias in the case of a compensated interface, a spin-flop type ordering has been proposed.[6] However, while evidence of a spin-flop arrangement in the AF has been observed, it is unclear whether the observed spin-flop is the cause of the exchange bias field H_E.[11] Furthermore, recent theoretical calculations show that such spin-flop ordering can contribute to an enhanced H_C but will not result in exchange bias field (i.e., $H_E=0$).[7] Interfacial roughness in a compensated surface can lead to the presence of a net moment. The difficulty of measuring such a moment notwithstanding, conflicting correlation between roughness and exchange bias has been reported.[12,13] To date, the issue of compensated and uncompensated AF surfaces remains unresolved.

In this work, we have addressed the question of compensated and uncompensated surfaces and the issue of roughness by studying exchange coupling using epitaxial AF CoO layers of different crystallographic orientations grown on single crystal substrates. We show that exchange coupling exists for both compensated and uncompensated AF interfaces. However, exchange bias ($H_E \neq 0$), the traditional signature of exchange coupling, is observed *only* for an *uncompensated* AF. Exchange coupling in a compensated AF spin structure results in an enhanced H_C but no bias field ($H_E=0$). We further show that for the uncompensated AF surface, the absence of exchange bias field depends on the quality of the epitaxial films.

Because of the simple crystal and spin structures, we have used CoO/Py bilayers (Py=$Ni_{81}Fe_{19}$) to address these key issues in exchange coupling in FM/AF bilayers. The simple NaCl-type fcc structure of CoO allows epitaxial films of different orientations to be grown on MgO and sapphire substrates. Neutron diffraction results show that bulk CoO has a simple spin structure with the AF ordering along one of the [111] directions. Hence, all {100} and {110} surfaces are completely compensated, whereas the {111} surfaces are generally uncompensated.[14] In this respect, AF materials with complex spin structures (e.g., FeMn, and IrMn) are unsuitable for the purpose of addressing the question of compensated AF surface. Among the AFs (e.g., CoO, NiO and FeF_2) with simple crystal and spin structures that exhibit exchange bias, CoO has the distinct advantage of having the highest AF anisotropy energy.[15]

The CoO films from thermally oxidized Co films or Co particles generally are not epitaxial nor with unique orientation.[1] Epitaxial CoO/Py bilayers were made by magnetron sputter deposition in a system with a base pressure of 3×10^{-8} Torr. Epitaxial CoO films of thicknesses up to 750 Å were deposited at 400 °C onto single-crystal substrates of

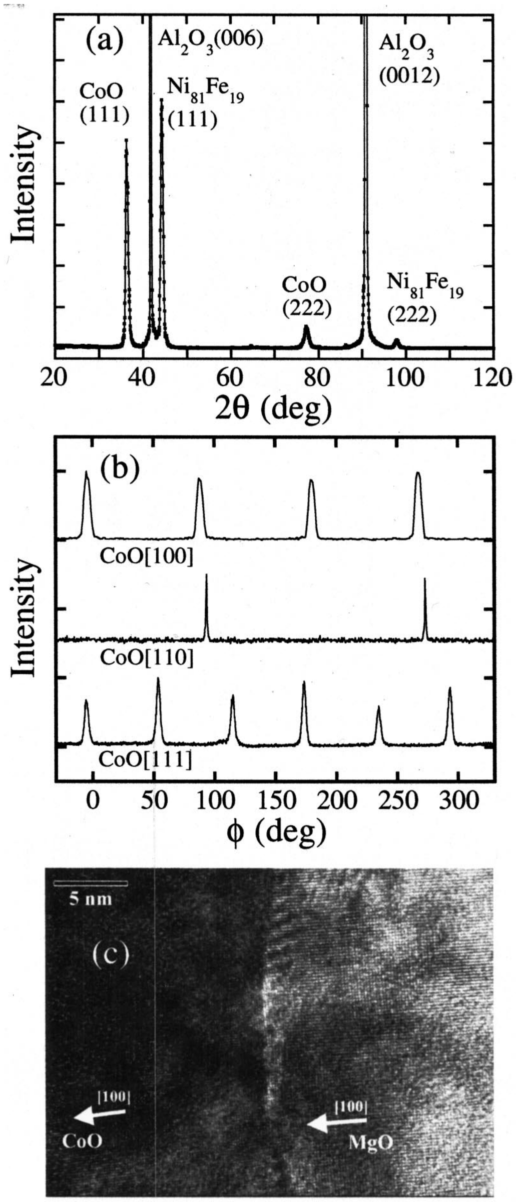

FIG. 1. (a) θ-2θ x-ray diffraction scan of (111)CoO film grown on (0001) sapphire. (b) (100) and (110) CoO films show twofold and fourfold symmetry, respectively, in the ϕ scan, whereas (111)CoO films show 6 instead of 3 peaks due to twinning. (c) cross-sectional TEM micrograph of (100)CoO on (100)MgO substrate.

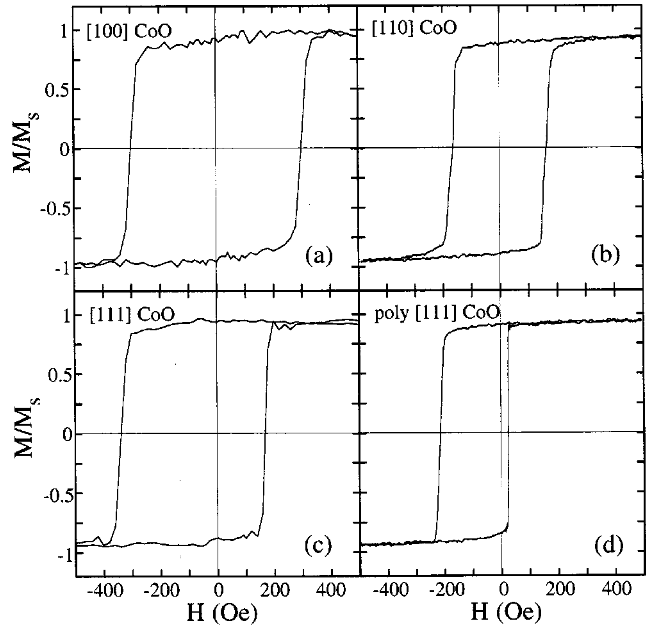

FIG. 2. Hysteresis loops of CoO/Py bilayers containing epitaxial (a) (100), (b) (110), (c) (111), and (d) polycrystalline CoO at 80 K after field-cooled from room temperature in a 10 kOe field. Note that for the compensated surfaces of (100) and (110)CoO, there is no exchange bias field.

[100] MgO, [110] MgO, [111] MgO, and [0001] sapphire. In addition, polycrystalline [111]-orientated CoO films were also made using Si substrates. To minimize variations in the fabrication conditions, all the samples were made in the same deposition run. Following the CoO deposition, samples were cooled to 200 °C, at which a 260 Å thick layer of Py was deposited. All samples were capped with 50 Å Ta to prevent oxidation.

The thin CoO layers, buried below the Py and Ta layers, were characterized by x-ray diffraction using the Cu $K_{\alpha 1}$ radiation with a resolution of 12 arcsec, Bragg-reflected off a (220) Ge crystal. The $\theta/2\theta$ scans show that the CoO layers deposited on [0001] sapphire [shown in Fig. 1(a)] and [111] MgO, [100] MgO, and [110] MgO are exclusively [111], [100], and [110] oriented, respectively. To establish the epitaxial relationship, ϕ-scan measurements of the off-axis reciprocal lattice vectors were made. The epitaxial nature of the [100] CoO film was confirmed by the four-fold symmetry in the ϕ scan of the (420) peak, as shown in Fig. 1(b). The same conclusion was reached by cross-sectional transmission electron microscopy (TEM) of the same sample, as shown in Fig. 1(c). The [110] MgO/CoO/Py films also reveal epitaxy as indicated by the twofold symmetry of the ϕ scan of the (111) peak [shown in Fig. 1(b)]. For the [0001] sapphire/CoO/Py films, the presence of 6, instead of 3, peaks in the ϕ scan of the (222) peak of CoO [as shown in Fig. 1(b)] indicates twinning of the crystals as commonly observed in [111] films. In addition to film orientation and epitaxy, we have further assessed the quality of the epitaxial CoO films by analyzing the correlation length in the growth direction from the peak width of the principal x-ray peak. The best samples used were those with correlation lengths close to the layer thickness.

To determine exchange coupling, each CoO/Py sample was field-cooled in a 10 kOe field from above T_N of CoO (about 290 K) to 80 K. Field cooling along different crystal directions did not make a noticeable difference. The hysteresis loops measured at 80 K of the CoO/Py samples containing the epitaxial [100], [110], and [111] CoO films with long correlation lengths, as well as polycrystalline [111] CoO films, are shown in Fig. 2. It is immediately evident that epitaxial [100] and [110] CoO films, as shown in Figs. 2(a) and 2(b), show no exchange bias field ($H_E=0$), whereas both the epitaxial [111] and polycrystalline [111] CoO films,

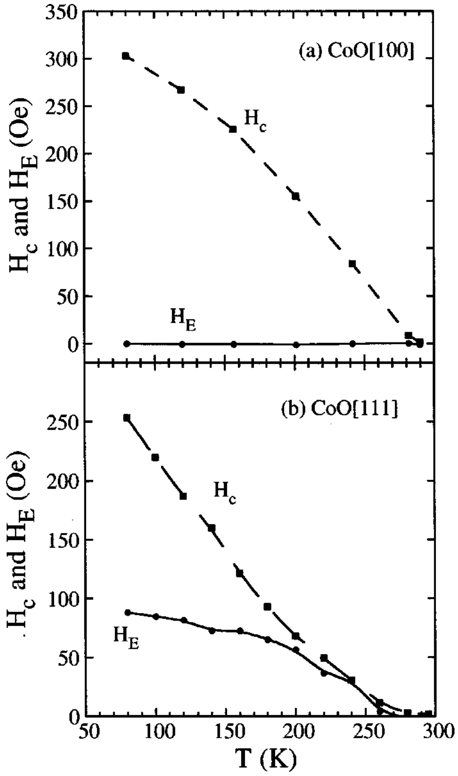

FIG. 3. Temperature dependence of exchange field (H_E) and coercivity (H_c) of (a) [100] and (b) [111] CoO/Py bilayers.

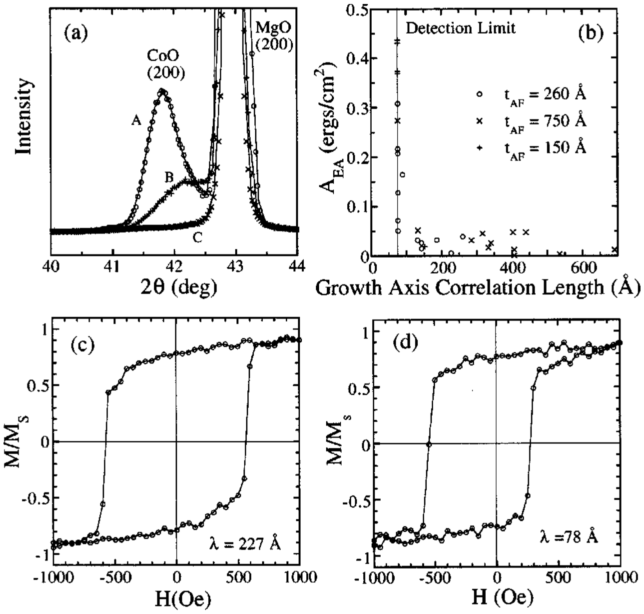

FIG. 4. (a) CoO[200] and MgO[200] peaks for samples grown under different substrate temperature (T_S), resulting in different correlation length of the CoO layer of 282, 153, and 89 Å for cases A, B, and C, respectively, (b) exchange anisotropy energy vs correlation length in the growth direction for a series of (100)MgO/CoO/Py samples with CoO thickness 260, 750, and 150 Å, and hysteresis loops at 80 K of 260 Å thick (100)CoO films with a correlation length of (c) 227 Å and (d) 78 Å.

as shown in Figs. 2(c) and 2(d), show large exchange bias fields ($H_E \neq 0$).

Although there is no exchange bias field for the completely compensated interfaces of [100] and [110], there is exchange coupling between the FM and the AF layers. This is indicated by the large coercivity H_c of the [100] and [110] interfaces, about 300 and 164 Oe, respectively. Both coercivities are much larger than the value of about 3 Oe measured for the uncoupled Py layer. This is further illustrated by the temperature dependence of H_E and H_c of the [100] CoO/Py samples shown in Fig. 3(a), where $H_E = 0$ has been observed at all temperatures. The large value of H_c gradually decreases to the value of the uncoupled Py layer as the temperature gradually approaches $T_N \approx 290\,\mathrm{K}$, as observed in many exchange-coupled FM/AF bilayers. As shown in Fig. 3(b), for the [111] CoO, both H_E and H_c decrease with temperature and vanish at T_N. Assuming that the spin structure of bulk CoO is preserved in epitaxial CoO film, these results show that exchange coupling exists for both uncompensated and compensated interfaces. Exchange bias field results only from an uncompensated surface of [111]. There is exchange coupling ($H_c \neq 0$) for compensated surfaces ([100] and [110]), but with no exchange bias field. The latter feature is in agreement with the theoretical studies of Schulthess and Butler.[2]

Of the three [111] CoO/Py samples with large exchange bias fields, the largest value (106 Oe) of H_E has been observed in polycrystalline [111] CoO, followed by 86 Oe in epitaxial [0001] sapphire/[111] CoO and 36 Oe in [111] MgO/[111] CoO. These variations correlate with the interface roughness, as determined by small-angle x-ray reflectivity. These results indicate that Si has the smoothest interface, [0001] sapphire has an interface of intermediate roughness, and [111] MgO has the roughest interface. This is consistent with the expectation that an increase in roughness would result in a decrease of the uncompensated moment, and thus a smaller H_E.

Having established that epitaxial films with a compensated interface (e.g., [100]) show no exchange bias field, we have investigated the consequence of reduced epitaxy, which leads to roughness, reduced spin compensation, and generally inferior quality. The resultant reduced spin compensation at the interface can be revealed by the appearance of H_E. During fabrication of these [100] CoO films, the deposition conditions of substrate temperature (from −200–200 °C) and sputtering gas pressure were deliberately altered from those for epitaxial growth. The quality of the resultant samples was determined by measuring the correlation length $\lambda = \xi_{\mathrm{AF}}$ in the growth direction using x-ray diffraction. A high quality AF layer would be that with $\xi_{\mathrm{AF}} \approx t_{\mathrm{AF}}$, whereas an inferior film would show $\xi_{\mathrm{AF}} \ll t_{\mathrm{AF}}$.

Three representative x-ray scans of various [100] CoO layers with $t_{\mathrm{AF}} = 150$, 260, and 750 Å are shown in Fig. 4(a). Whether the (200) peak of CoO can be resolved from the (200) peak of MgO, depends on the width of the CoO (200) peak, which provides a measurement of ξ_{AF}. In Fig. 4(a), the (200) peak of CoO can be resolved in two cases but not the third because of short $\xi_{\mathrm{AF}} = 89\,\text{Å}$ in the latter. Considering all the CoO films of 260 Å for example, the samples with long correlation lengths [e.g., $\xi_{\mathrm{AF}} = 227\,\text{Å}$ shown in Fig.

4(c)] show $H_E=0$, whereas those with short correlation lengths [e.g., $\xi_{AF}=78$ Å shown in Fig. 4(d)] show a large value of H_E. To compare samples with different layer thicknesses, the exchange energy per unit area $A_{EA}=H_E t_{FM} M_{FM}$ has been calculated and plotted in Fig. 4(b) as a function of the correlation length in the growth direction, where t_{FM} and M_{FM} are the thickness and magnetization of the Py layer. Because the x-ray resolution of the CoO (200) peak places a lower limit of about 78 Å for the correlation length, all data on the vertical line at 78 Å in Fig. 4(b) correspond to $\xi_{AF} \leqslant 78$ Å. As shown in Fig. 4(b), the samples with long correlation lengths do not exhibit H_E and only samples with short correlation lengths exhibit a significant exchange bias. These results further reinforce the conclusion that the interface with a compensated spin structure does not exhibit an exchange bias field. Only when the compensated interface is significantly altered with the appearance of interfacial moments, does the exchange bias field appear.

In summary, we have shown conclusively that FM/AF bilayers containing epitaxial [100] CoO and [110] CoO films with a compensated spin structure do not exhibit an exchange bias field H_E, even though the exchange coupling is in place as revealed by the enhanced coercivity. Only samples with an uncompensated spin structure, either inherent to the crystal structure, as in [111] CoO, or due to inferior epitaxy, show an exchange bias field H_E. These results clearly establish that uncompensated AF interfacial spin structure is essential for exchange bias.

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[1] W. H. Meiklejohn and C. P. Bean, Phys. Rev. **102**, 1413 (1956); **105**, 904 (1957).

[2] See, e.g., J. Nogues and I. K. Schuller, J. Magn. Magn. Mater. **192**, 203 (1999).

[3] B. Dieny, V. S. Speriosu, S. S. P. Parkin, B. A. Gurney, D. R. Wilhoit, and D. Mauri, Phys. Rev. B **43**, 1297 (1991).

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

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

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

[7] T. C. Schulthess and W. H. Butler, Phys. Rev. Lett. **81**, 4516 (1998).

[8] M. D. Stiles and R. D. McMichael, Phys. Rev. B **59**, 3722 (1999).

[9] J. Nogues, D. Lederman, T. J. Moran, and I. K. Schuller, Phys. Rev. Lett. **76**, 4624 (1996).

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

[11] Y. Ijiri, J. A. Borchers, R. W. Irwin, S. H. Lee, P. J. van der Zaagm, and R. M. Wolf, Phys. Rev. Lett. **80**, 608 (1998).

[12] J. Nogues, D. Lederman, T. J. Moran, I. K. Schuller, and K. V. Rao, Appl. Phys. Lett. **68**, 3186 (1996).

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

[14] W. L. Roth, Phys. Rev. **110**, 1333 (1958).

[15] J. Kanamori, Prog. Theor. Phys. **17**, 197 (1957).