Strong exchange bias by a single layer of independent antiferromagnetic grains: The CoO/Co model system

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CoO/Co bilayers with only 20 Å thin CoO reveal an extremely strong exchange bias characterized by magnetic hysteresis loops of rectangular shape and an ideal dependence on Co layer thickness. Quantitative information on important structural and magnetic parameters are given including the exact lateral size and shape of the fine grains constituting the films and a characteristic temperature dependence of the exchange bias. The high coupling energy originates from a single dense layer of independent antiferromagnetic grains in connection with a large number of uncompensated spins in the CoO.

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One of the striking discoveries in magnetism made more than 40 years ago during the study of surface oxidized particles of cobalt was the observation of hysteresis loops substantially shifted away from zero field.[1] The loop shift was attributed to exchange coupling at the interface of the ferromagnetic (FM) and antiferromagnetic (AFM) materials. The phenomenon was therefore termed exchange anisotropy or exchange bias (EB). Since then a great variety of AFM/FM systems with different AFM and FM materials have been found to exhibit the EB effect.[2] These systems have been studied with a large number of experimental techniques, and several theories have been put forward to describe the physical properties. However, a complete understanding of the underlying interfacial coupling mechanism is still missing and a quantitatively satisfactory model for a specific AFM/FM system has not been reported up to now.

The most widely studied type of EB systems are thin AFM/FM films. One fundamental problem in analyzing the observed effects is to accurately determine the structure and morphology at the AFM/FM interface. These properties strongly depend on the preparation methods which include sputter deposition of AFM materials, metal evaporation in a reactive gas atmosphere, or oxidation after metal film deposition.[2] In this paper, we present a detailed analysis of CoO/Co bilayers prepared by *in situ* oxidation.[3] This particular AFM/FM system exhibits an extraordinarily strong EB which will be shown to originate from independent grains with an extremely small AFM thickness. Quantitative information on the most relevant structural and magnetic parameters will also be provided.

Long-range ordered, ideally hydrogen terminated H/Si(111)1$\times$1 substrates were prepared *ex situ* by wet chemical treatment and subsequently introduced into an ultrahigh vacuum (UHV) apparatus with a base pressure in the low 10^{-10}-mbar range.[3] After cleanliness and crystalline orientation of substrate surfaces had been checked by Auger electron spectroscopy (AES) and low-energy electron diffraction (LEED) Co films with lateral dimensions of 4 $\times$4 mm^2 were deposited by molecular-beam epitaxy (MBE) at a low rate of 1–2 Å/min. Film thickness was monitored by a water-cooled quartz microbalance. CoO/Co bilayers with a constant CoO thickness (20 Å) were obtained by a recently introduced *in situ* oxidation method using a controlled exposure of high-purity oxygen gas at low partial pressure.[3] Scanning tunneling microscopy (STM) images were carried out *in situ* by a commercial Omicron instrument. Magnetic measurements were made with a Quantum Design superconducting quantum interference device magnetometer. For *ex situ* investigations the samples were capped with 40 Å of Au.

Pure Co layers deposited on H/Si(111)1$\times$1 substrates are soft magnetic exhibiting corcivities of 20–30 Oe only [Fig. 1(a)]. Their magnetization M_s in magnetic fields up to 1500 Oe amounts approximately 5% less than the magnetization given for bulk Co (1440 emu/cm^3). The magnetic behavior of Co layers drastically changes if they are covered by AFM CoO [Fig. 1(b)]. CoO/Co bilayers exhibit a very strong EB effect which considerably varies with Co layer thickness. The coercivities for decreasing and increasing external fields, H_{CA} and H_{CP}, behave very differently. $|H_{CA}|$ continuously decreases with increasing Co thickness whereas H_{CP} features an almost constant value between -30 Oe and zero field. Using the formula $H_E=|H_{CA}+H_{CP}|/2$ the strength of the effect can quantitatively be described by the EB field H_E. For a Co layer of 164 Å thickness, H_E is as large as 460 Oe.

The dependence of H_E on inverse Co layer thickness is

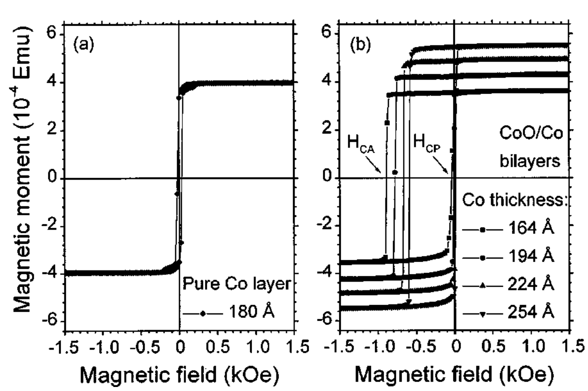

FIG. 1. Magnetic hysteresis loops at $T=10$ K for (a) a pure 180-Å Co layer and (b) 20-Å CoO/Co bilayers with increasing Co thickness (164, 194, 224, and 254 Å) performed after cooling in a field $H^{COOL}=+4000$ Oe from $T=320$ K. The loops start with a positive external field of $H=+1500$ Oe.

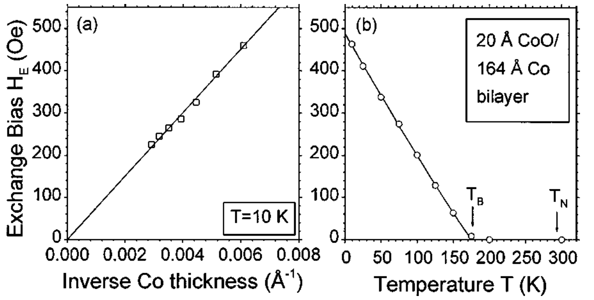

FIG. 2. (a) Exchange bias field H_E as function of inverse Co layer thickness obtained for 20-Å CoO/Co bilayers with different Co thickness. (b) Exchange bias field H_E as a function of temperature T from 10 to 300 K for a 20-Å CoO/164-Å Co bilayer. The solid line is a fit to the data (see text).

summarized in Fig. 2(a). The data reveal an inverse proportionality between the strength of the EB and the thickness of the FM layer which convincingly demonstrates that EB is an interfacial effect. An inverse behavior has been found for many systems studied, but in most cases only roughly.[2] In the present case the experimentally found relation between H_E and Co thickness shows only minor deviations from a linear dependence. Using the product of the EB field H_E, the magnetization M_s and the Co thickness $d_{\rm CO}$, $H_E \times M_s \times d_{\rm CO}$, as a lower limit for the interfacial energy we obtain a value of $1.03 \pm 0.03\,{\rm erg/cm^2}$ (at $T = 10\,{\rm K}$). This is one of the highest values ever reached in EB systems.[2,4] The extraordinary strength of the coupling is further emphasized by the fact that the FM magnetization reverses into the biased (cooling field) direction for oppositely directed external fields which is evidenced by the negative values of the coercivity H_{CP}.

Furthermore, it should be noted that the hysteresis loop maintains the same typical rectangular shape throughout the whole FM thickness range considered here. Similar behavior only exists for AFM/FM bilayers containing soft magnetic materials such as permalloy.[2,5] A closer inspection of the loops depicted in Fig. 1(b) provides interesting details of the underlying magnetic reversal mechanisms. For decreasing fields, the magnetization remains in saturation up to relatively high coercivities H_{CA} before a sudden reversal takes place. Contrary to this, the return to the remanent state for increasing fields leads to rounded edges of the hysteresis loop. The occurrence of asymmetric reversal mechanisms for decreasing and increasing fields has been found experimentally for several EB systems and has also been addressed theoretically.[6–9] The presently observed behavior resembles results from a simple one-dimensional model for EB based on domain-wall formation in the AFM.[6] Roughly speaking, the asymmetric reversal is interpreted by an irreversible jump of the magnetization in one direction opposed to a small region of reversible rotation in the other.

Figure 3 shows an *in situ* STM image of the surface of a 180-Å-thick Co layer which is representative for all films considered in this letter. Obviously, room-temperature deposition yields roundly or slightly elliptically shaped Co islands. Most of the islands closely touch each other but do not coalesce into a flat, uniform film. However, the film is very dense with only small areas reaching down to underlying grains (black areas). That is, the top of the Co films can be considered as a single dense layer of Co grains with a typical lateral diameter of 100 ± 10 Å. The MBE grown Co films are characterized by a fine grain structure with an extremely narrow distribution of lateral grain size. It should be noted that the height difference between the highest (white) and the lowest (black) areas amounts only 24 Å. A characteristic height profile of the Co film surface [Fig. 3(b)] proves that height variations along the surface and therefore also along the interface are relatively smooth. From the top to the edge of each island where it usually touches neighboring islands the height difference amounts only 8 Å on average. Height variations between separated Co grains and island height profiles at low coverages performed by the authors indicate a height of the grains of only 20 Å although a higher value cannot be excluded.

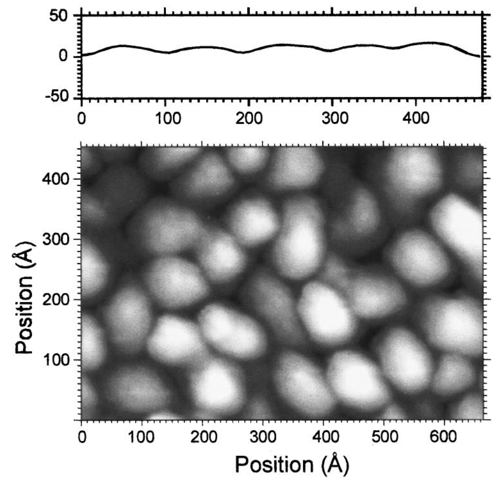

FIG. 3. (a) Bottom: Topographic STM image of a pure 180-Å Co layer deposited on a H/Si(111) 1×1 substrate at room temperature ($U = +0.1$ V and $I = 1.0$ nA). The size of the image is 460 $\times 670$ Å. (b) Top: Typical height profile along a straight line in (a). The lowest point at the right has arbitrarily been chosen as zero.

As already mentioned above the present CoO/Co bilayers are prepared by a controlled *in situ* exposure of clean metal layers to an extremely small amount of pure oxygen gas.[3] The thickness of the CoO obtained by this method has accurately been determined to 20 Å.[3,10] STM imaging of the surface of such a 20-Å CoO/Co bilayer reveals no apparent changes of surface morphology compared to the pure Co layer (apart from the necessary change of the tunneling parameters). Thus it can be concluded that the CoO film consists of a single dense layer of small grains which originate from the oxidation of the top layer of Co grains in the pure film.

According to a sophisticated model for EB very recently put forward by Stiles and McMichael (SM),[7,11] the FM layer is suggested to interact with independent AFM grains. In this model each grain is assumed to be in a single AFM state and it is assumed to be small enough not to break into domains. Rotation of the FM magnetization winds up partial domain walls in the AFM grains due to the interfacial coupling of the AFM spins at the interface. Although this interpretation of

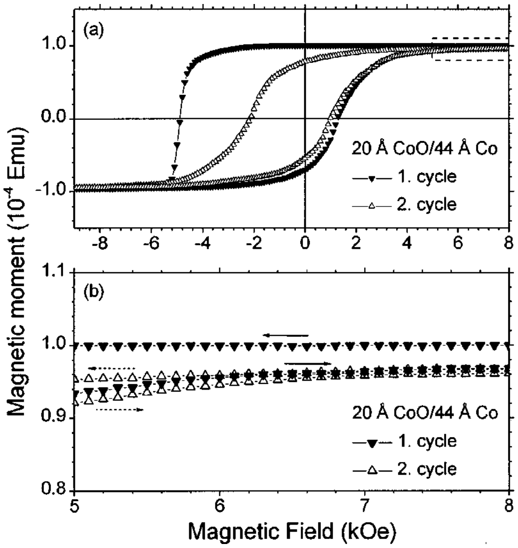

FIG. 4. (a) Two magnetic hysteresis cycles for a 20-Å CoO/44-Å Co bilayer performed immediately after each other at a temperature $T=10\,\mathrm{K}$ after cooling in a field $H^{\mathrm{COOL}}=+4000\,\mathrm{Oe}$ from $T=320\,\mathrm{K}$. (b) Detail of (a) for external fields between $+5000$ and $+8000$ Oe inside the dashed border line in (a). Arrows mark the direction of the two branches (increasing or decreasing) of the hysteresis loop (solid for the first cycle, dashed for the second).

the EB effect appears to be particularly appropriate for the small sized CoO grains described above, the data presented so far do not provide evidence for the proposed mechanism of domain-wall formation. Therefore it is necessary to consider other characteristic physical properties. One of these is the temperature dependence of the EB.

The behavior of H_E in the range from 10 to 300 K is shown in Fig. 2(b). The data can accurately be modeled by a linear falloff proportional to $(1-T/T_B)^n$. The fitting of $H_E=H_{E0}(1-T/T_B)^n$ results in $H_{E0}=490\,\mathrm{Oe}$, $T_B=175\,\mathrm{K}$, and $n=1.06$ [solid line in Fig. 2(b)]. EB vanishes above the blocking temperature $T_B=175\,\mathrm{K}$ which is far below the Néel temperature T_N of bulk CoO($T_N=293\,\mathrm{K}$). The linear temperature dependence of H_E with $n\approx 1$ can theoretically be reproduced by assuming a cubic anisotropy for the AFM material.[12] However, it can also be explained by the SM model without symmetry restrictions:[11] the experimental fact that EB occurs below a temperature $T_B\approx\frac{2}{3}T_N$ is quantitatively explained by thermal instabilities of the AFM state in grains smaller than a certain size.

Figure 4(a) shows two magnetic hysteresis cycles performed one after the other for a 20-Å CoO/44-Å Co bilayer. For the first loop, magnetization reversal with decreasing fields starts at -2000 Oe and covers a comparatively wide range up to fields higher than -5000 Oe. For increasing fields, the range in which reversal takes place even expands. More importantly, large external fields (H_{CP}) have to be applied to achieve a complete magnetization reversal back into the positive direction. This is completely different from the behavior observed for thicker Co films [Fig. 1(b)] where the backward transition occurs spontaneously at low negative or zero fields. Obviously, already the first magnetization reversal into the negative field direction substantially changes the magnetic behavior for thinner Co films. The second cycle proceeds with a drastic reduction of H_{CA} to only -2200 Oe (compared to -4900 Oe for the first loop) and an overall mode which rather resembles a hard axis reversal of an uniaxial FM film. Referring to the results obtained for larger Co thickness (Figs. 1 and 2) where the product $H_E\times M_s\times d_{\mathrm{CO}}$ and $H_{CA}\times M_s\times d_{\mathrm{CO}}$, respectively leads to a constant value magnetization reversal would be expected at a coercivity $H_{CA}\approx -3300\,\mathrm{Oe}$. Contrary to this, the reversal starts at considerably lower fields and extends over a wide field range. This type of behavior can be observed for a thickness below the critical value of $d_{\mathrm{CO}}^{\mathrm{CRIT}}\approx 80\,\mathrm{Å}$. It reveals that beyond exchange coupling and domain-wall formation thickness dependent properties of thin FM films are of crucial importance for the observation of ideal EB. The critical behavior may be explained by a change in the type of reversal mechanism which has already been predicted by a micromagnetic theory for certain bilayer systems in the ultrathin film regime.[13]

More importantly, the data obtained for low Co thickness supply quantitative information about the interfacial exchange. According to the data in Fig. 4(b) which shows a detail of the complete hysteresis loops, the total magnetic moment at high positive external fields is reduced by approximately $3.5\times 10^{-6}\,\mathrm{emu}$ due to the performance of the first hysteresis cycle. Thus it can be concluded that the 20-Å CoO/44-Å Co bilayer in its initially strongly biased state possesses a moment along the axis of unidirectional anisotropy which is considerably higher than in its unbiased state which is more or less already reached after one hysteresis cycle has been performed. Because the magnetization of the Co layer is in saturation at external fields as high as 8000 Oe, the observed difference in moment of $2.2\times 10^{-5}\,\mathrm{emu/cm^{-2}}$ has to be attributed to uncompensated spins in the CoO which correspond to approximately half a monoatomic layer of bulk CoO assuming a moment of $3.8\mu_B$ for each Co^{2+} ion.[14]

The experimental finding of a high density of uncompensated spins in the CoO is similar to the determination of about 0.8 monolayer of uncompensated Fe moments in FeMn/Co bilayers by x-ray dichroism[15] suggesting that this property is in common with other AFM/FM systems. Following an ideal interface model which relies on Heisenberg exchange across an epitaxial atomically smooth AFM/FM interface the density of uncompensated spins determined for the CoO/Co system would lead to EB fields which are more than one order of magnitude higher than the experimentally observed values. It can therefore be concluded that even if a high density of uncompensated AFM spins exists the EB effect cannot quantitatively be explained by an ideal interface model. This is consistent with the assumption, that the interfacial energy is rather determined by partial domain-wall formation in the AFM grains.[6,7,11,12] The present results are at variance with the interpretation of experiments using sputter deposited CoO as AFM material. In this study, it has been found that the spins responsible for the uncompensated AFM moment constitute only about 1% of the spins in a monoatomic layer of CoO.[14]

In conclusion, with the present CoO/Co bilayers we have found an AFM/FM system exhibiting remarkably simple characteristics of the magnetic behavior and an extraordinarily strong EB effect. The most relevant physical parameters responsible for EB have also been determined: The AFM consists of a single dense layer of small roundly shaped grains with a typical lateral diameter of 100 ± 10 Å and an average CoO thickness of approximately 20 Å. In the EB state half a monoatomic layer of uncompensated spins can be attributed to the AFM. These results and a characteristic temperature dependence of the EB strongly support the formation of partial domain walls or spin twisting in independent AFM grains to be responsible for the experimentally observed EB.[7,11] The present CoO/Co system provides a model system for studying EB within the large class of granular or polycrystalline AFM/FM bilayers. A unique advantage for theoretical analysis is the simple structure of the fine CoO grains with an extremely narrow distribution of lateral size and a CoO thickness corresponding to only about 8–9 Co layers.

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