Reversing the Training Effect in Exchange Biased CoO/Co Bilayers

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We performed a detailed study of the training effect in exchange biased CoO/Co bilayers. Highresolution measurements of the anisotropic magnetoresistance (AMR) display an asymmetry in the first magnetization reversal process and training in the subsequent reversal processes. Surprisingly, the AMR measurements as well as magnetization measurements reveal that it is possible to partially reinduce the untrained state by performing a hysteresis measurement with an in-plane external field perpendicular to the cooling field. Indeed, the next hysteresis loop obtained in a field parallel to the cooling field resembles the initial asymmetric hysteresis loop, but with a reduced amount of spin rotation occurring at the first coercive field. This implies that the antiferromagnetic domains, which are created during the first reversal after cooling, can be partially erased.

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The exchange bias (EB) effect is observed when a layer of a ferromagnet (FM) makes contact with a layer of an antiferromagnet (AFM), which introduces an exchange coupling at their interface. This results in a unidirectional shift of the hysteresis loop when the bilayer is grown in a magnetic field or cooled in a field below the Néel temperature (T_N) of the AFM. The EB in the AFM-FM bilayers also gives rise to an enhanced coercivity as well as to an asymmetric reversal of the magnetization, which can be strongly affected by "training," i.e., by going through consecutive hysteresis loops. Exchange bias was discovered almost 50 years ago by Meiklejohn and Bean [1]. Only recently the origin of exchange bias was linked to a fraction of uncompensated interfacial spins (about 4% to 7% of a monolayer) that are pinned in the vicinity of the interface, inside the AFM, and are not affected by an external field [2,3]. A reliable theoretical understanding is, however, still lacking [4-7]. Therefore, and because of technological applications such as spin valves in magnetic reading heads and magnetic random access memories, the EB effect remains at the forefront of research in thin film magnetism.

In this Letter, we report on the results of a detailed study of the training effect in CoO(AFM)/Co(FM) bilayers. Polycrystalline CoO/Co bilayers are selected due to their very pronounced training effects: the coercivity decreases and the shape of the magnetization loop changes considerably. Several theoretical models have been put forward to explain the training effect, but a detailed understanding of the effect is missing. The domain state model, which states that the EB shift results from an exchange field provided by irreversible magnetization of the AFM, allows us to explain the training effect in terms of domain wall formation *perpendicular to the interface* in the AFM [8,9]. When going through the hysteresis loop, a rearrangement of the AFM domain structure results in a partial loss of the domain state magnetization and causes a reduction of the EB effect. Irreversible training effects can also be related to the symmetry of the antiferromagnetic anisotropies and the inherent frustration of the interface [10]. Radu et al. [11] argued that the asymmetry is caused by interfacial domain formation (parallel to the interface) during the very first reversal. These interfacial domains serve as seeds for the subsequent magnetization reversals. Here, we show that the untrained state can be reinduced by going through a hysteresis loop with the applied magnetic field perpendicular to the cooling field direction without raising the temperature above T_N . This surprising effect is directly reflected by magnetization measurements performed with a superconducting quantum interference device (SQUID) magnetometer. High-resolution measurements of the magnetoresistance allow us to further elucidate this partial reversibility of the training effect.

In a FM layer the resistance depends on the angle between the magnetization and the current direction. This angle-dependent resistance is known as the anisotropic magnetoresistance (AMR) [12,13]. In a saturated FM layer, the AMR effect can be expressed as

$$R(\theta) = R_{\perp} + \triangle R_0 \cos^2(\theta), \qquad (1)$$

where R_{\perp} is the resistance with the magnetization perpendicular to the current and $\triangle R_0$ is the difference in resistance with the magnetization parallel and perpendicular to the current, respectively. The origin of the AMR effect is related to spin-orbit scattering. For the present study AMR measurements are performed to probe in detail the switching behavior of the CoO/Co bilayers for different subsequent hysteresis loops.

For the preparation of the CoO/Co bilayers a 20 nm thick Co layer is dc magnetron sputtered on top of an oxidized Si wafer with a typical deposition rate of 0.1 nm/s. The base pressure of the vacuum sputter cham-

ber is 10^{-7} mbar, while the working pressure for the Ar sputter gas is 10^{-3} mbar. After deposition, the Co layer is oxidized in situ for 2 min in a partial oxygen pressure of 10^{-3} mbar, which results in the formation of a 2 nm thick CoO top layer. For the SQUID magnetization measurements the sample is cooled in a field of +100 mT applied in the sample plane from above the Néel temperature of bulk CoO (291 K) to 10 K, which is well below the blocking temperature of our oxidized Co layers. After field cooling, the magnetic field is increased to +200 mT and two subsequent hysteresis loops [Fig. 1(a)] are measured with the field parallel to the cooling field. The first reversal at -100 mT is more abrupt, while all subsequent reversals are more rounded. This asymmetric behavior is typical for the training effect in CoO/Co and can be directly linked to a change in the magnetization reversal mechanism. Initially, domain wall nucleation and domain wall propagation govern the reversal, leading to a sudden change of the magnetization. The following more rounded reversals are dominated by a rotation of the magnetization [11,14]. This training effect can be understood as being the result of the splintering of the AFM into a collage of domains during the first reversal at negative fields [15]. Throughout field cooling the ferromagnetic Co layer consists of a single FM domain, which induces a uniform state in the AFM CoO. During the first reversal, the uniform FM



FIG. 1. SQUID magnetization measurements of a CoO/Co bilayer at 10 K after cooling in a field of +100 mT. The upper panel (a) shows the first and second hysteresis loops with the magnetic field applied in the direction of the cooling field. Panel (b) represents the subsequent two hysteresis loops when the magnetic field is applied perpendicular to the cooling field. The lower panel (c) shows the next two hysteresis loops with the magnetic field again applied along the cooling field direction. A reentry of the untrained state without heating the sample above the blocking temperature is observed.

Co magnetization is broken up, and via the exchange coupling at the CoO/Co interface this results in a torque acting on the CoO spins. As a result, the metastable uniform AFM state lowers its interfacial energy by splitting up into domains. The latter AFM domain structure will affect all subsequent magnetization reversals [16,17]. Figure 1(b) shows the subsequent two SQUID magnetization measurements of the hysteresis loop with the magnetic field perpendicular to the cooling field. Almost no EB or training effect is observed. The rotation of the exchange anisotropy was studied by Gredig et al. [18] where external fields are applied at different azimuthal angles with respect to the unidirectional anisotropy. Finally, when the external magnetic field is again applied along the cooling field direction, we surprisingly observe the reappearance of an asymmetric hysteresis loop. Remarkably, the untrained state can be partially reinduced by changing the orientation of the applied magnetic field, and this effect is obtained without heating the sample above the Néel temperature.

To further elucidate the partial reappearance of the untrained state, measurements of the AMR were performed. The AMR provides direct information about the domain configuration of the FM and, as a result of the pinning, also about the AFM. For the high-resolution magnetoresistance measurements we fabricate narrow stripes of CoO/Co using electron-beam lithography and lift-off techniques. After exposure and development of the resist layer, a CoO(2 nm)/Co(20 nm) bilayer is deposited by sputtering and subsequent in situ oxidation. Finally, the lift-off is performed by immersing the sample in a bath of hot acetone. In order to increase the sensitivity of our magneto resistance measurement, 2 μ m wide and 120 μ m long stripes are fabricated. Both ends of a stripe are connected to larger predefined Au contact pads to which we are able to attach the voltage and current leads by ultrasonic wire bonding. High-resolution four-terminal magnetoresistance measurements are performed in a helium flow cryostat by integrating the sample into an Adler-Jackson bridge. The ac measuring current for the lock-in detection has a frequency of 27.7 Hz and a root-mean-square (rms) amplitude of 3.5 µA.

The results of our magnetization measurements with a vibrating sample magnetometer (VSM) on an unpatterned CoO/Co reference film, which is deposited simultaneously with a CoO/Co stripe, are shown in Fig. 2. The sample is cooled from 300 to 5 K in an in-plane field of +400 mT. The first reversal in the decreasing field branch at -130 mT is very abrupt while all subsequent reversals are more rounded, in agreement with the results obtained with SQUID magnetometry for the CoO/Co sample discussed above.

Figure 3 shows the magnetoresistance measurements of the CoO/Co stripe after cooling from 300 to 10 K in a field of \pm 100 mT parallel to the stripe. After field cooling, the magnetic field is increased to \pm 700 mT and three subse-



FIG. 2. Hysteresis loops measured at 5 K with VSM magnetometry of a CoO/Co bilayer cooled in a field of +400 mT. The first reversal at negative field is dominated by domain wall nucleation and domain wall propagation and is abrupt. All subsequent reversals are dominated by rotation of the magnetization and are more rounded.

quent hysteresis loops are measured with the field parallel to the CoO/Co stripe. A smaller AMR effect (less rotation) is observed for the first reversal when compared to the subsequent reversals. These AMR results are consistent with our VSM magnetometry (see Fig. 2) as well as with previous results [19,20]. More interesting is the direct indication for the existence of magnetic domains in the Co layer. After field cooling and before passing through the first magnetization reversal in the descending field branch, the resistance in saturation reaches its maximum because all spins are oriented along the cooling field. After going through a complete hysteresis loop, the resistance at saturation is reduced (see right inset of Fig. 3), indicating that spins in the FM are rotated away from the cooling field. This is consistent with the presence of an interfacial domain structure in the FM [11]. These interfacial domains nucleate at the interface with the AFM, which is strongly coupled to the FM by the exchange interaction. Therefore, our AMR results are consistent with the fact that the AFM splits up into domains after the first reversal. As reported before [19], the training effect in CoO/Co bilayers depends on the thickness of the AFM layer. Bilayers with thicker CoO (thickness larger than 5 nm) reveal less training and relatively square hysteresis loops. In thinner CoO layers (thickness smaller than 5 nm) similar to our CoO layers, changes in the spin alignment of the AFM grains are possible because of their smaller magnetocrystalline anisotropy. As revealed by our measurements, the training effect in this type of film is consistent with the altering of the CoO spin structure. Quantitatively, the resistance in saturation is reduced by 1.6% after going through a complete hysteresis loop (inset, Fig. 3). Using Eq. (1) we find that such a reduction is consistent with the formation of domain walls parallel to the AFM-FM interface, where the domain walls extend over a few monolayers [11].

Our magnetoresistance measurements confirm that it is possible to partially reinduce the untrained state without heating the sample above the Néel temperature. This implies that the magnetic state obtained after field cooling is less irreversible and unique than generally accepted. Figure 4 shows two hysteresis loops along the cooling field direction after field cooling to 10 K in a field of +100 mT. After going through several hysteresis loops, a reversed



FIG. 3 (color). Field dependence of the magnetoresistance of a CoO/Co stripe at 10 K after cooling in a field of +100 mT applied along the length of the stripe. A smaller resistance change (less rotation) is observed during the first reversal when compared to the subsequent reversals. The insets compare the resistance at saturation to the maximum resistance (reference line), which ideally corresponds to the case that all spins are oriented along the cooling field direction.



FIG. 4 (color). Field dependence of the magnetoresistance of the CoO/Co stripe at 10 K after cooling in a field of +100 mT along the stripe. The blue line illustrates the reappearance of the training effect without any heating of the sample. This reappearance is achieved by going through a hysteresis loop with the magnetic field in the sample plane but perpendicular to the cooling field direction (not shown). The insets show the resistance at saturation when compared to the maximum resistance (reference line).

training effect can be achieved by going through a hysteresis loop with the magnetic field in the sample plane but perpendicular to the cooling field direction (not shown). After performing the loop in the perpendicular field, a hysteresis loop is measured with the field again applied along the cooling field direction. It is clear from Fig. 4 that the untrained state has been partially reinduced without any heating of the sample. The exchange bias field is increased and the amount of magnetization rotation in the descending field branch is reduced when compared to the trained reversals. An indication for the mechanism governing this partial reappearance of the untrained state can be obtained from the magnetoresistance at saturation (see right inset of Fig. 4). After performing the hysteresis loop in the perpendicular field, the initial magnetoresistance at saturation is again higher than the magnetoresistance after the trained reversal. From these results we conclude that performing a hysteresis loop in a field perpendicular to the cooling field alters or partially removes the FM domains. Because the AFM domains, which are coupled by a fraction of uncompensated interfacial spins [2,3] to the FM, are inducing the FM domains, it is very likely that the domain structure of the CoO is also altered by the application of the perpendicular field. When performing a hysteresis loop in a perpendicular field for the second time, we observe a similar behavior although the partial revival of the untrained state is less pronounced when compared to the revival after the first loop in a perpendicular field. A more detailed analysis of our results [21] indicates that the external field not only affects the AFM domain size distribution, but also induces a collective rotation of the AFM spins.

In conclusion, the results of our magnetization and magnetoresistance experiments demonstrate that it is possible to partially reinduce the untrained state in an exchange biased CoO/Co structure. A clear increase in exchange bias field and a reduction in the amount of magnetization rotation is observed after performing a hysteresis loop in a magnetic field perpendicular to the cooling field direction. This surprising result can be explained by a change in the magnetic domain structure in the antiferromagnetic CoO layer by the application of the perpendicular field. The presence of antiferromagnetic domains is confirmed by a careful inspection of the magnetoresistance data at saturation.

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- [1] W.H. Meiklejohn and C.P. Bean, Phys. Rev. **102**, 1413 (1956).
- [2] H. Ohldag, A. Scholl, F. Nolting, E. Arenholz, S. Maat, A. T. Young, M. Carey, and J. Stöhr, Phys. Rev. Lett. 91, 017203 (2003).
- [3] P. Kappenberger, S. Martin, Y. Pellmont, and H. J. Hug, Phys. Rev. Lett. **91**, 267202 (2003).
- [4] J. Nogués and I. K. Schuller, J. Magn. Magn. Mater. 192, 203 (1999).
- [5] A.E. Berkowitz and K. Takano, J. Magn. Magn. Mater. 200, 552 (1999).
- [6] R.L. Stamps, J. Phys. D 33, R247 (2000).
- [7] M. Kiwi, J. Magn. Magn. Mater. 234, 584 (2001).
- [8] U. Nowak, K. D. Usadel, J. Keller, P. Miltényi, B. Beschoten, and G. Güntherodt, Phys. Rev. B 66, 014430 (2002).
- [9] J. Keller, P. Miltényi, B. Beschoten, G. Güntherodt, U. Nowak, and K. D. Usadel, Phys. Rev. B 66, 014431 (2002).
- [10] A. Hoffmann, Phys. Rev. Lett. 93, 097203 (2004).
- [11] F. Radu, M. Etzkorn, R. Siebrecht, T. Schmitte, K. Westerholt, and H. Zabel, Phys. Rev. B 67, 134409 (2003).
- [12] I. A. Campbell and A. Fert, in *Ferromagnetic Materials*, edited by E. P. Wohlfarth (North-Holland, Amsterdam, 1982), Vol. 3.
- [13] T. R. McGuire and R. I. Potter, IEEE Trans. Magn. 11, 1018 (1975).
- [14] M. Gierlings, M. J. Prandolini, H. Fritzsche, M. Gruyters, and D. Riegel, Phys. Rev. B 65, 092407 (2002).
- [15] U. Welp, S. G. E. te Velthuis, G. P. Felcher, T. Gredig, and E. D. Dahlberg, J. Appl. Phys. 93, 7726 (2003).
- [16] F. Nolting et al., Nature (London) 405, 767 (2000).
- [17] H. Ohldag, T.J. Regan, J. Stöhr, A. Scholl, F. Nolting, J. Lüning, C. Stramm, S. Anders, and R. L. White, Phys. Rev. Lett. 87, 247201 (2001).
- [18] T. Gredig, I. N. Krivorotov, C. Merton, A. M. Goldman, and E. D. Dahlberg, J. Appl. Phys. 87, 6418 (2000).
- [19] T. Gredig, I. N. Krivorotov, and E. D. Dahlberg, J. Appl. Phys. 91, 7760 (2002).
- [20] M. Gruyters, J. Appl. Phys. 95, 2587 (2004).
- [21] F. Radu, H. Zabel, S. Brems, D. Buntinx, K. Temst, and C. Van Haesendonck (unpublished).