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Magnetization reversal in trained exchange biased multilayers

Amitesh Paul[1], **Thomas Brückel, Emmanuel Kentzinger and Ulrich Rücker**

Institut für Festkörperforschung, Forschungszentrum Jülich GmbH, D-52425 Jülich, Germany

E-mail: A.Paul@fz-juelich.de

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Abstract

FM magnetization may be collinear (responsible for nonuniform reversal) or noncollinear (responsible for uniform reversal) to the direction of the applied field in ferromagnetic–antiferromagnetic (FM–AF) exchange-biased systems depending upon the number of field cycles it has undergone. Usually noncollinearity sets in right after the first field cycle, i.e. when the system is in the trained state. Here we show that in case of polycrystalline multilayers (MLs) of continuous AF–FM interfaces (ML-C), e.g. in $[Co/CoO]_{20}$, this collinearity of the FM magnetization remains not only in the untrained state but *even in the trained state* as each FM layer in the ML remagnetizes symmetrically for both field branches via the nonuniform mode only. Thus the state of magnetization can remain virtually unaffected by repeated field cycling, and this can be exploited in building stable state spin-valve systems.

(Some figures in this article are in colour only in the electronic version)

Direct exchange coupling, which 'locks' the magnetization in a certain direction between the ferromagnet (FM) and antiferromagnet (AF) layers, gives rise to a unidirectional magnetic anisotropy called the exchange bias, H_{EB} [1]. This is realized when a FM in contact with an AF is cooled below the blocking temperature T_B of the AF in an external field H_{FC}. For a magnetic sensor based upon giant magnetoresistance (GMR) in spin-valve structures [2], a large H_{EB} of the locked layer is used to yield a well-defined response over a wide range of field strengths and directions. Thus exchange bias is an integral part for the development of spintronic devices. Such a device, however, is unable to withstand the effect of repeated field cycles due to *training* effects in such systems [3].

The whole phenomenon of exchange bias is dependent upon the state of the interface in which the AF or FM spins are frozen as they are field cooled, as in the case of similar bulk

[1] Author to whom any correspondence should be addressed.

AFM domains. After the first field cycle the system relaxes into a so-called 'trained state'. This results in a reduction of the H_{EB} [4] along with a decrease in the coercivity and has been observed to vary with the number of cycles of the hysteresis loop. Rearrangement of the AF domain structure has been suggested to be connected to the loss of magnetization in the AF and thereby the magnitude of H_{EB}. Training effects can be observed in epitaxial as well as in polycrystalline systems. In spite of its technological importance, understanding of the basic mechanism of exchange bias [5, 6] has remained elusive for half a century.

Among the various systems showing exchange bias, Co(FM)/CoO(AF) bilayer systems have been studied extensively by various groups [7–9]. Here we define various combinations of AF–FM interfaces which can be realized from a bilayer AF–FM unit. These can be of four types: (i) a single bilayer (BL); (ii) a stack of AF–FM bilayers separated by a non-magnetic layer (ML-S); (iii) a FM layer sandwiched between two AF layers and separated by a non-magnetic layer (ML-T); and (iv) a continuous stack of alternating FM and AF layers (ML-C) [9]. ML-S basically represent a BL as all the characteristics are expected to remain similar across a reasonably thick non-magnetic, e.g. Au, layer where the magnetism is multiplied by the number of repeats in the stack. A ML of the type ML-C, however, cannot be considered as independent units of AF–FM interfaces. This makes the ML-C more complicated and also more interesting [9]. Here we discuss our results for ML-C type MLs.

Exchange bias is regarded as negative (positive) and is set by convention as a shift of the hysteresis loop in the opposite direction (in the same direction) to the field cooling direction. Thus a negative exchange bias can be achieved for $H_{FC} = +4.0/-4.0$ kOe during the first field cycle. The first cycle of the field sweeping direction is the cycle when the applied field starts from the same direction in which the system has been field cooled. For example, if the system is cooled in the negative direction (-4.0 kOe in the present case), then to observe the first cycle one should start from a field in the negative direction only. This way one can complete the full cycle with the applied field from negative to positive (decreasing branch: the H_{FC} direction is opposite to the H_a) and then from positive to negative (increasing branch: the H_{FC} direction is in the same direction as H_a) fields. The same cycle is repeated for the second cycle to measure the bias in the *trained* state. If one opts to field cool in the negative direction and start the field sweeping from the positive direction, then the state of the specimen observed is already in the trained state.

Interestingly, asymmetric reversal mechanisms are observed in BL or ML-S type bilayers for the *first* field cycle or in the untrained state—as domain wall motion (nonuniform reversal) occurs along the decreasing branch and magnetization rotation (uniform reversal) occurs along the increasing branch of the hysteresis loop. For a *trained* sample on the other hand, i.e. in the second-half of the first field cycle (increasing branch) and the second cycle of the hysteresis loop, without altering the field cooling history, the magnetization reversal always proceeds *symmetrically via magnetization rotation* for both loop branches [10, 11].

The numerical simulations of Hoffmann [12], in a BL system, have suggested an inherent frustration which can lead to perpendicular/noncollinear arrangement of AF sublattice moments with respect to the H_{FC} direction—'stabilized by the net moment of the noncollinear configuration of the AF sublattices'. Such an arrangement is broken after the first field half-cycle (decreasing branch), as the system relaxes to an antiparallel configuration of the sublattice AF magnetization. The FM magnetization (M_{FM}), being coupled to the AF, is closely collinear ($\parallel$ or anti-$\parallel$ to the H_a direction) during the first field cycle and therefore flips with an external applied field (H_a). After the first field cycle, the FM magnetization direction has been shown to generally vary from 45°–135° which would favour its rotation with increasing H_a. Such a variation follows from the collinearity of the AF sublattice moments. However, such perpendicular/noncollinear arrangement of AF sublattice moments in CoO is difficult to achieve

since its anisotropy is very large. Moreover, it has been observed experimentally [8] that after field cooling there is no trace of interfacial noncollinearity between CoO and FM layers.

Neutron scattering under grazing incidence with polarization analysis has been proven decisive for identification of the reversal mechanism in exchange bias systems [7–10, 13]. For one-dimensional analysis four different cross sections can be distinguished, namely non-spin flip (R_{++} and R_{--}) and spin flip (SF) channels (R_{+-} and R_{-+}). Here subscripts $+$ and $-$ represent the polarization parallel and antiparallel to the guiding field respectively. $R_{++/--}$ contains the sum/difference between the nuclear and magnetic scattering, whereas the SF signal contains only the magnetic information. Magnetization rotation is identified by a significant increase in the specular SF reflectivities, which corresponds to in-plane magnetization components perpendicular to the guiding field H_a applied collinear to H_{FC}. Reversal by domain nucleation and propagation do not provide enhanced SF intensities, because the magnetization is always collinear to H_a.

Very recently, Paul *et al* showed symmetric and sequential reversal in two different exchange biased polycrystalline multilayers (MLs) of the type ML-C with two different combinations of AF and FM layers. For both systems, $[IrMn/CoFe]_{10}$ [14] and $[CoO(7.0\,nm)/Co(11.0\,nm)]_{20}/Au(50.0\,nm)$ [9], exchange bias field strength has been seen to evolve through the stack of the ML. This evolution was related to the decreasing domain sizes mediated by their respective grain sizes [14]. Here, sequential refers to the reversal of layer magnetizations one after the other due to increasing or decreasing bias fields along the stack. Thus the sequential switching was proved to be universal in MLs. In the present paper we investigate the same CoO/Co ML along a full magnetization loop by specular and off-specular (not shown) polarized neutron reflectometry (PNR), but for the *trained* state, and compare it with that of the untrained state. Details of the sample preparation and instrument and method used for neutron measurements were discussed previously [9, 15]. Magnetization loops are measured by means of a superconducting quantum interference device (SQUID) at 10 K after field cooling in an external field of ± 5 kOe from room temperature to 10 K, i.e. well below the $T_B \approx 220$ K of CoO. It may be noted that the magnetization behaviour of our CoO films is comparable to ideal stoichiometric oxides as we compare our BL or ML-S samples with the theoretical loops [12, 15].

Figure 1 shows SQUID magnetization loops for the specimens cooled down to 10 K at $H_{FC} = -4.0$ kOe for a few cycles. Our coercive field values are in accordance with earlier results [16, 17] as we found no change in H_{C2} (increasing branch) but a decrease in H_{C1} (decreasing branch) by 80 Oe up to the second cycle. Thus the exchange bias field H_{EB} was reduced from 435 to 395 Oe after the first field cycle. Training effects in polycrystalline samples are supposed to be common as they have multiple equivalent easy magnetization axes [12]. A step around 0.7 kOe is seen in the decreasing branch—probably caused by some unoxidized Co within the CoO layers [15] and obviously not related to any effect due to decreasing domain size as has been suggested earlier when observed on patterned samples [18]. It is important to note that we could regain the coercive field of the untrained state after heating the sample above its T_B of 220 K and cooling down again to 10 K. We could not find any vertical shift [4, 19] which can be correlated with the decrease in sublattice AF magnetization with subsequent cycles, neither could we find any decrease in exchange bias after the second loop as predicted by the model of Binek [20] which was for a BL system.

All four polarization channels of the specular reflectivities are measured at different external fields H_a, three of which are shown in figure 2 (circled numbers in figure 1) together with least-square fits [9]. The intensity maps for two channels (NSF and SF) at some representative fields are shown in figure 3 (circled numbers in figure 1) for (a) untrained and (b) trained specimens. The specular reflectivities (NSF and SF) are extracted from the

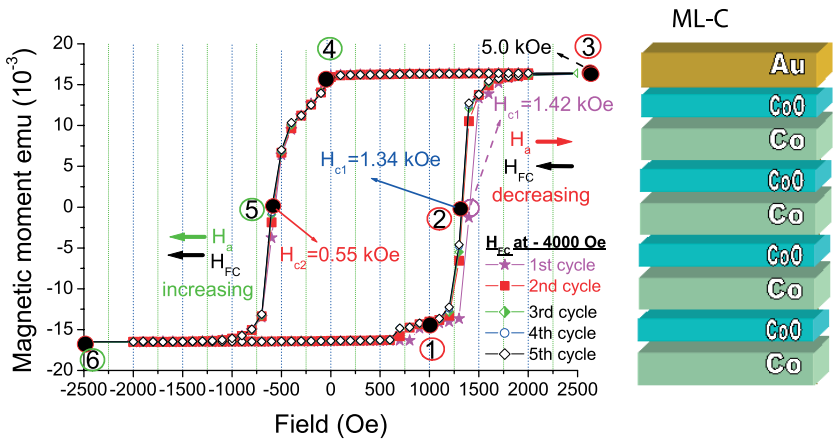

Figure 1. SQUID hysteresis loops at 10 K for the ML-C: SiO_2/[Co(11.0 nm)/CoO(7.0 nm)]$_{\times 20}$/Au(50.0 nm) ML when field cooled at −4.0 kOe for a few cycles. The filled circles (black) are the field values for which we show the PNR spectra during the second cycle. $H_{C1} = 1.34$ kOe for the decreasing branch in the second cycle (blue script) whereas $H_{C2} = 0.55$ kOe for the increasing branch (red script) which remains the same for both the first and second cycles. The open circle (magenta) in the decreasing branch shows the reversal point during the first cycle $H_{C1} = 1.42$ kOe (magenta script).

intensity maps integrating the intensities along the specular region. The intensity maps show the possibility of our instrument simultaneously measuring the specular and the off-specular intensities.

The three peaks of the ML in the NSF channels (R_{++} and R_{--}) are the first-, second- and third-order Bragg reflections of the ML. The corresponding peaks in the SF channels (R_{+-} and R_{-+}) are solely due to polarization inefficiencies of our set-up (this is evident as we compare the intensities with that of the saturated state in panels (③) and (⑥) of figure 2. Unlike other groups, we do not present the data subtracted from the contribution due to instrumental inefficiencies (our polarizer and analyser efficiencies are around ≈95%) for a direct comparison of the raw data. R_{++} and R_{--} are almost equal at $H_a = 1300$ Oe (panel ②) on the decreasing and $H_a = 550$ Oe (panel ⑤) on the increasing branch, signifying the respective reversal points. For all other fields R_{++} or R_{--} dominates and reflects a net magnetization collinear with H_a, while the SF intensities always remain in the background. Therefore, the absence of any SF processes throughout the field cycle strongly indicate the absence of any coherent rotation, even in the trained state.

The details of the fitting of the specular intensities (considering deviations from the purely collinear, single domain configurations, i.e. $\theta_i = 0°$ or 180°) have been described earlier in [9, 14]. In our model we neglect any uncompensated magnetization contribution from the AF layers considering them to be lower than the detectable limit. We find the magnetization reversal mechanism to be strikingly similar to that for the first field cycle as observed and discussed earlier [9]: decreasing H_a switches the Co layers sequentially from the top to the bottom, and on increasing H_a the reversal proceeds in the opposite direction. Figure 4 shows the layer switching sequence for the first and second field cycles. This symmetric magnetization reversal process—without coherent rotation, which remains similar and stable for the second field cycle, i.e. in the *trained* state—is an exciting observation and is different from any earlier observations [10, 11]. We also confirm this observation for other similar systems,

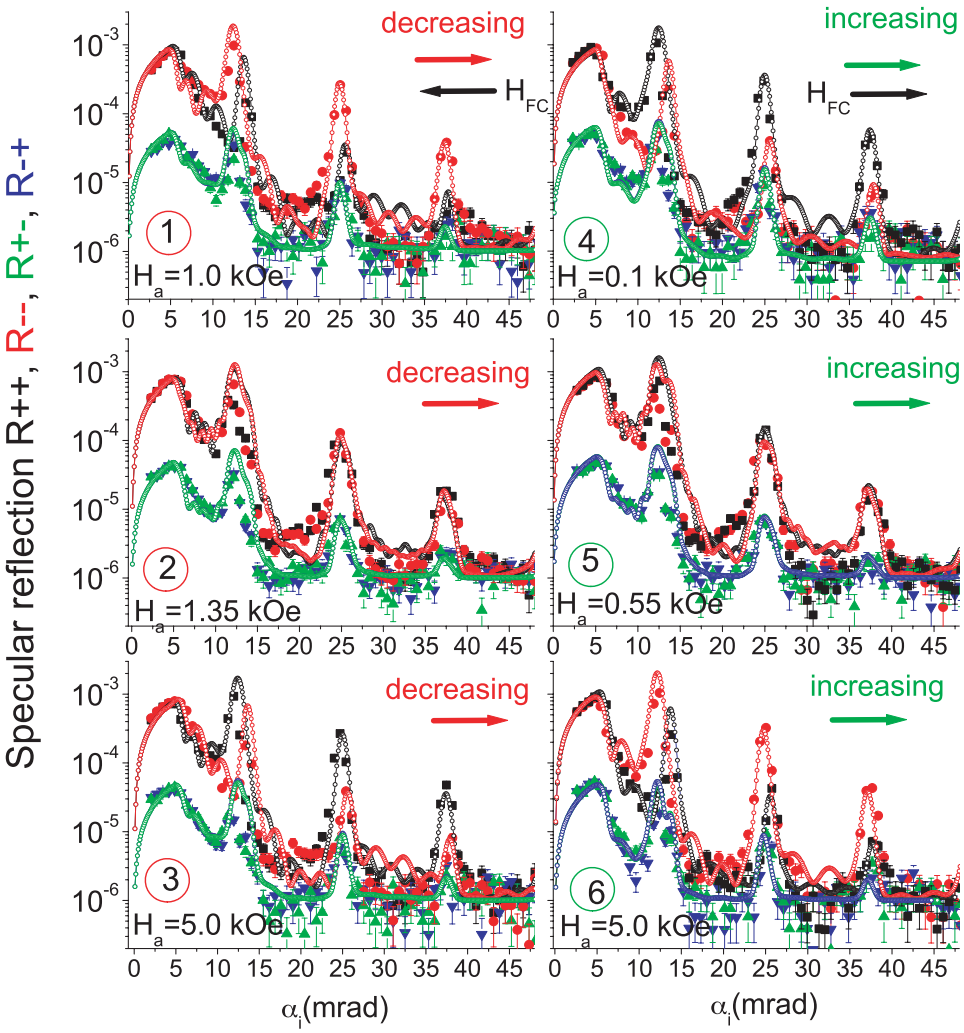

Figure 2. Measured (solid symbols) and fitted (open circle) NSF (R_{++} (black square) and R_{--} (dark grey/red circle)) and SF (R_{+-} (light grey/green triangle) and R_{-+} (black/blue inverted triangle)) reflectivity patterns of the ML-C: SiO_2/[Co(11.0 nm)/CoO(7.0 nm)]$_{\times 20}$/Au(50.0 nm) ML at different applied fields H_a along increasing/decreasing branches of the hysteresis loops for the second field cycle, where $H_{FC} = -4.0$ kOe and measured at 10 K. The numbers in circles correspond to the applied fields on the hysteresis loop in figure 1 along the two branches during the second cycle. No SF signal can be observed as we compare the signal for other fields with that of the saturation state at 5.0 kOe where all the SF are due to non-ideal polarization only.

e.g. [IrMn/CoFe]$_{10}$ [14]. An earlier report of reversal in a similar system [21] lacks any data from the trained state during the second cycle and most importantly the measurements were done at 240 K which is very close to the blocking temperature of the AF (≈250 K [21] or below [9]) and is therefore probably ambiguous.

We have measured at least seven spectra within a field range of 0.03–0.08 kOe covering the reversal regime of around 500 Oe for the increasing field branch and five spectra for a range of 1.25–1.5 kOe (reversal regime of around 250 Oe) along the decreasing branch. Each spectrum was analysed separately with the parameters from the previous field spectra starting from the

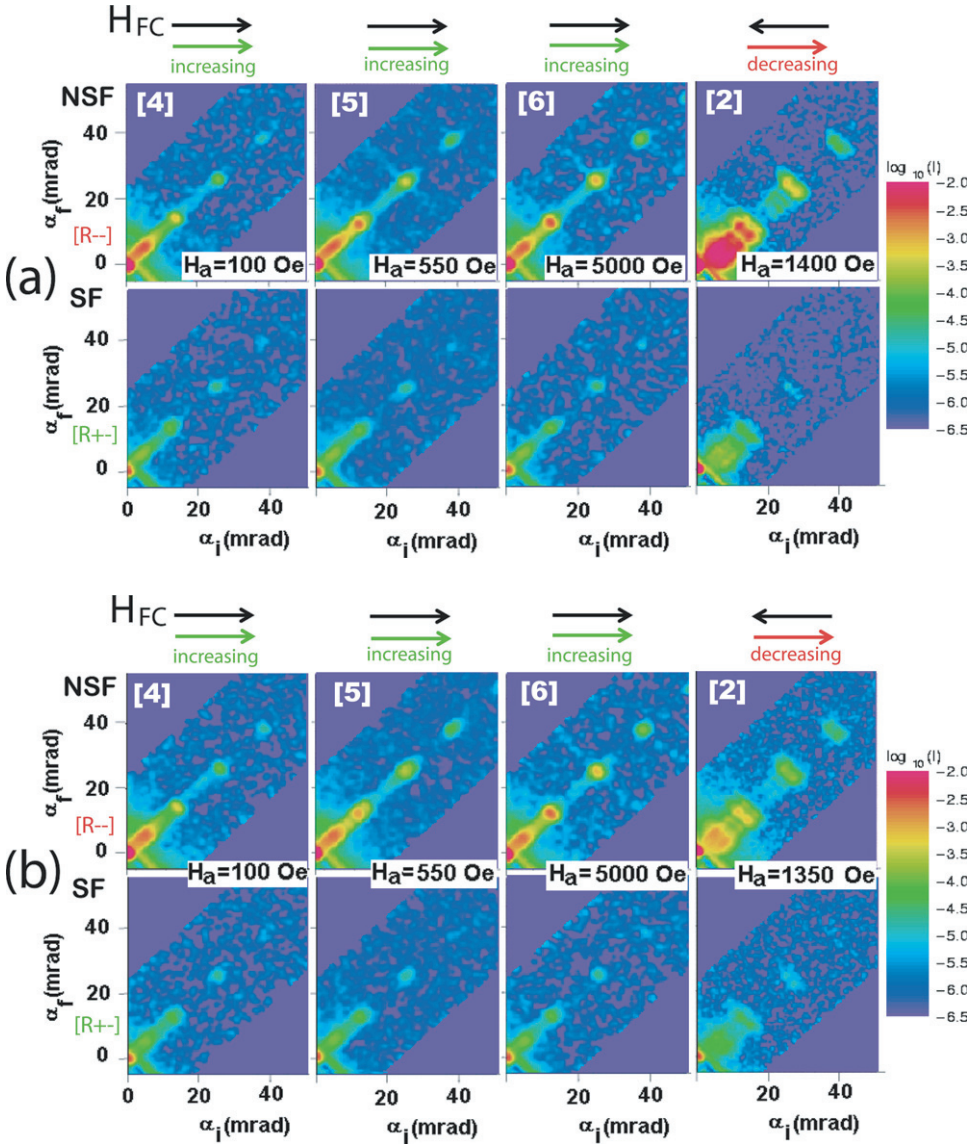

Figure 3. NSF and SF intensity maps of the ML-C: $SiO_2/[Co(11.0\,nm)/CoO(7.0\,nm)]_{\times 20}/Au(50.0\,nm)$ ML at different applied fields H_a along increasing/decreasing branches of the hysteresis loops for (a) the first and (b) the second field cycles, where $H_{FC} = -4.0$ kOe and measured at 10 K. The numbers correspond to the applied fields on the hysteresis loop in figure 1 along the two branches during the cycles. No SF signal can be observed as we compare the signal for other fields to that of the saturation state at 5.0 kOe where all the SF are due to non-ideal polarization only. The splitting of the specular intensity in the decreasing branch is purely instrumental and has been properly taken care of during the data analysis.

remanence state to the saturation state of the layers and vice versa. The spectra are seen to be highly sensitive to the orientation of magnetization direction for each and every layer. This has enabled us to identify if any transverse component is present or absent in any of the layers in the stack. This is particularly visible in the SF channels of the spectra. We show as an example

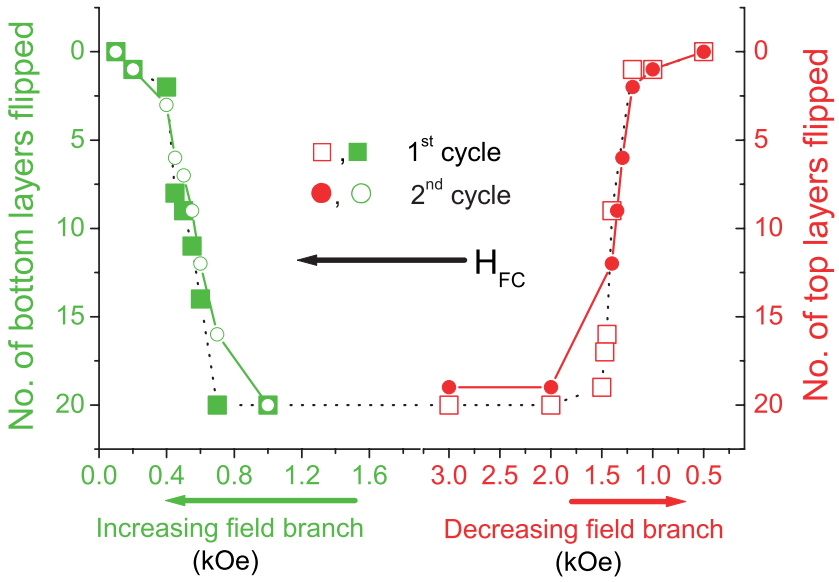

Figure 4. Layer switching sequence for the first cycle (from [9]) and second cycle of field sweeping along increasing/decreasing branches of the hysteresis loops of ML-C.

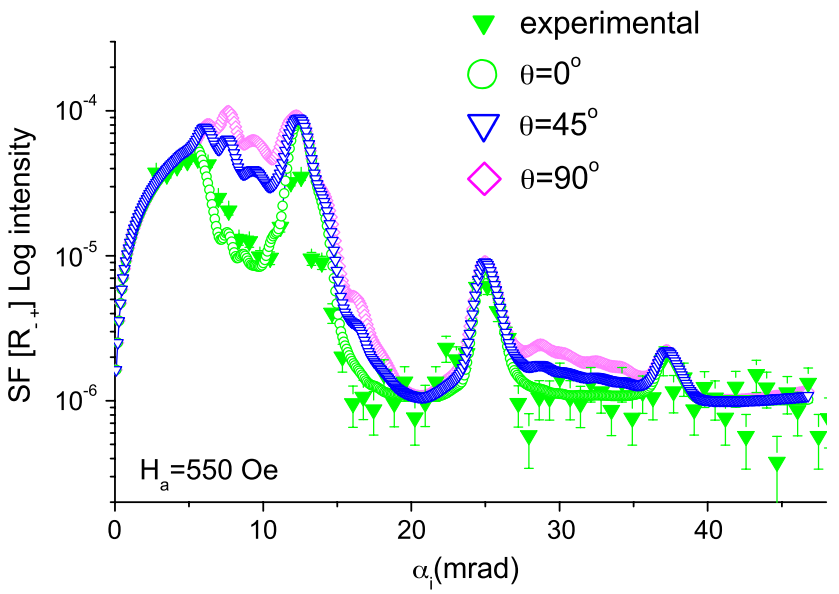

Figure 5. Simulated spectra of the ML considering three different orientations of magnetization (θ) for, e.g., (i) 0° (which is the fitted curve to the experimental data), (ii) 45° and (iii) 90° for the seventh layer (arbitrarily chosen) in the stack at an applied field of 0.55 kOe along the increasing branch. The simulations clearly show the sensitivity of measurement due to the magnetization orientation of a single layer in the whole stack.

in figure 5 the sensitivity of the direction of the magnetization component perpendicular to the applied field on the SF channel (R_{-+}). Here we simulate the spectra of the ML considering

three different orientations of magnetization (θ) such as (i) 0° (which is the curve fitted to the experimental data), (ii) 45° and (iii) 90° for the seventh layer (arbitrarily chosen) in the stack at an applied field of 0.55 kOe along the increasing branch. The simulations for the SF channels (particularly for lower momentum transfer range) clearly show the influence on the overall intensity profile of magnetization orientation of a single layer out of 20 layers in the stack.

Our results indicate that the M_{FM} remain $\parallel$ or anti-$\parallel$ to the H_a direction and is independent of the number of field cycles [12]. Now, if we consider a $\perp$/noncolliner configuration of sublattice AF magnetization which remains stable even after the first field cycle, then we should not observe any training effect. Alternatively, if we consider the initial arrangement to be broken then it is perhaps not instrumental in deciding the magnetization reversal mechanism in subsequent cycles. Moreover, our AF layer thickness is 7.0 nm, which is above the thickness for which one observe saturation in H_{EB} where also no training effect would have been expected (experimentally verified). Thus as we already observe some training effects we can think of some local rearrangement of spins. Such rearrangements or magnetic training due to the magnetic roughness was predicted earlier [22], but due to the limited probing range of magnetic correlations for our instrument [23] we could not observe any spin-dependent diffuse scattering from our sample. This situation therefore leads to the possibility that the coupling between the M_{FM} and the sublattice AF magnetization is weaker than the antiferromagnetic coupling between the two sublattice magnetizations. But this would lead to the breakdown of the conditions for the frustrated state, which requires a comparable coupling between sublattice AF magnetization and the coupling of M_{FM} to the sublattice AF magnetization, resulting in an initial collinear sublattice AF magnetization.

It has been proven that the exchange interaction at the FM–AF interface is antiferromagnetic [6, 24] for Fe/FeF_2 or Co/FeF_2 systems. However, recent results of x-ray resonant scattering on Fe/CoO systems showed that the coupling may also be ferromagnetic [25]. A ferromagnetic coupling cannot be considered to explain the so-called positive exchange bias as coupling can also be influenced by the field cooling strength [24, 26]. Therefore, assuming an antiferromagnetic coupling, as M_{FM} is expected to follow the H_{FC} direction (the AF moments remain antiparallel to H_{FC}), they start to flip direction with increasing H_a. One may consider that the AF coupling between the M_{FM} and the pinning AF moments is weakened after the first cycle; this will cause the M_{FM} to rotate back with applied field as is usually seen for ML-S type MLs. However, in the present case of ML-C type MLs, the AF–FM antiferromagnetic coupling may not be weakened even after repeated field cycles, and is probably stabilized by the successive interfaces of AF and FM layers. This can explain the flipping of FM layers (collinearity of FM layers) in ML-C even in the trained state.

In conclusion we have investigated the remagnetization behaviour of trained state AF–FM interfaces in CoO–Co exchange-biased MLs and compared it with that of the untrained state. The magnetization reversal is seen to proceed very similarly for the untrained and trained states. The FM layer magnetization always remain collinear to the direction of the applied field in the case of continuous MLs. Any rearrangements of AF sublattice magnetization, after field cycling, do not necessarily define the magnetization reversal process. Technologically we found a stable configuration of the magnetization state in our ML which can make it a potential candidate for spin-valves—virtually unaffected by unlimited field sweeping.

References

[1] Meiklejohn W H and Bean C P 1956 *Phys. Rev.* **102** 1413
[2] Paul A *et al* 2005 *J. Appl. Phys.* **97** 223910
[3] Paccard D *et al* 1966 *Phys. Status Solidi* **16** 301

[4] Keller J *et al* 2002 *Phys. Rev.* B **66** 14431
[5] Kiwi M *et al* 2001 *J. Magn. Magn. Mater.* **234** 584
[6] Ohldag H *et al* 2006 *Phys. Rev. Lett.* **96** 27203
[7] Gierlings M *et al* 2002 *Phys. Rev.* B **65** 92407
[8] Radu F *et al* 2003 *Phys. Rev.* B **67** 134409
[9] Paul A *et al* 2006 *Phys. Rev.* B **73** 92410
[10] Lee W-T *et al* 2002 *Phys. Rev.* B **65** 224417
[11] Zhang K *et al* 2001 *J. Appl. Phys.* **89** 6910
[12] Hoffmann A 2004 *Phys. Rev. Lett.* **93** 97203
[13] Fitzsimmons M R *et al* 2000 *Phys. Rev. Lett.* **84** 3986
[14] Paul A *et al* 2004 *Phys. Rev.* B **70** 224410
Paul A *et al* 2005 *Eur. Phys. J.* B **45** 249
[15] Paul A *et al* 2004 *Europhys. Lett.* **70** 224410
[16] Radu F *et al* 2002 *J. Magn. Mater.* **240** 251
[17] Hochstart A *et al* 2002 *Phys. Rev.* B **66** 92409
[18] Girgis E *et al* 2003 *Phys. Rev. Lett.* **91** 187201
[19] Nogues J *et al* 2000 *Phys. Rev.* B **61** 1315
[20] Binek C *et al* 2004 *Phys. Rev.* B **70** 14421
[21] Radu F *et al* 2002 *Appl. Phys.* A **74** S1570
[22] Velthuis S G E *et al* 2000 *J. Appl. Phys.* **87** 5046
[23] Paul A *et al* 2006 *Phys. Rev.* B **73** 94441
[24] Nogues J *et al* 1999 *J. Magn. Magn. Mater.* B **192** 203
[25] Radu F *et al* 2006 *J. Magn. Magn. Mater.* **300** 206
[26] Paul A *et al* 2007 unpublished