Switching of the exchange bias in Fe/Cr(211) double-superlattice structures

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The reversal of the direction of the exchange bias in a ''double-superlattice'' system which consists of an Fe/Cr antiferromagnetic (AF) superlattice which is ferromagnetically coupled with an Fe/Cr ferromagnetic (F) superlattice through a Cr spacer layer, is observed. Magnetometry and polarized neutron reflectometry show that a switch in the bias direction occurs at a field (~447 Oe) well below the field (14 kOe) necessary to saturate the AF superlattice and well below the field (2 kOe) where the AF superlattice initiates a spin–flop transition. The switching of the exchange bias cannot be explained in terms of a model of uniform rotation, but rather by breakdown into domains and reversal of the AF layers. The transparency of magnetic behavior of the double superlattice may be useful in understanding the behavior of traditional exchange bias systems. © *2000 American Institute of Physics.* [S0003-6951(00)00240-0]

Exchange bias, first discovered in 1956 by Meiklejohn and Bean[1] in assemblies of Co–CoO particles, refers to the occurrence of a unidirectional magnetic anisotropy that manifests itself in shifted hysteresis loops for coupled ferromagnet (F)–antiferromagnet (AF) field cooled through the Néel temperature T_N of the AF. Once the system is cooled, the exchange bias field is frozen. Applying laboratory fields in the opposite direction does not change the orientation of the bias. The only way to reverse the bias is to warm up the sample above the Néel temperature or, more exactly, the blocking temperature. No systematic study has been made of applying increasingly high field just below the blocking temperature. Some experiments, however, are proceeding along this direction.[2]

We studied the reversal of the exchange bias in a ''double superlattice.'' This system consists of one F superlattice of Fe and Cr(211) layers, and one AF superlattice obtained similarly but with a different Cr thickness t_{Cr} (since the interlayer exchange coupling oscillates with t_{Cr}). The coupling between the AF and F superlattices is governed by the value of t_{Cr} between the two superlattices. The sample has a layer sequence $[Fe(50\,Å)/Cr(20\,Å)]^F{}_5/[Fe(14\,Å)/Cr(11\,Å)]^{AF}{}_{20}$ with $t_{Cr}=20\,Å$ between the F and AF superlattices, to provide a ferromagnetic intersuperlattice coupling. A uniaxial anisotropy is introduced by epitaxially growing the sample via dc magnetron sputtering onto single-crystal MgO(110) substrates.[3] This artificial exchange bias system was constructed in order to attain a predictable and controllable exchange bias. We recall here the main magnetic properties,[4–6] and show how the switching of the exchange bias was obtained.

Figure 1 gives the magnetization of the sample at room temperature with the field H applied along the easy axis. Above 14 kOe the magnetic moments of all layers in both superlattices are aligned with H (in Fig. 1 the magnetization is normalized to this value). For descending H the magnetization decreases as the Fe layers in the AF superlattice first enter a spin-flop state,[7,8] and then become antiferromagnetically aligned. Below 2 kOe the magnetic moments of all layers are again collinear, and are aligned along the easy axis and H. However, for modest negative fields, the FM layers turn toward the field while the magnetic structure of the AF layers remains unaltered. The exchange bias for descending fields of the order of 40 Oe has the value expected[4] for the ferromagnetic coupling between the two superlattices. The goal of the present experiment was to determine at which field the bias switches.

A series of magneto-optic Kerr effect (MOKE) minor loops were taken between the turning points H_{max} and H_{min}, where H_{max} was greater than the saturation field, while H_{min} was increased in magnitude from −45 Oe to −14 kOe in steps of 5 Oe. Figure 2 gives the measured MOKE signal as a function of field for two critical values of H_{min}. The figure shows that for $H_{min}=-406$ Oe, the loop is biased around $-H_E=-38.5$ Oe, and the magnetization in the F superlattice is reversed back to its original orientation at $H=-H_E+H_c=-33.6$ Oe. This loop is representative for all loops measured with values of H_{min} between −34 and −406 Oe. However, for $H_{min}=-447$ Oe to −14 kOe, the F superlattice magnetization does not reverse back until $H=H_E+H_c$

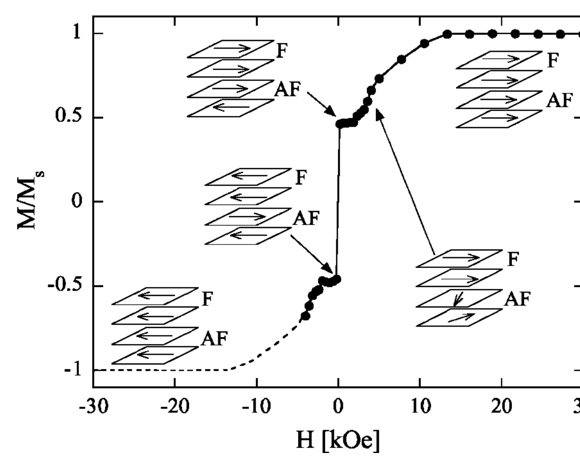

FIG. 1. The normalized magnetization curve measured with a superconducting quantum interference device magnetometer and with decreasing field. The sets of four arrows indicate the magnetic orientation in the F superlattice (top two) and in the AF superlattice (bottom two), at different stages of the magnetization curve.

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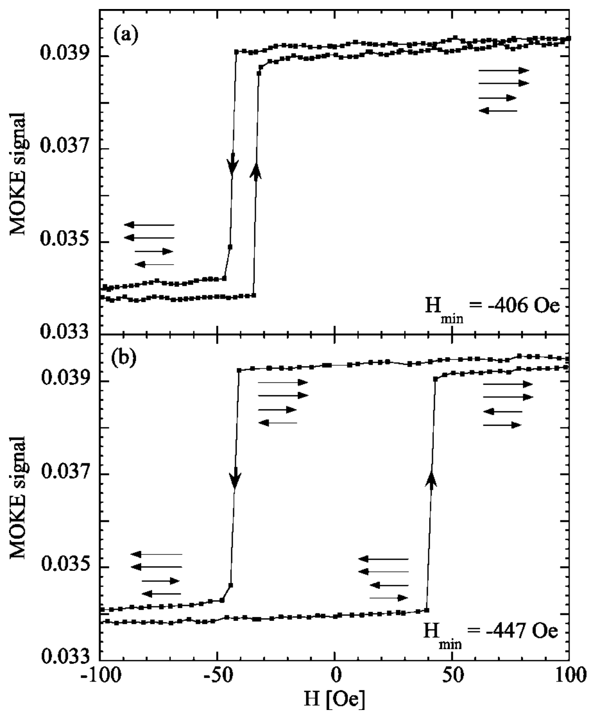

FIG. 2. Minor magnetization curves measured by means of MOKE. Both curves are measured for descending field from $H = 14$ kOe down to H_{min}, with (a) $H_{min} = -406$ and (b) -447 Oe, and then ascending back up to $H = 14$ kOe. The sets of four arrows indicate the magnetic orientation in the F superlattice (top two) and in the AF superlattice (bottom two), at different stages of the magnetization curve.

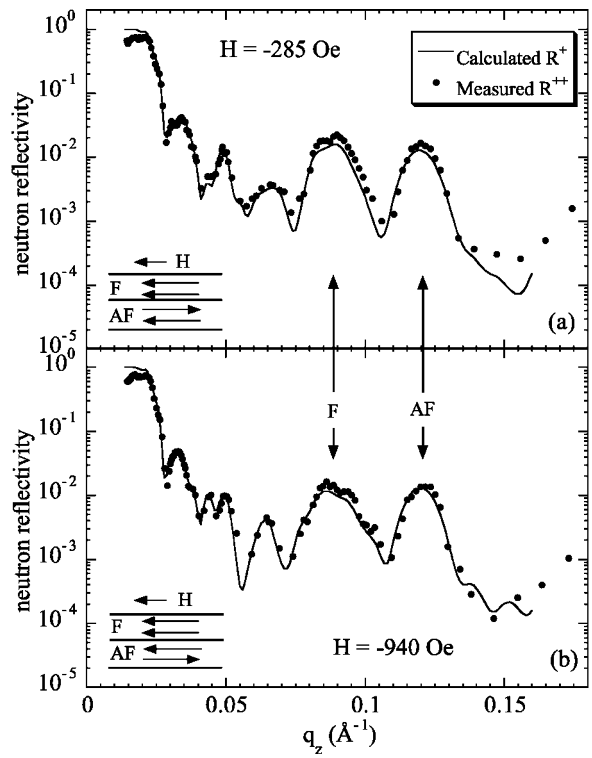

FIG. 3. Measured and calculated polarized neutron reflectivity for (a) $H = -285$ and (b) -940 Oe. The measured reflectivity is given for neutron with an initial and reflected polarization parallel to the applied field (R^{++}). The calculations are exactly the same as presented in Ref. 5. Since it is assumed all moments are collinear, $R^{++} = R^{+}$.

$= +40.7$ Oe, indicating a change in the direction of the bias, which implies that the AF superlattice has reversed its orientation. At intermediate values of H_{min}, the loop was either the same as for $H_{min} = -406$ Oe, or it was an average of the two loops.

The reversal of the AF superlattice was directly determined by polarized neutron reflectivity (PNR). The measurements were performed at Argonne's intense pulsed neutron source. The spin-dependent neutron reflectivity gives information about the magnetic and structural profile perpendicular to the surface. R^{+} and R^{-} denote reflectivities for neutrons polarized parallel and antiparallel to H, respectively. If polarization analysis of the reflected beam is performed, four intensities are measured, two nonspin flip: R^{++}, R^{--}, and two spin flip: R^{+-}, R^{-+}, reflectivities, where $R^{+} = R^{++} + R^{+-}$ and $R^{-} = R^{-+} + R^{--}$. If the magnetization of all layers is collinear to H, then $R^{-+} = R^{-+} = 0$. In this case R^{+} is an optical transform of $n(z) + m(z)$, where n is a depth-dependent nuclear scattering amplitude, m is the depth-dependent magnetization, and R^{-} is an optical transform of $n(z) - m(z)$. By alternatively measuring with neutrons in either spin state, the magnitude and direction of the layer-by-layer magnetization can be determined. If $R^{-+} = R^{-+} \neq 0$, there are components of the magnetization perpendicular to H.

It has been shown[4,5] that the polarized neutron reflectivity for the two magnetic configurations measured at $H = 166$ and -72 Oe (after saturation in $H = 30$ kOe) illustrate the reversal of the magnetization in the F superlattice. Here PNR measurements, performed with polarization analysis, are presented that were made with $H = -285$ and -940 Oe, successively, after saturation at 15 kOe. All measurements were performed at room temperature.

The spin-flip reflectivities (R^{-+}, R^{-+}) are equal to 0, within the resolution of the experiment, indicating that all magnetizations are collinear with the applied field. Figure 3 gives the measured reflectivities R^{++} as a function of momentum transfer (q_z) for both fields. The Bragg reflection at $q_z = 0.09$ Å^{-1} arises from interference between the Fe layers in the F superlattice. The reflection at 0.12 Å^{-1} arises from the interference between the Fe layers within the AF superlattice, and corresponds to a periodicity twice that of the structural ordering. In the q region between total reflection and the Bragg reflections there are clear differences in R^{++} for the two applied fields. In Ref. 5, the measured reflectivities could be described by calculations based on the two configurations of collinear moments (the magnetization in the F superlattice is either parallel or antiparallel to the top layers in the AF superlattice). In Fig. 3 these same calculations are presented along with the new data. For $H = -285$ Oe, the magnetization of the top layer of the AF superlattice is *antiparallel* to that of the F superlattice and to that of the applied field, just as was the case for $H = -72$ Oe. However, the measured reflectivity at $H = -940$ Oe agrees with the configuration where the magnetization of the top layer of the AF superlattice is *parallel* to that of the F superlattice and that of the applied field. In both cases the antiferromagnetic arrangement in the AF superlattice is maintained.

A first model to understand the mechanism of the bias reversal is that of uniform rotation of the magnetization in the AF superlattice. After the magnetization of the F has been switched and is again aligned with the applied field, the magnetization of the first layer of the AF superlattice and that of the adjacent F superlattice are opposite. Although the state in which they are parallel has lower energy, the transition has to take place through the spin-flopped state, where the moments are roughly at 90° with the field, which has considerably higher energy. However, if the applied field is increased, always in the negative direction, the energy of the two extremal states, with the magnetization aligned along the field, stay the same, while that of the spin-flopped state decreases until, at a certain field, the switching of the bias is permitted. Only at higher fields, the AF structure settles in a spin-flopped state. Although qualitatively this model explains the sequence of observed magnetic transitions, its validity has to be tested on the basis of a quantitative comparison.

The energy of the AF superlattice without the F superlattice can be written as[9]

$$E=\frac{1}{2}\sum_{i=1}^{N-1}\cos(\theta_i-\theta_{i+1})-\frac{\alpha}{2}\sum_{i=1}^{N}\cos^2\theta_i-h\sum_{i=1}^{N}\cos\theta_i. \quad (1)$$

Here the energy is normalized on $g\mu_B H_{\mathrm{E}}^{\mathrm{AF}}S$, where $H_{\mathrm{E}}^{\mathrm{AF}}$ is the exchange field between the layers of the AF superlattice. The first term accounts for the exchange interaction between adjacent layers, the second is the anisotropy energy with $\alpha=H_{\mathrm{A}}/H_{\mathrm{E}}^{\mathrm{AF}}$ as the normalized uniaxial anisotropy field, and the third is the Zeeman energy with $h=H/H_{\mathrm{E}}^{\mathrm{AF}}$ as the normalized applied field. θ_i is the angle between the magnetization of layer i in the superlattice and the easy axis. It is assumed that H is directed along the easy axis.

An AF system becomes unstable,[9] coinciding with the onset of the surface spin–flop transition, when the determinant of the matrix $\hat{m}$ composed of the second derivative of the energy with respect to θ_i of layer $i(m_{ij}=d^2E/d\theta_i d\theta_j)$, with both sublattices aligned in the field direction, becomes zero.

For the double superlattice a term is added to the energy of the AF superlattice: $E^*=E+\beta/2\cos(\theta_F-\theta_1)$, where θ_F is the angle between the magnetization in the F superlattice and the easy axis and β is the exchange field across the interface between the AF and F superlattice normalized on the exchange field in the AF superlattice. Again, the first zero point of the determinant of the matrix of second derivatives $\hat{m}^*$ is used as a criterion for the instability of the collinear AF structure. The determinant is now calculated numerically for increasing magnetic fields, using values for α, β, and h obtained from Ref. 3.

Starting with a top layer magnetization of the AF superlattice opposite to that of the F superlattice and the field, the determinant of $\hat{m}^*$ becomes zero at $H^*=2169$ Oe. While if the top layer is along the F superlattice magnetization then $H^*=2470$ Oe. The latter value is quite close to that obtained for the surface spin flop of the AF superlattice alone—uncoupled to the ferromagnet. The difference between these two conditions may seem obvious, since the surface spin flop will start on the side that is antiparallel to the field. In the first case this side is adjacent to the F superlattice, which due to the exchange interaction is increasing the effective field on the top layer of the AF superlattice. In the second case, the surface spin flop will start on the free side of the AF superlattice, which means the F superlattice has little influence.

The calculated surface spin-flop field is approximately that inferred from the magnetization measurements (Fig. 1). The reversal of the AF superlattice magnetization and the switch in the bias direction is observed at a much lower field than calculated. Therefore there must be another mechanism that is driving this transition at this low field of -447 Oe.

The fact that a uniform rotation model does not explain the switch of the AF is not totally unexpected. The width of the FM hysteresis loop is only $\sim$5 Oe, which is much smaller than the anisotropy field of the ferromagnetic superlattice. This indicates that in the double superlattices the magnetization reversal of the F superlattice is not by coherent rotation, but rather by nucleation and growth of reverse magnetic domains.[4,10] The present experiment indicates that a similar magnetization reversal takes place for the AF superlattice layers. A scenario of nucleation and growth of reverse domains in exchange bias systems is discussed briefly in Ref. 11. Furthermore, Stiles and McMichael[12] suggest that in the case of AF domains of limited size, it is possible for the moments to rotate out of the plane, decreasing the field for the reversal.

In summary, we have shown the reversal of the direction of the exchange bias in a double-superlattice system. The switch in the bias direction is the result of the reversal of the magnetization in the AF superlattice and takes place via domain nucleation and growth at about $H=-447$ Oe, well below the surface spin-flop transition of the AF superlattice. One of the descriptions of the exchange bias in traditional exchange coupled systems involves the existence of laterally limited antiferromagnetic domains.[13,14] Our work seems to indicate that, in adequate magnetic fields, it should be possible to switch those domains. The shape of the hysteresis loop will reflect the distribution of effective domain bias fields with their population.

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[1] W. H. Meiklejohn and C. P. Bean, Phys. Rev. **105**, 904 (1957).
[2] J. Nogués, L. Morellon, C. Leighton, M. R. Ibarra, and I. K. Schuller, Phys. Rev. B **61**, R6455 (2000).
[3] E. E. Fullerton, M. J. Conover, J. E. Mattson, C. H. Sowers, and S. D. Bader, Phys. Rev. B **48**, 15755 (1993); J. Appl. Phys. **75**, 6461 (1994).
[4] J. S. Jiang, G. P. Felcher, A. Inomata, R. Goyette, C. Nelson, and S. D. Bader, Phys. Rev. B **61**, 9653 (2000).
[5] S. G. E. te Velthuis, G. P. Felcher, J. S. Jiang, A. Inomata, C. S. Nelson, A. Berger, and S. D. Bader, Appl. Phys. Lett. **75**, 4174 (1999).
[6] L. Lazar, J. S. Jiang, G. P. Felcher, A. Inomata, and S. D. Bader (unpublished).
[7] R. W. Wang, D. I. Mills, E. E. Fullerton, J. E. Mattson, and S. D. Bader, Phys. Rev. Lett. **72**, 920 (1994).
[8] S. Rakhmanova, D. L. Mills, and E. E. Fullerton, Phys. Rev. B **57**, 476 (1998).
[9] A. L. Dantas and A. S. Carriço, Phys. Rev. B **59**, 1223 (1999).
[10] J. S. Jiang, G. P. Felcher, A. Inomata, R. Goyette, C. Nelson, and S. D. Bader, J. Vac. Sci. Technol. A **18**, 1264 (2000).
[11] T. C. Schulthess and W. H. Bulter, Phys. Rev. Lett. **81**, 4516 (1998).
[12] M. D. Stiles and R. D. McMichael, Phys. Rev. B **59**, 3722 (1999).
[13] A. P. Malozemoff, Phys. Rev. B **35**, 3679 (1987).
[14] K. Takano, R. H. Kodama, A. E. Berkowitz, W. Cao, and G. Thomas, Phys. Rev. Lett. **79**, 1130 (1997).