PHYSICAL REVIEW B **66**, 220406(R) (2002)

Induced magnetic moments at a ferromagnet-antiferromagnet interface

A. Hoffmann
Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439
and Los Alamos National Laboratory, Los Alamos, New Mexico 87545

J. W. Seo*
Institut de Physique, Université de Neuchâtel, CH-2000 Neuchâtel, Switzerland
and IBM Research Division, Zürich Research Laboratory, CH-8803 Rüschlikon, Switzerland

M. R. Fitzsimmons
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

H. Siegwart, J. Fompeyrine, and J.-P. Locquet
IBM Research Division, Zürich Research Laboratory, CH-8803 Rüschlikon, Switzerland

J. A. Dura and C. F. Majkrzak
National Institute of Standards and Technology, Gaithersburg, Maryland 20899

(Received 12 September 2002; published 31 December 2002)

We have studied the magnetization depth profile in a Co/LaFeO$_3$ exchange bias system with polarized neutron reflectometry. In the exchange biased state we observe differences between the reflectivity profiles when the magnetization of the ferromagnetic layer is saturated either parallel or antiparallel to the cooling field. This difference vanishes above the blocking temperature. Since the reflectivity profiles are directly related to the Fourier components of the magnetization depth profile, this data suggest that a net moment develops within the antiferromagnetic layer close to the interface with the ferromagnetic layer, which remains unchanged during the magnetic-field cycling and is coupled antiferromagnetically to the ferromagnet.

DOI: 10.1103/PhysRevB.66.220406 PACS number(s): 75.70.Cn, 61.12.Ha

The coupling between a ferromagnet and an antiferromagnet gives rise to so-called exchange bias, which manifests itself in a magnetic hysteresis curve, which is not symmetric around zero magnetic field.[1] Even though this effect was first observed almost half a century ago,[2] there is still an ongoing discussion about the microscopic mechanism of exchange bias. Already early on it was recognized, that if one assumes that the ferromagnetic spins only couple to one sublattice of the antiferromagnet (i.e., at an uncompensated interface) and if this coupling is of similar magnitude as the typical exchange coupling in ferromagnetic and antiferromagnetic materials, then the expected shift of the hysteresis loop H_E would be one or two orders of magnitude bigger than the experimentally observed one.[3] To alleviate this discrepancy, there are generally two approaches to a microscopic model.

One idea is that the coupling energy is spread across a domain wall either in the antiferromagnet[4] or the ferromagnet,[5] which effectively reduces the coupling. Another idea is that the ferromagnet couples to a small net magnetic moment in the antiferromagnet instead of exclusively to one sublattice. This net moment may occur either due to modifications of the antiferromagnetic spin structure at the interface[6] or due to an imbalance of coupling to the sublattices.[7] A preferential coupling to one of the antiferromagnetic sublattices may arise from defects or roughness at the interface,[8] grain boundaries,[9] domain structure of the antiferromagnet,[10] or the eptaxial relationship at the interface.[11]

Experimentally the origin of exchange bias remains unclear. Tests of the first microscopic model with polarized neutron reflectometry measurements of Fe$_3$O$_4$/NiO multilayers suggested the possibility of an interfacial domain wall,[12] while similar measurements on Fe/MnF$_2$ suggested a homogeneous magnetization throughout the ferromagnetic layer.[13] On the other hand, magnetometry data from Ni$_{81}$Fe$_{19}$/FeMn/Co trilayers suggested indirectly the existence of a partial domain wall in the antiferromagnetic layer.[14] Conversely, there have been many investigations into any net magnetic moments within the antiferromagnetic layer. A net magnetization has been observed in field cooled CoO antiferromagnetic films without coupling to a ferromagnetic layer.[9] The temperature dependence of this moment is the same as the exchange bias in Ni$_{81}$Fe$_{19}$/CoO bilayers. In Fe/FeF$_2$ and Fe/MnF$_2$ there is besides the horizontal shift of the hysteresis loop, also a vertical shift, which might stem from a fixed net moment within the antiferromagnetic layer.[15] Recent experiments with doped antiferromagnetic CoO layers suggest that the domain structure of the antiferromagnet, which might give rise to a net moment at the interface, is a key parameter for the exchange bias.[16] More compelling evidence for a net moment in the antiferromagnet comes from x-ray dichroism[17] and x-ray resonant scattering,[18] which suggest weak magnetic moments in the antiferromagnetic layers. However, the spatial distribution and the field dependence of these moments remain unclear. Here, we report on polarized neutron reflectometry measurements of an exchange bias system consisting of an antiferromagnetic LaFeO$_3$ and a ferromagnetic Co layer. The results of these measurements cannot be attributed to formation of a

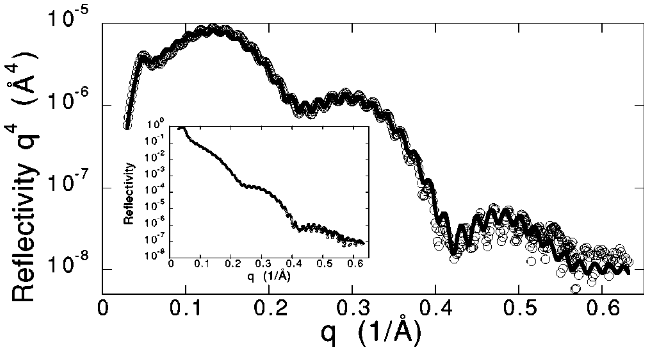

FIG. 1. X-ray reflectivity data. The open symbols are the data, while the solid line is fit obtained from a model (inset is a conventional reflectivity profile plot).

domain wall parallel to the interface in either the ferromagnet or the antiferromagnet in the exchange biased state. However, a comparison with simulated data suggests, that a net magnetization forms in the antiferromagnet close to the interface, which is antiferromagnetically coupled to the ferromagnetic layer and unchanged upon reversing the magnetic field.

For the measurements we prepared a 25-Å-thick ferromagnetic Co layer on a 350-Å thick antiferromagnetic $LaFeO_3$ layer. The $LaFeO_3$ film was grown with molecular-beam epitaxy by means of a block-by-block method[19] on a (001) $SrTiO_3$ substrate at 750 °C with a partial oxygen pressure of 5×10^{-6} Torr. This method has shown to yield high-quality epitaxial oxide films.[20] The $LaFeO_3$ was covered by Co grown at room temperature, which was capped by 10 Å of Pt to prevent oxidation. X-ray-diffraction and transmission electron microscopy analysis[21] showed that the $LaFeO_3$ layer grew epitaxial along [110] with a twinned in-plane structure of its *c* axis along the [100] and [010] direction of the $SrTiO_3$, while the Co layer was polycrystalline. Figure 1 shows x-ray reflectivity data and a model fit. From this fit[22] we can extract the individual layer thicknesses, which agree well with the nominal thicknesses, and the interface roughness for each layer, which is for all interfaces less than 7 Å. In particular, the roughness at the Co/$LaFeO_3$ interface is 3.1 Å, which indicates together with the TEM measurements that interdiffusion is nearly absent in these samples. It should be noted that such high-quality interfaces with very low roughness are beneficial to the polarized neutron reflectometry, since the interface roughness gives an inherent lower limit to resolving any spatial features in the magnetization depth profile.

Polarized neutron reflectometry is tailor made to determine magnetization depth profiles in magnetic heterostructures with high sensitivity and spatial resolution of a few atomic monolayers.[23] The polarized neutron reflectometry measurements were performed on the NG-1 reflectometer of the NIST Center for Neutron Research. For the measurements, a polarized neutron beam is reflected specularly from the sample onto a polarization analyzer. Use of a polarized beam with polarization analysis permits determination of four spin-dependent neutron cross sections. Two of these are the non-spin-flip reflectivities R^{--} and R^{++} (with the neutron spin antiparallel and parallel to the applied field, respectively). The difference between these two cross sections is determined by the component of the magnetization depth profile, which is parallel to the applied magnetic field. The remaining two cross sections are the spin-flip reflectivities R^{-+} and R^{+-}, which are related to the magnetization components perpendicular to the applied field. The momentum transfer *q* dependence of these profiles is related to the Fourier components of the magnetization depth profile, providing depth sensitivity. In all our measurements presented here the spin-flip reflectivities were negligible, indicating that the magnetization depth profile is collinear with the magnetic field.

Figures 2(a) and 2(b) show polarized neutron reflectivities measured at 300 K with magnetic fields of -7.5 kOe and $+7.5$ kOe, respectively. These fields are well above the coercive fields of 35 Oe and sufficient to saturate the sample. At 300 K the sample has no exchange bias and thus one does not expect any difference between the neutron data taken for negative and positive saturation (no difference is observed). In fact Figs. 2(a) and 2(b) exemplify the good reproducibility of the reflectivity measurements. Even though the Néel temperature of $LaFeO_3$ is 750 K,[24] a macroscopic exchange bias of $H_E=25$ Oe can be established by field cooling the sample from 300 K to 18 K in an applied field of $+7.5$ kOe. The blocking temperature for the exchange bias is 100 K. The measurements were then repeated at 18 K for applied fields of ±7.5 kOe. A distinct difference between the two reflectivity profiles is observed [compare Figs. 2(c) and 2(d)]. While the profile at -7.5 kOe is little changed from the room-temperature measurements, there is a significant increase in reflectivity at higher-momentum transfer *q* for the measurement taken at $+7.5$ kOe. It should be noted that the applied fields are again well above the coercivity of $H_c=500$ Oe at 18 K, and thus are sufficient to saturate the magnetization of the ferromagnetic layer. In order to determine whether or not the observed difference was an experimental artifact, we repeated the measurements after field cooling in negative fields and observed the reversed difference for the reflectivity at negative and positive saturation, respectively. Thus the enhanced reflectivity at high *q* is correlated with the direction of field cooling and the corresponding exchange bias.

The difference between the polarized neutron reflectivities for positive and negative saturation after field cooling clearly indicates that the magnetization depth profiles in the two saturated states are not identical. Without any further analysis of the reflectivity data one can already exclude a domain wall in either ferromagnetic or antiferromagnetic layer as the source of the observed difference. For the case of a domain wall parallel to the ferromagnetic-antiferromagnetic interface[4,5] one would expect after field cooling, that the biggest change in the magnetization depth profile occurs for the magnetic field being antiparallel to the cooling field and no change for the field parallel to the cooling field. However, the experimental data show exactly the opposite trend; namely, the change of the reflectivity profile is most pronounced for the magnetic field parallel to the cooling field.

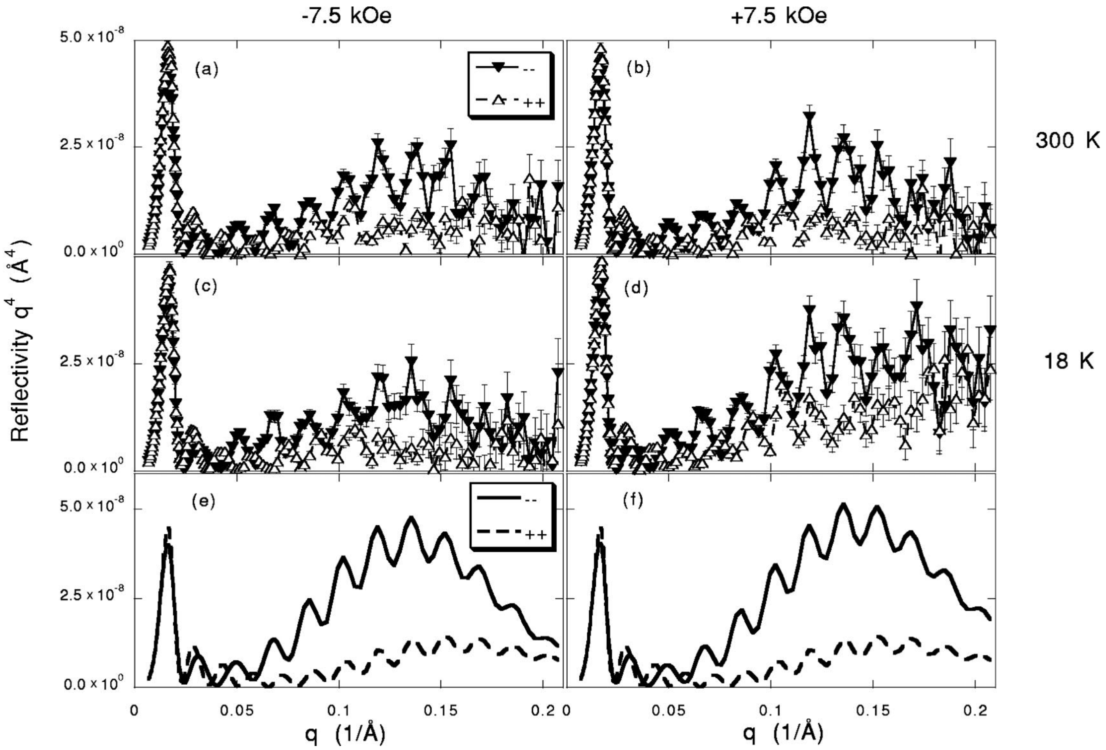

FIG. 2. (a)–(d) polarized neutron reflectivity measured for applied fields of -7.5 kOe [(a),(c)] and $+7.5$ kOe [(b),(d)]], respectively. The closed symbols are R^{--} data and the open symbols are R^{++} data. (a),(b) Measurements taken at 300 K. (c),(d) Measurements taken at 18 K after field cooling in $+7.5$ kOe field. (e),(f) Simulations of polarized neutron reflectivities assuming a net magnetic moment in the antiferromagnetic layer within 10 Å of the interface with the ferromagnetic layer. The magnetic moment in the antiferromagnetic layer is either parallel (e) or antiparallel (f) to the magnetization in the ferromagnetic layer. The solid line shows R^{--}, while the dashed line shows R^{++}. The simulations correspond to the low-temperature polarized neutron reflectivity data.

On the other hand the data are consistent with an additional net moment forming in the antiferromagnetic layer close to the interface with the ferromagnet. Figures 2(e) and 2(f) show simulated polarized neutron reflectivities, which were calculated using the structural parameters obtained from the fit in Fig. 1 and assuming a net moment of $2\mu_B$/Fe atom in the $LaFeO_3$ within 10 Å of the interface with the Co layer. Notice, that since in $LaFeO_3$ the moment of Fe is $4.6\mu_B$/atom,[24] this is equivalent to roughly every fourth spin pointing opposite to its bulk orientation. As can be seen in Fig. 2 the simulations reproduce the main feature of the low-temperature data, which is the increased reflectivity for high-momentum transfer q particularly for R^{--}. Intuitively this can be easily understood, since a net moment in the antiferromagnet opposite to the ferromagnetic magnetization gives a higher contrast for the magnetic scattering at the antiferromagnet/ferromagnet interface than if they were aligned. Furthermore, a comparison of simulated data with no net moment shows that an additional moment modifies the reflectivity profiles more for an antiparallel orientation between the ferromagnet and the antiferromagnet moments [Fig. 2(f)] than for the parallel orientation [Fig. 2(e)]. This result is also in agreement with the experimental observations [Figs. 2(a–d)].

While the simulations show qualitatively correct trends to explain the low-temperature data, they do not fit the data precisely and thus precise quantitative statements about the magnitude and spatial dependence of the magnetic moment in the antiferromagnet are not possible. One difficulty is that the thickness of the capping layer (10 Å) and the spatial extent of the net magnetic moment in the antiferromagnet (a few angstrom) are comparable to the interface roughness (<7 Å), and thus the model used for calculating the reflectivity profile approaches the limits of its validity.[22] Furthermore, a model with a moment in the antiferromagnet either being parallel or antiparallel to the ferromagnetic magnetization may oversimplify the actual magnetization depth profile in these exchange bias structures. Recent numerical simulations suggest that while the antiferromagnet may develop a net moment at the interface with the ferromagnet, only part of this moment may be ‘‘frozen,’’ while the rest of it reverses upon magnetic-field reversal.[25]

Nevertheless, we can draw several conclusions from comparing the simulated and measured reflectivity profiles. There is an asymmetry in the polarized neutron reflectivities between positive and negative saturation, which only occurs in the exchange biased state. Furthermore this asymmetry is most pronounced for high-momentum transfer, indicating a difference in the magnetization depth profiles at short length scales. The comparison with the simulations show that, first, the net moment in the antiferromagnet is opposite to the cooling field, meaning that the coupling is antiferromagnetic between the ferromagnetic and antiferromagnetic layers. Such antiferromagnetic coupling has also been indirectly inferred from other experiments.[18,26,27] Second, the net moment is confined close to the interface (i.e., within just a few monolayers). If it would be more extended or even persist throughout the entire thickness of the antiferromagnetic layer, then the low-temperature reflectivity profiles would be modified at low q, contrary to the experimental observation. Third, the net moment at the interface of the antiferromagnet has to be rather large, i.e., of the order of $1\mu_B$ per Fe atom. It should be noted that numerical simulations[25,28] and recent experimental results[17,18,29–31] suggest that the interface spin structure of the antiferromagnet may be significantly altered when coupled to a ferromagnet. In addition, strong correlations between the ferromagnetic and antiferromagnetic spins have been observed on similar Co/$LaFeO_3$ samples.[32] The tendency of $LaFeO_3$ towards weak ferromagnetism[33] may also be enhanced at the interface.

In conclusion we have shown that the polarized neutron reflectivity profiles develop an asymmetry between positive and negative saturation in exchange biased Co on $LaFeO_3$. The observed asymmetry is inconsistent with a domain wall developing in either the ferromagnet or the antiferromagnet. A comparison with simulated neutron reflectivities suggests that the antiferromagnet develops a net moment close to the interface, which is antiferromagnetically coupled to the ferromagnet and largely unchanged during the magnetization reversal. By modifying the layer thicknesses of this system, it may be possible to obtain more quantitative measurements of the magnitude and spatial extend of this frozen moment as well as temperature and cooling field dependencies.

We thank B. Beschoten, J. A. Borchers, G. Güntherodt, U. Nowak, T. C. Schulthess, and K. D. Usadel for many stimulating discussions. This work was supported by the U.S. Department of Energy, BES-DMS under Contracts Nos. W-7405-Eng-36 and W-31-109-ENG-38.

---

*Present address: Institut de Physique de la Matière Complexe, EPFL, CH-1015 Lausanne, Switzerland.

[1] J. Nogués and I.K. Schuller, J. Magn. Magn. Mater. **192**, 203 (1999).

[2] W.H. Meikeljohn and C.P. Bean, Phys. Rev. **102**, 1413 (1957).

[3] W.H. Meikeljohn and C.P. Bean, Phys. Rev. **105**, 904 (1957).

[4] D. Mauri *et al.*, J. Appl. Phys. **62**, 3047 (1987).

[5] M. Kiwi *et al.*, Europhys. Lett. **48**, 573 (1999).

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

[7] L. Néel, Ann. Phys. (Paris) **2**, 61 (1967).

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

[9] K. Takano *et al.*, Phys. Rev. Lett. **79**, 1130 (1997).

[10] A.P. Malozemoff, Phys. Rev. B **35**, 3679 (1987).

[11] P.J. van der Zaag *et al.*, J. Appl. Phys. **79**, 5103 (1996).

[12] A.R. Ball *et al.*, Appl. Phys. Lett. **69**, 1489 (1996).

[13] A. Hoffmann, M.R. Fitzsimmons, J. Nogués, C. Leighton, V. Leiner, H. Lauter, and I.K. Schuller (unpublished).

[14] F.Y. Yang and C.L. Chien, Phys. Rev. Lett. **85**, 2597 (2000).

[15] J. Nogués, C. Leighton, and I.K. Schuller, Phys. Rev. B **61**, 1315 (2000).

[16] P. Miltényi *et al.*, Phys. Rev. Lett. **84**, 4224 (2000).

[17] W.J. Antel, Jr., F. Perjeru, and G.R. Harp, Phys. Rev. Lett. **83**, 1439 (1999).

[18] T.P.A. Hase *et al.*, Appl. Phys. Lett. **79**, 985 (2001).

[19] J.-P. Locquet *et al.*, Appl. Phys. Lett. **64**, 372 (1994).

[20] J.-P. Locquet *et al.*, Nature (London) **394**, 453 (1998).

[21] J. W. Seo *et al.*, in *Transport and Microstructural Phenomena in Oxide Electronics*, edited by D.S. Ginsley, M.E. Hawley, D.C. Paine, D.H. Blank, and S.K. Streiffer, MRS Symposia Proceedings No. 666 (Material Research Society, Pittsburgh, 2001), p. F8.8.1.

[22] L.G. Parratt, Phys. Rev. **95**, 359 (1954).

[23] G.P. Felcher, J. Appl. Phys. **87**, 5431 (2000).

[24] W.C. Koehler and E.O. Wollan, J. Phys. Chem. Solids **2**, 100 (1957).

[25] U. Nowak, A. Misra, and K.D. Usadel, J. Appl. Phys. **89**, 7269 (2001).

[26] J. Nogues *et al.*, Phys. Rev. Lett. **76**, 4624 (1996).

[27] C.-C. Kao, as discussed in T.P.A. Hase, *et al.*, Appl. Phys. Lett. **79**, 985 (2001).

[28] T. C. Schulthess (private communication).

[29] H. Ohldag *et al.*, Phys. Rev. Lett. **86**, 2878 (2001).

[30] W. Zhu *et al.*, Phys. Rev. Lett. **86**, 5389 (2001).

[31] M. Gruyters and D. Riegel, Phys. Rev. B **63**, 052401 (2000).

[32] F. Nolting *et al.*, Nature (London) **405**, 767 (2000).

[33] H. Watanabe, J. Phys. Soc. Jpn. **14**, 511 (1959).