Three-Dimensional Noncollinear Antiferromagnetic Order in Single-Crystalline FeMn Ultrathin Films

Wolfgang Kuch,* Liviu I. Chelaru, Francesco Offi,† Jing Wang,‡ Masato Kotsugi,§ and Jürgen Kirschner

Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany

(Received 14 July 2003; published 9 January 2004)

We present experimental evidence for a three-dimensional noncollinear antiferromagnetic spin structure in ultrathin single-crystalline fcc $Fe_{50}Mn_{50}$ layers using magnetic circular dichroism photoelectron emission microscopy and x-ray magnetic linear dichroism. Layer-resolved as-grown domain images of epitaxial trilayers grown on Cu(001) in which FeMn is sandwiched between ferromagnetic layers with different easy axes reveal the presence of antiferromagnetic spin components in the film plane and normal to the film plane. An FeMn spin structure with no collinear order in the film plane is consistent with the absence of x-ray magnetic linear dichroism in Fe L_3 absorption in FeMn/Co bilayers.

DOI: 10.1103/PhysRevLett.92.017201 PACS numbers: 75.70.Ak, 75.30.Gw

Antiferromagnetism has remained a fascinating field of research since its first experimental verification by Shull and Smart in 1949 [1]. The spin order in antiferromagnets is characterized by a periodic modulation described by wave vectors of the order of inverse atomic distances. Whereas the spin structure of many bulk antiferromagnets has been experimentally well studied, the spin structure of ultrathin antiferromagnetic films has mainly remained concealed. This is due to constraints of the employed experimental techniques such as neutron diffraction or Mössbauer spectroscopy, which require thick samples to obtain sufficient signal. The lack of knowledge about the spin structure of antiferromagnetic (AF) thin films is in sharp contrast to the renewed interest in the coupling between ferromagnetic (FM) and AF thin films [2,3], which has rapidly increased in the past few years due to the effect of exchange bias [4] and its use in magnetoresistive devices.

Theories trying to describe this coupling [5] need the antiferromagnetic spin structure as a key ingredient. Often just the spin structure of the respective AF bulk material is assumed, although already the AF spin structure at the surface of a NiO single crystal is significantly influenced by the presence of an FM overlayer [6]. Even stronger modifications may be expected in ultrathin AF films. It is thus of basic importance to gain knowledge about the spin structure of ultrathin AF films.

In this Letter, we present an x-ray magnetic circular dichroism photoelectron emission microscopy (XMCD-PEEM) study of single-crystalline ultrathin $Fe_{50}Mn_{50}$ layers (FeMn in the following), sandwiched between several combinations of FM layers with in-plane or out-of-plane magnetization. Information about the FeMn spin structure is obtained from layer-resolved magnetic domain images of the FM layers. Previous XMCD-PEEM studies have shown that the antiferromagnetism of FeMn leads to the occurrence of very small irregular FM domains in bilayers in which Co is grown on top of ultrathin FeMn films [7]. This signature of the AF layer is ascribed to a laterally fluctuating pinning of FM spins by locally uncompensated AF spins of the same component at the surface of the FeMn layer. We show here that in FM/AF/FM trilayers with identical easy axes of the FM layers, in contrast, the top FM layer assumes a regular domain pattern originating from an oscillatory magnetic coupling between parallel and antiparallel alignment of the two FM layers with two atomic monolayers (ML) period, typical for interlayer coupling across an AF layer. We probe the AF spin structure by depositing trilayers with orthogonal easy axes of the FM layers or by changing the easy axis of the top FM layer from a collinear to an orthogonal configuration by deposition of additional magnetic material. If the AF spins were all collinear with the bottom FM layer spin axis, no influence of the AF layer on the orthogonal component would be expected in such trilayers. We observe, however, that the orthogonal top FM layer spin component behaves exactly the same as in FM/AF bilayers and exhibits very small irregular domains indicative of fluctuating pinning of that spin component by the AF layer. The simultaneous presence of both oscillatory coupling of the spin component parallel to the bottom FM layer magnetization and laterally fluctuating pinning of the spin component perpendicular to it provides direct experimental evidence that a three-dimensional noncollinear antiferromagnetic spin structure, reminiscent of that of bulk FeMn, must also be present in its near-two-dimensional realization. This result is further corroborated by the absence of linear magnetic dichroism in the Fe L_3 x-ray absorption signal from the FeMn layer.

Chemically disordered FeMn assumes a noncollinear antiferromagnetic spin structure in the bulk [8,9]. The so-called $3Q$ noncollinear spin structure is characterized by four different sublattices in which the spins point along four different $\langle 111 \rangle$ directions [9]. This spin structure has been quoted for the discussion of recent experimental observations in ultrathin Co/FeMn [7] and $Fe_{20}Ni_{80}$/FeMn bilayers [10]. However, no experimental or theoretical proof for such a noncollinear spin structure in ultrathin films has been presented so far.

FeMn, Co, and Ni layers were grown epitaxially at room temperature by thermal evaporation on Cu(001), as described in Ref. [11]. Single-crystalline, virtually unstrained fcc FeMn films with no indications for chemical order are obtained due to the low lattice mismatch between FeMn and Cu ($\approx 0.4\%$) [11] and between FeMn and the in-plane lattice spacings of Co/Cu(001) [12] and Ni/Cu(001) [13]. XMCD-PEEM was used for the layer-resolved imaging of the FM layers [14]. Some of the layers have been prepared as microwedges suitable for microscopic imaging. The measurements were performed at the UE56/2 beam line of BESSY in Berlin. Details of the experimental setup and the instrument (Focus IS-PEEM) can be found in Ref. [15]. The instrument settings were identical to the ones used in Ref. [7]. Since the incidence angle of x rays is 60° with respect to the surface normal, magnetic contrast is obtained for both in-plane and out-of-plane magnetized domains. Domain images are presented as gray-scale coded absorption asymmetry upon helicity reversal of the exciting radiation, i.e., the difference of two images acquired for opposite helicity divided by their sum.

We first show in Fig. 1 layer-resolved domain images of a 15 ML Ni/FeMn/Co trilayer, in which the bottom Co layer and the FeMn layer have been prepared as crossed wedges. The Co thickness increases in the lower part of the images up to 8 ML from bottom to top, as indicated, and stays constant at 8 ML in the upper part. The FeMn thickness increases within the displayed area from 1.2 to 11.7 ML from left to right. While the easy axis of magnetization for Co films on Cu(001) is along in-plane $\langle 110 \rangle$ directions, it is out of plane for 15 ML Ni/Cu(001). Figure 1(a) shows the domain image of the Co layer, panel 1(b) the domain image of the Ni layer at the same position. Magnetization directions are indicated in some of the domains. The Co magnetization is along $[\bar{1}\,\bar{1}\,0]$ and $[\bar{1}10]$ for FeMn thicknesses below 7 ML and changes by 45° to $[0\bar{1}0]$ above that thickness. This is an indication for AF order in the FeMn layer above that thickness [7]. The Ni magnetization is oriented out of the film plane at FeMn thicknesses above 3 ML. At 7 ML FeMn thickness, the same thickness at which the Co magnetization direction changes by 45°, small out-of-plane domains appear in the Ni layer. These domains are qualitatively identical to the in-plane domains observed in Co/FeMn/Cu(001) [7] and are the signature for a fluctuating out-of-plane spin component at the surface of the AF FeMn layer.

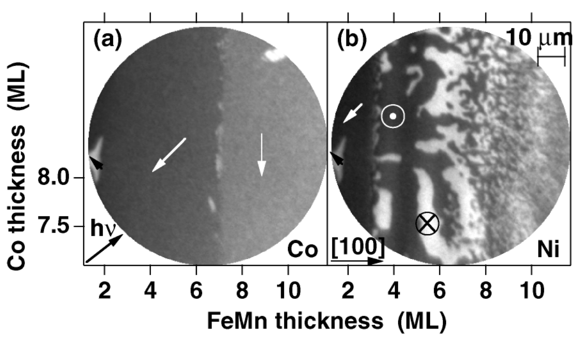

FIG. 1. Layer-resolved magnetic domain images of 15 ML Ni on top of a FeMn/Co/Cu(001) crossed wedge. The Co thickness increases up to 8 ML from bottom to top (left axis), then stays constant; the FeMn thickness increases from left to right (bottom axes). (a) Domain image obtained at the Co L_3 edge; (b) domain image obtained at the Ni L_3 edge.

Figure 2(b) shows a domain image of the Ni layer of the same sample at a different position on the wedge, for higher FeMn thicknesses. The corresponding Co domain image is displayed in Fig. 2(a). The same small domains are recognized in the Ni layer. A periodic vertical stripe pattern is seen superimposed onto the pattern of small domains, which shows a phase jump right at the domain boundary of the Co layer. Images taken for different incidence azimuth confirm that this contrast is due to an in-plane component of magnetization. It is thus interpreted as a canting of the Ni magnetization direction away from pure out-of-plane directions by an oscillatory coupling to the Co layer across the AF FeMn layer with a coupling period of 2 ML. The top Ni layer thus experiences two effects at the same time from the underlying FeMn/Co bilayer: a spatially fluctuating pinning of

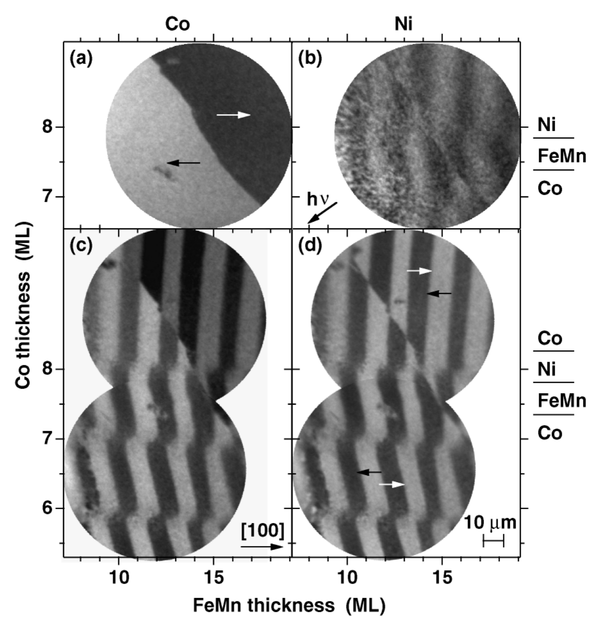

FIG. 2. (a),(b) Layer-resolved magnetic domain images of a sample consisting of 15 ML Ni on top of a FeMn/Co/Cu(001) crossed wedge; (c),(d) same as (a) and (b) but after deposition of 3 ML Co on top of the Ni layer. The Co bottom layer thickness increases up to 8 ML from bottom to top (left axis), then stays constant; the FeMn thickness increases from left to right (bottom axes). (a),(c) Domain images obtained at the Co L_3 edge; (b),(d) domain images obtained at the Ni L_3 edge.

the out-of-plane component of magnetization and an oscillatory coupling to the bottom FM layer of the in-plane component of magnetization.

Deposition of 3 ML of Co on top of the Ni layer switches its anisotropy from out of plane to in plane. Element-resolved domain images for Co and Ni of the same area of the sample taken afterwards are shown in Figs. 2(c) and 2(d), respectively. The domain configuration of the top FM layer can be seen in the Ni image, panel 2(d). The magnetization is now fully in plane; hence, the small domains have disappeared. The Co image, panel 2(c), now contains information from both the bottom and the top FM layers. The top FM layer domain pattern shows the same vertical stripes as before, but now with sharp boundaries. Analysis of the XMCD asymmetry reveals that the magnetization in these stripes is indeed parallel and antiparallel to the magnetization of the bottom Co layer. We have observed exactly the same behavior in FM/AF/FM trilayers in which both FM layers have in-plane anisotropy from the very beginning. The sawtoothlike modulation of the stripes in the range of the Co wedge, which is periodic with Co thickness with 1 ML period, is attributed to the periodic modulation of the bottom interface roughness due to the layer-by-layer growth of the films and will be discussed in a forthcoming publication.

Up to now we have considered the effect that the surface of an ultrathin FeMn film on top of an in-plane magnetized FM layer has on a second FM layer growing on top. Next we consider a system where the bottom FM layer is magnetized out of plane. Figure 3 shows the domain image obtained at the Ni L_3 edge of a Ni/FeMn/Ni trilayer in which the FeMn layer was deposited as a wedge. After deposition of the first Ni layer the sample had been annealed to 450 K to smoothen the surface and saturated by an out-of-plane 500 Oe external field. After

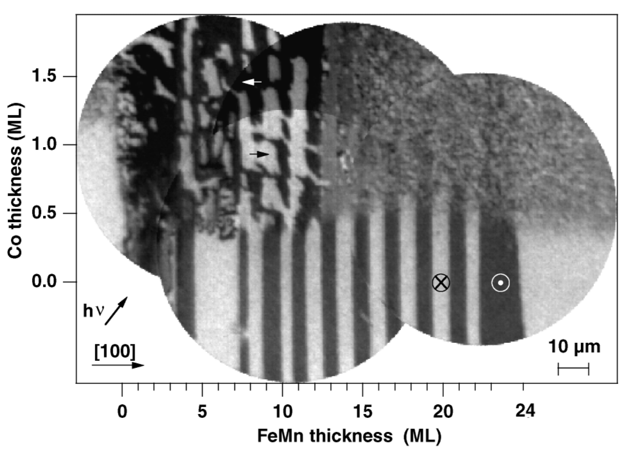

FIG. 3. Magnetic domain image at the Ni L_3 edge of Co/Ni/FeMn/Ni/Cu(001), in which FeMn and Co were deposited as crossed wedges. The Co thickness increases from bottom to top starting at "0.0" (left axis), the FeMn thickness from 0 to 24 ML from left to right (bottom axis).

preparation and acquisition of a first series of images, a Co wedge perpendicular to the FeMn wedge was deposited on top. This induces a spin-reorientation transition in the top FM layer from out of plane to in plane. Figure 3 shows the sample at the beginning of the Co wedge, as indicated at the left axis. The FeMn thickness increases from 0 to 24 ML from left to right. In the bottom part of the image, up to a Co thickness of ≈ 0.5 ML, the same domain pattern is found as was observed before deposition of the Co wedge. Regular vertical stripes are observed above an FeMn thickness of about 5 ML, which become more regular above 9 ML, with a period of 2 ML. The magnetization in these stripes is alternatingly pointing out and into the film plane as confirmed from measurements under different light incidence azimuth. The magnetization direction of the bottom Ni layer corresponds to the "white" direction as known from regions outside the FeMn wedge. Notice the absence of any small domains in the bottom part of the image, contrary to the previous case. This indicates that the growing top Ni layer does not sense a fluctuating pinning of the out-of-plane component of magnetization, but only the regular oscillatory coupling with 2 ML period.

Above a Co layer thickness of 0.5 ML the magnetization of the top FM layer has turned to in plane, as was verified by images acquired for opposite light incidence azimuth. For FeMn thicknesses above 12 ML FeMn this change to in-plane magnetization is accompanied by the occurrence of small domains, exactly as they are observed in as-grown Co/FeMn bilayers [7]. This means that in the apparently out-of-plane magnetized Ni/FeMn/Ni trilayer, there must be still a statistically fluctuating in-plane component, which can force the in-plane direction of magnetization of the top FM layer to a small domain configuration. Such small domains are energetically unfavorable and disappear as soon as the AF layer is heated above the Néel temperature [7]. They do not occur in trilayers with identical anisotropy of both FM layers or in bilayers in which the FM layer is deposited first. In these two cases the magnetization direction of the bottom FM layer locally sets the exchange bias and, by the oscillatory coupling, determines the pinning experienced by FM layers at the top AF interface. The presence of small domains in the top FM layer in the spin-engineered trilayers with different anisotropy, however, shows that the perpendicular spin component behaves independently, as if the bottom FM layer were not present. A three-dimensional spin structure in the AF FeMn layer has consequently to be considered. If the FeMn layer is deposited onto an in-plane magnetized Co film, it is the out-of-plane component of the FeMn spins which is not ordered by the Co bottom layer and gives rise to small out-of-plane domains in the top Ni layer [cf. Figs. 1(b) and 2(b)]. In the case of FeMn being deposited onto an out-of-plane magnetized Ni layer, the in-plane component of the FeMn spins is laterally statistically fluctuating, leading to small

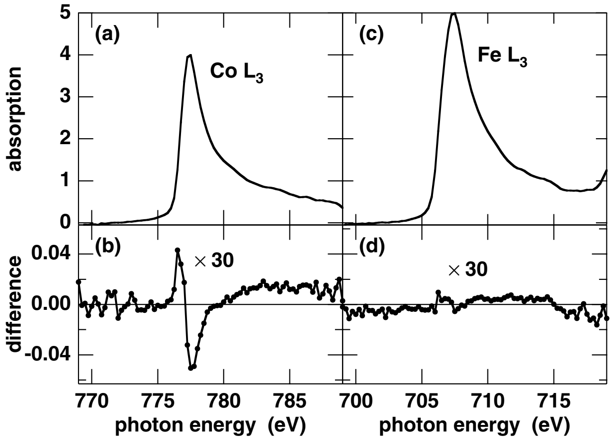

FIG. 4. XMLD spectra of 15 ML FeMn/6 ML Co/Cu(001), (a),(c) L_3 absorption intensity for Co and Fe, respectively. (b),(d) Corresponding difference between absorption spectra for parallel and perpendicular orientation of light polarization vector and Co magnetization direction.

in-plane domains after capping the top Ni layer by Co (cf. Fig. 3) [16].

The action of AF spins on the FM magnetization direction is probably mediated by uncompensated AF spins at step edges or kinks at the interface. If the AF layer is deposited on top of an FM layer, the AF spin structure will be modified to exhibit a nonvanishing magnetic moment with alternating sign in each atomic layer [17], which in turn acts on the top FM layer to display the oscillatory coupling behavior.

Further indication for a noncollinear spin order in the ultrathin FeMn layers comes from measurements of the x-ray magnetic linear dichroism in absorption (XMLD). XMLD measures the anisotropic charge distribution induced by a collinear (FM or AF) spin alignment in a structurally fourfold symmetric sample [18]. We present in Figs. 4(a) and 4(c) the Co and Fe L_3 absorption signals, respectively, of a 15 ML FeMn/6 ML Co bilayer for normal incidence as measured by total electron yield. The Co film had been saturated by an external field before deposition of the FeMn layer. The difference between absorption for parallel and perpendicular orientation of polarization vector and the Co magnetization direction has been measured for two orthogonal orientations of the linear x-ray polarization. The result is given in panels 4(b) and 4(d). Whereas at the Co L_3 edge [4(b)] a small but sizeable XMLD signal similar to that of Ref. [18] is detected, this is not the case at the Fe L_3 edge [4(d)]. A signal similar in size to that of Co would be expected for a collinear orientation of Fe spins in Fe/Cu(001) [18]. The absence of XMLD points towards an FeMn spin structure with noncollinear order also in the in-plane component.

The experimental results provide evidence for the presence of a three-dimensional spin structure in ultrathin, near-two-dimensional single-crystalline FeMn layers. This has implications on theoretical models of the AF–FM interaction and for understanding the exchange bias effect along different magnetization components [3]. Although statements about the exact spin structure and the size of the AF spin component perpendicular to the FM pinning direction cannot be made on the basis of our experiments, it is plausible to consider the same nearest-neighbor exchange interaction as in bulk FeMn, statistically averaged over all possible local chemical environments, to be the driving mechanism. It will be interesting to see whether the compositional disorder, which reduces the translational symmetry, is a necessary ingredient for such a noncollinear spin structure in ultrathin films.

We acknowledge technical help by B. Zada, W. Mahler, and the BESSY staff and financial support by the BMBF (No. 05 KS1EFA6).

*Electronic address: kuch@mpi-halle.de
†Present address: INFM, Unità Roma Tre, Roma, Italy.
‡Present address: The University of Hong Kong, Hong Kong, China.
§Present address: Hiroshima Synchrotron Radiation Center, Hiroshima, Japan.

[1] C. G. Shull and J. S. Smart, Phys. Rev. **76**, 1256 (1949).
[2] W. J. Antel, Jr., F. Perjeru, and G. R. Harp, Phys. Rev. Lett. **83**, 1439 (1999); P. Miltényi *et al.*, *ibid.* **84**, 4224 (2000); F. Y. Yang and C. L. Chien, *ibid.* **85**, 2597 (2000); W. Zhu *et al.*, *ibid.* **86**, 5389 (2001).
[3] S. Maat *et al.*, Phys. Rev. Lett. **87**, 087202 (2001).
[4] For a review, see J. Nogués and I. K. Schuller, J. Magn. Magn. Mater. **192**, 203 (1999), and references therein.
[5] For a review, see R. L. Stamps, J. Phys. D: Appl. Phys. **33**, R247 (2000), and references therein.
[6] H. Ohldag *et al.*, Phys. Rev. Lett. **86**, 2878 (2001).
[7] W. Kuch *et al.*, Phys. Rev. B **65**, 140408(R) (2002).
[8] J. S. Kouvel and J. S. Kasper, J. Phys. Chem. Solids **24**, 529 (1963).
[9] H. Umebayashi and Y. Ishikawa, J. Phys. Soc. Jpn. **21**, 1281 (1966).
[10] R. Coehoorn *et al.*, Physica (Amsterdam) **319B**, 141 (2002).
[11] F. Offi, W. Kuch, and J. Kirschner, Phys. Rev. B **66**, 064419 (2002).
[12] J. R. Cerdá *et al.*, J. Phys. Condens. Matter **5**, 2055 (1993).
[13] W. Platow *et al.*, Phys. Rev. B **59**, 12 641 (1999).
[14] J. Stöhr *et al.*, Science **259**, 658 (1993).
[15] W. Kuch *et al.*, J. Vac. Sci. Technol. B **20**, 2543 (2002).
[16] The FeMn thickness for the appearance of small domains in Fig. 3 (12 ML) is larger than in Fig. 1(b) (8 ML). This could be due to different AF ordering temperatures of FeMn in the different trilayers, but could as well indicate a minimum thickness for the occurrence of the three-dimensional noncollinear spin structure in FeMn/Ni/Cu(001).
[17] This does not necessarily imply a collinear layered spin structure of the AF, but would equally occur for other (for example, the 3Q) AF spin structures.
[18] S. S. Dhesi, G. van der Laan, and E. Dudzik, Appl. Phys. Lett. **80**, 1613 (2002).