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Magnetic interface coupling in single-crystalline Co/FeMn bilayers

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The magnetic coupling between epitaxial single-crystalline Co and FeMn layers on Cu(001) was investigated by element-resolved magnetic circular dichroism domain imaging using a photoelectron emission microscope. As-grown Co domain patterns reveal the presence of many small domains in the antiferromagnet. The coupling of the Co layer is found to be along ⟨100⟩ crystallographic directions. This is discussed in terms of a 45° coupling due to frustrations at topological 90° domains in the FeMn layer. Coercivity oscillations as a function of FeMn thickness with atomic monolayer period support the importance of such step-induced domains in the coupling.

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The magnetic coupling across the interface between a ferromagnet and an antiferromagnet is the key for the understanding of the phenomena summarized under the term ''exchange bias.'' They comprise of a shift of the hysteresis loop along the field axis and an increase in coercivity. Great effort has recently been devoted to experimental[1] and theoretical investigations[2] of these phenomena due to their importance in a variety of magnetoresistive applications. Because of the complex nature and incomplete characterization of the interface between granular sputtered or polycrystalline films, which are commonly used in most of the experimental studies,[3–8] details of this coupling are poorly understood. An important point, for example, is the coupling across so-called fully compensated antiferromagnetic interfaces, i.e., surfaces of the antiferromagnet at which the antiferromagnetic spins sum to zero. Koon suggested a model for ideally compensated interfaces involving a 90° coupling between the spins in the ferromagnet and the spins in the antiferromagnet.[9] In contrast to this, Ohldag *et al.* studied the spin structure at a Co/NiO(001) single crystal interface, and observed an alignment of the Co domain structure with the axial orientation of the NiO antiferromagnetic spins in the surface plane.[10] In other theories a local uncompensation, invoked by interface roughness of defects, is assumed.[2]

In order to address the coupling at the interface between an antiferromagnet and a ferromagnet, we performed an experimental study of single crystalline Co/FeMn bilayers with structurally well-defined interfaces, grown epitaxially on Cu(001). FeMn layers have been used as the antiferromagnet in numerous studies and applications of exchange bias.[3–6] Bulk FeMn has a tetragonal antiferromagnetic spin structure, in which the spins of the corner atom of the fcc unit cell and three adjacent face-centering atoms point towards the center of the common tetrahedron.[11] The (001) surface is accordingly nominally fully compensated with respect to the in-plane component of the spins. The growth of Co and FeMn on Cu(001), as well as the growth of Co on FeMn/Cu(001) and of FeMn on Co/Cu(001) is accompanied by clear intensity oscillations in medium energy electron diffraction, indicating pseudomorphic growth in a layer-by-layer fashion.[12] Scanning tunneling microscopy studies revealed that not more than three atomic levels are simultaneously exposed during the growth of FeMn on Cu(001).[13] We observe a moderate exchange bias at room temperature in these bilayers, which is in line with experiments by Jungblut *et al.*, published in 1994 and 1995.[14] By using all-metal bilayers we avoid oxidation/reduction reactions that might occur at metal/oxide interfaces,[15] and which would lead to a further complication of the interface coupling.

Element-resolved magnetic domain imaging by photoelectron emission microscopy (PEEM) with x-ray magnetic circular dichroism (XMCD) as the contrast mechanism allows to observe the characteristic changes in Co magnetization at the antiferromagnetic ordering transition of the FeMn layer, either as a function of temperature or of FeMn film thickness. We show that the Co layer magnetization is pinned along crystallographic ⟨100⟩ directions by the antiferromagnetism of the FeMn layer, 45° to the Co in-plane easy axes. We attribute this to frustrations at atomic steps of the *locally compensated* FeMn(001) surface. It is demonstrated that as a consequence of the periodic change in step density during layer-by-layer epitaxial growth, the resulting increase in coercivity shows monolayer period oscillations as a function of FeMn layer thickness.

Co/FeMn bilayers were grown at room temperature on Cu(001) in zero field. $Fe_{50}Mn_{50}$ films were obtained by the coevaporation of Fe and Mn. Composition and film thickness were calibrated by oscillations of the diffracted medium energy electron intensity, and cross checked by Auger electron spectroscopy. The accuracy of the thicknesses is estimated as 5%. The bilayers were grown either as continuous films or as crossed wedges with 155 μm width, as described in Ref. 16. Circularly polarized x rays from the helical undulator beamline UE56-2 PGM2 of BESSY II in Berlin were used, incident to the sample under an angle of 60° from the surface normal. The setup of the electrostatic PEEM is identical to that described in previous publications.[17] Parameters were set to result in a lateral resolution of 400 nm and a field of view of 90 μm. Images are presented in the form of gray-scale coded absorption asymmetry for opposite light helicity at the maxima of the L_3 edges. Vectorial analysis of the local magnetization direction was done by comparing images of the same area on the sample acquired for different light incidence azimuths.

Since direct imaging of the antiferromagnetic domains of

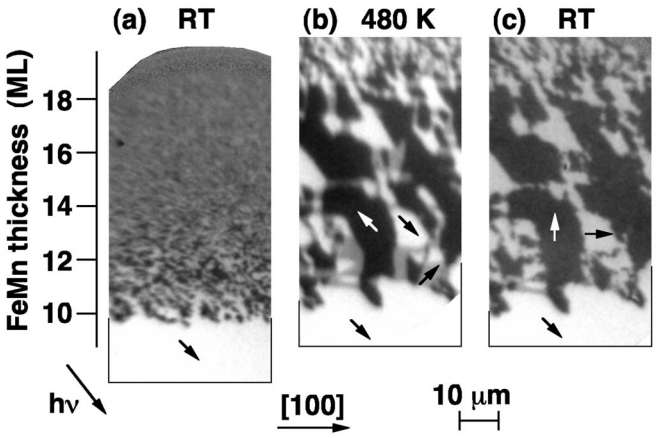

FIG. 1. (a) As-grown Co domain image of 6-ML Co/FeMn/Cu(001). The FeMn thickness is indicated on the left axis. (b) Co domain image of the same area of the sample at a temperature of 480 K. Note that in this image some blurring occurs due to thermal movement of the sample during exposure. (c) Co domain image of the same area of the sample after cooling back to room temperature.

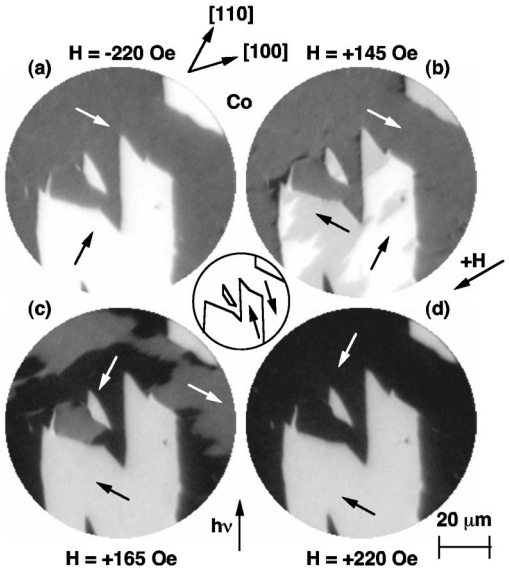

FIG. 2. (a) Co domain pattern of a 20-ML Co/15-ML FeMn/Cu(001) bilayer. Magnetization directions are indicated by arrows. (b), (c), (d) Same position of the sample after application of an external magnetic field of 145 Oe (b), 165 Oe (c), and 220 Oe (d) in the direction indicated by *H*. Although the Co magnetization is along ⟨110⟩ directions, the pinning by the FeMn layer is found to be along [010] and [$0\bar{1}0$], as indicated in the sketch in the center.

metallic FeMn by linearly polarized light is hampered by vanishing crystal-field splitting in the Fe and Mn $L_{2,3}$ edges, we focus here on ferromagnetic domain images of the Co layer. XMCD images acquired at the Fe L_3 and Mn L_3 edges always showed identical domain patterns as the corresponding Co images, however with a strongly reduced magnetic signal. These ferromagnetic moments in FeMn were found independent of whether the FeMn layer was below or above its Néel temperature, and will be published elsewhere.

Figure 1(a) shows the as-grown Co domain image of six atomic monolayers (ML) of Co on top of a wedge-shaped FeMn/Cu(001) film. The FeMn thickness increases from the bottom to top, as indicated at the left axis. Below approximately 10 ML FeMn thickness, a single domain with a white contrast is observed. Above 10 ML FeMn thickness the as-grown domain pattern starts to decay into small domains. The average domain size is decreasing with increasing FeMn thickness, and the magnetic contrast vanishes towards the top of the image, when the domains are becoming smaller than the instrumental resolution. 10 ML is exactly the thickness at which a sudden increase of more than two orders of magnitude in coercivity is observed.[12] We conclude thus that 10 ML corresponds to the thickness of the antiferromagnetic ordering transition at room temperature, and attribute the observed Co domain pattern to domains in the antiferromagnetic FeMn film. Local exchange interaction between domains in the FeMn layer and the Co layer induce a replicated domain pattern in the Co layer in the initial stages of growth, which is then partially frozen when the Co film becomes thicker. The as-grown Co domain pattern is thus an indicator for the presence of small domains in the antiferromagnet.

Because of the many domain walls this Co domain pattern is metastable, i.e., energetically unfavorable. We demonstrate this by heating the sample to 480 K in zero field [Fig. 1(b)]. At this temperature the antiferromagnetic ordering transition is shifted to a thickness of about 20 ML, as evidenced from the jump in coercivity.[12] The FeMn layer is thus paramagnetic in the region shown in Fig. 1, whereas the Co layer is still ferromagnetic at that temperature.[18] Without being coupled to an antiferromagnet, the Co magnetization rearranges into bigger domains to reduce domain-wall energy. In these domains Co is magnetized along ⟨110⟩ in-plane directions, as indicated by arrows, which are the easy axes for Co/Cu(001),[19,20] and obviously are also the easy axes for Co on top of *paramagnetic* FeMn. After cooling back to room temperature, the domain pattern stays qualitatively largely identical [Fig. 1(c)]. Analysis of the magnetization direction, however, reveals that on top of *antiferromagnetic* FeMn, the Co magnetization is now along ⟨100⟩ directions, as seen in Fig. 1(c) from the lower contrast in this region of the sample, and indicated by arrows. The transition between Figs. 1(b) and 1(c) has been tested to be reversible by repetitive heating and cooling, except for some domain coarsening. The change in Co magnetization direction from ⟨110⟩ to ⟨100⟩ is thus directly related to the antiferromagnetism of the FeMn film. A 45° rotation occurs in all of the Co domains, which shows that the pinning by antiferromagnetic FeMn is along ⟨100⟩ directions.

For high Co thicknesses above 15 ML, the Co magnetization on top of antiferromagnetic FeMn is found to be again along ⟨110⟩ directions. We can attribute this to the increasing total anisotropy energy in the Co film as the films become thicker.[20] The pinning of the underlying FeMn layer in this case, however, is still along ⟨100⟩ directions, as is demonstrated in Fig. 2. Figure 2(a) shows the Co domain pattern of a 20-ML Co/15-ML FeMn/Cu(001) bilayer after heating to 480 K and subsequent application of a 220 Oe external magnetic field in the negative "*H*" direction at room tempera-

ture. The magnetization is along [110] and $[1\bar{1}0]$, as indicated by arrows. Panel (b) shows the remnant domain image after application of a 145 Oe field in the positive direction indicated by *H*, panel (c) after 165 Oe, and panel (d) after 220 Oe external field. It is seen that the magnetization is only rotated by 90° at each position, and that 220 Oe is not enough to reverse the magnetization by 180°. This is explained by assuming a pinning by the antiferromagnetic FeMn layer in the [010] and $[0\bar{1}0]$ directions, as indicated in the sketch in the center of Fig. 2. The anisotropy of the 20-ML Co layer is consequently strong enough to force the magnetization into the ⟨110⟩ directions, 45° away from the pinning direction of the FeMn layer. It is then relatively easy to flip the Co magnetization between the two adjacent ⟨110⟩ directions by domain-wall motion, but stronger fields are needed to reverse the Co magnetization by 180°, since in the latter case an irreversible rearrangement of antiferromagnetic spins is involved.

This change in easy axis from the ⟨100⟩ pinning direction to ⟨110⟩ for thicker Co films allows a rough estimate of the pinning strength. Assuming that the Co fourfold in-plane anisotropy in the Co/FeMn bilayers is similar to that of single Co films on Cu(001), we take the experimental value for the anisotropy of 15-ML Co/Cu(001) from Ref. 20, namely 2.2×10^{-4} J/m^2. At that Co thickness the anisotropy energy and the coupling energy are of the same magnitude. This estimate for the coupling strength, about 2×10^{-4} J/m^2, is at the upper end of the range of interface energies reported in literature for sputtered FeMn films.[1]

The above observations can be explained by assuming a noncollinear bulklike tetragonal spin structure in the FeMn films.[11] In that structure, the ideal (001) terminated surface consists of a checkerboard arrangement of spins along one ⟨110⟩ axis, with in-plane components alternatingly pointing in opposite directions, for example, along ±[110]. The spins in the next monatomic level above or below are then oriented with their in-plane components at 90° with respect to this level, i.e., along $\pm[1\bar{1}0]$. Each monatomic step in the surface gives thus rise to a topological 90° domain in the surface layer of the antiferromagnet. Such topological domains do not cost domain-wall energy, and provide thus a sensible explanation for the many small domains observed in the as-grown Co/FeMn bilayers. Coupling of a ferromagnetic Co layer to 90° FeMn domains results in a net coupling at intermediate 45° directions, in analogy to the fluctuation mechanism used by Slonczewski for explaining the biquadratic interlayer exchange coupling, in which spatial fluctuations between 0° and 180° coupling lead to an effective 90° coupling.[21] In extension of that mechanism to 90° coupling fluctuations, exchange averaging over monatomic steps leads to an effective 45° coupling of Co to FeMn in the present case, i.e., to a pinning along ⟨100⟩. In the measurements of Jungblut *et al.* a change in the easy axis of permalloy films on Cu(110) by 90° has been observed when in contact with antiferromagnetic FeMn films,[14] which was analogously interpreted in terms of step-induced topological 180° domains in the uncompensated (110) surface of the FeMn bulk spin structure,[22] and quantified in a micromagnetic model following the line of Slonczewski's model.[23] A similar mechanism has been claimed to be also responsible for 90° magnetization rotations in Fe on stepped surfaces of antiferromagnetic Cr(001).[24,25]

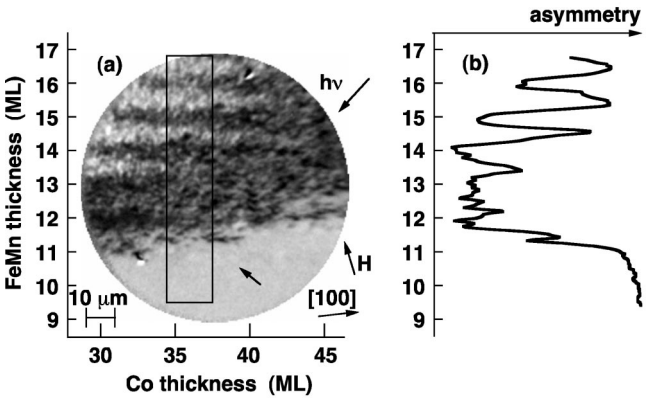

FIG. 3. (a) Co domain image of a crossed double wedge of Co and FeMn on Cu(001) after application of a 330 Oe external field in the direction indicated by *H*. The Co thickness increases from left to right (bottom axis), the FeMn thickness from top to bottom (left axis). Bright stripes with monolayer FeMn thickness period are observed on the left side. (b) Vertical line scan of the XMCD asymmetry averaged over the rectangle shown in panel (a).

The resulting spin configuration and coupling type resemble Koon's model,[9] although an angle of 45° instead of 90° is involved. This type of coupling emphasizes the importance of 90° domain boundaries in the antiferromagnet, induced by monatomic steps. Scanning tunneling microscopy investigations support this view. A large number of small, ≈1 nm wide islands and holes of single atomic step height are observed in otherwise atomically flat terraces.[13] The layer-by-layer growth of the FeMn films on Cu(001) implies that the number of surface steps oscillates with monolayer periodicity, where minima are located at integer monolayer thicknesses, and maxima at half filled monolayers. If step edge atoms are important for the coupling between ferromagnets and antiferromagnets, there should consequently be also an oscillation in the coupling strength. This is indeed the case. In Fig. 3 we show in panel (a) the domain configuration of a Co/FeMn bilayer, here shaped as a crossed double wedge, after application of a 330 Oe external magnetic field in the direction indicated by the arrow labeled *H*. The Co thickness increases from left to right, as indicated at the bottom axis, the FeMn thickness from bottom to top, as indicated at the left axis. At low FeMn thicknesses at the bottom of the image (in the paramagnetic region and in the beginning of the antiferromagnetic region) the Co layer has switched into a [110] single domain state, as indicated by the arrow. At FeMn thicknesses above ≈12 ML, the Co layer is not saturated, but shows still many small domains. At the left-hand side of the image, i.e., at lower Co thicknesses, bright stripes are observed. In these stripes the as-grown domain pattern before application of the external field, which looked quite similar to Fig. 1(a), is largely conserved. Outside these stripes many domains in the Co have switched to give a darker contrast. Figure 3 (b) shows a line scan along

the vertical direction of the image, horizontally averaging over the rectangle printed in panel (a). It is seen that the stripes appear with a periodicity of 1-ML FeMn.[26] We attribute these stripes to coercivity modulations as a function of FeMn thickness. Since the coercivity in Co layers on top of antiferromagnetic FeMn/Cu(001) is nearly exclusively caused by the antiferromagnetism of the FeMn layer, it must depend on the strength of the interface coupling between the ferromagnet and the antiferromagnet. The monolayer periodicity of the coupling strength is the consequence of the oscillation in the number of step edge atoms caused by layer-by-layer growth, and directly proves the importance of monatomic steps in the Co-FeMn interface coupling.

In conclusion, by using high-quality single-crystalline epitaxial Co/FeMn bilayers, we show that the pinning direction of a FeMn(001) layer is along $\langle 100 \rangle$ directions, with a coupling strength of the order of 10^{-4} J/m^2. The direction of the coupling experienced by a ferromagnetic film during growth on FeMn(001) is rapidly changing spatially, leading to small domains in the as-grown ferromagnetic domain pattern. This can be explained by the noncollinear FeMn spin structure. Monatomic steps at the surface of the FeMn layer cause topological 90° domains in the antiferromagnet, which are seen partly reflected in the as-grown domain pattern of the Co overlayer. Exchange averaging over these domains leads to an effective coupling along $\langle 100 \rangle$ directions. Oscillations in the coercivity of Co/FeMn with a monolayer period in FeMn thickness are related to the oscillations of the number of monatomic steps related to the layer-by-layer growth mode of the single-crystalline FeMn films on Cu(001), further supporting this model.

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[1] See, for example, J. Nogués and I.K. Schuller, J. Magn. Magn. Mater. **192**, 203 (1999), and references therein.

[2] See, for example, R.L. Stamps, J. Phys. D **33**, R247 (2000), and references therein.

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

[4] V.I. Nikitenko, V.S. Gornakov, A.J. Shapiro, R.D. Shull, K. Liu, S.M. Zhou, and C.L. Chien, Phys. Rev. Lett. **84**, 765 (2000).

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

[6] A. Anguelouch, B.D. Schrag, G. Xiao, Y. Lu, P.L. Trouilloud, R.A. Wanner, W.J. Gallagher, and S.S.P. Parkin, Appl. Phys. Lett. **76**, 622 (2000).

[7] P. Miltényi, M. Gierlings, J. Keller, B. Beschoten, G. Güntherodt, U. Nowak, and K.D. Usadel, Phys. Rev. Lett. **84**, 4224 (2000).

[8] C. Leighton, M.R. Fitzsimmons, P. Yashar, A. Hoffmann, J. Nogués, J. Dura, C.F. Majkrzak, and I.K. Schuller, Phys. Rev. Lett. **86**, 4394 (2001).

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

[10] H. Ohldag, A. Scholl, F. Nolting, S. Anders, F.U. Hillebrecht, and J. Stöhr, Phys. Rev. Lett. **86**, 2878 (2001).

[11] H. Umebayashi and Y. Ishikawa, J. Phys. Soc. Jpn. **21**, 1281 (1966).

[12] F. Offi, W. Kuch, and J. Kirschner (unpublished).

[13] L.I. Chelaru *et al.* (unpublished).

[14] R. Jungblut, R. Coehoorn, M.T. Johnson, J. aan de Stegge, and A. Reinders, J. Appl. Phys. **75**, 6659 (1994); R. Jungblut, R. Coehoorn, M.T. Johnson, C. Sauer, P.J. van der Zaag, A.R. Ball, T.G.S.M. Rijks, J. aan de Stegge, and A. Reinders, J. Magn. Magn. Mater. **148**, 300 (1995).

[15] G.H. Yu, C.L. Chai, F.W. Zhu, J.M. Xiao, and W.Y. Lai, Appl. Phys. Lett. **78**, 1706 (2001).

[16] W. Kuch, J. Gilles, F. Offi, S.S. Kang, S. Imada, S. Suga, and J. Kirschner, J. Electron Spectrosc. Relat. Phenom. **109**, 249 (2000).

[17] W. Kuch, R. Frömter, J. Gilles, D. Hartmann, Ch. Ziethen, C.M. Schneider, G. Schönhense, W. Swiech, and J. Kirschner, Surf. Rev. Lett. **5**, 1241 (1998).

[18] C.M. Schneider, P. Bressler, P. Schuster, J. Kirschner, J.J. de Miguel, and R. Miranda, Phys. Rev. Lett. **64**, 1059 (1990).

[19] P. Krams, F. Lauks, R.L. Stamps, B. Hillebrands, and G. Güntherodt, Phys. Rev. Lett. **69**, 3674 (1992).

[20] W. Weber, R. Allenspach, and A. Bischof, Appl. Phys. Lett. **70**, 520 (1997).

[21] J.C. Slonczewski, Phys. Rev. Lett. **67**, 3172 (1991).

[22] S. Riedling, M. Bauer, C. Mathieu, B. Hillebrands, R. Jungblut, J. Kohlhepp, and A. Reinders, J. Appl. Phys. **85**, 6648 (1999).

[23] M.J. Dekker and K. Ramstöck, J. Appl. Phys. **82**, 5633 (1997).

[24] E.J. Escorcia-Aparicio, H.J. Choi, W.L. Ling, R.K. Kawakami, and Z.Q. Qiu, Phys. Rev. Lett. **81**, 2144 (1998).

[25] H. Hopster, Phys. Rev. Lett. **83**, 1227 (1999).

[26] Note that while the 5% error in the thickness is negligible in the determination of the period, it amounts to more than 0.5 ML in the absolute thickness, therefore impeding us to draw definite conclusions about the phase of the oscillations.