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Direct measurement of depth-dependent Fe spin structure during magnetization reversal in Fe/MnF_2 exchange-coupled bilayers

W. A. A. Macedo,[1] B. Sahoo,[2,*] J. Eisenmenger,[3] M. D. Martins,[1] W. Keune,[2,4] V. Kuncser,[5] R. Röhlsberger,[6] O. Leupold,[7,†] R. Rüffer,[7] J. Nogués,[8] Kai Liu,[9] K. Schlage,[10] and Ivan K. Schuller[11]

[1]*Centro de Desenvolvimento da Tecnologia Nuclear, 31270-901 Belo Horizonte, MG, Brazil*

[2]*Universität Duisburg-Essen, 47048 Duisburg, Germany*

[3]*Universität Ulm, 89081 Ulm, Germany*

[4]*Max-Planck-Institut für Mikrostrukturphysik, 06120 Halle, Germany*

[5]*National Institute of Materials Physics, 77125 Bucharest-Magurele, Romania*

[6]*Deutsches Elektronen-Synchrotron, 22067 Hamburg, Germany*

[7]*European Synchrotron Radiation Facility, 38043 Grenoble Cedex, France*

[8]*ICREA and Institut Català de Nanotecnologia, Campus of the University Autònoma de Barcelona, E-08193 Bellaterra, Spain*

[9]*University of California–Davis, Davis, California 95616, USA*

[10]*Universität Rostock, 18051 Rostock, Germany*

[11]*University of California–San Diego, La Jolla, California 92093, USA*

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We measured directly the depth-dependent Fe spin rotation upon magnetization reversal in exchange-coupled Fe/MnF_2 bilayers using nuclear resonant scattering of synchrotron radiation from an ^{57}Fe-probe layer buried at different depths within the Fe film. Our results show that the exchange-biased ferromagnetic layer develops a noncollinear spin structure along the film normal direction, reminiscent of a partial domain wall parallel to the Fe/MnF_2 interface. This is contrary to most theoretical models of exchange bias which assume a collinear spin structure in the ferromagnetic layer.

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I. INTRODUCTION

Exchange bias (EB), originating from the interface coupling between a ferromagnet (F) and an antiferromagnet (AF), gives rise to shifted hysteresis loops along the magnetic-field (x) axis.[1–4] This effect, discovered 50 years ago,[1] is used nowadays to set a reference magnetization direction in thin-film spin-valve devices and, therefore, is of great interest for a broad range of applications based on spintronics.[5] Despite the enormous technological impact and the intense research efforts, one of the main challenges to understand the microscopic mechanisms of EB is the investigation of the magnetic structure at the F/AF interface and its depth dependence perpendicular to the interface (z direction). In fact, in some EB models a spiraling AF spin structure along the z axis, i.e., planar AF domain walls (DW) or incomplete (partial) domain walls (IDW) upon magnetization reversal have been predicted.[6–10] Such DW or IDW in the AF, parallel to the interface, have been inferred from experiments for several systems.[11–14] Although similar noncollinear spin structures in the F layer were theoretically predicted by Néel[15] and used to explain certain experimental results,[16,17] they were included only in few EB models[3,18] and have been rarely systematically studied.[19]

Experimentally, different techniques allow the study of buried magnetic structures. However, it is always difficult and challenging to obtain a detailed description of the depth-dependent magnetic structure at the atomic scale. For example, polarized neutron reflectometry (PNR) studies give rarely indication of EB-induced depth-dependent magnetization in the F layer although the presence of parallel DW in the Co layer of Co/FeF_2 has been demonstrated.[20] Magneto-optical Kerr effect measurements probing both the F/AF and F/air interfaces give also strong indication of the existence of depth-dependent structure in the F layer.[19] Interestingly, conversion electron Mössbauer spectroscopy (CEMS) studies using ^{57}Fe probe layers at different depths of the F in exchange-biased Fe/MnF_2 do not indicate depth-dependent spin structure in Fe.[21,22] However, the CEMS measurements were conducted in remanence[21,22] due to the inherent difficulties of detecting electrons while applying strong external magnetic fields. Coherent nuclear resonant scattering (NRS) of synchrotron radiation is a particularly promising technique which uses buried ultrathin (a few angstroms thick) isotopic ^{57}Fe probe layers in combination with wedge-type Mössbauer inactive ^{56}Fe layers to provide, with high lateral resolution, depth-dependent properties in magnetic films and multilayers.[23,24]

Here, we report the direct measurement of the depth-dependent Fe spin rotation in an exchange-coupled Fe/MnF_2 bilayer using NRS of synchrotron radiation from an ^{57}Fe-probe layer buried at different depths within the Fe layer [Fig. 1(a)]. The results show that when exchange biased, the F layer develops a partial domain wall upon magnetization reversal, in contrast to most EB theories which do not take into account the z dependence of the F-spin structure.

II. EXPERIMENTAL PROCEDURE

The Fe/MnF_2 thin-film system of composition 40 Å Cu cap/70 Å Fe (=60 Å ^{56}Fe+10 Å ^{57}Fe)/520 Å $MnF_2(110)$/160 Å $ZnF_2(110)$ was grown on MgO(100) by molecular-beam epitaxy.[21] The 10-Å-thick ^{57}Fe layer (95.5%

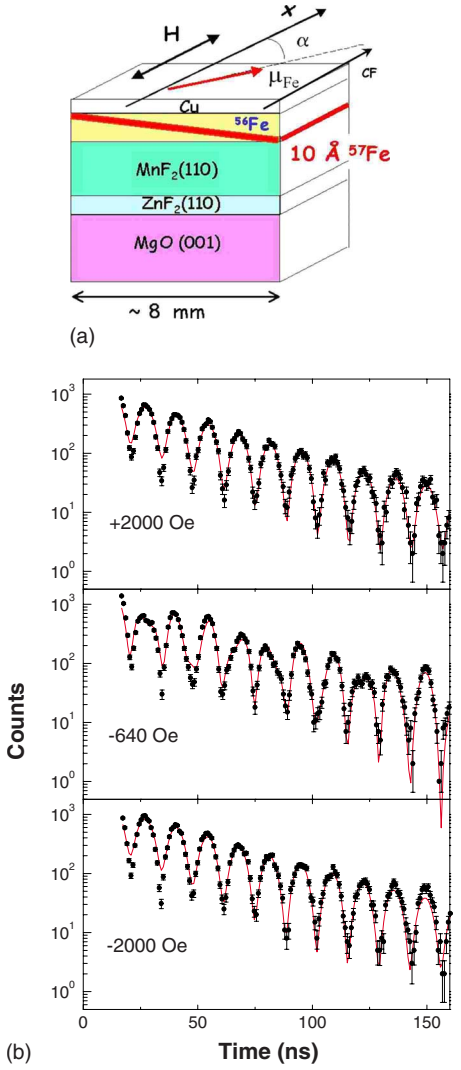

FIG. 1. (Color online) (a) Schematic diagram of the wedge sample and NRS experimental geometry: the incident photon beam, reflected at an angle of 4 mrad relative to the surface, is oriented along the x direction (note that all the arrows are in the sample plane). (b) Typical 10 K NRS time spectra measured in decreasing magnetic fields with the 14.4 keV photons probing the ^{57}Fe center position. The red solid lines are least-squares fits to the experimental data. The +2000 Oe cooling field (CF) (applied from 150 to 10 K) and the sweeping field H were applied in plane along the MgO[100] (x) direction. α is the angle between the in-plane Fe spin direction and the $+x$ direction.

isotopical enrichment) was inserted diagonally, i.e., between two wedge-shaped ^{56}Fe layers, as illustrated in Fig. 1(a). The MnF_2 film grows as (110)-oriented pseudotwinned quasiepitaxial layer and forms a compensated AF surface with the Mn spins in the interface plane, whereas the Fe is polycrystalline.[21] X-ray diffraction indicated a rocking curve width $\Delta\Theta=3°$ for the MnF_2 layer. Room-temperature CEMS studies (not shown) indicate a dominant α-Fe sextet and also contributions of subspectra with reduced magnetic hyperfine fields (B_{HF}), attributed to chemical intermixing (corresponding to 3 Å of ^{57}Fe) at and close to the Fe/MnF_2 interface. Moreover, the CEMS results indicate a fully in-plane magnetized Fe layer, in agreement with previous works.[21,22] Magnetic hysteresis loops below the MnF_2 Néel temperature ($T_N=67$ K) were measured using superconducting quantum interference device (SQUID) magnetometry. EB was always established by field cooling (FC) the sample from 150 to 10 K in an external field (H) of 2000 Oe applied in plane along the MgO[001] direction [x direction, Fig. 1(a)]. Measurements were conducted between +2000 and −2000 Oe along the same direction.

The NRS experiments were performed at beamline ID18 of the European Synchrotron Radiation Facility (ESRF), with the incident photon beam oriented along the MgO[001] direction [the x direction shown in Fig. 1(a)], which is at ±45° relative to the two AF MnF_2 [110] twin easy axes.[21] The sample was illuminated in grazing incidence geometry at an angle of about 4 mrad. A cryomagnet system on top of a two-circle element and a two-axis adjustable table allowed fine adjustment and scanning of the sample position (together with the whole cryostat unit) in front of the 14.4 keV beam [cross section 50 μm(vertical) × 300 μm (horizontal)]. This provides the depth selectivity of the measurements along the wedge with the ^{57}Fe probe layer at different distances from the F/AF interface. We have selected two beam positions: the *center* position, which is 24 Å away from the F/AF interface, and the *top* position, which is 60 Å away. In NRS, the time response of the forward scattered intensity reflected from the ultrathin ^{57}Fe probe layer is measured.[23,24] We assume the magnetization to be confined in the sample plane.[21] For the 14.4 keV ^{57}Fe resonance the standard NRS geometry (with incident σ polarization and no polarization analysis in the detection) was employed.[24] The relative orientation of the in-plane Fe magnetic moment (μ_{Fe}) with respect to the incident wave vector directed along x (azimuthal angle α) is determined from the time-dependent delayed scattered intensity.[23,24] This provides the same information as obtained from the line intensity ratio in conventional Mössbauer spectroscopy.[21] The measured time spectra were least-squares fit using the simulation and least-squares fitting procedure of the CONUSS program.[25]

III. RESULTS AND DISCUSSION

Figure 1(b) shows typical 10 K NRS time spectra for selected magnetic fields along the decreasing-field branch of the hysteresis loop. From the fitting of the NRS data, a B_{HF} value of 34.1 ±0.2 T at 10 K was obtained for all time spectra of the center and top positions in the whole applied magnetic-field range. This is the expected value of B_{HF} for bcc Fe, and it agrees well with CEMS results on ^{57}Fe probe layers in Fe/MnF_2 bilayers.[21] Assuming a unidirectional collinear Fe spin structure within the depth interval sensed by the x-ray beam at either position, the in-plane spin-rotation angle α of the Fe magnetic moment as a function of H was determined. Although NRS of linearly polarized x rays cannot distinguish between angles $\pm\alpha$ and angles $(180° \pm \alpha)$ (Fe spin components along positive or negative H, respectively), the sign of the SQUID magnetization provides the magnetization reversal points [where the Fe spins change from being predominantly along the positive field ($+x$) direc-

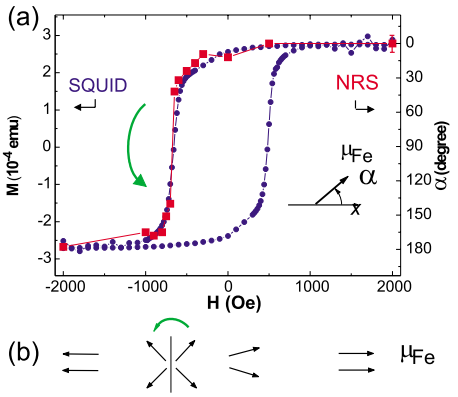

FIG. 2. (Color online) (a) Comparison between the SQUID magnetization loop (blue circles) at 10 K and the NRS in-plane Fe spin-rotation angle α (red squares) at 10 K for the descending-field branch at the ^{57}Fe top position. (b) Schematics of the possible spin configuration within the ^{57}Fe probe layer along the loop. The curved arrows indicate the jump of the Fe spin configuration at the magnetization reversal. The error bars of α in the region of reversal are $\pm 3°$.

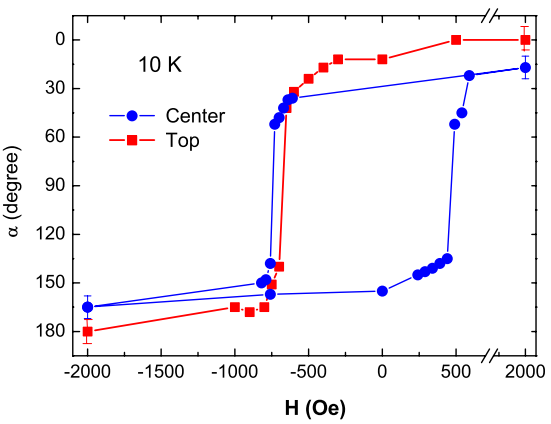

FIG. 3. (Color online) Complete 10 K NRS angular hysteresis loop at the ^{57}Fe center position (24 Å from the Fe/MnF_2 interface, blue full circles) and descending-field branch (at 10 K) at the ^{57}Fe top position (60 Å from the interface, red full squares). The depth dependence of the Fe spin structure at 10 K is reflected in the 40 Oe shift and in the different values of α at ± 2000 Oe between the curves. The center and top data were measured in two independent NRS runs after FC in +2000 Oe from 150 to 10 K. The error bars of α near the reversal are $\pm 3°$.

tion to predominantly along the negative field ($-x$) direction]. Following this approach a NRS angular hysteresis loop can be constructed. Note that due to interface intermixing and the resulting complicated hyperfine-field distribution,[22] time spectra corresponding to the very Fe/MnF_2 interface could not be interpreted in this straightforward manner. Thus, NRS results corresponding to ^{57}Fe atoms only at 24 Å away from the interface (center position) and close to the top of the Fe film (60 Å away from the interface, the top position) are presented. This has no effect on the results presented here. Moreover, the Cu cap layer has no influence on the hyperfine parameters of the bcc ^{57}Fe probe layer located 10 Å below the Cu layer (top position), as checked on test samples by CEMS and in agreement with previous work.[26]

The 10 K SQUID magnetization loop and the NRS in-plane Fe spin-rotation angle α for the top position (decreasing-field branch) as a function of H are shown in Fig. 2(a), where the typical error bar for α is indicated. Good agreement is obtained between the SQUID and the NRS result. We observe that the in-plane Fe spins first rotate continuously and weakly from an alignment along the positive applied field direction ($\alpha=0°$) at +2000 Oe to a direction with $\alpha=42°$ and subsequently show an unexpected "jump" to $\alpha=138°$, near $H=-H_C$, followed by a gradual rotation to finally align along the negative field at −2000 Oe ($\alpha=180°$). The corresponding Fe spin configuration within the ^{57}Fe probe layer (angle α) and the observed directional spin-jump near $-H_C$ is sketched in Fig. 2(b) (based also on earlier CEMS studies on similar samples[21]). Similarly, along the increasing-field branch, a jump of the Fe spin direction is observed near $H=+H_C$, as shown in Fig. 3 for the center position (full circles). Remarkably, with our results we have identified the angles between which the Fe magnetization jumps.

Strikingly, α values near the magnetization reversals are close to the AF easy axes directions (45° and 135°) of the MnF_2 twin domains. Apparently, due to the strong interfacial exchange coupling of Fe spins along (or perpendicular to) the AF easy axis,[27] the Fe spins jump by 90° (spin flip) near $H=-H_C$ along the decreasing field branch. This indicates that there is an angular instability range perpendicular to the cooling-field direction for the Fe spins on top of twinned AF MnF_2(110).

The depth-dependent magnetization reversal at 10 K manifests itself in the field dependence of α along the decreasing-field branches measured by NRS at the ^{57}Fe top position (i.e., 60 Å from the Fe/MnF_2 interface) and the center position (i.e., 24 Å from the interface). Figure 3 shows the full 10 K angular hysteresis loop for the center position (full blue circles) and the descending-field branch at the top position (full red squares). The top layer ^{57}Fe spins revert earlier (by ~40 Oe) than the Fe spins at the center layer, implying stronger pinning of the Fe atoms closer to the exchange-biased Fe/MnF_2 interface.

Moreover, a clear difference in the magnetic-field response at 10 K can be observed between the ^{57}Fe spins at different depths. At +2000 Oe, the top layer spins are fully aligned with the field ($\alpha=0°$), but the spins at the center of the film (i.e., 24 Å from the interface) are misaligned with respect to the field direction ($\alpha=17°$). The same tendency is observed at −2000 Oe.

It is interesting to note also that at the different depths (center or top) both site-selective NRS loops (at 10 K) are characterized by continuous rotations, followed by jumps of the Fe spin direction at $H=-H_C$ (or $H=+H_C$) from about $\alpha=\pm 45°$ (or $\alpha=\pm 135°$) to $\alpha=\pm 135°$ (or $\alpha=\pm 45°$) at magnetization reversal as the applied magnetic field is decreased (increased). This is a direct observation of the fourfold magnetic anisotropy induced by the exchange interaction between the Fe and MnF_2 layers, as observed earlier by (global) magnetoresistance hysteresis loops.[28] The EB field ($H_E=-50\pm 10$ Oe), coercivity ($H_C=620\pm 10$ Oe), and loop

shapes obtained from the ^{57}Fe probe layer NRS (Fig. 3) are in reasonable agreement with $H_E=-70$ Oe and $H_C=550$ Oe measured at 10 K by SQUID magnetometry of the whole sample. In contrast, above the MnF_2 Néel temperature (at 150 K) the Fe spins seem to rotate more continuously during magnetization reversal according to NRS (not shown), without evidence for a depth dependence of the Fe spin rotation angle α.

IV. SUMMARY

In conclusion, we found a clear depth-dependent Fe spin rotation in exchange biased Fe/MnF_2 bilayers using NRS of synchrotron radiation and an ^{57}Fe probe layer at different distances from the Fe/MnF_2 interface. Well below the AF Néel temperature (at 10 K), the Fe spins rotate continuously from $\alpha=0°$ toward $\pm 45°$, then jump discontinuously to $\alpha=\pm 135°$, and follow a continuous rotation to $\alpha=180°$ as the field is decreased. The jumps occur from one of the twinned MnF_2 easy axis ($\pm 45°$) to the other ($\pm 135°$). Moreover, the reversal and saturation are easier to achieve at 60 Å than at 24 Å from the interface. This implies that the spin rotation in an exchange-biased system is like in a spring magnet forming a partial domain wall, as predicted by Kiwi *et al.*[29,30] This result sets decisive restrictions to theories of exchange bias.

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*Present address: Swiss Light Source, Paul Scherrer Institut, 5232 Villigen, Switzerland.

†Present address: Deutsches Elektronen-Synchrotron, 22067 Hamburg, Germany.

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