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Observation of the bulk spin-flop in an Fe/Cr superlattice

L. Bottyán[a,]*, L. Deák[a], J. Dekoster[b], E. Kunnen[c], G. Langouche[b],
J. Meersschaut[b], M. Major[a,b], D.L. Nagy[a], H.D. Rüter[d], E. Szilágyi[a], K. Temst[c]

[a] KFKI Research Institute for Particle and Nuclear Physics, P.O. Box 49, 1525 Budapest, Hungary
[b] Instituut voor Kern-en Stralingsfysica, K.U. Leuven, Celestijnenlaan 200D B-3001, Leuven, Belgium
[c] Laboratorium voor Vaste-Stoysica en Magnetisme, K.U. Leuven, Celestijnenlaan 200C B-3001, Leuven, Belgium
[d] II. Institut für Experimentalphysik Universität Luruper Chaussee 149 D-22761, Hamburg, Germany

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Abstract

The layer magnetisation reorientation transition (spin-flop, SF) was studied in an artificial layer antiferromagnet (AF), namely in MgO(0 0 1)/[^{57}Fe(2.6 nm)/Cr(1.3 nm)]$_{20}$ epitaxial superlattice (SL) by synchrotron Mössbauer reflectometry and Kerr effect (SMOKE). The SF occurs simultaneously in the entire SL stack (bulk SF) in an increasing field of $H_{\mathrm{SF}} = 13\,\mathrm{mT}$ along the easy direction parallel to the layer magnetisations. It is recognised by the kink in the SMOKE loop and by the sharp up-rise of the AF Bragg peak in the delayed Mössbauer reflectivity. The moderate value of observed H_{SF} is compared with estimations from a spin-chain model and interpreted as due to intraplane domain-wall motion during SF. © 2001 Published by Elsevier Science B.V.

Keywords: Artificial superlattices; Interlayer coupling; Kerr measurements; Mössbauer spectroscopy; Synchrotron radiation

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An interesting model system of an ‘artificial layer antiferromagnet’ is a periodic Fe/Cr antiferromagnetic (AF) superlattice (SL) with even number of Fe layers. When the external magnetic field is aligned along the easy axis of the Fe layers parallel/antiparallel to the magnetizations $\underline{\mathrm{M}}_k (k = 1, 2, 2n)$, the anisotropy-stabilised configuration becomes energetically unfavourable at a certain critical in-plane field strength and a sudden magnetisation reorientation is expected in a finite multilayer stack [1–4] with *surface spin-flop* [5,6] or *bulk spin-flop* (BSF) [7] scenarios, in the cases of uniaxial and four-fold in-plane anisotropy, respectively.

Synchrotron Mössbauer Reflectometry (SMR, [8–11]) is sensitive to the alignment of local hyperfine fields in the film. Consequently, in an ^{57}Fe-containing magnetic SL, the Fe-layer magnetisation directions can be determined relative to the photon’s propagation and polarisation vectors [8,9]. Time integral (TI) SMR records the total number of delayed photons as a function of the angle of grazing incidence Θ. Structural Bragg peaks due to the electronic SL periodicity are observed in the prompt and in the delayed signal, but the magnetic (hyperfine) super-cell doubling in an AF SL appears only in the delayed TISMR. The AF Bragg-peak intensity in TISMR is at maximum for the photon’s wave vector $\underline{\mathrm{k}}$, parallel/antiparallel to $\underline{\mathrm{M}}_k$, and zero for $\underline{\mathrm{k}} \perp \underline{\mathrm{M}}_k$ [3]. Therefore, SMR is especially suitable for studying the spin-flop (SF) phenomena. Here, we report on TISMR of the (bulk) SF in a Fe/Cr AF SL with a four-fold in-plane anisotropy. The observed H_{SF} is compared with a spin-chain calculation with the aim of elucidating the magnetisation reorientation mechanism.

The [^{57}Fe(2.6 nm)/Cr(1.3 nm)]$_n$ ($n = 20$) periodic multilayer was grown on a MgO(0 0 1) substrate at 450 K by MBE using an electron beam gun (Cr) and a Knudsen cell (^{57}Fe) at a base pressure of 1×10^{-9} mbar following a degassing of the substrate at 873 K for 30 min. RHEED patterns and high-angle X-ray diffractograms confirmed the epitaxial quality and excellent layering of the SL film. Low-angle X-ray diffraction at $\lambda = 0.086\,\mathrm{nm}$ (Fig. 1a) showed extended Kiessig-fringes

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*Corresponding author. Tel.: +36-1-392-2761; fax: +36-1-395-9151.

E-mail address: battyan@rmki.kfki.hu (L. Bottyán).

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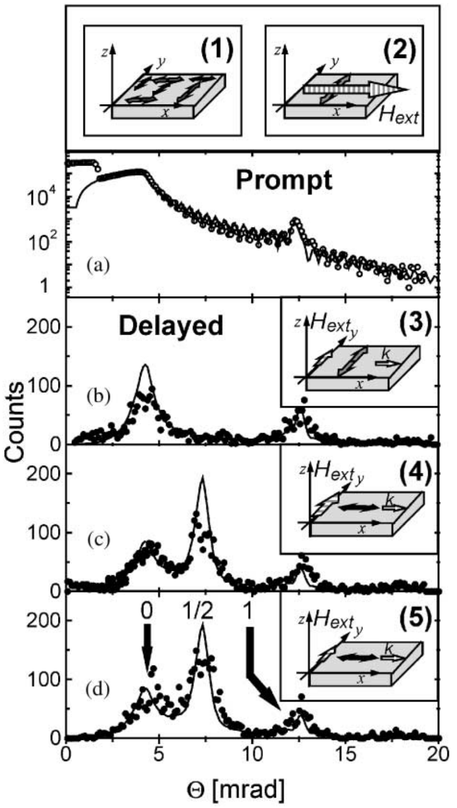

Fig. 1. Prompt (a) and TISMR (b–d) scans of MgO(0 0 1)/[^{57}Fe(2.6 nm)/Cr(1.3 nm)]$_{20}$ superlattice taken in (b): 3 mT, (c): 35.3 mT and (d): repeated 3 mT magnetic fields, respectively. The appearance of the $\frac{1}{2}$-order AF Bragg peak shows the reorientation of the layer magnetisations at a bulk SF transition field H_{SF}. The system of co-ordinates in the insets is fixed to the substrate.

and structural SL reflections up to the third order (not shown) with a bilayer period of 3.9 nm and root-mean-square interface roughness of 0.43 nm. The Fe/Cr thickness ratio was determined by Rutherford backscattering. Conversion electron Mössbauer spectroscopy revealed an in-plane orientation of the Fe moments, an expected consequence of the shape anisotropy.

^{57}Fe SMR experiments were performed on the BW4 nuclear resonance beamline in HASYLAB, Hamburg, at room temperature in vertical scattering geometry. Motorized permanent magnets provided horizontal fields between 3 and 46 mT perpendicular to $\underline{k}$. TISMR scans were recorded at grazing angles between 0 and 20 mrad. The SMR results are shown in Fig. 1. The solid lines in (a)–(d) are simulations [9,12]. Peaks labelled '0', '$\frac{1}{2}$' and '1' are the total reflection peak [13,14], the AF Bragg peak and the structural Bragg peak, respectively. The presence or absence of the $\frac{1}{2}$ peak reveals if $\underline{k} \parallel \underline{M}_k$ or $\underline{k} \perp \underline{M}_k$.

In the SL, $\underline{M}_k$ points parallel or anti-parallel to either of the Fe[0 1 0] or Fe[1 0 0] easy axes in the film plane, with AF domains oriented at random, parallel/antiparallel with either of those (inset (1) in Fig. 1). The initial magnetic state was carefully prepared by aligning $\underline{M}_k$ along a *single* easy axis: the SL film was magnetised to 46 mT in order to induce an SF (inset (2)). Then, the field was decreased to 3 mT and the sample was rotated through $\pi/4$. At this point, TISMR scan $1/b$ was recorded. Since in this state (inset (3)), $\underline{k} \perp \underline{M}_k$, no AF superreflections were observed (Fig. 1b, inset (4)). Having increased the field to 35.3 mT, the $\frac{1}{2}$-order AF Bragg peak appeared (Fig. 1c) as a direct evidence of the BSF. Due to the four-fold anisotropy, this state was *preserved* when the magnetic field was decreased again to 3 mT (inset (5)). Accordingly, the $\frac{1}{2}$-order AF Bragg-peak intensity did not change (Fig. 1d).

The BSF transition was also confirmed by SMOKE (Fig. 2). High-field loops (see inset in Fig. 2) were indicative of AF coupling and a saturation field of $H_S \approx 0.9$ T. First, the $\underline{M}_k$ were prepared in an easy direction of Fe (by exerting and releasing a saturating field), afterwards, the sample was rotated through $\pi/4$. A kink was observed in the first loop around $H_{SF} = 13$ mT, which did not re-occur until the sample was turned to the perpendicular direction. This is in full agreement with the TISMR scans. A difference between SMOKE and TISMR is that the latter probes the entire multilayer stack at the AF Bragg angle, while the former remains more sensitive to the upper layers. The agreement indicates that the SF reorientation occurs simultaneously in the entire SL stack (bulk SF).

In order to relate the layer parameters to the measured H_{SF}, for simplicity, an infinite 'two-sublattice' spin-chain scheme is invoked. The energy E per unit area of a SL with quadratic anisotropy (experimentally found for Fe/Cr on MgO(0 0 1)) in an external

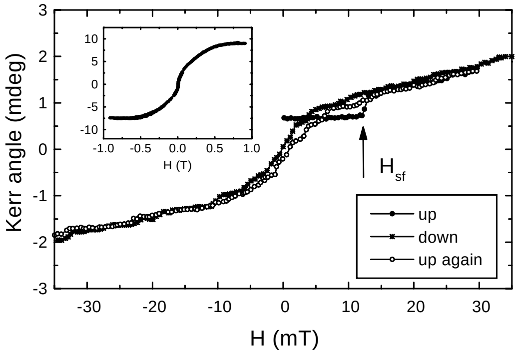

Fig. 2. Surface magneto-optical Kerr loops of a superlattice MgO(0 0 1)/[^{57}Fe(2.6 nm)/Cr(1.3 nm)]$_{20}$. SF occurs around $H_{SF} = 13$ mT only once following a 90° rotation of the substrate relative to the field direction.

field H is

$$\begin{aligned}E(H,\vartheta_1,\vartheta_2) = & -J_1\cos(\vartheta_1-\vartheta_2) - J_2\cos^2(\vartheta_1-\vartheta_2)\\ & + A(\sin^2 2\vartheta_1 + \sin^2 2\vartheta_2)\\ & - HM(\cos\vartheta_1 + \cos\vartheta_2),\end{aligned} \quad (1)$$

where J_1, J_2 and A are the bilinear and biquadratic coupling coefficients between the two sublattices and the magneto-crystalline energy (J_1, $J_2 < 0$, $A > 0$). The bulk anisotropy energy $K_1 = 4A/t_{\mathrm{Fe}}^{\mathrm{tot}}$, with $t_{\mathrm{Fe}}^{\mathrm{tot}}$and M being the Fe *sublattice* layer thickness (i.e. 26 nm in the present case), and moment per unit area $M = n|M_k|/2$, $k = 1, 2$. $\underline{\mathrm{M}}_1$ and $\underline{\mathrm{M}}_2$ decline by ϑ_1 and ϑ_2, respectively, from the field (the latter pointing along an easy direction of Fe). The SF and saturation occur in increasing and decreasing fields, at which the energy given by Eq. (1) is no longer positive-definite. The respective field values are $H'_{\mathrm{SF}} = 4\sqrt{A(4A - J_1 + 2J_2)}/M$ and $H_{\mathrm{S}} = -2(J_1 + 2J_2 + 4A)/M$. (As shown by Dantas and Carriço [4], H_{S} remains unaffected, while the SF field H'_{SF} is lowered in a finite SL stack due to the dangling surface layers. For a strongly AF-coupled finite SL ($-J_1 + 2J_2 \geqslant A$), this lower value of the SF field is $H_{\mathrm{SF}} \approx H'_{\mathrm{SF}}/\sqrt{2}$. This latter H_{SF} value is considered in the following estimations.) From the SMOKE loops, $H_{\mathrm{S}} \approx 0.9\,\mathrm{T}$. Using this and literature value of $K_1 = 4.5\,\mathrm{kJ/m^3}$ [15], H_{SF} was calculated. Assuming a pure bilinear coupling ($J_2/J_1 = 0$), this gives $H_{\mathrm{SF}}^{\mathrm{calc}} = 260\,\mathrm{mT}$. Allowing for a variation of $0 < J_2/J_1 < 0.45$, a range of $H_{\mathrm{SF}}^{\mathrm{calc}}$ was estimated. For $K_1 = 4.7\,\mathrm{kJ/m^3}$, $H_{\mathrm{SF}}^{\mathrm{calc}} > 130\,\mathrm{mT}$. Varying K_1 in a range as broad as $2.4\,\mathrm{kJ/m^3} < K_1 < 4.7\,\mathrm{kJ/m^3}$, $H_{\mathrm{SF}}^{\mathrm{calc}}$ remains by a factor of 5 above the measured value. These facts imply that, as expected, rather than by coherent rotation of the sublattice magnetisations, the SF is likely to occur by intralayer domain wall motion in this artificial layer antiferromagnet. The latter requires much lower field to overcome the anisotropy barrier. The balance between the Zeeman energy and the anisotropy energy at the SF field was found to be essential in shaping the AF domain structure [16].

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References

[1] F.C. Nötermann, R.L. Stamps, A.S. Carriço, R.E. Camley, Phys. Rev. B 46 (1992) 10847.

[2] M. Major, L. Bottyán, L. Deák, D.L. Nagy, in: E.A. Görlich, A. Pedziwiatr (Eds.), Proceedings of the XXXIV, Zakopane School of Physics, Jagellonian University, Cracow, 1999, p. 165.

[3] M. Major, Master’s Thesis, Eötvös Loránd University, Budapest, 1999 (in Hungarian).

[4] A.L. Dantas, A.S. Carriço, Phys. Rev. B 59 (1999) 1223.

[5] R.W. Wang, D.L. Mills, E.E. Fullerton, J.E. Mattson, S.D. Bader, Phys. Rev. Lett. 72 (1994) 920.

[6] N.S. Almeida, D.L. Mills, Phys. Rev. B 52 (1995) 13504.

[7] K. Temst, E. Kunnen, V.V. Moshchalkov, H. Maletta, H. Fritzsche, Y. Bruynseraede, Physica B 276–278 (2000) 684.

[8] D.L. Nagy, L. Bottyán, L. Deák, E. Szilágyi, H. Spiering, J. Dekoster, G. Langouche, Hyperfine Interactions 126 (2000) 349.

[9] L. Deák, L. Bottyán, D.L. Nagy, H. Spiering, Phys. Rev. B 53 (1996) 6158.

[10] R. Röhlsberger, Hyperfine Interactions 123/124 (1999) 455.

[11] A.I. Chumakov, D.L. Nagy, L. Niesen, E.E. Alp, Hyperfine Interactions 123/124 (1999) 427.

[12] H. Spiering, L. Deák, L. Bottyán, Hyperfine Interactions 125 (2000) 197.

[13] A.Q.R. Baron, J. Arthur, S.L. Ruby, A.I. Chumakov, G.V. Smirnov, G.S. Brown, Phys. Rev. B 50 (1994) 10354.

[14] L. Deák, L. Bottyán, D.L. Nagy, Hyperfine Interactions 92 (1994) 1083.

[15] H.-P. Klein, E. Keller, Phys. Rev. 144 (1966) 372.

[16] D.L. Nagy, L. Bottyán, B. Croonenborghs, L. Deák, B. Degroote, J. Dekoster, H.J. Lauter, V. Lauter–Pasyuk, O. Leupold, M. Major, J. Meersschaut, O. Nikonov, A. Petrenko, R. Rüffer, H. Spiering, E. Szilágyi, Phys. Rev. Lett, submitted for publication.