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Specular and Off-Specular Synchrotron Mössbauer Reflectometry: Applications to Thin Film Magnetism

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Synchrotron Mössbauer Reflectometry (SMR) is a novel tool for studying the magnetic structure of multilayers. The orientation of the layer magnetisation in an antiferromagnetically coupled multilayer is determined from the intensity of the pure nuclear reflection in specular time-integral SMR experiments. The value of the saturation field is estimated with high accuracy. The bulk spin-flop transition in an Fe/Cr superlattice of fourfold in-plane magnetocrystalline anisotropy is demonstrated. The width of the off-specular (diffuse) scattering peak is a measure of the in-plane antiferromagnetic domain size. The domain correlation length of 2.6 μm measured in remanence on the Fe/Cr superlattice following magnetic saturation is in good agreement with semi-empirical model calculations.

Introduction Since the first observation of the total external reflection (TER) of X-rays [1] and thermal neutrons [2], their grazing-incidence reflection from flat surfaces has been widely used to investigate the chemical, isotopic and magnetic structure of thin films and multilayers (ML). The real part of the index of refraction n of most materials for thermal neutrons and of all materials for non-resonant X-rays is by about 10^{-5} less than unity. The intensity of the reflected specular beam for $\Theta > \Theta_c = \sqrt{2(1-n)}$ rapidly decreases with increasing wave-vector transfer $Q = 2k \sin \Theta$, where k is the length of the wave vector of the incident radiation and Θ_c is the critical angle of the TER. The interference of the beams reflected from the interfaces of a stratified medium leads to patterns of the reflectivity versus wave-vector-transfer spectrum $R(Q)$ that bear information on the depth profile of the index of refraction $n(z)$, the argument z being the coordinate perpendicular to the sample surface. $R(Q)$ can be calculated from $n(z)$, e.g. using the method of *characteristic matrices* [3]. Therefore, in frames of a given model for the stratified system, $n(z)$ can be reconstructed (the parameters of the model can be fitted) from $R(Q) = |r(Q)|^2$, where $r(Q)$ is the reflectivity amplitude. As shown by Lax [4], the index of refraction is related to the scattering amplitude f by

$$n = 1 + \frac{2\pi N}{k^2} f , \tag{1}$$

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where N is the density of scatterers. This is the basic idea of specular *X-ray* and *neutron reflectometry*, two methods that can be used for mapping the electron density and the isotopic/magnetic structure of thin films, respectively. Indeed, the electron density for non-resonant X-rays or nuclear and magnetic scattering length density for neutrons is implied in f.

Mössbauer Reflectometry (MR) is X-ray reflectometry performed with nuclear resonant photons. Close to the nuclear resonance, f is strongly energy-dependent and contains the matrix elements of the hyperfine interactions. This is how MR is applied to study the magnetic structure of thin films. Due to the small ($\sim 10^{-5}$) solid angle involved in grazing incidence experiments, the well-collimated synchrotron radiation (SR) is much better suited for reflectometric experiments than radioactive sources.

Synchrotron Mössbauer Reflectometry (SMR) is the application of grazing incidence nuclear resonant scattering of SR [5] to thin film and ML structure analysis. SMR has recently been reviewed in various papers [6–10]. SMR and polarised neutron reflectometry can be mapped onto each other and a common optical formalism exists [11]. Starting with the pioneering work by Toellner et al. [12], SMR has by now become an established technique the most recent development being its extension to off-specular (diffuse) scattering [13].

In the present paper, we will show how time-integral SMR can be used for magnetic structure analysis of antiferromagnetically (AF) coupled MLs. The intensity of the AF superreflection turns out to be a sensitive measure of the orientation of the layer magnetisation. The 90° reorientation of the layer magnetisation on bulk spin-flop transition results in full appearance/disappearance of the AF reflection. Finally, off-specular SMR is used to measure the size of AF domains in a Fe/Cr superlattice after coercivity-limited domain ripening and spin-flop-induced domain coarsening. All SMR measurements have been performed at the nuclear resonance beamline ID18 of the European Synchrotron Radiation Facility, Grenoble.

Synchrotron Mössbauer Reflectometry The arrangement of SMR (Fig. 1) is very similar to that of any grazing-incidence scattering experiment. Photons from the high-resolution monochromator hit the sample mounted on a two-circle goniometer of adjustable height at an angle of grazing incidence ω. The scattered photons are detected by an avalanche photo diode (APD) the aperture of which may be limited by a slit in front of the detector. The adjustable detector height defines the scattering angle 2Θ.

Any nuclear resonant scattering experiment utilises the delayed photons which follow the primary excitation by the synchrotron radiation bunch within the lifetime of the resonant nuclear level. An SMR measurement is performed in either time integral or time differential regime. Time integral SMR (TISMR), which we shall use throughout this paper, records the total number of delayed photons from t_1 to t_2 as a function of ω and/or Θ. Here t_1 is a few nanoseconds determined by the bunch quality of the radiation source and by the dead time of the detector and the electronics, while t_2 is set to a value somewhat below the bunch repetition time of the storage ring.

Position-sensitive detectors have recently been increasingly applied in X-ray and neutron reflectometry in order to map the scattered intensity in a single experiment on the whole (ω, Θ) plane. Although APD arrays are being tested at various synchrotron facilities, as yet, TISMR experiments have only been performed in two different single-parameter geometries, viz., Θ–2Θ scan and ω scan. In a Θ–2Θ scan the sample orienta-

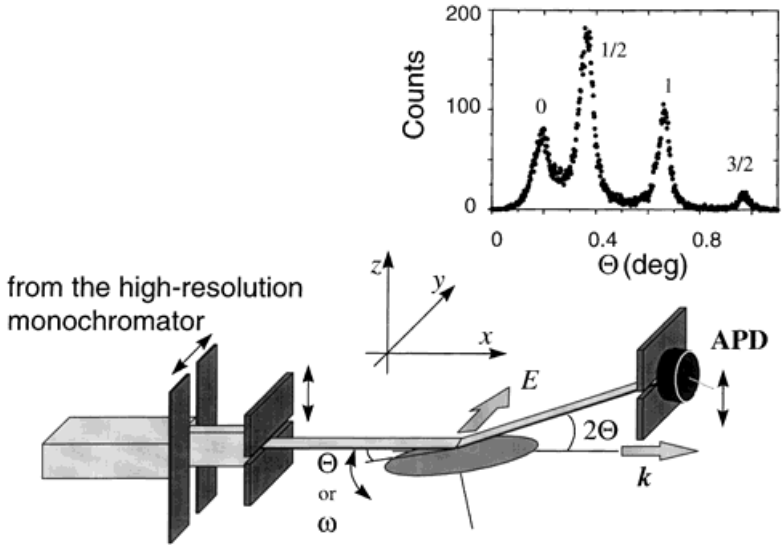

Fig. 1. Experimental set-up of an SMR experiment. The inset shows a Θ–2Θ scan measured on a MgO(001)/[^{57}Fe(26 Å)/Cr(13 Å)]$_{20}$ multilayer with layer magnetisation parallel to the photon beam. The order of reflections is indicated, half-order reflections being the antiferromagnetic peaks of pure nuclear origin

tion and the detector height are simultaneously changed fulfilling the constraint of specular reflection, $\omega = \Theta$. In an ω-scan experiment 2Θ is fixed and ω is varied. As a rule, a scan of the *prompt* photons (i.e., X-ray reflectometry) is recorded along with a delayed TISMR scan.

In a Θ–2Θ experiment the wave-vector transfer Q is perpendicular to the sample surface. For a periodic ML, in the first Born approximation (kinematic theory), Bragg maxima of m-th order appear at $Q = \sqrt{(2m\pi/d)^2 + Q_c^2}$, where d is the bilayer thickness and Q_c is the critical wave vector transfer of the TER (typically about 0.5 nm^{-1}). Thus, a Θ–2Θ scan reveals the plane-perpendicular structure. Akin to neutron reflectometry, the AF superstructure of a ML may result in superreflections of half-integer order m in a TISMR Θ–2Θ experiment.

In an ω scan experiment the condition of specular reflection is not fulfilled for $\omega \neq \Theta$. Off-specular scattered intensity is only significant in case of lateral inhomogeneities. In fact, for small values of ω and Θ in such an experiment, the perpendicular-to-plane component of the wave vector transfer is constant ($Q_z = 2k\Theta$) while varying ω, the in-plane parallel-to-beam (longitudinal) component of the wave vector transfer is scanned: $Q_x = 2k\Theta(\omega - \Theta)$. In order to have significant intensity, the detector height is set to meet the Q_z value of a Bragg peak. The width of the ω-scan (i.e., Q_x scan) is, in first Born approximation, inversely proportional to the lateral, longitudinal correlation length ξ of the quantity the perpendicular-to-plane periodicity of which the Bragg peak is related to

$$\xi = \frac{2\pi}{\Delta Q_x} = \frac{\pi}{k\Theta\,\Delta\omega}\,, \tag{2}$$

where ΔQ_x and $\Delta\omega$ are the peak widths of the Q_x and ω scans, respectively. Therefore, setting 2Θ in an ω-scan experiment to an electronically forbidden pure nuclear reflection the lateral correlation length of inhomogeneities of the hyperfine interaction (magnetic roughness, magnetic domains) can be determined.

Direction of the Layer Magnetisation in Coupled Multilayers, the Bulk Spin-Flop The magnetic-field dependence of the layer magnetisation direction in AF-coupled MLs bears information on interlayer coupling and magnetic anisotropy. The usual approach is measuring the net magnetisation by vibrating sample magnetometry, magnetooptic Kerr effect (MOKE), SQUID, etc. These methods yield quite accurate values of the susceptibility in small fields. It may not be, however, easy to determine the value of the saturation field H_s. In fact, due to biquadratic layer–layer coupling or a distribution of H_s, the magnetisation curve $M(H)$ is slowly saturating and H_s (or its mean value) may not be determined with a satisfactory accuracy.

The inset of Fig. 2 shows the Kerr loop taken on a MgO(001)/[^{57}Fe(26 Å)/Cr(13 Å)]$_{20}$ ML. The ML was grown on MgO(001) substrate at 450 K by MBE alternately depositing ^{57}Fe from a Knudsen cell and Cr from an electron gun at a rate of 0.1 and 0.35 Å/s, respectively, and base pressure of 4×10^{-10} mbar. Using high and low-angle X-ray diffraction as well as Rutherford backscattering a MgO(001)/[^{57}Fe(26 Å)/Cr(13 Å)]$_{20}$ epitaxial superlattice structure was found with MgO(001)[110] || Fe(001)[100]. The saturation field estimated from the Kerr loop is 0.6 T < H_s < 1.0 T a more accurate estimation from the Kerr loops being hardly possible.

TISMR can be efficiently used to determine the layer magnetisation direction in AF-coupled multilayers. In fact, the AF superreflection is suppressed for layer magnetisation perpendicular to the photon wave vector **k** and it is maximum for the parallel/antiparallel orientation [9]. In a magnetic field **H** applied perpendicular to **k**, the saturation is shown by the disappearance of the AF reflections and is, thereby, more easy to detect than from the magnetisation curve $M(H)$. Moreover, as numerical calculations

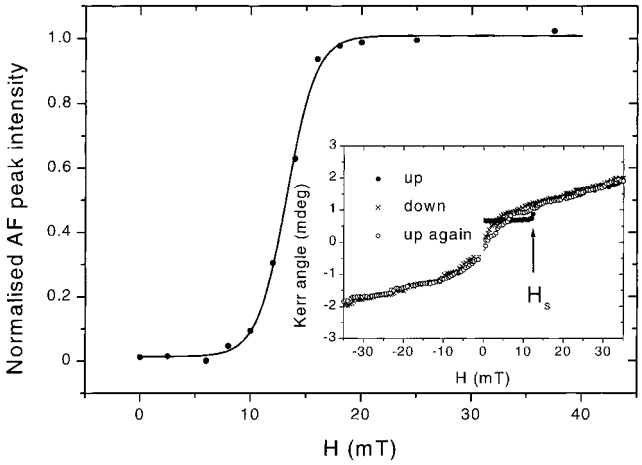

Fig. 2. Normalised intensity of the AF peak of a MgO(001)/[^{57}Fe(26 Å)/Cr(13 Å)]$_{20}$ multilayer vs. magnetic field H perpendicular to the photon beam during bulk spin-flop. The continuous line is guiding the eyes. The inset shows the MOKE angle vs. magnetic field

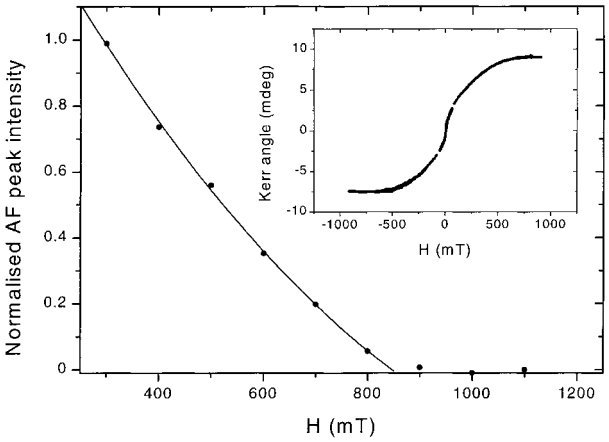

Fig. 3. Normalised intensity of the AF peak of a MgO(001)/$[^{57}Fe(26\,Å)/Cr(13\,Å)]_{20}$ multilayer in increasing magnetic field H perpendicular to the photon beam. The continuous line is guiding the eyes. The inset shows the MOKE angle vs. magnetic field

[14] for the above ML structure performed with an anisotropic optical algorithm [15, 16] show, the AF-peak intensity scales approximately with $\sin|\varphi|$ where φ is the angle between **H** and the layer magnetisation **M**. Therefore, the AF-peak intensity in this geometry is a very sensitive measure of the direction of **M**, and H_s can be determined with a high accuracy. Figure 3 shows the normalised intensity of the AF peak in increasing external field. The intensity decreases almost linearly with increasing field and $H_s = (840 \pm 30)$ mT is obtained from the extrapolation of $M(H)$ to $M = 0$.

The sensitivity of the AF-peak intensity on the direction of M has recently been used to directly demonstrate [17, 18] the *bulk spin-flop* (BSF) transition [19] on the same Fe/Cr ML of fourfold in-plane magnetocrystalline anisotropy. Indeed, when the external magnetic field is aligned along the easy axis of the Fe layers parallel/antiparallel to M, the anisotropy-stabilised configuration becomes energetically unfavourable at a certain critical in-plane field strength and a sudden magnetisation reorientation to the perpendicular easy direction associated with a sudden appearance of the AF peak takes place (cf. Fig. 3). The BSF transition is, at the same time, only associated with a minute change of the net magnetisation (cf. the inset in Fig. 3). Detailed analysis of the SMR study of the BSF transition will be published elsewhere [20].

Antiferromagnetic Domains in Multilayers Domain structure of AF-coupled multilayers is an issue of both theoretical and technological importance. Domain-size-dependent resistance noise, for example, may be as large as to limit GMR-sensor applications [21]. It is extremely difficult to visualise *in-plane AF domains* in a multilayer of few nm thickness. In fact, Kerr-microscopy has been performed so far only on thick trilayers [22, 23]. Therefore, indirect methods like resistance noise [21] and magnetoresistance [24] measurements, off-specular non-polarised [25] and polarised neutron reflectometry [26, 27] and, recently, soft-X-ray resonant magnetic diffuse scattering [28] have been used to estimate the AF-domain-size distribution in magnetic MLs.

Off-specular SMR is, as shown in Section 2, also suitable to investigate the in-plane correlation length of AF domains in coupled multilayers. Figure 4 shows off-specular SMR scans of the same Fe/Cr multilayer at the AF reflection of $\Theta = 0.4°$ in two different states, depending on the magnetic prehistory [13]. The domain size or, more precisely, the correlation length can be evaluated from the width of the off-specular ω scan

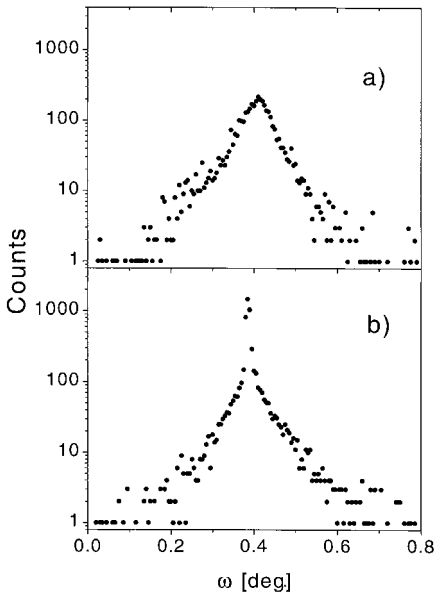

Fig. 4. Off-specular SMR scans of a MgO(001)/[^{57}Fe(26 Å)/Cr(13 Å)]$_{20}$ multilayer at the AF reflection: a) small-domain state, b) large-domain state

using Eq. (2). The broad line in scan Fig. 4a corresponds to AF microdomains of correlation length $\xi \approx 2.6$ μm. This state has been measured in remanence having released the sample from previous saturation ('unsaturation'). In contrast to this, scan Fig. 4b having been measured after a more sophisticated magnetic prehistory [13], is the sum of a broad diffuse shoulder (22% of the total area) and a narrow specular line (78%). In this state 22% of the multilayer consists of microdomains ($\xi \approx 2.6$ μm) while the majority of the multilayer contains large domains. Due to the finite aperture of the detector, only a lower limit of the correlation length ($\xi > 16.5$ μm) can be deduced from the width of the specular peak.

The expected size of the microdomains formed during unsaturation can be easily estimated. Leaving the saturation region, two kinds of AF patch domains are formed differing in the sense of rotation of their odd and even layers. The domain correlation length ξ on formation is determined by the structural lateral correlation length, i.e. by the terrace length of the ML. Supposing that the domain-wall density of the nested patch domains scales with $1/\xi$, these domains are spontaneously increasing in order to decrease the domain-wall energy. Small inclusions of one kind of domains in a large domain of the other kind have a domain-wall density scaling with ξ and will spontaneously decrease, in other words the large domains will increase at the expense of the embedded small ones. The spontaneous growth is limited by the coercivity H_c. There exists a critical size ξ_c so that the nested patch domains cannot grow beyond ξ_c and embedded domains disappear if smaller than ξ_c. Consequently, after this *ripening* process the domain correlation length will be close to ξ_c.

Supposing that the domain walls are Neél walls and neglecting the effect of anisotropy on the magnetisation direction within the domain wall, the domain-wall energy of a circular domain of diameter ξ is

$$E_w = \frac{\xi \pi p t_{\mathrm{Fe}}}{2} \left(\frac{A_{\mathrm{ex}} \pi^2}{l} + \frac{lK}{4} \right), \tag{3}$$

where p is the number of bilayers, t_{Fe} is the total thickness of Fe layers in the ML, $A_{\mathrm{ex}} = 2.1 \times 10^{-11}$ J/m is the exchange constant of bulk iron, $K = 4.7 \times 10^4$ J/m^3 is the anisotropy constant of iron and l is the width of the domain wall. For interlayer-coupling-dominated MLs [29] $l = (\pi/2)\,(A_{\mathrm{ex}} t_{\mathrm{Fe}}/J)^{1/2}$, where J is the interlayer coupling

constant ($J = 1.0$ mJ/m^3 for the ML under study). The energy dissipation by coercivity on changing the diameter of the domain by $\delta\xi$ is

$$\delta E_M = \xi \pi t_{Fe} M \mu_0 H_c \, \delta\xi \,, \tag{4}$$

where $M = 1.7 \times 10^6$ A/m is the saturation magnetisation of Fe and H_c is the coercivity. ξ_c can be determined from the condition $\delta E_w = \delta E_M$ leading to

$$\xi_c = \frac{\frac{A_{ex}\pi^2}{l} + \frac{lK}{4}}{2\pi M \mu_0 H_c} \,. \tag{5}$$

Approximating the nested patch domains with a chessboard-like structure, the same consideration results in the same value for ξ_c.

H_c cannot be directly measured on a ML of pure AF coupling. Using values measured on similar ferromagnetically coupled MLs ranging from $H_c = 2$ to 30 Oe, 0.6 μm $< \xi_c <$ 8.4 μm in excellent agreement with the value $\xi = 2.6$ μm measured after domain ripening.

Summary SMR has become an efficient tool in studying magnetic structure of multilayers. AF coupling results in pure nuclear superstructure reflections in specular TISMR experiments. The intensity of the AF reflection is a sensitive measure of the in-plane orientation of the layer magnetisation. Bulk spin-flop transition, i.e. a 90° reorientation of the layer magnetisation in increasing easy-axis field in a ML of fourfold in-plane anisotropy results in a sudden change of the AF peak intensity. The width of the off-specular (diffuse) scattering peak is inversely proportional to the in-plane antiferromagnetic domain correlation length. The average domain size of 2.6 μm measured in remanence on an Fe/Cr superlattice following magnetic saturation is the result of coercivity-limited domain-wall-energy-driven spontaneous growth (ripening) of the domains in decreasing field.

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