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Off-Specular Synchrotron Mössbauer Reflectometry: A Novel Tool for Studying the Domain Structure in Antiferromagnetic Multilayers

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Abstract. The off-specular (diffuse) nuclear resonant reflectivity of synchrotron radiation is a sensitive measure of the lateral autocorrelation of the magnetisation in thin films and multilayers. The width of the diffuse scattering peak measured at an electronically forbidden reflection is inversely proportional to the in-plane correlation length of the magnetisation direction. The average size of the in-plane antiferromagnetic domains is determined in different states of the same Fe/Cr superlattice. The hyperfine magnetic fields in coexisting small and large domains are measured independently.

Key words: magnetic multilayers, antiferromagnetic domains, off-specular scattering, Mössbauer reflectometry, nuclear resonant scattering of synchrotron radiation.

Grazing-incidence reflection of X-rays and neutrons from flat surfaces (X-ray and neutron reflectometry, the former also including soft X-ray resonant magnetic scattering) has been widely used to investigate the chemical, isotopic and magnetic structure of thin films and multilayers (ML). The sensitivity of nuclear resonant scattering of γ-radiation to hyperfine interactions enables a special kind of X-ray reflectometry, viz. that performed with nuclear resonant radiation (*Mössbauer reflectometry*, MR [1]). A serious limitation of MR with conventional sources [2] is the small ($\sim 10^{-5}$) solid angle involved. *Synchrotron Mössbauer reflectometry* (SMR) is the application of grazing incidence nuclear resonant scattering of SR [3] to thin film and ML structure analysis. SMR has recently been reviewed in various papers [4–8]. SMR and polarised neutron reflectometry can be mapped onto each other and a common optical formalism exists [9]. Starting with the pioneering work by Toellner *et al.* in which the existence of electronically forbidden superreflections in the grazing-incidence nuclear resonant reflectivity of synchrotron radiation of an antiferromagnetically (AF) coupled Fe/Cr ML was demonstrated [10], SMR has by now become an established technique.

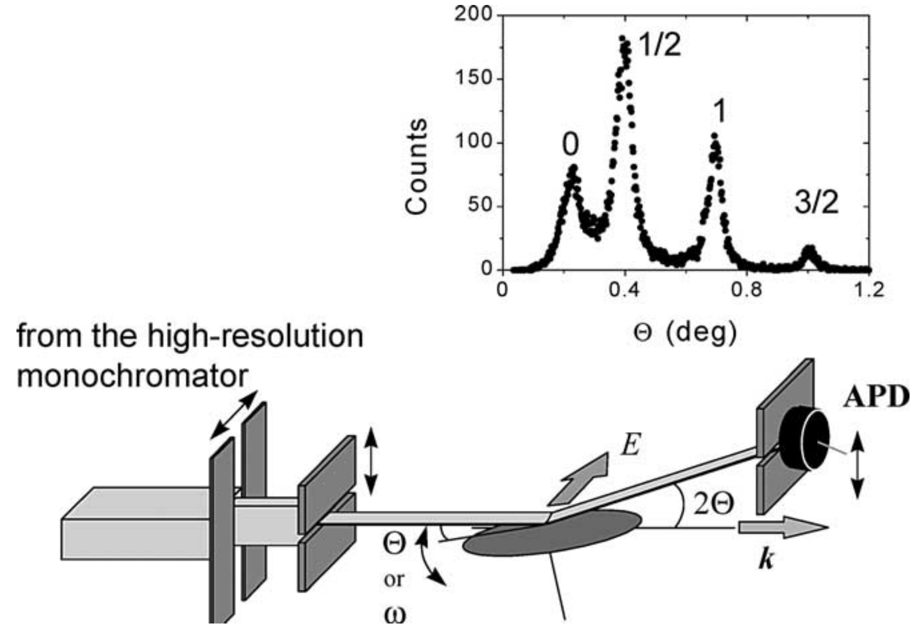

Figure 1. Experimental setup of an SMR experiment. The inset shows a Θ–2Θ scan measured at room temperature on a MgO(001)/[^{57}Fe(26Å)/Cr(13Å)]$_{20}$ ML with layer magnetisation parallel to the photon beam. The order of reflections is indicated, the half-order reflections being the antiferromagnetic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

Time-integral SMR can be efficiently used for magnetic structure analysis of AF coupled MLs. The intensity of the AF superreflection turns out to be a sensitive measure of the orientation of the layer magnetisation [6, 7]. Particularly, the 90° reorientation of the layer magnetisation on bulk-spin-flop transition results in full appearance/disappearance of the AF reflection [11].

In the present paper we will show that off-specular SMR can be used to measure the size of AF domains in coupled MLs. Especially the in-plane correlation length of the layer magnetisation in two different states of the same Fe/Cr ML will be determined. The quantum-beat patterns taken in specular and off-specular reflection will be used to compare the hyperfine fields of small and large domains. All measurements have been performed at the nuclear resonance beamline ID18 of the European Synchrotron Radiation Facility, utilising the 14.4 keV transition of ^{57}Fe.

The arrangement of an SMR setup (Figure 1) is very similar to that of any grazing-incidence scattering experiment. The 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. The nuclear resonant scattering process results in delayed photons, which are used in the SMR experiment.

As a rule, an SMR experiment is performed in Θ–2Θ geometry, i.e., so that the sample orientation and the detector height are simultaneously changed fulfilling the constraint of specular reflection, $\omega = \Theta$. One can, however, set the value of 2Θ and vary ω, a geometry called 'ω-scan'.

Since the scattering is elastic, 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), mth order Bragg maxima 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 total external reflection (typically about 0.5 nm^{-1}). Thus a Θ–2Θ scan reveals the plane-perpendicular structure. The AF superstructure of a ML may result in superreflections of half-integer order m in a time integral SMR Θ–2Θ experiment (i.e., when all delayed photons are recorded as a function of the angle of incidence).

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 (cf. the case of mosaicity in conventional X-ray diffraction). In fact, for the small values of ω and Θ in a grazing-incidence experiment, the perpendicular-to-plane component of the wave vector transfer is constant ($Q_z = 2k\Theta$) and by 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., of the 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:

$$\xi = \frac{2\pi}{\Delta Q_x} = \frac{\pi}{k\Theta\Delta\omega}. \tag{1}$$

Here 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.

Domain structure of AF-coupled MLs is an issue of both theoretical and technological importance. Off-specular non-polarised [12] and polarised neutron reflectometry [13, 14], soft-X-ray resonant magnetic diffuse scattering [15] and, very recently, off-specular SMR [16] have been used to estimate the AF-domain-size distribution in magnetic MLs.

The MgO(001)/$[^{57}Fe(26Å)/Cr(13Å)]_{20}$ ML was grown by MBE [16]. Figure 2 shows off-specular SMR scans of the same Fe/Cr ML taken at room temperature at the AF reflection of $\Theta = 0.39°$ in two different states, depending on the magnetic prehistory [16]. The domain size or, more precisely, the correlation length can be evaluated from the width of the off-specular ω-scan using Equation (1). The broad line in scan (a) corresponds to AF microdomains of correlation length $\xi \approx 2.6$ μm.

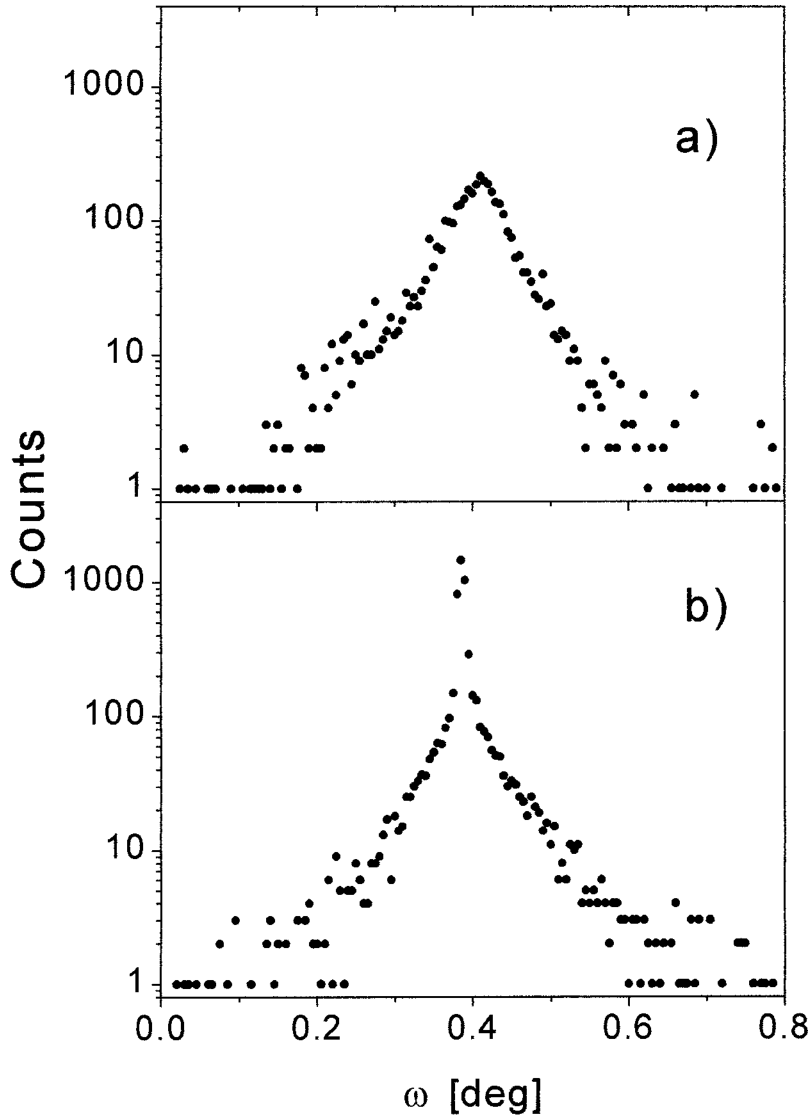

Figure 2. Off-specular SMR scans of a MgO(001)/[^{57}Fe(26Å)/Cr(13Å)]$_{20}$ ML at the AF reflection: (a) small-domain state, (b) large+small-domain state.

In contrast to this, scan (b) 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 ML consists of microdomains ($\xi \approx 2.6$ μm) while the majority of the ML 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.

It is possible to study the hyperfine interactions on the large and small domains of the same sample separately. Indeed, in first approximation, the specular and off-

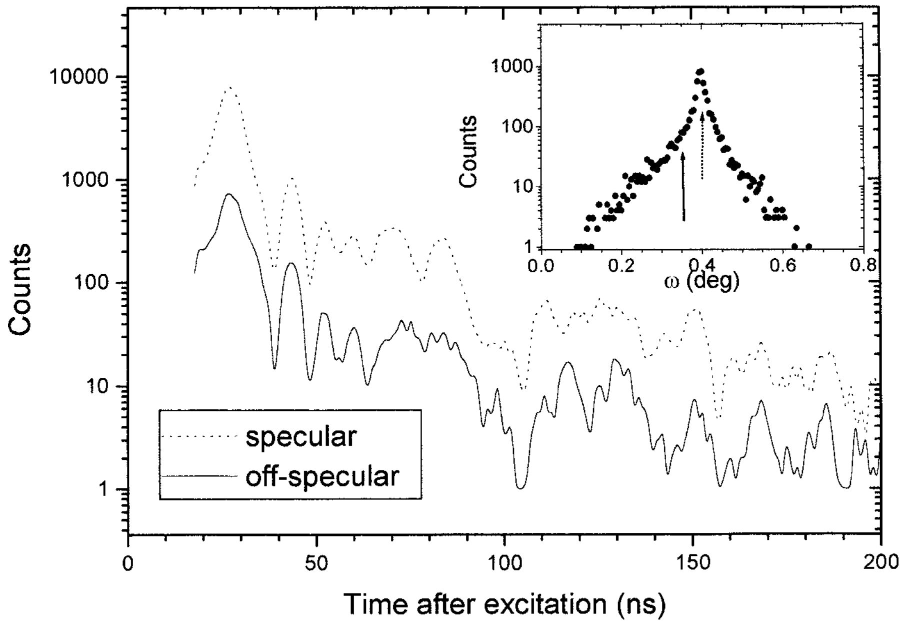

Figure 3. Time-differential SMR patterns of a MgO(001)/[57Fe(26Å)/Cr(13Å)]20 ML at the AF reflection ($2\Theta = 0.78°$) in the 'large+small'-domain state. Dotted line: $\omega = 0.39°$ (from large domains); full line: $\omega = 0.35°$ (from small domains). The inset shows the ω-values on the ω-scan corresponding to Figure 2(b).

specular scattering stems mainly from the large and small domains, respectively. Figure 3 shows time-differential SMR patterns taken at room temperature at the AF reflection ($2\Theta = 0.78°$) in the 'large+small'-domain state in the specular ($\omega = 0.39°$) and in an off-specular ($\omega = 0.35°$) direction. The beating frequencies of small and large domains are identical and correspond to an average hyperfine field of (34 ± 1) T. The difference in the beating depth will be analysed in the near future when the theory of time differential off-specular SMR will have been developed.

In conclusion, we used off-specular SMR to determine the lateral correlation length of the magnetisation direction in an AF-coupled Fe/Cr multilayer. Significant difference in the diffuse scattering width was found in two different states of the same sample showing a difference of at least one order of magnitude in the AF domain size. The magnitude of the hyperfine fields in coexisting small and large domains showed no significant difference.

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