Layer magnetization canting in ^{57}Fe/FeSi multilayer observed by synchrotron Mössbauer reflectometry

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Synchrotron Mössbauer reflectometry and CEMS results on a [^{57}Fe(2.55 nm)/FeSi (1.57 nm)]$_{10}$ multilayer (ML) on a Zerodur substrate are reported. CEMS spectra are satisfactorily fitted by α-Fe and an interface layer of random α-(Fe, Si) alloy of 20% of the ^{57}Fe layer thickness on both sides of the individual Fe layers. Kerr loops show a fully compensated AF magnetic layer structure. Prompt X-ray reflectivity curves show the structural ML Bragg peak and Kiessig oscillations corresponding to a bilayer period and total film thickness of 4.12 and 41.2 nm, respectively. Grazing incidence nuclear resonant Θ–2Θ scans and time spectra ($E = 14.413$ keV, $\lambda = 0.0860$ nm) were recorded in different external magnetic fields ($0 < B_{\text{ext}} < 0.95$ T) perpendicular to the scattering plane. The time integral delayed nuclear Θ–2Θ scans reveal the magnetic ML period doubling. With increasing transversal external magnetic field, the antiferromagnetic ML Bragg peak disappears due to Fe layer magnetization canting, the extent of which is calculated from the fit of the time spectra and the Θ–2Θ scans using an optical approach. In a weak external field the Fe layer magnetization directions are neither parallel with nor perpendicular to the external field. We suggest that the interlayer coupling in [Fe/FeSi]$_{10}$ varies with the distance from the substrate and the ML consists of two magnetically distinct regions, being of ferromagnetic character near substrate and antiferromagnetic closer to the surface.

1. Introduction

The Fe/Si system has attracted recent attention for exotic magnetic properties [1–5]. Fe MLs with FeSi spacer of CsCl (B2) structure show strong antiferromagnetic (AF) Fe–Fe layer coupling. Much consideration has been given to whether the coupling in the Fe/Si system has the same origin as in metal/metal MLs [1,3] and whether solely bilinear or bilinear + biquadratic layer coupling has to be considered [4–6]. In order to reveal details of the character and variation of the interlayer coupling, application of methods yielding information on the sublayer magnetization is of advantage.

Here we report on studies of a [^{57}Fe/FeSi]$_{10}$ ML on Zerodur substrate by the recently developed synchrotron Mössbauer reflectometry (SMR) [7–9]. SMR displays the effect of hyperfine interactions in form of time domain quantum beats. Through the

strong angular dependence of the penetration depth of the synchrotron radiation, SMR spectra sample the hyperfine interactions in the surface layer of the specimen in integrally increasing depth. In contrast to magnetometry, SMR yields information on the alignment of the individual sublayer magnetization, too. In Zerodur/$[^{57}\mathrm{Fe/FeSi}]_{10}$ ML we find a gradual decrease of the AF contribution in increasing external in-plane magnetic fields. The analyzed individual sublayer magnetization alignments are indicative of a two-block model.

2. Experimental results

A series of $[^{\mathrm{nat},57}\mathrm{Fe}/^{\mathrm{nat},57}\mathrm{FeSi}]_n$ MLs ($n = 10$, 15; 'nat' stands for natural isotopic abundance) was grown by molecular beam epitaxy on Zerodur substrates starting with an ^{57}Fe layer and FeSi layer on top. ^{57}Fe layers were evaporated from a Knudsen cell using a wire of 95% enriched ^{57}Fe isotope. The FeSi spacer layers with Fe of natural abundance were evaporated from electron beam guns at a distance of about 40 cm from the substrate. During iron deposition the target was rotated. Layer thickness was controlled by on-line mass spectroscopy. On $[\mathrm{Fe}/^{57}\mathrm{FeSi}]_n$ MLs, conversion electron Mössbauer spectroscopy (CEMS) showed a characteristic resonance of the metastable FeSi phase of CsCl (B2) structure [2,10]. $[^{57}\mathrm{Fe/FeSi}]_n$ MLs showed an α-Fe CEMS pattern with 0, 1 and 2 Si nearest neighbors (NN) of the probe nuclei (figure 1). Kerr loops of Zerodur/$[^{57}\mathrm{Fe/FeSi}]_{10}$ ML sample revealed a fully compensated AF state and no saturation up to the available maximum field of ± 0.35 T.

Grazing incidence prompt and delayed time integral (10–300 ns) Θ–2Θ scans (figure 2) as well as time spectra at selected angles were recorded at room temperature

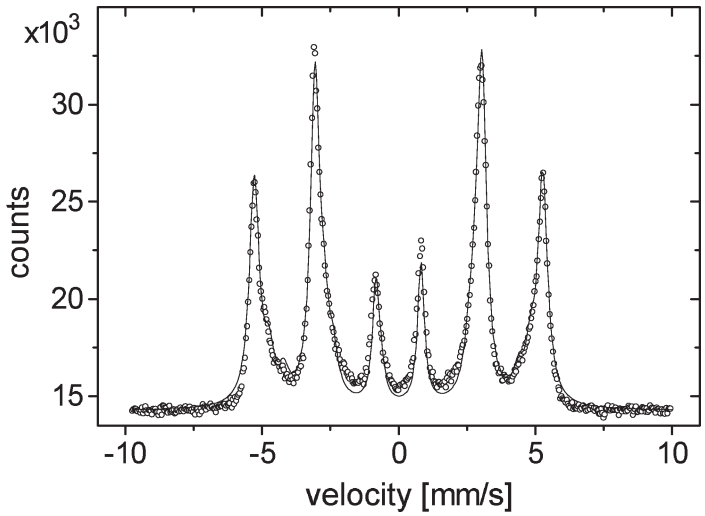

Figure 1. CEMS spectrum of Zerodur/$[^{57}\mathrm{Fe}/^{\mathrm{nat}}\mathrm{FeSi}]_{10}$ ML at room temperature. Contribution from the B2-$^{\mathrm{nat}}$FeSi spacer layer is not seen due to low isotopic abundance. Fit curve (solid line) accounts for 0, 1 and 2 Si NNs.

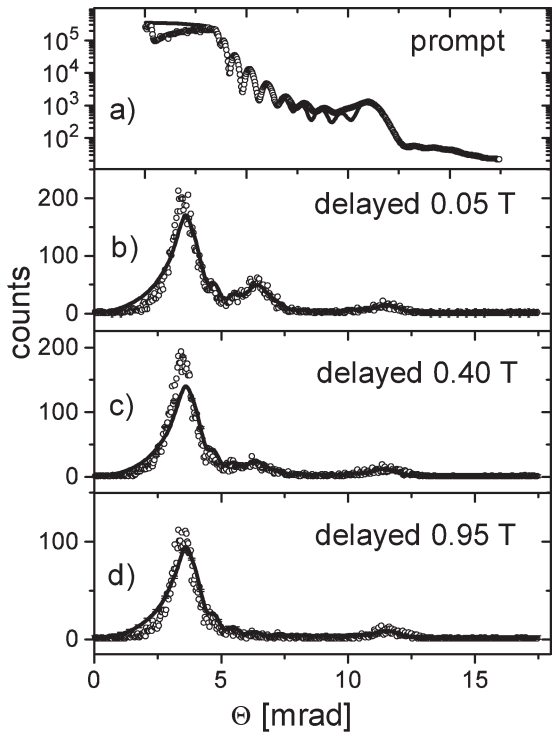

Figure 2. Prompt (a) and time integral (10–300 ns) delayed Θ–2Θ reflectivity scans of Zerodur/ $[^{57}\text{Fe/FeSi}]_{10}$ ML in various external magnetic fields (b)–(d). The fit curves represent the model layer and magnetic structure described in the text.

on an $[^{57}\text{Fe/FeSi}]_{10}$ ML at the ID18 nuclear resonance beamline ($E = 14.413$ keV, $\lambda = 0.0860$ nm) of the ESRF [11] in various external magnetic fields ($0 < B_{\text{ext}} < 0.95$ T) perpendicular to the scattering plane. The prompt X-ray reflectivity curve (figure 2(a)) shows the structural ML Bragg peak at 11.7 mrad and damped Kiessig oscillations corresponding to the total film thickness of 41.2 nm and effective surface roughness of $R_{\text{q}} = 0.89$ nm. The delayed nuclear scattering, in contrast, reveals the apparent magnetic ML period doubling (AF ML Bragg peak at 6.28 mrad) which gradually disappears with increasing transversal magnetic field.

3. Discussion

The majority component in the CEMS spectrum of Zerodur/$[^{57}\text{Fe/FeSi}]_{10}$ is α-Fe. The satellites on the low-field side of the main peaks originate from the ^{57}Fe environments in which 1, 2, ... of the 8 Fe NNs in α-Fe are exchanged for Si. We use a random alloy approach for modeling the interface. Allowing for a constant negative and positive contribution of each Si NN to the Fe hyperfine field and isomer shift, respectively, a quite satisfactory fit is obtained (figure 1) for $\Delta(B_{\text{hf}}) = -2.71(3)$ T

(-8.2%) and Δ(IS) $= 0.046(3)$ mm/s per NN Si, respectively, to be compared with -8.3% and about 0.0 mm/s published by Stearns for a random α-(Fe, Si) alloy [12]. The relative intensities (within the satellite subspectra) and the line widths were kept from those of the main α-Fe component. Consideration is restricted to ^{57}Fe environments with 0, 1 and 2 Si NNs, the measured fraction of which are 0.750, 0.164 and 0.086, respectively. The former is the fraction of the unperturbed environments that stems from the α-Fe *and* from the zero Si NN environments in the interface layer. In the present study, attention is focused on the Fe layer magnetization alignment in the ML and the issue of the minority hyperfine components corresponding to the various possible local surroundings at the real Fe/FeSi interface is not addressed.

Therefore the following layer structure of Zerodur/[^{57}Fe/FeSi]$_{10}$ ML was adapted in accordance with the CEMS fit. The ten Fe layers consist of a "core" sublayer of α-Fe and – on each side – an interface layer of random α-(Fe, Si) alloy of Si concentration c_{Si}. The distribution of the number of Si NNs in the interface layer is assumed binomial. The fraction of zero Si NNs in the random alloy from the measured fractional ratio of the 1 and 2 Si NN environments is $(2/7)(1 - c_{\mathrm{Si}})/c_{\mathrm{Si}} = 0.164/0.086 = 1.907$, from where $c_{\mathrm{Si}} = 0.13$. The corresponding values of the binomial distribution are

$$b(i) = \binom{8}{i} c^i (1-c)^{8-i} = 0.327,\ 0.392 \text{ and } 0.205$$

for $i = 0$, 1 and 2 Si NNs, respectively. Assuming a uniform Mössbauer–Lamb factor, the α-Fe fraction, a of all ^{57}Fe environments satisfies the equation $0.75 = a + (1-a)b(0)$. From the above, $a = 0.629$, which imposes a constraint on the relative thickness

$$\frac{d\,a}{a + (1-a)/(1-c_{\mathrm{Si}})} = 0.596\,d$$

and

$$\frac{(d/2)\,(1/a)}{(1-c_{\mathrm{Si}})/[a + (1-a)/(1-c_{\mathrm{Si}})]} = 0.202\,d,$$

of the ^{57}Fe and α-(^{57}Fe, Si) layers, respectively, d being the total thickness of an ^{57}Fe-containing layer within a ML period. Within the described model each "core" layer of α-^{57}Fe of $0.6\,d$ thickness is, therefore, sandwiched between two layers of α-(^{57}Fe$_{0.87}$Si$_{0.13}$) of $0.2\,d$ thickness (figure 4). In the following we attempt to describe the external field SMR spectra with the above layer model. The hyperfine field in the interface layer – being of conduction electron polarization origin – is assumed to be parallel to the hyperfine field in the adjacent Fe layer. In accordance with the Kerr results, the ferromagnetic (FM) contribution to the magnetization is ignorable – at least within the penetration depth of He–Ne laser light.

The program used for evaluating prompt reflectivity, delayed reflectivity and time spectra of stratified media at grazing incidence uses an optical approach, the principles of which were described elsewhere [8]. Each non-resonant layer is characterized by its layer thickness as well as real and imaginary part of the complex (electronic) index of

refraction. Nuclear resonant layers are further described by $p_{\rm hf}$ hyperfine parameters per non-equivalent nuclear resonant sites in the layer. In general $p_{\rm hf} = 12$, for the zero field gradient case of ^{57}Fe/FeSi, $p_{\rm hf} = 7$ per site (f-factor, isomer shift, spectral width, magnitude and polar angles of magnetic hyperfine field). The total intensity, the background, an additive time shift and an effective interface/surface roughness parameter of the ML were also fitted. The polarization of the incident synchrotron radiation was assumed to be 100%.

The number of parameters of the problem is reduced by introducing linear constraints: (a) of periodicity, (b) of the "core/interface" layer model in accordance with the CEMS spectrum, (c) by fixing the hyperfine field in the interface layer in the same direction as in the adjacent ^{57}Fe layers, and (d) of the same magnitude as determined from the CEMS spectrum, (e) directly relating the electronic contribution to the index of refraction for Fe and (α- and B2-)FeSi layers to the elemental photo-absorption coefficients and the chemical composition. The layer thicknesses and directions of the ^{57}Fe hyperfine fields were fitted. The fit to the prompt X-ray reflectivity curve (solid line in figure 2(a)) corresponds to the model with the above constraints. The time spectra at various grazing angles and the time integral delayed Θ–2Θ scans were successively calculated using the same parameter file allowing for self-consistency.

In a weak external field of 50 mT the Fe moments, consequently the hyperfine fields in this periodic ML of even number of FM layers, are expected to align perpendicular to the external field and in pairs antiparallel to each other in an AF-aligned ML. The fit curves (thin solid lines in figure 3) corresponding to this $\phi(\mathrm{AF}_1) = 0^\circ$,

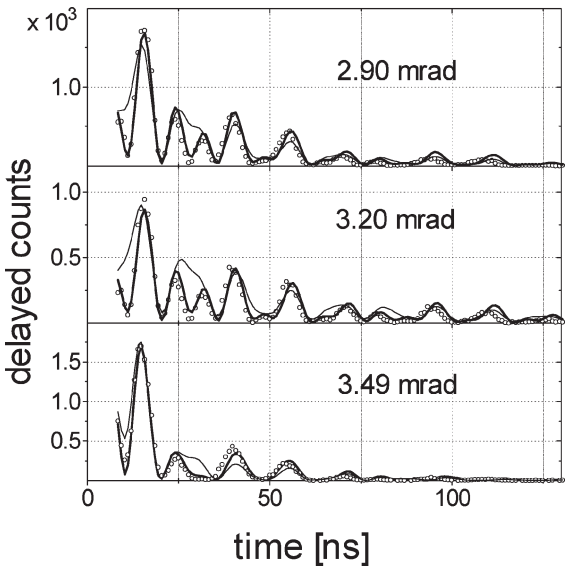

Figure 3. SMR time spectra of Zerodur/[^{57}Fe/FeSi]$_{10}$ ML in an external field of 0.05 T perpendicular to the scattering plane at different angles of grazing incidence. Thin solid fit curve for an "all AF" (0–180°) sublayer magnetization alignment relative to scattering plane, thick solid fit curve for the 8AF–2FM model.

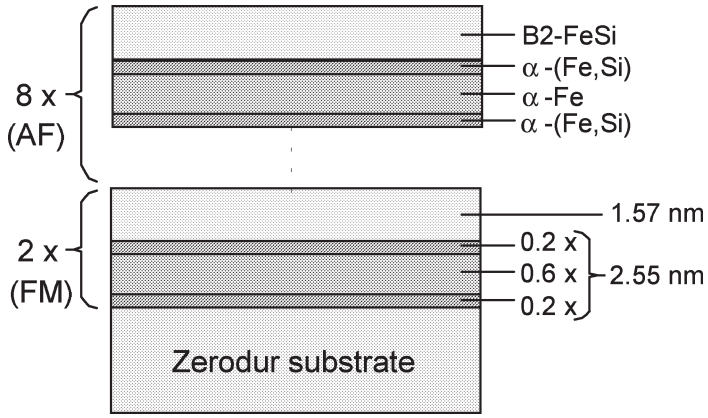

Figure 4. Model layer structure of Zerodur/[^{57}Fe/FeSi]$_{10}$ in accordance with CEMS and prompt reflectivity results as used in the 8AF–2FM model in fitting delayed reflectivity data.

$\phi(\mathrm{AF}_2) = 180^\circ$ alignment, however, show a surprisingly poor agreement with the experimental data. Allowing for the variation of the angle of the layer magnetizations, a much better agreement is achieved at $\phi(\mathrm{AF}_1) = 62(\pm 2)$ and $\phi(\mathrm{AF}_2) = 118(\pm 2)$ or, equivalently, $\phi(\mathrm{AF}_2) = 242(\pm 2)$ degrees (thick solid lines in figure 3), $\phi(\mathrm{AF}_1)$ and $\phi(\mathrm{AF}_2)$ being the respective azimuth angles of the AF sublayer magnetizations of the ML with respect to **k**, the wave vector of the incident radiation. For symmetry reasons the 118° and the 242° alignments cannot be distinguished solely by SMR. The former, however, corresponds to an 88% FM component of the total magnetization, in disagreement with our Kerr result and the latter ($\phi(\mathrm{AF}_2) = 242^\circ$) alignment – although not contradicting to the Kerr effect of limited penetration – corresponds to a massive 62 degrees misalignment of the observed AF magnetizations sampled by Kerr effect and CEMS with respect to the external field of 50 mT. Provided that all ten Fe layers are AF aligned this is a contradiction.

In a recent work Kohlhepp et al. [6] studied Fe/FeSi MLs grown on glass substrate by vibrating sample magnetometry (VSM) and Kerr effect. Both top and bottom Fe layers were both Kerr-tested, the latter through the glass substrate. The ML was found majority FM on bottom and majority AF on top. They allowed for a positional variation of the bilinear coupling coefficient, J_1, and showed this variation to result in magnetization curves resembling a strong biquadratic coupling.

The mentioned contradiction can be lifted by applying a "top-AF–bottom-FM" scheme. A detailed analysis of SMR time spectra and delayed reflectivity curves as function of external field will be published elsewhere [13] only results are summarized here. It is shown that, for some presently unknown microscopic reason, the coupling between the top layers in [Fe/FeSi]$_{10}$ ML is AF ($J_1^{(\mathrm{top})} \ll 0$), and between bottom layers is FM ($J_1^{(\mathrm{bottom})} \gg 0$). The AF components, in a weak external field will align at an angle between parallel and perpendicular to the field according to the coupling strength between AF and FM regions. Increasing the external field, an increasing FM

moment is induced in the AF-coupled top block and a more pronounced canting is observed on the surface. In a field of 0.95 T, all layers line up with the field within the experimental error. The detailed analysis of Kerr and SMR results is in favor of an "8AF–2FM" scheme [13] (8 AF coupled layer on top, two FM layers on bottom) as compared to the widely discussed phenomenological bilinear + biquadratic model, which provides no ground for an asymmetric alignment of the top layers with respect to the external field. However, in the 8AF–2FM model, the misalignment is due to the coupling between the two blocks.

In summary, by analyzing CEMS spectra, Kerr loops, SMR time spectra and SMR time integral delayed reflectivity curves of a Zerodur/[^{57}Fe(2.55 nm)/^{nat}FeSi (1.57 nm)]$_{10}$ we find evidence of two magnetically distinct regions in the ML perpendicular to the film surface, a minority FM and a majority antiferromagnetically aligned block, close to the substrate and close to the ML surface, respectively, rather than the widely accepted uniform combined bilinear + biquadratic coupling between the Fe layers.

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