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Abstract. Mössbauer reflectivity experiment with polarization selection are reported. The use of the LiF polarization analyzer allows us to get the $\pi \rightarrow \sigma^*$ peak on the reflectivity curve at the critical angle and Mössbauer $\pi \rightarrow \sigma^*$ reflectivity spectra of reasonable quality at the critical angle and also at the Bragg angles of superlattice. The impressive difference of the reflectivity spectra measured near the critical angle with and without polarizations analysis is observed for $[\text{57Fe}_{10}/\text{V}_{10}]_{20}$ multilayer characterizing by the ferromagnetic interlayer coupling. The combined fit of the whole set of spectra measured at different angles reveals the existence of antiferromagnetic Fe oxide phases in the top three bilayers. The experiment demonstrates benefits of the Mössbauer reflectivity with polarization analysis in ultrathin surface layer investigations.

1. Introduction

Mössbauer reflectivity (or nuclear resonant reflectivity NRR) is the very effective method for magnetization depth profile investigations (see e.g. [1-5]). As distinct from X-ray and neutron reflectivity, Mössbauer experiments are supplemented with spectrum measurements at different grazing angles revealing the details of the hyperfine field depth-profiles.

Specific polarization dependence of the nuclear resonant scattering on the hyperfine field orientation predetermines the importance of the polarization analysis of the reflected radiation for magnetic structure determination. It can be referred to the polarized neutron reflectivity (PNR), in which the four reflectivity curve measurements (neutron spins “up” $\rightarrow$ “up”, “up” $\rightarrow$ “down”, “down” $\rightarrow$ “up” and “down” $\rightarrow$ “down”) are standard for the magnetic depth profiling [6].

Our first experimental attempt on polarization selection in Mössbauer reflectivity experiments [7] revealed that Si polarization analyzers (double (840) reflections), previously used in the nuclear resonant forward scattering [8,9], are not suitable in reflectivity geometry. They have too narrow angular acceptance (~ 2°) while scattering from multilayers is typically accompanied with a rather...
broad angular divergence (60 – 200°). In spite of the huge loss of the reflected intensity during polarization analysis the signal of rotated π→σ’ polarization in reflectivity from magnetized [Fe/Cr] multilayer was detected and specific features of π→σ’ reflectivity curve were observed [7]. The experiment demonstrated advantages of polarization selection in Mössbauer reflectometry: simpler shape of Mössbauer spectrum, suppression of electronic scattering, enhanced depth selectivity, etc. However, the great loss of the intensity with Si analyzer made the data acquisition with polarization selection too time-consuming.

In this work we report the new results on the Mössbauer reflectivity with polarization selection performed with much suitable polarization analyzers (LiF crystals).

2. Experiment
The experiment was performed at the ID18 beamline [10] of the European synchrotron (ESRF) making use of the purely π-polarized radiation from Synchrotron Mössbauer Source (SMS) [11].

We tested several analyzer crystals for 14.4 keV resonant radiation of the Mössbauer transition in 57Fe nuclei (table 1). In addition, we decided to use not double but just single reflection from analyzer in order to diminish the intensity loss, so the detector had been moved from the forward direction (see figure 1).

![Figure 1. Experimental set-up for selection of the rotated π→σ’ polarization in reflected signal. HHLM is the high-heat-load monochromator; CRL are the compound refractive lenses; HRM is the high-resolution monochromator; 57FeBO3 is the nuclear monochromator (SMS).](image)

The best choice was LiF crystal with (622) reflection, which has angular acceptance ~ 100° and reasonable reflection coefficient. Moreover, the Bragg angle of (622) reflection for 14.4 keV radiation is much closer to 45° than for (840) reflection of Si crystal, and it supplies us with better π→π’ suppression.

Table 1. Considered polarization analyzers for the 14.4 keV radiation and their parameters: θBr - Bragg angle, |b| - asymmetry coefficient, Δθ - measured angular width of reflection (FMHW), R - measured reflection coefficient (at maximum). LiF (1) was provided by A.Rogalev (ID12, ESRF), LiF (2) was fabricated in the Institute of Crystallography RAS (Moscow).

| Analyser  | (h k l) | θBr (°) | cos²θBr | 1/|b| | Δθ (°) | R % |
|-----------|--------|---------|----------|---|------|----|
| Si_{ch-c} | (840)  | 45.10   | 1.2×10⁵  | 29 | 2.3  | 40 |
| Ge       | (664)  | 45.51   | 3.1×10⁴  | 8.1 | 4.0  | 15 |
| LiF (1)  | (622)  | 44.98   | 3.9×10⁷  | 2.8 | 100  | 5  |
| LiF (2)  | (622)  | 44.98   | 3.9×10⁷  | 2.9 | 90   | 7  |
| Graphite | (0 0 10) | 39.87   | 0.032    | 1  | 800  | 1.4 |
| Graphite | (0 0 12) | 50.29   | 0.034    | 1  | 800  | 0.7 |

We investigated several magnetic multilayers. The samples were placed into cryomagnet and cooled down to 4 K to get magnetic ordering for the samples with ultrathin Fe layers. For the samples
with ferromagnetic interlayer coupling the external field (up to 5 T) was applied along the beam in order to enhance the dichroic signal.

Here we present the results for $[{^{57}}\text{Fe}_{10}/\text{V}_{10}]_{20}$ multilayer, epitaxially grown in Uppsala University [12, 13], which have shown an impressive difference in Mössbauer reflectivity spectra measured with and without polarization selection.

3. Results and discussion

The measured X-ray reflectivity curve for $[{^{57}}\text{Fe}_{10}/\text{V}_{10}]_{20}$ sample confirms its periodicity (figure 2) and the fit gives the superlattice period $D_{\text{chem}} = 3.21$ nm.

Mössbauer reflectivity was measured as an integral over Mössbauer spectrum at each grazing angle, and no additional “magnetic” Bragg maxima were observed in this curve (figure 2), so the magnetic period of the multilayer does not differ from the chemical period. Mössbauer reflectivity curve with $\pi \rightarrow \sigma'$ polarization selection was measured only near critical angle and the peak at the critical angle (instead of common plateau in the total reflection region) was clearly seen (figure 2) in agreement with the theoretical prediction [14].

Mössbauer reflectivity spectra were measured sequentially in the increasing external magnetic field from 0 T up to 5 T applied along the beam and it was detected that their shape did not change after 1 T. The measured spectra for 1 T at the critical angle and at the first order Bragg angle without polarization analysis and with $\pi \rightarrow \sigma'$ polarization selection are shown in figure 3.

The impressive difference between Mössbauer reflectivity spectra measured near the critical angle without and with polarization selection takes place (compare spectra in figure 3 (a) and (b)). It is not only the shape difference (peaks or dips), these spectra differ by contributions. Notice that it would be very complicated to identify the spectrum contributions without the polarization analysis.

For the $\pi$-polarized incident radiation the $\pi \rightarrow \sigma'$ reflectivity spectrum should contain only the 1st, 3rd, 4th and 6th resonance lines of a magnetic Mössbauer sextet in the case of the planar orientation of hyperfine fields $B_{hf}$ [1,7]. If magnetization of the sample is saturated along the beam (as we suppose at 1 T) the spectra without polarization analysis should contain the same lines only. A fit of the data gives the $B_{hf}$ field distribution for all spectra shown in figure 3 (b), (c), (d) (excluding the first one in figure 3 (a)) with maximal splitting corresponding to $B_{hf} \approx 34$ T which we approximate by three sextets ($B_{hf} = 34.58, 31.03$ and $24.27$ T), shown in figure 3 (e). These values of $B_{hf}$ correspond to $\alpha$-Fe at 4 K.
(34.58 T), the existence of one V atom in surroundings of $^{57}$Fe nuclei (31.03 T) and the deficiency of half Fe atoms in the first coordination sphere (24.27 T) typical for interface regions respectively.

An additional sextet corresponding $B_{hf} \approx 46$ T is clearly seen in figure 3a. The fit gives several multiplets (figure 3 (f)) not aligned by the external field (because the ratio of their sextet line intensities is 3:2:1:2:3). Therefore, they do not contribute to the $\pi \rightarrow \sigma'$ reflectivity spectrum. It can be suggested that these contributions correspond to the antiferromagnetic or superparamagnetic oxidized iron phases. Antiferromagnetic order in e.g. Fe$_3$O$_4$ phase could not be destroyed by external field 1 T and the scattered radiation by $^{57}$Fe nuclei in this phase has no component with “rotated” polarization. The obtained depth distribution for these additional phases restricts only by the three top bilayers and they do not noticeably contribute to the reflectivity spectra at the Bragg angle (figure 3 (c), (d)). These spectra are formed by the whole multilayer which is mostly ferromagnetically aligned, the contributions from the top bilayers are negligible.

The spectra measured at the Bragg angle without and with $\pi \rightarrow \sigma'$ polarization selection do not differ essentially, because in both cases the sextets are presented by the 1st, 3rd, 4th and 6th lines. The most noticeable difference refers to the “tails”. The spectrum without polarizations analysis has asymmetric background caused by the small addition of the electronic scattering. The $\pi \rightarrow \sigma'$ reflectivity spectrum practically has zero background. Notice that signal to noise ratio is much better than with using Si analyzer in [7]. The asymmetry of lines in the $\pi \rightarrow \sigma'$ spectrum is explained by the refraction effect (for details see [7, 15]).

4. Conclusion
The success with polarization selection of the resonantly reflected radiation has been achieved with new LiF polarization analyzers. The good quality Mössbauer $\pi \rightarrow \sigma'$ reflectivity angular curve and $\pi \rightarrow \sigma'$ spectra have been measured. The results for $[^{57}$Fe$_{10}$/V$_{10}]_{20}$ sample show the possibility to select ferro- and antiferromagnetic phases of Fe and Fe oxides in the surface layers. Our Mössbauer reflectivity experiment with polarization selection demonstrates the strength of the improved method for ultrathin surface layer investigations.
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