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Probing magnetic structures by neutron reflectometry: Off-specular scattering from interfaces and domains in FeCoV/TiZr multilayers

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Abstract

Polarized neutron reflectometry (PNR) is commonly used to characterize the depth-dependent nuclear density and magnetization profiles of thin films and multilayers. While specular reflectivity is connected with characteristics perpendicular to the sample surface, in-plane features cause off-specular scattering. In this work we present time-of-flight PNR results on (FeCoV/TiZr) multilayers with applied fields parallel to the in-plane easy axis of magnetization. Our measurements at low applied fields clearly show diffuse scattering. We attribute this scattering to randomly oriented magnetic domains, with no coupling between the magnetic FeCoV layers. With increasing applied field the domain scattering decreases. At the same time, off-specular scattering with a well-defined relation between the wavelength and the scattering angle increases with increasing applied field. This type of scattering can be attributed to correlated interfaces. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Magnetic thin films and multilayers are commonly used in devices for magnetic recording and neutron beam-line optics. To develop and optimize such devices, there is an increasing demand to characterize the structural and magnetic properties of these systems. The depth-dependent characteristics are often obtained by polarized neutron reflectometry (PNR) because of its unique ability to obtain simultaneously the nuclear density and the magnetization profiles. Recent advances in beam-line optics, neutron flux and neutron detection equipment have made it possible to detect off-specular scattered neutrons that are linked to perturbations of densities in the plane of the film.

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Magnetic off-specular neutron scattering is therefore useful for receiving information about changes in the magnetic domain structure and about correlations in interfacial roughness in magnetic multilayers [1,2].

We have studied the behavior of $60\times(10$ nm $Fe_{31}Co_{68}V_1/x\,Ti_{75}Zr_{25})$, magnetron sputtered on a 5 mm thick glass substrate, with $x = 10$ nm and $x = 30$ nm. (FeCoV/TiZr) multilayers are good candidates for neutron polarization devices because of the contrast in density for spin-up and spin-down neutrons [3]. Although actual polarizing devices are designed with varying layer thickness throughout the multilayer, constant layer thickness simplify the interpretation of the PNR results. In this work, we show off-specular PNR measurements on (FeCoV/TiZr) multilayers and discuss the observed off-specular scattering.

2. Results and discussion

Previously, we have reported on specular PNR measurements on $60\times$(FeCoV/TiZr) multilayers [4]. It was shown that the multilayers exhibit clear Bragg reflections resulting from the bilayer repetition. The spin-dependent magnetic contrast between FeCoV and TiZr resulted in high intensity Bragg reflections for one spin state and vanishing Bragg reflections for the other. In the current study we used the CRISP reflectometer at ISIS, which has a neutron wavelength range of $\lambda = 0.1 - 0.6$ nm for PNR experiments. The combination of a high-brilliance pulsed neutron beam and the presence of a one-dimensional position sensitive detector rapidly yields a contour map of scattering angle versus wavelength at a fixed incident beam angle. In this paper, we define the horizon of the sample at detector angle $\theta = 0$. For the PNR measurements on $60\times(30$ nm FeCoV/ 10 nm TiZr) we used an incident angle of $\theta_{in} = -4$ mrad. At this incident angle, the available wavelength region includes the total reflection region and the first two Bragg reflections. All spectra presented in this work have been normalized to intensities measured by a monitor in the incident beam.

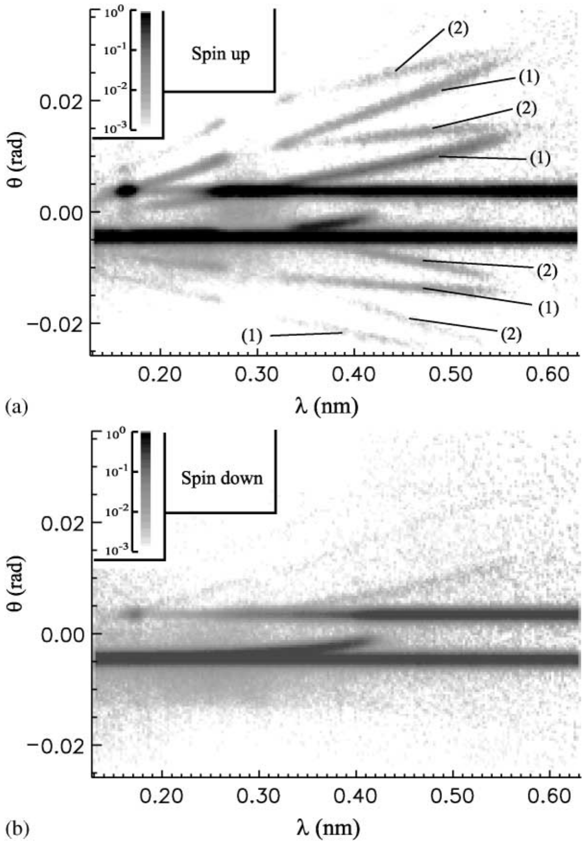

Fig. 1. Contour map of spin-up (a) and spin-down (b) neutron scattering from a (FeCoV/TiZr) multilayer, with an applied field of 5.1 kOe along the easy axis.

We performed measurements with applied field in-plane along the easy axis, starting with the remnant magnetization after saturating the magnetization in a negative field and then taking data up to fields of 5 kOe. From magnetization measurements we expect the multilayer to be magnetically saturated above 500 Oe. Fig. 1a shows the contour map for spin-up neutrons at an applied field of 5.1 kOe. The specularly reflected beam is recognizable at $\theta_{out} = 4$ mrad. The transmitted beam (together with that part of the incident beam that does not illuminate the sample) is visible at detector angle $\theta_{out} = \theta_{in} = -4$ mrad. Off-specular scattering is also visible in the form of lines that show a clear relation between θ_{out} and λ. Such lines are the result of scattering from *correlated* rough interfaces, and are characterized by

$$k_{\perp,\mathrm{out}} - k_{\perp,\mathrm{in}} = n\,\frac{2\pi}{d}, \tag{1}$$

where $k_{\perp,\mathrm{in(out)}} = (2\pi/\lambda)\sin\theta_{\mathrm{in(out)}}$, n is the order of the Bragg reflection and d is the bilayer thickness.

For a reflectometer with constant incident angle and variable wavelength, we can write for small angles $\theta_{\mathrm{out}} \simeq \theta_{\mathrm{in}} + n\lambda/d$. With $\theta_{\mathrm{in}} = \mathrm{constant} = -4$ mrad, this translates into the straight lines marked (1), converging at the incident beam position $\theta_{\mathrm{out}} = \theta_{\mathrm{in}}$ for $\lambda \to 0$. In addition to this interfacial scattering connected to the *incident beam*, also visible is a set of off-specular straight lines marked (2), that converge at $\theta_{\mathrm{out}} = 4$ mrad for $\lambda \to 0$. We attribute these lines to off-specular interfacial scattering caused by the *specularly reflected beam.* One special feature we would like to note is that Fig. 1a also shows off-specular scattering *below* the sample horizon. These lines have a negative slope and therefore (see Eq. (1)) have negative values of n.

We would like to stress that Eq. (1) is only valid for sufficiently high values of $k_{\perp,\mathrm{in(out)}}$, when refraction inside the layers is negligible. Corrections for refraction can be taken into account, but ultimately fail at high wavelengths where the wavevector of the incident and/or scattered beam can be imaginary inside a layer. For such cases, a distorted wave born approximation (DWBA) is more successful in predicting the off-specular scattering patterns [5,6].

Fig. 1b shows the contour map for spin-down neutrons, again at an applied field of 5.1 kOe. The off-specular interfacial scattering is much less intense than in the case of spin-up neutrons. However, even if the interfacial roughness would be fully non-magnetic in origin, the off-specular scattering would still be spin-dependent due to the spin-dependent neutron density distribution throughout the multilayer, as predicted by DWBA calculations. The presence of magnetic roughness may be studied by a rigorous spin-analysis of the off-specular scattered neutrons.

The magnetization measurements (see inset Fig. 3b) indicate a coercive field $H_C \approx 120$ Oe. Fig. 2 shows the contour map for spin-up neutrons for an applied field of 120 Oe. Besides the direct beam and the specularly reflected beam, neutrons

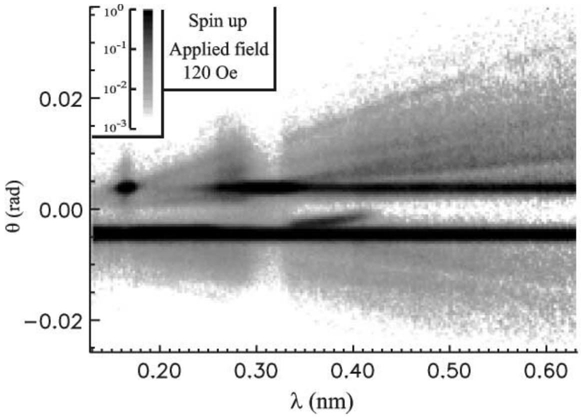

Fig. 2. Contour map of spin-up neutron scattering from a (FeCoV/TiZr) multilayer, with an applied field of 120 Oe along the easy axis.

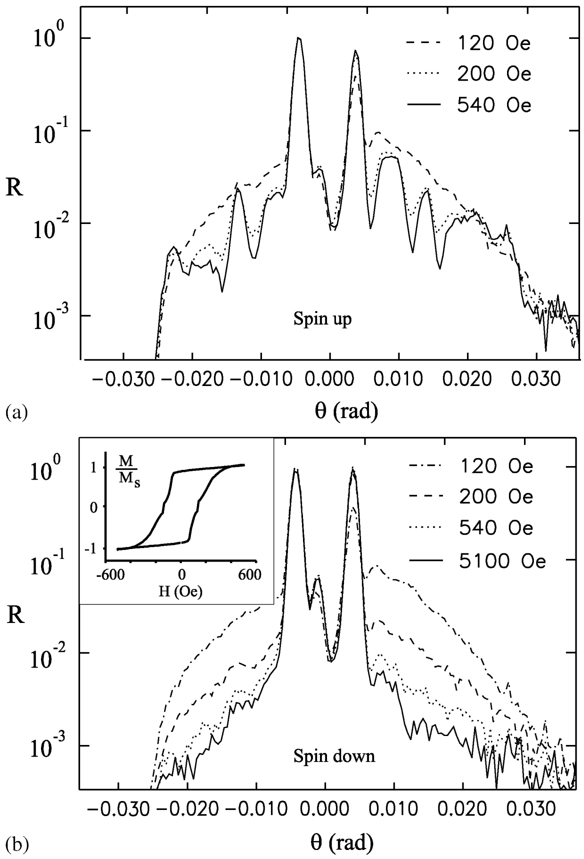

Fig. 3. Field dependence of spin-up (a) and spin-down (b) neutron scattering. All intensities are averaged for 0.4 nm ≪ 0.5 nm. The inset shows the easy axis hysteresis loop.

are scattered over a wide angular range *at all wavelengths*. We attribute this to small-angle scattering from *uncorrelated* magnetic domains, with no coupling between magnetic layers throughout the multilayer stack. Coupled magnetic domains inside multilayers are reported to show increased off-specular scatting close to the specular Bragg conditions where $\lambda = (2d\theta_{\mathrm{Bragg}})/n$ [7]. The decrease in off-specular intensity observed at the first order Bragg reflection ($\lambda \approx 0.3$ nm) is explained by the fact that most of the reflected intensity is coming from the top bilayers at that wavelength. Model calculations for this system show that for six bilayers the first-order specular Bragg reflection is already close to unity. Therefore, the neutron beam does not penetrate deeply into the multilayer and encounters less domains and interfaces, causing less off-specular scattering. This decrease in off-specular scattering is also observable in Fig. 1a as gaps in the lines of off-specular scattering. For spin-down neutrons the spectrum looks almost identical to that for spin-up neutrons, confirming that the average magnetization is close to zero and indeed $H \approx H_C$.

The contour map of θ versus λ can be transformed into a contour map of Q_X versus Q_Z, with Q_X and Q_Z the momentum transfer, respectively, parallel and perpendicular to the film plane. At constant Q_Z, the width of the scattering profile as a function of Q_X is inversely retated to the in-plane correlation length [8,9]. This correlation length, estimated at 5–10 μm, is connected to the average magnetic domain size and is confirmed by neutron depolarization studies on similar systems [10].

From the specular reflectivity at $\theta_{\mathrm{out}} = 4$ mrad we obtain that total reflection occurs from the substrate for $\lambda > 0.4$ nm. Similarly, total reflection occurs from the average multilayer potential for $\lambda > 0.5$ nm. Fig. 3a shows the spin-up intensity as a function of θ_{out}, averaged for $0.4\ \mathrm{nm} < \lambda < 0.5\ \mathrm{nm}$. In this region, the neutrons are able to penetrate the entire multilayer, before being totally reflected from the substrate [4]. The direct and specular reflected beam are easily recognizable at $\theta_{\mathrm{out}} = -4$ mrad and $\theta_{\mathrm{out}} = 4$ mrad. At low applied fields the presence of randomly oriented magnetic domains shows as a broad peak in the small-angle scattering. At higher fields, the domains grow and this results in a decrease of this kind of off-specular domain scattering. Finally, only interfacial scattering is visible at the highest fields.

We have already observed (Fig. 1b) that almost no interfacial scattering occurs at high applied field for spin-down neutrons. Fig. 3b indeed only shows a continuous decrease in domain scattering with increasing applied field. It is interesting to note that the multilayer is clearly not saturated at 500 Oe: the magnetization in the layers is still not completely aligned and causes off-specular scattering.

3. Conclusion

We have clearly observed off-specular scattering from interfaces and magnetic domains in periodic $60 \times$(FeCoV/TiZr) multilayers. At high fields we observe spin-dependent interfacial scattering, caused by correlated interface roughness. At lower fields this interfacial scattering is overshadowed by spin-independent scattering from uncorrelated domains. The domain scattering is shown to be very sensitive to small deviations from the total alignment of the domains.

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