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Physica B 283 (2000) 194–198

PHYSICA B

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Magnetic off-specular neutron scattering from Fe/Cr multilayers

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Abstract

The magnetic structure of a [Cr(9 Å)/^{57}Fe (68 Å)] $\times$ 12 multilayer on sapphire substrate has been studied with polarized neutron reflectometry. The intensity distribution was measured over a broad range of incident and outgoing angles with spin analysis. The specular and off-specular intensities of the first and second-order Bragg-peaks (determined by the bi-layer thickness) and those at the half-order positions (due to an antiferromagnetic coupling) were measured. Off-specular scattering arranged into sheets running across the antiferromagnetic half-order positions is spread over an appreciable range perpendicular to the specular line. It consists of almost totally spin-flip scattering. Nearly no experimental evidence of the peak at the position of antiferromagnetic superstructure is found in the spin-flip specular channel. The intensity of the diffuse scattering scales with the intensity of the Bragg peaks, which is a function of the external magnetic field. These findings result in a picture of antiferromagnetic domains, rather than in the model of homogeneous magnetized neighbouring layers with a magnetization coupling angle. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Off-specular scattering; Magnetic multilayer; Interface roughness; DWBA

There has been great interest in magnetic multilayers which, being new kinds of artificial magnetic materials, display a wide array of exciting properties including oscillating interlayer exchange coupling [1], giant magnetoresistance [2], and non-collinear magnetic ordering [3]. Also polarized neutron reflectometry (PNR) was often applied to Fe/Cr multilayers and gave interesting results [4] in particular with respect to the canted state of the magnetization in successive Fe layers. We will add further insight due the spin analysis of specular and off-specular scattering showing the spin-flip nature of the off-specular scattering which influences the interpretation of the specular scattering. Both effects strongly suggest the presence of magnetic domains as primary effect.

The (0 0 1) superlattice [Cr(9 Å)/^{57}Fe (68 Å)] $\times$ 12 was grown with molecular beam epitaxy. A Al_2O_3 substrate was annealed at 700°C and covered with a 65 Å Cr buffer at 300°C. The multilayer was grown at the substrate temperature 180°C starting with an Fe layer. The sample was characterized in situ with reflection high energy electron diffraction (RHEED) and ex situ with X-ray Diffraction. The in-plane magnetization measurements were carried out at room temperature with a vibrating sample magnetometer (VSM) and revealed extremely strong in-plane anisotropy in the sample. The magnetization curves measured at different magnetic field orientations relative to the hard axis are shown in Fig. 1. The hard magnetic axis is attributed as usual to the measurement showing the widest low-field hysteresis-loop (shown

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PII: S0921-4526(99)01938-9

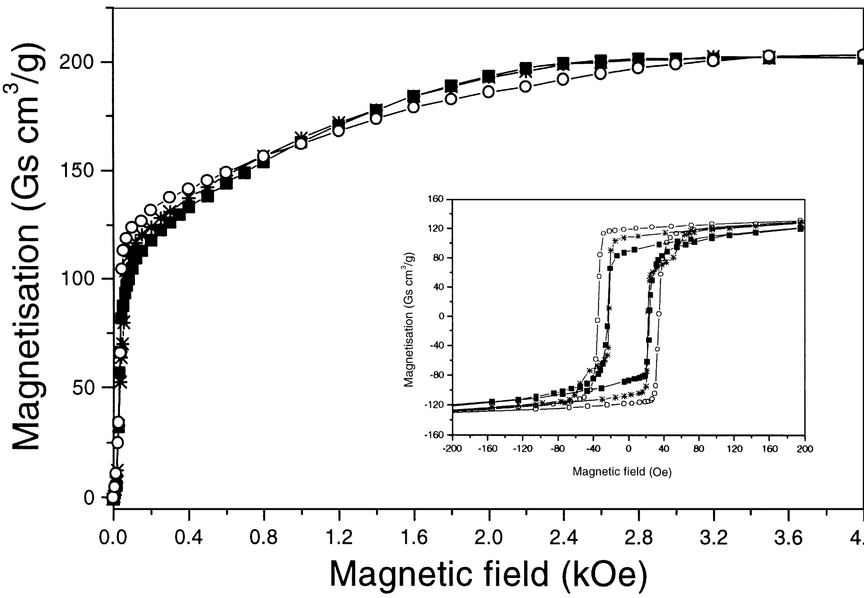

Fig. 1. Magnetization curves for magnetic field applied at an angle of 0° (circles), 22° (crosses) and 45° (squares) relative to the 'hard' magnetization axis. The low-field hysteresis loops are shown in the inset.

in the inset of Fig. 1). The low-field hysteresis behavior is typical for (0 0 1) Fe/Cr systems and it can be described in terms of fourfold in-plane anisotropy [5,6]. For the high-field magnetic behavior, we note some unusual features. It is very surprising that there is a significant increase in the anisotropy strength in contrast to the expected one for (0 0 1) Fe films [5]. The effective anisotropy field $2K/M_s$ is estimated to be about 3.5 kOe (see Fig. 1) compared with 0.55 kOe for bulk Fe. In addition, the hard axis magnetization curve (circles) lies under the soft axis one (squares) in large fields. Such kind of transformation from hard to soft axis magnetization could be described in terms of fourfold in-plane anisotropy. The multilayer system behaves as if there were two different mechanisms responsible for the anisotropy, one being more determining at small magnetic fields and another at higher fields. The interplay between the different mechanisms is likely to cause a complicated magnetic domain structure throughout the multilayer.

The PNR experiments were performed on the ADAM spectrometer at ILL [7]. The magnetic field H was applied parallel to the sample surface. The reflected intensity was recorded on a multidetector. The spin analysis of the scattered intensity was performed. The experiments were made for $H = 0.428$, 1.59, and 5.48 kG increasing the field from negative saturation field. The Figs. 2a and b and Figs. 3a and b show the scattering from the Fe/Cr multilayer for $H = 0.428$ and 5.48 kG, respectively, in the coordinate system of α_f, α_i, α_i being the angle between the incident neutron beam with the sample surface and α_f the angle between the outgoing neutrons and the sample surface. The scattered intensity is represented by a grey log-scale shown in the bar on the right-hand side of the figure. For Figs. 2a and 3a the orientation of the incident neutron spin and magnetic field is parallel (+) and for Figs. 2b and 3b antiparallel (−). For $\alpha_i = \alpha_f$ the reflectivity curve is clearly visible in all figures with its intensity oscillations. The intensity at the total reflection region and from the straight-through beam are not shown in the figures in order to make more features visible in the off-specular regions.

In Figs. 3a and b the position of the first order Bragg-peak at $\alpha_i = \alpha_f = 0.029$ rad and of the second-order peak at 0.058 rad is clearly seen by the high intensity. In addition, the fringes arising from the total film thickness emerge from the critical angle along $\alpha_i = \alpha_f$. Nearly no off-specular scattering is visible around these Bragg-peaks manifesting the extremely good quality of the sample. This picture agrees with the sample being in a saturation field with ferromagnetic magnetisation in the Fe-layers. The light horizontal lines crossing the pictures are background effects.

In Fig. 2 for the lower field of $H = 0.428$ kG additional intensity is observed going through the two half-order positions at $\alpha = 0.0145$, 0.0435 rad in comparison with the saturated state in the higher field. This additional intensity is off-specular scattering distributed along lines perpendicular to the reflectivity curve forming a superstructure Bragg-sheet. However in contrast to the usual case when the Bragg sheet crosses the specular line at the Bragg peak position no appreciable enhancement of the intensity at the superstructure position was observed. The nature of this phenomena will become clear after the discussion of the polarization analysis. The enhanced

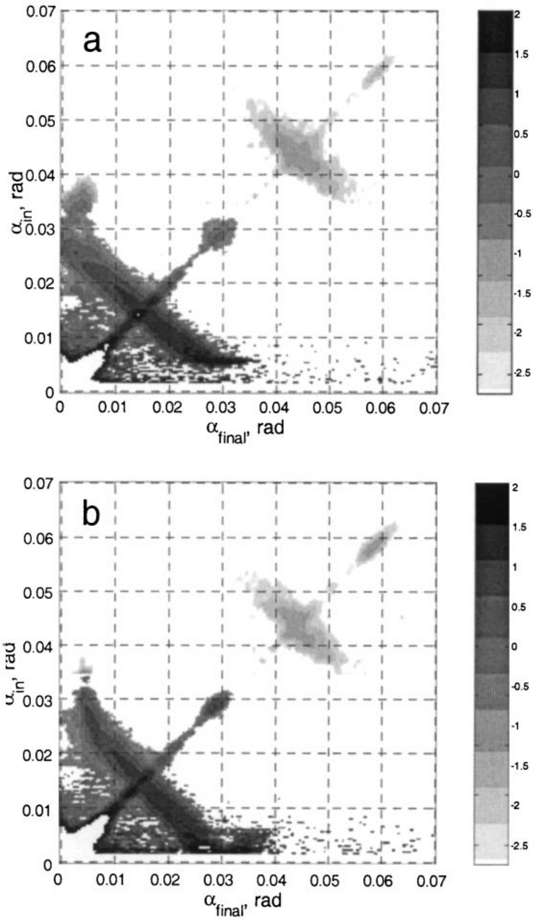

Fig. 2. Intensity map of specular and off-specular scattered neutrons from the Fe/Cr multilayer at $H = 0.428$ kG as a function of α_i and α_f the incident and outgoing scattering angles, respectively; neutron wavelength $\lambda = 4.41$ Å (a) for $R^+ = R^{++} + R^{+-}$ and (b) for $R^- = R^{--} + R^{-+}$.

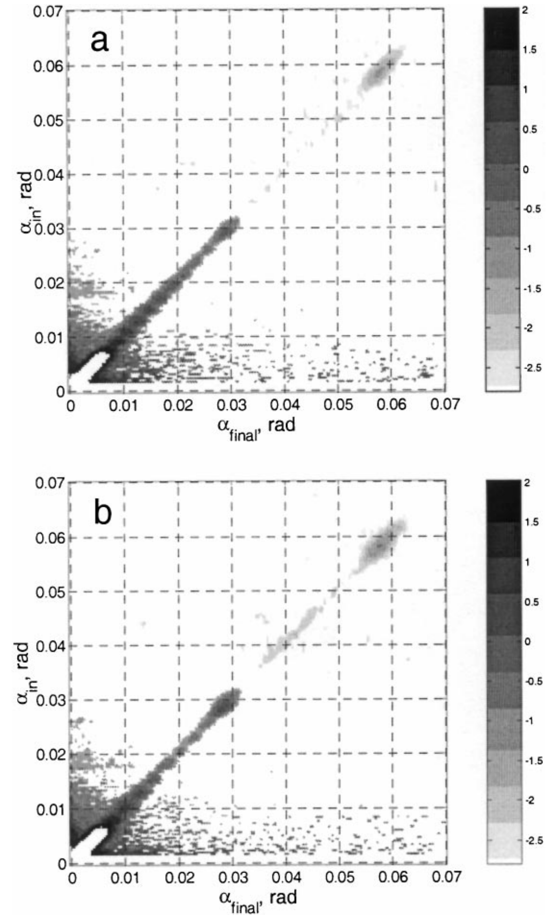

Fig. 3. Intensity map as in Fig. 2 but for the field $H = 5.48$ kG. (a) for $R^+ = R^{++} + R^{+-}$ and (b) for $R^- = R^{--} + R^{-+}$.

tiny intensity spots which can be seen within the crossing of the off-specular sheets and the specular line originate from the total thickness fringes. At the limits near the border lines of the figure the line shape of this off-specular scattering change is very visible in the figure for the off-specular scattering passing through the first half-order position, that means the line width and angular intensity distribution is influenced by the corresponding critical angle. We want to point to this effect in context with the usually performed transverse scans to get the Q_x dependence of the off-specular scattering. These transverse scans are probably even more influenced by this feature than expected [8] in particular due to instrumental resolution effects. A complete picture of the off-specular scattering was calculated in Ref. [9] for surface diffraction and we observe it here for the first time in magnetic off-specular scattering.

Figs. 4a and b show the scattered intensity as in Figs. 2a and b, but for the case when the analyser is placed between the sample and the detector centred on the position of the reflected beam. The analyser operates simultaneously in transmission and in reflection, so that one spin state is transmitted and the opposite one is reflected. The footprint of the analyser, marked with the dashed lines in Fig. 4, covers only a certain angular band of neutrons scattered from the sample. The specular line in Fig. 4b transmitted through the analyser corresponds to R^{--} and does not manifest any appreciable peaks at the half-order positions. Moreover the intensity in the off-specular half-order scattering is not transmitted by the analyser and the spin-flipped superstructure Bragg-sheet is terminated in the range of the analyser footprint shadow.

Fig. 4a, complementary to Fig. 4b shows along the specular ridge the intensity transmitted through the analyser, i.e. the spin-flip reflectivity R^{+-}. This spin-flip intensity is quite low along all the specular ridge and is

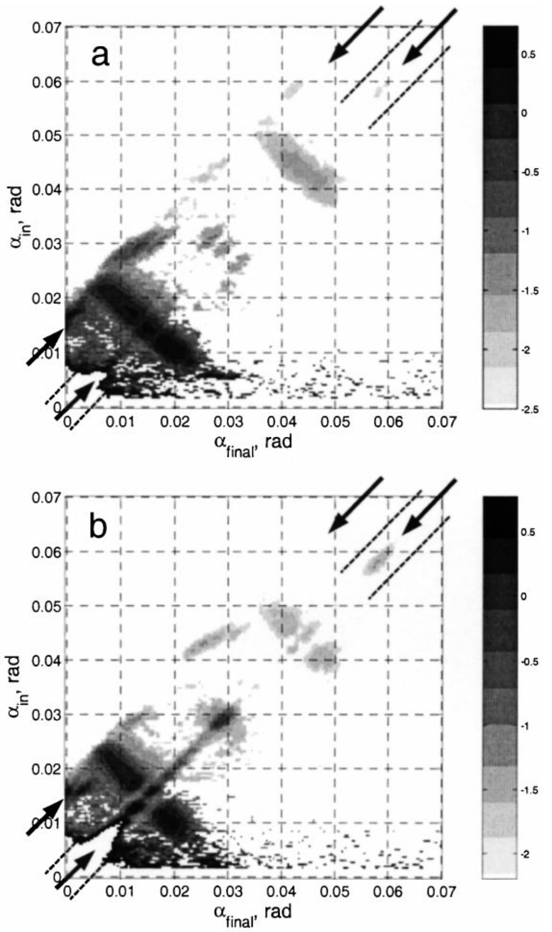

Fig. 4. Intensity map as in Fig. 2 but with the analyser. The arrows indicate the specular line in one spin state and the specular line in the opposite spin state; (a) in transmission R^{+-} and in reflection R^{++}; (b) in transmission R^{--} and in reflection R^{-+}; the width between the dashed lines indicates the footprint of the analyser.

certainly much lower than spin-flip off-specular scattering along the superstructure Bragg sheet. So here it is confirmed that the intensity at half-order Bragg-positions consists of spin-flipped neutrons [4]. However, at the crossing point between the superstructure Bragg sheets and the specular ridge the spin-flipped component is attributed in our case to off-specular scattering, but not to the specular reflection.

The absence of true superstructure spin-flip reflection would mean, that the lateral extension of lateral order in the multilayer may persist only within the range smaller than the corresponding projection of the neutron coherence length which can be estimated to be in the order of a few microns. Therefore, the model of homogeneously magnetized layers stacked into a sequence with magnetization direction varying between next Fe-layers is rather unsuitable. This may easily be demonstrated in a direct calculation of the spin-flip and non-spin-flip reflectivity, using the super-matrix formalism [9] generalizing conventional matrix routine for the case of spin-$\frac{1}{2}$ particle reflection.

Therefore we can make two important statements.

The first one is that to fit specular reflectivities and to make conclusions about the coupling angle with a model which takes into account *only* the specular reflection is not a reliable procedure. In such a case the intensity of the off-specular and specular scattering cannot be separated correctly, therefore only a global treatment and analysis of the experimental data including off-specular scattering will give correct results [10].

The second one is that the strong off-specular spin-flip scattering concentrated into the superstructure Bragg-sheets as shown in our experiment unambiguously manifests quite perfect antiferromagnetic ordering across the multilayer structure. Thus one may assume that the sample is decomposed into a set of domains in which alternating iron layer magnetization complete an antiferromagnetic sequence. However, the in-plane magnetization is homogeneous only within a certain, relatively small, range. A small domain size is supported by electron diffraction on a similar sample [11]. This range of a small domain size, however, cannot be simply deduced from the extension range of the Bragg sheets, which in Figs. 2, 4 run through the whole α_i–α_f map. It would also be incorrect just to fit them as a function of the lateral component of the momentum transfer due to the refraction effects clearly seen in Fig. 2 close to the critical angles of incidence or scattering. Moreover, as shown in Ref. [9], in the vicinity of those angles the birefringence effect splits the spin states in the scattered beam. All these effects are taken into account in the supermatrix routine developed in Ref. [9] for grazing incidence polarized neutron magnetic diffraction from layered structures. A similar routine may also be used to treat off-specular scattering from antiferromagnetic domains. Then, by fitting the theoretical model to the data with off-specular magnetic scattering details of the domain structure will be obtained [10]. The results of such calculation are presented in Fig. 5b in comparison with the experimental contour map plotted in Fig. 5a. Remarkable agreement between theory and experiment is reached using a model of domains with average lateral dimension of the order of 3000 Å and the magnetisation in successive Fe layers is predominantly and anti-ferromagnetically ordered through all the multilayer stack. As one can see from Fig. 5, this simple model not only reproduces correctly the general features of the experiment, such as spin-flip off-specular scattering sheets along with the non-spin-flip reflectivity ridge, but it also describes the effect of birefringent splitting (peak at $\alpha_{in} \sim 0.003$ and $\alpha_{final} \sim 0.005$), slight asymmetry of the superstructure sheets as well as

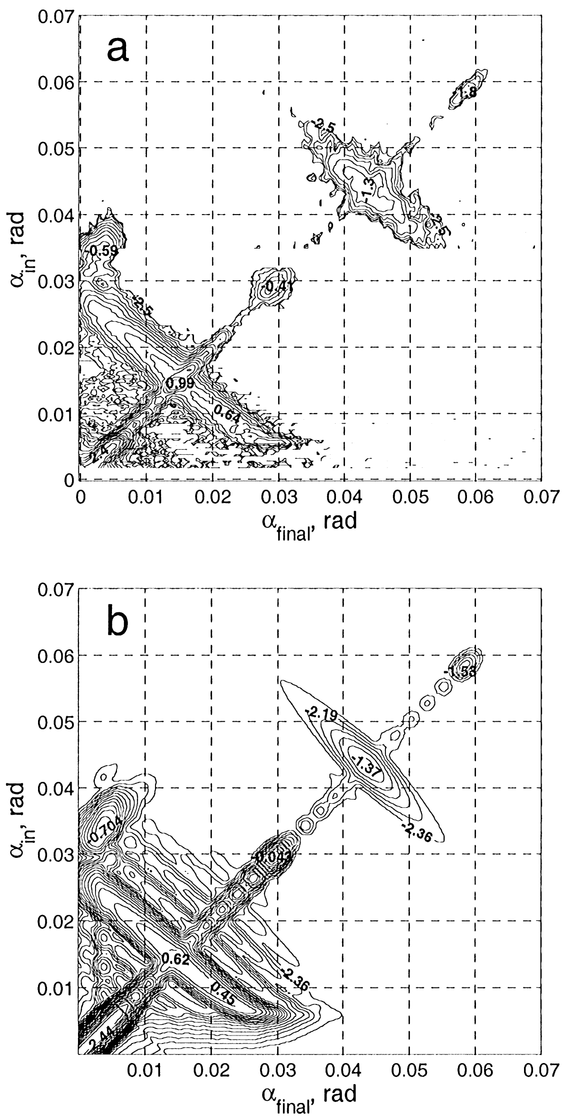

Fig. 5. Intensity map of off-specular spin-flipped and specular non-spin-flipped neutrons from Fe/Cr multilayer (a) experiment as in Fig. 2a presented with contour lines; (b) result of the supermatrix calculation with the model of antiferromagnetic domains. The numbers correspond to the log-gray-scale bar from Fig. 2.

the overall behavior of intensities along and across the sheets.

From the discussion of the data the following first picture arises: the magnetic disorder is by far dominating. In a model of magnetic domains, the domain size must be rather small and could agree with a domain size of 3000 Å as deduced from measurements of a similar sample [11]. Within the domains antiferromagnetic order must be predominant to create the intensity at the half-order position of the nuclear Bragg-peaks, which is also determinative for the off-specular scattering.

Acknowledgements

We thank R. Siebrecht for help on the ADAM spectrometer. The work performed in Ekaterinburg was supported in part by RFBR (Grant No. 98-02-17517).

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