

Journal of Magnetism and Magnetic Materials 226-230 (2001) 1694-1696

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Magnetic neutron off-specular scattering for the direct determination of the coupling angle in exchange-coupled multilayers

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Abstract

A new method is presented for the direct determination of the coupling angle in magnetic exchange multilayers. Strong anomalies in the off-specular neutron spin-flip scattering intensity at critical angles give a new tool, which is tested with Fe/Cr multilayers. In addition, a complete two-dimensional data analysis of specular reflection and off-specular scattering has been employed to verify atomic spin correlation in Fe/Cr multilayers, a typical system showing the GMR effect. \odot 2001 Elsevier Science B.V. All rights reserved.

Keywords: Polarized neutron reflection; Thin film—multilayers; Non-collinear exchange coupling

The investigation of non-collinear magnetic ordering is of basic interest in magnetic multilayered systems [1]. Methods investigating magnetic ordering, such as magnetooptics, magnetometry, etc. (see, e.g., Ref. [2]), use essential model assumptions. Also polarized neutron re flectometry (PNR) was applied to determine the coupling angle from specular reflection (see review in Ref. [3]). In this case, the magnetic structure is obtained through the model fit to the reflectivity line. However, the specular reflection cannot be evaluated alone without regarding the magnetic off-specular scattering in the half-order Bragg-peaks position. In order to provide a complete description of the magnetic structure, the specular and the off-specular scattering over a large range of incident and outgoing angles $(\alpha_{in} - \alpha_f)$ should be analyzed [4].

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Here we show that critical scattering effects in the spinflipped off-specular Bragg-sheet are determined by the magnetic spin configuration in the multilayer. Thus, the experimentally measured polarized neutron off-specular scattering can be used for the direct determination of the coupling angle θ in magnetic multilayers.

The PNR experiments on a MBE-grown $\lceil5^7\text{Fe}(68\text{\AA})/\text{Cr}(9\text{\AA})\rceil \times 12$ multilayer were performed on the spectrometer ADAM at ILL, $¹$ with the external mag-</sup> netic field H applied along the sample plane (see scheme in top of Fig. 1). In Fig. $1a-c$ the presented experimental data are obtained for the neutron spin parallel $(4 + \cdot)$ state) and anti-parallel $($ '-state) to *H*. The general features of the specular reflection ($\alpha_{\rm in} = \alpha_{\rm f}$) like the oscillations arising from the total thickness and the full-order Bragg peak ($\alpha_{\text{in}} = \alpha_{\text{f}} = 0.029 \text{ rad}$) are discussed in Ref. [4]. Here we focus on the off-specular scattering of only

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Fig. 1. Experimental intensity map of specular and off-specular scattered neutrons from ⁵⁷Fe/Cr multilayer as a function of $\alpha_{\rm in}$ and $\alpha_{\rm f}$, the incident and outgoing scattering angles, respectively: (a) for 4 and (b) for 4 - α -spin state at *H* = 0.428 kOe; (c) for 4 α -spin state at $H = 1.59$ kOe (a')–(c') are the corresponding calculations [4]. Top row schemes: (left and right) layer magnetic moment arrangement for $H = 0.428$ and 1.59 kOe, respectively; (middle) schematic presentation of the Fe/Cr multilayer with two idealized domains in a magnetic field *H* and the scattering geometry with the orientation of neutron polarization.

the half-order Bragg-sheet crossing the specular line around $\alpha_{\rm in} = \alpha_{\rm f} = 0.0145 \text{ rad}$, and which has a spin-flip origin caused by the magnetic fluctuations in the lateral direction (column-like magnetic domains with perfect non-collinear alignment across the film) $[4]$. It should be noted that due to a very good quality of the sample neither off-specular scattering across the full-order Bragg peak nor Yoneda scattering arising from interface roughness were detected.

The main feature of the off-specular scattering is its asymmetry in the vicinity of the axes $\alpha_{in} = 0$ and $\alpha_f = 0$. The critical values, at which the off-specular scattering is cut, are related to two scattering length densities *Nb* being different for the two neutron spin states: $Nb^{\pm} = Nb_n \pm Nb_m$, where *N* is the atomic number density, b_n and b_m are nuclear and magnetic parts of the scattering lengths, respectively. The critical angle

 $\alpha_{\text{in}}^{\text{c}} = 5 \times 10^{-3}$ rad for the incoming neutrons for the $\overline{+}$ '-state in Fig. 1a is related to \overline{Nb}^{\pm} and is determined by the equation $\sin \alpha_{\text{in},f}^{\text{c}} = \lambda \sqrt{Nb^{\frac{1}{2}}/\pi}$, with λ being the neutron wavelength ($\lambda = 4.41$ A). The critical angle α_f^e for the spin–flipped outgoing neutrons is related to $Nb^{-} < 0$, and therefore the off-specular scattering extends down to $\alpha_f = 0.$

In the case of non-collinear ordering, the magnetic moments in successive Fe layers are rotated by the angle \pm $\theta/2$ with respect to the magnetic field (and the neutron polarisation; see scheme above Fig. 1). The perpendicular component of the Fe layer magnetisation causes the spin-flip process. The parallel component determines the magnetic part of the neutron scattering length density. From this parallel component, the coupling angle is reconstructed (see the left diagram on top of Fig. 1) assuming an atomic magnetic Fe-moment $\mu_{Fe} = 2.2 \mu_B$.

Thus the relation $Nb_m = cN\mu\cos(\theta/2)$ is used with $c = 0.2692 \times 10^{-4} \text{ Å}/\mu_\text{B}$ and μ the atomic magnetic moment. So, for $H = 0.428$ kOe (Fig. 1a) a coupling angle of 130° was determined.

For $H = 1.59 kOe$ the critical angle increases to $\alpha_{\text{in}}^{\text{e}} = 6.1 \times 10^{-3}$ rad and the coupling angle reduces to 70 $^{\circ}$ (Fig. 1c). Further, at saturation at *H* = 5.48 kOe the magnetic moments in the Fe layers align parallel ($\theta = 0^{\circ}$) [4].

So far, the coupling angle was obtained using the $'+$ '-state (Fig. 1a and c). In Fig. 1b, the $-$ "-state is depicted. It is evident that all off-specular intensity is mirrored at the specular line and the same result for θ is obtained. In order to prove the validity of the abovedescribed procedure, the complete 2D data analysis was performed. The calculations of the intensity maps in Fig. $1a'-c'$ with the supermatrix formalism [4,5] reproduce quantitatively all the features of the experimental intensity maps using the above-mentioned values of the coupling angle and other structural parameters. In particular, the cut-off of the half-order off-specular Bragg-sheet is reproduced.

In summary, the anomalies in the off-specular scattering intensity allow to conclude already without spin analysis about the spin-flip character of the off-specular half-order Bragg sheet scattering. In addition, the quantitative evaluation of the value, at which the off-specular intensity is cut, is directly related to the parallel component of the layer magnetic moment and thus to the coupling angle. This gives a simple and effective tool to measure directly the coupling angle in exchange-coupled multilayers and does not require a complete data analysis.

We thank R. Siebrecht for help during the experiment. This work was supported by the German BMBF (No. 03DUOTU14) and RFBR (Grant No. 00-15-96745).

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