ELSEVIER

Physica B 267–268 (1999) 198–202

PHYSICA B

Polarized neutron grazing angle birefrigent diffraction from magnetic stratified media

B.P. Toperverg[b,c,*], A. Rühm[a], W. Donner[a], H. Dosch[a]

[a] *Max-Planck-Institut für Metallforschung, D-70569 Stuttgart, Germany*
[b] *Petersburg Nuclear Physics Institute, Gatchina, St. Petersburg 188350, Russia*[1]
[c] *Institut Laue–Langevin, Theory Group, B.P. 156, F-38042 Grenoble Cedex 9, France*

Abstract

Polarized neutron diffraction in multilayers is considered in the framework of the Supermatrix formulation of the distorted wave born approximation (DWBA). The general equations for the polarized neutron scattering cross sections are derived and illustrated by the example of diffraction from the regular ferromagnetic multilayered structure. It is shown that, due to the resonance enhancement of the neutron wave field inside the films, the intensity of birefrigent spin-flip and non-spin-flip diffraction is significantly amplified, if the wavelength is matched with the multilayers period.

PACS: 75.70; 68.48; 68.65; 78.70

Keywords: Polarized neutrons; Multilayer

The interplay between wave-like and geometrical optic limits, as well as between the behavior of a quantum spin-$\frac{1}{2}$ particle and its optic counterpart, is nicely demonstrated experimentally and fairly understood theoretically by the example of neutron grazing angle diffraction from a single magnetic film on a non-magnetic substrate [1]. As is well known [2], if the neutron wave impinges on a solid at grazing incidence its propagation perpendicular to the surface is mostly described by geometrical optics, while the diffraction in the direction parallel to the surface plane manifests its wave-like behavior and occurs if the lateral component $\boldsymbol{Q}$ of the momentum transfer is equal to a reciprocal lattice vector $\boldsymbol{\tau}$ displayed within the surface plane. Due to the optical effects the diffracted beam intensity shows a sharp maximum if the normal to the surface components, the incoming, p^{i}, and outgoing, p^{f}, wave vectors are close to the critical wave number of the total reflection. Due to the Zeeman effect [3,4] the magnetized media is optically active, and the incident and diffracted waves, refracted at the surface, are split into two components with different neutron spin projections onto the field direction

* Corresponding author. Present address: Institut Laue–Langevin, Theory Group, B.P. 156, F-38042 Grenoble Cedex, France. Fax: + 33-476-882416.

E-mail address: boristop@ill.fr (B.P. Toperverg)

[1] Permanent address. Tel.: + 7-812-323-38-98; fax: + 7-812-713-19-63; e-mail: boristop@thd.pnpi.spb.ru.

0921-4526/99/$ – see front matter
PII: S0921-4526(99)00066-6

and with different momentum transfer components perpendicular to the surface. Magnetic scattering by the atomic spins mixes neutron spin states and causes the spin-flip process. This results in the partial polarization of the four diffracted beams recorded in Ref. [1] in the scan over the $(p^{\mathrm{i}}, p^{\mathrm{f}})$-plane. As we shall show, similar effects are dramatically enhanced along the Bragg sheets in multilayered structures if either the incoming or the outgoing glancing angle is close to the angle of the total reflection. General equations for the polarized neutron diffraction will be derived and illustrated by the example of the perfectly ordered single crystalline multibilayer.

The neutron wave falling onto the surface at low angle $\alpha \ll 1$ and having a small wave vector component $p_0 \approx k\alpha$ orthogonal to the surface averages out almost all details of the crystal structure over its wavelength $\lambda_\perp = 2\pi/p \gg a$, where a is the interatomic spacing and $k = 2\pi/\lambda$ is the neutron wave number. This is a reason to decompose the Hamiltonian $\mathscr{H}(\boldsymbol{r})$ of the neutron interaction with the magnetic crystal into two parts: $\mathscr{H}(\boldsymbol{r}) = \mathscr{H}_0(z) + \tilde{\mathscr{H}}(\boldsymbol{r})$, where the first term $\mathscr{H}_0(z)$ depends upon the only coordinate z perpendicular to the surface and accounts for the interaction with the potential averaged over the crystalline structure. The second one $\tilde{\mathscr{H}}(\boldsymbol{r})$ varies with the period of the lattice spacing and causes the Bragg diffraction. The Hamiltonian $\tilde{\mathscr{H}}(\boldsymbol{r})$ is regarded as a perturbation for the eigenfunctions of the Hamiltonian $\mathscr{H}_0(z)$. The latter describes the optical effects, i.e. the mirror reflection from, refraction by, and transmission through the mean interaction potential: $\mathscr{H}_0(z) = V(z) - \hat{\boldsymbol{\mu}}\boldsymbol{B}(z)$, where $V(z) = \langle V(\boldsymbol{r})\rangle$ is the mean value of the nuclear potential $V(\boldsymbol{r})$, $\boldsymbol{B}(z) = \langle \boldsymbol{B}(\boldsymbol{r})\rangle$ is the mean magnetic field, $\hat{\boldsymbol{\mu}} = \mu\hat{\boldsymbol{\sigma}}$ is the neutron magnetic moment, $\hat{\boldsymbol{\sigma}} = \{\hat{\sigma}_x, \hat{\sigma}_y, \hat{\sigma}_z\}$ is a set of the Pauli matrices. In stratified media the Hamiltonian is written as sums: $\mathscr{H}_0(z) = \sum\{\mathscr{H}_m(z) + \tilde{\mathscr{H}}_m(z)\}$, where $\mathscr{H}_m(z) = \bar{V}_m - \hat{\boldsymbol{\mu}}\bar{\boldsymbol{B}}_m$, is the optical potential of the mth layer of the thickness $d_m = z_m - z_{m-1}$, z_m is the coordinate of the mth interface, and $z_0 = 0$. The eigenvectors of the reference Hamiltonian $\mathscr{H}_0(z)$ are factorized into the products: $|\Psi(\boldsymbol{r})\rangle = \exp(\mathrm{i}\boldsymbol{\kappa}\boldsymbol{\rho})|\psi(z)\rangle$, where $\boldsymbol{\kappa}$ is the in-plane component of the wave vector $\boldsymbol{k}$, $\boldsymbol{\rho}$ is the lateral coordinate, and $|\psi(z)\rangle$ is the two-component vector in the neutron spin space.

The interaction with the reference Hamiltonian $\mathscr{H}_0$ causes the specular reflection, and transforms the vector of the incoming neutron state $|\psi_{\mathrm{i}}(z)\rangle = \exp(\mathrm{i}p_0 z)|t_0\rangle$ into the final state vector $\psi_r(z) = \exp(-\mathrm{i}p_0 z)\hat{R}|t_0\rangle$, where $\hat{R}$ is the matrix of the reflection amplitudes and $|t_0\rangle$ is the vector of the neutron initial spin state. Then, the probability amplitude to find a reflected neutron in a certain state $|r\rangle$ is equal to the element $\langle r|\hat{R}|t_0\rangle$ of the 2×2 matrix $\hat{R}$. The reflectivity $\mathscr{R}$ is defined as a mean value $\mathscr{R} = \overline{|\langle r|\hat{R}|t_0\rangle|^2} = \frac{1}{4}\mathrm{Tr}\{\hat{\rho}_0\hat{R}^+\hat{\rho}\hat{R}\}$, where the density matrices of the polarizer, $\hat{\rho}_0$, and the analyzer $\hat{\rho}$ are parametrized as $\hat{\rho}_0 = \overline{|t_0\rangle\langle t_0|} = \{1 + \boldsymbol{P}_0\hat{\boldsymbol{\sigma}}\}$ and $\hat{\rho} = \overline{|r\rangle\langle r|} = \{1 + \boldsymbol{P}\hat{\boldsymbol{\sigma}}\}$, i.e. by the initial polarization vector $\boldsymbol{P}_0$ and the vector of the polarization analysis $\boldsymbol{P}$.

The cross section of the diffraction is proportional to the averaged over the spin states modulus of the matrix element $\langle\Psi^{\mathrm{f}}|\tilde{\mathscr{H}}|\Psi^{\mathrm{i}}\rangle$ squared, with superscripts (i, f) referred to the initial or final neutron states. Due to the in-plane periodicity of the Hamiltonian $\tilde{\mathscr{H}}$ the diffraction occurs at $\boldsymbol{Q} = \boldsymbol{\tau}$, and in its amplitude

$$f(p^{\mathrm{f}}, p^{\mathrm{i}}; \boldsymbol{Q}) \propto \langle\psi^{\mathrm{f}}|\tilde{\mathscr{H}}|\psi^{\mathrm{i}}\rangle = \sum_m \langle\psi^{\mathrm{f}}_m|\tilde{\mathscr{H}}_m|\psi^{\mathrm{i}}_m\rangle, \qquad (1)$$

where $|\psi_m^{\mathrm{i,f}}\rangle$ are the vectors of the neutron states within the mth layers. These vectors may generally be represented as a linear superposition

$$|\psi_m(z)\rangle = \{\exp(\mathrm{i}\hat{\varphi}_m(z))\hat{t}_m + \exp(-\mathrm{i}\hat{\varphi}_m(z))\hat{r}_m\}|t_0\rangle, \quad (2)$$

of the transmitted and reflected waves propagating inside the layer. Here $\hat{\varphi}_m(z) = \hat{p}_m(z - z_m)$, $\hat{p}_m^2 = p_0^2 - \hat{q}_{mc}^2$, $\hbar^2\hat{q}_{mc}^2 = 2m\mathscr{H}_m(z)$, and $|t_m\rangle = \hat{t}_m|t_0\rangle$, $|r_m\rangle = \hat{r}_m|t_0\rangle$, $\hat{t}_m = |t_m\rangle\langle t_0|$ is the transmission, while $\hat{r}_m = |r_m\rangle\langle t_0|$ is the reflection matrix. For arbitrary non-collinear multilayered structure their elements can, for instance, be computed by use of the supermatrix approach which generalizes the conventional matrix formalism [6] for the case of the spin-$\frac{1}{2}$ particle interacting with a stratified magnetic media. Such a generalization is needed due to the fact that entering the magnetic layer the neutron wave is birefrigent into two with the wave numbers $p_{m\pm} = \{p_0^2 - q_{mc\pm}^2\}^{1/2}$, which are the eigenvalues of the operator $\hat{p}_m$ and $q_{mc\pm}$ are the critical wave numbers of the total reflection for one or the other spin component.

The reflected wave is also split and four waves with the wave numbers $\pm p_{m\pm}$ are travelling in the layer. However, in the course of refraction neutron spin may be flipped and the matrices $\hat{t}_m$ and $\hat{r}_m$ are, in general, not diagonal in the representation with the quantization axis along the mean magnetic field inside the layer.

After substitution $|\psi(z)\rangle$ from Eq. (2) into Eq. (1) one can express the amplitude $f_m = f_m(p^f, p^i; \boldsymbol{\tau})$ of diffraction from the mth layer via the atomic scattering operator $\hat{f}_m = F^N_m + F^M_m(\boldsymbol{m}^{\perp}_m\hat{\boldsymbol{\sigma}})$, where $F^N_m = b^N_m S^N_m$, is the nuclear and $F^M_m = b^M_m S^N_m$ is the magnetic scattering amplitude, b^N_m is the nuclear, and b^M_m is the magnetic scattering length, S^N_m is the nuclear structure factor, and S^M_m is the magnetic form-factor of the unit cell; $\boldsymbol{m}^{\perp}_m = \boldsymbol{m}_m - \boldsymbol{e}(\boldsymbol{e}\boldsymbol{m}_m)$ is the component of the unit vector $\boldsymbol{m}_m = \boldsymbol{M}_m/M_m$, perpendicular to the unit vector $\boldsymbol{e}$ directed along the momentum transfer, and $\boldsymbol{M}_m$ is the magnetic moment of the ions in the mth layer. The operator $\hat{f}_m$ does not commute with either $\hat{p}_m$, or with $\hat{t}_m$ and $\hat{r}_m$. Therefore the equation for the scattering amplitude contains a number of terms which may be classified if f_m is represented as $f_m = \langle r|\hat{\mathscr{F}}_m|t_0\rangle$, where the operator $\hat{\mathscr{F}}_m$, which transforms the incoming state into the final, is decomposed into the sum: $\hat{\mathscr{F}}_m = \hat{\mathscr{F}}^{tt}_m + \hat{\mathscr{F}}^{tr}_m + \hat{\mathscr{F}}^{rt}_m + \hat{\mathscr{F}}^{rr}_m$. The partial scattering operators $\hat{\mathscr{F}}^{tt}_m = \hat{t}^f_m \hat{F}^{tt}_m \hat{t}^i_m$, transform the transmitted into the layer wave to the wave first scattered and then transmitted through the layer, $\hat{\mathscr{F}}^{rr}_m = \hat{r}^f_m \hat{F}^{rr}_m \hat{r}^i_m$ transforms the initially reflected wave into that which is scattered and then reflected, while the operators $\hat{\mathscr{F}}^{tr}_m = \hat{t}^f_m \hat{F}^{tr}_m \hat{r}^i_m$, and $\hat{\mathscr{F}}^{rt}_m = \hat{r}^f_m \hat{F}^{rt}_m \hat{t}^i_m$, mix reflected and transmitted waves. The component of the (super)matrix $\hat{F}^{\alpha\beta}_m$ (with $\{\alpha, \beta\} = \{t, r\}$) are written as

$$\begin{aligned}\hat{F}^{\alpha\beta}_m = \tfrac{1}{2}\{&[F^N_m + F^M_m(\boldsymbol{b}_m\boldsymbol{m}^{\perp}_m)][1 + (\boldsymbol{b}_m\boldsymbol{\sigma})]G^{\alpha\beta}_{++}\\ &+ [F^N_m - F^M_m(\boldsymbol{b}_m\boldsymbol{m}^{\perp}_m)][1 - (\boldsymbol{b}_m\hat{\boldsymbol{\sigma}})]G^{\alpha\beta}_{--}\\ &+ F^M_m[(\boldsymbol{b}^{\perp}_m\hat{\boldsymbol{\sigma}})(G^{\alpha\beta}_{+-} + G^{\alpha\beta}_{-+})\\ &- \mathrm{i}(\boldsymbol{b}^a_m\hat{\boldsymbol{\sigma}})(G^{\alpha\beta}_{+-} - G^{\alpha\beta}_{-+})]\}. \end{aligned} \qquad (3)$$

Here $\boldsymbol{b}_m$, $\boldsymbol{b}^{\perp}_m = \boldsymbol{m}^{\perp} - \boldsymbol{b}_m - (\boldsymbol{b}_m\boldsymbol{m}^{\perp})$ and $\boldsymbol{b}^a_m = [\boldsymbol{m}^{\perp} \times \boldsymbol{b}_m]$ are three orthogonal vectors, and $G^{\alpha\beta}_{\mu\nu}$ are the Laue functions:

$$\begin{aligned} G^{\alpha\beta}_{++} &= \{\mathrm{e}^{\mathrm{i}(\varphi^f_{m\alpha} + \varphi^i_{m\beta})} - 1\}/\{\mathrm{e}^{\mathrm{i}(\phi^f_{m\alpha} + \phi^i_{m\beta})} - 1\},\\ G^{\alpha\beta}_{+-} &= \{\mathrm{e}^{\mathrm{i}(\varphi^f_{m\alpha} - \varphi^i_{m\beta})} - 1\}/\{\mathrm{e}^{\mathrm{i}(\phi^f_{m\alpha} - \phi^i_{m\beta})} - 1\},\\ G^{\alpha\beta}_{--} &= G^{\alpha\beta}_{++}\mathrm{e}^{-2\mathrm{i}[(\varphi^f_{m\alpha} + \varphi^i_{m\beta}) - (\phi^f_{m\alpha} + \phi^i_{m\beta})]},\\ G^{\alpha\beta}_{-+} &= G^{\alpha\beta}_{+-}\mathrm{e}^{-2\mathrm{i}[(\varphi^f_{m\alpha} - \varphi^i_{m\beta}) - (\phi^f_{m\alpha} - \phi^i_{m\beta})]}, \end{aligned} \qquad (4)$$

where $\phi_{m\alpha} = p_{m\alpha}a_m$, and a_m is the unit cell constant.

If one represents the matrices $\hat{t}_m$ and $\hat{r}_m$ in the form [5]: $\hat{t}_m = t_{0m} + (\boldsymbol{t}_m\hat{\boldsymbol{\sigma}})$, where $t_{0m} = \frac{1}{2}\mathrm{Tr}\{\hat{t}_m\}$ and the vector $\boldsymbol{t}_m = \frac{1}{2}\mathrm{Tr}\{(\hat{t}_m\hat{\boldsymbol{\sigma}})\}$, and $\hat{r}_m = r_{0m} + (\boldsymbol{r}_m\hat{\boldsymbol{\sigma}})$, where $r_{0m} = \frac{1}{2}\mathrm{Tr}\{\hat{r}_m\}$ and $\boldsymbol{r}_m = \frac{1}{2}\mathrm{Tr}\{(\hat{r}_m\hat{\boldsymbol{\sigma}})\}$, then using Eqs. (3) and (4) one can readily calculate the matrix elements $\hat{\mathscr{F}}^{\alpha\beta}_m$. However, the result looks quite lengthy, and we do not bring it into the paper. Instead, we note, that these calculations are needed to determine the parameters of scattering operator $\hat{\mathscr{F}}_m$. If it is represented as $\hat{\mathscr{F}}_m = \mathscr{F}_{0m} + (\mathscr{F}_m\hat{\boldsymbol{\sigma}})$, and $\mathscr{F} = \boldsymbol{b}\mathscr{F}_1$, with $|\boldsymbol{b}| = 1$ then $\mathscr{F}_{0,1} = \frac{1}{2}[\mathscr{F}_+ \pm \mathscr{F}_-]$, where $\mathscr{F}_{\pm}$ are the eigenvalues of the matrix $\hat{\mathscr{F}}$.

This representation, leads us to the general equation for the polarized neutron scattering cross section:

$$\begin{aligned}\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \tfrac{1}{2}\{&|\mathscr{F}_0|^2[1 + (\boldsymbol{P}_0\boldsymbol{P})] + |\mathscr{F}|^2[1 - (\boldsymbol{P}_0\boldsymbol{P})]\}\\ &+ \mathrm{Re}\{\mathscr{F}^*_0(\mathscr{F}[\boldsymbol{P}_0 + \boldsymbol{P}]) + (\mathscr{F}^*\boldsymbol{P}_0)(\mathscr{F}\boldsymbol{P})\}\\ &- \mathrm{Im}\{\mathscr{F}^*_0(\mathscr{F}[\boldsymbol{P}_0 \times \boldsymbol{P}])\\ &+ \tfrac{1}{2}(\boldsymbol{P}_0 - \boldsymbol{P})[\mathscr{F}^* \times \mathscr{F}]\}, \end{aligned} \qquad (5)$$

where $\mathscr{F}_0$ and $\mathscr{F}$ are the sums of $\mathscr{F}_{m0}$ and $\mathscr{F}_m$ over the whole sequence of layers.

Usually only the projections $\pm P = \pm(\boldsymbol{P}\boldsymbol{b}_0)$ onto the direction of the incident polarization $\boldsymbol{b}_0$ can be analyzed for initial polarization vectors $\boldsymbol{P}_0 = \pm P_0\boldsymbol{b}_0$, where $|\boldsymbol{b}_0| = 1$, P_0 and P are the efficiencies of the polarizing and analyzing devices, respectively. Hence, in an ideal case of $P = P_0 = 1$ the equations for four scattering cross sections $(\mathrm{d}\sigma/\mathrm{d}\Omega)_{\mu\nu}$: non-spin-flip if $\{\mu\nu\} = \{\pm\pm\}$, and spin-flip, if $\{\mu\mu\} = \{\pm\mp\}$, have the form:

$$\frac{\mathrm{d}\sigma^{\mathrm{fi}}}{\mathrm{d}\Omega_{++}} = \left|\mathscr{F}_+\cos^2\frac{\gamma}{2} + \mathscr{F}_-\sin^2\frac{\gamma}{2}\right|^2, \qquad (6)$$

$$\frac{\mathrm{d}\sigma^{\mathrm{fi}}}{\mathrm{d}\Omega_{--}} = \left|\mathscr{F}_+\sin^2\frac{\gamma}{2} + \mathscr{F}_-\cos^2\frac{\gamma}{2}\right|^2, \qquad (7)$$

$$\frac{d\sigma^{fi}}{d\Omega_{+-}} = \frac{d\sigma^{if}}{d\Omega_{-+}} = \frac{1}{4}|\mathscr{F}_{+} - \mathscr{F}_{-}|^2 \sin^2\gamma, \tag{8}$$

where γ is the angle between the vectors $\boldsymbol{b}_0$ and $\boldsymbol{b}$.

For the regular multilayered structure the summation over m can be performed analytically [7] and the results for the case of the magnetized ferromagnetic multibilayer are illustrated in Fig. 1, where the diffraction intensities for the periodic stacking of 20 bilayer (70Fe/30Si) are plotted as a functions of the incident, α_i, and scattered, α_f, glancing angles.

In Fig. 1 one can observe a set of features usual for such systems: peaks of intensity in the range of total reflection, quasi-Bragg sheets [8], etc. However, we should note some new features, which are not appropriate for a non-magnetic system. First is that the position of the peak intensities are different for + + and − − scattering. This is clearly due to the different refraction indices for the neutrons with the positive and negative spin projections onto the magnetization direction. The second effect is the spin-flip diffraction. It occurs in the fairly saturated magnetic state even at $\boldsymbol{b}_0 \| \boldsymbol{b}$, when the spin-flip process is impossible in the specular channel. In the case $\tau \neq 0$ spin may be flipped by the microscopic magnetic fields created by the atomic magnetic moments. In the spin-flip diffraction neutron experience transition between the states split by the Zeeman effect and, thus, changes the absolute value of its momentum projection normal to the surface. This brings distortion into the intensity distribution over the (α_i, α_f)-plane, as it was first observed in the recent experiments and clearly seen in Figs. 3 and 4 of Ref. [1]. The effect is the most pronounced if at least one of the angles, α_i, or α_f, is small.

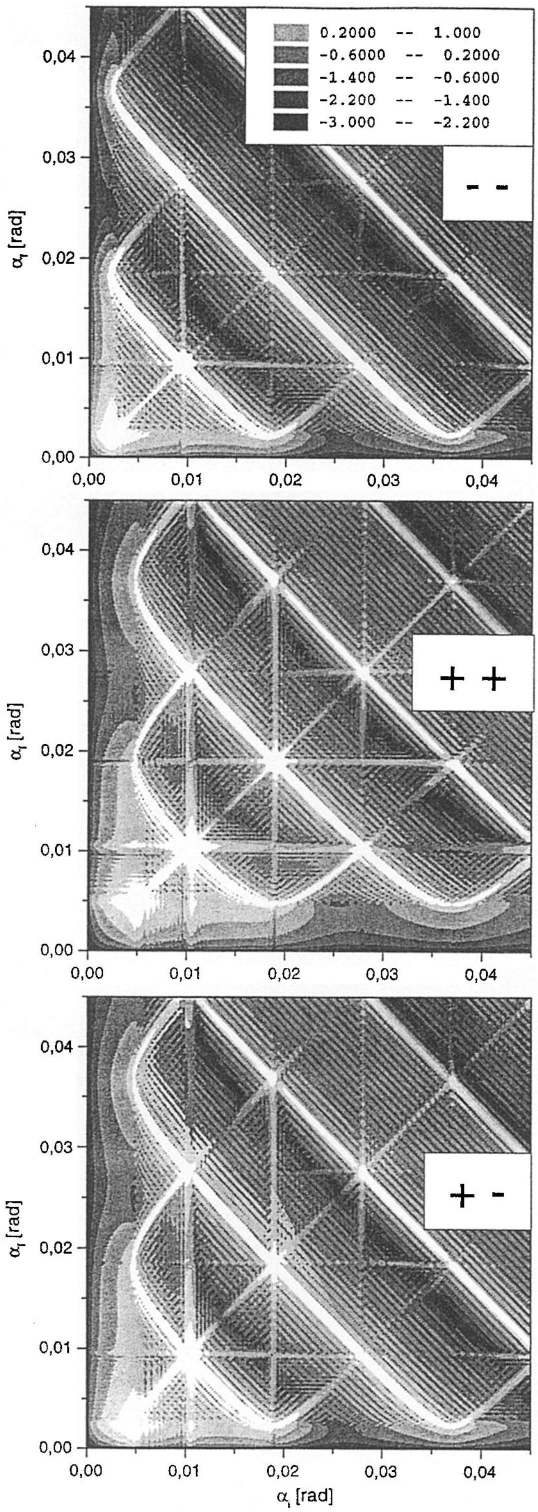

Fig. 1. Contour plots of the cross sections of: (+ +) and (− −) non-spin-flip, (+ −) spin-flip, neutron diffraction from (70Fe/30Si) multilayer computed in accordance with Eqs. (6)–(8) as a function of the scattering angles α^f at the incidence angles α^i.

Acknowledgements

We thank G. Felcher for helpful discussions. This work is supported by the BMBF-Grant No. 03-DO4WUP-2), Russian State Program "Neutron

Research of Condensed Matter" and RFFI-Grant No. L-EN-96-15-96775.

References

[1] R. Günther, W. Donner, B. Toperverg, H. Dosch, Phys. Rev. Lett. 81 (1998) 116.
[2] H. Dosch, Springer Tracts in Modern Physics, vol. 126, Springer, Heidelberg, 1992.
[3] G.P. Felcher et al., Nature 377 (1995) 409.
[4] C.S. Schneider, C.G. Shull, Phys. Rev. B 3 (1971) 830.
[5] L.D. Landau, E.M. Lifshits, Quantum Mechanics, third ed., Pergamon Press, Oxford, 1977.
[6] M. Born, E. Wolf, Principles of Optics, Pergamon, New York, 1980.
[7] B.P. Toperverg, O. Schärpf, I.S. Anderson, unpublished.
[8] V. Holý, T. Baumbach, Phys. Rev. B 49 (1994) 10 669.