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Specular reflection and off-specular scattering of polarized neutrons

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Abstract

The general situation with polarized neutron reflectivity, diffraction on a crystalline structure and off-specular scattering from magnetic inhomogeneities in thin films is thoroughly discussed. It is argued that only a combination of birefringence (spin-dependent refraction) of a neutron wave in the mean magnetic field of the film and spin-flip magnetic diffraction or scattering can lead to the splitting of the outgoing (diffracted or scattered) beam into two beams with different wave vectors and spin states. An effect of interplay between the lateral projection of the coherence length and the in-plane extension of magnetic inhomogeneities (size of domains, roughness correlation length etc.) is outlined. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Recent spectacular achievements and bright perspectives in polarized neutron (specular) reflectometry (PNR) applications for solution of the variety of intriguing problems of thin-film magnetism have received exhaustive coverage in the lecture by Felcher [1]. Among the advantages of PNR method he pointed out its ability to probe the magnetization depth profile and “unimpeachably” deduce non-collinear magnetization arrangements in magnetic multilayers. This was illustrated by the famous results [2–3] (quoted earlier [5] from Ref. [6]) on determination of the angles of 90° or

50° between the magnetization directions in neighboring Fe layers of Fe/Cr multilayers. It was also demonstrated [1] that using supermatrix routine [7] proposed in Ref. [8] one is able not only to simulate the PNR data for 70° coupling angle expected at other thickness of the Cr layers [9], but even to verify whether the magnetization of the subsequent iron layers forms a spiral structure, similar to that imprinted [10] into La/Fe system.

The only disadvantage of PNR mentioned in Ref. [1] is that it is insensitive to the magnetization component parallel to the momentum transfer which under specular conditions is normal to the surface. This missing information can be retrieved from the experiments on grazing incidence neutron diffraction (GIND) [11] carried out in addition to PNR measurements. The other problem, which was intentionally not touched in the overview [1], is that PNR does not provide any direct information

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on lateral properties of the films, completely ignores their crystalline structure and, what is more important, possible large-scale inhomogeneities: either in bulk of the layers, or/and at the interfaces. The authors [1] are certainly aware of these problems [12], but left the opportunity to fill the gap in their review solely devoted to specular PNR.

In fact, considering PNR data, it is silently anticipated that both, the nuclear and magnetic scattering length density are essentially functions of only the coordinate perpendicular to the surface, while the layers are perfectly magnetized. The latter assumption is rather inappropriate for any magnetic systems with spontaneous magnetization except for a trivial case of full saturation in a strong magnetic field parallel to the surface. Otherwise, in order to reduce a demagnetizing field outside the sample, in-layer magnetization is broken into domains. Then the interpretation of the PNR data invokes an additional hypothesis on the properties of the domains. Indeed, if the domain size much exceeds the lateral coherence length of a neutron beam, then one should first calculate the reflectivity for each of these domains, assuming that, or another type of magnetization arrangement in the direction across the multilayer stack. After that, the result should be incoherently averaged over the domain-magnetization distribution along the surface of the sample. All this, however, complicates the modeling, and in Ref. [3] the domain distribution was finally fixed, but the data were fitted by varying the coupling angle.

In the case of large domains almost no off-specular magnetic scattering should be seen, and this would deliver a direct proof of the hypothesis [3]. If on the contrary, an amount off-specular magnetic signal is recorded [13–14], then one can, in principle, determine their size and the lateral magnetization distribution. Quantitative analysis of both, PNR and off-specular scattering collected over a broad range of incident and scattering angles was performed [14] for the Fe/Cr multilayer similar to that investigated in Ref. [3]. This analysis has led the authors to an alternative model of the magnetic state in their sample. In accordance with Ref. [14], lateral magnetization is decomposed into a set of domains with dimensions of only few thousands of Angströms, remaining perfectly correlated antifer-

romagnetically across the multilayer. This result was obtained due to unambiguous experimental refinement of true specular non-spin-flip reflection from spin-flip diffuse scattering in the range where they are totally overlapped [16]. We shall see, that otherwise separation of non-spin-flip signal from spin-flip scattering does not always make problems due the effect of “autoseparation” in space, as observed in Refs. [5,17,18]. Here it is only necessary to underline that for this separation, one needs more than only a Zeeman splitting of the neutron spin states in a magnetic field. Off-specular scattering [5,17,18] absolutely demands a cause by which the translational symmetry in lateral direction is violated on the scale smaller than the lateral projection of the coherence length. One of the causes can be the formation of magnetic domains [19] of a proper size.

2. From Schrödinger equation to Larmor precession

Let us first guide the initial polarization vector \mathbf{P}_0 in the external magnetic field $\mathbf{H}(\mathbf{r})$ from the polarizer to the sample. The coherence length l_c of a monochromatic neutron beam is usually estimated via the uncertainty principle as $l_c \sim 1/\delta k$, where $\delta k = (2\pi/\lambda)\delta\Omega$, λ is the wave length and $\delta\Omega$ is the angular divergence defined by the collimation conditions. Typically, $l_c \sim 10^3\text{--}10^4$ Å. However, this value is related to the coherence across the beam, while at low angles of incidence $\alpha_i \ll 1$, an uncertainty in the lateral projection of the incoming wave vector is proportional to $\sin \alpha_i$, and the lateral projection of the coherence length $l_{||} \sim l_c/\alpha_i \gg l_c$ may reach a scale of millimeters [21]. At a scale smaller than $l_{||}$ one can use the plane wave approximation [20] for the incoming neutron and should solve the Schrödinger equation

$$\{\nabla^2 + k_0^2 - (2m/\hbar^2)\hat{V}(\mathbf{r})\}|\Psi(\mathbf{r})\rangle = 0, \quad (1)$$

where $|\Psi(\mathbf{r})\rangle$ is two-component vector of spin states, $k_0 = 2\pi/\lambda$, and the interaction operator

$$\hat{V}(\mathbf{r}) = V_N(\mathbf{r}) - \hat{\mu}\mathbf{B}(\mathbf{r}) \quad (2)$$

has 2×2 matrix representation in spin space. The first term describes the interaction with nuclei (unit

matrix for spinless nuclei is omitted), while the second one corresponds to the magnetic part of the interaction: $\mathbf{B}(\mathbf{r})$ is magnetic field, $\hat{\boldsymbol{\mu}} = \mu\boldsymbol{\sigma}$ is the neutron magnetic moment operator, $\mu = \gamma\mu_N$, μ_N is the nuclear magneton, $\gamma = -1.91$, $\frac{1}{2}\boldsymbol{\sigma} = \hat{\mathbf{s}}$ is the neutron spin operator, $\boldsymbol{\sigma} = \{\sigma^x, \sigma^y, \sigma^z\}$, σ^z are the Pauli matrices, and outside of the sample $\mathbf{B}(\mathbf{r}) = \mathbf{H}(\mathbf{r})$.

Following customary procedure in reflectometry, one should find solutions of Eq. (1) in the free space above the film and inside the substrate and match them with solutions inside the film. In the first approximation, it is assumed that $\mathbf{B}(\mathbf{r}) \approx \mathbf{B}(z)$ depends on the only coordinate z , perpendicular the surface normal. Then Eq. (1) is easily solved. However, the external field $\mathbf{H}(\mathbf{r})$ is often a function of all three coordinates. This brings enormous difficulties to finding an exact solution of Eq. (1). Nonetheless, due to the smooth variation of $\mathbf{H}(\mathbf{r})$ on a scale of l_c , one can employ a quasi-classical approach and neglect very small deviations of the incoming and reflected beams in the external field gradients. These deviations are certainly different for different spin components and result in the Stern–Gerlach effect which is observed at high-field gradients [22], but can be neglected (except for that part of the neutron beam which hits edges of a sample, where the gradients cause an appreciable effect). Then, in Eq. (1), one can approximate $\mathbf{H}(\mathbf{r}) \approx \mathbf{H}(\xi)$, where $\xi = (\mathbf{k}_0\mathbf{r})/k_0 \leq 0$, \mathbf{k}_0 is the incident wave vector, and \mathbf{r} is a radius vector pointing from the position of the sample to the neutron beam source. At shallow incidence $\xi \approx x$, where the x -axis is directed along the incoming (reflected) beam projection onto the surface plane. Under these circumstances the initial spin state vector, $|\psi_0^i\rangle = |\psi_0^i(\xi)\rangle$ is developed into the vector

$$|\Psi^i(0)\rangle = \hat{\mathcal{S}}(\xi, \boldsymbol{\sigma})|\psi_0^i\rangle, \quad (3)$$

$$\hat{\mathcal{S}}(\xi, \boldsymbol{\sigma}) = T_\xi \exp\left\{i \int_\xi^0 d\xi' \sqrt{k_0^2 - \hat{p}_M^2(\xi')}\right\}, \quad (4)$$

where T_ξ is the “chronological” operator and $\hbar^2 \hat{p}_M^2 = 2m\hat{\boldsymbol{\mu}}\mathbf{B}(\xi')$ is a function of the coordinate along the beam trace. Like any 2×2 matrix, $\hat{\mathcal{S}}(\xi)$ can be decomposed over orthogonal set of matrices

including three Pauli matrices and the unit matrix [23]:

$$\hat{\mathcal{S}}(\xi, \boldsymbol{\sigma}) = \mathcal{S}_0(\xi) + (\boldsymbol{\sigma}\mathbf{b})\mathcal{S}_b(\xi), \quad (5)$$

where $\mathcal{S}_0 = (\mathcal{S}^+ + \mathcal{S}^-)/2$ and $\mathcal{S}_b = (\mathcal{S}^+ - \mathcal{S}^-)/2$, \mathcal{S}^\pm are the eigenvalues of the $\hat{\mathcal{S}}$ -matrix, which is diagonal in the representation with the z -axis pointing along the 3D unit vector \mathbf{b} . Due to the unitarity of the \mathcal{S} -matrix its elements are not independent and $\mathcal{S}_{0,b}$ obeys the equation $|\mathcal{S}_0|^2 + |\mathcal{S}_b|^2 = 1$. Therefore, one can define

$$\mathcal{S}_0(\xi) = \cos \Phi \quad \text{and} \quad \mathcal{S}_b(\xi) = i \sin \Phi, \quad (6)$$

where the phase $\Phi = \Phi(\xi)$ is expressed via the eigenvalues $\mathcal{S}^\pm(\xi)$.

These eigenvalues and components of the vector \mathbf{b} , or directly all four elements of the $\hat{\mathcal{S}}$ -matrix, can easily be computed numerically for any configuration of the field, if one represents the integral in Eq. (4) as a finite sum of integrals over ranges of $\xi_l \leq \xi \leq \xi_{l-1}$ ($1 \leq l \leq L, \xi_0 = 0, \xi_L = \xi$), along which $\mathbf{B}(\xi)$ does not significantly change direction. Then,

$$\hat{\mathcal{S}}(\xi) = \hat{\mathcal{S}}_1(0, \xi_1)\hat{\mathcal{S}}_2(\xi_1, \xi_2) \cdots \hat{\mathcal{S}}_L(\xi_{L-1}, \xi). \quad (7)$$

At each of those intervals a quantization axis z_l can be chosen parallel to the direction $\mathbf{b}_l = \mathbf{B}_l/|\mathbf{B}_l|$ of the local field $\mathbf{B}_l = \mathbf{B}(\xi_l)$, and then

$$\hat{\mathcal{S}}_l = \frac{1}{2}\{(\mathbf{e}^{i\phi_l^+} + \mathbf{e}^{i\phi_l^-}) + (\boldsymbol{\sigma}\mathbf{b}_l)(\mathbf{e}^{i\phi_l^+} - \mathbf{e}^{i\phi_l^-})\}, \quad (8)$$

where $\phi_l^\pm \approx \sqrt{k_0^2 \mp p_M^2(\xi_l)}(\xi_{l-1} - \xi_l)$.

The Zeeman splitting of the neutron spin states in the external field is usually rather small with respect to the neutron kinetic energy. Consequently, the eigenvalues $\pm p_M^2(\xi_l)$ of the operator $\hat{p}_M^2(\xi_l)$ are also rather small, and $\{k_0^2 \mp p_M^2(\xi_l)\}^{1/2} \approx k \mp (\omega_l/2v)$, where $\omega_l = \mu\mathbf{B}_l/\hbar$ is the Larmor frequency of neutron spin precession in the field \mathbf{B}_l , and $v = \hbar k_0/m$ is the neutron velocity. Then Eq. (8) is substantially simplified, and

$$\hat{\mathcal{S}}_l = \cos \bar{\phi}_l/2 + i(\boldsymbol{\sigma}\mathbf{b}_l)\sin \bar{\phi}_l/2, \quad (9)$$

where $\bar{\phi}_l \approx \omega_0(\xi_{l-1} - \xi_l)/v$. As soon as the \mathcal{S} -matrix is computed, one can follow an evolution of the incoming polarization vector $\mathbf{P}_i = \mathbf{P}_i(0)$, which is defined as an expectation value $\mathbf{P}_i = \langle \Psi^i(0)|\mathbf{s}|\Psi^i(0)\rangle$, averaged over the spin states

produced by the polarizer. Taking into account Eq. (3) one has

$$\mathbf{P}_i = \frac{1}{2} \text{Tr} \{ \hat{\mathcal{S}}(\xi) \hat{\rho}_0(\xi) \hat{\mathcal{S}}^+(\xi) \boldsymbol{\sigma} \} = \frac{1}{2} \text{Tr} \{ \hat{\rho}_i(0) \boldsymbol{\sigma} \}, \quad (10)$$

where $\hat{\rho}_0 = \overline{|\psi_0\rangle\langle\psi_0|} = \{1 + (\mathbf{P}_0 \boldsymbol{\sigma})\}/2$ is the initial density matrix and $\mathbf{P}_0 = \mathbf{P}_0(\xi)$ is the initial polarization. Then, the initial density matrix $\hat{\rho}_0 = \rho_0(\xi)$ is transformed into the matrix $\hat{\rho}_i = \overline{|\Psi_i\rangle\langle\Psi_i|} = \{1 + (\mathbf{P}_i \boldsymbol{\sigma})\}/2$ with

$$\mathbf{P}_i = \mathbf{b}(\mathbf{P}_0 \mathbf{b}) + [\mathbf{P}_0 - \mathbf{b}(\mathbf{P}_0 \mathbf{b})] \cos \Phi + [\mathbf{b} \times \mathbf{P}_0] \sin \Phi. \quad (11)$$

This equation corresponds to the inhomogeneous classical precession of the polarization vector \mathbf{P}_i around the vector \mathbf{b} . The direction of this vector, as well as the total precession phase $\Phi(\xi)$, should be found via the routine proposed above. The result of Eq. (11) should be averaged over the field variation across the beam. This would result in depolarization.

As a conclusion one should admit that the Zeeman splitting of the spin states, in the local (on a scale of l_c) magnetic field, results in a rather small difference in the wave vectors $k_{\pm} = \{k_0^2 \mp p_M^2(\xi)\}^{1/2}$ of the neutron wave components with the positive, or negative spin projections onto the local field direction. The associated slight difference in the phase velocities $v_{\pm} = \hbar k_{\pm}/m$ and coherent transitions between those components in a smoothly varying field are resumed into the Larmor precession, but not into the splitting of the beam in space. All the consideration above can, word by word, be repeated for the reflected or scattered wave $|\Psi(\mathbf{r}')\rangle$, given by the equation

$$|\Psi(\mathbf{r}')\rangle = |\Psi(\xi')\rangle = \hat{\mathcal{S}}(\xi', \boldsymbol{\sigma}) |\Psi^i(0)\rangle, \quad (12)$$

similar to Eq. (3), in which $|\Psi^i(0)\rangle$ is substituted by the vector of spin states in the reflected (scattered) wave just above the surface, \mathbf{k}_0 is changed for the outgoing wave vector \mathbf{k}_0^f , $\xi' = (\mathbf{k}_0^f \mathbf{r}')/k_0^f \geq 0$ and \mathbf{r}' pointing from the sample position to the direction of observation.

3. Refraction, reflection and transmission

The situation is changing drastically at the surface, which causes rapid variation of the potential

$\hat{V}(\mathbf{r})$ along the z -coordinate normal to the surface and almost perpendicular to the beam at shallow incidence. If this variation occurs on a scale much smaller than l_c then the potential gradient in this direction can be considered as an infinite and this corresponds to the definition of an ideal surface. Such a surface can be regarded as ideally flat, if on the scale greater than $l_{||}$, the interaction operator in Eq. (1) is $\hat{V}(\mathbf{r}) \approx \hat{V}_0(z)$. Then, due to the translational invariance of Eq. (1) along the surface and negligible splitting in the wave numbers $\pm k_0$, the solution of Eq. (1) just above the film ($z < 0$) is to be written as

$$|\Psi(\mathbf{r})\rangle = \exp(i\boldsymbol{\kappa}\boldsymbol{\rho}) \{ e^{ip_0 z} + \hat{R} e^{-ip_0 z} \} |\psi^i(0)\rangle, \quad (13)$$

where $\boldsymbol{\kappa}$ is the in-plane (conserving) projection of wave vector \mathbf{k} , $\boldsymbol{\rho}$ is the lateral coordinate, $p_0 = \sqrt{k_0^2 - \kappa^2}$ is the normal to the surface component of the wave vector \mathbf{k}_0 , \hat{R} is the reflectance matrix, which transforms two-spin components of the incoming wave function $|\psi^i(0)\rangle$ into the components of the reflected wave $|\psi^r(0)\rangle = \hat{R} |\psi^i(0)\rangle$. Inside the material the exponential factor of the wave function $|\Psi(\mathbf{r})\rangle$ is exactly the same, as in Eq. (13), while the vector of spin states $|\psi^i(0)\rangle$ at the front face of the film is transformed into the vector

$$|\psi(z)\rangle = \hat{\mathcal{S}}_{\perp}(z, \boldsymbol{\sigma}) |\psi^i(0)\rangle, \quad (14)$$

of states inside the film ($z > 0$). The $\hat{\mathcal{S}}_{\perp}$ -matrix has, however, a form different from that in Eq. (4). In the simplest case of a homogeneous medium $\hat{V}_0(z) = \hat{V}_0$ at $z > 0$, from Eqs. (1), (2), (13) and (14) it immediately follows that the $\hat{\mathcal{S}}_{\perp}$ -matrix is written as

$$\hat{\mathcal{S}}_{\perp}(z, \boldsymbol{\sigma}) = \hat{t} e^{i\hat{\phi}(z)} + \hat{r} e^{-i\hat{\phi}(z)}, \quad (15)$$

where $\hat{\phi}(z) = \hat{p}z$. The “transmission”, \hat{t} , and “reflection”, \hat{r} , operators are to be found from boundary conditions, while the wave vector operator $\hat{p} = \sqrt{p_0^2 - \hat{p}_c^2}$ is determined by Eqs. (1), (2), where $\hat{p}_c^2 = p_N^2 + \hat{p}_M^2$, $\hbar^2 \hat{p}_c^2 = 2m\hat{V}_0$, $\hbar^2 \hat{p}_M^2 = 2m\hat{\boldsymbol{\mu}}\mathbf{B}$, $\mathbf{B} = \mathbf{H} + 4\pi\mathbf{M}$ is the mean magnetic field, \mathbf{M} is the mean magnetization (in-plane component), $p_N^2 = 4\pi n\bar{b}_N$, and \bar{b}_N is the nuclear scattering length density. The operator \hat{p}_c is diagonal in the representation with the quantization axis along \mathbf{B} , and its eigenvalues are $p_{\pm}^{\pm} = \sqrt{p_0^2 - p_N^2 \mp p_M^2}$, where $p_M^2 = 4\pi n\bar{b}_M$, and $\bar{b}_M = (m/2\pi)\boldsymbol{\mu}\mathbf{B}$ is the magnetic

scattering length density of the homogeneous field \vec{B} .

We see that the presence of the sharp interface produces, in contrast to the smooth field variation, a dramatic effect at small p_0 . The wave at the surface is birefringent, and the two different spin components have different wave vector projections normal to the surface, while their lateral projections are equal. This situation is just opposite to the Larmor precession case. If, in particular, one of the normal projections becomes zero and the corresponding spin component is totally reflected then the beam inside the material is completely polarized along the mean-field direction.

The continuity conditions for the wave function at the front interface result in a pair of equations in a customary form

$$1 + \hat{R} = \hat{t} + \hat{r} \quad \text{and} \quad (1 - \hat{R})p_0 = (\hat{t} - \hat{r})\hat{p}. \quad (16)$$

To complete the set of equations one, as usual, needs to match the wave function at the interface with that in the substrate. If it is non-magnetic, then the wave function in the substrate is $|\psi_t(z)\rangle = \hat{T} \exp\{ip_s(z - z_s)\}$, where \hat{T} is the transmittance matrix, z_s denotes an interface coordinate, $p_s = \{p_0^2 - \hat{p}_{cs}^2\}^{1/2}$, and \hat{p}_{cs} is the critical wave number of the substrate material. A final pair of equations also has a well-known form

$$\hat{T} = \hat{t}e^{i\hat{\phi}} + \hat{r}e^{-i\hat{\phi}} \quad \text{and} \quad \hat{T}p_s = (\hat{t}e^{i\hat{\phi}} + \hat{r}e^{-i\hat{\phi}})\hat{p}, \quad (17)$$

where $\hat{\phi} = \hat{p}d$, and d is the layer thickness.

Due to the fact, that all the matrices in Eqs. (15–17) commute, they are diagonal in the representation with the quantization axis along the vector $\mathbf{b}_0 = \vec{B}/|\vec{B}|$ and this set of equation has a rather simple solution:

$$\begin{aligned} R^\pm &= \Delta_\pm^{-1} \{ (p_0 - p^\pm)(p^\pm + p_s) \\ &\quad + (p_0 + p^\pm)(p^\pm - p_s)e^{2i\varphi^\pm} \}, \\ \Delta_\pm &= (p_0 + p^\pm)(p^\pm + p_s) + (p_0 - p^\pm)(p^\pm - p_s)e^{2i\varphi^\pm}, \\ t^\pm &= \frac{2p_0(p^\pm + p_s)}{\Delta_\pm}; \quad r^\pm = \frac{2p_0(p^\pm - p_s)e^{2i\varphi^\pm}}{\Delta_\pm}, \end{aligned} \quad (18)$$

$T^\pm = 4p_0p^\pm/\Delta_\pm$, $\varphi^\pm = p^\pm d$ and d in the film thickness.

If $d \ll l_c$, then due to the reasons discussed above, the beam transmitted into the nonmagnetic thick

substrate is not split, while inside the film there exist a pair of transmitted waves with different wave vectors p_\pm and, correspondingly, a pair of reflected ones. These waves are split in the reciprocal, but not in the real space. The eigenvalues R_\pm , T_\pm , t_\pm and r_\pm of corresponding matrices are simply the amplitudes of either reflected or transmitted waves with positive, or negative spin projections onto the vector \mathbf{b}_0 .

The situation is slightly more complicated for multilayers with N layers whose magnetization is not parallel to each other. In this case one should introduce a set of $\mathcal{S}_{n\perp}$ -matrices similar to Eq. (15), but with the matrices \hat{t}_n and \hat{r}_n , $\hat{\phi}_n(z) = \hat{p}_n(z - z_{n-1})$ marked by the index $1 \leq n \leq N$ numerating the layers. Here z_{n-1} is the coordinate of the interface between $(n-1)$ st and n th layers, $z_0 = 0$, $\hat{p}_n^2 = p_0^2 - \hat{p}_{nc}^2$ and \hat{p}_{cn} is a critical wave number matrix for n th layer.

Matching \mathcal{S}_\perp^n and \mathcal{S}_\perp^{n+1} matrices and their derivatives at the interfaces between neighboring layers, one derives a chain of coupled equations for the matrices \hat{t}_n and \hat{r}_n :

$$\begin{aligned} \hat{t}_n e^{i\hat{\phi}_n} + \hat{r}_n e^{-i\hat{\phi}_n} &= \hat{t}_{n+1} + \hat{r}_{n+1}, \\ (\hat{t}_n e^{i\hat{\phi}_n} - \hat{r}_n e^{-i\hat{\phi}_n})\hat{p}_n &= (\hat{t}_{n+1} - \hat{r}_{n+1})\hat{p}_{n+1}, \end{aligned} \quad (19)$$

where $\hat{\phi}_n = \hat{p}_n d_n$, and d_n is the n th layer thickness.

A solution of these equations can be found via the recursion Parrat procedure, or equivalently, via the supermatrix formalism [8,7]. It should be noticed that the matrices \hat{t}_n , \hat{r}_n and \hat{p}_n are, in general, not commutative, i.e. \hat{t}_n and \hat{r}_n are not diagonal in the representation with the quantization axis parallel to the in-layer field \vec{B}_n . However, if the partial $\hat{\mathcal{S}}_{n\perp}$ -matrix is written in a form similar to Eq. (5):

$$\hat{\mathcal{S}}_{n\perp} = \mathcal{S}_{n\perp} + (\boldsymbol{\sigma} \mathcal{S}_{n\perp}) \quad (20)$$

and the direction of the (complex) vector $\mathcal{S}_{n\perp}$ is found (via the supermatrix routine), then the $\hat{\mathcal{S}}_{n\perp}$ -matrix is diagonal in the coordinate system with the quantization axis parallel to this direction, different for each layer and varying with p_0 . This means, that reflection and transmission are accompanied by spin-flip transition between the states with the eigenvalues $p_n^\pm = \sqrt{p_0^2 - p_{Nn}^2 \mp p_{Mn}^2}$ of the wave vector operator \hat{p}_n . As a result of that, as well as of the interference between transmitted and

reflected waves (within the coherence length), the expectation value $\mathbf{P}_n = \langle |s| \rangle_n$, i.e., the in-layer “polarization” undergoes rather complicated evolution, which may be called “quantum Larmor precession”, but it does not resemble its classical analogue guided by Eq. (13).

Each of the matrices \hat{t}_n and \hat{r}_n can be represented similar to Eq. (20) and, in particular,

$$\hat{R} = R_0 + (\sigma \mathbf{R}), \quad (21)$$

where $R_0 = \text{Tr}\{\hat{R}/2\}$ and $\mathbf{R} = \text{Tr}\{\sigma \hat{R}/2\}$ are to be found via the supermatrix routine [8,7].

The reflectance matrix \hat{R} transforms the incident vector of states $|\psi^i(0)\rangle$ into the final vector $|\Psi^f(0)\rangle = \hat{R}|\psi^i(0)\rangle$, which is then guided to the point of observation by Eq. (12), and $|\Psi^f(\xi')\rangle = \hat{\mathcal{S}}(\xi')\hat{R}\hat{\mathcal{S}}(\xi)|\psi_0\rangle$. The polarization vector $\mathbf{P}_r(0) = \langle |s| \rangle_f$ of the reflected beam just above the surface, but beyond the interference zone (coherence length) with the incoming beam, can be not collinear with the polarization $\mathbf{P}_i(0)$ and with the external field. Then it experiences the Larmor precession, on the way to the analyzer accounted for in the equation

$$\mathbf{P}_r(\xi') = \frac{1}{2} \frac{\text{Tr}\{\hat{\rho}_i(\xi)\hat{R}^+ \sigma(\xi')\hat{R}\}}{\text{Tr}\{\hat{\rho}_i(\xi)\hat{R}^+ \hat{R}\}}, \quad (22)$$

where $\sigma(\xi') = \hat{\mathcal{S}}^+(\xi')\sigma\hat{\mathcal{S}}(\xi')$.

In fact, one can simply calculate the polarization vector $\mathbf{P}_r(0)$ and then rotate it around the unit vector \mathbf{b}' for the angle Φ' according to Eq. (11), but for the neutron path from the sample to the analyzer.

The ratio $\mathbf{P}_r(0) = \mathcal{P}(0)/\mathcal{R}(0)$ in Eq. (22) contains in the denominator the reflectivity

$$\begin{aligned} \mathcal{R} = & \{|R_0|^2 + |\mathbf{R}|^2\} + 2 \text{Re}\{(\mathbf{P}_i \mathbf{R})R_0^*\} \\ & + \text{Im}\{(\mathbf{P}_i [\mathbf{R} \times \mathbf{R}^*])\}, \end{aligned} \quad (23)$$

for the case with no polarization analysis. The nominator in Eq. (22) has the form

$$\begin{aligned} \mathcal{P}(0) = & \mathbf{P}_i \{|R_0|^2 - |\mathbf{R}|^2\} + 2 \text{Re}\{\mathbf{R}[R_0^* + (\mathbf{P}_i \mathbf{R}^*)]\} \\ & + 2 \text{Im}\{[\mathbf{P}_i \times \mathbf{R}]R_0^* - \frac{1}{2}[\mathbf{R} \times \mathbf{R}^*]\}. \end{aligned} \quad (24)$$

This equation contains three different types of terms. The first one corresponds to the component of the reflected polarization vector parallel to the

vector \mathbf{P}_i , while the others are either parallel or perpendicular to the vector \mathbf{R} , and in general, are not collinear with the incoming polarization. These components will be precessing in the field guiding the polarization to the analyzer, and finally one may detect some spin-flip signal, which originated not in the reflection process, but totally due to this precession. It is important to note that among different terms in Eq. (24) there are two which are independent of the incident polarization and describe part of the polarization arising at reflection. The most interesting is the last one, which is absent at a collinear magnetization arrangement, but which should be present if $[\mathbf{b}_n \times \mathbf{b}_m] \neq 0$.

In the collinear case $R_0 = \{R_+ + R_-\}/2$ and the vector $\mathbf{R} = \mathbf{b}_0\{R_+ - R_-\}/2$, where $R_\pm = |R_\pm| \exp(i\chi_\pm)$ are complex eigenvalues of the reflectance matrix $\hat{\mathcal{R}}$, $\mathbf{b}_0 = \hat{\mathbf{B}}/|\hat{\mathbf{B}}|$ is not necessarily collinear with \mathbf{P}_i . Indeed, in the case of domains larger than the lateral projection of the coherence length one should be able to calculate the reflectivity for each of them, and then average over the lateral distribution of the magnetization direction \mathbf{b}_0 . The reflectivity \mathcal{R} and the vector $\mathcal{P}(0)$ for each of domains looks quite simple:

$$\mathcal{R} = \frac{1}{2}\{|R_+|^2(1 + P_i^{\parallel}) + |R_-|^2(1 - P_i^{\parallel})\}, \quad (25)$$

$$\begin{aligned} \mathcal{P}(0) = & \frac{1}{2}\{\mathbf{b}_0[|R_+|^2(1 + P_i^{\parallel}) - |R_-|^2(1 - P_i^{\parallel})] \\ & + 2|R_+||R_-|[\mathbf{P}^{\perp} \cos \chi - \mathbf{P}^a \sin \chi]\}, \end{aligned} \quad (26)$$

where $P_i^{\parallel} = (\mathbf{P}_i \mathbf{b}_0) = P_i \cos \gamma$ is the incoming polarization vector projection onto the direction \mathbf{b}_0 , $\mathbf{P}^{\perp} = [\mathbf{P}_i - \mathbf{b}_0(\mathbf{P}_i \mathbf{b}_0)]$ is the a component of \mathbf{P}_i perpendicular to \mathbf{b}_0 , $\mathbf{P}^a = [\mathbf{P}_i \times \mathbf{b}_0]$ is a vector perpendicular to both the incoming polarization and the field, and $\chi = \chi_+ - \chi_-$ is a phase shift. The polarization at reflection \mathbf{P}_r is then defined as a ratio $\mathbf{P}_r = \langle \mathcal{P}(0) \rangle_\gamma / \langle \mathcal{R} \rangle_\gamma$ of the quantities given in Eqs. (25) and (26) averaged over the angle γ .

4. Diffraction and off-specular scattering

All the consideration above was essentially based on the assumption that the domain size is greater than l_{\parallel} . This assumption should be proven by absence of scattering in off-specular directions. If it is detected then one may expect that there exists

a certain amount of small domains or of other magnetic inhomogeneities smaller than l_{\parallel} . A presence of domains can also affect the grazing incidence magnetic diffraction on the crystalline structure.

If off-specular scattering is small with respect to the incoming intensity, then it can be regarded as a perturbation and accounted for in the framework of the distorted wave Born approximation (DWBA) using the reference wave functions from the previous Section. Due to the perturbation interaction potential operator $\hat{\mathcal{V}}(\mathbf{r}) = \hat{V}(\mathbf{r}) - \hat{V}_0(z)$, where $\hat{V}_0(z) = \langle \hat{V}(\mathbf{r}) \rangle_{\rho}$ is operator averaged over the lateral coordinate ρ , the initial state with the wave vector $\mathbf{k}^i = \{\kappa^i; p_{\pm}^i\}$ may be scattered into the state with the wave vector $\mathbf{k}^f = \{\kappa^f; p_{\pm}^f\}$, where $p_{\pm}^{i,f}$ are defined by the same equations as p_{\pm} with p_0 substituted by the normal to the surface components $p_0^{i,f}$ of the initial or final wave vectors $\mathbf{k}_0^{i,f} = \{\kappa^{i,f}; p_0^{i,f}\}$. These scattering processes may be accomplished by the transitions between neutron spin states.

Within the first order of DWBA the scattering amplitude $\hat{f}(\mathbf{k}^f, \mathbf{k}^i)$ (operator in spin space) is proportional to the matrix element: $\langle \Psi^f | \hat{\mathcal{V}}(\rho, z) | \Psi^i \rangle$, where the vectors $|\Psi^{i,f}\rangle = \exp(i\mathbf{\kappa}^{i,f} \rho) |\psi^{i,f}(z)\rangle$ and $|\psi^{i,f}(z)\rangle$ depend only on the coordinate z normal to the surface. Due to this factorization, the scattering amplitude is written as

$$f(\mathbf{Q}_{\parallel}; p^f, p^i) = A(\mathbf{Q}_{\parallel}) \langle \psi_0^f | \hat{F}^{fi} | \psi_0^i \rangle, \quad (27)$$

where $\mathbf{Q}_{\parallel} = \mathbf{\kappa}^f - \mathbf{\kappa}^i$ is the lateral momentum transfer, $A(\mathbf{Q}_{\parallel}) = \sum_j e^{i\mathbf{Q}_{\parallel} \rho_j}$, ρ_j are the lateral coordinates of atoms, and at $a_{\perp} p_0^{i,f} \ll 1$, $\hat{F}^{if} = \hat{F}(\mathbf{Q}_{\parallel}; p^i, p^f)$ is defined as

$$\hat{F}^{fi} = \sum_j \hat{\mathcal{S}}_{\perp}^f(z_j) \hat{F}(\mathbf{Q}_{\parallel}) \hat{\mathcal{S}}_{\perp}^i(z_j). \quad (28)$$

Here, $\hat{\mathcal{S}}_{\perp}(z)$ -matrices are given in Eq. (15), $z_j = a_{\perp} j$ are atomic coordinates and a_{\perp} is the unit cell constant in the direction perpendicular to the surface, $\hat{F}(\mathbf{Q}_{\parallel}) = \hat{F}(\mathbf{Q}_{\parallel}, 0) \approx \hat{F}(\mathbf{Q})$, and

$$\hat{F}(\mathbf{Q}) = F_N(\mathbf{Q}) + (\sigma \mathbf{m}_{\perp}) F_M(\mathbf{Q}). \quad (29)$$

In Eq. (29), $F_N = b_N \mathcal{F}_N(\mathbf{Q})$ is a product of the nuclear scattering length b_N and the nuclear structure factor $\mathcal{F}_N(\mathbf{Q})$, while $F_M = b_M \mathcal{F}_M(\mathbf{Q})$ is a product of the magnetic scattering length b_M and the

magnetic cell form factor $\mathcal{F}_M(\mathbf{Q})$, $\mathbf{m}_{\perp} = \mathbf{m} - e(\mathbf{em})$ is the component of the unit vector $\mathbf{m} = \mathbf{M}/M$ perpendicular to the momentum transfer direction given by the unit vector $\mathbf{e} = \mathbf{Q}/|Q|$.

Substitution of Eq. (15) into Eq. (28) yields

$$\begin{aligned} \hat{F}^{fi} = & \frac{1}{4} \{ G_{++} [1 + \sigma \mathbf{b}] \hat{F}(\mathbf{Q}) [1 + \sigma \mathbf{b}] \\ & + G_{+-} [1 + \sigma \mathbf{b}] \hat{F}(\mathbf{Q}) [1 - \sigma \mathbf{b}] \\ & + G_{-+} [1 - \sigma \mathbf{b}] \hat{F}(\mathbf{Q}) [1 + \sigma \mathbf{b}] \\ & + G_{--} [1 - \sigma \mathbf{b}] \hat{F}(\mathbf{Q}) [1 - \sigma \mathbf{b}] \}, \quad (30) \end{aligned}$$

where $G_{\mu\nu} = G_{\mu\nu}^{fi}$, $\mu = \pm$, $\nu = \pm$, and

$$G_{\mu\nu}^{fi} = t_{\mu}^f t_{\nu}^i g_{\mu\nu}^{tt} + r_{\mu}^f r_{\nu}^i g_{\mu\nu}^{rr} + r_{\mu}^f t_{\nu}^i g_{\mu\nu}^{rt} + t_{\mu}^f r_{\nu}^i g_{\mu\nu}^{tr},$$

$$g_{\mu\nu}^{tt} = \frac{\exp[i(\phi_{\mu}^f + \phi_{\nu}^i)] - 1}{i(\phi_{\mu}^f + \phi_{\nu}^i)},$$

$$g_{\mu\nu}^{rr} = \frac{\exp[i(\phi_{\mu}^f - \phi_{\nu}^i)] - 1}{i(\phi_{\mu}^f - \phi_{\nu}^i)},$$

$$g_{\mu\nu}^{rt} = g_{\mu\nu}^{tt} \exp[-i(\phi_{\mu}^f + \phi_{\nu}^i)],$$

$$g_{\mu\nu}^{tr} = g_{\mu\nu}^{rr} \exp[-i(\phi_{\mu}^f - \phi_{\nu}^i)], \quad (31)$$

with $\phi_{\mu}^{(f,i)} = p_{\mu}^{(f,i)} a_{\perp} \ll 1$, $\phi_{\mu}^{(f,i)} = p_{\mu}^{(f,i)} d$.

The Laue functions $g_{\pm\pm}^{tt}$ correspond to non-spin-flip processes in which the wave with $+$ or $-$ spin projection is transmitted into the film, scattered by inhomogeneities, and the scattered wave is transmitted out of the film. The functions $g_{\pm\pm}^{rr}$ correspond to non-spin-flip transitions between reflected waves. If $d/a_{\perp} \gg 1$, then each of these functions reveal two sharp maxima at $p_{\pm}^f + p_{\pm}^i = 0$. Two other functions, $g_{\pm\mp}^{tt}$ and $g_{\pm\mp}^{rr}$, reach their maximum values d/a_{\perp} at $p_{\pm}^f + p_{\mp}^i$ corresponding to spin-flip scattering in off-specular directions. The other Laue functions $g_{\mu\nu}^{rt}$ and $g_{\mu\nu}^{tr}$ correspond to the processes with transitions between transmitted and reflected waves. They show maxima at $p_{\pm}^f - p_{\pm}^i$ (non-spin-flip), or at $p_{\pm}^f - p_{\mp}^i$ (spin-flip).

Calculating further products of the Pauli matrices in Eq. (30) one has

$$\hat{F}^{fi} = F_0 + (\mathbf{F}\sigma), \quad (32)$$

where $F_0 = \text{Tr}\{\hat{F}^{fi}/2\}$ and $\mathbf{F} = \text{Tr}\{\sigma \hat{F}^{fi}/2\}$.

If the coordinate system is fixed by three orthogonal vectors $\mathbf{b} = \mathbf{B}/|\mathbf{B}|$, $\mathbf{b}_{\perp} = \mathbf{m}_{\perp} - \mathbf{b}(\mathbf{m}_{\perp} \mathbf{b})$, and

$\mathbf{b}_a = [\mathbf{b} \times \mathbf{b}_\perp]$, then the vector \mathbf{F} is decomposed as follows:

$$\mathbf{F} = F_{\parallel} \mathbf{b} + F_{\perp} \mathbf{b}_\perp + F_a \mathbf{b}_a. \quad (33)$$

Finally, the zero matrix element F_0^{fi} and the components of the vector \mathbf{F}^{fi} can be written explicitly as

$$\begin{aligned} F_0 &= \frac{1}{2} \{ (G_{++} + G_{--}) F_N \\ &\quad + (G_{++} - G_{--}) F_M(\mathbf{b} \mathbf{m}_\perp) \}, \\ F_{\parallel} &= \frac{1}{2} \{ (G_{++} - G_{--}) F_N \\ &\quad + (G_{++} + G_{--}) F_M(\mathbf{b} \mathbf{m}_\perp) \}, \\ F_{\perp} &= \frac{1}{2} (G_{+-} + G_{-+}) F_M, \\ F_a &= \frac{1}{2} (G_{+-} - G_{-+}) F_M, \end{aligned} \quad (34)$$

where $(\mathbf{b} \mathbf{m}_\perp) = (\mathbf{b} \mathbf{m}) - (\mathbf{e} \mathbf{b})(\mathbf{m} \mathbf{e})$. From these equations it follows that the amplitudes F_0 and F_{\parallel} in Eq. (33) do not contain terms responsible for transitions between the states with p_+ and p_- , while the other two, F_{\perp} and F_a , correspond to purely magnetic scattering and provide such transitions.

Substitution of Eq. (33) into the definition of the polarization and scattering cross section

$$\begin{aligned} \frac{d\sigma}{d\Omega} &= \overline{\langle |f(\mathbf{k}^f, \mathbf{k}^i)|^2 \rangle}, \\ \frac{d\sigma}{d\Omega} \mathbf{P} &= \overline{\langle f^+(\mathbf{k}^f, \mathbf{k}^i) s f(\mathbf{k}^f, \mathbf{k}^i) \rangle}, \end{aligned} \quad (35)$$

immediately yields the final equations which become identical with Eq. (23) and (24), if R_0 and \mathbf{R} are substituted by F_0 and \mathbf{F} , respectively, and the additional factor $|A(\mathbf{Q}_{\parallel})|^2$ is introduced.

If the periodical crystalline lateral structure is regarded as a perturbation, then $|A(\mathbf{Q}_{\parallel})|^2 = (2\pi)^2 / s_0 \mathcal{N}_0 \delta(\mathbf{Q}_{\parallel} - \boldsymbol{\tau})$, where $\boldsymbol{\tau}$ is the in-plane reciprocal vector, s_0 is the area of the unit cell cross section by the film surface, $\mathcal{N} = S_0 / s_0$ and S_0 is the foot print of the beam.

In the saturated state and at the incident polarization parallel to the magnetization any spin-flip signal and, correspondingly, splitting of the diffracted beam is totally due to the local atomic field components perpendicular to the mean magnetic field direction. Then the effect is usually small due to the atomic magnetic form factor $\mathcal{F}_M(\tau \neq 0) \ll 1$

[11]. All the considerations above are actually valid for a large domain, and in multidomain samples, the scattering cross section and polarization should be averaged over domains. This will dramatically increase the spin-flip signal at specular reflection and at diffraction, but it does not increase a contribution from the two last terms in Eq. (33) responsible for the spatial splitting of the diffracted beam. If, on the contrary, there exists an amount of small domains, then the splitting is determined by the mean magnetic field, $\bar{\mathbf{B}}$ averaged over domains. It may be slightly smaller, than at saturation, but the transitions between the states with p_+ and p_- are more efficiently provided by the domain field components perpendicular to $\bar{\mathbf{B}}$, than by the local atomic scale fields [11]. In this case, magnetic scattering contributes not only into the true diffraction, but rather to diffuse scattering concentrated around the Bragg peak positions. In particular, relatively small domains (on a scale of large lateral coherence length) create low angle scattering manifested in off-specular directions. This can, in principle, explain the experiments [17–18]. Corresponding equations have exactly the same structure as the equation for the Bragg diffraction discussed above. The only difference is that the $\delta(\mathbf{Q}_{\parallel} - \boldsymbol{\tau})$ function should be substituted for the lateral domain form factor [16] $\langle |A(\mathbf{Q}_{\parallel})|^2 \rangle_{\text{domains}}$.

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