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Off-specular polarized neutron scattering from magnetic fluctuations in thin films and multilayers

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Abstract. Polarized neutron off-specular scattering from magnetic fluctuations (magnetic domains, roughnesses, and dynamical spin correlations) in thin films and multilayers is theoretically considered within the super-matrix approach of the distorted wave Born approximation. General equations relating the scattering cross section with the pair spin correlation function, the correlator of the nuclear scattering potential fluctuations, and the magnetic–nuclear cross correlator are derived and brought into a form easy for applications.

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One of the greatest advantages of neutron scattering for a wide range of problems in magnetism is the transparency of the data interpretation and the reliability of the quantitative analysis. Relatively weak interactions with matter often guarantee a single-event scattering process, which can be described within the framework of the Born approximation (BA). Then, the magnetic scattering cross section is simply proportional to the Fourier transform of a known combination of the pair spin correlation tensor (related to the dynamical susceptibility) components. Explicit equations derived in the BA [1–4] for the neutron polarization at magnetic scattering have allowed the invention of the vector or 3D polarization analysis [5, 6], widely employed [7] not only to separate nuclear scattering from magnetic scattering and from their interference, but also to determine different components of the susceptibility tensor, as well as to distinguish between static and dynamical correlations. In thick crystals the BA may not be valid [8], and then one needs to develop a dynamical scattering theory of polarized neutrons [9], which appears to be cumbersome and quite difficult in practical use. For scattering at grazing incidence on a flat surface the BA also breaks down, but the situation is handled much more easily. In this case interaction with the mean potential averaged over the lateral projection of the neutron coherence length is strong and leads to the optical effects: specular reflection and refraction, which are exactly accounted for. According to the neutron spin states the exact solution incorporates birefringence [10–12] of the neutron wave refracted into the homogeneous magnetic media. Lateral inhomogeneities of the interaction potential giving rise to off-specular scattering are considered as a perturbation and described within the distorted wave Born approximation (DWBA) [13]. Examples of these inhomogeneities can include interfacial roughnesses [14], magnetic domains [15], inclusions [16], thermal spin fluctuations, or even lateral crystalline structure [10]. Each violates the translational invariance of the system with respect to the lateral shift and causes off-specular scattering, which in magnetic materials may be associated with spin-flip processes. This paper is devoted to the derivation of the explicit equations for (spin-flip and non-spin-flip) scattering cross sections. It generalizes earlier results [11, 12, 17, 18] used in [10, 15, 16, 19–23] to treat various experimental situations.

1 Refraction, transmission, and reflection

The neutron interaction with a magnetic material is formally described by the operator $\hat{V}(\boldsymbol{r}, t) = \hat{1} V_{\mathrm{N}}(\boldsymbol{r}, t) + \hat{\boldsymbol{\mu}} \boldsymbol{B}(\boldsymbol{r}, t)$, where $V_{\mathrm{N}}(\boldsymbol{r}, t)$ is the nuclear scattering potential, $\boldsymbol{B}(\boldsymbol{r}, t)$ is the microscopic magnetic field, $\boldsymbol{\mu} = \mu \hat{\boldsymbol{\sigma}}$, μ is the neutron magnetic moment, $\hat{\boldsymbol{\sigma}}$ is the vector of the Pauli matrices, and $\hat{1}$ is a unit matrix in the neutron spin, $s = \hat{\boldsymbol{\sigma}}/2$, space. A neutron wave impinging onto a surface at the angle α_i of grazing incidence averages out most of the microscopic details of the interaction potential $\hat{V}(\boldsymbol{r}, t)$ over the lateral projection $l_{\parallel} \sim l_{\mathrm{c}} / \sin \alpha_i$ of the coherence length $l_{\mathrm{c}} \sim \lambda / \delta\theta$, which is mostly related to the primary divergence $\delta\theta$ of the beam (with the wavelength λ). As far as the mean value $\langle \hat{V}(\boldsymbol{r}, t) \rangle_{\boldsymbol{\rho}} = \hat{V}(z)$ is a function of the coordinate z normal to the surface and independent of the lateral projection ρ of the neutron coordinate $\boldsymbol{r}$, the solution of the Schrödinger equation with $\hat{V}(z)$ is factorized into the product $|\Psi(\boldsymbol{r}, t)\rangle = \mathrm{e}^{-\mathrm{i}Et} \mathrm{e}^{\mathrm{i}\boldsymbol{\kappa}\boldsymbol{\rho}} |\psi(z)\rangle$, where $\boldsymbol{\kappa}$ is the lateral projection of the wave vector $\boldsymbol{k}$, $2mE = \hbar^2 k^2$, m is the neutron mass, and $|\psi(z)\rangle$ is the two-component vector in the neutron spin

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space. In the layered structure $\hat{V}(z) = \sum_n \hat{V}_n(z - z_{n-1})$ and $|\psi(z)\rangle = \sum_n |\psi_n(z - z_{n-1})\rangle$, where n enumerates the layers, $z_{n-1} \le z \le z_n$, and $z_0 = 0$. If $\hat{V}_n(z - z_{n-1}) = \hat{V}_n$ is independent of z, then $|\psi_n\rangle = \hat{S}_n(z - z_{n-1})|\psi_0\rangle$, where $|\psi_0\rangle$ is the vector of the initial states, and the $\hat{S}_n$-matrix is a linear combination of the operators:

$$\hat{S}_n(z - z_n) = \mathrm{e}^{\mathrm{i}\hat{p}_n(z - z_{n-1})}\hat{A}^t_n + \mathrm{e}^{-\mathrm{i}\hat{p}_n(z - z_{n-1})}\hat{A}^r_n \,. \quad (1)$$

It develops $|\psi_0\rangle$ into the vector of neutron states within the nth layer. The refraction effects are taken into account by the operator $\hat{p}_n = \sqrt{\hat{p}_0^2 - \hat{p}_{nc}^2}$, where $\hbar^2\hat{p}_{nc}^2 = 2m\hat{V}_n$, $\hat{p}_0 = \hat{1}p_0$, and $p_0 = k\sin\alpha$ is the normal component of the wave vector $\boldsymbol{k}$. If the quantization axis is chosen along with the mean magnetic field $\boldsymbol{B}_n$ inside the layer, then $\hat{p}_n$ is a diagonal 2×2 matrix with the eigenvalues $p_n^{\pm} = \sqrt{p_0^2 - p_{n\pm}^2}$, where $p_{n\pm}^2 = p_{n\mathrm{N}}^2 \pm p_{n\mathrm{M}}^2$, $p_{n\pm}$ are the critical wave numbers for the total reflection of the positive or negative spin projection onto the $\boldsymbol{B}_n$ direction, and $p_{n\mathrm{N}}^2 = 4\pi(Nb)_{n\mathrm{N}}$, $p_{n\mathrm{M}}^2 = 4\pi(Nb)_{n\mathrm{M}}$ and $(Nb)_{n\mathrm{N}}$, $(Nb)_{n\mathrm{M}q}$ are the nuclear and magnetic scattering length densities, respectively. If the vectors $\boldsymbol{B}_n$ are not collinear, then 2×2 matrices of transmittance, $\hat{A}^t_n$, and reflectance, A^r_n, do not commute with the wave-number matrix $\hat{p}_n$ and can be computed via the super-matrix routine [11, 12].

2 Neutron scattering cross section in the DWBA

Interaction with the mean multilayer structure manifests itself in only specular reflection from and transmission through the sample. If the system contains magnetic domains laterally extended beyond the range of $l_\|$ they can provide a spin-flip contribution to the specular reflection and transmission, which should be calculated for each of the domains, and the result should be averaged over their distribution. If, on the contrary, inhomogeneities are relatively small, then the effect of the residual part $\hat{\tilde{V}}(r, t) = \hat{V}(\boldsymbol{r}, t) - \hat{V}(z)$ of the interaction operator can be accounted for as a perturbation for the neutron states found in the previous section. This perturbation causes off-specular scattering with the double-differential cross section given by the standard equation [1]:

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\omega\mathrm{d}\Omega} = C\overline{|\langle\Psi^f|\hat{\tilde{V}}_{ll'}(r)|\Psi^i\rangle|^2}\delta(\omega - \varepsilon_{l'} + \varepsilon_l) \,, \quad (2)$$

where ω is the energy transfer, $C = (k^f/k^i)(m/2\pi\hbar^2)^2$, and averaging runs over neutron states and over the specimen states l and l' with energies ε_l and $\varepsilon_{l'}$, respectively. In the DWBA the vectors $|\Psi^{f(i)}\rangle$ of the final, f (or initial, i), neutron states are to be calculated as indicated in the previous section with the wave vector $\boldsymbol{k} = \{\boldsymbol{\kappa};\, p_0\}$ substituted for $\boldsymbol{k}_{f(i)} = \{\boldsymbol{\kappa}_{f(i)};\, p_{f(i)}\}$.

This immediately yields:

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\omega\mathrm{d}\Omega} = \frac{C}{2\pi\hbar}\mathrm{Tr}\int \mathrm{d}t\, \mathrm{e}^{-\mathrm{i}\omega t}\left(\hat{\rho}_i \hat{\tilde{V}}^{fi+}(t)\hat{\rho}_f \hat{\tilde{V}}^{fi}(0)\right) , \quad (3)$$

where $\hat{\rho}_{f(i)} = \overline{|\psi_{f(i)}\rangle\langle\psi_{f(i)}|} = [\hat{1} + \hat{\boldsymbol{\sigma}}\boldsymbol{P}_{f(i)}]/2$ is the density matrix of the final (initial) neutron state, $\boldsymbol{P}_i$ is the incoming beam polarization vector, and $\boldsymbol{P}_f$ is the vector of the polarization analysis efficiency. In (3) $\hat{\tilde{V}}^{fi}(t) = \sum_n \hat{\tilde{V}}^{fi}_n(t)$, where $\hat{\tilde{V}}^{fi}_n(t) = \hat{\tilde{V}}_n(\boldsymbol{q}_\|,\, p_f,\, p_i;\, t)$ is a function of the lateral momentum transfer $\boldsymbol{q}_\| = \boldsymbol{\kappa}_f - \boldsymbol{\kappa}_i$, and of the normal to the surface projections p_i and p_f of the incoming and outgoing wave vectors, respectively:

$$\hat{\tilde{V}}^{fi}_n(t) = \int \mathrm{d}\boldsymbol{\rho}\, \mathrm{e}^{\mathrm{i}\boldsymbol{q}_\|\boldsymbol{\rho}} \int_0^{d_n} \mathrm{d}z\, \hat{S}^f_n(z)\hat{\tilde{V}}(\boldsymbol{\rho}, z;\, t)\hat{S}^i_n(z) \,, \quad (4)$$

where d_n is the layer thickness. Taking into account (1), the integration over coordinates in (4) can readily be performed, and

$$\hat{\tilde{V}}^{fi}_n(t) = \hat{A}^{f\alpha}_n \hat{\Lambda}^\mu_n \hat{\tilde{V}}_n(\boldsymbol{q}_\|,\, p^{\alpha\mu}_{nf} - p^{\beta\nu}_{ni};\, t)\hat{\Lambda}^\nu_n \hat{A}^{i\beta}_n \,, \quad (5)$$

where $\alpha = t, r$, $\beta = t, r$, $\mu = \pm$, $\nu = \pm$, and summation over the repeating twice indexes is anticipated (the Einstein rule), $\hat{\Lambda}^\pm_n = [\hat{1} \pm \hat{\boldsymbol{\sigma}}\boldsymbol{b}_n]/2$, $\boldsymbol{b}_n = \boldsymbol{B}_n/B_n$, $p^{t\pm}_{nf(i)} = \sqrt{p^2_{f(i)} - p^2_{n\pm}}$ represent two eigenvalues of the normal projections of the outgoing (incoming) wave vector in the nth layer, and $p^{r\pm}_{nf(i)} = -p^{t\pm}_{nf(i)}$.

If $\hat{\tilde{V}}_n(r, t) = \hat{1}\tilde{V}_{\mathrm{N}n}(\boldsymbol{r}, t) + \hat{\boldsymbol{\sigma}}\tilde{\boldsymbol{b}}_n(\boldsymbol{r}, t)$, then (5) allows separation of different contributions in the scattering cross section in (3), giving the result:

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\omega\mathrm{d}\Omega} = C\sum_{nn'}\{I^{\mathrm{M}}_{nn'} + I^{\mathrm{N}}_{nn'} + I^{\mathrm{NM}}_{nn'}\} \,, \quad (6)$$

where I^{N} and I^{M} respectively describe nuclear and magnetic off-specular scattering, while I^{NM} refers to their interference. Two first terms in (6) can be expressed via corresponding correlation functions of fluctuations of the magnetic or nuclear scattering potential, while the last is related to the correlations between these two types of fluctuations:

$$I^{\mathrm{M}}_{nn'} = \mu^2 T^{\{\zeta\}}_{ifnn'}\langle\tilde{\boldsymbol{B}}^\xi_n(\boldsymbol{q}_\|,\, q^{\{\eta\}}_n,\, t)\tilde{\boldsymbol{B}}^{\xi'+}_{n'}(\boldsymbol{q}_\|,\, q^{\{\eta'\}}_{n'},\, 0)\rangle_\omega \,, \quad (7)$$

$$I^{\mathrm{N}}_{nn'} = T^{\{\phi\}}_{ifnn'}\langle\tilde{V}_{\mathrm{N}n}(\boldsymbol{q}_\|,\, q^{\{\eta\}}_n,\, t)\tilde{V}^+_{\mathrm{N}n'}(\boldsymbol{q}_\|,\, q^{\{\eta'\}}_{n'},\, 0)\rangle_\omega \,, \quad (8)$$

$$\begin{aligned} I^{\mathrm{MN}}_{nn'} &= \mu T^{\{\chi\}}_{ifnn'}\langle\tilde{\boldsymbol{B}}^\xi_n(\boldsymbol{q}_\|,\, q^{\{\eta\}}_n,\, t)\tilde{V}^+_{\mathrm{N}n'}(\boldsymbol{q}_\|,\, q^{\{\eta'\}}_{n'},\, 0)\rangle_\omega \,, \\ &+ \mu T^{\{\theta\}}_{ifnn'}\langle\tilde{V}_{\mathrm{N}n}(\boldsymbol{q}_\|,\, q^{\{\eta\}}_n,\, t)\tilde{\boldsymbol{B}}^{\xi'+}_{n'}(\boldsymbol{q}_\|,\, q^{\{\eta'\}}_{n'},\, 0)\rangle_\omega \,. \end{aligned} \quad (9)$$

Here the transverse momentum transfer $q^{\{\eta\}}_n = p^{\alpha\mu}_{nf} - p^{\beta\nu}_{ni}$ is represented in the tensor form, accounting for all possible spin-flip and non-spin-flip transitions between transmitted and reflected waves inside the layer. The superscript notation $\{\zeta\}$, $\{\phi\}$, $\{\chi\}$, and $\{\theta\}$ becomes clear if the tensors depending on the density matrices and reflection–transmission amplitudes are written explicitly:

$$T^{\{\zeta\}}_{ifnn'} = \mathrm{Tr}\{\hat{\rho}^{\beta'\beta}_{in'n}\hat{\Gamma}^{\nu\xi\mu}_n\hat{\rho}^{\alpha\alpha'}_{fnn'}\hat{\Gamma}^{\mu'\xi'\nu'}_{n'}\} \,, \quad (10)$$

$$T^{\{\phi\}}_{ifnn'} = \mathrm{Tr}\{\hat{\rho}^{\beta'\beta}_{in'n}\hat{\Gamma}^{\nu\mu}_n\hat{\rho}^{\alpha\alpha'}_{fnn'}\hat{\Gamma}^{\mu'\nu'}_{n'}\} \,, \quad (11)$$

$$T^{\{\theta\}}_{ifnn'} = \mathrm{Tr}\{\hat{\rho}^{\beta'\beta}_{in'n}\hat{\Gamma}^{\nu\xi\mu}_n\hat{\rho}^{\alpha\alpha'}_{fnn'}\hat{\Gamma}^{\mu'\nu'}_{n'}\} \,, \quad (12)$$

$$T^{\{\chi\}}_{ifnn'} = \mathrm{Tr}\{\hat{\rho}^{\beta'\beta}_{in'n}\hat{\Gamma}^{\nu\mu}_n\hat{\rho}^{\alpha\alpha'}_{fnn'}\hat{\Gamma}^{\mu'\xi'\nu'}_{n'}\} \,, \quad (13)$$

where $\hat{\Gamma}^{\nu\xi\mu}_n = \hat{\Lambda}^\nu_n\hat{\sigma}^\xi\hat{\Lambda}^\mu_n$, $\hat{\Gamma}^{\nu\mu}_n = \hat{\Lambda}^\nu_n\hat{\Lambda}^\mu_n$, and

$$\hat{\rho}^{\beta'\beta}_{in'n} = \hat{A}^{\beta'}_{in'}\hat{\rho}_i[\hat{A}^\beta_{in}]^+ \,;\; \hat{\rho}^{\alpha\alpha'}_{fnn'} = [\hat{A}^\alpha_{fn}]^+\hat{\rho}_f[\hat{A}^{\alpha'}_{fn}]^+ \,. \quad (14)$$

At first sight, these equations look quite complicated. However, as soon as the amplitude matrices are represented as $\hat{A}_n^\alpha = [\hat{1}A_n^\alpha + \hat{\boldsymbol{\sigma}}\boldsymbol{A}_n^\alpha]/2$ and the quantities $A_n^\alpha = \mathrm{Tr}\hat{A}_n^\alpha$ and $\boldsymbol{A}_n^\alpha = \mathrm{Tr}(\hat{\boldsymbol{\sigma}}\hat{A}_n^\alpha)$ are found, the traces can easily be calculated either analytically (and expressed via a combination of $\boldsymbol{P}_{f(i)}$, $\boldsymbol{b}_n$, A_n^α, and $\boldsymbol{A}_n^\alpha$ [11]), or computed numerically [15, 18, 21–23].

Since all information about the neutron spin behavior in the mean multilayer structure is absorbed in (10)–(13), one can concentrate on the functions around the mean value. Corresponding quantities enter (7)–(9) in the form of the Fourier transforms:

$$\tilde{\boldsymbol{B}}_n(\boldsymbol{q}_\parallel, q_n^{\{\eta\}}, t) = \int_0^{d_n} \mathrm{d}z\, \mathrm{e}^{\mathrm{i}q_n^{\{\eta\}}z}\, \tilde{\boldsymbol{B}}_n(\boldsymbol{q}_\parallel, z, t)\,, \tag{15}$$

$$\tilde{V}_{\mathrm{N}n}(\boldsymbol{q}_\parallel, q_n^{\{\eta\}}, t) = \int_0^{d_n} \mathrm{d}z\, \mathrm{e}^{\mathrm{i}q_n^{\{\eta\}}z}\, \tilde{V}_{\mathrm{N}n}(\boldsymbol{q}_\parallel, z, t)\,. \tag{16}$$

Further calculations, as usual, require that or another model for correlations of fluctuations in real space:

$$\langle \tilde{B}_n^\xi(\boldsymbol{\rho}, z, t)\tilde{B}_{n'}^{\xi'}(0, z', 0)\rangle\,, \tag{17}$$

$$\langle \tilde{B}_n^\xi(\boldsymbol{\rho}, z, t)\tilde{V}_{\mathrm{N}n'}(0, z', 0)\rangle\,, \tag{18}$$

$$\langle \tilde{V}_{Nn}(\boldsymbol{\rho}, z, t)\tilde{V}_{\mathrm{N}n'}(0, z', 0)\rangle\,, \tag{19}$$

which are in a known way related to the atomic pair spin correlation function, the correlator of nuclear density fluctuations, and the cross correlator of those two types of fluctuations. This separate task is beyond the scope of the paper, while some examples can be found in [15, 18, 21–23].

3 Conclusions

In conclusion, it is important to note that the BA definitely fails either in the vicinity of the total reflection for incoming (outgoing) wave vectors, or in the range of the Bragg reflection from the mean multilayer structure. Then one can use the DWBA, which automatically takes into account the neutron spin behavior during coherent neutron wave propagation inside a magnetic multilayer. This behavior is, however, rather complex, especially in the case of a non-collinear layer magnetization arrangement. In the present paper it is shown that, nonetheless, the problem of calculations of the polarized neutron cross section of off-specular neutron scattering can be reduced to a modeling of a pair spin correlation function, i.e. to the general problem in the polarized neutron data interpretation, independent of the fact whether the BA is valid or not.

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