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A common optical algorithm for the evaluation of specular spin polarized neutron and Mössbauer reflectivities

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Abstract

Using the general approach of Lax for multiple scattering of waves a 2×2 covariant expression for the reflectivity of polarized slow neutrons of a magnetic layer structure of arbitrary complexity is given including polarization effects of the external magnetic field. The present formalism is identical to the earlier published one for the (nuclear) resonant X-ray (Mössbauer) reflectivity and properly takes the effect of the external magnetic field of arbitrary direction on the neutron beam into account. The form of the reflectivity matrix allows for an efficient numerical calculation. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

The detectable information on a thin or stratified structure by the reflectometric techniques is the one-dimensional scattering amplitude density profile perpendicular to the surface, which in turn can be related to the chemical/isotopic/magnetic, etc. profile within the penetration depth of the corresponding radiation. X-ray and neutron reflectometry, therefore, have become standard tools in studying surfaces and thin films. In nonresonant X-ray or unpolarized neutron reflectometry, the scattering processes being independent of the polarization of the incident wave, any stratified medium can be described by a scalar complex index of refraction. There are other important cases, however, in which the scattering medium is birefringent for the corresponding radiation, and the polarization-dependent multiple scattering leads to non-scalar optics. These cases include polarized neutron reflectometry (PNR) and (synchrotron) Mössbauer reflectometry (SMR), the latter being only a special but well-studied case of the anisotropic (resonant) X-ray scattering problem. Beyond the trivial analogy between the scalar cases of neutron and X-ray multiple scattering, the generalization to polarization-dependent scattering of *any* waves [1] is not straightforward and in fact, as we point out below, it cannot be performed in general. It is the purpose of this paper to show that, indeed, such analogy, i.e. a common optical formalism exists for the anisotropic neutron and anisotropic nuclear resonant

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X-ray transmission and reflection for the case of forward scattering and that of grazing incidence.

2. General considerations

In this section, starting from the general theory of Lax [1], we shall obtain some general formulae for the scattering of multicomponent waves. Description of the various theories of scattering processes on a single scatterer lead to an inhomogeneous wave equation

$$[(\Delta + k^2)I - U(\boldsymbol{r})]\Psi_1(\boldsymbol{r}) = 0, \quad (1)$$

where k is the vacuum wave number, I is the unit matrix, $U(\boldsymbol{r})$ is the scattering potential and $\Psi_1(\boldsymbol{r})$ is the amplitude of the scattered wave, an electromagnetic field vector or quantum mechanical spinor state. For many scattering centers the coherent field fulfils the

$$[(\Delta + k^2)I + 4\pi Nf\,]\Psi(\boldsymbol{r}) = 0, \quad (2)$$

three-dimensional wave equation, where f is the coherent forward scattering amplitude, N is the density of the scattering centers per unit volume and $\Psi(\boldsymbol{r})$ is the coherent field defined by an average of the field vectors over the positions and states of the scattering centers [1]. Eq. (2) shows that from the point of view of the coherent field the system of randomly distributed scattering centers can be replaced by a homogeneous medium, with an index of refraction $n = I + (2\pi N/k^2)f$. Since n for both X-rays and slow neutrons hardly differs from I, it is better to use the susceptibility tensor defined by $\chi = (4\pi N/k^2)f$ [2].

By choosing a simple homogeneous layer with the above susceptibility χ and z-axis normal to the layer, one gets the well-known 1D wave equation:

$$\Psi''(z) + k^2 \sin\theta\left[I\sin\theta + \frac{\chi}{\sin\theta}\right]\Psi(z) = 0 \quad (3)$$

with θ being the angle of incidence. Defining Φ via $(\mathrm{i}k\sin\theta)\Phi'(z) := \Psi''(z)$, we get a system of first-order differential equations:

$$\frac{\mathrm{d}}{\mathrm{d}z}\begin{pmatrix}\Phi\\ \Psi\end{pmatrix} = \mathrm{i}kM\begin{pmatrix}\Phi\\ \Psi\end{pmatrix}, \quad (4)$$

where

$$M = \begin{pmatrix} 0 & I\sin\theta + \dfrac{\chi}{\sin\theta} \\ I\sin\theta & 0 \end{pmatrix} \quad (5)$$

is commonly called the "differential propagation matrix" in optics [2,3]. Eq. (4) was derived without specifying the scattering process.

For an arbitrary multilayered film with homogeneous layers of thicknesses $d_1, d_2, \ldots, d_S$ and differential propagation matrices $M_1, M_2, \ldots, M_S$, χ in Eq. (5) is replaced by the susceptibility χ_l of layer l. The solution of the differential equation (4) can be expressed in terms of the total characteristic matrix

$$L = L_S \cdot \cdots \cdot L_2 \cdot L_1 \quad (6)$$

of the multilayer, where

$$L_l = \exp(\mathrm{i}kd_l M_l) \quad (7)$$

is the characteristic matrix of the lth individual layer. The 2×2 reflectivity matrix R is derived from the total characteristic matrix L by

$$R = (L_{[11]} - L_{[12]} - L_{[21]} + L_{[22]})^{-1} (L_{[11]} + L_{[12]} - L_{[21]} - L_{[22]}), \quad (8)$$

where $L_{[ij]}$ ($i, j = 1, 2$) are 2×2 blocks of the 4×4 total characteristic matrix L [2]. The reflected intensity

$$I^r = \mathrm{Tr}(R^\dagger R\rho) \quad (9)$$

can be calculated by using the arbitrary polarization density matrix ρ of the incident beam and the reflectivity matrix [4].

3. Numerical considerations

The numerical problem in evaluating the reflectivity is the calculation of the exponential of the 4×4 matrices in Eq. (7). Here we cite our previous results [5] proving that it is possible to get a closed solution to the general problem requiring the calculation of 2×2 matrices only. The characteristic matrix is of the form

$$L_l = \begin{pmatrix} \cosh(kd_l F_l) & (1/x)F_l \sinh(kd_l F_l) \\ xF_l^{-1}\sinh(kd_l F_l) & \cosh(kd_l F_l) \end{pmatrix}, \quad (10)$$

where the 2×2 $F_l = \sqrt{-I\sin^2\theta - \chi_l}$ and $x = \mathrm{i}\sin\theta$ [2].

To evaluate Eq. (10), first we have to calculate the 2×2 square root of the F matrices. This can be made by using the identity

$$G^{1/2} = \frac{G + I\sqrt{\det G}}{\sqrt{\mathrm{Tr}\, G + 2\sqrt{\det G}}}, \tag{11}$$

where G is any nondiagonal 2×2 matrix [3]. The sinh and cosh functions are calculated from their definition with the exponential functions. Moreover, the exponential of the 2×2 matrix G can be expressed by itself and its scalar invariants:

$$\exp G = \exp(\tfrac{1}{2}\mathrm{Tr}\, G) \times \left[\cos\sqrt{\det \bar{G}}\, I + \frac{\sin\sqrt{\det \bar{G}}}{\sqrt{\det \bar{G}}}\bar{G}\right], \tag{12}$$

where $\bar{G} = G - \frac{1}{2} I\,\mathrm{Tr}\, G$ [4].

In order to calculate the characteristic matrix of a semi-infinite layer (substrate) S, we have to find its $L_S \to L^\infty$ limit for $d_S \to \infty$. From Eqs. (5)–(13) follows that the corresponding limit is given by

$$L^\infty = \begin{pmatrix} I & p\sqrt{I + \dfrac{\chi_S}{\sin^2\theta}} \\ p\left(\sqrt{I + \dfrac{\chi_S}{\sin^2\theta}}\right)^{-1} & I \end{pmatrix} \tag{13}$$

where $p = \mathrm{sgn}[\mathrm{Re}(\mathrm{Tr}\, F_S)]$ is the sign of the real part of the trace of F_S.

The above algebra turns out to be numerically very stable, therefore this approach is suitable for fast numerical calculations of the characteristic matrices for anisotropic stratified media. In fact, the exponential of the matrix in Eq. (5) can be calculated exactly without solving any eigenvalue problem. The program based on this calculus is freely available [5,6].

4. Mössbauer and polarized neutron reflectometries

A simple application of Eq. (4) to nuclear resonant X-ray scattering is not possible, since the anisotropic Maxwell equations and the spin-dependent Schrödinger-equation lead to different results [3,7] and the 3×3 susceptibility tensor cannot be expressed by the 2×2 forward scattering amplitude f in general. However, starting from the Maxwell equations and using the 3×3 nuclear susceptibility tensor given by Afanas'ev and Kagan [8] the nuclear resonant X-ray reflectivity could be derived [2] for forward scattering and grazing incidence in terms of the coherent forward scattering amplitude. The dynamical theory of X-ray scattering [9,10] provide an equivalent result in the grazing incidence limit [2,5]. However, in Ref. [2] both an upper and a lower limit was found for the grazing angle θ for this approximation to apply, which limits are not present in the original theory of Lax [1]. The forward scattering amplitude matrix was expressed for the nuclear resonant X-ray case in Refs. [4,11] in terms of the hyperfine interactions.

The application of the above optics for PNR implies specifying f (or χ) for the interaction potential U in Eq. (3). We use the potential $U(\boldsymbol{r}) = U_\mathrm{p}(\boldsymbol{r}) + U_\mathrm{m}(\boldsymbol{r})$ as the sum of the isotropic nuclear potential

$$U_\mathrm{p}(\boldsymbol{r}) = 4\pi b\delta(\boldsymbol{r}) I, \tag{14}$$

and the anisotropic magnetic potential

$$U_\mathrm{m}(\boldsymbol{r}) = -\frac{2m}{\hbar^2}\mu_\mathrm{m}[\boldsymbol{B}_\mathrm{a}(\boldsymbol{r}) + \boldsymbol{B}_\mathrm{ext}] = -\frac{2m}{\hbar^2}\mu_\mathrm{m}\boldsymbol{B}(\boldsymbol{r}) \tag{15}$$

with m being the mass of the neutron, b the nuclear scattering length of the nucleus in the laboratory system, $\mu_\mathrm{m} = g\mu_\mathrm{N}\sigma$ the magnetic moment operator of the neutron, $g = -1.9132$, $\mu_\mathrm{N} = 5.050\times 10^{-27}\ \mathrm{Am}^2$, σ the Pauli operator, $\boldsymbol{B}_\mathrm{a}$ the atomic magnetic field, $\boldsymbol{B}_\mathrm{ext}$ the (homogeneous) external magnetic filed. In the first Born approximation

$$f = -\frac{1}{4\pi}\int_\Omega \mathrm{d}^3\boldsymbol{r}\, U(\boldsymbol{r}), \tag{16}$$

where Ω is the volume of the interaction (in fact the atomic volume). By using $\chi = (4\pi N/k^2) f$ we get

$$\chi = \frac{1}{k^2}\left[\frac{2m}{\hbar^2} g\mu_\mathrm{N}\sigma\bar{\boldsymbol{B}} - 4\pi N\sum_i \alpha_i b_i I\right], \tag{17}$$

where index i accounts for the different types of scattering centers, and α_i for the relative abundance of the ith nucleus. The mean magnetic field $\bar{\boldsymbol{B}} = \boldsymbol{B}_{\text{ext}} + \bar{\boldsymbol{B}}_{\text{a}} = \boldsymbol{B}_{\text{ext}} + \frac{1}{\Omega}\int_\Omega \mathrm{d}^3\boldsymbol{r}\boldsymbol{B}_{\text{a}}(\boldsymbol{r})$.

In neutron reflectometry the scattering vector, $Q = 2k\sin\theta$ and the scattering length density

$$K = k^2\chi = \frac{2m}{\hbar^2} g\mu_{\text{N}}\sigma\bar{\boldsymbol{B}} - 4\pi N \sum_i \alpha_i b_i I \tag{18}$$

are more often used than θ and χ. With these notations, Eq. (4) reads:

$$\frac{\mathrm{d}}{\mathrm{d}z}\begin{pmatrix}\Phi\\ \Psi\end{pmatrix} = \mathrm{i}\begin{pmatrix} 0 & \dfrac{Q}{2}I + \dfrac{2K}{Q} \\ \dfrac{Q}{2}I & 0 \end{pmatrix}\begin{pmatrix}\Phi\\ \Psi\end{pmatrix}. \tag{19}$$

Using the definition of the Pauli matrices, the scattering length density matrix Eq. (18) is expressed by the physical quantities

$$K = \frac{2m}{\hbar^2} g\mu_{\text{N}}\begin{pmatrix} \bar{B}_{z'} & \bar{B}_{x'} - \mathrm{i}\bar{B}_{y'} \\ \bar{B}_{x'} + \mathrm{i}\bar{B}_{y'} & -\bar{B}_{z'} \end{pmatrix} - 4\pi N \sum_i \alpha_i b_i I, \tag{20}$$

where $\bar{B}_{x'}$, $\bar{B}_{y'}$, $\bar{B}_{z'}$ are the components of the magnetic field $\bar{\boldsymbol{B}}$ in an arbitary coordinate system with $z' \| B_{\text{ext}}$.

Having K from Eqs. (20) or (18) for each layer l, Eq. (10) is used to calculate the exponential of the differential propagation matrix of Eq. (19). With this (by applying Eqs. (10)–(13)) first Eq. (7) characteristic matrices, then Eq. (6) total characteristic matrix L, from which Eq. (8) complex reflectivity matrix R is calculated. For the sake of brevity, we dropped the layer index l in K, $\bar{\boldsymbol{B}}$, N, α_i and b_i in Eqs. (16)–(20).

An elegant covariant treatment of specular PNR [12] including earlier matrix methods of restricted form [13,14] recently published by Rühm et al. turns out to be equivalent to the present results. Indeed, substituting $p_0 = k\sin\theta$ and $\mathscr{H}_l = -(\hbar^2k^2/2m)\chi_l$ for layer l in Eq. (7) of Ref. [12] we obtain Eq. (10), an equation equivalent to Eq. (3.20) of Ref. [2]. Consequently, what we have shown here is the equivalence [15] of the supermatrix formalisms developed for SMR [2] and PNR [12].

5. The external magnetic field as an anisotropic medium

Although their general treatment would have allowed for, Rühm et al. [12] did not explicitly studied the effect of the (guiding or polarizing) external magnetic field on the neutron beam, what we briefly outline in this section in the standard manner borrowed from anisotropic optics [3].

Reflectivity expression (8) is only valid for a neutron beam incident on the layer system ($l = 1, 2, 3, \ldots, S$) from the vacuum ($l = 0$). In the typical experimental setup, however, guiding fields and often strong external magnetic fields are used in order to eliminate depolarization of the neutrons and to ensure polarization of the sample, respectively. The effect of the external magnetic field was studied by Pleshanov [16] and Fermon [17] in detail. From Eq. (20) it follows, that the vacuum, in presence of an external magnetic field, is an anisotropic 'medium'. Consequently, the incoming beam is given in this 'medium' instead of being given in the vacuum. In order to treat this problem, following Borzdov [3] (for a brief outline in English see Ref. [2]), for the case of neutron reflectometry we introduce an impedance tensor γ by the following relationship:

$$\gamma^{0,r,t}\Psi^{0,r,t} := \Phi^{0,r,t}, \tag{21}$$

where indexes $0, r$ and t indicate incident, reflected and refracted waves, respectively (see Eq. (3.4) of Ref. [2]). Substituting Eq. (21) into Eq. (19), we get the impedance tensors

$$\gamma \equiv \gamma^0 = -\gamma^r = \sqrt{I + \frac{4K}{Q^2}}, \tag{22}$$

where K is calculated from Eq. (20) for the given external magnetic field. We dropped γ^t because the substrate is taken as a semi-infinite layer with Eq. (13). From expressions (18) and (20) for γ we get

$$\gamma(Q) = \frac{1}{Q}\begin{pmatrix} \sqrt{Q_+} & 0 \\ 0 & \sqrt{Q_-} \end{pmatrix}, \tag{23}$$

where

$$Q_\pm^2 = Q^2 \pm \frac{8m}{\hbar^2} g\mu_{\text{N}}|\boldsymbol{B}_{\text{ext}}| \tag{24}$$

is the momentum $Q_\pm = 2k\sin\theta_\pm$ measured in the external magnetic field. Due to the birefringence of anisotropic media (including vacuum in presence of external magnetic field), the beam propagation directions for the different polarizations necessarily differ from each other, consequently the angles of incidence and the momentum of the beams with different polarizations (sign '$+$' and '$-$') are also different ($\theta_\pm$ and $Q_\pm$). The vacuum momentum Q can be calculated backwards from $Q_\pm^2$ by applying the Fresnel refraction law [Eqs. (20) and (3), as well as the definition of K and Q by Eq. (18)].

Having the impedance tensors of the individual layers, we simply apply the modified general 2×2 reflectivity expression

$$R = [(L_{[11]} - L_{[21]})\gamma - L_{[12]} + L_{[22]}]^{-1}$$
$$[(L_{[11]} - L_{[21]})\gamma + L_{[12]} - L_{[22]}] \quad (25)$$

which takes the effect of the external magnetic field into account through the impedance tensor γ [2]. The reflected intensity I^r is calculated from Eq. (9) using the reflectivity matrix R and the polarization density matrix $\rho = \overline{|\Psi\rangle\langle\Psi|}$ of the incident beam, where the bar represents the average over the polarizations [4,12].

6. Conclusion

In summary, a common optical formalism of (nuclear) resonant X-ray (Mössbauer) reflectometry and polarized neutron reflectometry was presented. Consequently, the strictly covariant formalism of [3] as published in Ref. [2] and the corresponding computer program [5,6] are readily available for neutron reflectometry of layered systems of arbitrary complexity. Taking the effect of the external magnetic field through the impedance tensor into account, a modified reflectivity expression is given. The form of the reflectivity matrix allows for a very efficient numerical algorithm for both SMR and PNR implemented in Refs. [5,6].

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