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Method of the reflections function in the X-ray reflectometry study of multilayers

N.V. Kourtina*, E.A. Kravtsov, V.V. Ustinov

Institute of Metal Physics, Ural Division of the Russian Academy of Sciences, 18, S. Kovalevskaya St., GSP-170, Ekaterinburg, 620219, Russia

Abstract

A theory of specular X-ray reflectivity from a rough interface based on the reflection function method is proposed. By using the approximation of the abruptly changing potential, we represent a reflectivity in the form of a series. Its first term reproduces the Nevot–Croce approximation and the second one gives the phase correction, which can be used to obtain the degree of interface asymmetry. The model X-ray reflectometry profiles for Fe/Cr superlattice are used to illustrate the method. © 2002 Elsevier Science B.V. All rights reserved.

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The X-ray reflectometry is a useful tool for studying surface and interface structure in thin films and multilayers. Usually, the rough surface X-ray reflection is analysed in the frame of the plane-wave Born approximation (PWBA) or the distorted-wave Born approximation (DWBA) [1,2]. In this work, we apply the reflection function method (RFM) [3] to the specular X-ray reflection from a rough surface or interface.

Let us consider the X-ray reflection on a non-ideal interface structure. We assume that this structure is homogeneous along the surface which is parallel to the (xy) plane and the media can be characterized by its dielectric susceptibility $\chi(z)$ depending only on the normal coordinate z, where $\chi(z)\to\chi_\pm$ when $z\to\pm\infty$. The change of the material occurs only in the z-direction perpendicular to the surface. Then one has to solve the one-dimensional Helmholtz equation

$$\left(\frac{\mathrm{d}^2}{\mathrm{d}z^2}+k^2\sin^2\theta\right)E(z)+k^2\chi(z)E(z)=0. \tag{1}$$

Here $E(z)$ is the electric field in the medium, θ is the incident angle and $k=2\pi/\lambda$, λ being a wave length of radiation. As the first step, we need to evaluate the scattering matrix

$$S_{12}=\begin{pmatrix} r_{11} & t_{12} \\ t_{21} & r_{22}\end{pmatrix}$$

related with the given interface between two subsequent layers, which are denoted as 1 and 2.

The RFM starts from the transformation of the linear second order differential equation (1) for the wave amplitude $E(z)$ into a non-linear first order equation of Riccati type for the reflection function $B(z)$. This transformation is not unique and can be performed in a number of different ways. An advantage of the RFM is that the perturbation expansion carried out in the framework of this scheme gives more rapid convergence in comparison with the conventional Born series. In particular, the first order approximation easily enables one to go beyond DWBA. We denote $q(z)=2k\sqrt{\sin^2\theta+\chi(z)}$, and represent the electric field $E(z)$ in the form:

$$E(z)=q^{-1/2}(z)\left[A(z)\exp\left(\frac{\mathrm{i}}{2}\int_{z_0}^{z}q(x)\,\mathrm{d}x\right)+C(z)\exp\left(-\frac{\mathrm{i}}{2}\int_{z_0}^{z}q(x)\,\mathrm{d}x\right)\right], \tag{2a}$$

*Corresponding author. Fax: +7-343-92-327-37.
E-mail address: hope@imp.uran.ru (N.V. Kourtina).

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where $A(z)$ and $C(z)$ are the amplitude functions. In addition, we apply the following condition:

$$\frac{d}{dz}E(z) = \frac{i}{2}q^{1/2}(z)\left[A(z)\exp\left(\frac{i}{2}\int_{z_0}^{z} q(x)\,dx\right) - C(z)\exp\left(-\frac{i}{2}\int_{z_0}^{z} q(x)\,dx\right)\right]. \quad (2b)$$

The reflection function $B(z)$ is defined as $B(z) = C(z)/A(z)$.Taking into account the continuity of $E(z)$ and Eq. (1) one can prove that $B(z)$ satisfies the first order non-linear differential equation

$$\frac{d}{dz}B(z) = \frac{q'(z)}{2q(z)}\left[\exp\left(i\int_{z_0}^{z} q(x)\,dx\right) - B^2(z)\exp\left(-i\int_{z_0}^{z} q(x)\,dx\right)\right] \quad (3)$$

Eqs. (1) and (3) should be supplemented by the boundary conditions. For example, the choice of $B(+\infty) = 0$ corresponds to the X-ray beam, incident from $z<0$, and in this case a reflection coefficient r_{11} is given by the relation $r_{11} = B(-\infty)$. We also take into consideration the dimensionless functions $g_{\pm}(z)$, which are related to $\chi(z)$ via equality $\chi(z) - \chi_{\pm} = \pm(\chi_- - \chi_+)g_{\pm}(z)$. The function $g_-(z)\to 0$, when $z\to -\infty$, and $g_-(z)\to 1$, if $z\to +\infty$ (See Fig. 1). The functions $g_{\pm}(z)$ obey the relation $g_+(z) + g_-(z) = 1$. One can regard $g_{\pm}(z)$ as a "shape" of the interface, which reproduces the gradual transition from the first layer to the second one. We shall call interface "symmetric", if $(\partial/\partial z)g_-(z)$ is an even function of z, otherwise interface is "asymmetric".

In case of grazing incidence angles, Eq. (3) can be solved in the approximation of the abruptly changing potential. The small parameter ε of this expansion is defined as $\varepsilon = aq_c/2\pi$, where a is the characteristic length corresponding to the variation of the potential

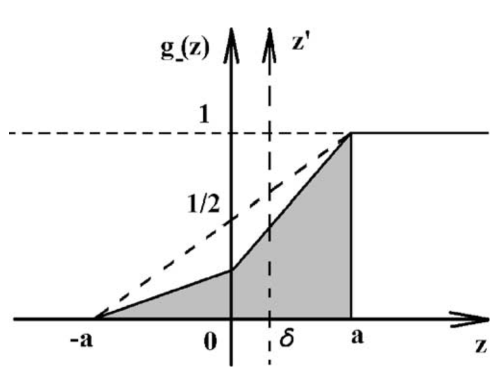

Fig. 1. The linear segment form of the profile $g_-(z)$, corresponding to the interface of a width $2a$. The "symmetric" case is shown by the dashed line, and the "asymmetric" one is depicted by the shaded region.

and $q_c = \max|q(z)|$. In the X-ray reflectometry studies a is of the order of mean-root-square interfacial roughness $\sigma = 2-8$ Å [4] and $q_c \sim (4\pi/\lambda)\sin\theta$. Therefore the condition $\varepsilon \ll 1$ holds over the scattering angle region $(0<\theta<4°)$. These estimates make it possible to find the solution of Eq. (3) in the form

$$B(z) = B_0(z)\exp(\beta(z)), \quad (4)$$

where

$$B_0(z) = (q(z) - q_2)/(q(z) + q_2),$$

$$q_{2(1)} = 2k\sqrt{\sin^2\theta + \chi_{\pm}}$$

and

$$\beta(z) = \sum_{n=1}^{\infty}\beta_n(z)\varepsilon^n.$$

The function $B_0(z)$ corresponds to the boundary condition $B(+\infty) = 0$ and it gives the Fresnel reflection coefficient $r_{11}^F = (q_1 - q_2)/(q_1 + q_2)$ from an ideal sharp interface. The series $\beta(z)$ yields the corrections due to the interfacial non-ideality.

The use of ansatz (4) is the essential step in the derivation. It enables us partially to sum up the reducible parts of the expansion $B(z)$ in powers of ε, so that the coefficients $\beta_n(z)$ are associated with the irreducible terms only. The series $\beta(z)$ can be found by means of subsequent iterations from Eq. (3). It turns out that, at each step n, one encounters the only linear inhomogeneous differential equation for $\beta_n(z)$. The details of this derivation will be presented elsewhere. As a result, up to the third order of ε, the elements of the matrix S_{12} can be written in the form

$$r_{11} = r_{11}^F\exp\{iq_1\delta - \tfrac{1}{2}q_1q_2\sigma^2 + iq_1[(q_1^2 + 3q_2^2)\mu_1 + (q_1^2 - q_2^2)\mu_2]\sigma^3\}, \quad (5)$$

$$r_{22} = r_{22}^F\exp\{-iq_2\delta - \tfrac{1}{2}q_1q_2\sigma^2 + iq_2[-(q_2^2+3q_1^2)\mu_1 + (q_1^2 - q_2^2)\mu_2]\sigma^3\}, \quad (6)$$

$$t_{12} = t_{21} = t_{12}^F\exp(\tfrac{1}{2}i(q_1 - q_2)\delta + \tfrac{1}{8}(q_1 - q_2)^2\sigma^2 + \tfrac{1}{2}i(q_1 - q_2)^2[(q_1 - q_2)\mu_1 + (q_1 + q_2)\mu_2]\sigma^3). \quad (7)$$

Here r^F, t^F are Fresnel's reflection and transmission amplitudes and parameters δ, σ, and $\mu_{1(2)}$ are expressed via $g_{\pm}(z)$ as follows

$$\delta = -\int_{-\infty}^{+\infty} zg_-'(z)\,dz, \quad (8)$$

$$\sigma^2 = -2I^{(2)}(-,+) = 2\int_{-\infty}^{+\infty} g_-(z_1)\,dz_1\int_{z1}^{+\infty} g_+(z_2)\,dz_2 \quad (9)$$

$$\mu_{1(2)} = [I^{(3)}(-,+,+)\mp I^{(3)}(-,-,+)]/4\sigma^3, \quad (10)$$

where

$$I^{(3)}(-,\pm,+) = \int_{-\infty}^{+\infty} g_-(z)\,\mathrm{d}z_1 \int_{z1}^{+\infty} g_\pm(z)\,\mathrm{d}z_2 \int_{z\infty}^{+\infty} g_+(z)\,\mathrm{d}z_3.$$

Consider now the physical meaning of Eqs. (5)–(10). First of all, the phase shift δ arises due to transmitting electromagnetic wave in the non-uniform interface region. This phase shift is equivalent to "effective" increase in the thickness of layer 1 to value δ: $z' = z - \delta$ (see Fig. 1). In the process of the numerical treatment of the X-ray reflectometry profiles this fact enables one to adjust the ratio between the layers' thicknesses in the periodical cell of the superlattice in order to obtain the best fit to experimental data. The second order correction to the amplitudes r^F, t^F in Eq. (5)–(7) reproduces the well-known Nevot–Croce [5] approximation. The magnitude σ has the meaning of the root-mean-square interfacial roughness and it is given by Eq. (9).

The phase correction corresponding to the third order terms of the expansion is a new feature in the question. In addition to σ^2 it contains two extra parameters μ_1 and μ_2. We found, that μ_2 is in general non-zero for a wide set of profiles $\chi(z)$, whereas μ_1 does not vanish in case of the asymmetric interfaces only. Hence, this property may be used to define μ_1 as the measure of the interface asymmetry. The parameter μ_2 has no such evident meaning as μ_1. But we may note that due to inequality $|(q_1 - q_2)/q_1| \ll 1$ the contribution from μ_2 in Eqs. (5) and (6) turns out to be less essential, than the asymmetric term, proportional to μ_1.

To test the obtained approximation we exploited the symmetric Epstein profile $g_-^E(z) = (1 + e^{-z/a})^{-1}$ for which the exact solution is known. In this case we obtained $\sigma = (\pi/\sqrt{3})a$ and $\mu_2 = (3\sqrt{3}/2\pi^3)\varsigma(3) \cong 0.100$. We have also found that μ_2 has the same order of magnitude regardless of the exact choice of $g_\pm(z)$ (one possible form from many others is shown in Fig. 1). Assuming further $\mu_2 = 0.1$ the model X-ray profile corresponding to the Al_2O_3/Cr(70 Å)/[Fe(20 Å)/Cr(9 Å)]$_8$ multilayer has been calculated, taking into account the possible asymmetry μ_1 in the interfacial structure. Provided the matrices $S_{k,k+1}$ are known, the solution of Eq. (1) and, hence, the scattering matrix S of the whole multilayer is found by means of recurrent scheme [6]. The results obtained are shown in Fig. 2. In agreement with Eqs. (5)–(7) the phase correction becomes essential with the increase of the incident angle θ and it provides a more adequate description of the reflectometry spectrum for the scattering vectors in the range from the first to the second Braggs' peaks. The more exhaustive account and the details of our numerical algorithm will be presented elsewhere. We would like to emphasize that the form of the scattering matrix as given in Eqs. (5)–(7) is rather general, i.e., it is irrelevant to the precise form of a reflectivity profile. Thus it provides the unification description of a large variety of possible symmetric as well as asymmetric interfaces.

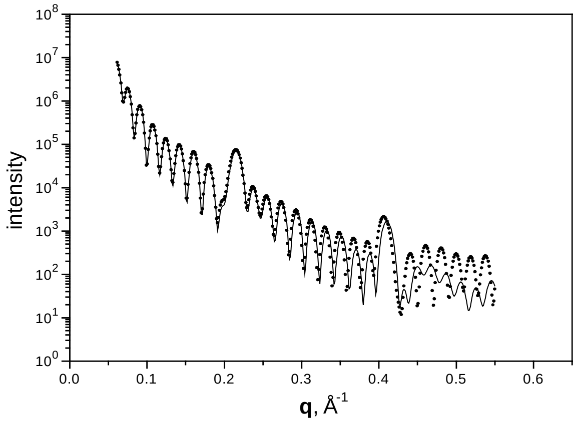

Fig. 2. Model X-ray reflectivity profiles for multilayer structure Al_2O_3/Cr(70 Å)/[Fe(20 Å)/Cr(9 Å)]$_8$ calculated without asymmetric phase corrections (points), and with asymmetric phase correction ($\mu_1 = 0.2$, solid line). Wave length $\lambda = 1.789$Å.

Summing up, we have developed the theory of specular X-ray reflectivity from a rough interface based upon the reflection function method. By using the approximation of the abruptly changing potential we have found the phase correction to the reflectivity due to interface roughness and asymmetry, which is essential for the description of the X-ray reflectivity spectra for greater incident angles.

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