Diffuse scattering of x rays from nonideal layered structures

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A new theory of nonspecular x-ray scattering from layered systems with random rough interfaces based on the distorted-wave Born approximation is presented. Calculations of the diffuse scattering from a single gold layer and two W/Si multilayer mirrors has been carried out. The theory explains the existence of maxima and minima in the angular distribution of diffusely scattered intensity resulting from standing-wave-enhanced scattering and other dynamical effects. The influence of the mutual correlation between individual interface profiles on x-ray scattering is discussed. © *1995 American Institute of Physics.*

I. INTRODUCTION

Nowadays, thin layers and synthetic layered structures are of an increasing significance in applied physics. The physical properties of layered systems depend not only on the composition and thickness of sublayers but can be strongly affected by the quality of interfaces.

Recent progress in the controlled deposition of ultrathin films has made it possible to fabricate nanometer-period multilayer structures which may be used as optical elements for soft x rays.[1] It is well known that the specular reflectivity can be considerably modified by boundary imperfections. Moreover, interfacial roughness can lead to a loss of contrast between specularly reflected and diffusely scattered radiation which is detrimental for imaging systems. For these applications, both the root-mean-square (rms) value and the lateral characteristics of interfacial profiles are important. The determination of interfacial roughness is therefore a necessary assumption for predicting scattering properties of multilayers and, prospectively, for improvement of deposition technology.

X-ray methods provide tools for investigating surfaces, thin layers, and layered structures. There is a large body of work discussing the specularly reflected component of x-ray scattering.[2,3] The reflectivity curves yield information on densities and thicknesses of sublayers and are frequently used for a rms roughness evaluation. Considerably less attention has been devoted to the diffuse component of the scattering, which depends on the rms value as well as on the correlation function.[4–8]

Several theories of nonspecular scattering from a single random rough surface have been developed. The Born approximation (BA) and the distorted-wave Born approximation (DWBA) are mainly used in the x-ray region.[4] The BA is valid at glancing angles much greater than the critical angle θ_c, whereas it breaks down as we approach the total external reflection region. On the contrary, the DWBA is suitable for glancing angles close to θ_c. However, at larger glancing angles, the expressions for the differential cross section derived in the DWBA reduce to the results in the BA.[4]

The generalization of a scattering theory for layered structures with rough interfaces is a very complicated problem. The scattered field is a superposition of many plane waves (modes) and it is impossible to take the multiple scattering of every mode within the system into account. Therefore a simplified model of interaction between electromagnetic radiation and stratified medium has to be considered.

The development of the BA for the case of x-ray scattering from nonideal multilayer structures has been carried out by Stearns.[5] His solution is based on the so-called "specular field approximation." In this approach, the coherent (specular) field within the multilayer is treated dynamically, including multiple reflection and extinction, and the incoherent (diffuse) field is treated kinematically, i.e., the total incoherent field is approximated by the sum of diffuse scattering from each interface.

More recently, the extension of the DWBA for layered systems with rough interfaces has been performed by Holý *et al.*[9] In their approach, the specular field within the layered system with smooth interfaces has been used for calculating the diffusely scattered intensity.

In this paper, a new theory of x-ray diffuse scattering from layered systems based on the DWBA is described. The real specular field and the real transmission coefficient of the radiation diffusely scattered from inner interfaces are considered. By means of this theory, the previously measured modulations in the diffusely scattered intensity are explained. The influence of mutual correlation between individual interface profiles on the x-ray diffuse scattering is discussed.

II. THEORY

Let us consider a plane wave $\mathbf{E}_1^T$ with wave vector ${}^1\mathbf{k}_1$ incident on a system of $N-1$ layers deposited on a thick substrate. This wave is specularly reflected and refracted (coherent field) and simultaneously diffusely scattered (incoherent field) from each interface.

A. The coherent field

We start by determining the coherent field within the layered structure which may be in each medium i expressed as the superposition of two plane waves $\mathbf{E}_i^T$ and $\mathbf{E}_i^R$ (Fig. 1). The continuity of the tangential components of the electric and magnetic vectors at the boundary i may be written as[10]

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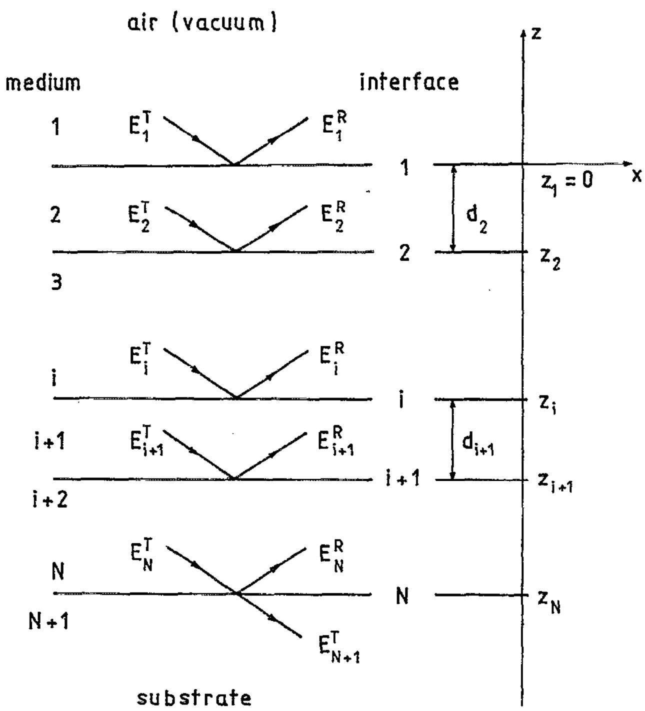

FIG. 1. Scheme of the coherent field within stratified medium.

$$E_i^T(\theta_1)+E_i^R(\theta_1)=\frac{1}{\varphi_{i+1}(\theta_1)}E_{i+1}^T(\theta_1)+\varphi_{i+1}(\theta_1)E_{i+1}^R(\theta_1), \tag{1a}$$

$$E_i^T(\theta_1)\,{}^1k_{iz}-E_i^R(\theta_1)\,{}^1k_{iz}=\frac{1}{\varphi_{i+1}(\theta_1)}E_{i+1}^T(\theta_1)\,{}^1k_{i+1z}-\varphi_{i+1}(\theta_1)E_{i+1}^R(\theta_1)\,{}^1k_{i+1z}, \tag{1b}$$

where ${}^1\mathbf{k}_i$ is the wave vector of the wave $\mathbf{E}_i^T$, φ_i the amplitude factor corresponding to the perpendicular depth d_i,

$$\varphi_i(\theta_1)=e^{-i\,{}^1k_{iz}d_i} \quad \text{for } i=2,\ldots,N, \tag{2}$$

and $\varphi_{N+1}=1$.

According to Snell's law, the wave vectors ${}^1\mathbf{k}_i$ are determined by the refractive indices n_i of individual layers, wavelength λ, and the glancing angle θ_1 of the incident wave:

$${}^1k_{ix}={}^1k_{1x}, \tag{3a}$$

$${}^1k_{iy}={}^1k_{1y}, \tag{3b}$$

$${}^1k_{iz}=-k_0\sqrt{n_i^2-\cos^2\theta_1}=-{}^1\xi_i, \tag{3c}$$

where $k_0=2\pi/\lambda$ is the magnitude of the vacuum wave vector. By dividing the difference of Eqs. (1a) and (1b) by their sum, the well-known recursion formula[10,11] may be obtained:

$$R_i(\theta_1)=\frac{R_{i+1}(\theta_1)\varphi_{i+1}^2(\theta_1)+\tilde{r}_{i,i+1}(\theta_1)}{R_{i+1}(\theta_1)\varphi_{i+1}^2(\theta_1)\tilde{r}_{i,i+1}(\theta_1)+1}. \tag{4}$$

$R_i=E_i^R/E_i^T$ is the reflection coefficient of the system below the boundary i. The reflection coefficient $\tilde{r}_{i,i+1}$ of this rough interface is related to the Fresnel reflection coefficient $r_{i,i+1}$ of a smooth interface by[12]

$$\tilde{r}_{i,i+1}=r_{i,i+1}\,\chi_i(\sqrt{q_{iz}q_{i+1z}}). \tag{5}$$

χ_i denotes the one-dimensional characteristic function of the rough interface i. Vector $\mathbf{q}_i(0,0,2\,{}^1\xi_i)$ represents the difference of wave vectors of waves $\mathbf{E}_i^R$, $\mathbf{E}_i^T$. The set of equations (4) may be solved by starting at the bottom medium $N+1$, where $R_{N+1}=0$ (since the thickness of the substrate is assumed to be infinite).

Further, the coherent field may be determined using formulas[13]

$$E_i^R(\theta_1)=R_i(\theta_1)E_i^T(\theta_1), \tag{6}$$

$$E_{i+1}^T(\theta_1)=\frac{1+R_i(\theta_1)}{1+R_{i+1}(\theta_1)\varphi_{i+1}^2(\theta_1)}\,\varphi_{i+1}(\theta_1)E_i^T(\theta_1). \tag{7}$$

The calculation is now carried out from top to bottom.

B. The diffuse x-ray scattering from a single rough interface

As the second step, the scattering of the coherent field from each rough interface has to be evaluated. Let us consider rough interface (surface) i between two homogeneous media i and $i+1$. The deviation of this interface from the average reference plane $z=z_i$ is described by the profile function $h_i(x,y)$.

The electric field for x rays polarized perpendicular to the plane of incidence satisfies the wave equation

$$\nabla^2\psi(\mathbf{r})+k_0^2\psi(\mathbf{r})-V(\mathbf{r})\psi(\mathbf{r})=0, \tag{8}$$

where $\mathbf{r}$ is the position vector and the scattering potential $V(\mathbf{r})$ is related to the refractive index $n(\mathbf{r})$ by

$$V(\mathbf{r})=k_0^2[1-n^2(\mathbf{r})]. \tag{9}$$

The polarization of x rays in the plane of incidence is not discussed since at grazing incidence the results are the same for both the polarization components.

Let us assume that the real rough interface represents a small perturbation from the smooth interface, for which the exact eigenfunctions are known. Then it is convenient to split the scattering potential into two parts:

$$V(\mathbf{r})=V_1(\mathbf{r})+V_2(\mathbf{r}), \tag{10}$$

where V_1 is the scattering potential of the system with a smooth interface and V_2 is regarded as the perturbation due to the roughness:

$$V_1=\begin{cases} k_0^2(1-n_i^2), & \text{for } z>z_i, \\ k_0^2(1-n_{i+1}^2), & \text{for } z<z_i \end{cases} \tag{11}$$

and

$$V_2=\begin{cases} k_0^2(n_i^2-n_{i+1}^2), & \text{for } z_i<z<z_i+h_i(x,y), \\ -k_0^2(n_i^2-n_{i+1}^2), & \text{for } z_i+h_i(x,y)<z<z_i, \\ 0, & \text{elsewhere.} \end{cases} \tag{12}$$

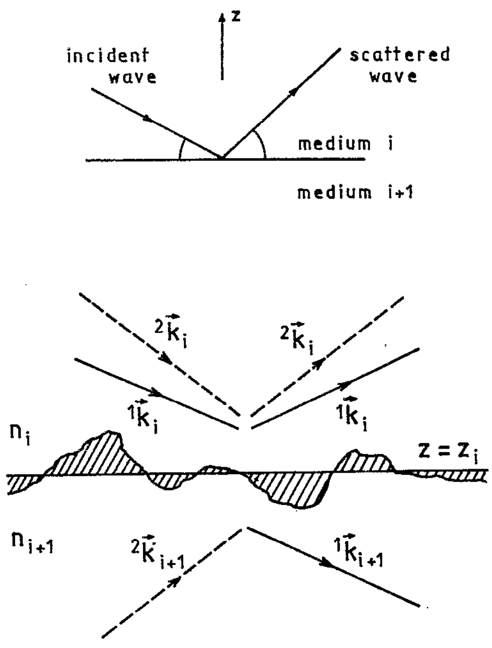

FIG. 2. X-ray scattering from random rough interface i. The incident wave ($^1\mathbf{k}_i$) is scattered into the same half space ($^2\mathbf{k}_i$).

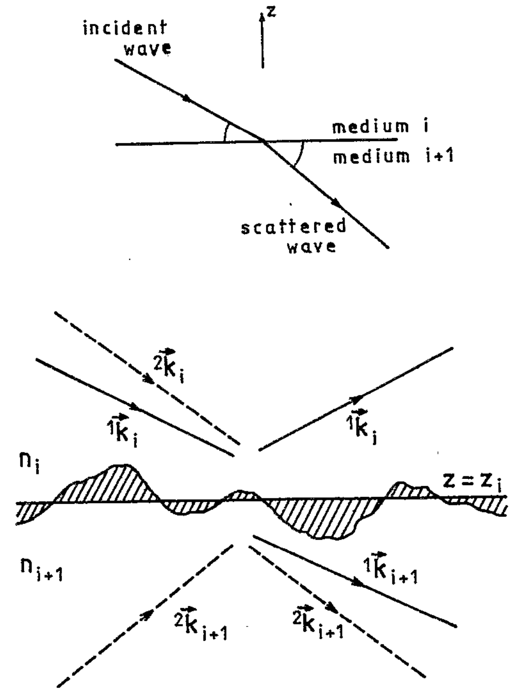

FIG. 3. X-ray scattering from random rough interface i. The incident wave ($^1\mathbf{k}_i$) is scattered into the opposite half space ($^2\mathbf{k}_{i+1}$).

Let the plane wave

$$^1\phi = e^{i\,^1\mathbf{k}_i\mathbf{r}} \tag{13}$$

with the wave vector $^1\mathbf{k}_i$ fall on the boundary i. According to the Fresnel theory, the eigenstate for the smooth interface is

$$^1\psi(\mathbf{r}) = \begin{cases} e^{i\,^1\mathbf{k}_i\mathbf{r}} + r_{i,i+1}(^1\mathbf{k}_i)e^{i\,^1\mathbf{k}_i'\mathbf{r}}, & \text{for } z>z_i, \\ t_{i,i+1}(^1\mathbf{k}_i)e^{i\,^1\mathbf{k}_{i+1}\mathbf{r}}, & \text{for } z<z_i, \end{cases} \tag{14}$$

where $^1\mathbf{k}_i'$, $^1\mathbf{k}_{i+1}$ are the wave vectors of the specularly reflected and transmitted waves. The amplitudes of these waves are given by the Fresnel reflection and transmission coefficients $r_{i,i+1}$ and $t_{i,i+1}$, respectively.

Sinha *et al.*[4] define another eigenstate for the smooth interface,

$$^2\bar{\psi}(\mathbf{r}) = \begin{cases} e^{i\,^2\mathbf{k}_i\mathbf{r}} + r^*_{i,i+1}(-^2\mathbf{k}_i)e^{i\,^2\mathbf{k}_i'\mathbf{r}}, & \text{for } z>z_i, \\ t^*_{i,i+1}(-^2\mathbf{k}_i)e^{i\,^2\mathbf{k}_{i+1}\mathbf{r}}, & \text{for } z<z_i, \end{cases} \tag{15}$$

which is a time reversed state for the incident plane wave with the wave vector $-^2\mathbf{k}_i$ (Fig. 2).

The T matrix for scattering between states $^1\mathbf{k}_i$ and $^2\mathbf{k}_i$ (proportional to the scattered electric field) has in the DWBA the form[14]

$$\langle 2|T|1\rangle = \langle ^2\bar{\psi}|V_1|^1\phi\rangle + \langle ^2\bar{\psi}|V_2|^1\psi\rangle. \tag{16}$$

The diffuse scattering is the consequence of the perturbation potential V_2 so it is related to the second term on the right-hand side of Eq. (16) only.

Substituting expressions (12), (14), and (15) for V_2, $^1\psi$, and $^2\bar{\psi}$, respectively, the perturbation matrix element can be evaluated as

$$\langle ^2\bar{\psi}|V_2|^1\psi\rangle = k_0^2(n_i^2 - n_{i+1}^2)t_{i,i+1}(^1\mathbf{k}_i)t_{i,i+1}(-^2\mathbf{k}_i) \times F_i(\mathbf{q}_{i+1}). \tag{17}$$

The functions F_i are given by

$$F_i(\mathbf{q}_{i+1}) = \frac{i}{q_{i+1z}} \int\int_{S_0} dx\, dy (e^{-iq_{i+1z}h_i(x,y)} - 1) \times e^{-i(q_x x + q_y y)} \tag{18}$$

and the wave-vector transfers

$$\mathbf{q}_i = {}^2\mathbf{k}_i - {}^1\mathbf{k}_i. \tag{19}$$

Note that the tangent components of all the vectors $\mathbf{q}_i$ are the same ($q_{ix} = q_x$, $q_{iy} = q_y$). The integration in Eq. (18) is carried out over the illuminated area S_0 of the size L_xL_y.

In a similar way, the field diffusely scattered from the interface i into the medium $i+1$ may be derived. Then the eigenstate $^2\bar{\psi}$ is chosen as the time reversed state for the wave incident on the interface i from the medium $i+1$ with the wave vector $^2\mathbf{k}_{i+1}$ (Fig. 3):

$$^2\bar{\psi}(\mathbf{r}) = \begin{cases} t^*_{i+1,i}(-^2\mathbf{k}_{i+1})e^{i\,^2\mathbf{k}_i\mathbf{r}}, & \text{for } z>z_i, \\ e^{i\,^2\mathbf{k}_{i+1}\mathbf{r}} + r^*_{i+1,i}(-^2\mathbf{k}_{i+1})e^{i\,^2\mathbf{k}_{i+1}'\mathbf{r}}, & \text{for } z<z_i, \end{cases} \tag{20}$$

leading now to the perturbation matrix element[13]

$$\langle ^2\bar{\psi}|V_2|^1\psi\rangle = k_0^2(n_i^2 - n_{i+1}^2)t_{i,i+1}(^1\mathbf{k}_i)t_{i+1,i}(-^2\mathbf{k}_{i+1})F_i(\mathbf{q}_{i+1}). \tag{21}$$

C. The diffuse x-ray scattering from a layered system

For the evaluation of the diffuse scattering from layered systems, the scattering of the waves $\mathbf{E}_i^T(\theta_1)$, $\mathbf{E}_{i+1}^R(\theta_1)\varphi_{i+1}(\theta_1)$ from each interface i both into the medium i and $i+1$ has to be taken into account. The wave vectors ${}^2\mathbf{k}_i$ of the scattered waves are again determined by the observation angle θ_2. Let us denote the pertinent matrix elements $\langle {}^2\bar{\psi}|V_2|{}^1\psi\rangle$ by $\mathcal{S}_i^{TR}$, $\mathcal{S}_i^{TT}$ (the scattering of the wave $\mathbf{E}_i^T$) and $\mathcal{S}_i^{RR}$, $\mathcal{S}_i^{RT}$ (the scattering of the wave $\mathbf{E}_{i+1}^R\varphi_{i+1}$). It follows from Eqs. (17) and (21) that

$$\mathcal{S}_i^{TR}(\mathbf{q}_1)=E_i^T(\theta_1)k_0^2(n_i^2-n_{i+1}^2)\times t_{i,i+1}(\theta_1)t_{i,i+1}(\theta_2)F_i({}^1\xi_{i+1}+{}^2\xi_{i+1}), \tag{22a}$$

$$\mathcal{S}_i^{TT}(\mathbf{q}_1)=E_i^T(\theta_1)k_0^2(n_i^2-n_{i+1}^2)\times t_{i,i+1}(\theta_1)t_{i+1,i}(\theta_2)F_i(-{}^1\xi_{i+1}+{}^2\xi_{i+1}), \tag{22b}$$

$$\mathcal{S}_i^{RR}(\mathbf{q}_1)=-E_{i+1}^R(\theta_1)\varphi_{i+1}(\theta_1)k_0^2(n_{i+1}^2-n_i^2)\times t_{i+1,i}(\theta_1)t_{i,i+1}(\theta_2)F_i({}^1\xi_i-{}^2\xi_i), \tag{22c}$$

$$\mathcal{S}_i^{RT}(\mathbf{q}_1)=-E_{i+1}^R(\theta_1)\varphi_{i+1}(\theta_1)k_0^2(n_{i+1}^2-n_i^2)\times t_{i+1,i}(\theta_1)t_{i+1,i}(\theta_2)F_i(-{}^1\xi_i-{}^2\xi_i), \tag{22d}$$

where the tangential components q_x, q_y of vectors $\mathbf{q}_i$, $\mathbf{q}_{i+1}$ are omitted in the argument of functions F_i. The variables ${}^2\xi_i$ are defined analogically to ${}^1\xi_i$ as

$${}^2\xi_i=k_0\sqrt{n_i^2-\cos^2\theta_2}. \tag{23}$$

If the multiple diffuse scattering is neglected, the variables $\mathcal{S}_i$ may be considered as "*independent sources of x rays.*" Further, the field outside the layered system produced by these "*sources*" has to be determined. In contrast to the kinematical approximation, the total incoherent field is not expressed as the sum of the waves scattered from each interface but the multiple reflections and thus the real transmission are included in the calculation. Consequently, besides $\mathcal{S}_i^{TR}$ and $\mathcal{S}_i^{RR}$, the terms $\mathcal{S}_i^{TT}$, $\mathcal{S}_i^{RT}$ have to be considered.

In the medium i, the sources $\mathcal{S}_{i-1}^{TT}$, $\mathcal{S}_{i-1}^{RT}$, $\mathcal{S}_i^{TR}$, $\mathcal{S}_i^{RR}$ produce the electromagnetic field, which is a superposition of two specularly related waves $\mathcal{D}_i^T$, $\mathcal{D}_i^R$. The boundary conditions for the interfaces $i-1$ and i yield

$$\mathcal{D}_i^R(\mathbf{q}_1)=R_i(\theta_2)\mathcal{D}_i^T(\mathbf{q}_1)+\mathcal{S}_i^{RR}(\mathbf{q}_1)+\mathcal{S}_i^{TR}(\mathbf{q}_1), \tag{24}$$

$$\frac{1}{\varphi_i(\theta_2)}\mathcal{D}_i^T(\mathbf{q}_1)=Q_{i-1}(\theta_2)\mathcal{D}_i^R(\mathbf{q}_1)\varphi_i(\theta_2)+\mathcal{S}_{i-1}^{RT}(\mathbf{q}_1)+\mathcal{S}_{i-1}^{TT}(\mathbf{q}_1), \tag{25}$$

where Q_i are the reflection coefficients of layered systems above the interface i. We can determine $R_i(\theta_2)$ from Eq. (4) and $Q_i(\theta_2)$ from the similar recursive formula

$$Q_i(\theta_2)=\frac{Q_{i-1}(\theta_2)\varphi_{i-1}^2(\theta_2)+\tilde{r}_{i,i-1}(\theta_2)}{Q_{i-1}(\theta_2)\varphi_{i-1}^2(\theta_2)\tilde{r}_{i,i-1}(\theta_2)+1}. \tag{26}$$

The values of Q_i are calculated in the direction from top ($Q_1=0$) to bottom.

If we express the term $\mathcal{D}_i^R$ from Eqs. (24) and (25), multiply it by the transmission coefficient $T_i(\theta_2)$ for the system above the interface i, and carry out the summation over all interfaces, the final matrix element $\mathcal{D}_0$ corresponding to the total diffusely scattered field outside the layered structure may be obtained:

$$\mathcal{D}_0(\mathbf{q}_1)=\sum_{i=1}^{N}T_i(\theta_2)\mathcal{D}_i^R(\mathbf{q}_1)=\sum_{i=1}^{N}T_i(\theta_2)\frac{R_i(\theta_2)\varphi_i(\theta_2)[\mathcal{S}_{i-1}^{RT}(\mathbf{q}_1)+\mathcal{S}_{i-1}^{TT}(\mathbf{q}_1)]+\mathcal{S}_i^{RR}(\mathbf{q}_1)+\mathcal{S}_i^{TR}(\mathbf{q}_1)}{1-R_i(\theta_2)Q_{i-1}(\theta_2)\varphi_i^2(\theta_2)}. \tag{27}$$

The transmission coefficients $T_i(\theta_2)$ may be determined in the following way: Let the plane wave $\mathbf{E}_{N+1}^R(\theta_2)$ in the substrate pass through the system of layers toward the surface. By analogy to Eq. (7), the amplitudes of the waves $\mathbf{E}_i^R(\theta_2)$ in each medium may be expressed using the recursive formula

$$E_i^R(\theta_2)=\frac{1+Q_i(\theta_2)}{1+Q_{i-1}(\theta_2)\varphi_i^2(\theta_2)}\varphi_{i+1}(\theta_2)E_{i+1}^R(\theta_2). \tag{28}$$

From Eq. (28), all the transmission coefficients $T_i(\theta_2)$ may be calculated as

$$T_i(\theta_2)=\frac{E_1^R(\theta_2)}{E_i^R(\theta_2)}. \tag{29}$$

If the amplitude of the wave $\mathbf{E}_i^R(\theta_2)$ in a medium i is too small, the calculation of the remaining transmission coefficients $T_j(\theta_2)$, $j\leq i$, has to be performed starting by the wave not in the substrate but in a layer closer to the surface.

By substituting Eqs. (22a)–(22d) for $\mathcal{S}_i$, the term $\mathcal{D}_0$ may be rewritten in the form

$$\mathcal{D}_0(\mathbf{q}_1)=\sum_{i=1}^{N}[\alpha_i(\mathbf{q}_1)F_i({}^1\xi_{i+1}+{}^2\xi_{i+1})+\beta_i(\mathbf{q}_1)F_i({}^1\xi_i-{}^2\xi_i)+\gamma_i(\mathbf{q}_1)F_i(-{}^1\xi_{i+1}+{}^2\xi_{i+1})+\delta_i(\mathbf{q}_1)F_i(-{}^1\xi_i-{}^2\xi_i)], \tag{30}$$

where α_i, β_i, γ_i, and δ_i are the following coefficients:

$$\alpha_i(\mathbf{q}_1)=k_0^2(n_i^2-n_{i+1}^2)E_i^T(\theta_1)t_{i,i+1}(\theta_1)t_{i,i+1}(\theta_2)\times\frac{T_i(\theta_2)}{1-R_i(\theta_2)Q_{i-1}(\theta_2)\varphi_i^2(\theta_2)}, \tag{31a}$$

$$\beta_i(\mathbf{q}_1)=k_0^2(n_i^2-n_{i+1}^2)E_{i+1}^R(\theta_1)\varphi_{i+1}(\theta_1)t_{i+1,i}(\theta_1)$$
$$\times t_{i,i+1}(\theta_2)\frac{T_i(\theta_2)}{1-R_i(\theta_2)Q_{i-1}(\theta_2)\varphi_i^2(\theta_2)}, \quad (31b)$$

$$\gamma_i(\mathbf{q}_1)=k_0^2(n_i^2-n_{i+1}^2)E_i^T(\theta_1)t_{i,i+1}(\theta_1)t_{i+1,i}(\theta_2)$$
$$\times\frac{T_{i+1}(\theta_2)R_{i+1}(\theta_2)\varphi_{i+1}(\theta_2)}{1-R_{i+1}(\theta_2)Q_i(\theta_2)\varphi_{i+1}^2(\theta_2)}, \quad (31c)$$

$$\delta_i(\mathbf{q}_1)=k_0^2(n_i^2-n_{i+1}^2)E_{i+1}^R(\theta_1)\varphi_{i+1}(\theta_1)t_{i+1,i}(\theta_1)t_{i+1,i}(\theta_2)$$
$$\times\frac{T_{i+1}(\theta_2)R_{i+1}(\theta_2)\varphi_{i+1}(\theta_2)}{1-R_{i+1}(\theta_2)Q_i(\theta_2)\varphi_{i+1}^2(\theta_2)}. \quad (31d)$$

The diffuse component of the differential cross section,[4]

$$\left(\frac{d\sigma}{d\Omega}\right)_{\rm diff}=\frac{\langle\mathscr{D}_0\mathscr{D}_0^*\rangle-\langle\mathscr{D}_0\rangle\langle\mathscr{D}_0^*\rangle}{16\pi^2}, \quad (32)$$

is then

$$\left(\frac{d\sigma}{d\Omega}\right)_{\rm diff}=\frac{1}{16\pi^2}\sum_{i=1}^{N}\sum_{j=1}^{N}[\alpha_i\alpha_j^*$$
$$\times F_{ij}({}^1\xi_{i+1}+{}^2\xi_{i+1},{}^1\xi_{j+1}+{}^2\xi_{j+1})+\ldots], \quad (33)$$

where the symbol $\langle\ \rangle$ denotes the process of averaging across the random rough interfaces. The term in parenthesis consists of 16 items, which differ only in the multiplicative factors and in the arguments of the functions F_{ij} defined as

$$F_{ij}(\kappa_1,\kappa_2)=\langle F_i(\kappa_1)F_j^*(\kappa_2)\rangle-\langle F_i(\kappa_1)\rangle\langle F_j^*(\kappa_2)\rangle. \quad (34)$$

Equation (33) is the general formula describing the relationship between the diffuse component of x-ray scattering from the layered system and the topography of the interfaces. The shape of function F_{ij} depends on the distribution of interface heights $h_i(x,y)$.

If $h_i(x,y)$ are Gaussian random variables with standard deviations σ_i, the function F_{ij} may be expressed in the form

$$F_{ij}(\kappa_1,\kappa_2)=\frac{L_xL_y}{\kappa_1\kappa_2^*}e^{-(\kappa_1^2\sigma_i^2+\kappa_2^{*2}\sigma_j^{*2})/2}$$
$$\times\int\int_{S_0}dX\,dY(e^{\kappa_1\kappa_2^*C_{ij}(X,Y)}-1)e^{i(q_xX+q_yY)}, \quad (35)$$

where

$$C_{ij}(X,Y)=\langle h_i(x+X,y+Y)h_j(x,y)\rangle \quad (36)$$

is the correlation function between height fluctuations of the interfaces i and j. We assume isotropic self-affine interfaces[15] described by the correlation functions

$$C_{ij}(R)=\sigma_{ij}^2e^{-(R/\tau_{ij})^{2H}}, \quad (37)$$

where the exponent H determines the texture of the roughness and takes values between 0 and 1. Values of H close to 1 describe smooth hills and valleys, whereas values of H approaching 0 characterize extremely jagged interfaces. The correlation function is exponential for $H=\frac{1}{2}$ and Gaussian for $H=1$. The parameters σ_{ij} and τ_{ij} are defined by means of the rms roughness σ_i and the correlation length τ_i of individual interfaces as

$$\sigma_{ij}^2=\frac{\sigma_i^2+\sigma_j^2}{2}e^{-|z_i-z_j|/\tau_z} \quad (38)$$

and

$$\tau_{ij}=\sqrt{\tau_i\tau_j}. \quad (39)$$

The parameter τ_z describes the tendency of individual layers to replicate the substrate surface (vertical correlation). For simplicity, τ_z is supposed to be the same for all layers of the system.

III. RESULTS AND DISCUSSION

In order to demonstrate results of the scattering theory presented in the previous section, the diffuse x-ray scattering (Cu K_α radiation) from a single layer and periodical multilayers has been calculated. The Gaussian random rough interfaces and the exponential shape of correlation functions have been assumed.

Three scanning modes were considered to map out the distribution of the diffusely scattered intensity in the reciprocal space.[16] In the θ_1 mode (transverse scan), the sample rotates and the scattering angle $\theta_1+\theta_2=2\theta$ is kept constant. The angle ω of the sample rotation is given by $\omega=(\theta_2-\theta_1)/2$. In the θ_2 mode, the angle of incidence remains fixed and the angular distribution of the scattered intensity is scanned. Finally, in the offset $(\theta,2\theta)$ scan the sample and analyzer are moved in such a way that the difference of angles θ_1 and θ_2 is conserved.

The theoretical θ_1 scans calculated for a gold layer on a silicon substrate are shown in Fig. 4. As follows from detailed numerical analysis, the contribution from the upper interface predominates in the nonspecularly scattered intensity and the correlation between the substrate and gold surface profiles has no appreciable effect on these scans. It is noteworthy that the diffuse scattering exhibits local maxima (known as Yoneda peaks)[17] if either θ_1 or θ_2 is equal to θ_c. This effect is caused by a standing wave with a maximum located at the surface, resulting in an enhanced diffuse scattering. Moreover, subsidiary maxima and minima may occur in the scattered intensity. These fringes arise due to the variation of the surface coherent field produced by the interference of the incident wave and the wave specularly reflected from the substrate. The differential cross sections are calculated in two ways. The former approach includes the influence of interfacial roughness on the local coherent field $\mathbf{E}_i^T$, $\mathbf{E}_i^R$ which affects the diffusely scattered field (30) through the coefficients (31). On the contrary, the latter approach corresponds to the use of exact eigenfunctions for the ideal layered structure for working out the perturbation theory (DWBA).[9] The difference between these two curves is more significant at larger angles [Fig. 4(b)]. Here, the reflection from the substrate is suppressed by the roughness, the modu-

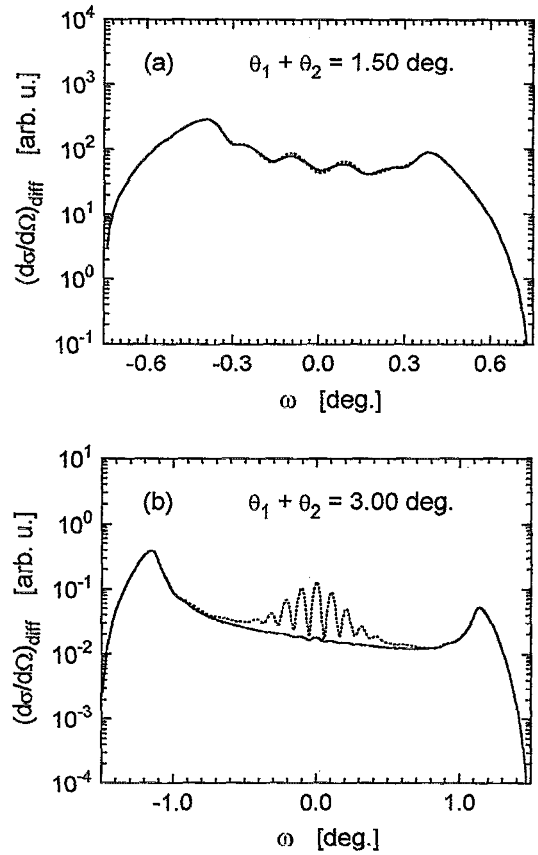

FIG. 4. Theoretical transverse scans for the gold layer (d_{Au}=20 nm) on the silicon substrate. The differential cross sections calculated using the coherent field within the stratified medium with rough (solid line) and smooth (dashed line) interfaces are compared. The rms roughness σ=2.0 nm and the correlation length τ=0.1 μm are assumed at both interfaces.

lation of the coherent field is therefore negligible, and the fringes disappear. Only the calculations starting from the coherent field within nonideal stratified medium give realistic results in this case.

Figure 5 depicts the influence of the vertical correlation[16,18,19] on the theoretical θ_2 scan. If the roughness is correlated (identical layered systems), the maxima and minima periodically alternate in the angular distribution of the scattered intensity. Similar to the Kiessig structure of reflectivity curves, these oscillations result from the interference between the waves diffusely scattered from the top and bottom interfaces. However, if the interface profiles are completely uncorrelated, nonspecular scattering monotonously decreases with the increasing angle θ_2.

For the demonstration of the diffuse scattering from more complicated structures, two W/Si multilayers have been chosen. These x-ray mirrors differ only in the thickness ratio of layers composed of heavy and light elements. The rms roughness σ=0.5 nm and the correlation length τ=1 μm are considered at each interface.

The series of theoretical transverse scans at 2θ values corresponding to the first-, second-, and third-order Bragg maxima are plotted in Figs. 6 and 7. The x-ray scattering from multilayers with correlated and uncorrelated roughness is compared. These plots show several interesting features. When the wave vector of the incident or scattered wave fulfils the Bragg condition,[20] the sharp maxima arise. However, satellite minima may sometimes occur in these positions. This somewhat surprising but already experimentally observed phenomenon[13,16,21] can be explained within the framework of the theory presented above. When one of the angles θ_1, θ_2 approaches the Bragg angle, not only are the interference effects and the coherent field modulations important but, in addition, the transmission of the incident wave or the waves scattered from individual boundaries exhibits a local extreme. Whether the increase or the decrease of the transmission occurs it depends particularly on the ratio of W and Si in the bilayer and on the number of periods. That is why no decision—which effect is dominant and whether the scattering in a given direction is enhanced or attenuated—may be done without carrying out the numerical calculations.

For uncorrelated roughness, $\tau_z \rightarrow 0$, $C_{ij} \rightarrow 0$ if $i \neq j$ and the sum in the relation (33) for the diffuse component of the differential cross section has only N nonzero terms. On the other hand, if the roughness is fully conformal, $\tau_z \rightarrow \infty$ and all terms contribute to the total diffuse scattering. Therefore the nonspecular intensity is usually in the former case much smaller. That is why the vertical correlation is very undesirable at layered systems used as image elements, because it causes the contrast degradation of the final image. But the opposite situation may also occur and the incoherent scattering from the layered structure having uncorrelated interfaces may be comparable or even higher [Fig. 6(c)] than for the identical system. The reason why the multilayer with correlated interface profiles exhibits a very low diffusely scattered intensity at the third-order Bragg maxima is the fact that the third order is nearly structure factor forbidden. Another interesting feature is also that incoherent scattering from systems with completely uncorrelated roughness may show fine structure but this is mostly less apparent. This can be explained by the effect of primary extinction in the vicinity of Bragg angles[22] (reduced or enhanced transmission if the nodes of the standing wave pass through the W or Si layers).

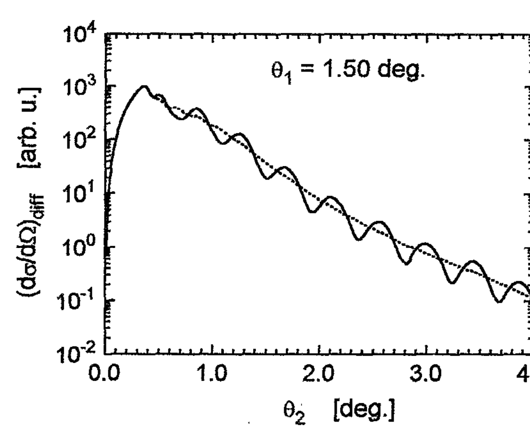

FIG. 5. Theoretical θ_2 scans for the gold layer (d_{Au}=20 nm) with correlated (solid line) and uncorrelated (dashed line) interface profiles. The rms roughness σ=1.0 nm and the correlation length τ=0.1 μm are considered at both interfaces.

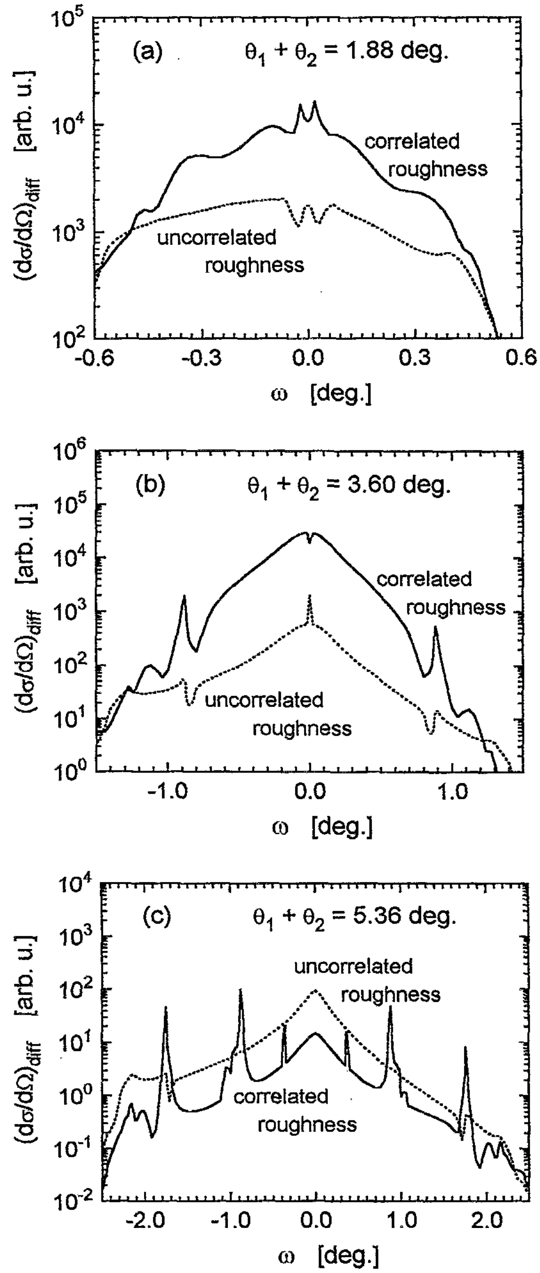

FIG. 6. Transverse scans calculated for the multilayer W/Si (d_W=1.7 nm, d_{Si}=3.3 nm, 40 periods) at (a) the first-, (b) second-, and (c) third-order Bragg maxima.

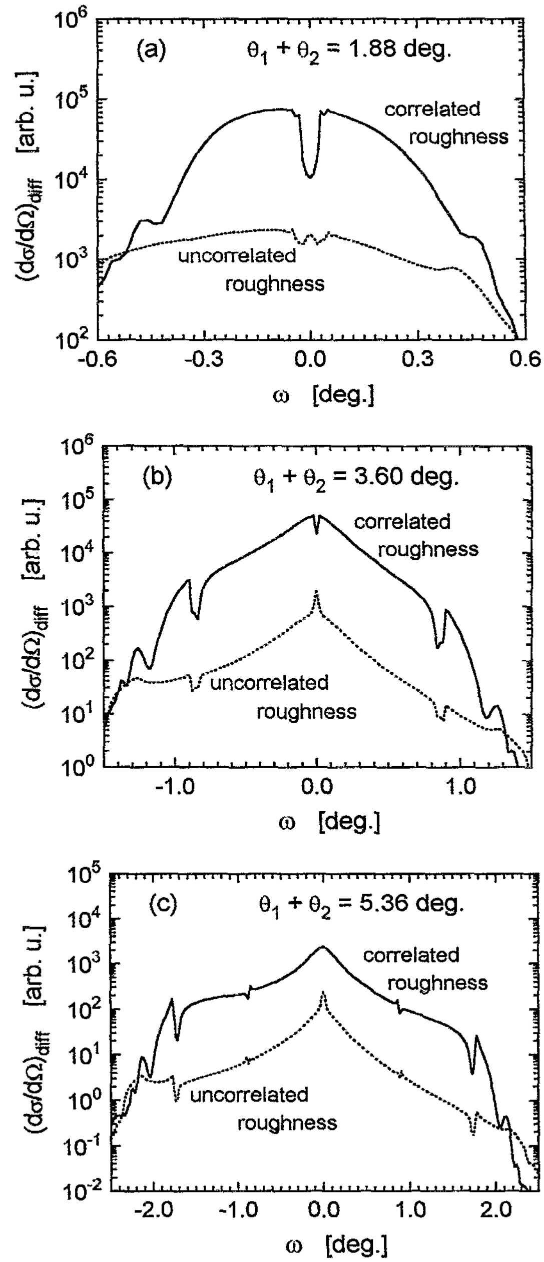

FIG. 7. Transverse scans calculated for the multilayer W/Si (d_W=1.2 nm, d_{Si}=3.8 nm, 40 periods) at (a) the first-, (b) second-, and (c) third-order Bragg maxima.

The off (θ,2θ) scan measurement appears to be a very promising method for determining vertical correlation. The numerical calculations were carried out for the multilayer consisting only of 10 pairs of Fe/C (Fig. 8). A smaller number of periods was chosen in order to reach a better resolution of subsidiary interference fringes. This scan exhibits no fine structure for completely uncorrelated interfaces. With increasing vertical correlation, the interference maxima start to appear in the regions where the angle of scattering 2θ is equal to twice the Bragg angle. These peaks correspond to the constructive interference of waves diffusely scattered from individual bilayers in a full analogy to the Bragg peaks in the specular reflection. For the parameter $\tau_z = 0.2$ μm, not only the profiles of adjoining boundaries are correlated but a certain degree of correlation also exists between the roughness of the substrate and of the surface. This again results in fast oscillations equivalent to the Kiessig ones.

The presented results show that the structure of layered systems has a strong impact on their scattering properties.

IV. SUMMARY

In this paper, the theory of diffuse x-ray scattering from rough layered systems based on the distorted-wave Born approximation has been presented. To author's knowledge, this

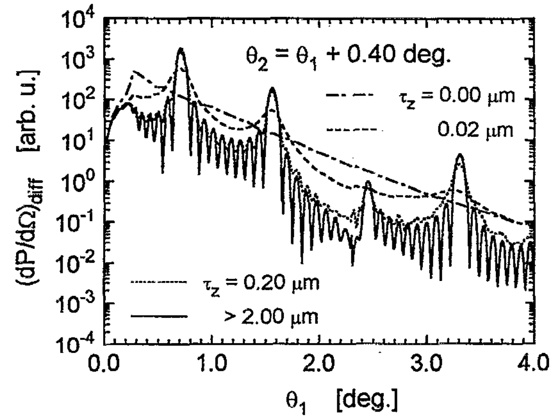

FIG. 8. Theoretical offset $(\theta,2\theta)$ scans for multilayers Fe/C ($d_{Fe}=1.77$ nm, $d_c=3.28$ nm, 10 periods) with different vertical correlation. The same values of $\sigma=0.5$ nm and $\tau=1.0$ μm are considered at each interface.

is the first model valid also at small glancing angles, where the calculation of the diffusely scattered intensity respects both the real coherent field within a nonideal stratified medium and a real transmission of waves scattered from inner boundaries.

By using the above-mentioned approach, the Yoneda anomalous scattering of x rays can be described. Further, the modulation in the nonspecularly scattered intensity can be explained as the consequence of dynamical effects. The scattering from periodical multilayers exhibits satellite maxima and minima when θ_1 or θ_2 approaches the Bragg angle. These features depend on the mutual correlation between individual boundaries but they can be observed for completely correlated as well as for uncorrelated interface profiles.

This scattering theory may be also used for fitting experimental data. In this way, basic statistical parameters (root-mean-square roughness, correlation function) of random rough surfaces and interfaces within layered structures may be quantified. Different scattering properties of systems with correlated and uncorrelated roughness enables one to specify the level of vertical correlation.

A paper devoted to the characterization of layered structures by coherent and incoherent scattering of x rays is being prepared.

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