

Method of Transfer Matrices and Dynamical Thick-Crystal Approximation in Surface X-ray Diffraction by Multilayer Structures¹

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Abstract – A method for simulating X-ray grazing-incidence (surface) diffraction from multilayer crystal structures and superlattices was developed. The method is based on the solution of the problem of dynamical X-ray diffraction in each layer, the calculation of 4×4 transfer matrices for layers, and the use of dynamical thick-crystal approximation. The respective computations showed that the results are reproducible for $\approx 10^2$ and even more layers. Therefore, this method can be very useful for theoretical calculations of surface structures by X-ray diffraction data.

1. INTRODUCTION

In¹ recent years, surface X-ray diffraction (SXRD) has been successfully used for studying the perfection of multilayer structures [1 - 7]. But until now, the theoretical models have been developed only for certain cases. Thus, Golovin *et al.* [1] suggested a method for interpreting surface diffraction for several amorphous layers on the crystal surface. Golovin *et al.* [2] also suggested a method for interpreting surface diffraction from crystalline surface layers for the limiting asymmetric case, while Pietsch [5] considered surface diffraction from ideal periodic superlattices. Melikyan [8] developed a method for simulating surface diffraction proceeding from the kinematical theory of X-ray diffraction. But this method cannot satisfactorily describe scattering in perfect structures because of the initial assumptions made. Andreeva *et al.* [9, 10] derived differential equations of surface diffraction in the matrix form, but the cumbersome and unstable solutions of the equations for thick crystals make them inapplicable for processing experimental data. Finally, Rhan and Pietsch [3] derived a system of equations for an arbitrary sequence of crystalline and amorphous layers on the crystal surface and suggested that this system can be solved numerically using the Gauss method. Unfortunately, in practice, this method cannot be used for more than five to six layers owing to the rounding-off errors in the numerical solution of the system of $4 \times N_l$ equations, where N_l is the number of layers. Thus, in the general case, quantitative analysis of the experimental data on surface diffraction from multilayer structures has been impossible.

In Section 2, I suggest a modification of the Rhan and Pietsch method [3]. It reduces the computations to 4×4 matrices, increases the accuracy of these compu-

tations, and extends them to structures with a large number of surface layers. Section 3 considers the dynamical thick-crystal approximation chosen for simulating surface diffraction from multilayer structures and illustrates its practical use.

2. THE MATRIX METHOD

Let us consider surface diffraction from a multilayer structure formed by an arbitrary sequence of amorphous and crystalline layers. I assume that the lattice constant is the same for all the surface crystalline layers, which is usually the case.² In the approximation of dynamical diffraction, the wave field in vacuum outside the crystal and in surface layers (for both polarizations) can be written in the following way (see, e.g., [3]):

in vacuum

$$E_v(r) = \{E_0 \exp(ix\Phi_0 z) + E_s \exp(-ix\Phi_0 z)\} \times \exp(ix_{\perp} r_{\perp}) + \{E_h \exp(ix\Phi_h z)\} \exp[i(x+h)_{\perp} r_{\perp}], \quad (1)$$

in crystalline layers

$$D_c^k(r) = \left\{ \sum_{j=1}^4 D_j^k \exp(ixu_j^k z) \right\} \exp(ix_{\perp} r_{\perp}) + \left\{ \sum_{j=1}^4 V_j^k D_j^k \exp[ix(u_j^k + \psi)z] \right\} \exp[i(x+h)_{\perp} r_{\perp}], \quad (2)$$

and in amorphous layers

² It should be noted that crystalline layers with spacings differing by $\Delta d/d > 10^{-4}$ give no contributions to diffraction and, therefore, can be considered as amorphous.

¹ In memory of Z.G. Pinsker.

$$D_a^k(r) = \{D_0^k \exp(ixu_0^k z) + D_{0s}^k \exp(-ixu_0^k z)\} \exp(ix_{\perp} r_{\perp}) + \{D_h^k \exp(ixu_h^k z) + D_{hs}^k \exp(-ixu_h^k z)\} \exp [i(x+h)_{\perp} r_{\perp}] \quad (3)$$

where $E_0 = 1$, E_s , and E_h are the amplitudes of the incident, specularly reflected, and diffracted waves, respectively. The superscript $k = 1, \dots, N$ is the layer number in a multilayer structure; x is the wavevector of X-rays ($x = |x|$); h is the reciprocal-lattice vector providing surface diffraction; Φ_0 and Φ_h are the angles of entrance and departure with respect to the surface; and u_j^k are the solutions of the fourth-order dispersion equation for dynamical diffraction in the k th crystalline layer ($j = 1, \dots, 4$):

$$(u_j^k - \Phi_0^2 - \chi_0^k) [(u_j^k + \psi)^2 - \Phi_h^2 - \chi_0^k] = \chi_h^k \chi_h^k, \quad (4)$$

where χ_0^k , χ_h^k , and χ_h^k are X-ray polarizabilities of the k th layer; $\psi = 2\delta \sin(\theta_B)$; δ is the angle of misorientation of the h vector with respect to the surface; $V_j^k = (u_j^k - \Phi_0^2 - \chi_0^k) / \chi_h^k$ are the amplitude ratios of the diffracted and transmitted waves for the j th solution (4);

and $u_{0,h}^k = \sqrt{\Phi_{0,h}^2 + \chi_0^k}$. It is seen that four wave fields are excited in each layer. It is known [11] that equation (4) has four solutions: two attenuating and two enhancing with the crystal depth. They are characterized by $\text{Im} u_j > 0$ and $\text{Im} u_j < 0$, respectively. The enhancing solutions are considered as the waves reflected from the lower layer boundary. Therefore, the waves with $\text{Im} u_j < 0$ for the last layer (substrate) are taken to be very weak and can be eliminated from formulas (2) and (3) (the so-called thick-crystal approximation).

Thus, for a structure consisting of N layers, I arrive at $4N$ unknown wave amplitudes: two in vacuum (E_s , E_h) and $4(N-1) + 2$ in the crystal. These amplitudes can be determined from the set of $4N$ boundary conditions at N layer boundaries for the fields and their derivatives [3]. In particular, the equations at the vacuum–first crystalline layer interface can be written in the following way:

$$E_0 + E_s = \sum_{j=1}^4 D_j^1, \quad E_h = \sum_{j=1}^4 V_j^1 D_j^1, \quad (5)$$

$$\Phi_0(E_0 - E_s) = \sum_{j=1}^4 u_j^1 D_j^1, \quad -\Phi_h E_h = \sum_{j=1}^4 w_j^1 D_j^1.$$

The equations for the boundaries between two crystalline layers and between two amorphous layers are written as

$$\sum_{j=1}^4 f_j^k D_j^k = \sum_{j=1}^4 D_j^{k+1}, \quad \sum_{j=1}^4 f_j^k V_j^k D_j^k g_k = \sum_{j=1}^4 V_j^{k+1} D_j^{k+1},$$

$$\sum_{j=1}^4 f_j^k u_j^k D_j^k = \sum_{j=1}^4 u_j^{k+1} D_j^{k+1}, \quad (6)$$

$$\sum_{j=1}^4 f_j^k w_j^k D_j^k g_k = \sum_{j=1}^4 w_j^{k+1} D_j^{k+1};$$

$$f_0^k D_0^k + f_0^{k-1} D_{0s}^k = D_0^{k+1} + D_{0s}^{k+1},$$

$$f_h^k D_h^k + f_h^{k-1} D_{hs}^k = D_h^{k+1} + D_{hs}^{k+1},$$

and

$$u_0^k (f_0^k D_0^k + f_0^{k-1} D_{0s}^k) = u_0^{k+1} (D_0^{k+1} + D_{0s}^{k+1}),$$

$$u_h^k (f_h^k D_h^k + f_h^{k-1} D_{hs}^k) = u_h^{k+1} (D_h^{k+1} + D_{hs}^{k+1}),$$

respectively. I used the following notation:

$f_j^k = \exp(ixr^k u_j^k)$, $g^k = \exp(ixr^k \psi)$, $f_{0,h}^k = \exp(ixr^k u_{0,h}^k)$; t^k is the thickness of the k th layer.

As I have already mentioned, Rhan and Pietsch [3] suggested that the system of $4N$ boundary conditions be solved by using the Gauss method. Here, I used a different method.

Let us rewrite the equations at each interface in matrix form. With this aim, the four-component D^k vectors and the 4×4 a^k and b^k matrices for the right- and left-hand sides of the boundary equations are introduced:

$$D_{cr}^k = \begin{pmatrix} D_1^k \\ D_2^k \\ D_3^k \\ D_4^k \end{pmatrix}, \quad D_{am}^k = \begin{pmatrix} D_0^k \\ D_h^k \\ D_{0s}^k \\ D_{hs}^k \end{pmatrix}; \quad (8)$$

$$a_{cr}^k = \begin{pmatrix} 1 & 1 & 1 & 1 \\ V_1^k & V_2^k & V_3^k & V_4^k \\ u_1^k & u_2^k & u_3^k & u_4^k \\ w_1^k & w_2^k & w_3^k & w_4^k \end{pmatrix}, \quad a_{am}^k = \begin{pmatrix} 1 & 0 & 1 & 0 \\ 0 & 1 & 0 & 1 \\ u_0^k & 0 & -u_0^k & 0 \\ 0 & u_h^k & 0 & -u_h^k \end{pmatrix}; \quad (9)$$

$$b_{cr}^k = G^k a_{cr}^k F^k, \quad b_{am}^k = a_{am}^k F^k; \quad (10)$$

where

$$F_{ij}^k = f_j^k \delta_{ij}, \quad G_{ij}^k = g_j^k \delta_{ij}, \quad g_i^k = \begin{cases} 1, & \text{if } i = 1, 3 \\ g_k, & \text{if } i = 2, 4. \end{cases}$$

In accordance with the thick-crystal approximation, the vector D^k for the substrate has only two components, and the matrix a^k has only two columns.

Let us also introduce the following quantities for the fields in vacuum (above the crystal surface): a four-component incident vector E^i , a two-component

vacuum vector E^v , and a 2×4 vacuum matrix b^v in the form:

$$E^i = \begin{pmatrix} 1 \\ 0 \\ \Phi_0 \\ 0 \end{pmatrix}, E^v = \begin{pmatrix} E_s \\ E_h \end{pmatrix}, \text{ and } b^v = \begin{pmatrix} 1 & 0 \\ 0 & 1 \\ -\Phi_0 & 0 \\ 0 & -\Phi_h \end{pmatrix}. \quad (11)$$

The boundary conditions can then be rewritten in the form

$$\begin{aligned} b^v E^v + E^i &= a^1 D^1, \\ b^1 D^1 &= a^2 D^2, \\ &\dots\dots\dots \\ b^k D^k &= a^{k+1} D^{k+1}, \\ &\dots\dots\dots \\ b^{N-1} D^{N-1} &= a^N D^N, \end{aligned} \quad (12)$$

where $D^N \equiv D^s$ and $a^N \equiv a^s$ are a two-component vector and a 2×4 matrix for the substrate, respectively.

Thus, it is evident that the complete system of equations is quasi-diagonal

$$\begin{pmatrix} b^v & -a^1 & 0 & 0 & \dots & 0 & 0 & 0 \\ 0 & b^1 & -a^2 & 0 & \dots & 0 & 0 & 0 \\ 0 & 0 & b^2 & -a^3 & \dots & 0 & 0 & 0 \\ 0 & 0 & 0 & b^3 & \dots & 0 & 0 & 0 \\ \dots & \dots \\ 0 & 0 & 0 & 0 & \dots & -a^{N-2} & 0 & 0 \\ 0 & 0 & 0 & 0 & \dots & b^{N-2} & -a^{N-1} & 0 \\ 0 & 0 & 0 & 0 & \dots & 0 & b^{N-1} & -a^N \end{pmatrix} \quad (13)$$

$$\times \begin{pmatrix} E^v \\ D^1 \\ D^2 \\ D^3 \\ \dots \\ D^{N-2} \\ D^{N-1} \\ D^N \end{pmatrix} = \begin{pmatrix} -E^i \\ 0 \\ 0 \\ 0 \\ \dots \\ 0 \\ 0 \\ 0 \end{pmatrix},$$

and, therefore, solution (12) can be substantially simplified. If one begins to solve system (12) with the last equation, one arrives at

$$b^v + E^i = a^1(b^1)^{-1} \dots a^{N-1}(b^{N-1})^{-1} a^N D^N \quad (14)$$

or

$$b^v + E^i = (T^1 \dots T^{N-1} a^N) D^N.$$

From now on, the products $T^k = a^k(b^k)^{-1}$ are to be called the transfer or T matrices of the layers. It follows from (10) that

$$T_{cr}^k = a^k(F^k)^{-1}(a^k)^{-1}G^k, \text{ and } T_{sm}^k = a^k(F^k)^{-1}(a^k)^{-1}. \quad (15)$$

Here $(F^k)_{ij}^{-1} = (f_i^k)^{-1}\delta_{ij}$, and $(G^k)_{ij}^{-1} = (g_i^k)^{-1}\delta_{ij}$.

If the layer thickness tends to zero, then

$$F^k \rightarrow I, \quad G^k \rightarrow I, \quad T^k \rightarrow [a^k(a^k)^{-1}] \rightarrow I.$$

In other words, the T matrix of an infinitely thin layer tends to the unit diagonal matrix.

Equation (14) has a simple physical interpretation. If all $i^k = 0$, the equation coincides with the equation of surface diffraction for a perfect crystal. If the crystal surface consists of several layers, the matrix of the substrate is modified by multiplying the T matrices of all the layers:

$$\bar{a}^s = T^1 \dots T^{N-1} a^s. \quad (16)$$

Equations (14), with regard for (16), form the system of four equations for four unknown amplitudes E_0, E_s, D_1^s , and D_2^s :

$$\bar{a}^s D^s - b^v E^v = E^i \quad (17)$$

or

$$\begin{cases} \bar{a}_{11}^s D_1^s + \bar{a}_{12}^s D_2^s - E_s = 1 \\ \bar{a}_{21}^s D_1^s + \bar{a}_{22}^s D_2^s - E_h = 0 \\ \bar{a}_{31}^s D_1^s + \bar{a}_{32}^s D_2^s - \Phi_0 E_s = \Phi_0 \\ \bar{a}_{41}^s D_1^s + \bar{a}_{42}^s D_2^s - \Phi_h E_h = 0. \end{cases} \quad (17a)$$

System (17) can easily be solved by any method.

Thus, the solution considered above has the advantage of using no operations with matrices of ranks higher than 4×4 and, therefore, should be stable in computations.

In order to implement the algorithm in practice, one must

- calculate the a^k matrices,
- determine the inverse matrices numerically by any standard algebraic method,
- calculate the T^k matrices and their product, and
- solve the system of equation (17) by the Gauss or any other method.

The reflection coefficients are calculated by the following standard formulas:

$$P_s = |E_s|^2, \quad (18)$$

$$P_h = (\Phi_h / \Phi_0) |E_h|^2. \quad (19)$$

The experimental P_h values are often measured as functions of angle Φ_h . In this case, one must multiply the right-hand side of (19) by $2\Phi_h$ [12].

3. DYNAMICAL THICK-CRYSTAL APPROXIMATION

Allow a multilayer structure to be sufficiently thick, for example, 10^4 Å. It is known that the X-ray field in surface diffraction attenuates in the crystal at a depth of about 10^2 Å. The question arising is whether it is possible to attain the maximum calculational accuracy if one takes into account the contributions that come to surface diffraction from all the layers. Below, I will show that, in most cases, the answer to this question is negative.

To clarify the situation, let us formally apply the algorithm described in the previous section to a multilayer structure consisting of N identical layers (i.e., to a perfect crystal). For the sake of simplicity, let us assume that $\psi = 0$, and, therefore, $G^k = I$. Then, equations (14) and (15) yield

$$\begin{aligned} \bar{a}^s &= \underbrace{(aF^{-1}a^{-1})(aF^{-1}a^{-1}) \dots (aF^{-1}a^{-1})}_{(N-1) \text{ times}} a^s \\ &= a(F^{-1})^{N-1}(a^{-1}a^s) = a^s(F^{-1})^{N-1}, \end{aligned} \quad (20)$$

where $(F^{-1})^{N-1}$ is the diagonal matrix consisting of two increasing exponents corresponding to two attenuating solutions with $\text{Im}u_j > 0$

$$[(F^{-1})^{N-1}]_{ji} = \delta_{ji} \exp[-ix(N-1)tu_j], \quad j, l = 1, 2. \quad (21)$$

It follows from (20) and (21) that, if the total thickness of all the layers $t_{\text{tot}} = (N-1)t$ is either small or comparable with the depth of X-ray field attenuation in surface diffraction [$t_{\text{at}} = \max(x\text{Im}u_j)^{-1}$], then the matrix elements \bar{a}^s and a^s are of the same order of magnitude and equation (17) can easily be solved numerically. If $t_{\text{tot}} \gg t_{\text{at}}$, then the exponents in (21) have high values, and $\|\bar{a}^s\| \gg \|a^s\|$. It follows from (17) that, in this case, the solutions $D_1^j(t_{\text{tot}})$ and $D_2^j(t_{\text{tot}})$ should be small. In accordance with (2), I have

$$D_j^s(t_{\text{tot}}) = D_j^s(0) \exp(ixt_{\text{tot}}u_j). \quad (22)$$

Substitution of (22) into (21) allows one to diminish the large exponents in (21) and to consider the equations for a perfect crystal. But the attempt to solve equation (17) on a computer without renormalization would result in errors of the accuracy loss type because of a large difference between the orders of magnitude of \bar{a}^s and a^s . Thus, if $t_{\text{tot}} = 10^4$ and $t_{\text{at}} = 50$ Å, the exponents in (19) attain unrealistic values of 10^{100} .

The problem of loss of accuracy can be solved in two ways:

(1) Similar to the previous example, I can renormalize the amplitudes in each layer of a multilayer structure with regard for attenuation. This method seems to

be quite natural for a perfect crystal, but meets obvious difficulties for multilayer structures characterized by different attenuation within each layer.

(2) A more physical approach takes diffraction into account only in those upper layers of the multilayer structure where X-ray fields are characterized by substantial amplitudes, but it ignores the contributions from deeper layers. This approach corresponds to the standard thick-crystal approximation known in X-ray diffraction.

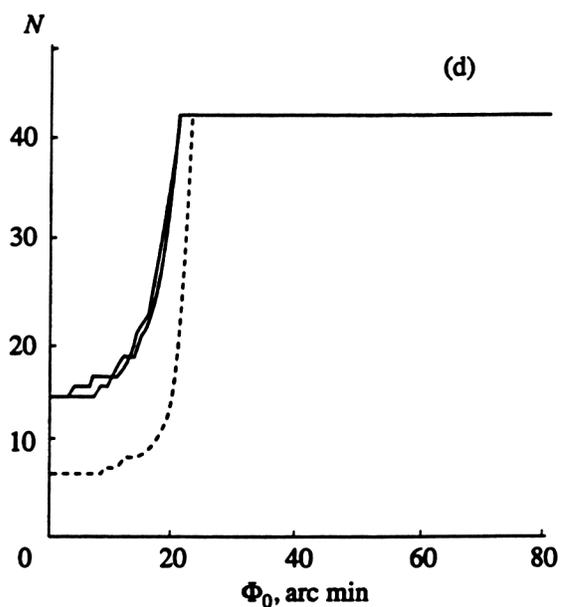
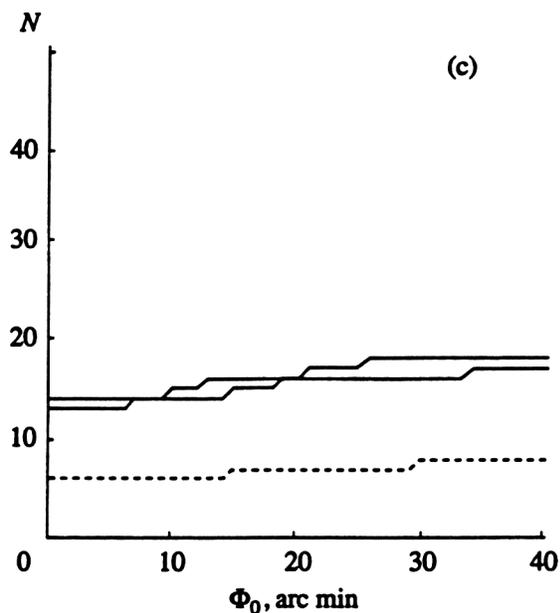
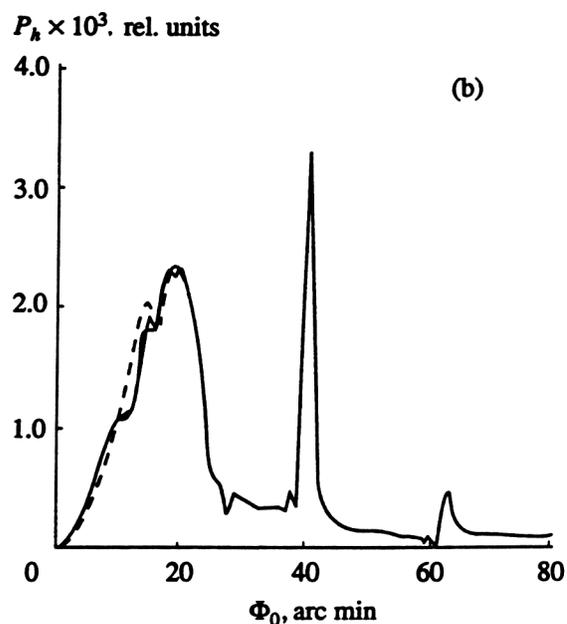
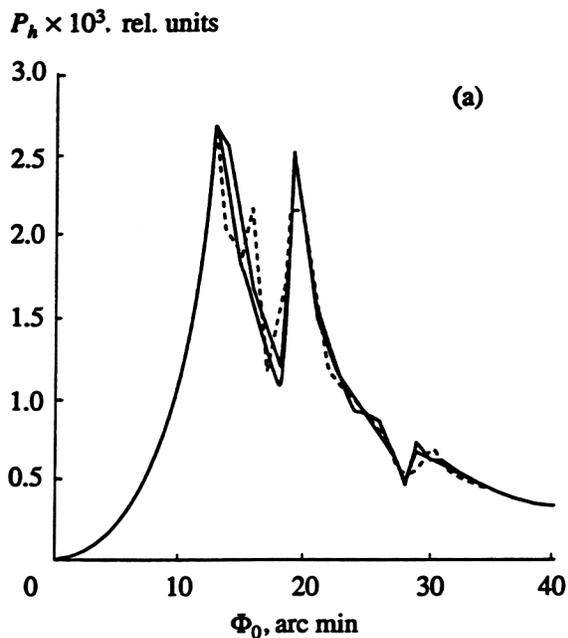
In practice, the thick-crystal approximation can be used in the following way: When calculating the product of T matrices in (16), one should attentively follow the behavior of the maximum element of the matrix-product after each matrix multiplication by the T matrix of each following layer (beginning with the first layer). If the product attains a very large value (for example, 10^{10} to 10^{15}), the X-ray field in the respective layer becomes very weak, and the reflection of X-ray waves from the lower boundary of this layer and all of the deeper layers can be neglected. In other words, the layer is considered as an infinitely thick substrate, and only the waves with $\text{Im}u_j > 0$ are taken into consideration.

Since the attenuation depth of X-ray waves in the crystal in surface diffraction is essentially dependent on angles Φ_0 and Φ_h , the number of the layers that should be taken into account varies from point to point on the diffraction curve (the figure). Therefore, the above suggested procedure can be called the dynamical thick-crystal approximation.

The figure shows the curves of surface diffraction from the AlAs/GaAs superstructures calculated by the matrix method in the dynamical thick-crystal approximation. The calculational parameters are (220) reflection, $\text{CuK}\alpha$ radiation, π -polarization, and $\psi = 0$. The substrate parameters are a GaAs crystal, the superstructure consisting of twenty (70 Å AlAs + 150 Å GaAs) periods. The figure (a and b) shows the diffraction curves at the incidence angles $\Phi_0 = 10'$ (the figure, a) and $\Phi_0 = 20'$ (the figure, b). The figures (c and d) shows the respective curves depending on the numbers of layers taken into account in the dynamical calculations.

It is seen that, on the whole, the suggested method correctly represents the surface diffraction pattern from the superlattice, i.e., the location of the peaks of Bragg reflections of various orders from a periodic layer structure (see, e.g., [6, 7]). If the incidence angle is $\Phi_0 = 10'$ (the penetration depth of X-rays is small), no more than 8 to 18 layers from the total 41 are taken into account. Therefore, the Bragg peaks from the superlattice at this angle are weakly marked. If $\Phi_0 = 20'$, the reflections from all the superlattice layers should be taken into account along virtually the entire diffraction curve, and we observe bright Bragg peaks.

The figure (a and b) shows the diffraction curves calculated for different thresholds (10^5 , 10^{13} , and 10^{14}) of the thick-crystal approximation with a computer accuracy of 18 significant digits. It is seen that



(a, b) The curves of surface X-ray diffraction from the superlattice consisting of twenty (70 \AA AlAs + 50 \AA GaAs) periods calculated by the matrix method, and (c, d) the diagrams illustrating the choice of the number of reflecting layers for dynamical calculations. Angles of incidence (a, c) 10° and (b, d) 20° . Solid lines correspond to 10^{13} and 10^{14} thresholds of the thick-crystal approximation; the dashed line corresponds to the threshold 10^5 .

the change of the threshold value by nine orders of magnitude does not qualitatively change the computational result. Therefore, this method is rather stable.

Thus, I developed an algorithm for simulating surface X-ray diffraction from superlattices and other multilayer structures. Unlike the algorithm suggested earlier [3], this algorithm is quite reliable for a large number of layers. In contrast to the algorithm suggested in [6], this algorithm can be applied to layer structures having no rigorous periodicity. In particular, the method allows one to perform the calculations for arbitrary $\chi_0(z)$, $\chi_h(z)$, and

$\delta r(z)$ distributions, i.e., provides the description of various inhomogeneities in superlattices. This makes the suggested algorithm very promising for studying surface structures by surface X-ray diffraction.

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