J. Moscow Phys. Soc. 1 (1991) 425–434. Printed in the UK

A theory of multiple Bragg diffraction of x rays in multilayer crystal systems

V G Kohn

I V Kurchatov Institute of Atomic Energy, ploshchad' Kurchatova 14, 123182 Moscow, USSR

Received 27 July 1991

Abstract. This paper presents a general theory of multiple diffraction of x rays in a perfect crystal on whose surface a system of epitaxial films has been grown, without restrictions on film thickness or diffraction geometry. In the general case some of the diffracted rays pass through the plate (Laue diffraction) and some are reflected and exit from the plate through the entrance surface (Bragg diffraction). The author develops a method for calculating the intensities of the diffracted waves in one crystal layer in terms of matrix algebra and the explicit isolation of Laue–Laue, Bragg–Bragg, and Laue–Bragg scattering. The general problem of calculating the parameters of multiple diffraction in a multilayer system is solved by the method of matrix recurrence formulae. A concrete example of applying the theory is given, namely, the three-wave (333, 113) diffraction of radiation Cu Kα in a crystal with a homogeneous epitaxial film in which the interplanar distance (111) is slightly changed. The method of direct calculation of the angular dependence of the intensity of the weak (113) ray is used to show the possibility of measuring directly the variation in the phase of the wave reflected from the (333) plane, the variation being caused by distortions of the crystal lattice in the film.

1. Introduction

The phenomenon of multiple diffraction of any radiation with wavelengths of the order of the interatomic distance in a crystal is widely known and can be easily observed experimentally. It emerges when a certain relation between the wavelength and the direction of propagation of a ray in the crystal is met and consists in a strong diffractive reflection of the plane wave from several systems of crystal planes simultaneously. For x rays of a given frequency ω multiple diffraction is realized inside a narrow cone near certain directions specified by a unit vector $\boldsymbol{s}_0$ whose two free components satisfy the Bragg law

$$(K\boldsymbol{s}_0 + \boldsymbol{h}_m)^2 = K^2 \tag{1}$$

where $K = \omega/c = 2\pi/\lambda$, with λ the wavelength, and $\boldsymbol{h}_m$ is the reciprocal lattice vector with number m.

The angular deviation of the ray from the direction of $\boldsymbol{s}_0$ must not exceed a quantity of the order of the scattering amplitude, whose role in the present case is played by the Fourier component of the crystal's polarizability, χ_h. In standard conditions the absolute value of χ_h is of the order of 10^{-5}. Hence, for the experimental

0960-0175/91/040425+10$03.50

investigation of the multiple diffraction of x rays in pure form, the collimation of the incident ray in two planes must be no worse than several fractions of an angular second. This is an extremely stringent requirement that has been realized in experiments only recently. Kov'ev and Simonov [1] were the first to use a doubly collimated ray in studies of multiple diffraction of x rays. By measuring the angular dependence of the intensities of the diffracted rays they found in a direct manner the phase of the scattering amplitude χ_h in a perfect silicon crystal, that is, the phase problem of structural analysis was solved (in this particular case). Applying synchrotron radiation [2] enabled shortening the time of the experiment drastically, which makes the method of multiple diffraction extremely promising for a great number of problems in which the structure of perfect crystals and near-the-surface layers is studied.

The basics of the theory of multiple diffraction of x rays were formulated in the works of Ewald and Laue in the first half of this century, but to this day only the theory of perfect crystals has been well developed [3]. Lately, however, science and technology have made extensive use of perfect crystals with a complex system of epitaxial films of various composition grown on the crystal's surface. The study of such systems by the multiple-diffraction method is currently hindered by the total absence of a theory or sufficiently clear understanding of the potential of the method.

The present paper is devoted to this problem. It presents a rigourous theory of multiple diffraction of x rays in multilayer crystal systems with layers of arbitrary thickness. The approach considers the most general and complicated (from the standpoint of analysis) case, namely, when a fraction of the rays are reflected back and leave the crystal through the entrance surface (Bragg diffraction). Owing to the complex nature of the boundary conditions, the problem proves to be self-consistent. The method of matrix recurrence formulae developed to solve this problem can, naturally, be employed in similar problems with any type of radiation (electrons, neutrons, or Mössbauer gamma rays). The discussion in this paper is restricted to the case where the diffraction angle is not very small and mirror-reflected waves can be ignored.

2. Statement and solution of the problem in one crystal plate

The propagation of x rays in a medium can be described fairly accurately by the Maxwell equations with allowance for the quantum mechanical nature of the response of the medium to electromagnetic radiation. For the Fourier components of the electric field, $\boldsymbol{E}(\boldsymbol{k},\omega)$, we have the following equation:

$$(k^2 - K^2)\boldsymbol{E}(\boldsymbol{k},\omega) - \boldsymbol{k}(\boldsymbol{k}\cdot\boldsymbol{E}(\boldsymbol{k},\omega)) = \frac{4\pi i\omega}{c^2}\boldsymbol{j}(\boldsymbol{k},\omega) \tag{2}$$

where $\boldsymbol{j}(\boldsymbol{k},\omega)$ is a Fourier component of the induced-current density and is calculated as the quantum mechanical average of the current-density operator over the state of the crystal in the radiation field [4]. Allowing for the periodicity of the crystal lattice and the weakness of the interaction (linear in the field strength), we can generally express the current in terms of the field as an expansion in reciprocal lattice vectors:

$$j^i(\boldsymbol{k}_0,\omega) = \frac{c^2K^2}{4\pi i\omega}\sum_{mj}\chi^{ij}_\omega(\boldsymbol{k}_0,\boldsymbol{k}_m)E^j(\boldsymbol{k}_m,\omega) \tag{3}$$

where $\boldsymbol{k}_m = \boldsymbol{k}_0 + \boldsymbol{h}_m$, and $\chi^{ij}_\omega(\boldsymbol{k}_0,\boldsymbol{k}_m)$ is a frequency dependent Fourier component of the crystal's complex-valued polarizability tensor, which allows for all types of

scattering of x rays, both the classical Thomson scattering and resonance scattering (the photoelectric effect, Compton scattering, and diffuse thermal scattering [5]).

Suppose that a plane wave with a wavevector $\boldsymbol{K}_e \approx K\boldsymbol{s}_0$, where $\boldsymbol{s}_0$ satisfies the Bragg law (1), is incident on the upper surface of a crystal shaped as a plate of thickness t. The only component of the wavevector that can change inside the crystal is the one parallel to the inward unit normal $\boldsymbol{n}_0$ to the surface of the plate, that is, $\boldsymbol{k}_0 = \boldsymbol{K}_e + \varepsilon\boldsymbol{n}_0/2$. The complex-valued quantity ε is defined in such a way that its imaginary part is equal to the absorption coefficient for the wave propagating to within the crystal. Next we substitute (3) into (2) and allow for the fact that $\boldsymbol{k}\cdot\boldsymbol{E}(\boldsymbol{k},\omega) = 0$ in the approximation linear in the field strength and that out of the infinite system of equations it is sufficient to isolate only those reciprocal lattice vectors that approximately satisfy the Bragg law (1). The transverse nature of the electric field makes it possible to introduce two scalar amplitudes,

$$\boldsymbol{E}(\boldsymbol{k}_m,\omega) = \gamma_m^{-1/2}\sum_{s=\pi,\sigma} B_{ms}\boldsymbol{e}_{ms} \tag{4}$$

where $\boldsymbol{e}_{m\pi}$ and $\boldsymbol{e}_{m\sigma}$ are mutually perpendicular unit polarization vectors normal to $\boldsymbol{k}_m$, and $\gamma_m = (\boldsymbol{k}_m\cdot\boldsymbol{n}_0)/K$.

Thus, the problem consists in finding the complex-valued refraction correction ε and the scalar amplitudes B_{ms}, which are, respectively, the eigenvalues and eigenvectors of a matrix,

$$\varepsilon B_{ms} = \sum_{m's'} K\gamma_m^{-1/2}\gamma_{m'}^{-1/2}(-\alpha_m\delta_{mm'}^{ss'} + \chi_{mm'}^{ss'})B_{m's'}. \tag{5}$$

Here $\delta_{mm'}^{ss'}$ is the Kronecker symbol, α_m the parameter that is the measure of the extent to which the reciprocal lattice vector with number m does not obey the Bragg law,

$$\alpha_m = \frac{1}{K^2}\left[(\boldsymbol{K}_e + \boldsymbol{h}_m)^2 - K^2\right] \tag{6}$$

and

$$\chi_{mm'}^{ss'} = \sum_{ij} e_{ms}^i\chi_\omega^{ij}(\boldsymbol{k}_m,\boldsymbol{k}_{m'})e_{m's'}^j \tag{7}$$

is the kinematic scattering matrix.

Obviously, condition (1) can be met with any two reciprocal lattice vectors for any given wavelength of the radiation. Actually, because of the symmetry of the crystal lattice, equation (1) is often satisfied by a combination of three, four, or a larger number of reciprocal lattice vectors. If it is satisfied with $N-1$ vectors, we speak of the N-wave case. Since there are two polarizations, the rank of matrix (7) is $2N$, and system (5) has $2N$ solutions. The total amplitude of the electric field in the crystal is

$$\boldsymbol{E}(\boldsymbol{r},\omega) = \sum_{msj}\lambda_j\exp(\mathrm{i}\boldsymbol{k}_{mj}\cdot\boldsymbol{r})\gamma_m^{-1/2}B_{msj}\boldsymbol{e}_{ms} \tag{8}$$

where $\boldsymbol{k}_{mj} = \boldsymbol{K}_e + \boldsymbol{h}_m + \varepsilon_j\boldsymbol{n}_0/2$, the quantity λ_j is the degree of excitation of the jth solution of system (5), and the eigenvectors B_{ms} are normalized to unity.

Parameters λ_j are determined by the boundary conditions. In the geometry of reflections a fraction of the scattered waves exit from the crystal through the upper (entrance) surface ($\gamma_m < 0$). We call these waves Bragg-scattered and denote them by the letter B. The waves exiting from the crystal through the lower surface of the plate are Laue-scattered and we denote them by the letter L. Formula (8) remains valid as well when any wave of the L-type with a wavevector $\boldsymbol{K}_e + \boldsymbol{h}_m$ is incident on the upper surface of the crystal and any wave of the B-type with an appropriate wavevector is incident on the lower surface.

Generally, the amplitudes of L-waves at the upper surface of the plate and those of B-waves at the lower are known. Out of these we build a vector with components $D_{ms}\gamma_m^{-1/2}$. To this end we must determine the missing amplitudes, which comprise a vector $R_{ms}\gamma_m^{-1/2}$. Averaging the amplitude (8) of the electric field over the unit cell at both surfaces of the crystal, we obtain the boundary conditions for each ray independently in the following form:

$$\sum_j B_{msj} \exp(i\varepsilon_j t_m/2)\lambda_j = D_{ms} \tag{9}$$

where $t_m = 0$ if γ_m is positive and $t_m = t$ if γ_m is negative. The required amplitudes of the scattered rays are

$$R_{ms} = \sum_j B_{msj} \exp(i\varepsilon_j z_m/2)\lambda_j \tag{10}$$

where $z_m = t$ if γ_m is positive and $z_m = 0$ if γ_m is negative. The experimentally measured reflection coefficient of wave (ms) transformed into wave $(m's')$ is

$$P(m's', ms) = \left|\frac{R_{m's'}}{D_{ms}}\right|^2 . \tag{11}$$

Formulae (5)–(11) determine the approach to solving the problem in one crystal plate implicitly. However, for further progress, we need an explicit form of solution. This requires using matrix algebra. To simplify the notation we will discard the polarization index s by incorporating it into m. Matrix B_{mj} is a square matrix of rank $2N$, and the parameters λ_j, D_m, and R_m are vectors in the same space. When the subscripts and/or superscripts are dropped from these quantities, the notation is understood to be in matrix form. The systems of equations (5), (9), and (10) in matrix form are

$$B \cdot \varepsilon = G \cdot B \qquad X \cdot \lambda = D \qquad R = Y \cdot \lambda \tag{12}$$

where $\varepsilon_{jj'} = \varepsilon_j \delta_{jj'}$ is a diagonal matrix. In explicit form the solution can easily be expressed in terms of a multiplicative inverse matrix as follows:

$$R = Y \cdot X^{-1} \cdot D = M \cdot D \qquad M = Y \cdot X^{-1}. \tag{13}$$

We call $M_{mm'}$ the dynamical scattering matrix. This matrix depends on the thickness t of the crystal. Obviously, in the limit of $t = 0$ we have $X = Y$ and $M = I$, where I is the identity matrix. We can show that

$$M_{mm'} \approx \delta_{mm'} + \frac{it}{2}\,\mathrm{sgn}(\gamma_m)\, G_{mm'} \tag{14}$$

for small values of t, with G the matrix on the right-hand side of (5). Thus, for small values of t the dynamical matrix is simply proportional to the kinematic, but already for $t > \chi_h^{-1}$ the relationship becomes nonlinear and fairly complex.

The solution in the form (13) is also unsatisfactory for thick crystals and in the limit as $t \to \infty$. The reason is that when diffraction is due to reflection in an absorbing crystal, the imaginary parts of the eigenvalues, ε_j'', may be either positive or negative. More than that, if in the N-wave case n rays undergo Bragg reflection, the number of eigenvalues for which $\varepsilon_j'' < 0$ is exactly $2n$, while in the other eigenvalues $\varepsilon_j'' > 0$.

The physics of this property of eigenvalues can easily be explained. To each value of j there corresponds a Bloch wave. The energy flux in $2n$ Bloch waves is directed from the lower surface of the crystal to the upper, and the waves decay when moving in the opposite direction. It is natural to call this group of Bloch waves Bragg propagating, similar to the scattered rays. In what follows the respective set of values of j is also denoted by a single letter B. The other Bloch waves are of the Laue type and are denoted collectively by the letter L.

All this suggests that as t grows, a fraction of the elements of the X and Y matrices decrease infinitely and the remainder become infinitely large. No numerical calculation is possible in this case. To obtain a solution suitable for computer processing we order the values of m according to decreasing values of γ_m and the values of j according to decreasing values of ε_j'' and then go over to the new notation L and B as described above. We also introduce the diagonal matrices

$$\begin{aligned} E_{\mathrm{LL}} &= \exp\left(\frac{\mathrm{i}\varepsilon_j t}{2}\right)\delta_{jj'} \qquad j \in \mathrm{L} \\ E_{\mathrm{BB}} &= \exp\left(\frac{\mathrm{i}\varepsilon_j t}{2}\right)\delta_{jj'} \qquad j \in \mathrm{B}. \end{aligned} \tag{15}$$

The system of equations (9) in the new notation splits into two subsystems and can be written in the following form:

$$\begin{aligned} &B_{\mathrm{LL}} \cdot \lambda_{\mathrm{L}} + B_{\mathrm{LB}} \cdot \lambda_{\mathrm{B}} = D_{\mathrm{L}} \\ &B_{\mathrm{BL}} \cdot E_{\mathrm{LL}} \cdot \lambda_{\mathrm{L}} + B_{\mathrm{BB}} \cdot E_{\mathrm{BB}} \cdot \lambda_{\mathrm{B}} = D_{\mathrm{B}} \end{aligned} \tag{16}$$

where B_{LL} and B_{BB} are, respectively, $2(N-n)\times 2(N-n)$ and $2n \times 2n$ square matrices, and B_{LB} and B_{BL} are generally rectangular matrices. This system is used to find λ_{L} and λ_{B}, bearing in mind that only square matrices can have multiplicative inverses. The values are then substituted into equation (10), which in the new notation has the form

$$\begin{aligned} R_{\mathrm{L}} &= B_{\mathrm{LL}} \cdot E_{\mathrm{LL}} \cdot \lambda_{\mathrm{L}} + B_{\mathrm{LB}} \cdot E_{\mathrm{BB}} \cdot \lambda_{\mathrm{B}} \\ R_{\mathrm{B}} &= B_{\mathrm{BL}} \cdot \lambda_{\mathrm{L}} + B_{\mathrm{BB}} \cdot \lambda_{\mathrm{B}}. \end{aligned} \tag{17}$$

The solution of system (16) must be transformed into a form that contains no increasing exponentials, that is, instead of matrix E_{BB} we must deal with the multiplicative inverse E_{BB}^{-1}. A simple calculation leads to the following result:

$$\begin{aligned} \lambda_{\mathrm{L}} &= Z_{\mathrm{LL}}^{-1} \cdot \left(D_{\mathrm{L}} - B_{\mathrm{LB}} \cdot E_{\mathrm{BB}}^{-1} \cdot B_{\mathrm{BB}}^{-1} \cdot D_{\mathrm{B}}\right) \\ \lambda_{\mathrm{B}} &= E_{\mathrm{BB}}^{-1} Z_{\mathrm{BB}}^{-1} \cdot \left(D_{\mathrm{B}} - B_{\mathrm{BL}} \cdot E_{\mathrm{LL}} \cdot B_{\mathrm{LL}}^{-1} \cdot D_{\mathrm{L}}\right) \end{aligned} \tag{18}$$

where

$$Z_{LL} = B_{LL} - B_{LB} \cdot E_{BB}^{-1} \cdot B_{BB}^{-1} \cdot B_{BL} \cdot E_{LL}$$
$$Z_{BB} = B_{BB} - B_{BL} \cdot E_{LL} \cdot B_{LL}^{-1} \cdot B_{LB} \cdot E_{BB}^{-1}. \tag{19}$$

Substituting (18) into (17), we obtain the solution in a new form:

$$R_L = M_{LL} \cdot D_L + M_{LB} \cdot D_B \qquad R_B = M_{BL} \cdot D_L + M_{BB} \cdot D_B \tag{20}$$

where the blocks of the dynamical scattering matrix have the following form:

$$M_{LL} = B_{LL} \cdot E_{LL} \cdot Z_{LL}^{-1} - B_{LB} \cdot Z_{BB}^{-1} \cdot B_{BL} \cdot E_{LL} \cdot B_{LL}^{-1}$$
$$M_{LB} = B_{LB} \cdot Z_{BB}^{-1} - B_{LL} \cdot E_{LL} \cdot Z_{LL}^{-1} \cdot B_{LB} \cdot E_{BB}^{-1} \cdot B_{BB}^{-1}$$
$$M_{BL} = B_{BL} \cdot Z_{LL}^{-1} - B_{BB} \cdot E_{BB}^{-1} \cdot Z_{BB}^{-1} \cdot B_{BL} \cdot E_{LL} \cdot B_{LL}^{-1} \tag{21}$$
$$M_{BB} = B_{BB} \cdot E_{BB}^{-1} \cdot Z_{BB}^{-1} - B_{BL} \cdot Z_{LL}^{-1} \cdot B_{LB} \cdot E_{BB}^{-1} \cdot B_{BB}^{-1}.$$

It is easy to verify directly that notwithstanding their unwieldy form these equations are fairly convenient for calculations with any crystal thickness including the limit of $t \to \infty$.

For thick crystals the block M_{BL} is the most interesting. In the zeroth order of the perturbation expansions in small exponentials we get

$$(M_{BL})_{t\to\infty} \approx B_{BL} \cdot B_{LL}^{-1}. \tag{22}$$

An approximation equivalent to (22) has been suggested in [6, 7]. The effect of the anomalous transmission of Laue rays in the reflection geometry is described by the matrix M_{LL}. In first-order perturbation theory directly from (21) we obtain

$$(M_{LL})_{t\to\infty} \approx (B_{LL} - B_{LB} \cdot B_{BB}^{-1} \cdot B_{BL}) \cdot E_{LL} \cdot B_{LL}^{-1}. \tag{23}$$

This result was obtained in [8]. In contrast to the approximate formulae (22) and (23), formula (21) gives an exact solution of the problem in the most general case and for all values of t.

3. Recurrence formulae for a multilayer crystal system

Let us consider a crystal on whose surface there has been grown a complicated system of heteroepitaxial layers of a different composition but with almost the same parameters of the crystal lattice, so that multiple diffraction is realized in all layers on the same reciprocal lattice vectors. We isolate the subsystem consisting of the n upper layers (figure 1). The amplitudes of the waves incident on this subsystem (from above and below) are denoted by D_L and D'_B, the amplitudes of the scattered waves by R'_L and R_B, and the blocks of the scattering matrix by $W_{kl}^{(n)}$ $(k, l = L, B)$. To this subsystem we add one more layer, to which the blocks $M_{kl}^{(n+1)}$ of the scattering

matrix correspond. We denote the amplitudes of the waves incident on this layer by D'_{L} and D_{B} and those scattered by the layer by R_{L} and R'_{B}. By definition,

$$\begin{aligned} R'_{\mathrm{L}} &= W^{(n)}_{\mathrm{LL}} \cdot D_{\mathrm{L}} + W^{(n)}_{\mathrm{LB}} \cdot D'_{\mathrm{B}} \\ R_{\mathrm{B}} &= W^{(n)}_{\mathrm{BL}} \cdot D_{\mathrm{L}} + W^{(n)}_{\mathrm{BB}} \cdot D'_{\mathrm{B}} \\ R_{\mathrm{L}} &= M^{(n+1)}_{\mathrm{LL}} \cdot D'_{\mathrm{L}} + M^{(n+1)}_{\mathrm{LB}} \cdot D_{\mathrm{B}} \\ R'_{\mathrm{B}} &= M^{(n+1)}_{\mathrm{BL}} \cdot D'_{\mathrm{L}} + M^{(n+1)}_{\mathrm{BB}} \cdot D_{\mathrm{B}}. \end{aligned} \tag{24}$$

For a system consisting of $n+1$ layers, as is evident from figure 1, not all the amplitudes on the right-hand sides of the formulae in (24) are known. Instead we have the obvious relations

$$D'_{\mathrm{L}} = R'_{\mathrm{L}} \qquad D'_{\mathrm{B}} = R'_{\mathrm{B}}. \tag{25}$$

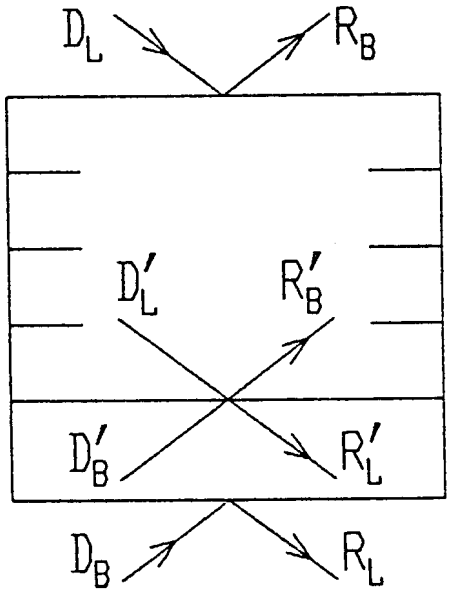

Figure 1. The ray pattern in a multilayer system.

The problem is to find the relationship between the amplitudes R_{L} and R_{B} of the waves scattered by the system of $n+1$ layers and the known amplitudes D_{L} and D_{B} of the waves incident on this system, that is, determine the blocks of the complete matrix that represents the scattering by the $(n+1)$th layer:

$$R_{\mathrm{L}} = W^{(n+1)}_{\mathrm{LL}} \cdot D_{\mathrm{L}} + W^{(n+1)}_{\mathrm{LB}} \cdot D_{\mathrm{B}} \qquad R_{\mathrm{B}} = W^{(n+1)}_{\mathrm{BL}} \cdot D_{\mathrm{L}} + W^{(n+1)}_{\mathrm{BB}} \cdot D_{\mathrm{B}}. \tag{26}$$

The solution method is the same as in section 2. Bearing in mind that the product of matrices is noncommutative and that only square matrices have multiplicative inverses, we must be careful in the calculations. Dropping the details, we can write

the answer as

$$
\begin{aligned}
W_{\mathrm{LL}}^{(n+1)} &= M_{\mathrm{LL}}^{(n+1)} \cdot \left(I_{\mathrm{LL}} - W_{\mathrm{LB}}^{(n)} \cdot M_{\mathrm{BL}}^{(n+1)}\right)^{-1} \cdot W_{\mathrm{LL}}^{(n)} \\
W_{\mathrm{LB}}^{(n+1)} &= M_{\mathrm{LB}}^{(n+1)} \\
&\quad + M_{\mathrm{LL}}^{(n+1)} \cdot \left(I_{\mathrm{LL}} - W_{\mathrm{LB}}^{(n)} \cdot M_{\mathrm{BL}}^{(n+1)}\right)^{-1} \cdot W_{\mathrm{LB}}^{(n)} \cdot M_{\mathrm{BB}}^{(n+1)} \\
W_{\mathrm{BL}}^{(n+1)} &= W_{\mathrm{BL}}^{(n)} \\
&\quad + W_{\mathrm{BB}}^{(n)} \cdot \left(I_{\mathrm{BB}} - M_{\mathrm{BL}}^{(n+1)} \cdot W_{\mathrm{LB}}^{(n)}\right)^{-1} \cdot M_{\mathrm{BL}}^{(n+1)} \cdot W_{\mathrm{LL}}^{(n)} \\
W_{\mathrm{BB}}^{(n+1)} &= W_{\mathrm{BB}}^{(n)} \cdot \left(I_{\mathrm{BB}} - M_{\mathrm{BL}}^{(n+1)} \cdot W_{\mathrm{LB}}^{(n)}\right)^{-1} \cdot M_{\mathrm{BB}}^{(n+1)}
\end{aligned}
\tag{27}
$$

where I_{LL} and I_{BB} are identity matrices of appropriate dimensions. Formulae (27) are the required recurrence formulae. If the crystal has N layers, after finding the scattering matrix for each layer by the procedure developed in section 2 we can easily find the scattering matrix for the entire multilayer system by successively applying (27).

The physical meaning of (27) is understood if we expand the matrices in power series, after which the result can be represented as the sum of processes of multiple reflection and transmission of the rays in the layers, which is schematically shown in figure 2 for the (a) LL, (b) BL, (c) LB, and (d) BB scattering processes. Summing the series by ordinary methods leads to equivalent results in a somewhat different form. For instance,

$$
W_{\mathrm{BL}}^{(n+1)} = W_{\mathrm{BL}}^{(n)} + W_{\mathrm{BB}}^{(n)} \cdot M_{\mathrm{BL}}^{(n+1)} \cdot \left(I_{\mathrm{BB}} - W_{\mathrm{LB}}^{(n)} \cdot M_{\mathrm{BL}}^{(n+1)}\right)^{-1} \cdot W_{\mathrm{LL}}^{(n)}. \tag{28}
$$

If the $(n+1)$st layer is very thick (the substrate), only matrix (28) is of interest, and in it we can employ approximation (22) for M_{BL}.

4. An example

The above theory makes it possible to give a meaningful description of a large number of new effects of multiple scattering of x rays in multilayer systems. As an illustration we apply the method to a fairly simple system, a thick single crystal with a homogeneous epitaxial film. Suppose that the relative variation in the interplanar distance in the film is $\Delta d/d$. This variation leads to an increase in the phase of the complex-valued amplitude of the wave reflected in the substrate, $\Delta\Phi = 2\pi(\Delta d/d)t/d$. If the film thickness is much less than the extinction length L_{ex}, in the angular region of total reflection the reflection coefficient P_{R} of two-wave diffraction is practically insensitive to the film, and there is no way in which the phase shift $\Delta\Phi$ can be measured. The film manifests itself only in a small maximum at the tail of the reflection curve.

The situation is quite different in the method of x-ray standing waves [9] when secondary radiation with a small escape depth is registered (photoelectrons, for instance). The shape of the curves representing the angular dependence of the yield of secondary radiation depends explicitly on the phase of the complex-valued reflection amplitude. As first demonstrated in [10], a similar pattern emerges in the event of three-wave diffraction in the angular region with strong reflection of the first

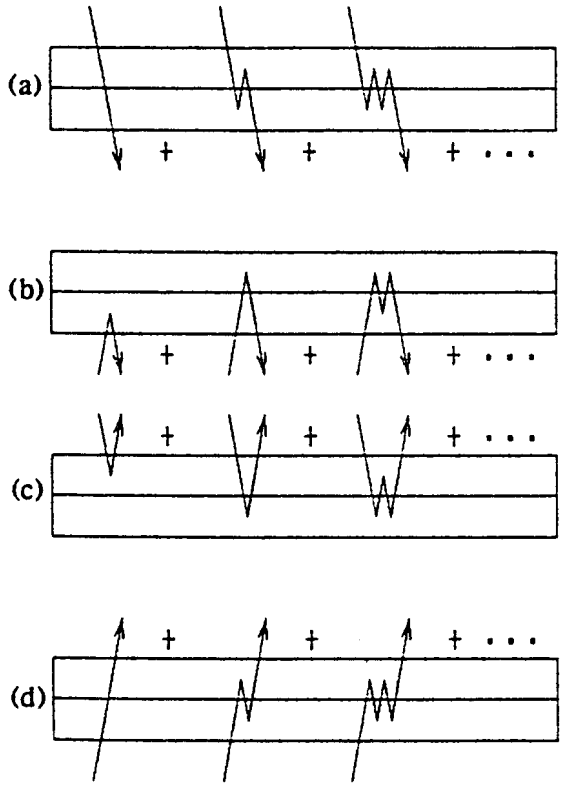

Figure 2. Summation of the processes of ray scattering in layers.

ray (h) and weak reflection of the second (g), where the intensity of the weak ray is approximately described by the following formula:

$$I_{gs} \approx I^0_{gs} \sum_{p=\pi,\sigma} \left| P^{ps}_{g0} + P^{ps}_{gh} \chi_{gh0} \exp(-i\Delta\Phi) \frac{E_{hs}}{E_{0s}} \right|^2 \tag{29}$$

with $I^0_{gs} = |\chi_{g0}/\alpha_g|^2 I_{0s}$ the intensity of the weak ray in the kinematic approximation in the absence of the first strong reflection, $P^{ps}_{mn} = e_{mp} \cdot e_{ns}$ the polarization factors, and $\chi_{gh0} = \chi_{gh}/\chi_{g0}$.

To demonstrate graphically this effect of the phase sensitivity of the curves, figure 3 shows the results of calculations of the coefficient of (113) reflection, P^{113}_R, for the case of (333, 113) diffraction of Cu Kα radiation in Si with an epitaxial film 1 μm thick. The entrance surface of the crystal plate is parallel to the (111) atomic planes. The calculation was done using formula (28), and the rotation axes of the crystal were chosen in such a way that parameter α_{333} was independent of $\Delta\varphi$.

For large values of $\Delta\varphi$, when $|\alpha_{113}| \gg |\chi_{g0}|$, the (113) beam has a low intensity and in the range of values of angle $\Delta\theta$ where $P^{333}_R \approx 1$, the (333) reflection curve is of the well-known 'Darwin table' type since $L^{333}_{ex} = 8.2\,\mu$m, which is considerably greater than the film's thickness. On the other hand, at $\Delta\Phi = 0$ the shape of the (113) reflection curves, as figure 3 clearly shows, has a distinct dispersive nature and strongly depends on the values of $\Delta\Phi$, changing in practically the same manner as photoemission curves [9].

Figure 3 shows 11 curves corresponding to different values of $\Delta\Phi$ from zero to 2π with an interval of $\pi/5$. To make the pattern more graphic, the curves have been shifted with respect to each other by one unit on the scale of the left vertical axis.

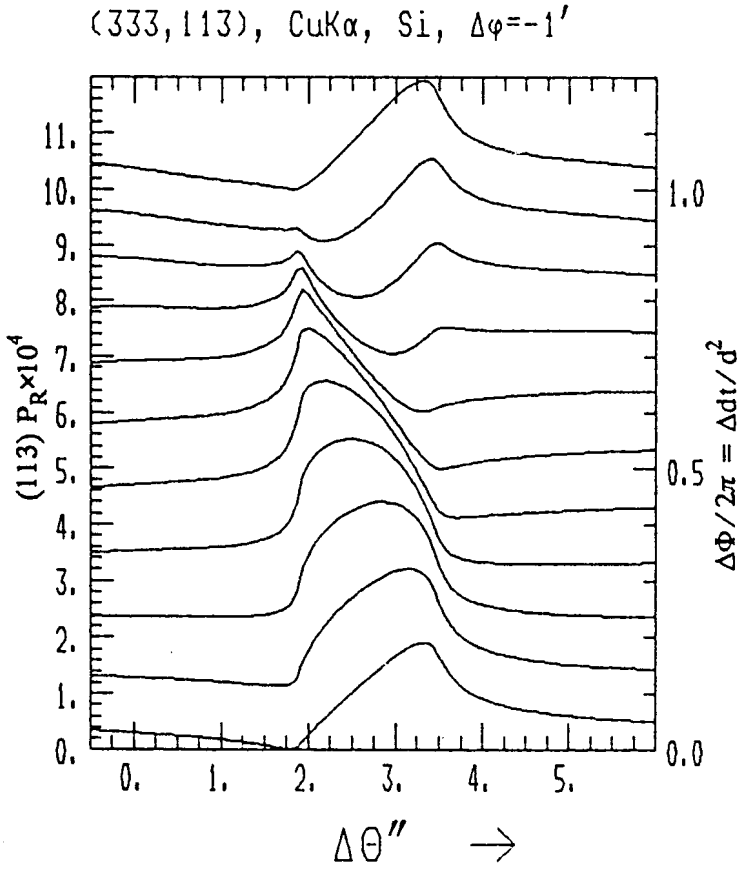

Figure 3. The curves representing the angular dependence of the coefficient of (113) reflection in the angular region of total reflection of the (333) ray but of weak reflection of the (113) ray for different values of the deformation of the crystal lattice in the epitaxial film, $\Delta d/d$. The respective variation of the phase of the (333) wave on the surface of the crystal is plotted along the right vertical axis.

The right vertical axis shows the values of $\Delta\Phi$ corresponding to different curves. Since the interplanar distance d is 1.05 Å for the (333) reflection, the specified interval of phase variation corresponds to $\Delta d/d$ varying from zero to 1.05×10^{-4}. The azimuthal angle of the crystal's rotation $\Delta\varphi$ is $-1'$. The effective escape depth of the (113) ray can be estimated by the formula $d_{\text{eff}} \approx \lambda \left|\gamma_g/\alpha_g\right| \approx C/\left|\Delta\varphi\right|$, with the coefficient $C = 0.11\,\mu$ms being very small in this case because $\left|\gamma_g\right|$ is small (a glancing beam). Thus, the demonstrated curve pattern will be observed for all values of $\Delta\varphi$ satisfying the condition $\left|\alpha_g\right| \gg \left|\chi_{g0}\right|$.

References

[1] Kov'ev É K and Simonov V I 1986 *Pis'ma Zh. Eksp. Teor. Fiz.* **43** 244
[2] Greiser N and Materlik G 1987 *Z. Phys.* B **66** 83
[3] Shin-Lin Chang 1984 *Multiple Diffraction of X-rays in Crystals* (Berlin: Springer)
[4] Afanasev A M and Kagan Yu 1968 *Acta Crystallogr.* A **24** 163
[5] Kolpakov A V, Bushuev V A and Kuz'min R N 1978 *Usp. Fiz. Nauk* **126** 479
[6] Kohn V G 1979 *Phys. Status Solidi* A **54** 375
[7] Shin-Lin Chang 1979 *Acta Crystallogr.* A **35** 543
[8] Toneyan A G, Kohn V G and Kuz'min R N 1984 *Kristallografiya* **29** 203
[9] Koval'chuk M V and Kohn V G 1986 *Usp. Fiz. Nauk* **149** 69
[10] Kohn V G 1988 *Phys. Status Solidi* A **106** 31