

Theory of X-Ray plane wave multiple diffraction

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1. INTRODUCTION

Despite the fact that a real coherent wave is a spherical monochromatic wave rather than a plane monochromatic wave, the theory of X-ray diffraction in single crystals as it has been developed by Ewald and Laue:

- P. P. Ewald, *Zur Begründung der Kristalloptik*.
 - Teil 1. - Ann. Phys., 1916, vol. 49, N. 1, p.1-38;
 - Teil 2. - Ann. Phys., 1916, vol. 49, N. 2, p. 117-143;
 - Teil 3. - Ann. Phys., 1917, vol. 54, N. 23, p. 519-556, N. 24, p. 557-597;
 - Teil 4. - Z. Kristall., 1937, vol. A97, N. 1/2, 1-27.
- M. Laue, *Ergebnisse der Exakten Naturwissenschaften*,
 - vol. 10, Berlin, Julius Springer, 1931, p. 133-158.

considers the plane monochromatic waves.

The reason is easy to understand taking into account that the single crystal is a macroscopically homogeneous object. Therefore the plane wave being scattered coherently conserves its form to a great extent. The other reason is a small value of interaction between X-rays and atoms of the crystal. So we need to take into account a large piece of crystal containing many atoms along the X-ray path for obtaining the significant effect.

Substituting Eq.(2) and Eq.(4) into Eq.(3) and taking into account Eq.(5) one obtains the infinite set of linear algebraic equations instead of differential equation

$$[k_h^2 - K^2] E_h^\alpha(\omega) = K^2 \sum_{\beta, g} X_{\omega}^{\alpha\beta}(\mathbf{k}_h, \mathbf{k}_g) E_g^\beta(\omega) \quad (6)$$

where $K = \omega/c$ as before. To obtain the understanding what kind of equation we have here we need to analyze first the coefficients which are, in general, Fourier components of the crystal susceptibility.

3. SUSCEPTIBILITY (KINEMATICAL SCATTERING AMPLITUDE)

3.1 Rayleigh scattering

Let us consider the structure of the susceptibility tensor $X_{\omega}^{\alpha\beta}(\mathbf{k}_h, \mathbf{k}_g)$. It allows us to understand better possible phenomena of X-ray diffraction. A modern science calculates the Fourier components of the susceptibility tensor considering the induced current density in a frame of quantum mechanics theory as a quantum mechanical average of the current density operator over the wave function of the crystal disturbed by electric field. Since the interaction between the X-rays and medium is small, the first order of the perturbation theory is enough (linear optics). We shall consider only elastic processes which don't change the frequency of the radiation. The inelastic processes cannot lead to coherent phenomena. In general case, the susceptibility turns out to be the complex value. The real part describes the *scattering processes* while the imaginary part describes the *absorption processes* which lead to decreasing the intensity of X-ray beam.

The main process which gives a large contribution is the *Rayleigh scattering* on the inhomogeneous electron density. This process is classic and it can be calculated to a great extent from the Newton mechanics. Let us consider first this way of derivation. One electron contributes the value ev to the induced current density, where e is the electron charge and v is the additional contribution to the electron velocity due to the electric field. The velocity has a direction along the direction of amplitude of electric field. We may obtain the value of velocity from the Newton equation. However, we shall keep the complex form of Fourier transformation to be consistent with Maxwell's equation where the same form of Fourier transformation is used. So we obtain

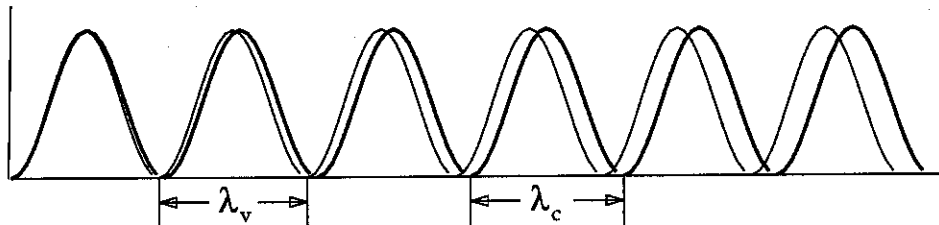


FIG. 1. Wavelength of X-rays in crystal is greater than in vacuum

is shown schematically in the Fig.2. In the case of face-centered cubic crystal lattice we have four atoms inside the unit cell. Such an approximation is rather good because all electrons of atom scatter the X-rays and the electrons of internal shells does not feel the influence of other atoms. As a result, the total expression of the Rayleigh contribution to the susceptibility can be written as

$$X_{\omega}^{\alpha\beta}(\mathbf{k}_h, \mathbf{k}_g)|_{\text{Rayleigh}} = \delta^{\alpha\beta} \chi_{h-g}, \quad (10)$$

where

$$\chi_h = -\frac{4\pi e^2}{m\omega^2 V_0} \sum_j f_j(\mathbf{h}) \exp(-i\mathbf{h}\mathbf{r}_j) \exp[-W_j(\mathbf{h})/2]. \quad (11)$$

Here the sum is carried out over the different atoms inside the unit cell of the crystal lattice (see Fig.2 for the face-centered cubic lattice), V_0 is the volume of the unit cell, therefore $1/V_0$ is a number of unit cells inside the unit volume, $f_j(\mathbf{h})$ is the space Fourier component of the electron density of atom having an index j on the reciprocal lattice vector \mathbf{h} (it is named usually atomic scattering factor or form-factor), \mathbf{r}_j is a coordinate of the j -th atom relative to an origin of the unit cell, $\exp[-W_j(\mathbf{h})]$ is the Debye-Waller factor which takes into account a random mean square displacement $\langle u_j^2 \rangle$ of the atom j from the equilibrium position inside the lattice $W_j(\mathbf{h}) = h^2 \langle u_j^2 \rangle$. The value $\langle u_j^2 \rangle$ depends on temperature.

3.2 Resonant scattering (photoelectron emission)

The expression of Eq.(11) is accurate in both classic and quantum mechanics. Let us introduce now the resonance (dispersion) contributions to the susceptibility, which is called, following to Laue, dispersion errors. The phenomenological theory of X-ray scattering considers the electron which moves over the circular orbit with a circular frequency ω_0 and is influenced by friction because it radiates. The corresponding Newton equation for one electron in this approach was taken in the form

$$m \left(\frac{d^2 x}{dt^2} + k \frac{dx}{dt} + \omega_0^2 x \right) = eE(\omega) \exp(-i\omega t), \quad (12)$$

where x is the electron coordinate, k is a friction coefficient. The velocity is $v = dx/dt$. The similar calculation as above leads to a following frequency dependence of the susceptibility

$$X(\omega) = -\frac{4\pi e^2}{m(\omega^2 - \omega_0^2 + ik\omega)} = X_0(\omega) [1 + A'(\omega) - iA''(\omega)] \quad (13)$$

where

$$X_0(\omega) = -\frac{4\pi e^2}{m\omega^2}, \quad A'(\omega) - iA''(\omega) = \frac{\omega_0^2(\omega^2 - \omega_0^2) - k^2\omega^2 - ik\omega^3}{(\omega^2 - \omega_0^2)^2 + k^2\omega^2}. \quad (14)$$

As it follows from Eq.(13) we obtain two additional terms in the expression of susceptibility compared to the pure Rayleigh scattering. One term add to a real part and it

The main reason of this fact is that the photoelectron emission is essentially quantum mechanical phenomenon. To described it in a frame of classic mechanics we need to make an additional assumption about the electron properties. So we need to suppose that the electron has no definite frequency of the resonance but it is in a state with the definite energy E_e . The frequency of the resonance corresponds to transition of electron from the state with one energy to the state with another energy. However, the photoelectron emission corresponds to transitions from the finite energy state to the free states of electron where the density of energy values is nearly constant and very high so the states form a continuum.

Therefore we need to consider a continuous distribution of virtual oscillators each having the frequency ω_j inside the interval from ω_0 up to infinity and introduce the new function $f_0(\omega_j)$ as the virtual oscillator strength normalized on the total number of electrons having the energy E_e

$$Z_0 = \int_{\omega_0}^{\infty} d\omega_j f_0(\omega_j) \quad (19)$$

after that the formula for the linear absorption coefficient becomes

$$\mu = \frac{6\pi c^2}{\omega^2} \int_{\omega_0}^{\infty} d\omega_j \frac{f_0(\omega_j)}{[1 + (\omega^2 - \omega_j^2)^2 \alpha^{-2} \omega^{-6}]} \quad (20)$$

As a result of approximate calculation of the integral (the denominator can be reduced to the Lorentz function and then replaced by delta-function) we obtain for ω far from ω_0 that

$$\mu(\omega) = \frac{2\pi^2 e^2}{mc} f_0(\omega) \quad (21)$$

Therefore the frequency dependence of the absorption coefficient now is determined completely by new quantity - strength of the virtual oscillator. However, in the frame of classic mechanics we have no the equations to calculate this. So we may use only the experimental results - the normal practice for the phenomenological theory. A more detailed topic on this subject may be found, for example, in the book

- B. K. Agarwal, *X-Ray Spectroscopy, An Introduction*,

Springer Series in Optical Science, vol.15, Springer-Verlag, Berlin, 1979.

where the following efforts to improve the classic approach are considered. Nevertheless the structure of the expression (13) for one electron is valid.

The summation over all electrons must be performed taking into account two assumptions. First, the atoms usually have many electron shells with different energies. Second, for high energy X-rays only the electrons which move over the internal circular orbits are essential and this electrons are very localized. Therefore, we may neglect their spacial distribution. As a result, the phenomenological theory deals with the Fourier coefficients of susceptibility as follows

$$\chi_{\mathbf{h}} = -\frac{4\pi e^2}{m\omega^2 V_0} \sum_j [f_j(\mathbf{h}) + \Delta f'_j(\omega) - i\Delta f''_j(\omega)] \exp(-i\mathbf{h}\mathbf{r}_j) \exp[-W_j(\mathbf{h})/2]. \quad (22)$$

• H. Sano, K. Ohtaka, Y.-H. Ohtsuki, J. Phys. Soc. Japan, 1969, vol. 27, p.1254.
This formula in general case of multiple diffraction may be written as

$$X_{\omega}^{\alpha\beta}(\mathbf{k}_h, \mathbf{k}_g)''|_{Compton} = \frac{8\pi r_0^2}{3V_0} \sum_j \exp(-i\mathbf{h}\mathbf{r}_j) \exp[-W_j(\mathbf{h})/2] G_j^{\alpha\beta}(\mathbf{k}_h, \mathbf{k}_g), \quad (25)$$

where

$$G_j^{\alpha\beta}(\mathbf{k}_h, \mathbf{k}_g) = \left\{ \delta^{\alpha\beta} f_j(\mathbf{h} - \mathbf{g}) - \frac{3}{8\pi} \int d\mathbf{s} [\delta^{\alpha\beta} - s^\alpha s^\beta] \sum_{m,n} F_j^{(mn)}(K\mathbf{s} - \mathbf{k}_h) F_j^{(nm)}(\mathbf{k}_g - K\mathbf{s}) \right\} \quad (26)$$

Here \mathbf{s} is an arbitrary unit vector which shows the direction of outgoing scattered wave and integration is performed over all directions of this vector, $F_j^{(mn)}(\mathbf{k})$ is the transition amplitude which is defined as

$$F_j^{(mn)}(\mathbf{k}) = \langle m | \sum_e \exp(i\mathbf{k}\mathbf{r}_e) | n \rangle \quad (27)$$

where $\langle m | \dots | n \rangle$ means the matrix element between the occupied electron states of the atom and a summation is carried out over the all electrons of j -th atom. Similar to photoelectric resonant scattering the Compton scattering is also a pure quantum mechanical process. Therefore the formula written above is obtained in a frame of quantum mechanics theory. The atomic scattering factor which appeared in the formulas for Rayleigh and resonance scattering, can be written as a sum of the transition amplitudes which are diagonal over electronic states

$$f_j(\mathbf{h}) = \sum_m F_j^{(mm)}(\mathbf{k}). \quad (28)$$

The formulas derived above allows us to estimate the value of interaction, i.e. the value of scattered wave as compared to the initial wave. For example, the amplitude of forward scattering in Silicon χ_0 for X-rays of energy $E = \hbar\omega = 14.41$ keV ($\lambda = 0.86$ Å) equals

- $\chi_0' |_{Rayl} = -4.62 \cdot 10^{-6}$, Rayleigh scattering,
- $\chi_0'' |_{photo} = 3.52 \cdot 10^{-8}$, the photoelectric contribution to the imaginary part,
- $\chi_0'' |_{Comp} = 3.86 \cdot 10^{-10}$, the Compton contribution to the imaginary part.

One can see that these values are very small therefore the X-ray beam must illuminate the large volume of the crystal to obtain a significant scattering wave.

4. BRAGG CONDITIONS AND GEOMETRY OF MULTIPLE DIFFRACTION

The electric field induction $\mathbf{D} = \varepsilon \mathbf{E}$, where ε is a dielectric function ($\varepsilon = 1 + \chi$) satisfies by definition the Maxwell's equation

$$\text{div } \mathbf{D} = \mathbf{E} \text{ grad } \varepsilon + \varepsilon \text{ div } \mathbf{E} = 0 \quad (29)$$

therefore

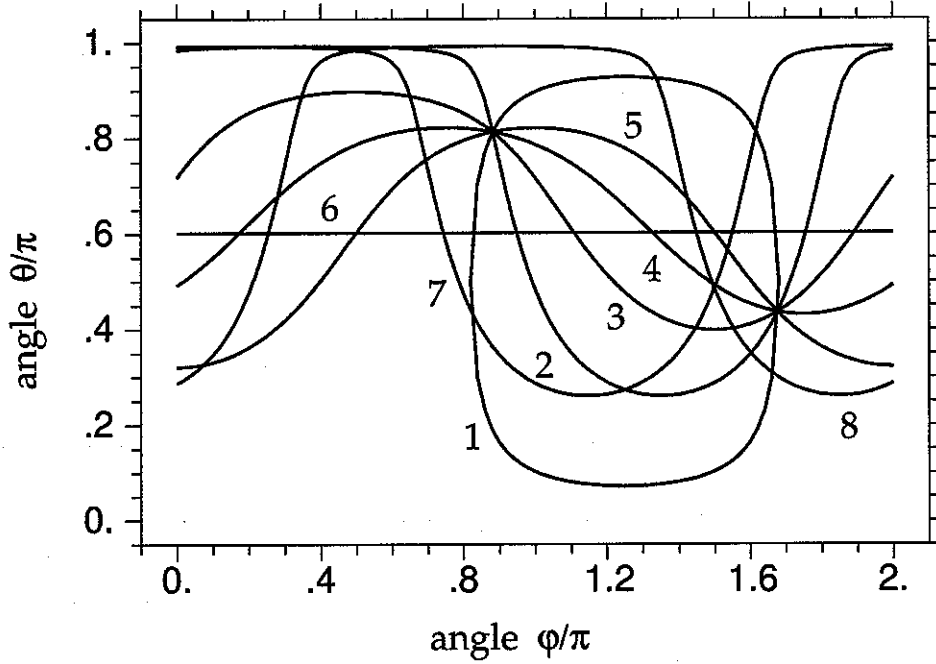


FIG. 3. The lines of Bragg conditions for different reciprocal lattice vectors of Si for $\lambda = 0.861 \text{ \AA}$, 1 = (220), 2 = (242), 3 = (044), 4 = ($\bar{2}$ 24), 5 = ($\bar{2}$ 02), 6 = (004), 7 = (422), 8 = ($\bar{4}$ 22)

a wavelength while d is an interatomic distance for the set of reflecting planes we may rewrite the Bragg condition in a scalar form

$$2d \sin \theta_B = \lambda \quad (35)$$

which allows to connect the direction of wave with the wavelength of the radiation.

The Eq.(35) defines only one angle of direction in the scattering plane while the values of second angle stay free. Using the spherical coordinates the vector \mathbf{k}_0 may be characterized as $k_{0x} = K \sin \theta \cos \varphi$, $k_{0y} = K \sin \theta \sin \varphi$, $k_{0z} = K \cos \theta$. For given frequency of the radiation the Bragg condition is fulfilled on the line inside the rectangle of angular coordinates. Taking into account that $K = 2\pi/\lambda$, $h = (2\pi/a)(h, k, l)$ in a cubic crystal lattice where a is a crystal lattice parameter, the Bragg condition can be written in the form

$$\frac{l \cos \theta + (\lambda/2a)(h^2 + k^2 + l^2)}{\sin \theta} = -(h \cos \varphi + k \sin \varphi). \quad (36)$$

Here the left-hand side depends only on θ , the right-hand side depends only on φ and the problem reduces to finding the possible θ values as a roots of equation for different φ values. There may be one or two roots. The functions $\theta_{hkl}(\varphi)$ are shown in Fig. 3 for several reciprocal lattice vectors of silicon and for ^{57}Fe nuclear resonant radiation with $\lambda = 0.861 \text{ \AA}$. One can see the line for (220) reciprocal lattice vector becomes closed. The

multiple diffraction. The systematic or frequency insensitive multiple diffraction with a number of waves larger than 3 may exist only due to symmetry of the crystal lattice when the additional reciprocal lattice vectors as a linear combination of \mathbf{h}_1 and \mathbf{h}_2 have their ends on the same circle. Such the cases are therefore *lattice-vector coplanar*. The frequency insensitive case is characterized by the property that a small difference in K may be compensated by a small difference of angles without destroying the Bragg conditions. Fig.5 shows an example of such a systematic six-beam case in Silicon where five reciprocal lattice vectors (220), (242), (044), ($\bar{2}$ 24) and ($\bar{2}$ 02) are circumscribed by the same circle as a section of the Ewald's sphere. In the Fig.3 and Fig.4 this situation reveals itself by intersection of five lines at the same point.

It is of interest to consider a simple algorithm which allows us to find all additional reciprocal lattice vectors of the systematic diffraction. This algorithm was elaborated and written by author in the book

- Z. G. Pinsker, *Dynamical scattering of X-Rays in Crystal*, Springer, Berlin, 1978. The two reciprocal lattice vectors \mathbf{h}_1 and \mathbf{h}_2 define the plane in the reciprocal lattice. Let \mathbf{b}_1 and \mathbf{b}_2 be two minimal reciprocal lattice vectors in this place which correspond to nonzero structure factors. Each reciprocal lattice vector can be represented as a linear combination of these vectors $\mathbf{h} = \alpha\mathbf{b}_1 + \beta\mathbf{b}_2$. The Bragg condition for the vector \mathbf{h} looks like $(\mathbf{K}_c + \mathbf{h})^2 = K_c^2$ where K_c is a critical wave vector which lies at the same plane as the reciprocal lattice vectors. In terms of parameters α and β the condition can be written as

$$\alpha^2 - 2\alpha(A - C\beta) - \beta(B - D\beta) = 0 \quad (37)$$

where

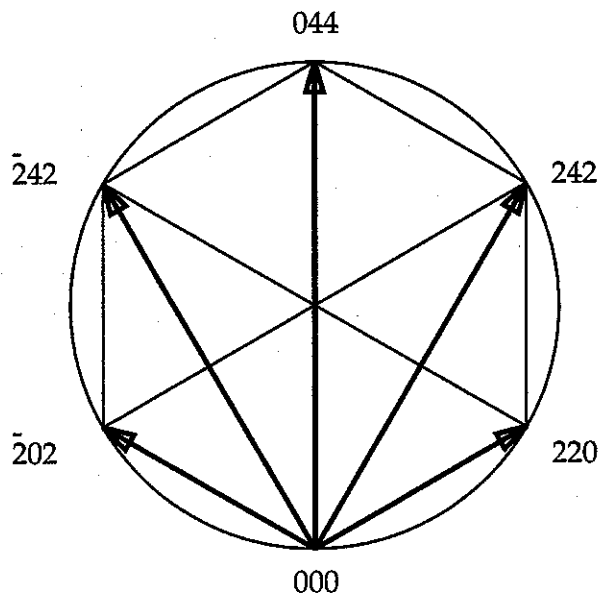


FIG. 5. The reciprocal lattice vectors of six-beam case in Silicon.

\mathbf{q}_m , namely, three mutually perpendicular unit vectors of polarizations \mathbf{e}_m^π , \mathbf{e}_m^σ and along the beam \mathbf{s}_m . The coordinates of the additional vector describe the angular deviation of the beam as well as the small relative change of the energy inside the bandwidth. In a linear approximation we obtain

$$\mathbf{q}_m/K_b = \Delta\theta_{1m}\mathbf{e}_m^\pi + \Delta\theta_{2m}\mathbf{e}_m^\sigma + \Delta\theta_\omega\mathbf{s}_m, \quad \Delta\theta_\omega = (\omega - \omega_b)/\omega_b, \quad (42)$$

We note that all the vectors \mathbf{K}_m have the same modulus $K_m^2 = K^2$ because the diffraction interaction is an elastic interaction which cannot change the energy of X-ray photon.

As it is shown in Fig.6, we may define the vectors \mathbf{e}_m^π and \mathbf{s}_m using the critical vector of coplanar diffraction \mathbf{K}_c and the unit vector of the normal to the reciprocal lattices plane \mathbf{n}_h as follows

$$\mathbf{s}_m = (\mathbf{K}_c + \mathbf{h}_m) \sin \theta_0 - \mathbf{n}_h \cos \theta_0, \quad \mathbf{e}_m^\pi = (\mathbf{K}_c + \mathbf{h}_m) \cos \theta_0 + \mathbf{n}_h \sin \theta_0 \quad (43)$$

while $\mathbf{e}_m^\sigma = [\mathbf{s}_m \times \mathbf{e}_m^\pi]$. Here θ_0 is an angle between the directions of the beams and $-\mathbf{n}_h$. The coplanar case corresponds to $\theta_0 = 90^\circ$.

In principle, each wave (for example, having the wave vector \mathbf{K}_i) may be treated as the incident wave. We define the geometrical parameters α_m describing the degree of deviation of m -th plane wave from the Bragg condition as

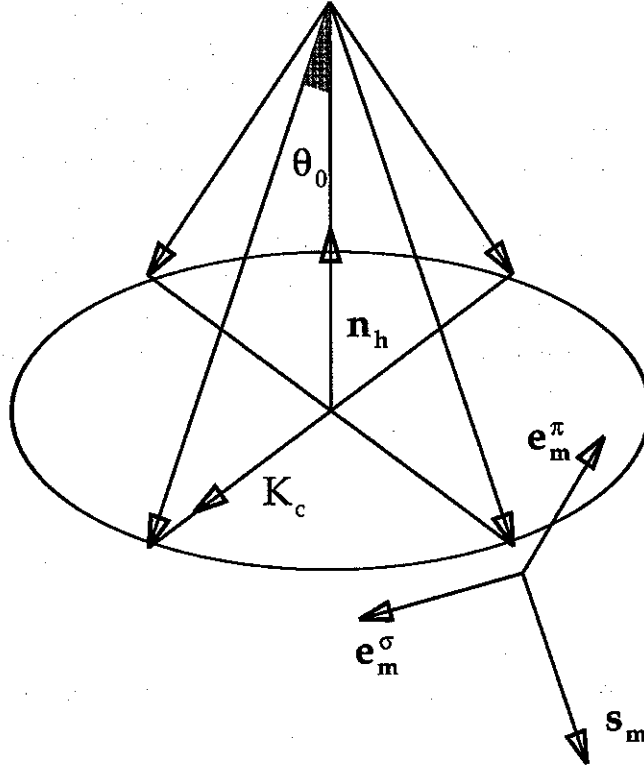


FIG. 6. The local coordinate system for a particular beam

$$G_{mm'}^{ss'}(\mathbf{q}_i) = \frac{K}{(\gamma_m \gamma_{m'})^{1/2}} (-\alpha_m(\mathbf{q}_i) \delta_{mm'}^{ss'} + g_{mm'}^{ss'}). \quad (48)$$

Here $\delta_{mm'}^{ss'}$ is the Kronecker symbol which equals unity if $m = m'$ and $s = s'$ and zero otherwise.

One can see that the equations (47) represent by themselves the eigen solution problem of the matrix $G_{mm'}^{ss'}$ having a rank $2n$. We shall call the matrix G the kinematical scattering matrix. Such a problem arises in any task investigated dynamical systems, for example, in the dynamical theory of the crystal lattice vibrations. Therefore we may use the standard mathematical methods to develop the dynamical theory of multiple diffraction. On the other hand, since even in three-beam case the rank of the matrix is 6 the analytical solution of the equation is not possible in general. Under these conditions the main purpose of the theory is to formulate the effective way of calculating all physical values measured in the experiment by means of computer, on one hand, and to formulate the general properties of the solutions, on the other hand. However, in some particular cases of high symmetry the analytical solutions are possible. These will be considered in another topic. Here we shall consider the general formulation of the problem which allows us to use a computer.

As well known, the eigen solution problem for the matrix of rank $2n$ has $2n$ solutions. We will distinguish them by the index j . On the other hand, to simplify the notation we will discard the polarization index s by incorporating it into m . So for each set of parameters value $\Delta\theta_{1i}$, $\Delta\theta_{2i}$ and $\Delta\theta_\omega = \Delta\omega/\omega$ we obtain $2n$ zones ε_j of refraction which may be used for drawing the dispersion surface and the matrix B_{mj} of the eigen vectors. We will suppose that the eigenvectors are normalized to unity. The total electric field inside the crystal is a linear combination of these eigen solutions

$$\mathbf{E}(\mathbf{r}, t) = \exp(i\mathbf{K}_0\mathbf{r} - i\omega t) \sum_m \exp(i\mathbf{h}_m\mathbf{r}) E_m(z) \mathbf{e}_m. \quad (49)$$

where

$$E_m(z) = \gamma_m^{-1/2} \sum_j \lambda_j B_{mj} \exp(i\varepsilon_j z/2).$$

Here $z = \mathbf{n}(\mathbf{r} - \mathbf{r}_s)$, where \mathbf{r}_s is the position of the point on the entrance surface.

The quantity λ_j defines the degree of excitation of the j -th eigen solution. The set of values λ_j is determined from the boundary conditions. The boundary conditions depends essentially on the geometry of the beams relative to boundary of the crystal. In general case, some of the values γ_m may be negative while others are positive. The fraction of the diffracted waves exit from the crystal through the upper (entrance) surface if $\gamma_m < 0$. We call these waves Bragg-reflected waves. We will denote them by one index B . The waves leave the crystal through the lower (exit) surface when $\gamma_m > 0$. Such waves are called Laue-reflected waves. We will denote them by one index L .

Up to now we assumed that only one plane wave $\exp(i\mathbf{K}_i\mathbf{r} - i\omega t)$ falls on the crystal plate. However, it is convenient to generalize the problem in a way which may be useful in considering the multilayer systems. Namely, we may suppose that all waves of L -type are known on the entrance surface of the plate ($z = 0$) and all waves of B -type are known on the exit surface ($z = t_p$). In this case the structure of the wave field inside the crystal,

i.e. the dynamical matrix is simply proportional to the kinematic matrix. However, already for $t_p > (K\chi_0)^{-1}$ the matrix elements become nonlinear on t_p and rather complex.

Let us consider the wave vectors of diffracted waves in air. Since the vector $\mathbf{K}_i = K_b \mathbf{s}_i + \mathbf{q}_i$ has an arbitrary value the other vectors cannot be arbitrary. These can differ from \mathbf{K}_i by the reciprocal lattice vector and the vector along the normal \mathbf{n} to the crystal surface. It is easy to verify that

$$\mathbf{q}_m = \mathbf{q}_i - \frac{K_b}{2\gamma_m} \alpha_m(\mathbf{q}_i) \mathbf{n}. \quad (56)$$

This equation allows us to obtain the correspondence between angular deviations of different beams from the reference values. For example,

$$\begin{aligned} \Delta\theta_{1m} = \Delta\theta_{1i} & \left((\mathbf{e}_m^\pi \mathbf{e}_i^\pi) - \frac{(\mathbf{e}_m^\pi \mathbf{n})(\mathbf{s}_m \mathbf{e}_i^\pi)}{(\mathbf{s}_m \mathbf{n})} \right) + \Delta\theta_{2i} \left((\mathbf{e}_m^\pi \mathbf{e}_i^\sigma) - \frac{(\mathbf{e}_m^\pi \mathbf{n})(\mathbf{s}_m \mathbf{e}_i^\sigma)}{(\mathbf{s}_m \mathbf{n})} \right) \\ & + \Delta\theta_\omega \left((\mathbf{e}_m^\pi \mathbf{s}_i) + \frac{(\mathbf{e}_m^\pi \mathbf{n})[1 - (\mathbf{s}_m \mathbf{s}_i)]}{(\mathbf{s}_m \mathbf{n})} \right) \end{aligned} \quad (57)$$

The formula is rather complicated. However, using the formulas of vector algebra

$$[(\mathbf{a} \times \mathbf{b})[\mathbf{c} \times \mathbf{d}]] = (\mathbf{ac})(\mathbf{bd}) - (\mathbf{ad})(\mathbf{bc}), \quad [\mathbf{s}_m \times \mathbf{e}_m^\pi] = \mathbf{e}_m^\sigma \quad (58)$$

we may transform this relation to a more simple form

$$\Delta\theta_{1m} \gamma_m = \Delta\theta_{1i} (\mathbf{n}[\mathbf{e}_i^\pi \times \mathbf{e}_m^\sigma]) + \Delta\theta_{2i} (\mathbf{n}[\mathbf{e}_i^\sigma \times \mathbf{e}_m^\sigma]) + \Delta\theta_\omega \{ (\mathbf{n} \mathbf{e}_m^\pi) + (\mathbf{n}[\mathbf{s}_i \times \mathbf{e}_m^\sigma]) \}. \quad (59)$$

In the important particular case of coplanar diffraction all the vectors \mathbf{e}_m^π equal to each other while all the vectors \mathbf{e}_m^σ and \mathbf{s}_m lie at the same plane. Let us assume that the vector \mathbf{n} lies also at the plane of \mathbf{s}_m (usually called the scattering plane). In this case the relation (58) shows that $\Delta\theta_{1m} = \Delta\theta_{1i}$. Therefore the angular property of the beam in the plane normal to the scattering plane becomes not changed.

The second angular parameter can be expressed in similar way

$$\Delta\theta_{2m} \gamma_m = \Delta\theta_{1i} (\mathbf{n}[\mathbf{e}_m^\pi \times \mathbf{e}_i^\pi]) + \Delta\theta_{2i} (\mathbf{n}[\mathbf{e}_m^\pi \times \mathbf{e}_i^\sigma]) + \Delta\theta_\omega \{ (\mathbf{n} \mathbf{e}_m^\sigma) + (\mathbf{n}[\mathbf{e}_m^\pi \times \mathbf{s}_i]) \}. \quad (60)$$

In the above mentioned coplanar case when \mathbf{n} lies in the scattering plane we obtain

$$\Delta\theta_{2m} = \Delta\theta_{2i} \frac{\gamma_i}{\gamma_m} + \Delta\theta_\omega \frac{(\mathbf{n}[\mathbf{e}_m^\sigma - \mathbf{e}_i^\sigma])}{\gamma_m} \quad (61)$$

We note that the last term can be transformed to another form taking into account that

$$(\mathbf{n}[\mathbf{e}_m^\sigma - \mathbf{e}_i^\sigma]) = (\mathbf{e}_m^\pi [\mathbf{n} \times \mathbf{h}_{mi}]) / K_b = 2 \sin \psi_{mi} \sin \theta_{Bmi} = (\gamma_m + \gamma_i) \tan \theta_{Bmi}$$

where $\mathbf{h}_{mi} = \mathbf{h}_m - \mathbf{h}_i$, ψ_{mi} is an angle between \mathbf{n} and \mathbf{h}_{mi} , θ_{Bmi} is the Bragg angle corresponding to the reciprocal lattice vector \mathbf{h}_{mi} , i.e. $\mathbf{h}_{mi} = 2K_b \sin \theta_{Bmi}$. As a result the Eq.(61) can be written as follows

$$\Delta\theta_{2m} = \Delta\theta_{2i} \frac{\gamma_i}{\gamma_m} + \Delta\theta_\omega \left(1 + \frac{\gamma_i}{\gamma_m} \right) \tan \theta_{Bmi} \quad (62)$$

This formula shows that the last term corresponds to a change of the Bragg angle with a change of frequency. The formula is valid in case of two-beam diffraction as well.

$$\begin{aligned} R_L &= B_{LL} \cdot E_{LL} \cdot \lambda_L + B_{LB} \cdot E_{BB} \cdot \lambda_B \\ R_B &= B_{BL} \cdot \lambda_L + B_{BB} \cdot \lambda_B \end{aligned} \quad (65)$$

The solution of Eq.(64) must be find in the form that contains no increasing exponentials, namely, instead of matrix E_{BB} we must deal with the multiplicative inverse E_{BB}^{-1} . A simple calculation leads to the following result

$$\begin{aligned} \lambda_L &= Z_{LL}^{-1} \cdot (D_L - B_{LB} \cdot E_{BB}^{-1} \cdot B_{BB}^{-1} \cdot D_B) \\ \lambda_B &= E_{BB}^{-1} \cdot Z_{BB}^{-1} \cdot (D_B - B_{BL} \cdot E_{LL} \cdot B_{LL}^{-1} \cdot D_L) \end{aligned} \quad (66)$$

where

$$\begin{aligned} Z_{LL} &= B_{LL} - B_{LB} \cdot E_{BB}^{-1} \cdot B_{BB}^{-1} \cdot B_{BL} \cdot E_{LL} \\ Z_{BB} &= B_{BB} - B_{BL} \cdot E_{LL} \cdot B_{LL}^{-1} \cdot B_{LB} \cdot E_{BB}^{-1} \end{aligned} \quad (67)$$

A substitution of Eq.(66) into Eq.(65) gives the solution in the new form

$$R_L = M_{LL} \cdot D_L + M_{LB} \cdot D_B, \quad R_B = M_{BL} \cdot D_L + M_{BB} \cdot D_B \quad (68)$$

with the blocks of the dynamical scattering matrix as follow

$$\begin{aligned} M_{LL} &= B_{LL} \cdot E_{LL} \cdot Z_{LL}^{-1} - B_{LB} \cdot Z_{BB}^{-1} \cdot B_{BL} \cdot E_{LL} \cdot B_{LL}^{-1} \\ M_{LB} &= B_{LB} \cdot Z_{BB}^{-1} - B_{LL} \cdot E_{LL} \cdot Z_{LL}^{-1} \cdot B_{LB} \cdot E_{BB}^{-1} \cdot B_{BB}^{-1} \\ M_{BL} &= B_{BL} \cdot Z_{LL}^{-1} - B_{BB} \cdot E_{BB}^{-1} \cdot Z_{BB}^{-1} \cdot B_{BL} \cdot E_{LL} \cdot B_{LL}^{-1} \\ M_{BB} &= B_{BB} \cdot E_{BB}^{-1} \cdot Z_{BB}^{-1} - B_{BL} \cdot Z_{LL}^{-1} \cdot B_{LB} \cdot E_{BB}^{-1} \cdot B_{BB}^{-1} \end{aligned} \quad (69)$$

It is easy to verify directly that notwithstanding their unwieldy form these equations are rather convenient for calculating the case of any crystal thickness including the limit $t_c \rightarrow \infty$. These formulas were derived for the first time in the papers

- V. G. Kohn, J. Moscow Phys. Soc., 1991, vol. 1, p. 425,
 - V. G. Kohn, Zh. Eksp. Teor. Fiz., 1994, vol. 105, p. 665
- (Sov. Phys. - JETP, 1994, vol. 78, p. 357)

For thick crystals the block M_{BL} is of most important which describes the reflected beams. A zero order of the perturbation expansion in small exponential leads to the approximate expression

$$\lim_{t_c \rightarrow \infty} M_{BL} = B_{BL} \cdot B_{LL}^{-1} \quad (70)$$

This is well known result which was obtained for the first time in the works

- V. G. Kohn, Phys. Stat. Sol. (a), 1979, vol. 54, p. 375,
- Shin-Lin Chang, Acta Crystall. A, 1979, vol. 35, p. 543.

On the other hand, the matrix M_{LL} describes the anomalous transmission effect of Laue beams under the existence of the Bragg beams. In the first order of the perturbation expansion we obtain from Eq.(69) the expression

$$\lim_{t_c \rightarrow \infty} M_{LL} = (B_{LL} - B_{LB} \cdot B_{BB}^{-1} \cdot B_{BL}) \cdot E_{LL} \cdot B_{LL}^{-1} \quad (71)$$

$$\begin{aligned}
Z_{LL} &= c_1 - c_2 e^{-i\varphi_2} \frac{1}{c_2 R_2} c_1 R_1 e^{i\varphi_1} = c_1 [1 - X e^{i\varphi_1} e^{-i\varphi_2}] \\
Z_{BB} &= c_2 R_2 - c_1 R_1 e^{i\varphi_1} \frac{1}{c_1} c_2 e^{-i\varphi_2} = c_2 R_2 [1 - X e^{i\varphi_1} e^{-i\varphi_2}]
\end{aligned} \tag{80}$$

where for a convenience the notation $\varphi_{1,2} = \varepsilon_{1,2} t_p / 2$, $X = R_1 / R_2$ is introduced. Finally we obtain (the notation t, \bar{t}, r, \bar{r} is used in the theory on crystalline multilayer)

$$\begin{aligned}
M_{LL} = t &= e^{i\varphi_1} \frac{(1 - X)}{(1 - X e^{i\varphi_1} e^{-i\varphi_2})}, & M_{BB} = \bar{t} &= e^{-i\varphi_2} \frac{(1 - X)}{(1 - X e^{i\varphi_1} e^{-i\varphi_2})}, \\
M_{BL} = r &= R_1 \frac{(1 - e^{i\varphi_1} e^{-i\varphi_2})}{(1 - X e^{i\varphi_1} e^{-i\varphi_2})}, & M_{LB} = \bar{r} &= \frac{1}{R_2} \frac{(1 - e^{i\varphi_1} e^{-i\varphi_2})}{(1 - X e^{i\varphi_1} e^{-i\varphi_2})}
\end{aligned} \tag{81}$$

Please note:

(1) the formulas is written in the form where $\exp(i\varphi_2)$ does not appear because it is impossible to calculate this number in a thick crystal by computer for the reason that it is very large,

(2) $t \neq \bar{t}$, $r \neq \bar{r}$ We only have

$$\frac{t}{\bar{t}} = e^{i\varphi_1} e^{i\varphi_2} = \exp \left(\frac{K t_p}{2\gamma_0} [\chi_0(1 + \beta) - \alpha\beta] \right), \quad \frac{r}{\bar{r}} = R_1 R_2 = -\frac{\chi_h}{\chi_{\bar{h}}} \tag{82}$$

Even in two beam case some authors failed to write the correct formulas. On the other hand, the approach based on the dynamical scattering matrix allows to solve more complicated tasks of calculating many crystal systems under the conditions of multiple diffraction. The examples of such systems are considered in a separate topic.