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On the Theory of Simultaneous X-ray Diffraction

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The dispersion equation for the four-wave case of X-ray diffraction is obtained in a compact form. The intermediate region between two-wave and arbitrary many-wave cases of diffraction is examined.

1. Introduction

In a recent paper by the present authors (Afanas'ev & Kohn, 1975) a simple method was suggested for the derivation of the dispersion equation in the three-wave case of X-ray diffraction. The method uses the obvious fact that the coefficients of the dispersion equation do not depend on the choice of the polarization vectors, but only on the angles between the wave vectors of incident and diffracting waves. In the present paper the dispersion equation for four-wave diffraction is derived in a compact form. Only a small modification of the method is necessary. The corresponding derivation is given in §2.

In §3 the intermediate region between two-wave and many-wave diffraction is examined, when all diffracting waves except one can be considered by perturbation theory. The dispersion equation is obtained in the form of a power series in the deviation from the Bragg conditions. The first terms of this series are determined by the dispersion determinants of the three-wave and four-wave cases. The possibility of the enhancement of the two-wave Borrmann effect in the simultaneous case is examined also.

2. Dispersion equation for four-wave diffraction

Let a monochromatic plane wave with the wave vector $\boldsymbol{\kappa}$ fall on a crystal having the form of a plate. The orientation of the crystal is such that three systems of planes (with vectors $\mathbf{K}_1$, $\mathbf{K}_2$ and $\mathbf{K}_3$, 2π times the reciprocal-lattice vectors) are near the Bragg position. Inside the crystal the space dependence of the electrical field vector $\mathbf{E}(\mathbf{r},\omega)$ is determined by the wave vector $\mathbf{k}_0=\boldsymbol{\kappa}+\kappa\varepsilon_0\mathbf{n}/\gamma_0$, where $\kappa=|\boldsymbol{\kappa}|$, $\mathbf{n}$ is the unit vector of the inner normal to the entrance surface, $\gamma_0=\mathbf{k}_0\mathbf{n}/|\mathbf{k}_0|$. The wave field is the superposition of the four plane waves with wave vectors $\mathbf{k}_0$ and $\mathbf{k}_h=\mathbf{k}_0+\mathbf{K}_h$, $h=1,2,3$.

The amplitudes of these waves $\mathbf{E}_h$ satisfy the transverse condition $\mathbf{k}_h\mathbf{E}_h=0$ and the next set of four vector equations

$$\begin{aligned}\tau_0\mathbf{E}_0+\chi_{01}\mathbf{E}_1+\chi_{02}\mathbf{E}_2+\chi_{03}\mathbf{E}_3&=0\\ \chi_{10}\mathbf{E}_0+\tau_1\mathbf{E}_1+\chi_{12}\mathbf{E}_2+\chi_{13}\mathbf{E}_3&=0\\ \chi_{20}\mathbf{E}_0+\chi_{21}\mathbf{E}_1+\tau_2\mathbf{E}_2+\chi_{23}\mathbf{E}_3&=0\\ \chi_{30}\mathbf{E}_0+\chi_{31}\mathbf{E}_1+\chi_{32}\mathbf{E}_2+\tau_3\mathbf{E}_3&=0\end{aligned}\qquad(2\cdot1)$$

where $\chi_{hh'}=\chi(\mathbf{k}_{h'}-\mathbf{k}_h)$ are the space Fourier components of 4π times the polarizability of a crystal,

$$\tau_h=\chi_{00}-2\varepsilon_h\,,\ \ \varepsilon_h=\tfrac{1}{2}\alpha_h+\frac{\varepsilon_0}{\beta_h}\,,\ \ \beta_h=\gamma_0/\gamma_h\,,\qquad(2\cdot2)$$

$$\alpha_h=\frac{(\boldsymbol{\kappa}+\mathbf{K}_h)^2-\boldsymbol{\kappa}^2}{\boldsymbol{\kappa}^2}\,,\ \ \gamma_h=\mathbf{s}_h\mathbf{n}\,,\ \ \mathbf{s}_h=\mathbf{k}_h/|\mathbf{k}_h|\,.$$

According to the transverse conditions every vector $\mathbf{E}_h$ has two independent components only. Let $\mathbf{e}_{h\pi}$, $\mathbf{e}_{h\sigma}$ and $\mathbf{s}_h$ be mutually perpendicular unit vectors. We write $\mathbf{E}_h=E_{h\pi}\mathbf{e}_{h\pi}+E_{h\sigma}\mathbf{e}_{h\sigma}$. Then (2·1) becomes a set of eight linear homogeneous equations for the scalar field amplitudes, which has the form

$$\begin{pmatrix} g_{hh'}\mathbf{e}_{h\pi}\mathbf{e}_{h'\pi} & g_{hh'}\mathbf{e}_{h\pi}\mathbf{e}_{h'\sigma}\\ g_{hh'}\mathbf{e}_{h\sigma}\mathbf{e}_{h'\pi} & g_{hh'}\mathbf{e}_{n\sigma}\mathbf{e}_{h'\sigma}\end{pmatrix}\begin{pmatrix}E_{h'\pi}\\ E_{h'\sigma}\end{pmatrix}=\dot{G}E=0\qquad(2\cdot3)$$

where $g_{hh'}=\tau_h\delta_{hh'}+\chi_{hh'}(1-\delta_{hh'})$, and $\delta_{hh'}$ is the Kronecker delta.

From the condition for the existence of a nontrivial solution of the set (2·3), we can deduce the eightfold equation for ε_0,

$$\Delta(\varepsilon_0)=\det(\dot{G})=0\,,\qquad(2\cdot4)$$

which is usually called the dispersion equation. To obtain the relevant form of equation (2·4), we must expand the determinant of the eighth-order matrix determined for some set of the polarization vectors $\mathbf{e}_{hs}$. However it is obvious that the determinant $\Delta(\varepsilon_0)$ does not depend on the specific choice of the polarization vectors.

First of all we note that the determinant $\Delta(\varepsilon_0)$, owing to the invariance, can contain only certain combinations of the quantities $s_{hh'}=\mathbf{s}_h\mathbf{s}_{h'}$. Moreover, it is easy to understand from the form of (2·3) that the every vector $\mathbf{s}_h$ must enter these combinations twice. The number of vectors can be no more than eight. To find all invariants satisfying these conditions, it is enough to expand the determinant of the fourth-power matrix $\{s_{hh'}\}$. Then we can determine the coefficients of the invariants from the consideration of a set of simple particular cases, where it is easy to calculate the determinant $\Delta(\varepsilon_0)$ directly.

The method described above was used by Afanas'ev & Kohn (1975) to obtain the dispersion equation in the three-wave case. In the four-wave case we have a large number of independent invariants and this method leads to a cumbersome expression. Nevertheless the compact form of the dispersion equation can be obtained in this case also.

Consider first the coplanar case in which all vectors $\mathbf{s}_h$ lie in the same plane. In this case, we choose all $\mathbf{e}_{h\pi}$ perpendicular to this plane and $\mathbf{e}_{h\sigma}$ in the plane. We can easily obtain from (2·3)

$$\Delta(\varepsilon_0)=\Delta_4\Delta_{4s} \quad (2\cdot5)$$

where

$$\Delta_4=\det(g_{hh'})\,, \quad \Delta_{4s}=\det(g_{hh'}s_{hh'})\,.$$

In the general case the determinant $\Delta(\varepsilon_0)$ contains some terms additional to that in (2·5). However, these terms must be proportional to the invariants, which are equal to zero in the coplanar case. There are seven invariants of this kind

$$\Omega^2_{012},\ \Omega^2_{013},\ \Omega^2_{023},\ \Omega^2_{123}, \quad \Omega_{hkl}=\mathbf{s}_h[\mathbf{s}_k\times\mathbf{s}_l]\,, \quad (2\cdot6)$$

$$V^2_{01},\ V^2_{02},\ V^2_{03}, \quad V_{hk}=\{[\mathbf{s}_h\times\mathbf{s}_k]\times[\mathbf{s}_l\times\mathbf{s}_m]\}\,. \quad (2\cdot7)$$

Hereafter $h,k,l,m=0,1,2,3,\ h\neq k\neq l\neq m$. Only three invariants in (2·6) are independent because the space is three-dimensional. It is convenient to choose the next six linear combinations (2·6) and (2·7) as linearly independent invariants

$$C_{01},\ C_{02},\ C_{03},\ C_{12},\ C_{13},\ C_{23},$$
$$C_{hk}=\tfrac{1}{2}[V^2_{hl}+V^2_{hm}-\Omega^2_{hkl}-\Omega^2_{hkm}]\,. \quad (2\cdot8)$$

In accordance with the above remarks we can at once write $\Delta(\varepsilon_0)$ in the form

$$\Delta(\varepsilon_0)=\Delta_4\Delta_{4s}+\sum_{h<k}C_{hk}B_{hk}(\tau,\chi)\,. \quad (2\cdot9)$$

To find the coefficients B_{hk} in (2·9) we consider six simple particular cases, where it is easy to calculate the determinant. Let, for example,

$$\mathbf{s}_0=\mathbf{s}_1\perp\mathbf{s}_2\perp\mathbf{s}_3\,. \quad (2\cdot10)$$

In this case, directing the polarization vectors so that

$$\mathbf{s}_0=\mathbf{s}_1=\mathbf{e}_{2\sigma}=\mathbf{e}_{3\pi},\ \mathbf{s}_2=\mathbf{e}_{0\pi}=\mathbf{e}_{1\pi}=\mathbf{e}_{3\sigma},$$
$$\mathbf{s}_3=\mathbf{e}_{0\sigma}=\mathbf{e}_{1\sigma}=\mathbf{e}_{2\pi}\,,$$

we obtain

$$\Delta(\varepsilon_0)=\Delta_{23}\Delta_{012}\Delta_{013} \quad (2\cdot11)$$

where

$$\Delta_{hkl}=\begin{bmatrix}\tau_h & \chi_{hk} & \chi_{hl}\\ \chi_{kh} & \tau_k & \chi_{kl}\\ \chi_{lh} & \chi_{lk} & \tau_l\end{bmatrix}, \quad \Delta_{hk}=\begin{bmatrix}\tau_h & \chi_{hk}\\ \chi_{kh} & \tau_k\end{bmatrix}. \quad (2\cdot12)$$

On the other hand in this case $C_{01}=1$ and the remaining $C_{hk}=0$. Taking into account that $\Delta_{4s}=\Delta_{01}\tau_2\tau_3$ we find finally

$$B_{01}=\Delta_{012}\Delta_{013}\Delta_{23}-\Delta_{01}\tau_2\tau_3\Delta_4\,. \quad (2\cdot13)$$

Another five coefficients can be determined similarly from consideration of particular cases of the kind (2·10) in which two vectors are equal and the rest are mutually perpendicular.

As a result, the dispersion equation for four-wave diffraction has the form

$$\Delta(\varepsilon_0)=\Delta_4\Delta_{4s}+\sum_{h<k}C_{hk}(\Delta_{hkl}\Delta_{hkm}\Delta_{lm}-\Delta_{hk}\tau_l\tau_m\Delta_4)=0\,. \quad (2\cdot14)$$

We note that the quantities C_{hk} can also be written in the form

$$C_{hk}=\tfrac{1}{4}[\Omega^2_{hlm}+\Omega^2_{klm}-\Omega^2_{hkl}-\Omega^2_{hkm}+2\Omega_{hkl}\Omega_{hkm}s_{lm}$$
$$+2\Omega_{hlm}\Omega_{klm}s_{hk}]=s_{hk}-s_{hk}s_{kl}s_{lh}-s_{hk}s_{km}s_{mh}$$
$$+s_{hk}s_{kl}s_{lm}s_{mh}+s_{hk}s_{km}s_{ml}s_{lh}-s_{hk}s^2_{lm}\,. \quad (2\cdot15)$$

The method outlined above allows the reduction in the general case of the determinant of order $2N$ to a sum of the products of determinants of $\leq N$. In the case $N=3$ we obtain the expression which was found firstly by Penning (1968). It can be obtained from (2·14) as a limit

$$\Delta^{(3)}(\varepsilon_0)=\lim\,[\tau_3^{-2}\Delta^{(4)}(\varepsilon_0)]$$
$$=\Delta_3\Delta_{3s}+\Omega^2_{012}(\Delta_{01}\Delta_{02}\Delta_{12}-\tau_0\tau_1\tau_2\Delta_3)\,. \quad (2\cdot16)$$

However, in the case $N=5$ the expression for the determinant of the form of (2·9) will contain already a large number of the terms in the sum over the invariants. We think that for $N\geq 5$ a direct numerical solution of the problem by means of a computer is more convenient.

3. Simultaneous diffraction in a nearly two-wave case

In Afanas'ev & Kohn (1975), the passage from two-wave to three-wave case was examined in detail when one parameter characterizing the deviation from the Bragg condition becomes large. In this section we consider the more general problem, namely, a perturbation of the two-wave solution by the nearby many-wave point. The number of diffracting waves can be arbitrary.

We note that this is the case of the most interest because parameters α_h (2·2) characterizing the deviation from the Bragg conditions are inevitably tied to one another owing to the three-dimensionality of the space. We restrict ourselves to the case where all reciprocal-lattice vectors lie in the same plane. Let $\boldsymbol{\kappa}_0$ be the vector exactly satisfying the simultaneous Bragg conditions, $\mathbf{a}_1,\mathbf{a}_2$ be the mutually perpendicular unit vectors in the plane which is perpendicular to $\boldsymbol{\kappa}_0$, and $\mathbf{a}_2\mathbf{K}_1=0$. Then

$$\boldsymbol{\kappa}=\boldsymbol{\kappa}_0+\kappa(\theta_1\mathbf{a}_1+\theta_2\mathbf{a}_2)\,,$$
$$\alpha_1=2\kappa^{-1}(\mathbf{a}_1\mathbf{K}_1)\theta_1\,,$$
$$\alpha_h=2\kappa^{-1}[(\mathbf{a}_1\mathbf{K}_h)\theta_1+(\mathbf{a}_2\mathbf{K}_h)\theta_2]\,,\quad h=2,\ldots,N-1\,. \quad (3\cdot1)$$

It is easy to see from (3·1) that if $|\theta_2|\to\infty$ then all α_h except α_1 become simultaneously large and we pass to the two-wave case with the reciprocal-lattice vector $\mathbf{K}_1$. On the whole, the many-wave region in the plane of parameters θ_1 and θ_2 is seen as a spot and the bunch of two-wave lines intersect it.

Let $|\theta_2| \gg |\chi_{00}|$. In this case the quantity $x=|\chi_{00}|/\theta_2$ is obviously a small parameter and we can express the whole many-wave determinant $\Delta^{(N)}(\varepsilon_0)$ as a power series in x. It is easy to understand that the first terms of this series have the form

$$\Delta^{(N)}(\varepsilon_0)=(\tau_2\ldots\tau_{N-1})^2\left[\Delta^{(2)}_{01}(\varepsilon_0)+\sum_{h=2}^{N-1}\left(\frac{\partial\Delta^{(3)}_{01h}}{\partial\tau_h}\right)_{\tau h=0}\frac{1}{\tau_h}+\tfrac{1}{2}\sum_{h,h'=2}^{N-1}\left(\frac{\partial^2\Delta^{(4)}_{01hh'}}{\partial\tau_h\partial\tau_{h'}}\right)_{\tau h=\tau h'=0}\frac{1}{\tau_h\tau_{h'}}+\ldots\right]. \quad (3\cdot2)$$

Here $\Delta^{(3)}_{01h}(\varepsilon_0)$ is the three-wave determinant of the vectors $\mathbf{k}_0$, $\mathbf{k}_1$, $\mathbf{k}_h$, and $\Delta^{(4)}_{01hh'}$ is the four-wave determinant of the vectors $\mathbf{k}_0,\mathbf{k}_1,\mathbf{k}_h,\mathbf{k}_{h'}$.

Using formulae (2·16), (2·14) and (3·2), one can examine the perturbation of the two-wave solutions up to the x^2 terms. As is known, the amplitudes of the diffraction waves decrease as $1/|\theta_2|$, *i.e.* only slightly. Therefore the formula (3·2) makes it possible to obtain the many-wave solutions (which are close to the two-wave solution) for a rather wide range of the angles and the contribution to the integral intensity from this region can just be dominant.

One of the principal problems in the theory of the simultaneous diffraction is the question of the enhancement of the two-wave anomalous-transmission effect. Qualitatively this question can be investigated easily from the formula (3·2). We assume that $|\theta_2|$ is large enough and restrict ourselves to the terms of the first power of x. Then the problem in fact reduces to the three-wave case which was examined by Afanas'ev & Kohn (1975).

Let us consider, as in Afanas'ev & Kohn (1975), the crystal with an inversion centre (when $\chi_{hh'}=\chi_{h'h}$). Assume that $|\chi_{01}|\geq|\chi_{hh'}|$, $h\neq h'$, and consider the point of the two-wave minimum of the imaginary part of ε_0, that is

$$\beta_1=1,\ \alpha_1=0,\ \tau_0=\tau_1=\chi_{01}+\varepsilon,\ \tau_h\approx-\alpha_h\,. \quad (3\cdot3)$$

Thei maginary part of the quantity ε introduced in (3·3) directly determines the difference between the many-wave minimum absorption coefficient and the two-wave one, namely,

$$\Delta\mu^{(N)}(\theta_2)=\mu^{(N)}(\theta_2)-\mu^{(2)}_{01\min}=-\kappa\varepsilon''\,. \quad (3\cdot4)$$

The analogous procedure, which has been used for the derivation of the formula (4·11) in Afanas'ev & Kohn (1975) gives the result:

$$\Delta\mu^{(N)}(\theta_2)=\frac{\kappa^2}{2\theta_2}\sum_{h=2}^{N-1}\frac{(\chi_{0h}-\chi_{1h})'(\chi_{0h}-\chi_{1h})''}{(\mathbf{a}_2\mathbf{K}_h)}\times\frac{(1-s^2_{01}-\Omega^2_{01h})}{(1-s^2_{01})}\,. \quad (3\cdot5)$$

For $N>3$ it follows from (3·5) that the simultaneous minimum of the absorption coefficient is smaller than the corresponding two-wave values, that is the enhancement of the effect always occurs. The symmetric three-wave case is the only exception. The formula (3·5) gives a convenient way for the qualitative estimation of the anomalous-transmission effect. Although the quantities $\mu^{(N)}_{\min}$ and $\Delta\mu^{(N)}(\theta_2)$ do not correlate directly with one another, we can nevertheless make a comparative estimate of the value $\mu^{(N)}_{\min}$ from knowledge of the speed of decrease of $\mu^{(N)}(\theta_2)$ with a large $|\theta_2|$. According to (3·5) $\Delta\mu^{(N)}$ is the larger, the stronger the asymmetry is of the particular three-wave combinations of the vectors $\mathbf{k}_0,\mathbf{k}_1,\mathbf{k}_h$. It is interesting that, in spite of this, the complete many-wave case can have a high symmetry.

The other consequence of the formula (3·5) is the asymmetry of the many-wave corrections. The absorption coefficient increases for one sign of θ_2 and decreases for the opposite sign. If the simultaneous case has the symmetry plane which is perpendicular to $\mathbf{a}_2$ then two roots with the same imaginary part $\sim\mu^{(2)}_{01}$ exist at large $|\theta_2|$. When $|\theta_2|$ decreases, the imaginary part of this root bifurcates and the two branches diverge symmetrically up and down according to (3·5) These are an example of configurations of the reciprocal-lattice vectors in the form of a symmetrical polygon.

References

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