

phys. stat. sol. (a) 28, 61 (1975)

Subject classification: 1.1; 22.1.1

I. V. Kurchatov Institute of Atomic Energy, Moscow

Borrmann Effect in the Three-Wave Case of X-Ray Diffraction

By

A. M. AFANASEV and V. G. KOHN

The general properties of the dispersion surface in the three-wave case are considered. A method of the derivation of the dispersion equation is suggested in an invariant form convenient for the concrete analysis. An analysis of the amplification of the anomalous transmission (Borrmann) effect in the three-wave case and a simple way for the estimation of this effect without large numerical calculations are given. The passage from two- to three-wave case is considered. This analysis allows to predict a general form of the dependence of the absorption coefficient on the orientation of the incident beam.

Рассмотрены общие свойства дисперсионной поверхности в трехволновом случае. Предложен метод вывода дисперсионного уравнения в инвариантной форме, удобной для анализа. Анализируется усиление эффекта аномального прохождения (эффекта Бормана) в трехволновом случае и предложен простой способ оценки этого эффекта, не требующий большого численного счета. Рассмотрен переход от трехволнового случая к двухволновому, что позволяет предсказать общий ход зависимости коэффициента поглощения от ориентации падающего пучка.

1. Introduction

In recent years, interest in three-wave diffraction of X-rays has continuously increased. Many papers have been published in which this phenomenon has been investigated both experimentally and theoretically [1 to 11]. In [1], it has first clearly been shown that the three-wave combination of the "bad" reflections in the lattice of a germanium crystal, which give a slight decrease of the absorption coefficient, provides an essential strengthening of the anomalous transmission effect when the Bragg conditions are exactly fulfilled. This case was examined in [2] theoretically. It turns out that in this case an analytical solution of the dispersion equation can be obtained for some values of the parameters characterizing the deviation from the Bragg conditions. In [3 to 5], other particular cases are investigated which allow for an analytical solution of the dispersion equation.

A general formulation of the problem has been given in papers [6, 7]. The physical principles of the theory of the three-wave case are the same as in the case of two strong waves. However, the wave field of X-rays, arising in a crystal in this case, has a much more complicated structure. The general analysis of the problem cannot be conducted completely in an analytical way, and numerical calculations are necessary to obtain concrete results.

One of the most interesting problems under consideration is the effect of anomalous transmission. Naturally, the question arises to what extent the presence of the third wave can amplify this effect. The particular case that the direct interaction between diffracted waves is absent, has been analysed in

papers [2, 6, 7]. However, in the general case this problem has not been examined. The present work is devoted, mainly, to the analysis of the Borrmann effect in the case of three-wave diffraction.

In Section 2, the derivation of the dispersion equation is given in an invariant form. The method of the derivation differs from that developed by Ewald and Heno [6], and it is convenient also for the multiple-wave problem for more than three waves.

The analysis of the Borrmann effects in an arbitrary case of three-wave diffraction requires the use of numerical methods for the solution of the dispersion equation. For finding the minimal absorption coefficient, it is necessary to consider, generally speaking, the whole region of values of the parameters characterizing the deviation from the Bragg conditions. A much more simple method for estimating the absorption minimum is offered in Section 3.

In Section 4, the passage from the two-wave case to the three-wave case is examined. In Section 5, the general properties of the dispersion surface and of a special point are considered.

2. Derivation of the Dispersion Equation

To describe the electromagnetic field of X-rays inside a crystal, we use Maxwell's equations for the electrical field vector. In terms of space and time Fourier components $\mathbf{E}(\mathbf{k}, \omega)$ we get

$$\frac{(k^2 - \kappa^2)}{\kappa^2} \mathbf{E}(\mathbf{k}, \omega) - i \frac{4\pi}{\omega} \mathbf{j}(\mathbf{k}, \omega) - \frac{1}{\kappa^2} \mathbf{k}(\mathbf{k} \cdot \mathbf{E}(\mathbf{k}, \omega)) = 0, \quad (2.1)$$

where $\kappa = \omega/c$, c is the light velocity, $\mathbf{j}(\mathbf{k}, \omega)$ the Fourier component of the current density. The expression for \mathbf{j} , which takes into account all possible interactions between the electromagnetic wave and the crystal, has been obtained by Afanasev and Kagan [12]. In the usual approximations, we have

$$\mathbf{j}(\mathbf{k}, \omega) = -\frac{i\omega}{4\pi} \sum_h \chi(\mathbf{k}, \mathbf{k}_h) \mathbf{E}(\mathbf{k}_h, \omega), \quad \mathbf{k}_h = \mathbf{k} + \mathbf{K}_h, \quad (2.2)$$

where χ is 4π times the polarizability of a crystal, \mathbf{K}_h is 2π times the reciprocal-lattice vector.

Let a plane wave with vector $\boldsymbol{\kappa}$ fall on a crystal in the form of a plate. Inside the crystal the space dependence of the field is determined by the vector $\mathbf{k}_0 = \boldsymbol{\kappa} + \kappa \varepsilon_0 \mathbf{n} / \gamma_0$, where \mathbf{n} is the inner normal to the entrance surface, $\gamma_0 = |\mathbf{k}_0 \mathbf{n}| / |\mathbf{k}_0|$. If the orientation of the crystal is such that two systems of planes (with the reciprocal-lattice-vectors \mathbf{K}_1 and \mathbf{K}_2) are near the Bragg position, then we can replace approximately the general set (2.1) by the set of three vector equations for the amplitudes $\mathbf{E}_h = \mathbf{E}(\mathbf{k}_h, \omega)$, $h = 0, 1, 2$:

$$\left. \begin{aligned} \tau_0 \mathbf{E}_0 + \chi_{01} \mathbf{E}_1 + \chi_{02} \mathbf{E}_2 + \mathbf{s}_0(\mathbf{s}_0 \mathbf{E}_0) &= 0, \\ \chi_{10} \mathbf{E}_0 + \tau_1 \mathbf{E}_1 + \chi_{12} \mathbf{E}_2 + \mathbf{s}_1(\mathbf{s}_1 \mathbf{E}_1) &= 0, \\ \chi_{20} \mathbf{E}_0 + \chi_{21} \mathbf{E}_1 + \tau_2 \mathbf{E}_2 + \mathbf{s}_2(\mathbf{s}_2 \mathbf{E}_2) &= 0, \end{aligned} \right\} \quad (2.3)$$

where

$$\left. \begin{aligned} \tau_h &= \chi_{00} - 2\varepsilon_h, & \varepsilon_h &= \frac{1}{2} \alpha_h + \frac{\varepsilon_0}{\beta_h}, & \alpha_h &= \frac{(\boldsymbol{\kappa} + \mathbf{K}_h)^2 - \kappa^2}{\kappa^2}, \\ \beta_h &= \gamma_0 / \gamma_h, & \gamma_h &= \mathbf{s}_h \mathbf{n}, & \mathbf{s}_h &= \mathbf{k}_h / |\mathbf{k}_h|, & \chi_{hh'} &= \chi(\mathbf{k}_h, \mathbf{k}_{h'}) \end{aligned} \right\} \quad (2.4)$$

Generally speaking, the transverse character of X-rays inside the crystal is not conserved. However, the longitudinal components are by a factor of χ smaller than the transverse ones, and they may be neglected. Nevertheless, it is convenient to preserve the last terms on the left-hand side of (2.3) and to consider formally the polarization of the electrical field to be arbitrary. Then (2.3) becomes a set of nine equations for the Cartesian components of the vectors \mathbf{E}_h . Its determinant must be equal to zero for the existence of a nontrivial solution

$$\Delta(\varepsilon_0) = \begin{vmatrix} (\tau_0 + s_0^x s_0^x) & \chi_{01} & \chi_{02} & s_0^x s_0^y & 0 & 0 & s_0^x s_0^z & 0 & 0 \\ \chi_{10} & (\tau_1 + s_1^x s_1^x) & \chi_{12} & 0 & s_1^x s_1^y & 0 & 0 & s_1^x s_1^z & 0 \\ \chi_{20} & \chi_{21} & (\tau_2 + s_2^x s_2^x) & 0 & 0 & s_2^x s_2^y & 0 & 0 & s_2^x s_2^z \\ s_0^y s_0^x & 0 & 0 & (\tau_0 + s_0^y s_0^y) & \chi_{01} & \chi_{02} & s_0^y s_0^z & 0 & 0 \\ 0 & s_1^y s_1^x & 0 & \chi_{10} & (\tau_1 + s_1^y s_1^y) & \chi_{12} & 0 & s_1^y s_1^z & 0 \\ 0 & 0 & s_2^y s_2^x & \chi_{20} & \chi_{21} & (\tau_2 + s_2^y s_2^y) & 0 & 0 & s_2^y s_2^z \\ s_0^z s_0^x & 0 & 0 & s_0^z s_0^y & 0 & 0 & (\tau_0 + s_0^z s_0^z) & \chi_{01} & \chi_{02} \\ 0 & s_1^z s_1^x & 0 & 0 & s_1^z s_1^y & 0 & \chi_{10} & (\tau_1 + s_1^z s_1^z) & \chi_{12} \\ 0 & 0 & s_2^z s_2^x & 0 & 0 & s_2^z s_2^y & \chi_{20} & \chi_{21} & (\tau_2 + s_2^z s_2^z) \end{vmatrix} = 0. \quad (2.5)$$

The determinant $\Delta(\varepsilon_0)$ does not depend on the choice of the coordinate system. Therefore, $\Delta(\varepsilon_0)$ depends on the quantities $s_{hh'} = (\mathbf{s}_h \mathbf{s}_{h'})$ only, and this dependence, as it is easy to understand from the form of (2.5), has the form

$$\Delta(\varepsilon_0) = F + G_0 s_{12}^2 + G_1 s_{02}^2 + G_2 s_{01}^2 + H s_{01} s_{02} s_{12}. \quad (2.6)$$

For a discussion of the coefficients in (2.6) we consider a few simple particular cases, where it is easy to calculate the determinant $\Delta(\varepsilon_0)$ directly. For example, let us consider a case in which the vectors \mathbf{s}_0 , \mathbf{s}_1 , and \mathbf{s}_2 are mutually perpendicular. In this case, we can choose the axes of the Cartesian coordinate system along these vectors, so that $s_0^x = 1$, $s_1^y = 1$, $s_2^z = 1$, and the other components of the vectors are equal to zero. Then, from (2.5), we get

$$\Delta(\varepsilon_0) = \begin{vmatrix} \tau_0 + 1 & \chi_{01} & \chi_{02} & \tau_0 & \chi_{01} & \chi_{02} & \tau_0 & \chi_{01} & \chi_{02} \\ \chi_{10} & \tau_1 & \chi_{12} & \chi_{10} & \tau_1 + 1 & \chi_{12} & \chi_{10} & \tau_1 & \chi_{12} \\ \chi_{20} & \chi_{21} & \tau_2 & \chi_{20} & \chi_{21} & \tau_2 & \chi_{20} & \chi_{21} & \tau_2 + 1 \end{vmatrix}. \quad (2.7)$$

On the other hand, from (2.6) it follows that the expression (2.7) determines the coefficient F . We shall be interested only in those roots which are connected with the transverse components of the fields. It is obvious in advance that these roots are small and are of the same order as χ . Therefore, we shall retain in (2.7) terms of the order of the least degree (six) of χ . As a result, we obtain

$$F = \Delta_{01} \Delta_{02} \Delta_{12}, \quad (2.8)$$

where

$$\Delta_{hh'} = \begin{vmatrix} \tau_h & \chi_{hh'} \\ \chi_{hh'} & \tau_{h'} \end{vmatrix}. \quad (2.9)$$

Analogously, considering other particular cases

2. $\mathbf{s}_0 = \mathbf{s}_1 \perp \mathbf{s}_2$
3. $\mathbf{s}_0 = \mathbf{s}_2 \perp \mathbf{s}_1$
4. $\mathbf{s}_0 \perp \mathbf{s}_1 = \mathbf{s}_2$
5. $\mathbf{s}_0 = \mathbf{s}_1 = \mathbf{s}_2$

we easily find

$$\left. \begin{aligned} 2. F + G_2 &= \tau_2 \Delta_{01} \Delta_3, \\ 3. F + G_1 &= \tau_1 \Delta_{02} \Delta_3, \\ 4. F + G_0 &= \tau_0 \Delta_{12} \Delta_3, \\ 5. F + G_0 + G_1 + G_2 + H &= \Delta_3^2, \end{aligned} \right\} \quad (2.10)$$

where

$$\Delta_3 = \begin{vmatrix} \tau_0 & \chi_{01} & \chi_{02} \\ \chi_{10} & \tau_1 & \chi_{12} \\ \chi_{20} & \chi_{21} & \tau_2 \end{vmatrix}. \quad (2.11)$$

As a result, according to (2.6), (2.8), (2.10), the dispersion equation has the form

$$\begin{aligned} \Delta(\varepsilon_0) &= \Delta_3[s^3 \Delta_3 + (s_{01}^2 - s^3) \tau_2 \Delta_{01} + (s_{02}^2 - s^3) \tau_1 \Delta_{02} + \\ &+ (s_{12}^2 - s^3) \tau_0 \Delta_{12}] + \Omega^2 \Delta_{01} \Delta_{02} \Delta_{12} = 0, \end{aligned} \quad (2.12)$$

where

$$s^3 = s_{01} s_{02} s_{12}, \quad \Omega^2 = 1 - s_{01}^2 - s_{02}^2 - s_{12}^2 + 2s^3 = (\mathbf{s}_0[\mathbf{s}_1 \times \mathbf{s}_2])^2. \quad (2.13)$$

The form of the dispersion equation obtained differs from that given by Ewald and Heno [6]. However, one can verify directly that these equations are identical. We think that the derivation presented above is a subject of independent interest because it can be useful in the n -wave case ($n > 3$) also, while the Ewald method is inconvenient even in the four-wave case [7].

Moreover, the dispersion equation in the form (2.12) is convenient also in cases when the parameter Ω^2 is close to zero. In these cases, for the solution of the problem, we consider the last term on the lefthand side of the equation to be small and treat it as a perturbation. In the zeroth approximation the solution of (2.12) is reduced to finding the roots of a third degree polynomial only.

The parameter Ω strictly equals zero when the three vectors \mathbf{s}_0 , \mathbf{s}_1 , and \mathbf{s}_2 lie in the same plane. Consequently, all cases which are close to this one can be investigated by means of the scheme outlined above.

3. Estimation of the Minimal Absorption Coefficient

One of the principal problems of the dynamical theory of X-ray diffraction is to find the points on the dispersion surface which correspond to a strong decrease of the absorption coefficient, that is to search for the cases where the Borrmann effect is realized most strongly.

For finding the minimal absorption coefficient $\mu_{\min}^{(3)}$, it is necessary to examine all values of α_1 , α_2 , and this is a rather hard work. Therefore it is interesting to find a way of estimating $\mu_{\min}^{(3)}$ without solving the total dispersion equation.

We write the set of equations (2.3) considering (2.4) in the form

$$\sum_{h'} \chi_{hh'} \mathbf{E}_{h'} = \left(\frac{2\varepsilon_0}{\beta_h} + \alpha_h \right) \mathbf{E}_h. \quad (3.1)$$

The matrix of the coefficients of (3.1) $\chi_{hh'}$ is known to have the following form (cf., for example, [12]):

$$\begin{aligned} \chi_{hh'} &= \chi_{rhh'} + i\chi_{ihh'} = \\ &= -\frac{4\pi r_0}{\chi^2 \Omega_0} \sum_j [f_j(|\mathbf{k}_{h'} - \mathbf{k}_h|) + i f'_{jh'}] \exp \{i(\mathbf{k}_{h'} - \mathbf{k}_h) \cdot \mathbf{Q}_j - M_j(|\mathbf{k}_{h'} - \mathbf{k}_h|)\}. \end{aligned} \quad (3.2)$$

Here the notations are taken from [12]. The matrixes $\hat{\chi}_r$ and $\hat{\chi}_i$, connected with the real and imaginary parts of the atomic formfactor f_j , are hermitean matrices to a good approximation. They are complex in a crystal with two and more atoms in the unit cell.

We multiply (3.1) by \mathbf{E}_h^* and take the sum over h

$$\sum_{hh'} \mathbf{E}_h^* (\chi_{rhh'} - \alpha_h \delta_{hh'}) \mathbf{E}_{h'} + i \sum_{hh'} \mathbf{E}_h^* \chi_{ihh'} \mathbf{E}_{h'} = 2\epsilon_0 \sum_h |\mathbf{E}_h|^2 \frac{1}{\beta_h}. \quad (3.3)$$

Then we separate the equation (3.3) into real and imaginary parts. Because the matrixes $(\hat{\chi}_r - \alpha)$ and $\hat{\chi}_i$ are hermitean, the first term on the left-hand side of (3.2) is real, and the second one is purely imaginary. Therefore

$$2\epsilon'_0 = \frac{\sum_{hh'} \mathbf{E}_h^* (\chi_{rhh'} - \alpha_h \delta_{hh'}) \mathbf{E}_{h'}}{\sum_h |\mathbf{E}_h|^2 \beta_h^{-1}}, \quad (3.4)$$

$$2\epsilon''_0 = \frac{\sum_{hh'} \mathbf{E}_h^* \chi_{ihh'} \mathbf{E}_{h'}}{\sum_h |\mathbf{E}_h|^2 \beta_h^{-1}}. \quad (3.5)$$

Here $\epsilon_0 = \epsilon'_0 + i\epsilon''_0$. The formulae (3.3), (3.4) determine the real and imaginary parts of the eigenvalues of the set (3.1) through its eigen vectors.

We are interested in the minimal value of the absorption coefficient $\mu_{\min}^{(3)} = 2\kappa\epsilon''_{0\min}$. This value is determined by the minimum of the right-hand side of equation (3.5) under the condition that the wave field amplitudes \mathbf{E}_h satisfy the transverse condition $\mathbf{s}_h \mathbf{E}_h = 0$ and (3.1). We neglect the requirement that the amplitudes satisfy (3.1) for the estimation of the minimal value of ϵ''_0 . Then we, naturally, get the lower limit only for $\epsilon''_{0\min}$, which we denote as $\tilde{\epsilon}''_{0\min}$. The knowledge of this limit gives us the possibility to estimate the magnitude of the Borrmann effect. Moreover, as it shall be shown below this limit lies near to the true value of $\epsilon''_{0\min}$ in a large number of cases.

For finding $\tilde{\epsilon}''_{0\min}$, we take the derivative of the right-hand side of (3.5) with respect to \mathbf{E}_h^* equal to zero

$$\frac{\delta 2\tilde{\epsilon}''_0}{\delta \mathbf{E}_h^*} = \left\{ \sum_{h'} \chi_{ihh'} \mathbf{E}_{h'} - \frac{\sum_{hh'} \mathbf{E}_h^* \chi_{ihh'} \mathbf{E}_{h'}}{\sum_h |\mathbf{E}_h|^2 \beta_h^{-1}} \frac{1}{\beta_h} \mathbf{E}_h \right\} \left/ \sum_h |\mathbf{E}_h|^2 \beta_h^{-1} = 0. \quad (3.6) \right.$$

According to (3.5) and (3.6), we obtain

$$\sum_{h'} \chi_{ihh'} \mathbf{E}_{h'} = \frac{2\tilde{\epsilon}''_0}{\beta_h} \mathbf{E}_h. \quad (3.7)$$

Consequently $\tilde{\epsilon}''_{0\min}$ is the minimal eigenvalue of the set (3.7) which differs from the general set (3.1) by the substitution of $\chi_{hh'}$ by $\chi_{ihh'}$ and by the absence of α_h .

The solution of the set (3.7) is a much simpler problem. In this case, the dispersion equation has real coefficients. Moreover, one must do the calculation only once without analysing the dependence of ϵ''_0 on the parameters α_1, α_2 . On the other hand, all results, obtained in the previous section for the general set (3.1), apply to the solution of the set (3.7). Particularly, the eigenvalues of this set are the roots of equation (2.12) with $\alpha_1 = \alpha_2 = 0$ and $\chi_{hh'} = \chi_{ihh'}$.

As an example of the application of the method developed above, we consider the symmetrical Laue-Laue case, $\beta_1 = \beta_2 = 1$, for a crystal with an inversion centre.

In such crystals, as it can be verified directly, the relation

$$\chi_{01}\chi_{12}\chi_{20} = \chi_{10}\chi_{02}\chi_{21} = \chi_1\chi_2\chi_3 \quad (3.8)$$

is satisfied. Here and later on we use the following abbreviations:

$$\begin{aligned} \chi_0 &= \chi_{00}, & \chi_1 &= \sqrt{\chi_{01}\chi_{10}}, & \chi_2 &= \sqrt{\chi_{02}\chi_{20}}, \\ \chi_3 &= \sqrt{\chi_{12}\chi_{21}}, & s_1 &= s_{01}, & s_2 &= s_{02}, & s_3 &= s_{12}. \end{aligned} \quad (3.9)$$

(We shall assume the principal root to have a positive imaginary part.) In accordance with (3.8), the dispersion equation (2.12) depends on four quantities χ_h only. Because the matrix $\chi_{ihh'}$ is hermitean, the corresponding quantities χ_{ih} are purely real.

For an estimate of the quantity $\tilde{\mu}_{\min}^{(3)}$, it is interesting to compare it with the value of the two-wave minimal absorption coefficient $\mu_{\min}^{(2)}$ for the same reflections. As it is well known, in the symmetrical Laue-case, $\beta = 1$,

$$\mu_{\min}^{(2)}(h, h') = \kappa(\chi_{00}'' - \sqrt{\chi_{hh'}\chi_{h'h}''}) = \kappa(\chi_{i0} - \chi_{ih+h'}). \quad (3.10)$$

Without restriction of generality (cf. Section 5) we can assume

$$\chi_{i1} > \chi_{i2} > \chi_{i3}. \quad (3.11)$$

Then the two-wave minimal absorption coefficient is realized on the first reflection. Therefore it is convenient to introduce the new quantity $\varepsilon = \chi_{i0} - \chi_{i1} - 2\varepsilon_0''$ which directly determines the difference between the three-wave minimal absorption coefficient and the two-wave one, namely,

$$\mu_{\min}^{(2)}(0, 1) - \tilde{\mu}_{\min}^{(3)} = \kappa\varepsilon. \quad (3.12)$$

In accordance with the above remarks we obtain from (2.12) the following equation for ε :

$$\Delta(\chi_{ih}, \varepsilon) = (\varepsilon^3 - 3\chi_{i1}\varepsilon^2 + B\varepsilon + C) [(1 - \Omega^2)(\varepsilon^3 + 3\chi_{i1}\varepsilon^2) + B_1\varepsilon + C_1] + \Omega^2\varepsilon(\varepsilon + 2\chi_{i1})(\varepsilon^2 + 2\chi_{i1}\varepsilon + \chi_{i1}^2 - \chi_{i2}^2)(\varepsilon^2 + 2\chi_{i1}\varepsilon + \chi_{i1}^2 - \chi_{i3}^2) = 0, \quad (3.13)$$

where

$$\left. \begin{aligned} B &= 2\chi_{i1}^2 - \chi_{i2} - \chi_{i3}, & C &= -\chi_{i1}(\chi_{i2} - \chi_{i3})^2, \\ B_1 &= 2\chi_{i1}^2(1 - s^3 - \Omega^2) + (\chi_{i1}^2 - \chi_{i2}^2)s_2^2 + (\chi_{i1}^2 - \chi_{i3}^2)s_3^2, \\ C_1 &= \chi_{i1}[(\chi_{i1}^2 - \chi_{i2}^2)s_2^2 + (\chi_{i1}^2 - \chi_{i3}^2)s_3^2 - 2(\chi_{i1}^2 - \chi_{i2}\chi_{i3})s_3^2]. \end{aligned} \right\} \quad (3.14)$$

The numerical solution of equation (3.13) is not difficult because the function $\Delta(\chi_{ih}, \varepsilon)$ is real.

It is an interesting question whether positive roots of equation (3.13) are possible. Qualitatively this question is easily investigated directly. Indeed, according to (3.11), we see that the second term in (3.13) is always positive for positive ε , and the first one has at least one positive root, if $\chi_{i2} \neq \chi_{i3}$, because $B > 0$, but $C < 0$ in this case. Consequently, under this condition, (3.13) has always one positive root, and this root is the larger the larger the asymmetry and the smaller the parameter Ω^2 . If $\chi_{i2} = \chi_{i3}$, then no positive roots exist, the maximum root equals zero. It is easy to understand, that the estimate is exact in this case, because the two-wave minimum is always reached in the limit $|\alpha_2| \rightarrow \infty$.

4. Passage from Two-Wave to Three-Wave Case

As it has been shown in the previous section, if the diffraction of X-rays takes place on two systems of planes simultaneously, under certain conditions an additional decrease of the anomalous absorption coefficient occurs. It is obvious that the point of the minimum of the three-wave absorption coefficient $\mu^{(3)}$ lies in the central region of the dispersion surface, $|\alpha_1|, |\alpha_2| \leq |\chi'_0|$, and the dependence of $\mu^{(3)}$ on α_1, α_2 is quadratic near this point. However, in the area far from the minimal point, $\mu^{(3)}$ depends on α_1, α_2 in a more complicated way.

The case $|\alpha_2| \gg |\chi'_0|$ is of special interest because it gives us a possibility to analyse clearly the passage from two-wave to three-wave case.

We write equation (2.12) in the form of a power series of τ_2 and use the abbreviations (3.9)

$$\Delta(\varepsilon_0) = \tau_2^2 D_2(\tau_0, \tau_1) + \tau_2 D_1(\tau_0, \tau_1) + D_0(\tau_0, \tau_1) = 0, \quad (4.1)$$

where

$$D_2 = (\tau_0 \tau_1 - \chi_1^2) (\tau_0 \tau_1 - \chi_1^2 s_1^2) \quad (4.2)$$

is the dispersion polynomial of the two-wave case, and

$$D_1 = (\tau_0 \tau_1 - \chi_1^2) (2\chi_1 \chi_2 \chi_3 s_3^3 - \tau_0 \chi_3^2 s_3^2 - \tau_1 \chi_2^2 s_2^2) + (\tau_0 \tau_1 - \tau_1 \chi_1^2 s_1^2) \times \\ \times (2\chi_1 \chi_2 \chi_3 - \tau_0 \chi_3^2 - \tau_1 \chi_2^2) - \Omega^2 (2\chi_1 \chi_2 \chi_3 \tau_0 \tau_1 - \tau_0 \chi_1^2 \chi_3^2 - \tau_1 \chi_1^2 \chi_2^2), \quad (4.3)$$

$$D_0 = (2\chi_1 \chi_2 \chi_3 - \tau_0 \chi_3^2 - \tau_1 \chi_2^2) (2\chi_1 \chi_2 \chi_3 s_3^3 - \tau_0 \chi_3^2 s_3^2 - \tau_1 \chi_2^2 s_2^2) + \\ + \Omega^2 \chi_2^2 \chi_3^2 (\tau_0 \tau_1 - \chi_1^2). \quad (4.4)$$

Here we use the relation (3.8) which is fulfilled for a crystal of Ge-type.

According to (2.4), in the limit $|\alpha_2| \gg |\chi'_0|$ we can assume $\tau_2 = -\alpha_2$ with good accuracy. Then the roots of the dispersion equation are determined by the first term in the right-hand side of (4.1). These roots represent four two-wave branches, that is roots of the equation $D_2(\tau_0, \tau_1) = 0$, and two branches with a normal absorption coefficient. We are interested in the addition to the two-wave solutions, which we call $\tilde{\tau}_{0j}$, $j = 1, 2, 3, 4$. We write $\tau_{0j} = \tilde{\tau}_{0j} + \varepsilon_j$, where ε_j is a small addition, and substitute it into (4.2) to (4.4). We can restrict ourselves to first-degree terms in ε_j because we have $|\varepsilon_j| \rightarrow 0$ in the limit $|\alpha_2| \rightarrow \infty$. As a result, we obtain

$$D_2 = \varepsilon_j B_2(\tilde{\tau}_{0j}) + \dots, \quad D_{0,1} = A_{0,1}(\tilde{\tau}_{0j}) + \varepsilon_j B_{0,1}(\tilde{\tau}_{0j}) + \dots \quad (4.5)$$

The quantity ε_j determines directly the addition to the two-wave absorption coefficient $\mu_{01}^{(2)}(j, \alpha_1)$, namely,

$$\Delta\mu^{(3)} = \mu^{(3)}(\alpha_1, \alpha_2) - \mu_{01}^{(2)}(j, \alpha_1) = -\varepsilon_j \varepsilon_j'. \quad (4.6)$$

We have from (4.1) and (4.5) the relation

$$\varepsilon_j = \frac{1}{\alpha_2} \frac{(A_1 - A_0 \alpha_2^{-1})}{(B_2 - B_1 \alpha_2^{-1} + B_0 \alpha_2^{-2})}. \quad (4.7)$$

Let us consider the case $A_1 \neq 0$ and $B_2 \neq 0$. Then we obtain from (4.7) that $\Delta\mu^{(3)}$ is always positive for a certain sign of α_2 and negative for the opposite sign. The analogous result can be easily obtained also for the two branches with normal absorption coefficient. In the central region of the dispersion surface (when $|\alpha_2|$ is small) the lower two-wave branches turn into the upper ones and also into branches with normal absorption coefficient when $|\alpha_2| \gg |\chi'_0|$, and vice versa. Moreover, we obtain that the minimum and the maximum of the three-wave curves $\mu^{(3)}(\alpha_2)$ are always smaller and larger, respectively, than the two-wave ones with the same α_1 .

The addition to the two-wave branch with minimal absorption coefficient (Borrmann-effect) is more interesting. Below we shall consider this case in more detail. For the sake of simplicity we shall not write the index j . The interesting solution satisfies the relation

$$\tilde{\tau}_0 \tilde{\tau}_1 = \chi_1^2. \quad (4.8)$$

The coefficients in (4.7), according to (4.8), are equal to

$$\left. \begin{aligned} B_2 &= 2t\chi_1^2(1 - s_1^2), \\ B_1 &= -2t\chi_1\chi_2\chi_3 \left[2(\Omega^2 - s^3) + s_2^2 \frac{1}{x} + s_3^2 x \right] - \chi_1^2(\chi_2^2 + \chi_3^2)(1 - s_1^2 - \Omega^2), \\ B_0 &= 2t\chi_2^2\chi_3^2\Omega^2 + \chi_1\chi_2\chi_3(\chi_2^2 + \chi_3^2) \left(s_2^2 \frac{1}{x} + s_3^2 x - 2s^3 \right), \\ A_1 &= -\frac{(1-x)^2}{x} \chi_1^2\chi_2\chi_3(1 - s_1^2 - \Omega^2), \\ A_0 &= \frac{(1-x)^2}{x} \chi_1^2\chi_2^2\chi_3^2 \left(s_2^2 \frac{1}{x} + s_3^2 x - 2s^3 \right), \end{aligned} \right\} \quad (4.9)$$

where

$$2t = \tilde{\tau}_0 + \tilde{\tau}_1, \quad x = \tilde{\tau}_0 \frac{\chi_3}{\chi_1\chi_2}. \quad (4.10)$$

For $x = 1$ it follows from (4.9) that the coefficients A_1 and A_0 strictly equal zero. In this case the two-wave value of τ_0 is strictly conserved for all α_2 and β_2 , and this result, naturally, does not depend on the condition $|\alpha_2| \gg |\chi_0|$.

Now we assume $\alpha_1 = 0$, $\beta_1 = 1$, but $\chi_2 \neq \chi_3$. In this case all coefficients in (4.7) differ from zero. Therefore, for large values of $|\alpha_2|$ $\Delta\mu^{(3)}$ is determined approximately by the expression

$$\Delta\mu^{(3)} = -\frac{\kappa}{\alpha_2} \left(\frac{A_1}{B_2} \right)'' = \frac{\kappa}{\alpha_2} (\chi_2' - \chi_3') (\chi_2'' - \chi_3'') \frac{(1 - s_1^2 - \Omega^2)}{(1 - s_1^2)}. \quad (4.11)$$

If the condition (3.11) is fulfilled, then (4.7) determines the additional decrease in the minimal two-wave absorption coefficient. As it follows from (4.11), in this case the region of the three-wave dispersion surface with small absorption coefficient is strongly stretched along the line $\alpha_1 = 0$ towards the side of positive α_2 . Moreover, according to (4.11), $|\Delta\mu^{(3)}|$ is the larger, the stronger the asymmetry is and the smaller the parameter Ω^2 . This is in accordance with the result of the previous section.

5. General Properties of Dispersion Surface

In this section we shall discuss the general properties of the dispersion surface $\varepsilon_0(\alpha_1, \alpha_2)$, namely, the symmetry properties of the function $\varepsilon_0'(\alpha_1, \alpha_2)$ and the question about the triply degenerate point on the dispersion surface. The evident consideration of this properties give us in some cases a possibility to understand physically the experimental results without solving the problem. It is essential also for the numerical solution of the dispersion equation.

Taking into account (2.9), (2.11), one easily sees that the left-hand side of equation (2.12) is not changed when replacing the indices 0, 1, 2, respectively

by h, k, l , where h, k, l is some permutation of the indices 0, 1, 2. This is because the vectors $\mathbf{k}_0, \mathbf{k}_1, \mathbf{k}_2$ enter (2.12) quite equivalently. Therefore all cases which differ from each other only by the permutation of these vectors, must have much in common.

It is easy to understand this generality physically. Indeed, from the experimental point of view the three-wave case is determined by two systems of planes, that is the reciprocal-lattice vectors \mathbf{K}_1 and \mathbf{K}_2 , which reflect the incident plane wave with the wave-vector $\boldsymbol{\kappa}$. In fact, however, an unresolvable superposition of waves arises in the crystal bulk, the individual components of which turn one into another being reflected on three systems of planes with the reciprocal-lattice vectors $\mathbf{K}_1, \mathbf{K}_2$ and $\mathbf{K}_3 = \mathbf{K}_2 - \mathbf{K}_1$. Only the distances between these planes, that is the moduli of these vectors, are important.

Let us consider, for the sake of simplicity, crystals of Ge-type. In this case we have, besides the zero Fourier component of the polarizability, only three independent coefficients $\chi_h = \chi(|\mathbf{K}_h|)$, $h = 1, 2, 3$, which are determined by (3.9). We note that the modulus of the structure amplitude depends on \mathbf{K}_h , of course, more complicatedly. However, this dependence can always be accounted for explicitly, and it does not change the conclusion given below. If all χ_h are not equal then, permuting the indices, we have six different cases. The following table shows the accordance between them. The quantities in any column of the table are equal to one another

$$\left. \begin{array}{l}
 1. \chi_1, \chi_2, \chi_3; \quad \tau_0, \tau_1, \tau_2, \\
 2. \chi_2, \chi_1, \chi_3; \quad \tau_0, \tau_2, \tau_1, \\
 3. \chi_1, \chi_3, \chi_2; \quad \tau_1, \tau_0, \tau_2, \\
 4. \chi_2, \chi_3, \chi_1; \quad \tau_2, \tau_0, \tau_1, \\
 5. \chi_3, \chi_1, \chi_2; \quad \tau_1, \tau_2, \tau_0, \\
 6. \chi_3, \chi_2, \chi_1; \quad \tau_2, \tau_1, \tau_0.
 \end{array} \right\} \quad (5.1)$$

The dispersion surfaces in all these cases show an one-to-one correspondence. We show how this correspondence is determined. For example, $\varepsilon_0 = E$ is a solution at any point $(\alpha_1, \alpha_2, \beta_1, \beta_2) = (A_1, A_2, B_1, B_2)$ for case 1 of (5.1). Then we can easily obtain the solution (2.12) at some point of the space $(\alpha_1, \alpha_2, \beta_1, \beta_2)$ for case 2 from (5.1). Indeed, according to (2.4), we find for case 1

$$\tau_0^{(1)} = \chi_0 - 2E, \quad \tau_{1,2}^{(1)} = \chi_0 - \frac{2E}{B_{1,2}} - A_{1,2}. \quad (5.2)$$

Passing to case 2, we notice that the following three values of τ_h are solutions of (2.12)

$$\tau_0^{(2)} = \chi_0 - 2E, \quad \tau_{1,2}^{(2)} = \chi_0 - \frac{2E}{B_{2,1}} - A_{2,1}. \quad (5.3)$$

We use formula (2.4) once more and obtain the same solution $\varepsilon_0 = E$, but at the point $(\alpha_1, \alpha_2, \beta_1, \beta_2) = (A_2, A_1, B_2, B_1)$.

The minimum of ε_0'' is more interesting. It is easy to understand that ε_0'' has the minimal value in cases of table (5.1) which correspond to $\beta_1 < 1$ and $\beta_2 < 1$. However, the quantity ε_0''/γ_0 is constant for all cases. Therefore all cases have the same minimal value of the quantity ε_0''/γ_0 .

Let us consider now the case of symmetrical Laue-Laue geometry. In this case the quantities $\beta_1 = \beta_2 = 1$ and ε_0' are the same for all cases in (5.1). Con-

sequently, the imaginary parts of the dispersion surfaces $\varepsilon_0''(\alpha_1, \alpha_2)$ in these cases have the same region of changing and turn into one another by means of rotations and inversions. If two coefficients χ_h are equal or all three coefficients are equal, then the function $\varepsilon_0''(\alpha_1, \alpha_2)$ is invariable with respect to these transformation.

As an example, we consider the case (220), (111), (111). According to the results of previous section, in this case the two-wave solution on (220) with the minimal imaginary part is conserved along the line $\alpha_1 = 0$. Moreover, because the cases 1 and 3 of (5.1) coincide, ε_0'' has the same values in the points $(\alpha_1, \alpha_2) = (A_1, A_2)$ and $(\alpha_1, \alpha_2) = (-A_1, A_2 - A_1)$. In the completely symmetrical case (220), (022), (202), when $\chi_1 = \chi_2 = \chi_3$, we obtain that the minimal imaginary part of the two-wave solution on (220) is conserved along the three lines $\alpha_1 = 0$, $\alpha_2 = 0$, $\alpha_1 = \alpha_2$, and ε_0'' has the same value at six points of the plane (α_1, α_2) .

Finally, we note one interesting property of the three-wave dispersion surface. If one neglects the imaginary part of the atomic form-factor, which is essential only for the absorption process, then always a point is on the plane α_1, α_2 in which the dispersion equation has a triply degenerate root. This fact does not depend on the parameters of the problem, namely, the geometry of the experiment and the structure of the crystal. It is obvious that in the vicinity of this point one may expect the unusual behaviour of the "pendellösung" effect.

It is easy to verify directly that the coordinates of this point are determined by the relation

$$\tau_0 = \frac{\chi_1 \chi_2}{\chi_3}, \quad \tau_0 \tau_1 = \chi_1^2, \quad \tau_0 \tau_2 = \chi_2^2. \quad (5.4)$$

According to (2.4), we obtain from (5.4)

$$\left. \begin{aligned} \alpha_1 &= \chi_0'(1 - \beta_1^{-1}) - \chi_1'(\chi_3'^2 - \chi_2'^2 \beta_1^{-1}) / \chi_2' \chi_3', \\ \alpha_2 &= \chi_0'(1 - \beta_2^{-1}) - \chi_2'(\chi_3'^2 - \chi_1'^2 \beta_2^{-1}) / \chi_1' \chi_3'. \end{aligned} \right\} \quad (5.5)$$

It must be noted that the point of degeneracy is absent when the structure amplitude for one of the reflections equals zero. It is easy to see that in this case one of the quantities α_1, α_2 in (5.5) equals ∞ . As it follows from the results of the previous section, this point is the point of intersection of three straight lines along which the two-wave solution with the minimal imaginary part is conserved.

References

- [1] G. BORRMANN and W. HARTWIG, *Z. Krist.* **121**, 401 (1965).
- [2] G. HILDEBRANDT, *phys. stat. sol.* **24**, 245 (1967).
- [3] E. J. SACCOGIO and A. ZAJAC, *Phys. Rev.* **139**, A255 (1965).
- [4] T. JOKO and A. FUKUHARA, *J. Phys. Soc. Japan* **22**, 597 (1967).
- [5] M. L. KSHEVETSKAYA, S. A. KSHEVETSKII, I. P. MIHAILYUK, and N. D. RARANSKII, *Ukr. fiz. Zh.* **18**, 578 (1973); **18**, 1171 (1973).
- [6] P. P. EWALD and Y. HENO, *Acta cryst.* **A24**, 5 (1968).
- [7] P. PENNING, *Philips Res. Rep.* **23**, 12 (1968).
- [8] T. I. BORODINA, A. A. KATSNELSON, and V. I. KISSIN, *phys. stat. sol. (a)* **3**, 105 (1970).
- [9] V. I. IVERONOVA, A. A. KATSNELSON, T. J. BORODINA, and I. G. SAPKOVA, *phys. stat. sol. (a)* **11**, 39 (1972).
- [10] S. BALTER, R. FELDMAN, and B. POST, *Phys. Rev. Letters* **27**, 307 (1971).
- [11] R. FELDMAN and B. POST, *phys. stat. sol. (a)* **12**, 273 (1972).
- [12] A. M. AFANASEV and YU. KAGAN, *Acta cryst.* **A24**, 163 (1968).

(Received November 20, 1974)