introducing absorption, the origin of the enhancement is manifest from the general resonance denominator $(K_h^2 - k_0^2)^{-1}$. In (II) absorption and atomic factors are introduced and a graphical and numerical rendering of the surface of dispersion is given for variable wavelength and, consequently, angular settings. Finally the effective coefficient of absorption, $\mu_{\rm eff}$, is given for two different three-beam cases in germanium for each of the six sheets of the surface of dispersion. The curves show $\mu_{\rm eff}$ as a function of λ/a for the cases $n = 1$, 2 and 3. They show strikingly to what extent a perfect crystal can become transmittant in one of its proper modes, while in others it becomes more opaque. The value of this analytical treatment lies in the general survey it provides, and as such it is satisfactory. On the other hand, it is labor saving to use computer methods in future cases.

Retrospect

I am lucky to have spent so many of my best years in doing research I have loved; in having been well prepared for Laue's discovery by my thesis work; by taking an active part in developing the theory of the subject; by gradually simplifying the mathematics and by arriving at an increasingly deeper understanding of the varied aspects of crystal optics.

On the other hand, my interest always centered on the perfect crystal in which I saw the preferred material for exacting optical investigation. Herein lies a strong limitation, and an abstraction which in important aspects is contrary to nature. I am happy to see how others have been, still are, and will be carrying on beyond the limitations I set for myself.

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On Extinction.

III. An Improvement of the Secondary-Extinction Theory*

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Abstract

The previous theory [Kato (1976). *Acta Cryst.* A**32**, 458–466] is improved by taking into account the higher-order correlations of lattice phase factors. The previous coupling constants in the energy-transfer equations have, in general, to be reduced. In the simplest case of non-absorbing crystals the reduction factor can be given as

$$R = 1 + (\tau_2)^{-1} \sum_{j=1}^{\infty} V_2^{(j+1)}(-\kappa_g \kappa_{-g})^j,$$

where $\kappa_{\pm g}$ is the kinematical diffraction amplitude per unit length for $\pm\mathbf{g}$ reflection, and $V_2^{(j)}$ is the correlation

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volume, which is defined for the $2j$th-order correlation function. The quantity $V_2^{(j)}$ is an extended concept of the correlation length τ_2 for the second-order correlation function in the sense that $V_2^{(1)} = \tau_2$. The set of $\{V_2^{(j)}\}$ characterizes the statistical nature of the crystalline medium. The present theory is conjectured to be applicable up to $\tau_2 \lesssim \Lambda$, in which Λ is the extinction distance.

I. Introduction

In previous papers [Kato, 1976*a*,*b*; (I) and (II) hereafter], the present author formulated an energy-transfer equation, from the wave equation of Takagi–Taupin type, for describing secondary extinction. In this formalism the coupling constants between the direct and the Bragg-reflected beams were given as being proportional to the kinematical diffraction power per unit length times the correlation length of the lattice phase factors at two distant positions. The formulation, however, was limited to the cases in which the correlation length is much shorter than the extinction distance.

A question then arises as to whether the theory can be extended to cover less imperfect crystals in which the correlation length is comparable to the extinction distance. In some of the literature (for example, Zachariasen, 1945), to meet this requirement it is proposed on the basis of physical intuition that the kinematical diffraction power be replaced by the dynamical diffraction power in the coupling constants.

In this paper, the previous theory [(I) and (II)] is developed within the framework of secondary-extinction theory. The approach takes into account the higher-order correlation functions of the lattice phase factors, of order higher than second, which was the highest considered in the previous theory. The coupling constants of the previous theory have to be reduced by a factor which is definable from the statistical nature of lattice distortion. Effects of absorption in secondary-extinction phenomena are also discussed. The present theory is conjectured to be applicable to less imperfect crystals.

II. Outline of the previous theory

The basic idea of the present formulation is similar to the previous one. The total wave field is given by the sum of the wavelets which are associated with all possible optical routes having O and G directions in a zigzag way [*cf.* Fig. 2 of (I)]. The measurable intensity is an ensemble average of the intensity field expected for a set of lattice phases at kink points of the optical route. Here, the lattice phase, which will be denoted by G below, means $2\pi(\mathbf{g}.\mathbf{u})$ where $\mathbf{g}$ is the reflection vector and $\mathbf{u}$ is the displacement vector of a lattice point referred to an ideally perfect crystal [*cf.* equation (3) of (I)]. As shown in (I), therefore, the intensities of the O and G beams, respectively, are written in the forms

$$\begin{aligned}\langle I_o\rangle = (A/a)^2 \sum_R \sum_{R'} (i\kappa_g)^k\,(i\kappa_{-g})^k\,(-i\kappa_g^*)^{k'}\,(-i\kappa_{-g}^*)^{k'} \\ \times (a)^{2k+2k'}\,\langle\exp\{-i[G(1,0)-G(1,1) \\ +G(2,1)\ldots -G(k,k)]\}\exp\{i[G(1',0) \\ -G(1',1')+G(2',1')\ldots -G(k',K')]\}\rangle,\quad (1a)\end{aligned}$$

$$\begin{aligned}\langle I_g\rangle = (A/a)^2|\kappa_g|^2 \sum_R \sum_{R'} (i\kappa_g)^k\,(i\kappa_{-g})^k\,(-i\kappa_g^*)^{k'} \\ \times (-i\kappa_{-g}^*)^{k'}\,(a)^{2k+2k'+2}\,\langle\exp\{-i[G(1,0) \\ -G(1,1)+G(2,1)\ldots G(r,r) \\ +G(r+1,r)\ldots +G(k+1,k)]\} \\ \times \exp\{i[G(1',0)-G(1',1')\ldots G(r',r') \\ -G(r'+1,r')\ldots +G(k'+1,k')]\}\rangle,\end{aligned}$$

where k is the number of kink points of the route R and the indexes (r,r) and $(r+1,r)$ in the phase G are the abbreviation of the positions (s_o^r,s_g^r) and (s_o^{r+1},s_g^r) respectively. The position coordinates (s_o,s_g) are defined by the oblique coordinate axes along the O and G beams with the origin at the entrance point [*cf.* Figs. 1 and 2 of (I)]. The quantity $\{\kappa_{\pm g}\}$ is diffraction amplitude per unit length, which is proportional to the structure factor $F_{\pm g}$ [*cf.* equation (2) of (I)] and (a) is an increment parameter introduced for numbering the position parameters. It will be reduced to zero after the calculation. The prime identifies the complex conjugate waves.

In order to evaluate the ensemble average of the phase factor, each route is divided alternately by vertical and horizontal segments.† In the previous theory, the following assumptions were made. (1) The vertical (horizontal) segments are composed of a sequence of horizontal (vertical) kink pairs (KP). (2) The end of a vertical (horizontal) segment is associated with an isolated kink (IK) of a (*b*) type. (3) All KP's and IK's are separated by distances larger than the correlation length, τ, and the correlations among KP's and IK's were therefore neglected. To justify the third assumption, it was necessary to assume that

$$\tau \ll |\kappa_g|^{-1} = \Lambda \quad \text{(extinction distance).} \qquad (2)$$

This restriction, however, can be relaxed to some extent. In fact, there are many possible routes composed of many KP's which are not sufficiently separated from other KP's and IK's so that correlations

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† For convenience, 'vertical' and 'horizontal' are used for the directions of the O and G beams [see also Fig. 1 of (II)].

among them cannot be neglected. The aim of this paper is to take such routes properly into consideration. In preparation, the higher-order correlation functions for a group of kink points are first discussed.

III. The correlation functions of higher order

If the kink points are well separated from other kink points, the phase factors can be averaged independently of other phase factors. For this reason, in (II) it was sufficient to introduce the second-order correlation function

$$f(z) = \langle \exp\{i[G(a,b) - G(a+z,b)]\}\rangle \quad (3a)$$

$$= \langle \exp\{i[-G(a,b) + G(a,b+z)]\}\rangle. \quad (3b)$$

From the statistical isotropy and homogeneity of the crystal, it is safe to assume that $f(z)$ is a real, even function. More detailed considerations are given in (II) and Appendix A in (II).

In the present treatment, one needs correlation functions of higher order in the forms

$$f(y_1,x_1) = \langle \exp\{i[-G(1,0) + G(1,1) - G(2,1)]\}\rangle, \quad (4a)$$

$$f(y_1,x_1,y_2) = \langle \exp\{i[-G(1,0) + G(1,1) - G(2,1) + G(2,2)]\}\rangle, \quad (4b)$$

⋮

odd:

$$f(y_1,x_1,\ldots,y_r,x_r,\ldots,x_n) = \langle \exp\{i[-G(1,0) + \ldots + G(r,r) - G(r+1,r) \ldots - G(n+1,n)]\}\rangle, \quad (4c)$$

⋮

even:

$$f(y_1,x_1,\ldots,y_r,x_r,\ldots,y_n) = \langle \exp\{i[-G(1,0) + \ldots - G(r,r-1) + G(r,r) \ldots + G(n,n)]\}\rangle, \quad (4d)$$

where

x_r = the distance between (r,r) and $(r+1,r)$, (5a)

y_r = the distance between $(r,r-1)$ and (r,r). (5b)

In the case of (4a), the phases of the nearest kink points can be cancelled out but the remaining phase is random, so that the correlation function $f(y_1,x_1)$ must be zero. Similarly, the correlation functions of the lattice phase factors of odd numbers are always zero.

In the case of (4b), on the other hand, the principal parts of the phase factors can be cancelled out (unless y_1 and y_2 are larger than τ). If the neighbouring kink pairs are well separated, $f(y_1,x_1,y_2)$ is reduced to $f(y_1)\,f(y_2)$. In general, however, it must have the form

$$f(y_1,x_1,y_2) = f(y_1)\,f(y_2) + f_2(y_1,x_1,y_2), \quad (6)$$

where f_2 represents the intrinsic part of the correlation of two kink pairs separated by x_1. Obviously, f_2 tends to zero as x_1 increases. With this scheme of notation, $f(y)$ is identical to $f_1(y)$.

Extending the above argument, one can write the correlation function of higher order, $2n$, as follows:

$$f(y_1,x_1,y_2,\ldots,y_n) = f_1(y_1)\,f_1(y_2)\ldots f_1(y_n) + \{\text{possible products of } f_1, f_2,\ldots, f_{n-1}\} + f_n(y_1,x_1,y_2,\ldots,y_n). \quad (7)$$

If one of $\{x_r\}$ is larger than τ, f_n tends to zero. We shall call $\{f_n\}$ the intrinsic correlation function. Another general relation between f and f_n is given in Appendix A.

The explicit form of f_n depends on the model of the lattice distortion. For the present formal theory, however, one needs only the correlation volume defined by

$$V_1^{(n)} = \int_0^\infty \cdots \int_0^\infty f_n(y_1,x_1,\ldots,y_n)\,\mathrm{d}\{x_r\}\,\mathrm{d}\{y_r\}. \quad (8a)$$

As will be seen later, more often used is another correlation volume defined by

$$V_2^{(n)} = \int_0^\infty \cdots \int_0^\infty \prod_{r=1}^{n} f(y_r)\,f_n(y_1,x_1,\ldots,y_n)\,\mathrm{d}\{x_r\}\,\mathrm{d}\{y_r\}. \quad (8b)$$

These correlation volumes are extended concepts of the correlation lengths defined by equation (2) of (II). In fact, it is obvious that

$$V_1^{(1)} = \tau_1; \quad V_2^{(1)} = \tau_2. \quad (9a,b)$$

In order to show physical applications of the theory conveniently, we sometimes neglect the end effects of a sequence of KP's. In this case, integrating $f(y_1,x_1,y_2,x_2,\ldots,y_r,x_r,\ldots)$ over an x_r gives the same factor for all r and integrating over a y_r gives a different factor which is, again, the same for all r. With this approximation, one can write the explicit expressions of equations (8a and b) as

$$V_1^{(n)} = (\tau_1)^{2n-1}\,\beta_1^{n-1}; \quad V_2^{(n)} = (\tau_2)^{2n-1}\,\beta_2^{n-1} \quad (n \geq 1). \quad (10a,b)$$

The subscripts 1 and 2 are introduced in order to distinguish the two cases of equations (8a and b). The dimensionless quantity β indicates the ratio of the correlation lengths of the kink pair itself and of the two kink pairs. The concrete discussion of β is left for further studies. However, it should be positive in any case, because there exists a positive correlation of lattice phase in a local sense. Also it is expected that β is less than unity because the correlation among kink pairs must be weaker than the correlation between the kink points themselves. As shown in Appendix A, the sets of correlation volumes (10) satisfy the general requirement (A.3). Therefore, they are regarded as approximate but acceptable forms of $V^{(n)}$.

We shall now introduce the concept of clusters in terms of the intrinsic correlation functions $\{f_n\}$.

(1) Cluster of kink pairs (CKP): This is a group of KP's which are associated with an intrinsic correlation function. A single KP may belong to many different clusters unless the KP concerned is well separated from the others.

(2) Isolated kink cluster (IKC): This is a special cluster which includes an excess kink point of types (a) or (b) at the tail of the cluster.

In the previous theory, the clusters associated with $\{f_n\}$ were neglected, except for f_1. The present problem is to take into account the neglected term in expression (7).

IV. Formulae for the intensities

In the present formulation, the routes R and R' are composed of a series of vertical and horizontal segments and the tails of the segments of R and R' form a correlated pair (CP) of IKC's. The expressions (1a and b), then, can be rewritten as

$$\begin{aligned}\langle I_o\rangle = (A/a)^2 \sum_R \sum_{R'} (-\kappa_g\,\kappa_{-g})^k\,(-\kappa_g^*\,\kappa_{-g}^*)^{k'}\,(a)^{2k+2k'} \\ \times \langle\exp(i[P_{\text{VS}}])\rangle\,\langle\exp(-i[P'_{\text{VS}}])\rangle \\ \times \langle\exp(i[P_{\text{IKC}}-P'_{\text{IKC}}]_a)\rangle\cdots\langle\exp(i[P_{\text{HS}}])\rangle \\ \times \langle\exp(-i[P'_{\text{HS}}])\rangle\,\langle\exp(i[P_{\text{IKC}}-P'_{\text{IKC}}]_b\rangle \\ \times \langle\exp(i[P_{\text{VS}}])\rangle\,\langle\exp(-i[P'_{\text{VS}}])\rangle,\end{aligned} \tag{11a}$$

$$\begin{aligned}\langle I_g\rangle = (A/a)^2|\kappa_g|^2 \sum_R \sum_{R'} (-\kappa_g\,\kappa_{-g})^k\,(-\kappa_g^*\,\kappa_{-g}^*)^{k'} \\ \times (a)^{2k+2k'+2}\,\langle\exp(i[P_{\text{VS}}])\rangle\,\langle\exp(-i[P'_{\text{VS}}])\rangle \\ \times \langle\exp(i[P_{\text{IKC}}-P'_{\text{IKC}}]_a)\rangle\ldots\langle\exp(i[P_{\text{VS}}])\rangle \\ \times \langle\exp(-i[P'_{\text{VS}}])\rangle\,\langle\exp(i[P_{\text{IKC}}-P'_{\text{IKC}}]_b)\rangle \\ \times \langle\exp(i[P_{\text{HS}}])\rangle\,\langle\exp(-i[P'_{\text{HS}}])\rangle.\end{aligned} \tag{11b}$$

In these formulae, $P_{\text{VS}}\,(P_{\text{HS}})$ denotes the total phase of a vertical (horizontal) segment and $[P_{\text{IKC}} - P'_{\text{IKC}}]_{a(b)}$ is the phase associated with a correlated pair of IKC's. The subscripts a and b specify the type of IKC.

The ensemble average $\langle\exp(i[P_{\text{VS}}])\rangle$ is nothing more than the correlation function given by equation (7). In the case of P_{HS}, the same expression can be used provided that $\{x_i\}$ and $\{y_i\}$ are interchanged.

The ensemble average for the correlated pair of IKC of type (a) can be written in the form

$$\begin{aligned}&\langle\exp(i[P_{\text{IKC}}-P'_{\text{IKC}}]_a)\rangle \\ &\quad = F_{m+m'+1}(v_1,u_1,\ldots,u_m;\,v,\bar{u};\,u'_{m'},v'_{m'},\ldots,u'_1,v'_1), && (12a) \\ &\quad \simeq f_{h+1}[v_1,u_1,\ldots,u_m,v,u'_{m'},\ldots,u'_1,v'_1)\,f(\bar{u}), && (12b)\end{aligned}$$

where $\bar{u}$ and v are the vertical and horizontal distances between the last (excess) kinks of the routes R and R', respectively, and F_{h+1} ($h = m + m'$) is the intrinsic correlation function of the total kinks included in a CP. For mathematical convenience, the factoring of $f(\bar{u})$ is approximated in equation (12b). With this approximation, the pair of excess kinks can be regarded as a kink pair, so that the remaining correlation function must be identical to f_{h+1}. It is worth noting that the intrinsic correlation function must be used here. In fact, $\langle\exp(i[P_{\text{IKC}} - P'_{\text{IKC}}])\rangle$ is regarded as a factor of the correlation function of the total KP's of the vertical segments of R and R' plus the correlated pair of IKC.

If the initial arms of (IKC) and (IKC)′ are separated by $\bar{v}$,

$$v = \bar{v} + \sum v_r - \sum v'_r. \tag{13}$$

Then, we shall use the following approximation:

$$f_{h+1}(\ldots,v,\ldots) = f_{h+1}(\ldots,\bar{v},\ldots)\prod^{m} f(v_r)\prod^{m'} f(v'_r). \tag{14}$$

The justification in the special case of $f_1(v)$ is discussed in Appendix B of (II). The justification of (14) is merely a matter of formal extension.

We shall now combine the ensemble average of the phases for the vertical segments of R and R' and the CP of IKC. Then, if the initial arms of the vertical segments of R and R' are separated by $\bar{y}$,

$$\bar{v} = \bar{y} + \sum y_r - \sum y'_r. \tag{15}$$

With the approximation used to obtain equations (14) and (12b), one obtains

$$\begin{aligned}&\langle\exp(i[P_{\text{VS}}])\rangle\,\langle\exp(-i[P'_{\text{VS}}])\rangle\langle\exp(i[P_{\text{IKC}}-P'_{\text{IKC}}]_a)\rangle \\ &\quad = f(y_1,x_1,\ldots,y_n)\,f(y'_1,x'_1,\ldots,y'_{n'}) \\ &\qquad \times f_{h+1}(v_1,u_1,\ldots,u_m,\bar{y},u'_{m'},\ldots,v'_1) \\ &\qquad \times [f(\bar{y})]\prod^{n} f(y_r)\prod^{n'} f(y'_r) \\ &\qquad \times \prod^{m} f(v_r)\prod^{m'} f(v'_r)\,f(\bar{x}).\end{aligned} \tag{16}$$

In this expression $\bar{u}$ has been replaced by $\bar{x}$ and the factor $[f(\bar{y})]$ is written explicitly. This factor comes from a calculation similar to that which produced factor $f(\bar{x})$ in equation (16) by application to the preceding horizontal segment.

The next problem is to sum (16) over all possible routes after multiplying by the relevant factors of $(a^2 \kappa_g \kappa_{-g})$ and $(a^2 \kappa_g^* \kappa_{-g}^*)$. Symbolically, one can write the sum as

$$\sum_R \sum_{R'} = \sum_{\text{IKC}} \sum_{\text{CP}} \sum_{\text{CKP}} \sum_{\text{CKP}}{}'. \tag{17}$$

First, we shall consider the operation $\sum_{\text{CKP}}$. The relevant factor to be summed is

$$f(y_1, x_1, \ldots, y_n) \prod{}^n f(y_r)\, (a^2 \kappa_g \kappa_{-g})^n.$$

As shown in equation (7), the correlation function f is the sum of many terms, each of which is a possible product of $f_1, f_2, \ldots$ and f_n or a series of CKP's.

A single term is specified by the numbers $\{p_1, p_2 \ldots\}$ of the intrinsic correlation function $\{f_1, f_2 \ldots\}$, and the total number $p = \sum p_j$. Fixing the series of CKP, we shall first sum (or integrate) over all variables $\{y_r/a\}$ and $\{x_r/a\}$. Then, the product of $\{f_j\}$ changes to the product of the correlation volumes $[V_2^{(1)}]^{p_1} [V_2^{(2)}]^{p_2} \ldots \times (1/a)^{2n-p}$. The terms which give the same product should appear $(p!/p_1! p_2! \ldots)$ times. Here, it is to be noted that $(p-1)$ variables of $\{x_r\}$ are missing in the expression of a p product of $\{f_j\}$. Adding a variable x_o, which indicates the position of the first CKP of the segment, one notices that p variables are hidden. This implies that the first kink position of each CKP is arbitrarily located on the vertical segment of the length l_o, although the order of sequence has to be fixed. Thus, the number of possible configurations of p clusters is $(l_o/a)^p/p!$

According to the above arguments, we have

$$\sum_{\text{CKP}} = \sum_{n=0}^{\infty} \sum_{p_j=0}^{n} \frac{(l_o)^{p_1+p_2 \ldots}}{p_1! p_2! \ldots} [(-\kappa_g \kappa_{-g})\, V_2^{(1)}]^{p_1}$$

$$\times [(-\kappa_g \kappa_{-g})^2\, V_2^{(2)}]^{p_2} \ldots \tag{18a}$$

$$= \exp\left[\sum_{j=1}^{\infty} (-\kappa_g \kappa_{-g})^j\, V_2^{(j)}\, l_o \right]. \tag{18b}$$

Here, the summation $\sum_{p_j}$ must be taken with the condition that the total number of kink pairs is

$$n = \sum j \times p_j. \tag{19}$$

The mathematical technique of deriving (18*b*) is given in Appendix *B*. Equation (18*b*) can be rewritten in the form

$$\sum_{\text{CKP}} = \exp[-\tau_2(\kappa_g \kappa_{-g})\, R_o\, l_o], \tag{20a}$$

where

$$R_o = 1 + (\tau_2)^{-1} \sum_{j=1}^{\infty} V_2^{(j+1)}\, (z)^j, \tag{21}$$

and z stands for $(-\kappa_g \kappa_{-g})$. In these expressions, the relation (9*b*) is employed.

Similarly, one can obtain

$$\sum_{\text{CKP}}{}' = \exp[-\tau_2(\kappa_g^* \kappa_{-g}^*)\, R_o^*\, l_o] \tag{20b}$$

for the conjugate wave. Similar expressions can also be obtained for the horizontal segment. There, the length of the segment l_o has to be replaced by l_g.

Next, we shall proceed to the operation $\sum_{\text{CP}}$. Since the correlated pair of type (*a*) is discussed here in detail, it is denoted by $\sum_{\text{CP}}^{(a)}$. The relevant factors to be summed are

$$|\kappa_g|^2\, a^2\, f(\bar{y})\, f_{h+1}(v_1, u_1, \ldots, u_m; \bar{y}; u'_{m'}, \ldots, v'_1)$$

$$\times \prod^{m} f(v_j) \prod^{m'} f(v'_j)\, (-\kappa_g \kappa_{-g} a^2)^m\, (-\kappa_g^* \kappa_{-g}^* a^2)^{m'},$$

where $h = m + m'$. Here, consider $S(m,m')$, which is part of $\sum_{\text{CP}}^{(a)}$ for a fixed pair of (m,m'), namely a single CP. $S(m,m')$, therefore, means the sum (or integration) over the variables $\{v_r/a\}$, $\{u_r/a\}$ and $(\bar{y}/a)$.† There is no hidden variable. One can also notice that the correlation volume $V_2^{(h+1)}$ corresponding to f_{h+1} is independent of the individual numbers of m and m'. Thus, one obtains

$$S(m,m') = 2|\kappa_g|^2\, a(-\kappa_g \kappa_{-g})^m (-\kappa_g^* \kappa_{-g}^*)^{m'}\, V_2^{(h+1)}. \tag{22}$$

Next, we shall sum $S(m,m')$ over the whole CP's. In other words we shall sum over (m,m') or (h,m), in which m' is replaced by $h - m$. As shown in Fig. 1, each CP is associated with a single horizontal arm of the route R. Since, as will be seen later, we are interested in the amplitude factor of CP per single horizontal arm, the factor due to the $(h+1)$ cluster must be:

$$2|\kappa_g|^2\, a(h+1)^{-1}\, V_2^{(h+1)}\, \{z^h + z^{h-1} z^* +, \ldots, (z^*)^{h+1}\}.$$

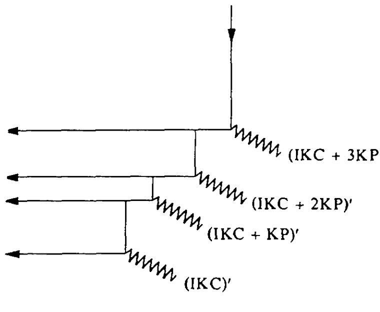

Fig. 1. The possible correlated pairs of IKC associated with a vertical segment R. $(\ldots)'$ indicates the cluster of the conjugate waves (for $h = 3$).

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† The variable $\bar{y}$ takes positive and negative values. Also, notice that $f_{h+1}(\ldots, \bar{y}, \ldots)$ is an even function.

Thus, the total amplitude factor for the possible CP's is

$$\sum_{\mathrm{CP}}{}^{(a)} = 2\tau_2 |\kappa_g|^2 aR_g, \tag{23}$$

where

$$\begin{aligned} R_g &= (\tau_2)^{-1} \sum_{h=0}^{\infty} (h+1)^{-1} V_2^{(h+1)} \\ &\quad \times \{z^h + z^{h-1} z^* + \ldots + (z^*)^h\} \\ &= (\tau_2)^{-1} \sum_{h=0}^{\infty} V_2^{(h+1)} \frac{(z)^{h+1} - (z^*)^{h+1}}{(z - z^*)(h+1)}. \end{aligned} \tag{24a}$$

It is worth noting that R_g is a real quantity and

$$R_g = R_o \quad (\text{when } z = z^*). \tag{24b}$$

Also, for CP's of type (*b*), we shall have the similar expression

$$\sum_{\mathrm{CP}}{}^{(b)} = 2\tau_2 |\kappa_{-g}|^2 aR_g. \tag{25}$$

The final step of the calculation is to perform the operation $\sum_{\mathrm{IKC}}$. What we need to do is to take the product of $S = \sum_{\mathrm{CP}} \sum_{\mathrm{CKP}} \sum_{\mathrm{CKP'}}$ for every vertical and horizontal segment and to sum over all possible configurations of the segments. The procedures are exactly the same as those described in §§ 3 and 4 of (II). The only difference is that the extra coefficients R_o and R_g are included in the expressions of (20*a*), (20*b*), (23) and (25). Thus, we obtain the intensity fields as follows:

$$\begin{aligned} \langle I_o \rangle &= |A|^2 \exp -2\tau_2 \operatorname{Re}\{(\kappa_g \kappa_{-g}) R_o\}(s_o + s_g) \\ &\quad \times |\kappa_g \kappa_{-g}| (s_o/s_g)^{1/2} I_1[4\tau_2 |\kappa_g \kappa_{-g}| R_g (s_o s_g)^{1/2}], \end{aligned} \tag{26a}$$

$$\begin{aligned} \langle I_g \rangle &= |A|^2 \exp -2\tau_2 \operatorname{Re}\{(\kappa_g \kappa_{-g}) R_o\}(s_o + s_g) \\ &\quad \times |\kappa_g|^2 I_0[4\tau_2 |\kappa_g \kappa_{-g}| R_g (s_o s_g)^{1/2}], \end{aligned} \tag{26b}$$

where I_0 and I_1 are the modified Bessel functions of the zeroth and the first order respectively. As explained in (II), the present solution satisfies the energy-transfer equation of the form

$$\frac{\partial\langle I_o\rangle}{\partial s_o} = -2\tau_2 \operatorname{Re}\{(\kappa_g \kappa_{-g}) R_o\} \langle I_o \rangle + 2\tau_2 |\kappa_{-g}|^2 R_g \langle I_g \rangle, \tag{27a}$$

$$\frac{\partial\langle I_g\rangle}{\partial s_g} = -2\tau_2 \operatorname{Re}\{(\kappa_g \kappa_{-g}) R_o\} \langle I_g \rangle + 2\tau_2 |\kappa_g|^2 R_g \langle I_o \rangle. \tag{27b}$$

The attenuation and diffraction powers (coupling constants) are reduced by the factors R_o and R_g, respectively, compared with the previous results.

V. Remarks and discussion

(*a*) *The reduction factors R_o and R_g*

The essence of the present results lies in the introduction of the reduction factors R_o and R_g [*cf.* equations (21) and (24*a*)]. If the power series can be terminated because of rapid decrease of the correlation volumes with their order, one can base a calculation of the factors, in principle, on the model of lattice distortion.

Here, however, we shall discuss the physical meanings of R_o and R_g based on an approximate but acceptable model of the correlation volumes $V_2^{(j)}$ which are given in equation (10*b*). We then obtain the expressions

$$R_o = 1 - \beta_2 \tau_2^2 z + \beta_2^2 \tau_2^4 z^2 + \ldots = (1 + \beta_2 \tau_2^2 \kappa_g \kappa_{-g})^{-1}, \tag{28a}$$

$$\begin{aligned} R_g &= 1 - \tfrac{1}{2}\beta_2 \tau_2^2 (z + z^*) + \tfrac{1}{3}\beta_2^2 \tau_2^4 (z^2 + zz^* + z^{*2}) + \ldots \\ &= [\beta_2 \tau_2^2 (\kappa_g \kappa_{-g} - \kappa_g^* \kappa_{-g}^*)]^{-1} \\ &\quad \times \log[(1 + \beta_2 \tau_2^2 \kappa_g \kappa_{-g})/(1 + \beta_2 \tau_2^2 \kappa_g^* k_{-g}^*). \end{aligned} \tag{28b}$$

Expressions (28*a*,*b*) are valid only when the series satisfy the condition $|\beta_2 \tau_2^2 \kappa_g \kappa_{-g}| < 1$, which leads to

$$\tau_2 < \Lambda/(\beta_2)^{1/2}. \tag{29}$$

With this result, the coupling constants can be reduced to half those given in the simpler theory described in (II).

Expressions (28*a*,*b*) are physically understandable, because the reduction of the coupling constants increases with increases of diffraction power $(\kappa_g \kappa_{-g})$ and correlation lengths τ_2 and $\tau_2\beta_2$, which are essentially measures of crystal perfection. The conventional and intuitive treatments which assume ideally perfect crystals for a mosaic block can be partially justified in the sense that the factors R_o and R_g are expressed by the power series of $(-\kappa_g \kappa_{-g})$. However, the wavelength dependence and the angular dependence are hardly justified. In fact, the degree of the reduction must depend upon the type and the degree of crystal perfection, which are introduced through the correlation length (volume) in the present theory. This point escapes the theory of the conventional treatment.

(*b*) *Applicability*

The present formulation eliminates the most essential approximation of the previous theory: the assumption of statistical independence of KP's. By introducing higher-order correlations of the lattice phase factors, all optical routes are taken into account. For this reason, certainly, the present theory can be applied to a wider range of crystal perfection than can the previous one.

The theory still assumes the factorization of the correlation functions [*cf.* equations (12) and (14)]. According to arguments similar to those described in Appendix B of (II) for $f_1(y)$, the factorization of $\{f_n\}$ can also be justified, provided that the $\{f_n\}$ decrease rapidly with increase of any one of their arguments.

In fact, the present formalism assumes integrability of $\{f_n\}$. This assumption is not always acceptable. For example, if the thermally vibrating crystals have an averaged perfect lattice, $f_1(y)$ must have the form

$$f_1(y) = \exp(-2M_s) + g(y), \tag{30}$$

where the first term is the Debye–Waller factor in the kinematical theory and $g(y)$ implies the correlation term. In this case, one cannot define a finite correlation length for $f_1(y)$. The present formalism cannot be applied to this case.

Even in the case when $V_2^{(n)}$ can be defined, the theory would be meaningless if the power series of the reduction factors were to diverge. For this reason, it seems reasonable to take the convergence criterion as a measure of the applicability of the present theory. In the particular case of $V_2^{(n)}$ given by equations (10*a*,*b*), the criterion is given by equation (29). It is conjectured, therefore, that the present theory is applicable when

$$\tau_2 \lesssim \Lambda. \tag{31}$$

(*c*) *Effects of absorption*

In non-absorbing crystals, where $\kappa_g \kappa_{-g} = |\kappa_g|^2 = |\kappa_{-g}|^2$ and consequently $R_o = R_g$, the energy conservation stated by

$$\frac{\partial\langle I_o\rangle}{\partial s_o} + \frac{\partial\langle I_g\rangle}{\partial s_g} = 0 \tag{32}$$

is automatically satisfied. In absorbing crystals, the relations for $\kappa_g \kappa_{-g}$ *etc.* must be modified, because of anomalous dispersion, as follows (see, for example, Zachariasen, 1945; Azaroff, 1974):

$$\kappa_g \kappa_{-g} = |\kappa_g^r|^2 - |\kappa_g^i|^2 + 2i|\kappa_g^r||\kappa_g^i| \cos(\varphi_1 - \varphi_2), \tag{33a}$$

$$|\kappa_g|^2 = |\kappa_g^r|^2 + |\kappa_g^i|^2 + 2|\kappa_g^r||\kappa_g^i| \sin(\varphi_1 - \varphi_2), \tag{33b}$$

$$|\kappa_{-g}|^2 = |\kappa_g^r|^2 + |\kappa_g^i|^2 - 2|\kappa_g^r||\kappa_g^i| \sin(\varphi_1 - \varphi_2), \tag{33c}$$

where κ_g^r and κ_g^i are the Fourier coefficients of κ^r and κ^i, respectively, in the phenomenological expression of the polarizability $\kappa = \kappa^r + i\kappa^i$, and φ_1 and φ_2 are the phase angles of κ_g^r and κ_g^i respectively. In centrosymmetric crystals, φ_1 is identical to φ_2. Moreover, in the cases where either $|\kappa_g^r| \gg |\kappa_g^i|$ or the reduction factors are close to 1, we can expect that $R_o = R_g$ (real, positive). To avoid complexity, we have discussed only these cases here.

One then obtains

$$\frac{\partial\langle I_o\rangle}{\partial s_o} + \frac{\partial\langle I_g\rangle}{\partial s_g} = E\{\langle I_o\rangle + \langle I_g\rangle\}, \tag{34}$$

and the enhancement coefficient E is given by

$$E = 4\tau_2 R_o |\kappa_g^i|^2. \tag{35}$$

The positiveness of E does not violate energy conservation, because the normal absorption coefficient $\mu_o = 2\kappa_o^i$ exceeds the enhancement coefficient. (Note that $|\kappa_o^i| > |\kappa_g^i|$ and $1 \geq 2\tau_2|\kappa_g \kappa_{-g}|^{1/2} \gg 2\tau_2 R_o |\kappa_g^i|$.)

The presence of a non-zero E implies anomalous transmission, in the case of secondary extinction. The physical meaning of this anomalous transmission is interpreted as Borrmann anomalous transmission in a perfect-crystal range of length τ_2. In the present treatment, however, the effect may have no significant role since $\mu_o \gg E$. In the very special cases such as $|\kappa_g^r| \simeq |\kappa_g^i|$, the anomalous transmission becomes significant. However, we need detailed mathematical analysis for the case of a complex reduction factor R_o.

In conclusion, the present theory derived the coupling constants of the energy-transfer equation from first principles of wave equations. It is highly desirable to check the theory experimentally in order to elucidate the nature of secondary extinction.

APPENDIX *A*

Correlation function and correlation volume

In § III, the correlation function $f(\mathbf{n})$ and the intrinsic correlation function $f_n(\mathbf{n})$ are introduced. Here, the abbreviated notation $\mathbf{n}$ is used for the arguments $(y_1, x_1, \ldots, x_{n-1}, y_n)$.

For an intrinsic correlation function, we have defined the correlation volume by equations (8*a*,*b*). Similarly, one can define the correlation volume for $f(\mathbf{n})$ as follows:

$$B_1^{(n)} = \int_0^\infty \cdots \int_0^\infty \mathrm{d}\{y_r\} \int_0^\Delta \cdots \int_0^\Delta \mathrm{d}\{x_r\}\, f(\mathbf{n}), \tag{A.1a}$$

$$B_2^{(n)} = \int_0^\infty \cdots \int_0^\infty \mathrm{d}\{y_r\} \int_0^\Delta \cdots \int_0^\Delta \mathrm{d}\{x_r\} \prod_{r=1}^{n} f(y_r)\, f(\mathbf{n}), \tag{A.1b}$$

where the integration limits Δ are introduced to avoid divergence. Here, Δ is a length which is sufficiently larger than the correlation length τ. In the following, we need not distinguish cases 1 and 2, so that the suffixes are omitted.

First, we obtain a relation between $\{f(\mathbf{n})\}$ and $\{f_n(\mathbf{n})\}$. As shown in equation (7), the last factor of every term of $f(\mathbf{n})$ in its decomposed form (right-hand side) must be one of $f_j(\mathbf{j})$. Thus, $f(\mathbf{n})$ can be expanded in terms of $f_j(\mathbf{j})$ and the coefficients must be $f(\mathbf{n}-\mathbf{j})$. Thus, one can see that

$$f(\mathbf{n}) = \sum_{j=1}^{n-1} f(\mathbf{n}-\mathbf{j})\, f_j(\mathbf{j}) + f_n(\mathbf{n}). \qquad (A.2)$$

In this expression, the sequence of KP's in $(\mathbf{n}-\mathbf{j})$ and $(\mathbf{j})$ must be kept identical to the original one of $(\mathbf{n})$.

Next, we integrate $(A.2)$ over the range $(0,\Delta)$ with respect to $\{x_r\}$ and over the range $(0,\infty)$ with respect to $\{y_r\}$. Since $\Delta > \tau$, the range $(0,\Delta)$ is equivalent to $(0,\infty)$ in the case of $V^{(j)}$. Then, we have the relation

$$B^{(n)} = \sum_{j=1}^{n-1} B^{(n-j)}\, \Delta V^{(j)} + V^{(n)}, \qquad (A.3)$$

where Δ appears due to the integration of a hidden variable x_{n-j}.

In the case of $B^{(n)}$, if we approximately neglect the end effects of a sequence of KP's, the integrations of $\{x_r\}$ and of $\{y_r\}$ are equivalent in the respective cases so that we expect the form of $B^{(n)}$ to be

$$B^{(n)} = \tau^n(\Delta + \beta\tau)^{n-1} \quad (n \geq 1). \qquad (A.4)$$

Here, the factor τ^n is due to the integrations of $\{y_r\}$ and $(\Delta + \beta\tau)^{n-1}$ is due to the integrations of $\{x_r\}$.

Substituting from $(A.4)$ into $(A.3)$, one can easily see that:

$$V^{(1)} = \tau, \quad V^{(2)} = \beta\tau^3, \quad V^{(3)} = \beta^2\tau^5 \ldots$$

By the use of mathematical reduction, we have

$$V^{(n)} = \beta^{n-1}\,\tau^{2n-1}. \qquad (A.5)$$

With the approximation stated in connection with $(A.4)$ for the correlation volume $B^{(n)}$, the correlation volume $V^{(n)}$ must have this form.

APPENDIX *B*

Evaluation of the multiple sum (18)

Here, we consider the multiple sum

$$S = \sum_{n=0}^{\infty} \sum_{p_j=0} \frac{1}{p_1!\,p_2!\ldots} [Q^{(1)}]^{p_1} [Q^{(2)}]^{p_2} \ldots, \qquad (B.1)$$

with the condition

$$n = \sum_j j \times p_j. \qquad (B.2)$$

In equation $(B.1)$, the notation

$$Q^{(j)} = V_2^{(j)}(-\kappa_g\,\kappa_{-g})^j\, l_0 \qquad (B.3)$$

is employed.

The method used here is well known as the method of Fowler and Guggenheim and the manner of its application is very similar to that in the theory of imperfect gas (Born & Fuchs, 1938). The summation $\sum_{p_j}$ in equation $(B.1)$ is equivalent to that to pick up the coefficient of ζ^n in the function of a complex variable ζ,

$$F(\zeta) = \sum_{p_j=0}^{\infty} \frac{1}{p_1!\,p_2!\ldots} [Q^{(1)}\,\zeta]^{p_1} [Q^{(2)}\,\zeta^2]^{p_2} \ldots, \qquad (B.4)$$

without any condition on $\{p_j\}$. This operation can be performed by the method of contour integral, and we obtain

$$S = \frac{1}{2\pi i} \oint \sum_{n=0}^{\infty} F(\zeta)\,(1/\zeta^{n+1})\,\mathrm{d}\zeta, \qquad (B.5)$$

where $\oint \mathrm{d}\zeta$ implies a closed-path integral around the origin.

The summation in equation $(B.4)$ can be performed easily as

$$F(\zeta) = \exp \sum_j Q^{(j)}\,\zeta^j. \qquad (B.6)$$

The summation in equation $(B.6)$ is given, provided that $|\zeta| > 1$, as

$$\sum_n (1/\zeta^{n+1}) = \frac{1}{\zeta - 1}. \qquad (B.7)$$

Since $F(\zeta)$ is regular, we have S as the residue of $F(\zeta)$ at $\zeta = 1$, which is only a pole within the integral path. Thus, we obtain

$$S = \exp \sum_j Q^{(j)}\,\zeta^j. \qquad (B.8)$$

This result leads to equation (18*b*).

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