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A Foundation for the Statistical Dynamical Theory of Diffraction

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Abstract

The statistical dynamical theory [Kato (1980). *Acta Cryst.* A36, 763-769] is reformulated on a sounder basis. The starting wave equation is free from the so-called Takagi-Taupin (T-T) approximation. Functional calculus, an operational technique and the concept of the Green function are used as mathematical tools. Integro-differential equations are derived for the coherent (averaged) wave field and the energy flow vector of the incoherent intensity field. The formulae are exact except for assuming a model in which the fluctuation of the lattice phase is a set of Gaussian random variables defined in three-dimensional space. The general framework of the previous theory is justified within the T-T approximation. In general, however, new terms must be added and some terms have to be revised by introducing a Green function matrix. The theory may be used as a starting point when any approximate theory is developed for practical purposes.

1. Introduction

The present author published a series of papers in order to unify primary and secondary extinction on the theoretical basis of optical coherence (Kato, 1976*a*, *b*, 1979, 1980*a*). Later, the theory was reformulated with a wider scope of diffraction such as the statistical characterization of crystals including invisible and/or indistinguishable lattice defects (Kato, 1980*b*, *c*). The theory, at present, is called the statistical dynamical theory.

In these studies, it was pointed out that the statistical nature of crystals is conveniently characterized in terms of the average of the lattice phase factor and its multiple correlation functions at different spatial positions. The observable intensity was postulated to be an ensemble average of possible intensities expected from the wave equation. Then it was shown that the observable intensity consists of coherent and incoherent parts. They can be represented in a formal manner by the statistical quantities of the wave field; namely the direct average of the field and the correlation function of the mutually complex conjugate fields. Thus, the problem was reduced to deriving the statistical quantities of the wave field from those of the lattice phase factor.

The attempt was partially successful but not always satisfactory. Moreover, a few intuitive arguments and even dubious treatments were introduced for developing an approximate theory. Some of them have been criticized by a few authors (Al-Haddad & Becker, 1988; Guigay, 1989).

In these circumstances, the present paper intends to reformulate the theory on a sounder basis. In the formulation, functional calculus, an operational technique and the concept of the Green function (propagator) are used. In fact, such methods have been applied to wave propagation in turbulent ionosphere and acoustic problems (*e.g.* Beran, 1968). In these problems, however, one-component (plane-wave-like) waves are dealt with and the statistical variable characterizing the medium is the refractive index. In the present work, we are concerned with two-component (Bloch-wave-like) waves and the statistical variable is the lattice phase.

In § 2, a brief review will be presented on mathematical subjects.* In § 3, starting from a wave equation which is free from the so-called Takagi-Taupin approximation, the exact equation for the characteristic functional (CFAL) of the wave field is derived. In § 4, the integro-differential equations are derived for both the averaged (coherent) wave field and the incoherent intensity field. Here, no approximation is made except that the model of a Gaussian random variable is assumed for the lattice phase at a certain stage of the theory. In the final section, the previous theories (Kato, 1980*b*) are discussed in the light of the present formalism.

2. The mathematical background

2.1. *The case of one random variable*

First, we shall show some elementary results. When a random variable (RV) q is specified through the probability density $\rho(q)$, the characteristic function (CF) is defined by

$$Z(p) = \langle \exp(ipq) \rangle \qquad (2.1a)$$

with the auxiliary condition

$$Z(0) = 1. \qquad (2.1b)$$

* The present author owes a great deal to the book of Furutsu (1982). Some details are described in the book of Beran (1968) and the papers of Furutsu (1972, 1975).

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Here, $\langle\ \rangle$ implies the average operation for the expression included in it and p is the conjugate variable of q. In other words, $Z(p)$ is the Fourier transform of ρ. The condition (2.1b) is equivalent to unit normalization of ρ.

Once the CF is obtained, one can calculate the nth moment from

$$\langle q^n\rangle = [(\partial/i\partial p)^n]Z(p)|_{p=0}. \tag{2.2}$$

The notation $[|_{p=0}]$ implies taking the limit at $p=0$ after the manipulation on the left-hand side. As a consequence of the relation (2.2), one can obtain the average of any analytic function $f(q)$ as

$$\langle f(q)\rangle = f[(\partial/i\partial p)]Z(p)|_{p=0}. \tag{2.3}$$

Furthermore, when $f(q)$ has the Fourier transform $\bar{f}(p)$,

$$\langle f(q)\rangle = (2\pi)^{-1}\int \bar{f}(p)Z(p)\,dp. \tag{2.4}$$

Next, some results of an operational technique are explained. Incidentally, the technique is very powerful when the probability density ρ is not known explicitly. Our problem is one such case. An operator can be defined from any analytic function of p by the rule

$$ip \to \partial/\partial c \tag{2.5}$$

where c is a dummy variable associated with q. Then we define an operator $\hat{Z}$ from $Z(p)$.* It has the property

$$\begin{aligned}\hat{Z}f(c) &= \langle \exp[(\partial/\partial c)q]f(c)\rangle \\ &= \left\langle \sum_{n=0}^{\infty}(1/n!)(q)^n(\partial/\partial c)^n f(c)\right\rangle \\ &= \langle f(q+c)\rangle.\end{aligned} \tag{2.6}$$

Then, we have

$$\langle f(q)\rangle = \hat{Z}f(c)|_{c=0}. \tag{2.7}$$

For later purposes, we also introduce the operator

$$\hat{q} = \hat{Z}c\hat{Z}^{-1}. \tag{2.8}$$

In symmetrical manner, the operator $\hat{p}$ can also be defined by

$$\hat{p} = \hat{Z}(\partial/i\partial c)\hat{Z}^{-1} \tag{2.9}$$

because $\hat{Z}$ and $\partial/\partial c$ must be commutative. We define also $\hat{\Theta}$ and $\hat{\Theta}_1$ by the rule (2.5) or (2.9) through the cumulant function and its derivative:

$$\Theta(p) = \log Z(p) \tag{2.10a}$$

$$\begin{aligned}\Theta_1(p) &= (\partial/i\partial p)\Theta(p) \\ &= (\partial Z/i\partial p)/Z(p).\end{aligned} \tag{2.10b}$$

* Henceforth, operators are denoted by ^.

According to the operator algebra familiar in quantum statistical mechanics (*e.g.* Louisell, 1973), we have

$$f(\hat{q}) = \hat{Z}f(c)\hat{Z}^{-1} \tag{2.11a}$$

$$\hat{q} = c + \Theta_1(\partial/i\partial c). \tag{2.11b}$$

By combining (2.6) and (2.11a), it follows that $\langle f(q+c)\rangle = f(\hat{q})\hat{Z}$. As a result, we have the useful relation

$$\langle (q+c)f(q+c)\rangle = \hat{q}f(\hat{q})\hat{Z} = \hat{q}\langle f(q+c)\rangle \tag{2.12}$$

which gives $\langle qf(q)\rangle$ when $\langle f(q)\rangle$ is known in the limit of $c=0$.

When $\Theta(p)$ has a quadratic form with respect to p and $\langle q\rangle = 0$, namely

$$\Theta(p) = -(1/2)\langle q^2\rangle p^2, \tag{2.13}$$

we have

$$\hat{q} = c + \langle q^2\rangle(\partial/\partial c). \tag{2.14}$$

In fact, it is the case that $\rho(q)$ [also $Z(p)$] has the Gaussian form. In the operator formalism, (2.14) is used as the definition of a Gaussian random variable (GRV).

2.2. *The case of N discrete RV's*

It is a formal matter to define the CF by

$$Z\{p_j\} = \left\langle \exp\left(i\sum_{j=1}^{N} p_j q_j\right)\right\rangle \tag{2.15a}$$

$$Z\{0\} = 1. \tag{2.15b}$$

Corresponding to (2.2), the $\{n_j\}$th moment can be given in the form

$$\mu\{n_j\} = \prod_{j=1}^{N}(\partial/i\partial p_j)^{n_j} Z\{p_j\}|_{\{p_j\}=0}. \tag{2.16}$$

We can also write down similar formulae corresponding to (2.3) and (2.4).

The operational technique can be developed in the same manner. The rule (2.5) is generalized as

$$ip_j \to \partial/\partial c_j \tag{2.17}$$

where $\{c_j\}$ are dummy variables associated with $\{q_j\}$. The operator corresponding to the CF has the form

$$\hat{Z} = Z\{\partial/i\partial c_j\}. \tag{2.18}$$

We introduce functions similar to (2.10a) and (2.10b),

$$\Theta\{p_j\} = \log Z\{p_j\} \tag{2.19a}$$

$$\Theta_r\{p_j\} = (\partial/i\partial p_r)\Theta\{p_j\}. \tag{2.19b}$$

Through these functions, the important operators $\{\hat{q}_r\}$ are defined as

$$\hat{q}_r = \hat{Z}c_r\hat{Z}^{-1} \tag{2.20a}$$

$$= c_r + \Theta_r\{\partial/i\partial c_j\}. \tag{2.20b}$$

Corresponding to (2.6) and (2.12), we have the relations

$$\hat{Z}f\{c_j\} = \langle f\{q_j + c_j\}\rangle \tag{2.21}$$

$$\langle (q_r + c_r) f\{q_j + c_j\}\rangle = \hat{q}_r f\{\hat{q}_j\}\hat{Z} = \hat{q}_r \langle f\{q_j + c_j\}\rangle. \tag{2.22}$$

If all of the $\{q_j\}$ are GRV and the averages of $\{\langle q_j\rangle\}$ are zero, the operator $\hat{q}_r$ has the explicit form

$$\hat{q}_r = c_r + \sum_{j=1}^{N} \langle q_r q_j\rangle (\partial/\partial c_j). \tag{2.23}$$

2.3. *The case of RV's defined on a continuous space*

In this subsection, for the sake of continuation of the argument, we shall consider RV's defined on one-dimensional space. Then, the continuous coordinate (s) corresponds to the index (j) in the previous case and the total RV's can be represented by a function $q(s)$.

It seems natural to write formally the following expression in the place of (2.15a).

$$Z[p] = \langle \exp[i \int p(s) q(s)\, ds]\rangle \tag{2.24}$$

where $p(s)$ is the conjugate variable of $q(s)$ specified at the same point s. The total of the variables can be regarded also as a function of s. However, we must be careful about the average operation.

For a set of discrete variables, the average of a function F is given by

$$\langle F\{q_j\}\rangle = \prod_{j=1}^{N} \int \rho\{q_j\} F\{q_j\}\, dq_j. \tag{2.25a}$$

Now, not only is N infinite, but the product index j is continuous so that we need the concept of functional calculus. According to the mathematical convention, we shall write the average as

$$\langle F\{q(s)\}\rangle = \int \mathcal{D}q[\rho\{q(s)\} F\{q(s)\}] \tag{2.25b}$$

where $\int \mathcal{D}q$ is called functional integration, which means the integration over all possible forms of $q(s)$. It is worth noting that the outcome after the manipulation is a number, as in the case of ordinary integration.

In (2.24), exp[ ] stands for the function $F[q(s)]$ to be averaged. It includes the function $p(s)$. Therefore, the outcome number depends explicitly upon the form of $p(s)$. For this reason, it is denoted by $Z[p]$ and called the 'functional of $p(s)$'.* For random variables defined on a continuous space, we must consider the characteristic functional (CFAL) in place of CF.

The next problem is to obtain a similar expression to (2.16). For this purpose, the functional derivative is introduced. It is the ratio of the functional variation $\delta Z[p]$ and an infinitesimal variation $\delta p(s)$ which results in δZ. For (2.24), it is

$$\delta Z[p]/\delta p(s) = i\langle q(s) \exp[i \int p(s') q(s')\, ds']\rangle. \tag{2.26}$$

It should be noted that the random variable $q(s)$ at the beginning of $\langle\ \rangle$ and the differential variable $p(s)$ on the left must be conjugate to each other and must be specified at the same point in s space.

We define the multiple correlation function (COF) by

$$M(s_1, s_2, \ldots, s_n) = \langle q(s_1) q(s_2) \ldots q(s_n)\rangle. \tag{2.27}$$

For the special case in which $s_1 = s_2 = \ldots = s_n$, $M(\ )$ is called the autocorrelation function. Averaged quantities with a mixed character are also conceivable. However, we shall not distinguish them, unless otherwise stated. Obviously, the COF is equivalent to the moment in the discrete case. Sometimes the notation M_n is used as an abbreviation. We can easily obtain the expression

$$M_n = (1/i)^n \frac{\delta^n Z[p]}{\delta p(s_1)\delta p(s_2) \ldots \delta p(s_n)}\bigg|_{p=0} \tag{2.28}$$

which constitutes the generalization of (2.16).

In the next stage, we must modify the formulae of the operational technique described in § 2.2. Since the modification is understandable as a matter of formality, here we shall present a glossary of equations.

The correspondence rule:

$$ip(s) \to \delta/\delta c(s) \qquad [2.17] \tag{2.29}$$

$$\hat{q}(s) = \hat{\mathbf{Z}} c(s) \hat{\mathbf{Z}}^{-1} \qquad [2.20a] \tag{2.30a}$$

$$= c(s) + \Theta_1[\delta/i\delta c;\, s] \qquad [2.20b] \tag{2.30b}$$

$$\Theta_1[p;\, s] = \delta(\log \mathbf{Z})/i\delta p(s) \qquad [2.19] \tag{2.31}$$

$$\langle (q(s) + c(s)) f[q + c]\rangle = \hat{q}(s)\langle f[q + c]\rangle. \qquad [2.22] \tag{2.32}$$

When $\hat{q}$ is a GRV operator,

$$\hat{q}(s) = c(s) + \int ds' \langle q(s) q(s')\rangle (\delta/\delta c(s')). \qquad [2.23] \tag{2.33}$$

The equation numbers in [ ] refer to the relevant equations in § 2.2. Bold letters are used for functionals when the argument (function) is suppressed. Henceforth, this convention will be used.

* 'Function' is defined by the mapping [a number → a number] whereas 'functional' implies the mapping [a function → a number]. It is the usual convention to use [ ] for denoting the original function, as has been done without notice in (2.24). A concise review of functional calculus is given in the book of Ryder (1985). More can be seen in the book of Beran (1968).

3. The wave equation and the related characteristic functionals

3.1. *The wave equation*

So far the mathematical subjects of RV's and the operational technique have been described. Now, we shall proceed to diffraction physics. We consider a wave field which obeys the coupled equations

$$(\Delta + k^2)d_o + \Phi M_{-g} d_g = 0 \qquad (3.1a)$$

$$(\Delta + k^2)d_g + \Phi^* M_g d_o = 0 \qquad (3.1b)$$

where k is the wave number in the crystal and $M_{\pm g}$ are proportional to the structure factor of the $\pm\bar{\mathbf{g}}$ reflection.† The wave field has two components $d_0(\mathbf{x})$ and $d_g(\mathbf{x})$ which are the transmitted and Bragg-reflected waves. They are treated as complex fields in three-dimensional space $\mathbf{x}$, Φ is called the lattice phase factor which will be explained later [see (3.5a)].

The equations (3.1) can be derived straightforwardly from the fundamental wave equation for distorted crystals (*e.g.* Kato, 1974) by equating separately the terms oscillating with approximate wave vectors $\bar{\mathbf{k}}_o$ and $\bar{\mathbf{k}}_g = \bar{\mathbf{k}}_o + 2\pi\bar{\mathbf{g}}(|\bar{\mathbf{k}}_o| = |\bar{\mathbf{k}}_g| = k)$. The procedure implies neglect of the *Umklappung* process (Kato, 1973). Also, in the term proportional to the polarizability of the crystal, an approximation is made to neglect the spatial variation of Φ and the amplitudes, a_o and a_g, as well as the polarization factor because the term concerned is itself very small.

Each component of the wave field can be written in the form

$$d_o(\mathbf{x}) = a_o(\mathbf{x}) \exp i(\bar{\mathbf{k}}_o \mathbf{x}) \qquad (3.2a)$$

$$d_g(\mathbf{x}) = a_g(\mathbf{x}) \exp i(\bar{\mathbf{k}}_g \mathbf{x}). \qquad (3.2b)$$

Then, in the case of the O wave, for example, it follows that

$$(\Delta + k^2)d_o(\mathbf{x}) = \{2i(\mathbf{k}_o \nabla)a_o + \Delta a_o\} \exp i(\mathbf{k}_o \mathbf{x}) \qquad (3.3)$$

where the differential operators ∇ and Δ are to be applied only to the amplitude $a_o(\mathbf{x})$. Therefore, (3.1) are equivalent to the equations

$$2ik\partial a_o/\partial s_o + \Delta a_o + \Phi M_{-g} a_g = 0 \qquad (3.4a)$$

$$2ik\partial a_g/\partial s_g + \Delta a_g + \Phi^* M_g a_o = 0 \qquad (3.4b)$$

where s_o and s_g are the coordinates along the directions $\mathbf{k}_o$ and $\mathbf{k}_g$ respectively. In this case, the phase factor $\exp[\pm 2\pi i(\bar{\mathbf{g}}\mathbf{x})]$ in $M_{\pm g}$ can be dropped.

Equation (3.4) has great merit because one need not bother with the phase factor of the carrier waves. Nevertheless, we shall proceed with (3.1) because their form is more familiar in applied mathematics. The equation of Takagi–Taupin (T–T) type is derived by neglecting Δa_o and Δa_g. The implication of retaining them in (3.4) and implicitly in (3.1) will be discussed further in § 5.

The wave equation includes the complex lattice phase factor defined by

$$\Phi(\mathbf{x}) = \exp[2\pi i(\bar{\mathbf{g}}\mathbf{u}(\mathbf{x}))] \qquad (3.5a)$$

where $\bar{\mathbf{g}}$ is the reflection vector of the standard perfect crystal and $\mathbf{u}(\mathbf{x})$ is the displacement vector of the lattice point. In general, $\langle\Phi\rangle$ is not zero, so that a new variable $Q(\mathbf{x})$ is introduced by

$$\Phi(\mathbf{x}) = E + Q(\mathbf{x}) \qquad (3.5b)$$

where E denotes $\langle\Phi\rangle$ and $\langle Q\rangle$ is zero. More will be mentioned on this point at the beginning of § 4. It can be mathematically proven that, if $(\bar{\mathbf{g}}\mathbf{u})$ is a RV, Q is also a complex RV.

For our purpose, it is more convenient to start with the slightly more general equations

$$(\Delta + k^2)d_o + (E + Q + C)M_{-g} d_g = J_o \qquad (3.6a)$$

$$(\Delta + k^2)d_g + (E + Q^* + C^*)M_g d_o = J_g. \qquad (3.6b)$$

As will be seen later, $C(\mathbf{x})$ is a dummy function introduced for the operator technique, and $J_o(\mathbf{x})$ and $J_g(\mathbf{x})$ are source functions of d_o and d_g at the present stage. Later, they will be assigned to the conjugate functions associated with d_o and d_g, respectively. After calculating any statistical quantity such as the correlation function of the wave fields, the physical value is given by taking the limit at $C = 0$ and $J_0 = J_g = 0$.

3.2. *The characteristic functionals*

First, the CFAL of $Q(\mathbf{x})$ and $Q^*(\mathbf{x})$ are considered. We need a generalization in two aspects. (i) Q and Q^* are functions of a point $\mathbf{x}$ in 3D space. Clearly, it is enough to replace s by $\mathbf{x}$ in the results listed at the end of § 2.3. (ii) There are two kinds of RV's $Q(\mathbf{x})$ and $Q^*(\mathbf{x})$. They can be dealt with separately by introducing the conjugate functions, $P(\mathbf{x})$ and $P^*(\mathbf{x})$, respectively. Alternatively, one may take the real and imaginary parts of Q as RV's. The final result, however, is not changed. Thus, an adequate CFAL for Q and Q^* has the form

$$Z[P^*, P] = \langle \exp[i \int (P^*(\mathbf{x})Q^*(\mathbf{x}) + P(\mathbf{x})Q(\mathbf{x}))\, d\mathbf{x}] \rangle \qquad (3.7a)$$

$$Z[0, 0] = 1. \qquad (3.7b)$$

Similarly one can define the CFAL for the wave fields (d_o and d_g) in the form

$$W[J_o^*, J_g^*; J_o, J_g] = \langle \exp[i\alpha \int \{J_o^*(\mathbf{x})d_o^*(\mathbf{x}) + J_g^*(\mathbf{x})d_g^*(\mathbf{x}) + J_o(\mathbf{x})d_o(\mathbf{x}) + J_g(\mathbf{x})d_g(\mathbf{x})\}\, d\mathbf{x}] \rangle \qquad (3.8a)$$

$$W[0, 0; 0, 0] = 1. \qquad (3.8b)$$

† In terms of $\kappa_{\pm g}$ defined by equation (2) of an earlier paper (Kato, 1976a),

$$M_{\pm g} = (2k\kappa_{\pm g}) \exp[\pm 2\pi i(\bar{\mathbf{g}}\mathbf{x})].$$

Here, the coefficient α is introduced to make the integration dimensionless. We distinguished the two kinds of CFAL by Z and W. However, also in the case of W, the average $\langle\ \rangle$ has to be taken ultimately over the functions $Q(\mathbf{x})$ and $Q^*(\mathbf{x})$, on which d_o and d_g *etc.* depend in the sense of a functional.

For abbreviation, $Z[P]$ and $W[J]$ are used for the expressions (3.7*a*) and (3.8*a*), respectively. Also, $\exp[P]$ and $\exp[J]$ are employed for the expression to be averaged in the respective cases.

As for the procedures discussed in § 2, the following operators are defined:

$$\mathbf{Z} = Z[\delta/i\delta C^*(\mathbf{x}),\ \delta/i\delta C(\mathbf{x})] \tag{3.9}$$

$$\hat{Q}(\mathbf{x}) = \hat{\mathbf{Z}} C(\mathbf{x}) \hat{\mathbf{Z}}^{-1} \tag{3.10}$$

with their complex conjugates. Then one can see, as the extension of (2.32),

$$\langle (Q(\mathbf{x}) + C(\mathbf{x})) f[Q+C,\ Q^*+C^*]\rangle = \hat{Q}(\mathbf{x}) \langle f[Q+C,\ Q^*+C^*]\rangle. \tag{3.11}$$

3.3. *The fundamental equation for* $W[J]$

It is straightforward to obtain the following results by taking the functional derivatives of (3.8*a*):

$$(\delta/\delta J_o(\mathbf{x}))\, W[J] = i\alpha \langle d_o(\mathbf{x}) \exp[J]\rangle \tag{3.12a}$$

$$(\delta/\delta J_g(\mathbf{x}))\, W[J] = i\alpha \langle d_g(\mathbf{x}) \exp[J]\rangle. \tag{3.12b}$$

If one takes $d_g(\mathbf{x}) \exp[J]$ in the place of the functional $f[Q+C,\ Q^*+C^*]$ in the relation (3.11) and with the use of (3.12*b*), it follows immediately that

$$\begin{aligned} &i\alpha \langle (Q(\mathbf{x}) + C(\mathbf{x})) d_g(\mathbf{x}) \exp[J]\rangle \\ &\quad = i\alpha \hat{Q}(\mathbf{x}) \langle d_g(\mathbf{x}) \exp[J]\rangle \\ &\quad = \hat{Q}(\mathbf{x}) (\delta/\delta J_g(\mathbf{x}))\, W[J]. \end{aligned} \tag{3.13a}$$

Similarly, we have

$$\begin{aligned} &i\alpha \langle (Q^*(\mathbf{x}) + C^*(\mathbf{x})) d_o(\mathbf{x}) \exp[J]\rangle \\ &\quad = i\alpha Q^*(\mathbf{x}) \langle d_o(\mathbf{x}) \exp[J]\rangle \\ &\quad = \hat{Q}^*(\mathbf{x}) (\delta/\delta J_o(\mathbf{x}))\, W[J]. \end{aligned} \tag{3.13b}$$

Operating with $(\Delta + k^2)$ on the expressions (3.12*a*) and (3.12*b*) and consulting (3.6*a*) and (3.6*b*) and (3.13*a*) and (3.13*b*), we finally obtain the fundamental equations for the CFAL, $W[J_o^*, J_g^*; J_o, J_g]$;

$$\begin{aligned} &[(\Delta + k^2)(\delta/\delta J_o(\mathbf{x})) \\ &\quad + M_{-g}(E + \hat{Q}(\mathbf{x}))(\delta/\delta J_g(\mathbf{x})) - i\alpha J_o]\, W[J] = 0 \end{aligned} \tag{3.14a}$$

$$\begin{aligned} &[(\Delta + k^2)(\delta/\delta J_g(\mathbf{x})) \\ &\quad + M_g(E + \hat{Q}^*(\mathbf{x}))(\delta/\delta J_o(\mathbf{x})) - i\alpha J_g]\, W[J] = 0. \end{aligned} \tag{3.14b}$$

The functional derivatives $(\delta/\delta J_o(\mathbf{x}),\ \delta/\delta J_g(\mathbf{x}))$ can be read as the operators ($\hat{d}_o(\mathbf{x})$ and $\hat{d}_g(\mathbf{x})$) onto $W[J]$, respectively. Then each term in [ ] has a one-to-one correspondence with each term in the wave equation (3.6).

The equation can be used to obtain systematically the relation among the multiple COF's of the wave field.

4. The statistical intensity

The crystal is statistically characterized by the multiple COF's of the lattice phase factor, such as

$$\langle \Phi(\mathbf{x})\rangle = \langle \Phi^*(\mathbf{x})\rangle = E \tag{4.1a}$$

$$\langle \Phi^2(\mathbf{x})\rangle = \langle \Phi^{*2}(\mathbf{x})\rangle = E_2 \tag{4.1b}$$

$$\begin{aligned} \langle \Phi(\mathbf{x}_1)\Phi^*(\mathbf{x}_2)\rangle &= \langle \Phi(\mathbf{x}_2)\Phi^*(\mathbf{x}_1)\rangle \\ &= E^2 + \langle Q(\mathbf{x}_1)Q^*(\mathbf{x}_2)\rangle \qquad (4.2a) \\ &= E^2 + (1-E^2)\tau(\mathbf{x}_1 - \mathbf{x}_2) \qquad (4.2b) \end{aligned}$$

$$\begin{aligned} \langle \Phi(\mathbf{x}_1)\Phi(\mathbf{x}_2)\rangle &= \langle \Phi^*(\mathbf{x}_2)\Phi^*(\mathbf{x}_1)\rangle \\ &= E^2 + \langle Q(\mathbf{x}_1)Q(\mathbf{x}_2)\rangle \qquad (4.3a) \\ &= E^2 + (E_2 - E^2)\sigma(\mathbf{x}_1 - \mathbf{x}_2) \qquad (4.3b) \end{aligned}$$

and the higher-order COF's. Here, E and E_2 are real constants and τ and σ are real functions of $|\mathbf{x}_1 - \mathbf{x}_2|$, provided that the statistical homogeneity and isotropy are fulfilled in the crystal. Also, $\tau(0) = \sigma(0) = 1$, which are equivalent to the condition of $\langle \Phi(\mathbf{x})\Phi^*(\mathbf{x})\rangle = 1$ and (4.1*b*), respectively. E and $\tau(\mathbf{x})$ were called static Debye-Waller factor and intrinsic correlation function, respectively (Kato, 1980*b*). It is outside the scope of this paper to derive them from the lattice distortion.

In general, the statistical properties of the wave field are also characterized by their multiple COF's defined as follows:

first order:

$$\langle d_o(\mathbf{x})\rangle, \quad \langle d_g(\mathbf{x})\rangle \quad \text{and c.c.} \tag{4.4a, b}$$

second order:

$$\langle d_o^*(\mathbf{x}_1) d_o(\mathbf{x}_2)\rangle, \quad \langle d_g^*(\mathbf{x}_1) d_o(\mathbf{x}_2)\rangle \tag{4.5a, b}$$

$$\langle d_o^*(\mathbf{x}_1) d_g(\mathbf{x}_2)\rangle, \quad \langle d_g^*(\mathbf{x}_1) d_g(\mathbf{x}_2)\rangle. \tag{4.5c, d}$$

The higher-order COF's may not be relevant to practical experiments in the present state of the art of X-ray diffraction.

Our aim is to find these COF's in terms of E and $\tau(\mathbf{x})$ *etc.* For this purpose, we shall define the Green functions of the wave equation (3.6). They are the solutions for special forms of $J_o(\mathbf{x})$ and $J_g(\mathbf{x})$; namely a point source at $\mathbf{x}_o$. We shall write them as

$$G_{oo}(\mathbf{x}|\mathbf{x}_o) = d_o(\mathbf{x}), \quad G_{go}(\mathbf{x}|\mathbf{x}_o) = d_g(\mathbf{x}) \quad \text{for } J_o(\mathbf{x}) = \delta(\mathbf{x} - \mathbf{x}_o) \quad \text{and} \quad J_g(\mathbf{x}) = 0 \tag{4.6a, b}$$

$$G_{og}(\mathbf{x}|\mathbf{x}_o) = d_o(\mathbf{x}), \quad G_{gg}(\mathbf{x}|\mathbf{x}_o) = d_g(\mathbf{x})$$
$$\text{for} \quad J_o(\mathbf{x}) = 0 \quad \text{and} \quad J_g(\mathbf{x}) = \delta(\mathbf{x}-\mathbf{x}_o). \tag{4.6c, d}$$

Because our wave equation is linear, it is straightforward to obtain the solution for general $J_o(\mathbf{x})$ and $J_g(\mathbf{x})$ as follows:

$$d_o(\mathbf{x}) = \int \{G_{oo}(\mathbf{x}|\mathbf{x}_o)J_o(\mathbf{x}_o) + G_{og}(\mathbf{x}|\mathbf{x}_o)J_g(\mathbf{x}_o)\}\, d\mathbf{x}_o \tag{4.7a}$$

$$d_g(\mathbf{x}) = \int \{G_{go}(\mathbf{x}|\mathbf{x}_o)J_o(\mathbf{x}_o) + G_{gg}(\mathbf{x}|\mathbf{x}_o)J_g(\mathbf{x}_o)\}\, d\mathbf{x}_o. \tag{4.7b}$$

We may write these in a neat form as

$$\mathbf{d}(\mathbf{x}) = \int G(\mathbf{x}|\mathbf{x}_o) \,.\, \mathbf{J}(\mathbf{x}_o)\, d\mathbf{x}_o \tag{4.7c}$$

where $\mathbf{d}$ and $\mathbf{J}$ are 2-component column matrices and $\mathbf{G}$ is a 2×2 square matrix, the elements being specified by o and g. The dot indicates the matrix product.

4.1. *The average wave field*

By virtue of (4.7c), the problem of obtaining the average fields (4.4a) and (4.4b) is reduced to finding the statistical Green function (SGF) $\langle \mathbf{G}(\mathbf{x}|\mathbf{x}_o)\rangle$. By definition, the Green function must satisfy the matrix equation

$$\mathbf{L}(\mathbf{x}) \,.\, \mathbf{G}(\mathbf{x}|\mathbf{x}_o) = \delta(\mathbf{x}-\mathbf{x}_o)\mathbf{I} \tag{4.8}$$

where $\mathbf{I}$ is the unit diagonal matrix. The matrix operator $\mathbf{L}(\mathbf{x})$ is decomposed as

$$\mathbf{L}(\mathbf{x}) = \mathbf{L}_0(\mathbf{x}) + \mathbf{M}(\mathbf{x}) \,.\, \mathbf{Q}(\mathbf{x}) \tag{4.9}\dagger$$

where

$$\mathbf{L}_0(\mathbf{x}) = \begin{pmatrix} \Delta + k^2 & EM_{-g}(\mathbf{x}) \\ EM_g(\mathbf{x}) & \Delta + k^2 \end{pmatrix} \tag{4.10a}\dagger$$

$$\mathbf{M}(\mathbf{x}) = \begin{pmatrix} M_{-g}(\mathbf{x}) & 0 \\ 0 & M_g(\mathbf{x}) \end{pmatrix} \tag{4.10b}$$

$$\mathbf{Q}(\mathbf{x}) = \begin{pmatrix} 0 & (Q(\mathbf{x}) + C(\mathbf{x})) \\ (Q^*(\mathbf{x}) + C^*(\mathbf{x})) & 0 \end{pmatrix}. \tag{4.10c}$$

In order to obtain the equation for SGF we shall take the average of (4.8). With (3.11), it follows that

$$\langle \mathbf{Q}(\mathbf{x}) \,.\, \mathbf{G}(\mathbf{x}|\mathbf{x}_0)\rangle = \hat{\mathbf{Q}}(\mathbf{x}) \,.\, \langle \mathbf{G}(\mathbf{x}|\mathbf{x}_0)\rangle \tag{4.11a}$$

where

$$\hat{\mathbf{Q}}(\mathbf{x}) = \begin{pmatrix} 0 & \hat{Q}(\mathbf{x}) \\ \hat{Q}^*(\mathbf{x}) & 0 \end{pmatrix}. \tag{4.11b}$$

Thus, SGF satisfies the equation

$$[\mathbf{L}_0(\mathbf{x}) + \mathbf{M}(\mathbf{x}) \,.\, \hat{\mathbf{Q}}(\mathbf{x})] \,.\, \langle \mathbf{G}(\mathbf{x}|\mathbf{x}_0)\rangle = \delta(\mathbf{x}-\mathbf{x}_0)\mathbf{I}. \tag{4.12a}$$

† $\mathbf{L}$ and $\mathbf{L}_0$ are in fact differential operators. Since, however, they have nothing to do with the operator technique developed previously, the notation $\hat{}$ is not used.

With (4.7c), we have the equivalent equation for the average field,

$$[\mathbf{L}_0(\mathbf{x}) + \mathbf{M}(\mathbf{x}) \,.\, \hat{\mathbf{Q}}(\mathbf{x})] \,.\, \langle \mathbf{d}(\mathbf{x})\rangle = [\![\mathbf{J}(\mathbf{x})]\!]. \tag{4.12b}$$

We may omit $[\![\ ]\!]$ when no external source exists at $\mathbf{x}$. These equations are exact and have the same style as the original equation (4.8) and equation (3.6), respectively. Now, however, the function $Q(\mathbf{x}) + C(\mathbf{x})$ is replaced by the operator.

In order to obtain a concrete result, we shall take the Gaussian model for RV's, $Q(\mathbf{x})$ and $Q^*(\mathbf{x})$. With this model, the corresponding operators must have the form

$$\hat{Q}(\mathbf{x}) = C(\mathbf{x}) + \int d\boldsymbol{\xi} \langle Q(\mathbf{x})Q(\boldsymbol{\xi})\rangle (\delta/\delta C(\boldsymbol{\xi})) + \int d\boldsymbol{\xi} \langle Q(\mathbf{x})Q^*(\boldsymbol{\xi})\rangle (\delta/\delta C^*(\boldsymbol{\xi})) \tag{4.13a}$$

$$\hat{Q}^*(\mathbf{x}) = C^*(\mathbf{x}) + \int d\boldsymbol{\xi} \langle Q^*(\mathbf{x})Q^*(\boldsymbol{\xi})\rangle (\delta/\delta C^*(\boldsymbol{\xi})) + \int d\boldsymbol{\xi} \langle Q^*(\mathbf{x})Q(\boldsymbol{\xi})\rangle (\delta/\delta C(\boldsymbol{\xi})). \tag{4.13b}$$

These can be derived straightforwardly as an extension of the relation (2.33) when CFAL has the form (3.7a).

The functional derivatives in (4.13) apply to $\langle \mathbf{G}(\mathbf{x}|\mathbf{x}_o)\rangle$. They have the following expressions (after a little cumbersome manipulation which is explained in the Appendix).

$$(\delta/\delta C(\boldsymbol{\xi})\langle \mathbf{G}(\mathbf{x}|\mathbf{x}_o)\rangle = -M_{-g}(\boldsymbol{\xi})\left\langle \mathbf{G}(\mathbf{x}|\boldsymbol{\xi}) \,.\, \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \,.\, \mathbf{G}(\boldsymbol{\xi}|\mathbf{x}_o)\right\rangle \tag{4.14a}$$

$$(\delta/\delta C^*(\boldsymbol{\xi})\langle \mathbf{G}(\mathbf{x}|\mathbf{x}_o)\rangle = -M_g(\boldsymbol{\xi})\left\langle \mathbf{G}(\mathbf{x}|\boldsymbol{\xi}) \,.\, \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \,.\, \mathbf{G}(\boldsymbol{\xi}|\mathbf{x}_o)\right\rangle. \tag{4.14b}$$

Based on these preparations, the matrix elements of $\mathbf{M} \,.\, \hat{\mathbf{Q}}(\mathbf{x}) \,.\, \langle \mathbf{G}(\mathbf{x}|\mathbf{x}_o)\rangle$ can be calculated explicitly. Then, SGF obeys a kind of integro-differential equation:

$$\mathbf{L}_0 \,.\, \langle \mathbf{G}(\mathbf{x}|\mathbf{x}_o)\rangle = \int \langle \mathbf{R}(\mathbf{x}|\boldsymbol{\xi}) \,.\, \mathbf{G}(\boldsymbol{\xi}|\mathbf{x}_o)\rangle\, d\boldsymbol{\xi} + \delta(\mathbf{x}-\mathbf{x}_o)I. \tag{4.15a}$$

Returning to the wave field [*cf.* (4.7c)], we also have the equation

$$\mathbf{L}_0\langle \mathbf{d}(\mathbf{x})\rangle = \int \langle \mathbf{R}(\mathbf{x}|\boldsymbol{\xi}) \,.\, \mathbf{d}(\boldsymbol{\xi})\rangle\, d\boldsymbol{\xi} + [\![\mathbf{J}(\mathbf{x})]\!]. \tag{4.15b}$$

In these equations, the matrix $\mathbf{R}$ is defined by

$$R_{oo}(\mathbf{x}|\boldsymbol{\xi}) = M_{-g}(\mathbf{x})M_g(\boldsymbol{\xi})\langle Q(\mathbf{x})Q^*(\boldsymbol{\xi})\rangle G_{gg}(\mathbf{x}|\boldsymbol{\xi}) \tag{4.16a}$$

$$R_{og}(\mathbf{x}|\boldsymbol{\xi}) = M_{-g}(\mathbf{x})M_{-g}(\boldsymbol{\xi})\langle Q(\mathbf{x})Q(\boldsymbol{\xi})\rangle G_{go}(\mathbf{x}|\boldsymbol{\xi}) \tag{4.16b}$$

$$R_{go}(\mathbf{x}|\boldsymbol{\xi}) = M_g(\mathbf{x})M_g(\boldsymbol{\xi})\langle Q^*(\mathbf{x})Q^*(\boldsymbol{\xi})\rangle G_{og}(\mathbf{x}|\boldsymbol{\xi}) \tag{4.16c}$$

$$R_{gg}(\mathbf{x}|\boldsymbol{\xi}) = M_g(\mathbf{x})M_{-g}(\boldsymbol{\xi})\langle Q^*(\mathbf{x})Q(\boldsymbol{\xi})\rangle G_{oo}(\mathbf{x}|\boldsymbol{\xi}). \quad (4.16d)$$

Here, the terms proportional to $C(\mathbf{x})$ and $C^*(\mathbf{x})$ are dropped.

In essence, the right-hand side of (4.15) represents statistical effects of the scattering processes at $\mathbf{x}$ and $\boldsymbol{\xi}$ on the wave equation. The four processes conceivable, each corresponding to one matrix element, are shown diagrammatically in Fig. 1. In general cases, the position $\boldsymbol{\xi}$ is arbitrary within the effective range of the correlation between $Q(\mathbf{x})$ and $Q(\boldsymbol{\xi})$ or $Q(\mathbf{x})$ and $Q^*(\boldsymbol{\xi})$. We shall discuss this point in § 5.3.

4.2. *Energy transfer equations*

4.2.1. *The flow vector and the conservation law.* We shall define the flow vector associated with each wave field as

$$F_o = (d_o^* \operatorname{grad} d_o - d_o \operatorname{grad} d_o^*)/2i \quad (4.17a)$$

$$F_g = (d_g^* \operatorname{grad} d_g - d_g \operatorname{grad} d_g^*)/2i. \quad (4.17b)$$

Meanwhile, the argument $\mathbf{x}$ will be suppressed. If d is a solution of the original wave equation (3.6), it follows that

$$\operatorname{div} F_o = (i/2)k^2(d_0^* d_o) + (i/2)M_{-g}(E+Q+C) \times (d_o^* d_g) + [\![S_o]\!] + \text{c.c.} \quad (4.18a)$$

$$\operatorname{div} F_g = (i/2)k^2(d_g^* d_g) + (i/2)M_g(E+Q^*+C^*) \times (d_g^* d_o) + [\![S_g]\!] + \text{c.c.} \quad (4.18b)$$

where

$$S_o = (-i/2)d_o^* J_o, \quad S_g = (-i/2)d_g^* J_g. \quad (4.19a, b)$$

F_o and F_g are real vectors. Moreover, if the crystal is non-absorbing [k real; $M_g^* = M_{-g}$] and no external source exists [$J_o = J_g = 0$], the total of the flow vectors satisfies the conservation law

$$\operatorname{div}(F_o + F_g) = 0. \quad (4.20)$$

Therefore, the expressions (4.17) are physically acceptable as the flow vector. Equations (4.18) are called the energy transfer equation in this paper.

When we adopt the form (3.2) for the wave function, it turns out that

$$\operatorname{div} F_o = 2ik(\partial/\partial s_o)(a_o^* a_o) + (a_o^* \Delta a_o - a_o \Delta a_o^*) \quad (4.21a)$$

$$\operatorname{div} F_g = 2ik(\partial/\partial s_g)(a_g^* a_g) + (a_g^* \Delta a_g - a_g \Delta a_g^*). \quad (4.21b)$$

By equating (4.18) and (4.21) and consulting the expression of $M_{\pm g}$ in the footnote on p. 4, the equivalent energy transfer equation to (4.18) can be obtained in terms of the amplitudes a_o and a_g and their complex conjugates. The neglect of the second term on the right-hand side of (4.21) will give the formulae (T-T approximation) used in the previous work.

The ensemble average of (4.18) will give the relation

$$\operatorname{div}\langle F_o\rangle = (i/2)k^2\langle d_o^* d_o\rangle + (i/2)M_{-g}(E+\hat{Q}) \times \langle d_o^* d_g\rangle + [\![\langle S_o\rangle]\!] + \text{c.c.} \quad (4.22a)$$

$$\operatorname{div}\langle F_g\rangle = (i/2)k^2\langle d_g^* d_g\rangle + (i/2)M_g(E+\hat{Q}^*) \times \langle d_g^* d_o\rangle + [\![\langle S_g\rangle]\!] + \text{c.c.} \quad (4.22b)$$

The average operation does not break the conservation law.

We also define similar flow vectors associated with the average field:

$$F_o^c = (\langle d_o^*\rangle \operatorname{grad}\langle d_o\rangle - \langle d_o\rangle \operatorname{grad}\langle d_o^*\rangle)/2i \quad (4.23a)$$

$$F_g^c = (\langle d_g^*\rangle \operatorname{grad}\langle d_g\rangle - \langle d_g\rangle \operatorname{grad}\langle d_g^*\rangle)/2i. \quad (4.23b)$$

They are called the coherent part of the flow vector and satisfy the relations

$$\operatorname{div} F_o^c = (i/2)k^2\langle d_o^*\rangle\langle d_o\rangle + (i/2)M_{-g}E\langle d_o^*\rangle\langle d_g\rangle + (i/2)M_{-g}\langle d_o^*\rangle\hat{Q}\langle d_g\rangle + [\![\langle S_o\rangle]\!] + \text{c.c.} \quad (4.24a)$$

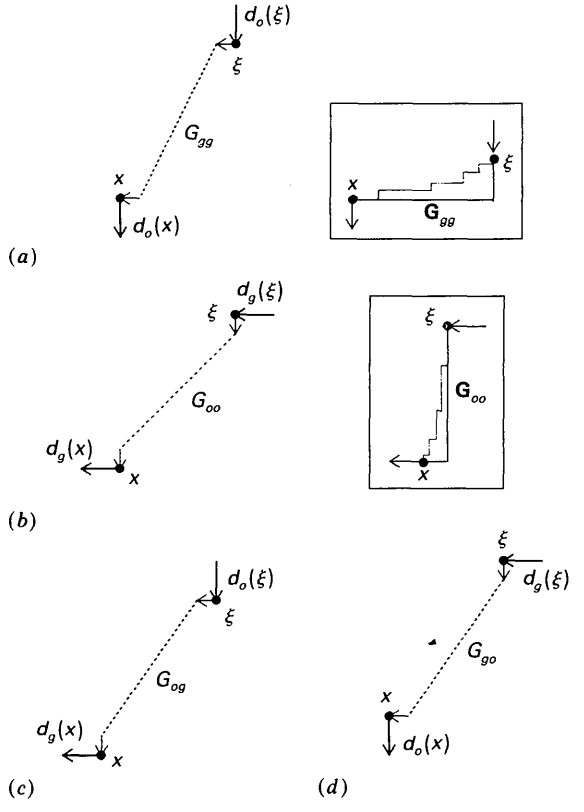

Fig. 1. Four types of scattering processes at $\mathbf{x}$ and $\boldsymbol{\xi}$. Each corresponds to the matrix element of $\mathbf{R}$ [see (4.16)]. The optical path (broken line) from $\boldsymbol{\xi}$ to $\mathbf{x}$ can be arbitrary within the range of intrinsic correlation of the lattice phases at the two points. The figures in the blocks illustrate the optical paths assumed in the previous papers (see § 5.3). The vertical and horizontal lines correspond to O and G waves. (*a*) R_{oo}, (*b*) R_{gg}, (*c*) R_{go}, (*d*) R_{og}.

$$\operatorname{div} F_g^c = (i/2)k^2\langle d_g^*\rangle\langle d_g\rangle + (i/2)M_g E\langle d_g^*\rangle\langle d_o\rangle + (i/2)M_g\langle d_g^*\rangle\hat{Q}^*\langle d_o\rangle + [\![\langle S_g\rangle]\!] + \text{c.c.} \quad (4.24b)$$

It is worth noting that this part does not satisfy the conservation law even when the crystal is non-absorbing because $\langle d_o^*\rangle$ and $\langle d_g^*\rangle$ do not commute with the operator $\hat{Q}$.

We shall also define the incoherent part of the flow vector by the relations

$$F_o^i = \langle F_o\rangle - F_o^c, \quad F_g^i = \langle F_g\rangle - F_g^c. \quad (4.25a, b)$$

4.2.2. *The flow vectors for the GRV model of Q and Q**. Since the operator $\hat{Q}$ is linear with respect to the functional derivatives $\delta/\delta C(\xi)$ and $\delta/\delta C^*(\xi)$, it follows that

$$\hat{Q}\langle d_o^* d_g\rangle = \langle(\hat{Q}d_o^*)d_g\rangle + \langle d_o^*(\hat{Q}d_g)\rangle. \quad (4.26)$$

On the other hand, $(\hat{Q}^* d_o)$ and $(\hat{Q}d_g)$ multiplied by a proper factor of $M_{\pm g}$ have been obtained in terms of the matrix $\mathbf{R}(\mathbf{x}|\xi)$ [see (4.16)]. With these results, (4.22) can be rewritten in the form

$$\begin{pmatrix}\operatorname{div}\langle F_o\rangle\\ \operatorname{div}\langle F_g\rangle\end{pmatrix} = \mathbf{A}(\mathbf{x}) + \mathbf{B}(\mathbf{x}) + \mathbf{T}_0(\mathbf{x}) + \mathbf{T}(\mathbf{x}) + [\![\langle\mathbf{S}\rangle]\!] + \text{c.c.} \quad (4.27)$$

where

$$\mathbf{A}(\mathbf{x}) = (i/2)\begin{pmatrix}EM_{-g}(\mathbf{x})\langle I_{og}(\mathbf{x}|\mathbf{x})\rangle\\ EM_g(\mathbf{x})\langle I_{go}(\mathbf{x}|\mathbf{x})\rangle\end{pmatrix} \quad (4.28a)$$

$$\mathbf{B}(\mathbf{x}) = (i/2)\int d\xi\left\langle \mathbf{S}_1(\mathbf{x}|\xi)\begin{pmatrix}I_{og}(\mathbf{x}|\xi)\\ I_{go}(\mathbf{x}|\xi)\end{pmatrix}\right\rangle \quad (4.28b)$$

$$\mathbf{T}_0(\mathbf{x}) = (i/2)k^2\begin{pmatrix}\langle I_{oo}(\mathbf{x}|\mathbf{x})\rangle\\ \langle I_{gg}(\mathbf{x}|\mathbf{x})\rangle\end{pmatrix} \quad (4.29a)$$

$$\mathbf{T}(\mathbf{x}) = (i/2)\int d\xi\left\langle \mathbf{S}_2(\mathbf{x}|\xi)\begin{pmatrix}I_{oo}(\mathbf{x}|\xi)\\ I_{gg}(\mathbf{x}|\xi)\end{pmatrix}\right\rangle. \quad (4.29b)$$

Here, the following notation is used:

$$I_{oo}(\mathbf{x}|\xi) = d_o^*(\mathbf{x})d_o(\xi) \quad (4.30a)$$

$$I_{gg}(\mathbf{x}|\xi) = d_g^*(\mathbf{x})d_g(\xi) \quad (4.30b)$$

$$I_{og}(\mathbf{x}|\xi) = d_o^*(\mathbf{x})d_g(\xi) \quad (4.30c)$$

$$I_{go}(\mathbf{x}|\xi) = d_g^*(\mathbf{x})d_o(\xi) \quad (4.30d)$$

and the matrices $\mathbf{S}_1$ and $\mathbf{S}_2$ are defined by

$$\mathbf{S}_1(\mathbf{x}|\xi) = \begin{pmatrix}-R_{og}(\mathbf{x}|\xi) & (M_{-g}^*(\mathbf{x})/M_g(\mathbf{x}))R_{go}(\mathbf{x}|\xi)\\ (M_g^*(\mathbf{x})/M_{-g}(\mathbf{x}))R_{og}(\mathbf{x}|\xi) & -R_{go}(\mathbf{x}|\xi)\end{pmatrix} \quad (4.31a)$$

$$\mathbf{S}_2(\mathbf{x}|\xi) = \begin{pmatrix}-R_{oo}(\mathbf{x}|\xi) & (M_{-g}^*(\mathbf{x})/M_g(\mathbf{x}))R_{gg}(\mathbf{x}|\xi)\\ (M_g^*(\mathbf{x})/M_{-g}(\mathbf{x}))R_{oo}(\mathbf{x}|\xi) & -R_{gg}(\mathbf{x}|\xi)\end{pmatrix}. \quad (4.31b)$$

It is worth noting that not only $\mathbf{A}$ and $\mathbf{T}_0$, but also $\mathbf{B}$ and $\mathbf{T}$ are functions of $\mathbf{x}$ after performing the integration of ξ.

The coherent part of the flow vector satisfies the energy transfer equation (4.24). For later purposes, however, we shall modify a little the third term on the right-hand side by the use of a relation similar to (4.26):

$$\langle d_o^*\rangle\hat{Q}\langle d_g\rangle = \hat{Q}(\langle d_o^*\rangle\langle d_g\rangle) - (\hat{Q}\langle d_o^*\rangle)\langle d_g\rangle. \quad (4.32)$$

Then, we shall have a relation similar to (4.27),

$$\begin{pmatrix}\operatorname{div} F_o^c\\ \operatorname{div} F_g^c\end{pmatrix} = \mathbf{A}^c(\mathbf{x}) + \mathbf{B}^c(\mathbf{x}) + \mathbf{T}_0^c(\mathbf{x}) + \mathbf{T}^c(\mathbf{x}) - \mathbf{U}(\mathbf{x}) + [\![\langle\mathbf{S}\rangle]\!] + \text{c.c.} \quad (4.33)$$

where $\mathbf{A}^c$, $\mathbf{B}^c$, $\mathbf{T}_0^c$ and $\mathbf{T}^c$ have the same structure as $\mathbf{A}$, $\mathbf{B}$, $\mathbf{T}_0$ and $\mathbf{T}$, respectively, and only I_{oo}, I_{gg} *etc.* are replaced by the following:

$$I_{oo}^c(\mathbf{x}|\xi) = \langle d_o^*(\mathbf{x})\rangle\langle d_o(\xi)\rangle \quad (4.34a)$$

$$I_{og}^c(\mathbf{x}|\xi) = \langle d_o^*(\mathbf{x})\rangle\langle d_g(\xi)\rangle \quad (4.34b)$$

$$I_{go}^c(\mathbf{x}|\xi) = \langle d_g^*(\mathbf{x})\rangle\langle d_o(\xi)\rangle \quad (4.34c)$$

$$I_{gg}^c(\mathbf{x}|\xi) = \langle d_g^*(\mathbf{x})\rangle\langle d_g(\xi)\rangle. \quad (4.34d)$$

The matrix $\mathbf{U}$, which comes from the term $(\hat{Q}\langle d_o^*\rangle)\langle d_g\rangle$ in (4.32), is given by

$$\mathbf{U}(\mathbf{x}) = (-i/2)\begin{pmatrix}|M_{-g}|^2\langle d_g^*(\mathbf{x})\rangle\int d\xi\{\langle R_{oo}d_o(\xi)\rangle + \langle R_{gg}d_g(\xi)\rangle\}\\ |M_g|^2\langle d_o^*(\mathbf{x})\rangle\int d\xi\{\langle R_{oo}d_o(\xi)\rangle + \langle R_{gg}d_g(\xi)\rangle\}\end{pmatrix}. \quad (4.35)$$

Here, the argument $(\mathbf{x}|\xi)$ is suppressed in the matrix elements of $\mathbf{R}$.

The subtraction of (4.33) from (4.27) gives the incoherent part of the flow vector as

$$\begin{pmatrix}\operatorname{div} F_o^i\\ \operatorname{div} F_g^i\end{pmatrix} = \mathbf{A}^i(\mathbf{x}) + \mathbf{B}^i(\mathbf{x}) + \mathbf{T}_0^i(\mathbf{x}) + \mathbf{T}^i(\mathbf{x}) + \mathbf{U}(\mathbf{x}) + \text{c.c.} \quad (4.36)$$

Notice that F_o^i and F_g^i are independent of the source $[\![\langle\mathbf{S}\rangle]\!]$ explicitly but related to it through $\mathbf{U}(\mathbf{x})$. The column matrices $\mathbf{A}^i$, $\mathbf{B}^i$, $\mathbf{T}_0^i$ and $\mathbf{T}^i$ have the same structure as $\mathbf{A}$, $\mathbf{B}$, $\mathbf{T}_0$ and $\mathbf{T}$ respectively, but depend upon the difference of the expressions (4.30) and (4.34) having the same double subscripts.

5. Discussion and concluding remarks

5.1. *The basic wave equation*

We started with equation (3.1) instead of the equation of T-T type which was assumed in the previous works mentioned in the *Introduction*. The new approach is required for the following reasons.

First, it seems that diffuse scattering plays significant roles in relevant experiments. In good

crystals, in which the amount per unit volume may not be large, the interference fringes in topography and/or the oscillatory profiles in goniometry have less contrast than the theoretical prediction for ideally perfect crystals. This is particularly true for higher-order reflections. [For examples, see Wada & Kato (1977) or Deutsch & Hart (1985).] The phenomena can be understood in principle from the fact that the diffuse scattering is essentially kinematical whereas the Bragg reflection is dynamical. In less-perfect crystals, also, there is evidence that the diffuse scattering is reflected by the Bragg net plane (Kashiwase, Kainuma & Minoura, 1982). Therefore, it is desirable to develop the theory of dynamical diffraction associated with diffuse scattering in a unified framework. The theory using the T–T approximation cannot treat properly these phenomena, because it assumes a single optical path for each of the O and G waves at the beginning.

Secondly, the equation of T–T type is a hyperbolic differential equation in which the Green function (the regular solution for a point source) cannot be defined in the true sense (*e.g.* Sommerfeld, 1949). On the other hand, the concept of the Green function is extremely useful for dealing with wave propagation. For these reasons, in this paper, wave equations of either elliptic type [(3.1)] or parabolic type [(3.4)] were adopted.

5.2. *Comparison with the previous theory*

In this section, we shall discuss the theoretical framework. The paper of Kato (1980*b*) is referred to as *P*.

First, we shall compare (4.15*b*) with equation (*P*-10) regarding the averaged wave field. Then, we notice that the terms related to the skew components of the **R** matrix (4.16) is missing in the previous treatment. They are proportional to $\langle Q(\mathbf{x})Q(\boldsymbol{\xi})\rangle$ which are given in expressions (4.3*a*) and (4.3*b*). When the crystal is very perfect the magnitude, estimated by $E_2-E^2\simeq\langle(\mathbf{gu})^2\rangle$, is small. Also, in another extreme case, each of E_2 and E must be very small because of the randomness of the lattice phase. Nevertheless, if the crystal is of intermediate perfection, the omitted terms may give a correction to the coupling constant of the wave equation.

The terms related to the diagonal elements, R_{oo} and R_{gg}, have expressions similar to the corresponding ones of (*P*-10). The difference will be discussed in the next subsection. They give a correction to the (complex) polarizability of the crystal which is included in k^2 of (3.1). As in the previous theory, essentially, they bring about an attenuation of the coherent wave field.

Next, we shall compare the expressions for the energy flow. Taking (*P*-16) and the present formula (4.22) or (4.27), we notice again the lack of the terms relevant to the $\mathbf{S}_1$ matrix [(4.31*a*)]; and, consequently, (*P*-16) is missing the term **B** of (4.27) which is given essentially by the skew elements of the **R** matrix. On the other hand, the two terms given by the matrix **T** correspond to the second and third terms of (*P*-16), respectively. They are related to the diagonal elements of the **R** matrix.

The terms relevant to **A** and $\mathbf{T}_0$ are dealt with in exactly the same manner as the previous theory.

Similar arguments can also be applied to the coherent and incoherent parts of the flow vector. If we put aside the terms $\mathbf{B}^c$ and $\mathbf{B}^i$, the general framework of the two theories are identical. In particular, the fact that the attenuation of the coherent part is compensated exactly by supplying the energy to the incoherent part is reserved also in the present theory. The matrix **U** in (4.33) and (4.36) describes this situation.

5.3. *Comparison of individual terms*

As an example, we shall take up the term involving R_{oo} on the right-hand side of (4.15*b*). With the use of (4.2*a*) and (4.2*b*), it has the form

$$I_n=(1-E^2)\int \mathrm{d}\boldsymbol{\xi} M_{-g}(\mathbf{x})M_g(\boldsymbol{\xi})\tau(\mathbf{x}-\boldsymbol{\xi}) \times\langle G_{gg}(\mathbf{x}|\boldsymbol{\xi})d_o(\boldsymbol{\xi})\rangle. \quad (5.1a)$$

We shall rewrite this using (3.2) and the relation in the footnote on p. 4 and the expression

$$G_{gg}(\mathbf{x}|\boldsymbol{\xi})=\bar{G}_{gg}(\mathbf{x}|\boldsymbol{\xi})\exp[i\bar{\mathbf{k}}_g(\mathbf{x}-\boldsymbol{\xi})]. \quad (5.1b)$$

After sorting out all phases of the carrier waves, we have

$$I_n=\exp[i\bar{\mathbf{k}}_o\mathbf{x}](1-E^2)(2k)^2\kappa_g\kappa_{-g} \times\int \mathrm{d}\boldsymbol{\xi}\tau(\mathbf{x}-\boldsymbol{\xi})\langle\bar{G}_{gg}(\mathbf{x}|\boldsymbol{\xi})a_o(\boldsymbol{\xi})\rangle. \quad (5.1c)$$

The corresponding one in (*P*-10*a*) can be written in the form

$$I_p=-(1-E^2)(2ik)\kappa_g\kappa_{-g}\int \mathrm{d}\eta g(\eta)\langle a_o(s_o, s_g-\eta)\rangle. \quad (5.2)$$

Here, $(2ik)$ is a multiplicative factor for adjusting the scale [see (3.4)]. The essential differences of I_p from I_n are (i) the omission of the Green function and (ii) that the integration is one-dimensional. Incidentally, the function $g(\eta)$ is the intrinsic COF of the lattice phase along the coordinate s_g, which is $\tau(\mathbf{x}-\boldsymbol{\xi})$ in the three-dimensional case.

These differences stem from the same origin; the T–T approximation. In fact, (5.2) can be derived from (5.1*c*) by assuming

$$\bar{G}_{gg}(\mathbf{x}|\boldsymbol{\xi})=(2ik)^{-1}\delta_2(\mathbf{x}_g-\boldsymbol{\xi}_g) \quad (5.3a)$$

where δ_2 is the two-dimensional δ function and $\mathbf{x}_g$ and $\boldsymbol{\xi}_g$ are the components of $\mathbf{x}$ and $\boldsymbol{\xi}$ perpendicular to the s_g direction. The expression (5.3*a*) is the solution of $(2ik)(\partial/\partial s_g)\bar{G}_{gg}=\delta(\mathbf{x}-\boldsymbol{\xi})$, which is nothing

else but an approximate form of (4.8) for the Green function under the T-T approximation and the omission of the Bragg reflection.

The full argument must be postponed to the future because the plausible form of the Green function is not worked out. Nevertheless, it is anticipated that the Green function will describe a diffuse wave propagation. This diffuseness is caused not only by diffuse scattering but also the dynamical diffraction which spreads over the Borrmann fan.

In this context, the optical paths used in the older papers (Kato, 1976*a*, *b*) are closer to the present ones. There, stepwise paths (due to the Bragg reflection) between x and ξ were assumed (see Fig. 1) rather than a straight path in the paper *P*. Now, the mathematical complications and approximations involved are eliminated by introducing the Green function.

The same argument can be applied to the term involving R_{gg} in (4.15*b*). At this stage, the lack of the skew term in the previous treatment can be interpreted as a consequence of the approximation $G_{og} = G_{go} = 0$.

In the previous work, furthermore, the amplitude $\langle a_o(s_o, s_g - \eta)\rangle$ in (5.2) is replaced by $\langle a_o(s_o, s_g)\rangle$. This approximation, which is called the gentle amplitude (GA) approximation, enables us to reduce the integro-differential equations (*P*-10) and (*P*-16) to the simple differential equations (*P*-12) and (*P*-17), respectively. The present theory also can be simplified in the same way by using $a_o(\mathbf{x})$ instead of $a_o(\xi)$ and neglecting the correlation of the Green function and the average wave field. Then, one needs an effective correlation volume defined by

$$v = \int \tau(\mathbf{x}-\xi)\langle G_{gg}(\mathbf{x}|\xi)\rangle \, \mathrm{d}\xi. \qquad (5.4)$$

It is worthwhile to comment that v must be λ dependent whereas the intrinsic correlation length τ, which is equivalently introduced in the previous theory, is strictly λ independent.

5.4. *Concluding remarks and future developments*

This paper has dealt with exactly the wave propagation and the energy flow when Bragg reflections are associated with diffuse scattering within the model of the GRV for the lattice phase. The model does not imply a small spatial fluctuation of the lattice phase. Also, no perturbation theory is used. The theory is free from the T-T approximation and the GA approximation. This approach seems necessary in order to develop a unified theory to cover large varieties of lattice distortions.

Much has to be done, however, for future developments to obtain more practical theories suitable for specific topics. In such studies, the present theory will serve as a starting point.

Finally, the present theory is strictly classical, although the mathematical techniques resemble those in quantum field theory. It seems necessary to keep this point in mind.

APPENDIX

The functional derivative of the Green function and the wave field

We shall start with equation (4.8). The notation ξ is used for the space coordinate $\mathbf{x}$. The inverse of $\mathbf{L}$ is written $\mathbf{L}^{-1}$. Then,

$$\mathbf{G}(\xi|\xi') = \mathbf{L}^{-1}(\xi)\delta(\xi-\xi')I \qquad (A.1a)$$

$$= \mathbf{L}^{-1}(\xi')\delta(\xi'-\xi)I \qquad (A.1b)$$

$$= \mathbf{G}(\xi'|\xi). \qquad (A.1c)$$

Equation (*A*.1*c*) implies the reciprocity of the Green function. It is obvious that the statistical Green function also has the same property.

Taking the functional variation of (4.8) with respect to $C(\xi)$, we shall see that

$$\mathbf{L}(\xi) \,.\, \delta\mathbf{G}(\xi|\xi') = -M(\xi) \,.\, \delta\mathbf{C}(\xi) \,.\, \mathbf{G}(\xi|\xi') \qquad (A.2a)$$

where $\mathbf{C}(\xi)$ is the skew matrix

$$\begin{pmatrix} 0 & C(\xi) \\ C^*(\xi) & 0 \end{pmatrix}.$$

Equation (*A*.2*a*) can be rewritten as

$$\delta\mathbf{G}(\xi|\xi') = -\mathbf{L}^{-1}(\xi) \,.\, M(\xi) \,.\, \delta\mathbf{C}(\xi) \,.\, \mathrm{G}(\xi|\xi'). \qquad (A.2b)$$

We multiply $\delta(\xi-\mathbf{x})$ from the right-hand side and integrate over ξ, and put $\xi' = \mathbf{x}_o$. The manipulations will give the relation

$$\delta\mathbf{G}(\mathbf{x}|\mathbf{x}_o) = -\int \mathbf{G}(\mathbf{x}|\xi) \,.\, M(\xi) \,.\, \delta\mathbf{C}(\xi) \,.\, \mathbf{G}(\xi|\mathbf{x}_o) \, \mathrm{d}\xi. \qquad (A.3)$$

In deriving this, (*A*.1*b*) is employed. Then, the functional derivative with respect to $C(\xi)$ is given by

$$(\delta/\delta C(\xi))\mathbf{G}(\mathbf{x}|\mathbf{x}_o) = -M_{-g}(\xi)\mathbf{G}(\mathbf{x}|\xi) \,.\, \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \,.\, \mathbf{G}(\xi|\mathbf{x}_o). \qquad (A.4a)$$

Similarly, we have

$$(\delta/\delta C^*(\xi))\mathbf{G}(\mathbf{x}|\mathbf{x}_o) = -M_g(\xi)\mathbf{G}(\mathbf{x}|\xi) \,.\, \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \,.\, \mathbf{G}(\xi|\mathbf{x}_o). \qquad (A.4b)$$

The average of this equation gives (4.14) in the text.

Multiplying $\mathbf{J}(\mathbf{x}_o)$ from the right and integrating over $\mathrm{d}\mathbf{x}_o$, we also obtain the functional derivatives for the wave field as

$$(\delta/\delta C(\xi))\mathbf{d}(\mathbf{x}) = -M_{-g}(\xi)\mathbf{G}(\mathbf{x}|\xi) \,.\, \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \,.\, \mathbf{d}(\xi) \qquad (A.5a)$$

$$(\delta/\delta C^*(\xi))\mathbf{d}(\mathbf{x}) = -M_g(\xi)\mathbf{G}(\mathbf{x}|\xi) \cdot \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \cdot \mathbf{d}(\xi). \quad (A.5b)$$

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Diffraction Streaks Due to Phase Disorder in One-Dimensional Displacive Modulation

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Abstract

It can be proved mathematically that the initial phase disorder in the wave of a one-dimensional displacive modulation introduces additional variations in the structure of reciprocal space and, as a result, causes characteristic diffraction streaks. Using a simple two-dimensional model, the occurrence of such streaks is confirmed in optical diffraction patterns. Electron diffraction streaks from two crystals with incommensurately modulated structures are presented and explained as the diffraction effect of the phase disorder.

1. Introduction

The wave of structural modulation is usually treated as a plane wave except for discommensurate-type modulation (Janssen & Janner, 1987; Steeds, Bird, Eaglesham, McKernan, Vincent & Withers, 1985). However, when the initial phase is not constant but varies depending on position in real space, an additional diffraction effect is expected. Recently, some streaks passing through the satellite spots were found in the electron diffraction patterns of some materials with a displacive modulated structure (Wu, Li & Hashimoto, 1990; Suzuki, Tanaka, Ishigame, Suemoto, Shibata, Onoda & Fujiki, 1986). It was proposed that there is a group of reflection planes in reciprocal space caused by disorder in the initial phase of the modulation wave.

In this paper, starting from the formula given by de Wolff (1974), it is shown that the initial phase disorder (IPD) changes the structure of reciprocal space and hence some extra diffraction streaks arise. This is also confirmed by optical diffraction for simple two-dimensional models. Finally, electron diffraction streaks observed from two kinds of crystals are shown and discussed from the viewpoint of IPD.

2. Reciprocal space of the crystal with IPD

de Wolff (1974) has given a formula to calculate the structure factor in the case of a one-dimensional displacive modulation. If the modulation wave is sinusoidal, atomic positions can be written as

$$\mathbf{r}_j = \mathbf{r}_{j0} + u_{j0} \sin 2\pi(\tau - \alpha_j), \quad (1)$$

where $\mathbf{r}_j$ is the average position of the jth atom, u_{j0} the displacement amplitude, τ the atom coordinate in four-dimensional space and α_j the initial phase. The structure factor is

$$F(hklm) = \sum_j f_j \exp 2\pi i[hx_{j0} + ky_{j0} + lz_{j0} + m(\alpha_j + \tfrac{1}{2})]J_m(2\pi\mathbf{g}\cdot\mathbf{u}), \quad (2)$$

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