

$$\langle I_{\text{Ni}}\{201\} \rangle = 4 \sum \{ \mathcal{S}_{\text{Ni}}^2 - d_{201}^2 [2b^2(\mathcal{S}_x^2 + \mathcal{S}_y^2) + b_3^2 \mathcal{S}_z^2] \}$$

$$\langle I_{\text{Ni}}\{001\} \rangle = 4 \sum (\mathcal{S}_x^2 + \mathcal{S}_y^2)$$

$\sum$  = summation over Ni<sub>1</sub> and Ni<sub>2</sub>.

### References

- BELOV, N. V., NERONOVA, N. N. & SMIRNOVA, T. S. (1965). *Tr. Inst. Kristallogr. Akad. Nauk. SSSR*, **11**, 33.  
 BERTAUT, E. F. (1968). *Acta Cryst.* **A24**, 217.  
 BERTAUT, E. F. (1970). *J. Phys. Radium C 1*, **32**, 462.

- DELORME, C. (1955). *C. R. Acad. Sci. Paris*, **241**, 1588.  
 FRUCHART, D., BERTAUT, E. F., SAYETAT, F., NASR EDDINE, FRUCHART, R. & SENATEUR, J. P. (1970), *Solid State Commun.* **8**, 91.  
*International Tables for X-ray Crystallography* (1969). Vol. I. Birmingham: Kynoch Press.  
 LOTGERING, F. K. (1956). *Philips Res. Rep.* **11**, 190.  
 OLBRYCHSKI, K. (1963). *Phys. Stat. Sol.* **3**, 1868.  
 OPECHOWSKI, W. & GUCCIONE, R. (1965). *Treatise on Magnetism*, Edited by RADO & SUHL, Vol IIA, Chap. 3. New York: Academic Press.  
 PRINCE, E. (1961). *J. Appl. Phys.* **32**, 68S.

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## The Dynamical Scattering Amplitude of an Imperfect Crystal. II. A Relation Between Takagi's Dynamical Equation and a More Exact Dynamical Equation

BY MASAO KURIYAMA

*National Bureau of Standards, Institute for Materials Research, Washington, D.C. 20234, U.S.A.*

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A basic equation of dynamical diffraction for an imperfect crystal is derived based on a general dynamical theory of diffraction. This equation is given in the form of a differential equation, and therefore can be considered to describe the diffraction processes locally inside a crystal. A phenomenological interpretation of this equation helps to fill in the gap between modern quantum mechanical treatments and ordinary treatments by dynamical theory of diffraction for a perfect crystal. In the approximation of poor resolution the more exact equation reduces to Takagi's equation. A necessary condition which makes Takagi's equation valid leads to the concept of local reciprocal lattice vectors.

### 1. Introduction

A general dynamical theory of diffraction for an imperfect crystal has been formulated previously by use of a quantum field theoretical technique\* (Ashkin & Kuriyama, 1966; Kuriyama, 1967*a,b*, 1968). The validity of this theory is not restricted by the magnitudes of strains (atomic displacements), and types of imperfections, nor by the state of the incident beam. This theory is constructed from an atomistic point of view: the generalized polarizability for X-rays or the generalized crystal potential for incoming electrons does not possess periodic translational invariance in imperfect crystals.

Much of the physics involved in diffraction from imperfect crystals has been discussed in a previous paper (Kuriyama, 1969) where a standard iteration technique is applied to this new formulation; the effects of crystal imperfections on dynamical diffraction have been treated correctly by properly accounting for the phase modulation of the diffracted beams, and not as a result of the assumption of modified Bloch waves.

Using this new formulation, a dynamical expression for the scattering amplitude of an imperfect crystal has been derived in a compact form (Kuriyama, 1970; hereafter this paper will be referred to as I).

On the other hand there have appeared a number of works on the dynamical theory of diffraction in imperfect crystals (Penning & Polder, 1961, 1964; Kato, 1963*a,b,c*, 1964*a,b*; Bonse, 1964; Kambe, 1965; Wilkens, 1966; Takagi, 1962, 1969; Balibar & Authier, 1967; Taupin, 1964; Chukhovskii & Shtolberg, 1970; Afanas'ev & Kohn, 1971; Howie & Whelan, 1961; Dederichs, 1966, 1967; and probably others). All but Dederichs's work appear to be phenomenological extensions of classical (Ewald-Laue-Bethe) perfect crystal theory to imperfect crystals and, hence, only find applications in those cases in which distortions are small.

Recent developments using such a phenomenological approach have led to equations such as Takagi's (1962, 1969). In this paper, therefore, the aim is to study the relation between Takagi's equation and the more exact dynamical equation.

### 2. Dynamical scattering amplitude

The scattering amplitude for an X-ray beam striking a crystal at position **R** with initial momentum **k** and

\* The dynamical theory of diffraction has also been formulated for a perfect crystal, using quantum field theoretical techniques, by Ohtsuki & Yanagawa (1966) and Hannon & Trammell (1968, 1969).

emerging at  $\mathbf{R}'$  with momentum,  $\mathbf{k}'$ , is given (Ashkin & Kuriyama, 1966; Kuriyama, 1967a,b, 1968) by

$$\langle \mathbf{k}'\mathbf{R}' ; \text{out} | \mathbf{k}, \mathbf{R}; \text{in} \rangle = \int d^3\mathbf{p}' \int d^3\mathbf{p} A^*(\mathbf{k}', \mathbf{p}'; \mathbf{R}') S(\mathbf{p}', \mathbf{p}) A(\mathbf{k}, \mathbf{p}; \mathbf{R}), \quad (2-1)$$

where  $A$  is the Fourier transform of the free photon wave packet depending on a parameter  $\mathbf{R}$  which indicates the spatial location for the maximum intensity. The function  $A$  determines the momentum and the energy distribution of the incoming X-ray beam.  $A^*$  is the complex conjugate of  $A$  unless additional slit systems are introduced to detect scattered photons.  $A^*$  may be replaced by a spectral function which characterizes the momentum and energy response of a detector when it is in use. The quantity  $S(\mathbf{p}'\mathbf{p})$  is the scattering matrix element of the crystal.

It has been shown in I that a compact expression of this matrix can be derived, under some conditions, for a crystal plate of thickness  $D$ :

$$S(\mathbf{p}', \mathbf{p}) = (p_z/|\mathbf{p}|) \delta(|\mathbf{p}'| - |\mathbf{p}|) \times \sum_{\mathbf{q}} \sum_{\mathbf{q}'} \delta(\mathbf{p}_t + \mathbf{J}_t + \mathbf{q}_t - \mathbf{p}'_t) \{ \exp[-i \frac{D}{2p'_z} M(\mathbf{p}') ] \}_{\mathbf{q}, \mathbf{q}'} \quad (2-2)$$

where the projection of a vector onto the crystal surface is described by a subscript  $t$ , and its projection along the normal of the surface by a subscript  $z$ . The matrix is a supermatrix, *i.e.* a matrix of matrices. The  $(I, J)$  element of a supermatrix is given by a matrix whose elements are specified by  $(q, q')$ . In this notation, a supermatrix  $S$  is written  $[S]_{\mathbf{q}, \mathbf{q}'}$ .

Expressing the matrix as a super matrix makes it possible to identify the subscript,  $\mathbf{I}$  or  $\mathbf{J}$ , as a reciprocal lattice vector defined in the perfect reference crystal. The perfect reference crystal is an imaginary crystal which is obtained by putting all atoms of a given type at ideally periodic lattice sites. Consequently, the superscript,  $\mathbf{q}$  or  $\mathbf{q}'$ , describes the deviation of an X-ray beam from an ideal direction of Bragg diffraction.

The matrix  $M$  is related to the Fourier transform of the generalized polarizability of the crystal. An element of  $M$  has the form (see I):

$$[M(\mathbf{p}')]_{\mathbf{q}, \mathbf{q}'} = [M_0 \Theta^{-2}]_{\mathbf{q}, \mathbf{q}'}, \quad (2-3)$$

where

$$[M_0(\mathbf{p}')]_{\mathbf{q}, \mathbf{q}'} = [(\mathbf{p}' - \mathbf{q} - \mathbf{I})^2 - p'^2] \delta_{\mathbf{q}, \mathbf{q}'} \delta_{\mathbf{I}, \mathbf{J}} - \gamma(\mathbf{J} + \mathbf{q}'; \mathbf{I} + \mathbf{q}), \quad (2-4)$$

and

$$[\Theta]_{\mathbf{q}, \mathbf{q}'} = \left\{ 1 - \frac{I_z + q_z}{p_z} \right\}^{1/2} \delta_{\mathbf{q}, \mathbf{q}'} \delta_{\mathbf{I}, \mathbf{J}}. \quad (2-5)$$

The  $\gamma$  is given by

$$\gamma(\mathbf{J} + \mathbf{q}'; \mathbf{I} + \mathbf{q}) = \frac{1}{N} \sum_l v_l(\mathbf{J} + \mathbf{q}'; \mathbf{I} + \mathbf{q}) \exp[-i(\mathbf{J} + \mathbf{q}' - \mathbf{I} - \mathbf{q}) \cdot (\mathbf{I} + \mathbf{u}_l)], \quad (2-6)$$

where  $N$  is the total number of atoms in the crystal,  $v_l$  is the 'atomic' polarizability of the atom at the  $l$ th site,  $\mathbf{I}$  is the position vector of the  $l$ th site in the perfect reference crystal, and  $\mathbf{u}_l$  is the displacement of the  $l$ th atom from its ideal position.

If all the positions of atoms and their atomic polarizabilities in the crystals are known, then the scattering amplitude can be calculated from the exponential matrix element given by (2-2), using equations (2-3) through (2-6). Therefore, our problem is reduced to calculating the following matrix:

$$[S(\sigma_f)]_{\mathbf{q}, \mathbf{q}'} = [\exp\{i\sigma_f M\}]_{\mathbf{q}, \mathbf{q}'}, \quad (2-7)$$

where

$$\sigma_f = -z/2p'_z = -z/2p \cos \Phi_f \quad (2-8)$$

and  $\cos \Phi_f$  is the direction cosine of the propagation vector  $\mathbf{p}'$ .

By definition the exponential matrix satisfies the differential equation:

$$\frac{dS(\sigma_f)}{d\sigma_f} = iM \cdot S(\sigma_f) \quad (2-9)$$

with the initial condition  $S(0) = \mathbf{I}$ . Therefore, as long as one can write the scattering matrix in an exponential matrix form, the calculation of the scattering matrix is equivalent to solving the differential equation (2-9). One may consider it as a basic diffraction equation.

As discussed in I there are at least two major approximations required to derive the scattering amplitude in form of an exponential matrix: one is that the reciprocal lattice vectors of interest, such as  $\mathbf{I}$  and  $\mathbf{J}$ , should lie very close to the Ewald sphere, and the second, that the double Fourier transform of the polarizability  $v(\mathbf{k}_1, \mathbf{k}_2)$  of the 'atomic' electrons depends only upon the momentum change  $(\mathbf{k}_1 - \mathbf{k}_2)$ , *i.e.* the scattering vector. The latter approximation holds almost always for usual diffraction problems except when resonance absorption takes place *via* atomic electrons inside the crystal. Unless the atomic polarizability term creates additional poles in the scattering amplitude, this second approximation has no effect on the results.

The first approximation was originally introduced to avoid mathematical complications due to backward scattering (specular reflection). A further investigation makes it possible to replace this approximation by a less restricted one. The well known two-wave approximation in ordinary dynamical theory may serve best as an example to give some idea about the backward scattering. The inverse photon Green's function (or the dispersion equation in ordinary dynamical theory) contains the energy term [see equation (2-18) in I]

$$D^{-1} = (\mathbf{k} - \mathbf{I})^2 - \omega^2 - [\text{Polarization term}] \equiv (\mathbf{k} - \mathbf{I})^2 - \mathbf{Q}^2, \quad (2-10)$$

where we consider diffraction in a perfect crystal and

$\mathbf{I}$  is a reciprocal lattice vector. Equation (2-10) can be written

$$D^{-1} = (\mathbf{Q} - \mathbf{I})^2 - \mathbf{Q}^2 + 2(\mathbf{Q} - \mathbf{I})(\mathbf{k} - \mathbf{Q}) + (\mathbf{k} - \mathbf{Q})^2. \quad (2-11)$$

It is customary in the two-wave approximation to discard the second order term,  $(\mathbf{k} - \mathbf{Q})^2$ , so that one need only determine two tie points on the dispersion surface. In the quantum mechanical formulation of the scattering amplitude, this second order term can, however, be retained. It gives rise to two additional poles in the scattering amplitude, and these poles give the backward scattering of the type similar to specular reflection. The ratio of backward scattering to Bragg scattering is quite negligible, being of the order of  $(1-n)^2$  where  $n$  is the refractive index of the crystal (Ashkin & Kuriyama, 1966).

This situation existed in the calculation of the scattering amplitude in I. The adoption of the above-mentioned first assumption enabled one to discard the second-order term. One could have calculated the scattering amplitude in I without this assumption. Then the result would have contained the additional terms, whose ratio to the leading term of an exponential form is again of the order of  $(1-n)^2$ . Consequently it follows that the scattering amplitude can be approximated by a compact form of matrix exponential if the backward scattering can be considered negligible. Ordinary diffraction satisfies this condition. It can be understood from this condition that the scattering amplitude for low-energy electron diffraction cannot be approximated by an exponential matrix.

### 3. Dynamical equation of diffraction

The previous discussion has shown that the restrictions on the validity of equation (2-9) are less severe than the ones originally imposed in I. It would be, therefore, most desirable to solve equation (2-9) as it is given for dynamical diffraction in an imperfect crystal. Its solution can be obtained in the form of a Feynman (1951) expansion using an iteration method. Such a solution would doubtlessly require a considerable amount of elaborate work. It would be worthwhile looking into the possibility of solving this equation in a different manner. Although it may cost the mathematical rigor which equation (2-9) enjoys, it is, nonetheless, interesting to know if equation (2-9) can be approximated by a simpler equation, and to find out how good the approximation will be.

Furthermore, one may benefit from such an approach by being able to interpret equation (2-9) in terms of phenomenological (or classical) concepts of diffracted wave fields. It may also help to find conceptual relations between existing approximations, most of which have been devised in one way or another as a result of extensions of the dynamical theory of diffraction in a perfect crystal.

Let us assume that (1) the atomic polarizability  $v(\mathbf{k}_1, \mathbf{k}_2)$  is given as a function of  $\mathbf{k}_1 - \mathbf{k}_2$ :  $v(\mathbf{k}_1, \mathbf{k}_2) \equiv$

$v(\mathbf{k}_1 - \mathbf{k}_2)$ , and (2) in the geometrical matrix elements (2-5),  $q_z$  can be neglected compared to  $p'_z + I_z$ . These two assumptions are almost equivalent to the restrictions used in I. For an imperfect crystal the reciprocal lattice points are not mathematical points, but have spatial dimensions. Consequently the momenta of possible scattered beams generally have directions deviated from the ideal directions defined by mathematical reciprocal points. This effect is known as line broadening in the kinematical diffraction theory. The assumption (2) implies that, even for the 'extended' reciprocal lattice points, the direction cosines of momenta of possible scattered beams can be approximated by those expected in the perfect reference crystal. It should be noted that the assumption (2) is applied only to the geometry, not to the matrix elements (2-4) which really describe the nature of dynamical interactions among scattered beams in the imperfect crystal.

The matrix  $\mathbf{S}$  is also a function of  $\mathbf{p}'$  in equation (2-2). In this section we will recover the explicit dependence of  $\mathbf{S}$  and  $\mathbf{M}$  on  $\mathbf{p}'$  in the expression (2-7) by writing, for example,

$$\mathbf{S}(\sigma_f) \equiv \mathbf{S}(\sigma_f; \mathbf{p}') = \mathbf{S}(\mathbf{p}'). \quad (3-1)$$

In the present approximation the matrix  $\mathbf{M}$  has the following property:

$$[\mathbf{M}(\mathbf{p}' + \mathbf{K} + \mathbf{q}_K)]_{\mathbf{i}, \mathbf{j}}^{\mathbf{q}, \mathbf{q}'} = \frac{p'_z + K_z}{p'_z} [\mathbf{M}(\mathbf{p}')]_{\mathbf{i} - \mathbf{K}, \mathbf{j} - \mathbf{K}}^{\mathbf{q}, \mathbf{q}'}. \quad (3-2)$$

It follows from equations (2-7) and (3-2) that

$$[\mathbf{S}(\mathbf{p}' + \mathbf{K} + \mathbf{q}_K)]_{\mathbf{i}, \mathbf{j}}^{\mathbf{q}, \mathbf{q}'} = [\mathbf{S}(\mathbf{p}')]_{\mathbf{i} - \mathbf{K}, \mathbf{j} - \mathbf{K}}^{\mathbf{q}, \mathbf{q}'}, \quad (3-3)$$

where another definition of matrix exponential

$$\exp \mathbf{A} = \sum_{n=0}^{\infty} \frac{1}{n!} \mathbf{A}^n$$

has been used.

It is easily understood from equation (2-2) that the matrix element  $[\mathbf{S}(\mathbf{p}')]_{\mathbf{i}, \mathbf{j}}^{\mathbf{q}, \mathbf{q}'}$  can be thought of as the amplitude of the beam scattered (or diffracted) in the direction of  $\mathbf{p}' = \mathbf{p} + \mathbf{J} + \mathbf{q}$  when the incident beam has the direction of  $\mathbf{p}$ . A general matrix element  $[\mathbf{S}(\mathbf{p}')]_{\mathbf{i}, \mathbf{j}}^{\mathbf{q}, \mathbf{q}'}$  can then be interpreted as the amplitude of the beam scattered by  $(\mathbf{J} - \mathbf{I} + \mathbf{p}' - \mathbf{q})$ -Bragg diffraction in the direction of  $\mathbf{p}' - \mathbf{I} - \mathbf{q}$  since

$$[\mathbf{S}(\mathbf{p}')]_{\mathbf{i}, \mathbf{j}}^{\mathbf{q}, \mathbf{q}'} = [\mathbf{S}(\mathbf{p}' - \mathbf{I} - \mathbf{q})]_{\mathbf{i}, \mathbf{j}}^{\mathbf{q}, \mathbf{q}'}. \quad (3-4)$$

by (3-3). Therefore, equation (2-9) can now be considered as a dynamical equation for multiple beams.

Let us take as an example a single Bragg diffraction condition where two final states are characterized by photon momenta  $\mathbf{p}' = \mathbf{p}_K = \mathbf{p} + \mathbf{K}$  with  $\mathbf{K} = \mathbf{0}$  for the transmitted beam and with  $\mathbf{K} = \mathbf{H}$  for the  $\mathbf{H}$ -Bragg diffracted beam. For simplicity we suppress the set of superscripts  $(\mathbf{q}, \mathbf{q}')$  without loss of generality. In this case, equation (2-9) produces two sets of differential equations depending upon which final state  $\mathbf{p}_K$  is used to write the matrix element:

(1)  $\mathbf{p}_H$ -representation

$$\left\{ \begin{array}{l} \frac{d}{d\sigma_H} [S(\mathbf{p}_H)]_{0, \mathbf{H}} = i[M(\mathbf{p}_H)S(\mathbf{p}_H)]_{0, \mathbf{H}} \\ \frac{d}{d\sigma_H} [S(\mathbf{p}_H)]_{\mathbf{H}, \mathbf{H}} = i[M(\mathbf{p}_H)S(\mathbf{p}_H)]_{\mathbf{H}, \mathbf{H}} \end{array} \right. \quad (3-5a)$$

$$\left\{ \begin{array}{l} \frac{d}{d\sigma_H} [S(\mathbf{p}_H)]_{0, \mathbf{H}} = i[M(\mathbf{p}_H)S(\mathbf{p}_H)]_{0, \mathbf{H}} \\ \frac{d}{d\sigma_H} [S(\mathbf{p}_H)]_{\mathbf{H}, \mathbf{H}} = i[M(\mathbf{p}_H)S(\mathbf{p}_H)]_{\mathbf{H}, \mathbf{H}} \end{array} \right. \quad (3-5b)$$

and (2)  $\mathbf{p}_0$ -representation

$$\left\{ \begin{array}{l} \frac{d}{d\sigma_0} [S(\mathbf{p}_0)]_{-\mathbf{H}, 0} = i[M(\mathbf{p}_0)S(\mathbf{p}_0)]_{-\mathbf{H}, 0} \\ \frac{d}{d\sigma_0} [S(\mathbf{p}_0)]_{0, 0} = i[M(\mathbf{p}_0)S(\mathbf{p}_0)]_{0, 0} \end{array} \right. \quad (3-6a)$$

$$\left\{ \begin{array}{l} \frac{d}{d\sigma_0} [S(\mathbf{p}_0)]_{-\mathbf{H}, 0} = i[M(\mathbf{p}_0)S(\mathbf{p}_0)]_{-\mathbf{H}, 0} \\ \frac{d}{d\sigma_0} [S(\mathbf{p}_0)]_{0, 0} = i[M(\mathbf{p}_0)S(\mathbf{p}_0)]_{0, 0} \end{array} \right. \quad (3-6b)$$

Here,  $\sigma_K$  is given by equation (2-8) where  $\Phi_f$  is either  $\Phi_0$  (the incident angle) or  $\Phi_H$  (the angle of emergence). Using the properties (3-2) and (3-3) and equation (2-8), we obtain, for example,

$$\begin{aligned} \frac{d}{d\sigma_0} [S(\mathbf{p}_0)]_{-\mathbf{H}, 0} &= \frac{\cos \Phi_0}{\cos \Phi_H} \frac{d}{d\sigma_H} [S(\mathbf{p}_H)]_{0, \mathbf{H}} \\ &= \frac{p_z}{p_z + H_z} \frac{d}{d\sigma_H} [S(\mathbf{p}_H)]_{0, \mathbf{H}} \end{aligned}$$

for the left hand term of (3-6a), and

$$i[M(\mathbf{p}_0)S(\mathbf{p}_0)]_{-\mathbf{H}, 0} = \left( \frac{p_z}{p_z + H_z} \right) i[M(\mathbf{p}_H)S(\mathbf{p}_H)]_{0, \mathbf{H}}$$

for the right hand term. This proves that equation (3-5a) is, in fact, equivalent to equation (3-6a). Similarly equation (3-5b) is proven to be equivalent to equation (3-6b).

The dynamical equation is therefore given in the mixed representation by

$$\frac{d}{d\sigma_K} [S(\mathbf{p}_K)]_{0, \mathbf{k}'} = i[M(\mathbf{p}_K)S(\mathbf{p}_K)]_{0, \mathbf{k}'}, \quad (3-7)$$

where

$$\mathbf{p}_K = \mathbf{p} + \mathbf{K} + \mathbf{q}_K. \quad (3-8)$$

$\mathbf{K}$ , of course, takes the value of  $\mathbf{0}$  and  $\mathbf{H}$  in the case of a single Bragg diffraction. Again, a rigorously exact solution of equation (3-7) would follow that of equation (2-9). However, equation (3-7) lends itself (more readily) to phenomenological interpretation; *viz.* studying the local behavior of the scattering amplitude.

#### 4. Approximation of poor momentum resolution

At this stage it may be worthwhile to think back to the nature of our assumptions which have helped to understand the physical meaning of the exponential matrix (2-7). Their effect on the scattering amplitude (2-1) has not yet been studied. Since the modulus squared of the scattering amplitude is the only observable in a strict sense, it is important to find out how our observable has been affected by our assumptions.

The assumption (2) has, in effect, meant that our geometrical resolution for the momenta of scattered

photons is insufficient to resolve individual scattered beams around a Laue spot. The incident beam, however, can still be an ideal plane wave, since its state is completely independent of both the state of scatterers, namely a crystal, and the state of detecting systems. The present geometrical condition can be written in the scattering amplitude (2-1) by the following spectral functions:

$$A(\mathbf{k}, \mathbf{p}) = \begin{cases} 1, & \text{if } \mathbf{k} = \mathbf{p} \\ 0, & \text{otherwise} \end{cases} \quad (4-1)$$

$$A^*(\mathbf{k}', \mathbf{p}') = A^*(\mathbf{k}', \mathbf{k} + \mathbf{K} + \mathbf{q}) = \begin{cases} 1, & \text{regardless of } \mathbf{q} \text{ if} \\ & \mathbf{K} = \mathbf{0} \text{ or } \mathbf{H} \\ 0, & \text{otherwise.} \end{cases} \quad (4-2)$$

The scattering amplitude is thus given by:

$$\langle \mathbf{k}'; \text{out} | \mathbf{k}; \text{in} \rangle = \frac{k_z}{k'_z} \sum_{\mathbf{q}} [S(\mathbf{k} + \mathbf{K} + \mathbf{q})]_{0, \mathbf{k}}^{\mathbf{q}, \mathbf{k}'} \quad (4-3)$$

where  $\mathbf{k}'$  is in the neighborhood of  $\mathbf{k} + \mathbf{K}$  and  $\mathbf{K}$  takes on the value of  $\mathbf{0}$  and  $\mathbf{H}$ . If we specify the momentum of the out-going wave by

$$\mathbf{k}' \equiv \mathbf{k}'_K = \mathbf{k} + \mathbf{K} + \mathbf{q}_F = \mathbf{k}_K + \mathbf{q}_F, \quad (4-4)$$

equation (4-3), using (3-3), can be rewritten

$$\begin{aligned} \langle \mathbf{k}'_K; \text{out} | \mathbf{k}; \text{in} \rangle &= \frac{k_z}{k_{K,z}} \sum_{\mathbf{q}} [S(\mathbf{k}'_K - \mathbf{q})]_{0, \mathbf{k}}^{\mathbf{q}, \mathbf{k}'_K} \\ &= \frac{k_z}{k_{K,z}} \sum_{\mathbf{q}} [S(\mathbf{k}'_K)]_{0, \mathbf{k}}^{\mathbf{q}, \mathbf{k}'_K}. \end{aligned} \quad (4-5)$$

One can always set  $\mathbf{q}_F$  equal to zero if one wishes.

Unfortunately this approximation is not of much help in simplifying equation (3-7). As demonstrated in a problem of X-ray interferometry (Kuriyama, 1971), the matrix  $S$  contains the exact phase relations between component waves. These exact relations are still retained in the approximate scattering amplitude (4-5) in spite of the gross approximation for momentum resolution. Therefore, we have to introduce some approximation about those phases to the same degree of order as in the approximation for resolution. This may be done by dropping the phase of  $S$  entirely.

To proceed, another important aspect must be taken into consideration. The matrix  $S$  of interest is mathematically described as a local function of  $\sigma_K$ , as seen in equation (2-7). Although we have not given any physical meaning to this quantity  $\sigma_K$ , the phases of  $S(\sigma_K)$  must be determined at the value of  $\sigma_K$ . Instead of the approximate scattering amplitude (4-5), we may define a local approximate scattering amplitude without the exact phases of component waves\* by

\* One may define  $\varphi_K(\sigma_K)$  by taking a double sum not only over  $q$ , but also over  $q_F$ . In doing so, the local approximate scattering amplitude will have more resemblance to the form of statistical ensemble average.

$$\begin{aligned}\varphi_K(\sigma_K) &= \frac{k_z}{k_{K,z}} \langle S(\sigma_K; k'_K) \rangle_{0,K} \\ &= \left( \frac{k_z}{k_{K,z}} \right) \frac{1}{N} \sum_{\mathbf{q}} [S(\mathbf{k}'_K)]_{0,K}^{q,q_F} \exp(-i\mathbf{q} \cdot \mathbf{R}_L) \\ &\quad \times \exp(+i\mathbf{q}_F \cdot \mathbf{R}_L),\end{aligned}\quad (4-6)$$

where  $\mathbf{R}_L$  is the actual position of an atom (or unit cell) of interest, being given by  $\mathbf{R}_L = \mathbf{L} + \mathbf{u}_L$ , and is considered to be a function of  $\sigma_K$ .

We thus obtain the differential equation for  $\varphi_K$ 's:

$$\begin{aligned}\frac{d\varphi_K}{d\sigma_K} &= i(\mathbf{k}_K^2 - \mathbf{k}^2)\varphi_K - i \sum_{\mathbf{J}} v(\mathbf{J}) \exp(-i\mathbf{J} \cdot \mathbf{R}_L)\varphi_{K-J} \\ &\quad - \frac{i}{N} \left( \frac{k_z}{k_{K,z}} \right) \sum_{\mathbf{q}} [2\mathbf{k}_K(\mathbf{q} - \mathbf{q}_F)] \\ &\quad + \frac{d}{d\sigma_K} \{(\mathbf{q} - \mathbf{q}_F)\mathbf{R}_L\} \times [S(\mathbf{k}'_K)]_{0,K}^{q,q_F} \exp(-i\mathbf{q}\mathbf{R}_L) \\ &\quad \times \exp(+i\mathbf{q}_F\mathbf{R}_L),\end{aligned}\quad (4-7)$$

where the dynamical equation (3-7) is used with the approximate form of  $v$  in M. In the derivation of the second term, the property (3-3) is used.

We may define another form of the approximate scattering amplitude by

$$\bar{\varphi}_K(\sigma_K) = \left( \frac{k_z}{k_{K,z}} \right) \frac{1}{N} \sum_{\mathbf{q}} [S(\mathbf{k}'_K)]_{0,K}^{q,q_F} \exp(-i\mathbf{q} \cdot \mathbf{R}_L) \times \exp[+i(\mathbf{K} + \mathbf{q}_F) \cdot \mathbf{R}_L], \quad (4-8)$$

where the cancellation of the phases is done including  $\mathbf{K}$ . For this quantity we obtain the differential equation:

$$\begin{aligned}\frac{d\bar{\varphi}_K}{d\sigma_K} &= i(\mathbf{k}_K^2 - \mathbf{k}^2)\bar{\varphi}_K - i \sum_{\mathbf{J}} v(\mathbf{J})\bar{\varphi}_{K-J} - \frac{i}{N} \left( \frac{k_z}{k_{K,z}} \right) \\ &\quad \times \sum_{\mathbf{q}} [2\mathbf{k}_K(\mathbf{q} - \mathbf{q}_F) + \frac{d}{d\sigma_K} \{(\mathbf{q} - \mathbf{q}_F - \mathbf{K})\mathbf{R}_L\}] \\ &\quad \times [S(\mathbf{k}'_K)]_{0,K}^{q,q_F} \exp(-i\mathbf{q} \cdot \mathbf{R}_L) \\ &\quad \exp[+i(\mathbf{K} + \mathbf{q}_F) \cdot \mathbf{R}_L].\end{aligned}\quad (4-9)$$

### 5. A further approximation: Takagi's equation

Equation (4-7) can be simplified drastically if its third term is somehow set equal to zero. This simplification yields Takagi's equation [equation (42) in Takagi (1962, 1969)]. Let us write

$$Q(\mathbf{q}) = 2\mathbf{k}_K\mathbf{q} + \frac{\partial}{\partial\sigma_K} (\mathbf{q} \cdot \mathbf{R}_L) = 2\mathbf{k}_K\{\mathbf{q} - \nabla(\mathbf{q} \cdot \mathbf{R}_L)\}, \quad (5-1)$$

where equation (2-8) is used to rewrite the second term. In an approximation where the sizes of the reciprocal lattice points in the imperfect crystal (see § 3) are much smaller than the size of the reciprocal unit cell, one may practically set  $Q(\mathbf{q}) = 0$ , regardless of the value of  $\mathbf{q}$ . This is one of the possible conditions making Takagi's equation valid. However, this approximation imposes a certain condition on the magnitude of the local

atomic displacement  $\mathbf{u}_L$ , where  $\mathbf{u}_L$  is given by  $\mathbf{R}_L = \mathbf{L} + \mathbf{u}_L$ .

Another approximation may be given which appears to relax this condition on the magnitude of local displacements. Suppose that the sum over  $\mathbf{q}$  in the third term of equation (4-7) is well represented by a single term for which  $\mathbf{q} = \bar{\mathbf{q}}(L)$ , being a function of the position of the atom of interest. Then we can select for this value of  $\bar{\mathbf{q}}(L)$

$$\mathbf{Q}(\bar{\mathbf{q}}) \equiv 0, \quad (5-2)$$

which implies

$$\bar{\mathbf{q}} - \nabla(\bar{\mathbf{q}} \cdot \mathbf{u}_L) \equiv 0. \quad (5-3)$$

The condition (5-3) at the local point  $L$  immediately indicates that a local perfect reference crystal is chosen at this point  $L$ . The local perfect reference crystal is an imaginary crystal made up of unit cells whose basis vectors are given by  $\mathbf{a} + \nabla(\mathbf{a} \cdot \mathbf{u}_L)$ . In this approximation, therefore,  $\sigma_K$  is considered to represent a coordinate at  $L$  and around  $L$  exists a local reference crystal in which Takagi's equation is valid.

This situation may be understood better from equation (4-9). This equation is reduced under the local condition (5-3) to the following form:

$$(-i) \frac{\partial \bar{\varphi}_K}{\partial \sigma_K} = [\mathbf{k}_K^2 - \mathbf{k}^2 - 2\mathbf{k}_K \nabla(\mathbf{K} \cdot \mathbf{u}_L)] \bar{\varphi}_K - \sum_{\mathbf{J} \neq 0} v(\mathbf{J}) \bar{\varphi}_{K-J}. \quad (5-4)$$

If the local perfect reference crystal is defined again at this point  $L$ , then the quantity

$$\mathbf{K}(L) = \mathbf{K} - \nabla(\mathbf{K} \cdot \mathbf{u}_L) \quad (5-5)$$

gives the local reciprocal lattice vectors at the point  $L$ . Thus equation (5-4) gives another form of Takagi's equation [See equation (40), Takagi (1969)].

Since equations (4-7) and (4-9) are differential equations defined locally, one can make the conditions (5-3) valid locally at various points,  $L$ , in the crystal. In other words a real imperfect crystal may be divided into local cells (or volume elements), each of which is characterized by different local perfect reference crystals. Within these reference crystals, the simplified forms of equations (4-7) and (4-9), which are Takagi's equations (42) and (40), are considered to be valid in the present approximations. Therefore the validity of Takagi's equations requires that the condition (5-3) be satisfied locally in addition to the other approximations which were required to derive equations (4-7) and (4-9). Takagi's formulation is based on infinitesimal mosaicity of crystals. However, the use of even the more traditional coarse mosaic picture for single crystals is of questionable value.

### 6. Conclusion and discussion

Starting with a rigorous expression for the scattering amplitude, we have proved that the calculation of the scattering amplitude is equivalent to solving the differential equation (2-9), if two approximations, one for

atomic polarizability and the other for backward scattering, are satisfied. Thus equation (2-9) can be considered as the basic diffraction equation for an imperfect crystal.

Instead of solving equation (2-9) in a standard way, we have simplified it by proposing approximate scattering amplitudes under the condition of poor resolution. Ultimately the simplified forms, equations (4-7) and (4-9), are reduced to Takagi's two forms by adding further the local conditions (5-3) for an arbitrary local point in the crystal.

In the dynamical theory of diffraction for a perfect crystal, the interactions between beams, characterized by reciprocal-lattice vectors  $\mathbf{K}$ , are thoroughly taken into account. For an imperfect crystal, beams should be characterized not by  $\mathbf{K}$ 's alone, but by  $\mathbf{K}$ 's and  $\mathbf{q}$ 's. Any dynamical treatment of diffraction in an imperfect crystal should, therefore, deal not only with the interactions between  $\mathbf{K}$  beams, but also with those between beams of different  $\mathbf{q}$ 's on an equal basis. However, we found that the approximations or assumptions which were required to obtain Takagi's forms almost forced us to abandon such a real dynamical property of multi-beam interactions; the interactions due to different  $\mathbf{q}$ 's were somehow blurred, and the resultant approximate forms of equation (2-9) contained only the interactions characterized by different  $\mathbf{K}$ 's. Thus they are treated dynamically only in a local sense, and in this, Takagi's theory is that of the perfect crystal applied to infinitesimally small mosaic blocks. But, from the derivation in § 5 it is seen that the size of blocks must have a physically meaningful minimum value. However, it is very difficult to define such a minimum unit, as is often the case in the mathematical transition from a discrete model to a continuum model.

As discussed previously by Kuriyama & Miyakawa (1970), the dynamical theory of diffraction for an imperfect crystal must properly deal with the phase modulation on diffracted beams, as a consequence of the presence of crystal imperfections. The approximations used in this paper have, in effect, replaced the phase modulation by a corresponding amplitude modulation. It is, therefore, not surprising that the approximate form of equation (2-9), that is, Takagi's equation, is of the same form as expected for dynamical diffraction in a perfect crystal. Naturally the approximations of amplitude modulation have a limited range of validity. In this approximation, the intensity of the diffracted beam diverges as the crystal becomes very imperfect (Köhler, Möhling & Peibst, 1970), while the correct treatment by phase modulation gives the intensity expected from the kinematical theory (Kuriyama & Miyakawa, 1969; 1970).

If a real crystal can be replaced by a model crystal which consists of local mosaic blocks, the solution of the more exact equation (2-9) will not be much different numerically from the solution of Takagi's equation, as understood from our derivation in the previous sections. In that case, his equation can probably be handled, analytically and numerically, more easily. If, however, solving his equation for an imperfect crystal requires as much effort as solving equation (2-9), then it is more desirable to solve equation (2-9) numerically, avoiding the approximation of poor resolution.

#### References

- AFANAS'EV, A. M. & KOHN, V. G. (1971). *Acta Cryst.* **A27**, 421.  
 ASHKIN, M. & KURIYAMA, M. (1966). *J. Phys. Soc. Japan*, **21**, 1549.  
 BALIBAR, F. & AUTHIER, A. (1967). *Phys. Stat. Sol.* **21**, 413.  
 BONSE, U. (1964). *Z. Phys.* **177**, 385.  
 CHUKHOVSKII, F. N. & SHTOLBERG, A. A. (1970). *Phys. Stat. Sol.* **41**, 815.  
 DEDERICHS, P. H. (1966). *Phys. Kondens. Mater.* **5**, 347.  
 DEDERICHS, P. H. (1967). *Phys. Stat. Sol.* **23**, 377.  
 FEYNMAN, R. P. (1951). *Phys. Rev.* **84**, 108.  
 HANNON, J. P. & TRAMMELL, G. T. (1968). *Phys. Rev.* **169**, 315.  
 HANNON, J. P. & TRAMMELL, G. T. (1969). *Phys. Rev.* **186**, 306.  
 HOWIE, A. & WHELAN, M. J. (1961). *Proc. Roy. Soc. A* **263**, 217.  
 KAMBE, K. (1965). *Z. Naturforsch.* **20a**, 770.  
 KATO, N. (1963a). *Acta Cryst.* **16**, 276.  
 KATO, N. (1963b). *Acta Cryst.* **16**, 282.  
 KATO, N. (1963c). *J. Phys. Soc. Japan*, **18**, 1785.  
 KATO, N. (1964a). *J. Phys. Soc. Japan*, **19**, 67.  
 KATO, N. (1964b). *J. Phys. Soc. Japan*, **19**, 971.  
 KÖHLER, R., MÖHLING, W. & PEIBST, H. (1970). *Phys. Stat. Sol.* **41**, 75.  
 KURIYAMA, M. (1967a). *J. Phys. Soc. Japan*, **23**, 1369.  
 KURIYAMA, M. (1967b). *Phys. Stat. Sol.* **24**, 743.  
 KURIYAMA, M. (1968). *J. Phys. Soc. Japan*, **25**, 846.  
 KURIYAMA, M. (1969). *Acta Cryst.* **A25**, 682.  
 KURIYAMA, M. (1970). *Acta Cryst.* **A26**, 56. (This is referred to as Paper I).  
 KURIYAMA, M. (1971). *Acta Cryst.* **A27**, 273.  
 KURIYAMA, M. & MIYAKAWA, T. (1969). *J. Appl. Phys.* **40**, 1697.  
 KURIYAMA, M. & MIYAKAWA, T. (1970). *Acta Cryst.* **A26**, 667.  
 OHTSUKI, Y. H. & YANAGAWA, S. (1966). *J. Phys. Soc. Japan*, **21**, 326, 502.  
 PENNING, P. & POLDER, D. (1961). *Philips Res. Rep.* **16**, 419.  
 PENNING, P. & POLDER, D. (1964). *Acta Cryst.* **17**, 950.  
 TAKAGI, S. (1962). *Acta Cryst.* **15**, 1311.  
 TAGAKI, S. (1969). *J. Phys. Soc. Japan*, **26**, 1239.  
 TAUPIN, D. (1964). *Bull. Soc. Franç. Miner. Crist.* **87**, 469.  
 WILKENS, M. (1966). *Phys. Stat. Sol.* **13**, 529.