affected. Since the factor is θ-dependent, failure to take account of it may have significant influence on temperature factors and extinction parameters derived from the measurement of a series of reflexions.

With the inbuilt flexibility of modern diffractometers, there is increased interest in the potential of this classical technique to measure structure factors of high precision. Establishment of such data on an absolute scale is only possible if all correction factors are recognized, assessed and included. In the past, the influence of surface-layer absorption on structure factors derived by this procedure has been largely overlooked.

In relation to this technique, Wooster & Macdonald (1948) have drawn attention to the trend of the two limiting values of intensity – that for the perfect crystal and that for the ideally imperfect crystal – to approach one another with increasing wavelength. They concluded that it is advantageous to use longer wavelengths for the determination of accurate structure factors. This region is obviously one in which inclusion of the correction factor for surface-layer absorption is essential.

I am most grateful to Mr. J. Cook, National Measurement Laboratory, CSIRO, Sydney, for supplying the boule of lithium fluoride.

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A Classical Derivation of the Dynamical Diffraction Equation for Imperfect Crystals Based on the Ewald–Laue–Bethe Theory

By Masao Kuriyama

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

(*Received* 31 *March* 1975; *accepted* 5 *May* 1975)

The basic equation of dynamical diffraction for imperfect crystals, which has been derived previously by a general dynamical theory of diffraction, is rederived classically based on the Ewald–Laue–Bethe concept, thus completing the formulation of a 'scattering matrix' theory. It is shown in this classical derivation that a series of assumptions is required at each stage of the mathematical formulation to allow it to proceed further. These assumptions are then viewed in terms of the general dynamical theory of diffraction, and found unnecessary in the rigorous formulation. This classical formulation provides a conceptual relation between the traditional Ewald–Laue–Bethe dynamical theory and the general dynamical theory, one that has been derived with the aid of quantum-field theory.

1. Introduction

A general dynamical diffraction theory for imperfect crystals has been formulated previously by use of a quantum-field theoretical treatment of scattering problems (Ashkin & Kuriyama, 1966; Kuriyama, 1967). This theory has succeeded in rigorously deriving a fundamental equation of dynamical diffraction in the momentum representation (Kuriyama, 1970, 1972) and a basic integral equation for topography in the spatial coordinate representation (Kuriyama & Early, 1974). In addition to rigor, another virtue of this theory is that the optical conditions are automatically included in the theory (for example, Ashkin & Kuriyama, 1966; Kuriyama, 1968*a*). This is particularly important because imperfect crystals lack periodic translational invariance; diffracted beams behave differently in their intensity and angular divergence, depending upon both the location of the incident beam on the crystal and the exit locations of the diffracted beams. This property of diffracted beams has made it possible to develop a new field of diffraction topography.

Much of the physics involved in diffraction from imperfect crystals has been explained in a series of papers based on this general dynamical theory for imperfect crystals (for example, Kuriyama, 1969). This theory is a 'scattering matrix' theory in the same sense as the many beam electron diffraction theory is. This equivalence has been demonstrated previously in an explicit form (Kuriyama, 1970). However, many have failed (for example, Kato, 1973) to recognize the direct equivalence between that general dynamical theory and the 'scattering matrix' theory, simply because the former has been derived *not* by the traditional method of dynamical diffraction theories, *but* by quantum mechanics. The purpose of this paper, therefore, is to demonstrate how one can derive the results of the general dynamical theory of diffraction for imperfect crystals from the traditional Ewald–Laue–Bethe dynamical theory of diffraction. In so doing, one cannot help noticing the necessity of many *ad hoc* assumptions which cannot be justified within the framework of the classical formulation.

These assumptions are to be justified only after the final results have been compared with experimental results. Since a series of assumptions has been introduced to obtain a final result, and moreover, since the result is given usually by numerical computations, it is not obvious whether or not those assumptions are really justified individually, even though they seem to be reasonable 'physically'. In contrast, most of these *ad hoc* assumptions are not required in the quantum-mechanical formulation. Certain mathematical restrictions on the classical derivation cannot be generalized because of mathematical difficulties. On the contrary, the general dynamical theory derived previously has proven that those restrictions are, in fact, not required.

The *ad hoc* assumptions and restrictions will be denoted in the text by Roman numerals, when they are introduced for the first time. These numbered assumptions and restrictions will later be discussed individually in comparison with the general dynamical diffraction theory. In the following classical formulation, the vector potential will be used to derive the wave equation of radiation fields for dynamical diffraction, unlike in the traditional derivation where electric or magnetic field strengths are used to represent the radiation fields.

2. A classical derivation of wave equations for radiation fields in a medium

In the following classical derivation, the basic principle is to reformulate the Ewald–Laue–Bethe theory for an imperfect crystal which does not possess periodic translational invariance. It is known that the wave equation for dynamical diffraction of electrons (or neutrons or other particles) is given in a form identical to the wave equation for dynamical diffraction of X-rays, except that the Coulomb crystal potential replaces the induced polarizability, and the Schrödinger equation is used instead of the Maxwell equations.* The radiation (X-rays) field, therefore, will be treated here as an example to obtain the wave equation for dynamical diffraction.

The radiation (electromagnetic) field is expressed in terms of the vector potential, **A**, by the electric and magnetic field strengths:

$$\mathbf{E} = -\frac{1}{c}\frac{\partial \mathbf{A}}{\partial t}, \quad \mathbf{H} = \nabla \times \mathbf{A}. \tag{2-1}$$

Since the radiation field is transverse, the radiation or Coulomb gauge is adopted:

$$\nabla . \mathbf{A} = 0. \tag{2-2}$$

Then the Maxwell equations lead to the following equation for the vector potential, **A**:

$$\Box \mathbf{A} = \frac{4\pi}{c}\mathbf{j}, \tag{2-3}$$

where

$$\Box = \frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \nabla^2.$$

The current density, **j**, induced in the medium by the radiation field is (I) assumed to be given by

$$\mathbf{j}(\mathbf{r}, t) = -\frac{e^2}{mc}\varrho(\mathbf{r})\mathbf{A}(\mathbf{r}, t). \tag{2-4}$$

Thus, (2-3) reduces to

$$[\Box - \phi(\mathbf{r})]\mathbf{A}(\mathbf{r}, t) = 0, \tag{2-5}$$

where

$$\phi(\mathbf{r}) = -\frac{4\pi e^2}{mc^2}\varrho(\mathbf{r}). \tag{2-6}$$

Since the medium (or crystal) does not have periodic translational invariance because of imperfections in it, the classical polarizability, $\phi(\mathbf{r})$, cannot be expanded into a Fourier series, in contrast with the polarizability in a perfect crystal. Under this condition, the polarizability (or crystal potential in the case of electrons) should (II) be described by a Fourier integral rather than by a Fourier series:

$$\phi(\mathbf{r}) = \int \frac{V d^3k}{(2\pi)^3}\phi(\mathbf{k}) \exp [i\mathbf{k}. \mathbf{r}], \tag{2-7}$$

where V is the crystal volume, and

$$\phi(\mathbf{k}) = \frac{1}{V}\int_{\text{all space}} d^3r\phi(\mathbf{r}) \exp [-i\mathbf{k} . \mathbf{r}]. \tag{2-8}$$

If the crystal (or medium) is perfect, then $\phi(\mathbf{k})$ is given by an integral within the unit cell:

$$\phi(\mathbf{k}) = -\frac{4\pi e^2}{mc^2}\frac{1}{v_a}\int_{\text{unit cell}} d^3r\varrho(\mathbf{r}) \exp [-i\mathbf{k} . \mathbf{r}] \tag{2-9}$$

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* Even though the Schrödinger equation is used to derive the wave equations for these particles, this derivation is considered to be classical, as opposed to the scattering formalism of contemporary quantum theory.

for a perfect crystal, as is known in the traditional dynamical theory.

Any function can be expanded by a complete set of eigenfunctions. The vector potential **A** is, therefore, written:

$$\mathbf{A}(\mathbf{r},t)=\int\frac{\mathrm{d}^3k}{(2\pi)^3}\int\frac{\mathrm{d}\omega}{2\pi}\sum_{\lambda}\ \boldsymbol{\varepsilon}(\mathbf{k},\lambda)u(\mathbf{k},\lambda)\exp\left[i\mathbf{k}\,.\,\mathbf{r}-i\omega t\right], \tag{2-10}$$

where $\boldsymbol{\varepsilon}(\mathbf{k},\lambda)$ represents the unit vector of the polarization direction of the plane wave propagating in the **k** direction. $u(\mathbf{k},\lambda)$ also depends on ω, but, for simplicity, this dependence is not explicitly written. Because of the transversality condition, (2-2), there are two independent directions specified by $\lambda=1$ and 2, which are perpendicular to each other and to the propagation direction, **k**. Substituting (2-7) and (2-10) into (2-5), multiplying by $\boldsymbol{\varepsilon}(\mathbf{k},\lambda)\exp\left[-i\mathbf{k}\,.\,\mathbf{r}+i\omega t\right]$ and integrating over all space and time, we obtain

$$[\mathbf{k}^2-(\omega/c)^2]u(\mathbf{k},\lambda)-\int\frac{V\mathrm{d}^3k'}{(2\pi)^3}\,\phi(\mathbf{k}-\mathbf{k}')\,[u(\mathbf{k}')]_{\mathbf{k},\,\lambda}=0, \tag{2-11}$$

where

$$[u(\mathbf{k}')]_{\mathbf{k},\,\lambda}=\sum_{\lambda'}\boldsymbol{\varepsilon}(\mathbf{k},\lambda)\,.\,\boldsymbol{\varepsilon}(\mathbf{k}'\lambda')u(\mathbf{k}'\lambda'). \tag{2-12a}$$

In the first term of (2-11), the relation

$$\boldsymbol{\varepsilon}(\mathbf{k},\lambda)\,.\,\boldsymbol{\varepsilon}(\mathbf{k},\lambda')=\delta_{\lambda\lambda'} \tag{2-13}$$

has been used. The polarization directions of the wave propagating in the $\mathbf{k}'$ direction can be chosen so that either the $\lambda'=1$ or $\lambda'=2$ direction may be perpendicular to the given polarization direction $\boldsymbol{\varepsilon}(\mathbf{k},\lambda)$ of the wave propagating in the **k** direction. Thus we can always express, in terms of the proper choice of the $\{\lambda'\}$ system,

$$\begin{aligned}[u(\mathbf{k}')]_{\mathbf{k},\,\lambda}&=\boldsymbol{\varepsilon}(\mathbf{k},\lambda)\,.\,\boldsymbol{\varepsilon}(\mathbf{k}'\lambda')u(\mathbf{k}'\lambda')\\&=P(\mathbf{k}\lambda;\mathbf{k}'\lambda')u(\mathbf{k}'\lambda'),\end{aligned} \tag{2-12b}$$

where λ' is either 1 or 2, depending upon which direction is not perpendicular to λ.

To proceed further at this stage, it is inevitable to introduce (III), a drastic assumption – a concept of lattice in an imperfect crystal. We assume that one can always assign to an imperfect crystal a perfect reference crystal whose reciprocal-lattice vector is denoted by **K**'s (**I**, **J** and **H** will also be used to represent a reciprocal-lattice vector). Then we can express a vector **k** in terms of **K** and the deviation from **K** as follows:

$$\mathbf{k}=\bar{\mathbf{k}}+\mathbf{K}+\mathbf{q}, \tag{2-14}$$

where **q** is restricted to within the reciprocal unit cell, and $\bar{\mathbf{k}}$ will be written hereafter as **k** without further confusion.* Thus the integral operation in (2-11) can be replaced by sums:

$$\int\frac{V\mathrm{d}^3k}{(2\pi)^3}\to\sum_{\mathbf{K}}\int'\frac{V\mathrm{d}^3q}{(2\pi)^3}\to\sum_{\mathbf{K}}\sum_{\mathbf{q}}, \tag{2-15}$$

where the prime mark on the integral sign indicates an integration within a reciprocal unit cell, and the summation over **q** is also limited to this volume.

To ensure elastic scattering in diffraction, it is necessary to introduce further (IV), the elastic scattering conditions:

$$(\omega/c)^2=|\mathbf{k}|^2\simeq|\mathbf{k}+\mathbf{K}+\mathbf{q}|^2 \tag{2-16}$$

and any vector **k** or $\mathbf{k}'$ can be constructed from **k** by adding **K** and **q**, namely

$$\left.\begin{aligned}&\mathbf{k}\to\mathbf{k}+\mathbf{K}+\mathbf{q}\\ \text{and}\qquad&\mathbf{k}'\to\mathbf{k}+\mathbf{K}'+\mathbf{q}'.\end{aligned}\right\} \tag{2-17}$$

We thus obtain classically the wave equation for dynamical diffraction of X-rays by substituting (2-16) and (2-17) into (2-11) and using (2-15):

$$\begin{aligned}&[(\mathbf{k}+\mathbf{K}+\mathbf{q})^2-\mathbf{k}^2]u(\mathbf{k}+\mathbf{K}+\mathbf{q};\lambda)\\&\quad-\sum_{\mathbf{K}'}\sum_{\mathbf{q}'}\phi(\mathbf{K}+\mathbf{q}-\mathbf{K}'-\mathbf{q}')P(\mathbf{K}+\mathbf{q},\lambda;\mathbf{K}'+\mathbf{q}',\lambda')\\&\qquad\times u(\mathbf{k}+\mathbf{K}'+\mathbf{q}';\lambda')=0.\end{aligned} \tag{2-18a}$$

The factor P is a polarization factor and λ' is chosen as discussed in (2-12b). For a single Bragg diffraction condition in a perfect crystal ($\mathbf{K}=\mathbf{0}$, **H** and $-\mathbf{H}$ are considered), (2-12b) yields, for instance,

$$P(\mathbf{k},1;\mathbf{k}+\mathbf{H},1)=\boldsymbol{\varepsilon}(\mathbf{k},1)\,.\,\boldsymbol{\varepsilon}(\mathbf{k}+\mathbf{H},1)=\cos 2\theta_B$$

and

$$P(k,2;\mathbf{k}+\mathbf{H},2)=\boldsymbol{\varepsilon}(\mathbf{k},2)\,.\,\boldsymbol{\varepsilon}(\mathbf{k}+\mathbf{H},2)=1.$$

Equation (2-18) can be written in a matrix form:

$$\mathsf{L}\,.\,\mathbf{u}=\mathbf{0}, \tag{2-18b}$$

where

$$\mathbf{u}=\begin{bmatrix}u(\mathbf{k}+\mathbf{K}+\mathbf{q},\lambda'')\\ \cdot\\ \cdot\\ \cdot\\ u(\mathbf{k},\lambda)\\ \cdot\\ \cdot\\ \cdot\\ u(\mathbf{k}+\mathbf{K}'+\mathbf{q}',\lambda')\end{bmatrix} \tag{2-19}$$

and

$$\begin{aligned}[\mathsf{L}]_{\mathbf{I}+\mathbf{q},\,\mathbf{J}+\mathbf{q}'}&=[(\mathbf{k}+\mathbf{I}+\mathbf{q})^2-\mathbf{k}^2-\phi(\mathbf{0})]\delta_{\mathbf{IJ}}\delta_{\mathbf{qq}'}\\&\quad-\phi(\mathbf{I}+\mathbf{q}-\mathbf{J}-\mathbf{q}')P(\mathbf{I}+\mathbf{q},\mathbf{J}+\mathbf{q}';\lambda')\\&\qquad\times(1-\delta_{\mathbf{qq}'}\delta_{\mathbf{IJ}}).\end{aligned} \tag{2-20}$$

For electrons, neutrons and other particles, the Schrödinger equation can be reduced to a form identical to (2-18), except for the polarization factors, to

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* The $\bar{\mathbf{k}}$ (later **k**) is still arbitrary at this stage. It will be defined physically by (2–16). See discussion (IV) in § 4.

give the wave equation for dynamical diffraction, if ϕ is understood as the crystal potential. We will, therefore, treat dynamical diffraction of all the particles and X-rays together hereafter. For simplicity, only a scalar field will be treated; for X-rays, a linearly polarized radiation is considered by suppressing $P(\mathbf{I}+\mathbf{q},\mathbf{J}+\mathbf{q}';\lambda')$.

Next we rewrite the diagonal elements of matrix L:

$$(\mathbf{k}+\mathbf{I}+\mathbf{q})^2-\mathbf{k}^2-\phi(0)\equiv(\mathbf{k}+\mathbf{I}+\mathbf{q})^2-\mathbf{Q}^2$$
$$=(\mathbf{Q}+\mathbf{I}+\mathbf{q})^2-\mathbf{Q}^2+2(\mathbf{Q}+\mathbf{I}+\mathbf{q})\,(\mathbf{k}-\mathbf{Q})+(\mathbf{k}-\mathbf{Q})^2, \tag{2-21}$$

where $\mathbf{Q}$ is a new vector whose magnitude is

$$|\mathbf{Q}|=Q=\sqrt{\mathbf{k}^2+\phi(0)},$$

and whose direction is defined to be parallel to $\mathbf{k}$. Here we introduce (V), another assumption, that the last term in (2-21), $(\mathbf{k}-\mathbf{Q})^2$, can be discarded. Then we can write

$$\mathsf{L}=\mathsf{M}_0+2Qz\Theta^2, \tag{2-22}$$

where

$$[\mathsf{M}_0]_{\mathbf{I}+\mathbf{q},\mathbf{J}+\mathbf{q}}=[(\mathbf{Q}+\mathbf{I}+\mathbf{q})^2-\mathbf{Q}^2]\delta_{\mathbf{IJ}}\delta_{\mathbf{qq}'}$$
$$-\phi(\mathbf{I}+\mathbf{q}-\mathbf{J}-\mathbf{q})\,(1-\delta_{\mathbf{qq}'}\delta_{\mathbf{IJ}}), \tag{2-23}$$

$$[\Theta]_{\mathbf{I}+\mathbf{q},\,\mathbf{J}+\mathbf{q}}=(\cos\theta_{\mathbf{I}+\mathbf{q}})^{1/2}\delta_{\mathbf{IJ}}\delta_{\mathbf{qq}'}, \tag{2-24}$$

and

$$z=|\mathbf{k}-\mathbf{Q}|. \tag{2-25}$$

The angle between $\mathbf{Q}$ and $\mathbf{Q}+\mathbf{I}+\mathbf{q}$ is denoted by $\theta_{\mathbf{I}+\mathbf{q}}$. Equation (2-22) can be rewritten, for convenience,

$$\mathsf{L}=\Theta[\Theta^{-1}\mathsf{M}_0\Theta^{-1}+2Qz\mathbf{I}]\Theta. \tag{2-26}$$

The matrices used here can be considered also as super-matrices. For example, the $(\mathbf{I},\mathbf{J})$ element of the super-matrix L is given by a matrix whose elements are specified by $(\mathbf{q},\mathbf{q}')$. In this notation, the element of a super-matrix L is written $[\mathsf{L}]^{\mathbf{q};\mathbf{q}'}_{\mathbf{I},\mathbf{J}}$, instead of $[\mathsf{L}]_{\mathbf{I}+\mathbf{q},\,\mathbf{J}+\mathbf{q}'}$.

Hereafter all the matrices are super-matrices.

3. Niehrs's solution for the Laue geometry and its integral expression

To solve (2-18) for given diffraction conditions, we need boundary conditions which define the geometric relation between the crystal and the incident beam. In this section, the solution of (2-18) will be (VI) sought under the boundary condition in the Laue geometry.

Niehrs (1965) obtained the solution in this geometry expressed in terms of the present notation in the following form:

$$[\mathbf{u}^{(i)}]_{\mathbf{K}+\mathbf{q}}=[\mathsf{A}(z_i)]^{\mathbf{q};\mathbf{o}}_{\mathbf{K},\mathbf{0}}\Big/\left[\frac{\partial}{\partial z}\det\mathsf{L}(z)\right]_{z=z_i}, \tag{3-1}$$

where z_i is the ith solution of $\det\mathsf{L}(z)=0$, and corresponds to the ith mode of the diffracted waves, and the matrix A is the adjoint of the matrix L. The (n,m) element of A is the algebraic complement, or the cofactor, of the (m,n) element of L. Consequently, the outgoing wave in a particular direction outside of the crystal is given by a superposition of individual modes whose amplitude is determined by (3-1). We can, therefore, consider the following function as the outgoing wave propagating in the $\mathbf{K}+\mathbf{q}$ direction:

$$\psi_{\mathbf{K}+\mathbf{q}}(\sigma_K)=\sum_i[u^{(i)}]_{\mathbf{K}+\mathbf{q}}f(z_i;\sigma_K), \tag{3-2}$$

where $f(z_i;\sigma_K)$ is an analytic function representing the wave state of each mode, and σ_K is a coordinate along the $\mathbf{K}+\mathbf{q}$ propagation direction.* The functions, $f(z_i;\sigma)$, are analytic in the domain of z, including z_i's. Then, it immediately follows from (3-1) and (3-2) that

$$\psi_{\mathbf{K}+\mathbf{q}}(\sigma)=\sum_i\frac{[\mathsf{A}(z_i)]^{\mathbf{q};\mathbf{o}}_{\mathbf{K},\mathbf{0}}}{\left[\frac{\partial}{\partial z}\det\mathsf{L}(z)\right]_{z=z_i}}f(z_i;\sigma)$$
$$=\oint\frac{[\mathsf{A}(z)]^{\mathbf{q};\mathbf{o}}_{\mathbf{K},\mathbf{0}}}{\det\mathsf{L}(z)}f(z;\sigma)\frac{\mathrm{d}z}{2\pi i}, \tag{3-3}$$

provided (VII) that all the characteristic values of $\mathsf{L}(z)$ are distinct: $z_i\neq z_j$. It is guaranteed by (2-20) or (2-26) that $\mathsf{A}(z)$ is analytic in the domain of this integral.

The inverse matrix of L is given in terms of its cofactors and, in turn, its adjoint matrix A by

$$\mathsf{L}^{-1}=\frac{\mathsf{A}}{\det\mathsf{L}}. \tag{3-4}$$

Equation (3-3) thus reduces to

$$\psi_{\mathbf{K}+\mathbf{q}}(\sigma)=\oint\frac{\mathrm{d}z}{2\pi i}[\mathsf{L}^{-1}(z)]^{\mathbf{q};\mathbf{o}}_{\mathbf{K},\mathbf{0}}f(z;\sigma)$$
$$=\int_{-\infty}^{+\infty}\frac{\mathrm{d}z}{2\pi i}[\mathsf{L}^{-1}(z)]^{\mathbf{q};\mathbf{o}}_{\mathbf{K},\mathbf{0}}f(z;\sigma), \tag{3-5}$$

where $f(z,\sigma)$ vanishes in the limit of large z's. From (2-26), we obtain

$$\mathsf{L}^{-1}(z)=\Theta^{-1}[\Theta^{-1}\mathsf{M}_0\Theta^{-1}+2Qz\mathbf{I}]^{-1}\Theta^{-1}. \tag{3-6}$$

Substituting this expression into (3-5), we obtain in the matrix form:

$$[\psi(\sigma)]^{\mathbf{q};\mathbf{o}}_{\mathbf{K},\mathbf{0}}=\frac{1}{2Q}$$
$$\times\left[\Theta^{-1}\int_{-\infty}^{+\infty}\frac{\mathrm{d}z}{2\pi i}\{\Theta^{-1}\mathsf{M}_0\Theta^{-1}+z\mathbf{I}\}^{-1}f\left(\frac{z}{2Q};\sigma\right)\Theta^{-1}\right]^{\mathbf{q},\mathbf{o}}_{\mathbf{K},\mathbf{0}} \tag{3-7}$$
$$=\frac{1}{2Q}\left[\Theta^{-1}f\left(-\frac{1}{2Q}\Theta^{-1}\mathsf{M}_0\Theta^{-1};\sigma\right)\Theta^{-1}\right]^{\mathbf{q},\mathbf{o}}_{\mathbf{K},\mathbf{0}},$$

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* The σ_K is merely a mathematical parameter. Its physical interpretation is arbitrary, indicating a shortcoming of the classical theory. See discussion (VIII) in § 4.

where the $(\mathbf{K},\mathbf{0};\mathbf{q},\mathbf{0})$ element of $\psi(\sigma)$ is given by $\psi_{\mathbf{K}+\mathbf{q}}(\sigma)$.

If we are (VIII) interested in a plane-wave state, then the function f is given by an exponential. Thus (3-7) reduces to the result obtained previously by a general dynamical diffraction theory (Kiruyama, 1970):

$$\begin{aligned}\psi(\sigma) &= \frac{1}{2Q}\,\Theta^{-1}\exp\left[-\frac{i\sigma}{2Q}\,\Theta^{-1}\mathsf{M}_0\,\Theta^{-1}\right]\Theta^{-1}\\ &= \frac{1}{2Q}\,\Theta^{-2}\exp\left[-\frac{i\sigma}{2Q}\,\mathsf{M}_0\Theta^{-2}\right],\end{aligned} \tag{3-8}$$

which is the scattering amplitude of the outgoing waves denoted by the S matrix in (2-30) in a previous paper (Kuriyama, 1970). Thus we can arrive classically at the basic dynamical diffraction equation for imperfect crystals:

$$\frac{\mathrm{d}\mathsf{S}}{\mathrm{d}\sigma}=(-i)\mathsf{MS}\,, \tag{3-9}$$

where

$$\mathsf{S}=2Q\Theta^2\psi \tag{3-10}$$

and

$$\mathsf{M}=\mathsf{M}_0\Theta^{-2}. \tag{3-11}$$

4. Comparison of the classical derivation with the general dynamical diffraction theory

It has been demonstrated in the two previous sections that a classical formulation based on the Ewald–Laue–Bethe concept can produce the fundamental equation of dynamical diffraction for imperfect crystals, thus recreating the results of the general dynamical diffraction theory previously derived by a quantum-field theoretical treatment. It has also been proven that the general dynamical diffraction theory is, in fact, a 'scattering matrix' theory,* even though it was formulated quantum mechanically. Once the fundamental equation is obtained, as shown in (3-9), in the momentum representation, all the previous results obtained by the general dynamical diffraction theory will be immediately reproduced. A general extinction theory described previously by Kuriyama & Miyakawa (1969, 1970) can be obtained by a perturbation method from (3-9). Both the 'ray' and the 'wave' theories can be reconstructed exactly in the same way as shown before (Kuriyama, 1972, 1973). The basic equation for topography (Kuriyama & Early, 1974) will be also obtained immediately, if proper optical conditions are added to the classical derivation.

Although the classical derivation has led to results equivalent to those from the quantum-field theoretical formulation, there is basic deficiency in the classical derivation; it requires a series of assumptions and restrictions one after another before that derivation yields the fundamental dynamical diffraction equation, (3-9). Those assumptions were numbered by Roman numerals, when they were introduced in the text for the first time. In the following it will be demonstrated item by item that these assumptions are not needed, but can rather be derived as theoretical consequences by the process of mathematical manipulation, if more rigorous formulation, such as a quantum-field theoretical treatment, is adopted from the outset.

(I) It is known that the use of second quantization or quantum-field theory makes it possible to write the electron current operator as a derivative of the Lagrangian density with respect to the photon field operator. The correct form of the current operator has been given by Ashkin & Kuriyama (1966) for X-ray diffraction, where the current operator includes part of the current that creates the Kramers–Heisenberg dispersion terms (Kuriyama, 1971).

(II) As Kuriyama & Miyakawa (1970) have discussed previously, the quantum-field theoretical technique for scattering problems, which deals with propagation of particles in matter, demands that polarizability should be a non-local function and hence represented by a double Fourier integral† (Kuriyama, 1967). In particular, if one ignores the Kramers–Heisenberg term in the polarizability, the double Fourier integral reduces to a single Fourier integral because of the presence of a delta function in the polarizability (Kuriyama, 1967).

(III) The above discussion, in itself, leads to the production of diffracted beams, not only in the Bragg-diffracted (by a perfect crystal) direction, but in any direction (in the extreme case). The dynamically diffracted waves in imperfect crystals cannot be described by Bloch waves. Only the correct calculation of the polarizability in imperfect crystals can interpret $\mathbf{J}$ and $\mathbf{q}$ properly through the geometrical structure factor (Kuriyama, 1967, 1968*b*):

$$g(\xi)=\frac{1}{N}\sum_n \exp\left[-i\xi\,.\,\mathbf{R}_n\right], \tag{4-1}$$

where N is the total number of atoms in the crystal, $\mathbf{R}_n$ is the position of the nth atom and ξ is a vector in the momentum space, eventually associated with $\mathbf{J}$ and $\mathbf{q}$. In terms of this geometrical factor, one can naturally introduce the concept of 'perfect reference crystal' without an assumption. The geometrical structure factor has maxima at values which are multiples of a set of vectors. These vectors can be identified as the basis vectors associated with the perfect reference crystal in the following way. Suppose that we know the positions of all the atoms in the crystal by use of an ideal lattice theory. Then we would know that the atomic spacings are close to a set

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* The words 'scattering matrix theory' are used here in the sense that Kato (1973) meant, although the correct terminology should belong to quantum mechanics.

† Since the non-local function contains at least two four-vector variables, it involves two (double) Fourier transforms with respect to these two four-vector spaces.

of constant values, which forms a unit cell. The lattice made up of these unit cells can be referred to as the perfect reference crystal in which we can define a reciprocal lattice and its basis vectors. We assign this physical meaning (the reciprocal-lattice vectors) to the vector $\mathbf{J}$. Thus, $\mathbf{q}$ is thought of as a vector describing the deviation from a reciprocal-lattice point, being similar to diffuse scattering.

(IV) The condition of elastic scattering cannot be introduced classically, because elastic scattering and inelastic scattering are successfully defined only by quantum-mechanical concepts. As shown by Ashkin & Kuriyama (1966) and Kuriyama (1971), the Lehmann, Symanzik & Zimmermann (1955, 1957) (LSZ) formulation (Low, 1955: Goldberger, 1955) or its equivalent can deal with the scattering condition in conjunction with boundary conditions. Energy conservation leads to elastic scattering, and momentum conservation leads to the Bragg diffraction condition. The momentum $\mathbf{k}$ in (2-14) represents all possible modes allowed inside the crystal; the specific set of $\mathbf{k}$'s (and, in turn, $\bar{\mathbf{k}}$) are not determined until the incident beam is brought into the crystal. In the classical derivation, this can be done by the boundary conditions with the elastic scattering assumption. In the LSZ formulation, this process is automatically contained. The arbitrariness of $\bar{\mathbf{k}}$ without the assumption of elastic scattering indicates another shortcoming of the classical theory.

(V) This assumption has been justified by the argument presented previously in terms of backward scattering (for example, Kuriyama, 1972). In the correct formulation, the scattering amplitude can be calculated without this assumption.

(VI) to (VIII) If one had started with the LSZ formulation, all of these assumptions and mathematical restrictions would not be required, as seen from previous papers (Kuriyama, 1967, 1970).

(VI) The Laue geometry was merely adopted for mathematical simplicity in derivation of the fundamental diffraction equation for imperfect crystals in previous papers (Kuriyama, 1970, 1972). However, in the present classical derivation, one could not derive the desired result without making use of Niehrs's work in the Laue geometry.

(VII) In the derivation of equation (3-3), the restriction that the characteristic values be distinct is mathematically indispensable. In contrast, the LSZ formula has yielded an integral expression essentially identical to (3-5) and, in turn, to (3-3) as the starting equation for the scattering amplitude without any restrictions (Ashkin & Kuriyama, 1966; Kuriyama, 1970).

(VIII) The function f has been introduced in (3-2) without any physical meaning and role. However, this quantity has a clear physical meaning in the LSZ formulation, as shown previously (Ashkin & Kuriyama 1966; Kuriyama, 1967). It should be emphasized that (3-5) is a classical counterpart of the LSZ formula

$$S=\iint \mathrm{d}^4x_1\mathrm{d}^4x_2[\Box_1\Box_2 D(x_1,x_2) - \Box_1\delta(x_1-x_2)]f^*(x_1)f(x_2), \quad (4\text{-}2)$$

which has been derived for dynamical diffraction by Ashkin & Kuriyama (1966). In this formulation, the functions f and f^* are well defined so that the optics in diffraction is explicitly contained in (4-2). There is no arbitrariness concerning the meaning of σ_K. This is another merit of the quantum-mechanical formulation.

As mentioned in the *Introduction*, the basic principle in a dynamical diffraction theory for imperfect crystals is the lack of periodic translational invariance. It is this lack also that necessitates the inclusion of the beam and detection optics at the onset of the rigorous theoretical formulation. With the aid of quantum-field theoretical technique, we can formulate a theory of dynamical diffraction complying with the two important requirements: no invariance under periodic translations and the inclusion of the beam optics. These two requirements are essential in diffraction topography (or microscopy). In contrast, the classical derivation fails to include the optics as a natural consequence, as seen in this paper.

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