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Dynamical Diffraction Equations for Imperfect Crystals

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Dedicated to Professor Dr. G. Borrmann on his 65th birthday

The ray theory of Kato and Kambe for imperfect crystals is derived in a formal way from a general dynamical theory of diffraction. This development together with the results from a previous paper concerning Takagi's equation (the wave theory) helps to clarify the meaning and limits of various phenomenological theories that have been extended to an imperfect crystal from the dynamical diffraction theory for a perfect crystal.

1. Introduction

In this past decade, there have appeared a number of dynamical theories of diffraction for imperfect crystals. These theories can be grouped into three categories; namely the *ray* (classical particle) theory, the *wave* theory and the *quantum* theory. Penning and Polder¹, Kato², Bonse³ and Kambe⁴ developed the *ray* theory, while the *wave* theory was derived by Takagi⁵ and Taupin⁶ in a phenomenological approach from the traditional Ewald-Laue-Bethe dynamical diffraction theory for perfect crystals. The *quantum* theory was formulated by Ashkin, Miyakawa and Kuriyama^{7,8}. In contrast to the two former theories, the *quantum* theory is capable of dealing with the dual property of particles such as photons and electrons.

It is rather interesting to learn from the history of quantum physics that the necessity and motivation for the development of these three theories resemble those found in the development of quantum physics. Therefore, it should not be surprising if the relationship among these three theories turns out to be very much similar to the well-known relations among Newtonian physics, de Broglie's wave mechanics and quantum physics.

A major complication in the dynamical diffraction problem for imperfect crystals is caused by the lack of periodic spatial translational invariance in the dynamic system of interest. Unlike dynamical diffraction in perfect crystals, there is no easy way to reduce the number of degrees of freedom which the dynamic system possesses for an imperfect crystal. The number of degrees of freedom in this system is equal to the number of atoms in the imperfect crystal; more precisely, the number of different displacements of the atoms comprising the crystal. In such a dynamic system, the equation of motion for scattered particles (or beams) consists of a set of many interacting equations, the number of which equals the number of degrees of freedom of this system. In other words, diffraction in the imperfect crystal is the consequence of interactions between many beams existing simultaneously. In reality, this implies that the diffracted beams (including the transmitted direction) exhibit line broadening around the propagation directions expected for a perfect crystal. Therefore, solving dynamical diffraction problems for imperfect crystals creates the same difficulty as solving many body problems familiar in solid state physics.



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Since various techniques have been developed for solving many body problems in modern physics, it is best to formulate a dynamical diffraction theory for imperfect crystals in a method commonly used in contemporary physics. Such an approach has produced the *quantum* theory mentioned previously.

The principle of approaching a *single* many-body problem is to reduce the problem to many single-body problems. In so doing, a single-body problem may turn out to be a tractable physical problem of a *fictitious* body (or particle). This is the method of normal coordinates in classical mechanics, and is known as second quantization in quantum mechanics, or the canonical transformation in the many-body problem.

In a previous paper⁸ (Paper II), we have demonstrated that the *quantum* theory reduces to the *wave* theory if we accept a certain set of approximations. The objective of this paper, therefore, is to study the relation between the *quantum* theory and the *ray* theory.

2. The Scattering Amplitude for Topography

In the scattering formalism of modern quantum mechanics, one deals with the exact quantum states of the system composed of the incoming particles and the scatterers. These states are called Heisenberg states which are given by the exact solution of the Schrödinger equation for the total Hamiltonian of the system. Dynamical variables such as field operators, therefore, obey the Heisenberg equation of motion. Before the incoming particles strike the scatterers, the exact Heisenberg state is called the *in* state, which implies that the incoming particles are free and the scatterers (electrons in the crystal) occupy energy levels below the Fermi energy. This condition for the scatterers defines the ground state of the crystal. After the particles are scattered out of the crystal, one can define an *out* state in a similar fashion. The *out* state represents the Heisenberg state in which the scattered particles are again free and the crystal has returned to its ground state.

When the crystal diffracts the incoming particles and a detector receives the scattered particles, the state of the system changes from the *in* state to the *out* state. Therefore, there is a quantum transition from the *in* state to the *out* state associated with the diffraction phenomenon^{7,9}. The scattering amplitude of the system that is equivalent to the diffract-

ed amplitude of the outgoing particles is, in terms of the *in* to *out* transition, given^{7,8} by

$$\langle \mathbf{k}' \mathbf{R}'; \text{out} | \mathbf{k}, \mathbf{R}; \text{in} \rangle \quad (2.1)$$

$$= \int d^3p \int d^3p' A^*(\mathbf{k}', \mathbf{p}'; \mathbf{R}') S(\mathbf{p}', \mathbf{p}) A(\mathbf{k}, \mathbf{p}; \mathbf{R}),$$

where the incoming particles of momentum \mathbf{k} strike the crystal in its ground state at position \mathbf{R} and the particles come out at position \mathbf{R}' with momentum \mathbf{k}' leaving the crystal in its ground state. The function A describes the momentum and the energy distribution of the incoming particles, and A^* is a function which characterizes the momentum and energy response of a detector. The quantity $S(\mathbf{p}', \mathbf{p})$ is the scattering matrix element of the crystal.

The scattering amplitude has been derived in a compact form in Paper I⁸:

$$S(\mathbf{p}', \mathbf{p}) = (|p_z|/|\mathbf{p}|) \delta(|\mathbf{p}'| - |\mathbf{p}|) \quad (2.2)$$

$$\sum_{\mathbf{q}} \sum_{\mathbf{J}} \delta(\mathbf{p}_t + \mathbf{J}_t + \mathbf{q}_t - \mathbf{p}_t') [\mathbf{S}]_{\mathbf{0}, \mathbf{J}}^{\mathbf{q}, \mathbf{q}'},$$

where the subscripts *t* and *z* designate the tangential and normal components of a vector relative to the crystal surface. The \mathbf{S} matrix is a super matrix, i. e., a matrix of matrices. The (\mathbf{I}, \mathbf{J}) element of a super matrix is given by a matrix whose element are specified by $(\mathbf{q}, \mathbf{q}')$. In this notation, a super matrix \mathbf{S} is written $[\mathbf{S}]_{\mathbf{I}, \mathbf{J}}^{\mathbf{q}, \mathbf{q}'}$. Expressing the matrix as a super matrix makes it possible to identify the subscript, \mathbf{I} or \mathbf{J} , with a reciprocal lattice vector defined for the perfect reference crystal. The perfect reference crystal is an imaginary crystal in which all atoms of a given type are located at ideally periodic lattice sites. Consequently, the superscript, \mathbf{q} or \mathbf{q}' , describes the deviation of the out-going particles from ideal Bragg diffraction directions.

The matrix \mathbf{S} satisfies the dynamical diffraction equation as shown in Paper II:

$$d\mathbf{S}(\sigma_f)/d\sigma_f = i \mathbf{M} \cdot \mathbf{S}(\sigma_f) \quad (2.3)$$

with the initial condition $\mathbf{S}(0) = \mathbf{I}$. In this equation, the only independent variable is σ_f , which is a mathematical coordinate along the propagation direction. The propagation direction of the transmitted beam is specified by $f=0$, and that of the \mathbf{K} -diffracted beam by $f=\mathbf{K}$.

The quantity \mathbf{M} is related to the generalized polarizability of an imperfect crystal and also contains the kinetic energy term, as described in detail in Paper II.

One of the practical applications of dynamical diffraction is topography of imperfections within

crystals. In this application, one observes the diffracted intensity distribution at the crystal surface where most of diffracted beams emerge. In the Laue geometry, this surface is called the exit surface.

For topography, one is not terribly concerned about the angular distribution of the out-going beams, except for their general Bragg diffraction directions, for example, a transmitted direction and an \mathbf{H} -Bragg diffraction direction. This condition allows us to introduce the following spectral distribution functions, A and A^* , described in Paper II:

$$A(\mathbf{k}, \mathbf{p}) = \begin{cases} 1, & \text{if } \mathbf{k} = \mathbf{p}, \\ 0, & \text{otherwise,} \end{cases} \quad (2.4)$$

$$A^*(\mathbf{k}', \mathbf{p}') = A^*(\mathbf{k}', \mathbf{k}_K' + \mathbf{q}) \quad (2.5)$$

$$= \begin{cases} 1, & \text{if } \mathbf{K} = \mathbf{O} \text{ or } \mathbf{H}, \\ 0, & \text{otherwise,} \end{cases} \quad \left. \begin{array}{l} \text{regardless of the} \\ \text{value of } \mathbf{q}. \end{array} \right\}$$

This implies that the geometrical resolution for the momenta of scattered particles is insufficient to resolve individual scattered beams around a diffraction spot.

If we specify the momentum of the out-going particle under observation by

$$\mathbf{k}_K' = \mathbf{k} + \mathbf{K} + \mathbf{q}_F \equiv \mathbf{k}_K + \mathbf{q}_F, \quad (2.6)$$

the scattering amplitude (2.2) reduces to

$$\langle \mathbf{k}_K', \mathbf{R}' ; \text{out} | \mathbf{k}, \mathbf{R} ; \text{in} \rangle = (k_z/k_{K,z}) \sum_q [\mathbf{S}(\mathbf{k}_K'; \sigma_K)]_{\delta, \mathbf{k}}^{q, q_F}. \quad (2.7)$$

One can always set q_F equal to zero or replace q_F by a distribution of q 's if one wishes (see Paper II). In diffraction spectroscopy where the precise momentum distribution of the scattered particles should be determined as well, the scattering amplitude for topography (2.7) is merely an approximation to the true one. Care must also be taken with the topographs taken at some distance from the exit surface of a crystal containing many severe imperfections, because of a geometrical enhancement of the broadened beam.

3. A Canonical Transformation (the Ray Theory)

To obtain the scattering amplitude for topography (2.7), the matrix \mathbf{S} is needed. Therefore, solving the dynamical diffraction problem is now equivalent to solving Equation (2.3). A possible way of handling Eq. (2.3) has been presented in Paper II, in

which the equation is replaced by a set of *local* differential equations accompanied by an assumption of a *local condition*. This local condition has led to the concept of local reciprocal lattice vectors. Paper II has thus reconstructed the *wave* theory from the generalized scattering theory (the *quantum* theory).

In this section a different approach will be discussed. But first, suppose that the matrix \mathbf{M} in Eq. (2.3) is diagonalized. Then this equation can be represented by a set of differential equations, each of which can be solved independently. Unfortunately, the matrix \mathbf{M} does not have such a simple form.

Let us introduce a unitary transformation which transforms \mathbf{S} to $\tilde{\mathbf{S}}$,

$$\tilde{\mathbf{S}}(\sigma_K) = \exp\{i \mathbf{W}(\sigma_K)\} \mathbf{S}(\sigma_K), \quad (3.1)$$

where, for the sake of simplicity, the matrix \mathbf{W} is assumed to be a diagonal matrix,

$$(\mathbf{W})_{q, q'} = W_q(\sigma_K) \delta_{qq'}. \quad (3.2)$$

Then Eq. (2.3) takes the form

$$d\tilde{\mathbf{S}}(\sigma_K)/d\sigma_K = i \mathbf{K}(\sigma_K) \tilde{\mathbf{S}}(\sigma_K), \quad (3.3)$$

where

$$\mathbf{K}(\sigma_K) = \frac{d\mathbf{W}(\sigma_K)}{d\sigma_K} + \exp\{i \mathbf{W}(\sigma_K)\} \mathbf{M} \cdot \exp\{-i \mathbf{W}(\sigma_K)\} \quad (3.4 a)$$

or

$$[\mathbf{K}(\sigma_K)]_{qq'} = (dW_q(\sigma_K)/d\sigma_K) \delta_{qq'} + \exp\{i[W_q(\sigma_K) - W_{q'}(\sigma_K)]\} [\mathbf{M}]_{qq'}. \quad (3.4 b)$$

The next task is to find the condition on W_q that renders \mathbf{K} diagonal. In order to diagonalize \mathbf{K} , one usually sets up the secular equation for \mathbf{K} :

$$|\mathbf{K} - \lambda \mathbf{I}| = 0. \quad (3.5 a)$$

Suppose that Eq. (3.5 a) is solved with respect to the λ 's; then one can write Eq. (3.5 a) in the equivalent form

$$\lambda_q = H(\partial W_q / \partial \sigma_K, \sigma_K; t) \equiv -\partial W_q / \partial t, \quad (3.5 b)$$

where we have introduced an extra arbitrary variable t , and $(d/d\sigma_K)$ has been replaced by $(\partial/\partial\sigma_K)$ since σ_K is no longer a single independent variable in this expression. Equation (3.5 b) is a differential equation for W_q , through which the canonical transformation (3.1) makes \mathbf{K} diagonal.

Before proceeding, we should remember that the dynamical diffraction Eq. (2.3) is merely a mathe-

matical equation which has been devised to calculate the matrix elements of \mathbf{S} in the scattering amplitude. Hence, the σ_K has been merely a mathematical variable and has not been given any physical meaning (see Paper II). The natural next step is to see if we can give a physical meaning to it in a formal way. In any physical system, there always are two sets of basic variables such as the coordinates and their canonically conjugate momenta (or the field operators and their conjugate) which describe motion of a physical particle (or determine the entire field). In this sense, if we can identify σ_K as, say, a coordinate, then we can consider that solving Eq. (2.3) is equivalent to solving for the motion of a "fictitious" particle (a quasi-particle) whose coordinate σ_K and its conjugate momentum satisfy some equation of motion.

Let us define the following function using the function H in (3.5 b) :

$$L \equiv \frac{d\sigma_K}{dt} \frac{\partial W_q}{\partial \sigma_K} - H\left(\frac{\partial W_q}{\partial \sigma_K}, \sigma_K; t\right). \quad (3.6)$$

It is imperative, if σ_K is to be the coordinate relating to the motion of a quasi-particle, that the variational principle apply to a function such as L . Let us apply the principle to L :

$$\delta \int_{t_0}^{t_1} L dt \equiv 0. \quad (3.7)$$

Then using partial integration and setting the variations $\delta\sigma_K(t_0)$ and $\delta\sigma_K(t_1)$ equal to zero, we obtain

$$\int_{t_0}^{t_1} \left[\left\{ \frac{d\sigma_K}{dt} - \frac{\partial H}{\partial (\partial W_q / \partial \sigma_K)} \right\} \delta \left(\frac{\partial W_q}{\partial \sigma_K} \right) - \left\{ \frac{d}{dt} \left(\frac{\partial W_q}{\partial \sigma_K} \right) + \frac{\partial H}{\partial \sigma_K} \right\} \delta \sigma_K \right] dt \equiv 0. \quad (3.8)$$

This has to be identically zero regardless of the variations $\delta(\partial W_q / \partial \sigma_K)$ and $\delta\sigma_K$. We, therefore, obtain

$$\begin{aligned} d\sigma_K/dt &= \partial H / \partial (\partial W_q / \partial \sigma_K) \\ \frac{d}{dt} \left(\frac{\partial W_q}{\partial \sigma_K} \right) &= - \frac{\partial H}{\partial \sigma_K}. \end{aligned} \quad (3.9)$$

These are Hamilton's canonical equations if the variable $\partial W_q / \partial \sigma_K$ is identified as the momentum canonically conjugate to σ_K .

It is therefore concluded that, when the canonical transformation (3.1) assumes its convenient form for our problem, it leads to the concept of quasi-

particles whose coordinates and momenta are given by σ_K and $\partial W_q / \partial \sigma_K$, respectively. Within this context, the functions, H , L and W , are, in effect, the Hamiltonian, the Lagrangian and Hamilton's principal function (Eikonal) for the quasi-particles, respectively. And Eq. (3.5 b) is, then, the Hamilton-Jacobi equation for the quasi-particles.

We have so far demonstrated that the calculation of the dynamical scattering amplitude can be simulated by the motion of the quasi-particles (rays) which satisfies the equation of motion given by Equation (3.9). Along the paths (trajectories) of these particles, Eq. (3.3) becomes diagonal. We have therefore created the *ray* theory from the *quantum* theory.

4. Discussion

The discussion in the previous section was rather formal and general. Emphasis was placed on the theoretical relation between the *ray* theory and a general dynamical scattering theory. Practical methods of solving Eq. (3.3) with the proper form of the canonical transformation have deliberately not been discussed. Ideally speaking, once one finds the trajectories of quasi-particles with different q 's by solving Hamilton's canonical Eq. (3.9), one should solve Eq. (3.3) with a diagonal matrix \mathbf{K} along each trajectory corresponding to different q 's. Then, knowing the explicit form of the canonical transformation, one can transform $\tilde{\mathbf{S}}$ back to \mathbf{S} to obtain the desired scattering amplitude for topography (2.7) by integrating over different q 's. Truthfully, this approach is just as difficult as solving the dynamical diffraction Eq. (2.3) by direct methods. However, the discussion in Section 3 has served the purpose of understanding the rigorous formulation of dynamical scattering in terms of classical concepts of diffracted ray trajectories.

Naturally there is a problem associated with practical benefits and limitations of the ray theory. In order to demonstrate it, we consider as an example the traditional two-wave (beam) approximation for an imperfect crystal. It should be noted here that this approximation when applied to imperfect crystals is no longer of a dynamical nature in the strict sense; only pairs of many possible beams in the crystal can be treated dynamically. In other words, the \mathbf{k} -beam and the $\mathbf{k} + \mathbf{H} + \mathbf{q}$ -beam with a specific \mathbf{q} interact dynamically, while the

$\mathbf{k} + \mathbf{H} + \mathbf{q}$ and $\mathbf{k} + \mathbf{H} + \mathbf{q}'$ beams may not. In the terminology of the local reciprocal lattice, this statement implies that the two-beams, \mathbf{k} and $\mathbf{k} + \mathbf{H}(r)$, are considered to be under dynamical diffraction (see Paper II).

Here we apply the two-wave approximation with the understanding described above. The canonical transformation of interest then functions only to diagonalize the matrix \mathbf{K} with respect to \mathbf{q} and \mathbf{q}' defined in Section 2, not with respect to \mathbf{I} and \mathbf{J} . From this point on, the subscripts q on W in (3.2)

really become identical to the q 's defined in Section 2. Equation (3.3) then gives a set of equations for the \mathbf{O} and \mathbf{H} beams just as for a perfect crystal. The Hamilton-Jacobi Eq. (3.5 b) now takes the form of the well-known dispersion equation for the two-wave case. Using the Hamiltonian defined by this equation, we can derive Hamilton's canonical equation in the two-wave approximation. This equation gives the "relativistic" equation for the rays as Kato² and Kambe⁴ derived previously.

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A Comparison of Diffuse Scattering by Defects Measured in Anomalous Transmission and Near Bragg Reflections*

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The diffuse scattering from neutron-irradiation-produced defects in copper was measured in anomalous transmission and near the 111 Bragg reflection. The results were related through the theory of Dederichs, and the intensities and forms of the scattering curves obtained from the two measurements were found to be in good agreement over a wide range of defect concentrations and sizes. It was shown that the intercept of a plot of the diffuse scattering intensity $I^s(q_0)$ vs $\ln(q_0)$ is at $q_0 = e^{1/2}/R$ for defects of uniform radius R and at $q_0 \cong 1/R_0 e$ for an exponential size distribution of average size R_0 .

Introduction

One of the consequences of the discovery of the anomalous (Borrmann) transmission of X-rays through nearly perfect crystals has been a renewed interest in the use of X-ray scattering for the investigation of defects in crystals. The decrease in anomalous transmission resulting from defects has been studied for impurities in silicon¹ and germanium², for defect clusters produced by neutron ir-

radiation in copper³, and for interstitials in copper⁴. This decrease in transmission results from the lattice strains introduced by the defects and the theory for describing this effect has been developed by Dederichs⁵, and we⁶ have shown the theory to be phenomenologically correct for neutron irradiated copper. The decrease in anomalous transmission is described in terms of an effective absorption coefficient μ^* which is the sum of two components, μ_{PE} due to enhanced photoelectric absorp-

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