Primary and Secondary Extinctions in the Dynamical Theory for an Imperfect Crystal

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Primary and secondary extinction are studied using the dynamical theory of X-rays diffracted by imperfect crystals. The transition from dynamical to kinematical scattering is explained in terms of fundamental processes in diffraction. Contrary to existing extinction theories, where the intensities diffracted dynamically by single coherent domains of a mosaic are combined using an *ad hoc* assumption of mosaic distributions, the present theory permits the dynamical *amplitudes* to change in response to disturbances of the dynamical interactions by imperfections. Neither the mosaic block model nor the statistical treatment of imperfections is used. The extinction of diffracted intensities is thereby treated as caused solely by *inhomogeneous* strains in a single coherent domain.

1. Introduction

The extinction of diffracted X-rays within a single and ideally perfect crystal block is called primary extinction (Darwin, 1914*a*,*b*; James 1954). A decrease in primary extinction is described by the fact that a crystal becomes slightly less perfect, without using the mosaic block concept for representing such an imperfect crystal. A crystal, which is neither ideally perfect nor ideally imperfect, diffracts X-rays with an intensity lying somewhere between the two values expected from the dynamical and from the kinematical theory of diffraction. This phenomenon is caused by secondary extinction (Darwin, 1914*a*, *b*; James, 1954).

To understand secondary extinction phenomenologically, it has been necessary so far to replace a crystal by mosaic blocks. Each block slightly misoriented mutually is assumed to be a perfect crystal which may have lattice constants different from those in other blocks. In real crystals where most imperfections are dislocations, defects, and impurities, it is the existence of inhomogeneous strains in the crystal that really affects the Bragg diffraction processes. Mosaic block models are too primitive to approximate real situations. It is also well known that primary and secondary extinction in general are inseparable in real crystals. The purpose of this paper is to study some fundamental aspects of extinction in X-ray diffraction from a new viewpoint, one based on a recently formulated dynamical theory of X-rays diffracted by imperfect crystals (Kuriyama, 1967; 1970). In the present treatment, primary and secondary extinctions always coexist, as one expects in real crystals. Since a decrease in extinction must be described by physical quantities characterizing imperfections, the mosaic block concept is not used to represent an imperfect crystal.

Much of the physics involved in diffraction from an imperfect crystal has been discussed in a paper by

one of the authors (Kuriyama, 1969). However, the change diffracted intensities from the dynamical to the kinematical value has been only discussed qualitatively. The present paper provides a quantitative discussion on this subject.

2. Scattering amplitude of an imperfect crystal

In the scattering formalism of modern quantum mechanics, the scattering amplitude for diffracted X-rays is given by the probability amplitude of a photon in the wave packet state $|\mathbf{k}, v; \mathbf{R}; \text{in}\rangle$ to make a transition to the state $|\mathbf{k}',v',\mathbf{R}'; \text{out}\rangle$, where $\mathbf{k}$ is momentum, $\omega = |\mathbf{k}| = 2\pi/\lambda$ is energy (with $c = \hbar = 1$), v the polarization direction of the photon, $\mathbf{R}$ is the center position of the X-ray beam on the crystal, and the primes indicate those quantities for outgoing waves (Ashkin & Kuriyama, 1966). When calculated for an imperfect crystal* (Kuriyama, 1967, 1970), the scattering amplitude, $\langle \mathbf{k}'v'\mathbf{R}'; \text{out} \mid \mathbf{k}v\mathbf{R}; \text{in}\rangle \equiv \langle \mathbf{k}'\mathbf{R}'|S|\mathbf{k}\mathbf{R}\rangle$ (with v and v' suppressed), consists of four terms (Kuriyama, 1969). The first term is essentially equivalent in form to the electric field intensity of radiation outside the crystal in the ordinary dynamical theory for an *absorbing* perfect crystal, while the other terms do not have a counterpart in the ordinary dynamical theory. For convenience we group these other three terms into one and call them the extinction term. This term vanishes when the crystal is perfect.

The extinction term cannot be derived rigorously by the ordinary dynamical theory or its extension. It is for this reason that a correct treatment by a modern

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* In the Appendix, an explicit expression of the scattering amplitude is given of a crystal undergoing an ideal thickness vibration. It is almost similar in form to that of a crystal having unspecified imperfections. To follow the discussion in this section, it may be helpful to refer to that expression.

scattering formalism has been required for an imperfect crystal. In an imperfect crystal the periodic spatial translational invariance does not hold, that is $f(\mathbf{r}) \neq f(\mathbf{r}+\mathbf{l})$, where $\mathbf{l}$ is the lattice vector. Since the ordinary dynamical theory requires this invariance, it is obvious that any attempted extension of the theory fails to produce the extinction term rigorously. In other words, the polarizability of the crystal, which is to the lowest order proportional to the charge distribution, $\varrho(r)$, cannot be represented by a single Fourier series:

$$\varrho(\mathbf{r}) = \sum_{\mathbf{K}} \varrho(\mathbf{K}) \exp (i \mathbf{K} \cdot \mathbf{r}),$$

where $\mathbf{K}$ is 2π times a reciprocal lattice vector. It should be at least represented by a single Fourier integral. However, the modern technique for scattering problems, which deals with propagation of particles in matter, demands that the polarizability should be a non-local function and represented by a double Fourier integral (Kuriyama, 1967). This leads to the production of diffracted beams, not only in the Bragg-diffracted (by a perfect crystal) direction but in any direction. These dynamically diffracted waves cannot be described by a Bloch wave function, contrary to the treatment in ordinary dynamical theory.

One might think, however, that for slightly distorted crystals the single Fourier series expansion of the polarizability would still be a good approximation if the Fourier coefficients were modified. In the modified expansion the 'Fourier' coefficients would be given by a slowly varying function of the position. Such a modified Fourier expansion would approximate the change in polarizability by an amplitude modulation; it could not provide any phase modulation. However, in a real imperfect crystal which does not have periodic translational invariance, the (classical) polarizability must be expressed by a Fourier integral; that is, the Fourier coefficients, $\varrho(\mathbf{k})$, exist not only for $\mathbf{k}=\mathbf{K}$, but for any arbitrary $\mathbf{k}$. It is clear, therefore, that the polarizability of an imperfect crystal should be considered that of the perfect crystal, but with phase modulation. As we know from a Taylor expansion of such a phase modulated function, an approximation of phase modulation by amplitude modulation is not only poor, but is sometimes unable to reproduce real physical situations. This argument provides the principal reason why an extension of ordinary dynamical theory is not sufficient to produce the extinction term rigorously (especially when distortions become large).

Next we turn to the physical meaning of the first term and the extinction term. Imperfections affect the first term in two ways (Kuriyama, 1969): the one is to change the effective absorption coefficient (here we consider the Laue geometry), and the other is to modify the Bragg diffracted propagators, which may be considered classically to be the electric field intensity of radiation in matter. The latter modification, in effect, causes narrowing of the dynamical diffraction range (Kuriyama, 1969). This means that the Bragg

diffracted propagators behave like nearly free field propagators (as seen in the kinematical scattering) resulting in an increase of the 'integrated' intensity contributed only from the first term (with the change in the effective absorption coefficient neglected). The modified Bragg diffracted propagator therefore causes a decrease in primary extinction as the crystal becomes less perfect. However, the first term in the scattering amplitude is not the sole term for primary extinction. Since the Bragg diffracted propagator is a basic quantity describing how a photon propagates in the material when the Bragg conditions are nearly satisfied, it appears also in the extinction term.

When the crystal becomes far less perfect, the extinction term becomes dominant, and therefore must be closely connected to secondary extinction. If one wishes to use the dispersion surface diagram, the extinction term involves a transition (jump) of a tie point from one branch (mode) to another. The frequency of the transition is determined by a set of actual (displaced) atomic positions in the crystal volume irradiated by the incident X-ray beam and by the modified Bragg diffracted propagators associated with the tie points on both branches (modes). The extinction term as well as the first term have been obtained in terms of atomic displacements in the crystal (Kuriyama, 1967, 1968, 1969). Since atomic displacements vary depending upon the types of imperfections, it is not easy to calculate scattering amplitudes for various types of imperfections.

The Fourier transforms of the atomic displacements can be uniquely determined for various types and arrangements of imperfections [see for instance, Mura (1964) for the continuum theory, Matsubara (1952) and Krivoglaz (1959) for the atomistic theory]. As discussed above, crystal imperfection essentially causes phase modulations in the generalized polarizability of the crystal. One might therefore conjecture that in diffraction problems it would be best to deal with atomic displacements by means of Fourier transforms. In fact, the fundamental processes in diffraction can
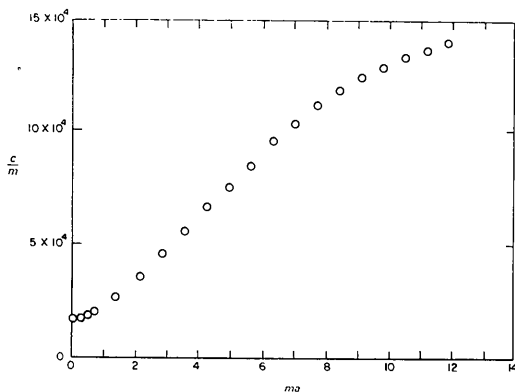

Fig. 1. Change of the diffracted intensity (counts.min⁻¹) as a function of the driving electric current (mA) for piezoelectric vibrations (Haruta, 1967).

be treated in terms of such Fourier transforms regardless of the specific type of imperfections (Kuriyama, 1969).

There still remain some complications in studying the properties of the scattering amplitude in practice. These are: (1) the complex functional form of the Fourier transforms for real imperfections, (2) the finite incident beam size which emphasizes the local behavior of the imperfections (Kuriyama, 1968, 1969) and (3) the superposition of different modes in the Fourier expansion of the atomic displacements. If these complications could be somehow eliminated without consequent loss of generality, the fundamental role of the extinction term would be clarified. We next consider one physical problem which provides a possibility for eliminating such complications.

3. Basic diffraction processes for extinction

For the purpose of studying the phase modulation produced by inhomogeneous strains, such as those associated with imperfections, we shall derive in this section the extinction effects produced by piezoelectric vibrations. In these vibrations the atoms are displaced from their ideal positions in a well-defined and controllable fashion. It is then expected that the intensity of X-rays diffracted by the crystal undergoing piezoelectric vibrations will change as a function of the vibrational amplitude. Recently, Haruta (1967) showed experimentally how the diffracted intensity changed as the driving electric current for piezoelectric vibrations increased. His result is reproduced in Fig. 1. This is a manifestation of the secondary extinction in diffraction.

Because the change in intensity can be studied quantitatively as a function of controllable lattice distortions, X-ray diffraction from a vibrating crystal allows a quantitative study of extinction. Throughout this paper an ideal thickness vibration is treated explicitly. The displacements of the atoms are given by

$$\mathbf{u}(l_z) = \mathbf{A_q} \sin (\mathbf{q} . \mathbf{l} - \Phi_q) \qquad (3 \cdot 1)$$

with

$$\mathbf{q}_t = 0 , \qquad (3 \cdot 2)$$

where the z axis is in the direction of the thickness L, $\mathbf{l}$ is a lattice vector in a perfect crystal, and a subscript t indicates the projection onto the X-ray entrance surface of the crystal. In so doing, the atomic displacements become independent of the lateral coordinates in a crystal plate of infinite lateral dimension. In this vibration diffraction is independent of the entry point of the incident X-rays. It is therefore sufficient to consider a plane wave incident on the crystal. The scattering amplitude is now given by $\langle \mathbf{k}'|S|\mathbf{k}\rangle$ without R and R'.

It is convenient in the following discussion to express some quantities necessary for the scattering amplitude in terms of the experimental variables which define the incident angle and the observation angles with reference to the Bragg angle. A reciprocal lattice point $\mathbf{H}$ is very close to the Ewald sphere. For the photons in the initial state (the incoming X-rays) the deviation from the Bragg condition is determined by the variable

$$2\varepsilon(\mathbf{k}) = \mathbf{k}^2 - (\mathbf{k} + \mathbf{H})^2 + (1 - \tau)v(0) \qquad (3 \cdot 3)$$

$$\simeq 2|\mathbf{k}|^2 \sin 2\theta_B \Delta\theta_i + (1 - \tau)v(0) , \qquad (3 \cdot 4)$$

where $\tau = 1 + (H_z/k_z)$ is a factor determining the asymmetrical geometry of reflection, $v(0)$ is related to the complex refractive index of the crystal, and $\Delta\theta_i$ is the angular deviation of the incident X-ray beam from the Bragg angle θ_B defined with respect to the diffracting plane.

In a similar way the deviation from the Bragg peak angle for the final states is determined by

$$2\eta(\bar{\mathbf{k}}) = \bar{\mathbf{k}}^2 - (\bar{\mathbf{k}} + \mathbf{H})^2 + (1 - \tau)v(0) \qquad (3 \cdot 5)$$

$$\simeq 2|\bar{\mathbf{k}}|^2 \sin 2\theta_B \Delta\theta_f + (1 - \tau)v(0) , \qquad (3 \cdot 6)$$

where, for convenience,

$$\mathbf{k}' = \bar{\mathbf{k}} + \mathbf{H} , \qquad (3 \cdot 7)$$

and $\Delta\theta_f$ is the angular deviation of the observation direction from the Bragg angle. Using the variable (3·4) for the initial state we obtain the solutions, α, to a dispersion equation for the initial state*

$$\alpha_i - k_z = \frac{1}{2k_z} [v(0) + \frac{1}{\tau} \{\varepsilon(\mathbf{k}) + (-1)^i R(\mathbf{k})\}] , \qquad (3 \cdot 8)$$

where

$$R(\mathbf{k}) = \sqrt{\{\varepsilon(\mathbf{k})\}^2 + \tau[J_0(\mathbf{H} . \mathbf{A_q})]^2 v(\overline{+\mathbf{H}})v(\overline{-\mathbf{H}})} \qquad (3 \cdot 9)$$

and the mode with $i = 1$ is the anomalous transmission mode while the mode with $i = 2$ is the anomalous (strong) absorption mode. The solutions, β, to the dispersion equation in the final states (outgoing X-rays) is given by the same form as (3·8) with β, $\bar{\mathbf{k}}$ and $\eta(\bar{\mathbf{k}})$ replacing α, $\mathbf{k}$ and $\varepsilon(\mathbf{k})$ respectively.

The scattering amplitude always contains the energy and the momentum conservation rules, the latter of which is sometimes implicit (Kuriyama, 1967). When the crystal is undergoing an ideal thickness vibration ($\mathbf{q}_t = 0$), the conservation rules are given by the two delta functions, one of which manifests the energy conservation rule while the other gives the same unmodified Bragg condition as expected in a perfect crystal (Kuriyama & Miyakawa, 1969). The momenta of the photons in the final states must satisfy the relation

$$\mathbf{k}'_t = \mathbf{k}_t + \mathbf{K}_t , \qquad (3 \cdot 10)$$

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* According to the usual terminology the dispersion equations define the possible X-ray modes in the unbounded crystal. Their solution is the set of wave-vectors from an arbitrary tie point on the surface of dispersion to the reciprocal lattice points. However, what we call here a dispersion equation for a specific state contains the selection of solutions of this set brought in by the existence of a crystal surface and an incident beam.

with $K=0$ and H to give nonvanishing scattering amplitude. Comparing equation (3·10) with (3·7) we obtain

$$\bar{k}=k \quad \text{(for an ideal thickness vibration).} \quad (3\cdot11)$$

It then follows that

$$\eta(\bar{k})=\varepsilon(k) , \quad (3\cdot12)$$

and

$$\alpha_{i=1}=\beta_{j=1}; \ \alpha_{i=2}=\beta_{j=2} . \quad (3\cdot13)$$

From the scattering amplitude of a crystal undergoing the ideal thickness vibration (Kuriyama & Miyakawa, 1969), the diffracted (or the transmitted) intensity for an incoming plane wave is obtained (see Appendix):

$$\sum_i |S^{(0)}(i)|^2+|S^{(1)}(1,2)|^2+|S^{(1)}(2,1)|^2$$

$$+\text{Interference terms,} \quad (3\cdot14)$$

where

$$|S^{(1)}(i,j)|^2=L^2|\sigma_{ij}(0)v(\pm H)|^2$$

$$\times [\{\cos (H . A_q)-J_0(H . A_q)\}^2 \left\{ \frac{\varepsilon(k)}{R(k)} \right\}^2$$

$$+ \sin^2 (H . A_q)]| F_{H-K}|^2 . \exp [-2\text{Im}\{\alpha_j-k_z\}L] . \quad (3\cdot15)$$

Here $v(K)$ is the Fourier transform of modified polarizability and J_0 is the Bessel function of zero order. The exact form of σ_{ij} will be given by (3·22) The quantities F_K are called the dynamical field functions which are obtained from the modified Bragg diffracted propagators. The explicit forms are written

$$F_O(k_t;i)= \frac{1}{2} \left[1-(-1)^i \frac{\varepsilon(k)}{R(k)} \right] \quad (3\cdot16)$$

$$F_H(k_t;i)= \frac{(-1)^i}{2} \frac{J_0(H . A_q)v(+H)}{R(k)} . \quad (3\cdot17)$$

In equation (3·15) the explicit expression is given only to the terms which are expected to be important for the extinction effect. It is worth noting that $S^{(1)}(1,2)$ gives the same value as $S^{(1)}(2,1)$ if the sign of ε is changed, except for the dependence of the effective absorption coefficients, $2\text{Im}\{\alpha_j\}$, on the ε value.

In numerical calculations of the intensity the dimensionless quantity x is chosen as a variable instead of the $\varepsilon(k)$:

$$x=[\tau v(+H)v(-H)]^{-1/2} \varepsilon(k) . \quad (3\cdot18)$$

Another variable $H . A_q$, instead of A_q alone, is denoted by y and used to represent the degree of crystal imperfection.* Since $|S^{(1)}(1,2)|^2$ and $|S^{(1)}(2,1)|^2$ are complementary, either one of them, when studied as a func-

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* The word 'imperfection' here refers to the amount of inhomogeneous strain in the crystal lattice and is not to be confused with topological 'lattice imperfections' such as vacancies and dislocations, which however can be detected through their (inhomogeneous) strain fields.

tion of those two variables, is sufficient to give a picture of the intensity behavior. We also omit the interference terms in the present discussion.

The quantities $v(0)$ and $v(\pm H)$, being complex, produce the imaginary parts of the dynamical field functions. It is due to the presence of absorption that the values of $v(0)$ and $v(\pm H)$ are complex, and consequently topographic images taken from imperfect
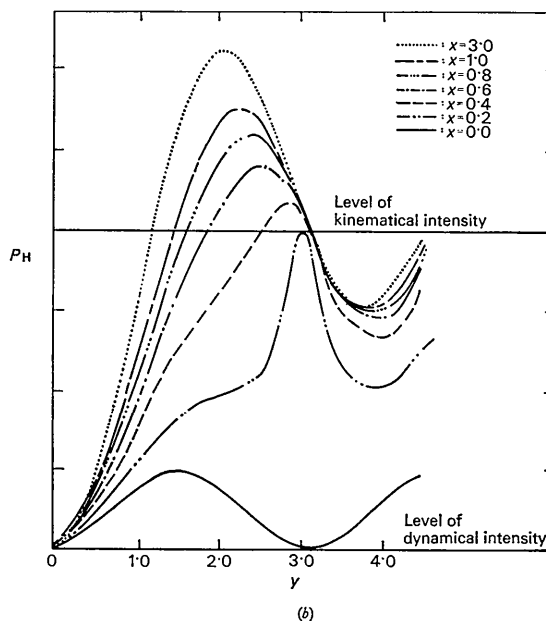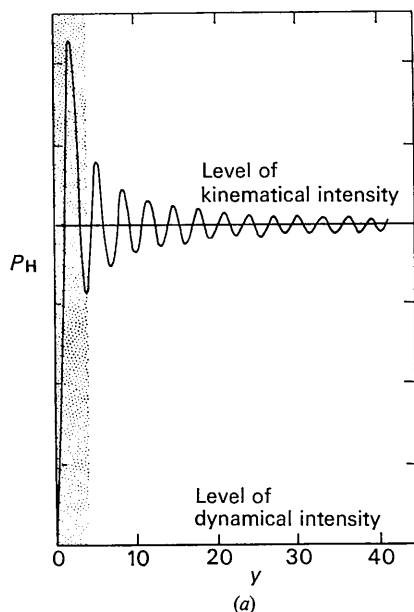

Fig. 2. (a) The overall behavior of the diffracted intensity P_H, in the Bragg diffracted direction, as a function of the degree of crystal imperfection. For this curve the incident angle of the X-rays is off the exact Bragg angle. (b) Enlargement of the shaded area in Fig. 2(a) for various values of the incident angle.

crystals become complicated (Kuriyama, 1969). In this paper, however, the imaginary parts of those functions are neglected except for the effective absorption coefficients appearing in the exponential function in (3·15). It follows from equation (3·15) that P_O, the power in the transmitted direction, is given by

$$P_0 = W(x,y) \frac{[J_0(y)]^2}{x^2 + [J_0(y)]^2} \qquad (3·19)$$

and P_H, the power in the Bragg diffracted direction, by

$$P_H = W(x,y) \left[1 + \frac{x}{\sqrt{x^2 + [J_0(y)]^2}} \right]^2, \qquad (3.20)$$

where

$$W(x,y) = \{\cos y - J_0(y)\}^2 \frac{x^2}{x^2 + [J_0(y)]^2} + \sin^2 y. \qquad (3·21)$$

The $|S^{(0)}|^2$ terms have been excluded in the expressions of P_0 and P_H. The effective absorption coefficient, $\text{Im}[\alpha]$, is also a function of x and y. In the above expressions, however, the damping factor due to absorption has been omitted.

At first glance it is found that P_0 becomes smaller with increasing y, while P_H remains at a large value. This implies that the crystal distortion effect is more prominent in the Bragg diffracted direction than in the transmitted direction. Therefore, the quantity P_H is first studied as a function of x and y. To look at the overall behavior of P_H in the wide range of the degree of imperfection (y), the value of P_H is plotted as a function of y in Fig. 2(a), where the value of x is so
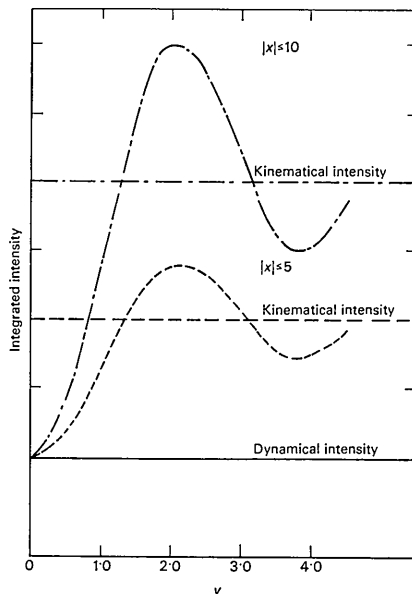

Fig. 3. Integrated intensity in the Bragg diffracted direction as a function of the degree of crystal imperfection. Two different ranges of x are chosen; the shapes of these two curves are almost identical, regardless of the ranges selected. Two levels of the kinematical intensity are shown, corresponding to the selected ranges of x. The dynamical intensity level is added schematically.

chosen that the incident angle of the X-ray is slightly off the exact Bragg angle. The value of P_H starts from zero and, though oscillating, reaches the value expected from the kinematical theory of diffraction. It should be remembered here that the P_H is only a part of the resultant intensity, although it becomes dominant when y increases. Therefore, the resultant intensity starts at the dynamical value for a perfect crystal and reaches the kinematical value as the degree of imperfection increases. The dynamical intensity term $|S^{(0)}(i)|^2$ also changes as a function of y.

The shaded area in Fig. 2(a) is enlarged to give Fig. 2(b), where the curves of P_H are shown for various values of x(the incident angle). All the curves increase like a squared sinusoidal function when y is small, and except for every small x, oscillate around the kinematical value when y increases. These curves would explain the intensity change which might be observed in an idealized experiment where the incident X-rays were given by an ideal plane wave having no intrinsic momentum dispersion. In practice, however, momentum dispersion in the incoming wave is inescapable for optical reasons, being of the almost same magnitude as the range of dynamical diffraction. It may be, therefore, reasonable to obtain the intensity integrated over a certain range of incident angles. The value of P_H integrated over two different ranges of x are plotted as a function of y in Fig. 3. The shapes of these two curves are almost identical, regardless of the ranges selected. Therefore, the curves in Fig. 3 should be compared with the experimental result. In this Figure the level of the dynamical integrated intensity is added schematically to the integrated value of P_H to give the whole feature in intensity. The oscillatory behavior in the integrated intensity will be discussed in the next section.

Fig. 4 shows the values of P_0 integrated over the same two regions of x as for the integrated P_H. The vertical scale is arbitrary in the Figures. If one uses the same scale for both Figures, the maximum value of the integrated P_0 is almost negligible compared to the maximum value of the integrated P_H. The integrated P_0 quickly approaches zero as y increases. This again confirms that the imperfection effect appears more prominently in the Bragg diffracted beam than in the transmitted beam.

The emphasis so far has been on the dynamical diffraction processes for extinction. In studying rocking curves, that is, the behavior of the resultant intensity as a function of x, another aspect must be taken into consideration. When y is very small and the crystal therefore nearly perfect, the rocking curve width is determined mainly by the dynamical terms $|S^{(0)}(i)|^2$ as expected. However, when y increases and the crystal becomes less perfect, the terms $|S^{(1)}(i,j)|^2$ dominate to determine the resultant rocking curve width. Since the Bragg diffracted propagator behaves like a free propagator as y increases, P_H does not decrease to zero, but remains unchanged with increasing x. In such a case the important factor in determining the rocking

curve for an imperfect crystal is the geometrical factor $\sigma_{ij}(0)$, which has been assumed to be independent of the value of q_z.

The quantity $\sigma_{ij}(0)$ is the approximate representation of $\sigma_{ij}(n)$ which has originally been defined (Kuriyama & Miyakawa, 1969) by

$$\sigma_{ij}(n) = [N(z)]^{-1} \sum_{l}^{N(z)-1} \exp\left[-i\{\alpha_j(\bar{\mathbf{k}}_t) - \alpha_i(\mathbf{k}_t)\}l_z\right]$$
$$\times \exp\left[-inq_z l_z\right], \quad (3\cdot22)$$

where $N(z)$ is the total number of unit cells in the z direction, l_z is the position of the lth cell measured from the X-ray entry surface of the crystal, and n is an integer. If it were not for q_z or n, the equation (3·22) would only be the size function and vanish unless $\alpha_j(\bar{\mathbf{k}}_t)$ is equal to $\alpha_i(\mathbf{k}_t)$, since $N(z)$ is quite large. However, in the present problem where the crystal is imperfect, the equation (3·22) is not given by a simple function, but becomes a function which is familiar in the kinematical line broadening problems of distorted crystals.

If $\sigma_{ij}(n)$'s are used instead of $\sigma_{ij}(0)$ to derive the more rigorous expression for (3·15), the quantities $\sin(\mathbf{H}.\mathbf{A}_q)$ and $\cos(\mathbf{H}.\mathbf{A}_q)$ in (3·15) are replaced by the lattice sum

$$[N(z)]^{-1} \sum_{l_z} \exp\left[-i\{\alpha_j(\bar{\mathbf{k}}_t) - \alpha_i(\mathbf{k}_t)\}l_z\right]$$
$$\left\{{\sin \atop \cos}\right\} (\mathbf{H}.\mathbf{A}_q \cos q_z l_z). \quad (3\cdot23)$$

This determines the effective range of $\alpha_j(\bar{\mathbf{k}}_t) - \alpha_i(\mathbf{k}_t)$, even for a set of modes for which $i \neq j$, to give non-vanishing values for (3·23). In the ideal thickness vibration, where $\bar{\mathbf{k}}_t = \mathbf{k}_t$, this effective range determines the range of x, that is the range of the incident angle, over which the resultant intensity does not vanish. In other words, this range is the rocking curve width. As y increases the profile of the rocking curve is determined by the product of the value of (3·23) and P_H as a function of x (the incident angle).

4. Discussion

Primary and secondary extinctions have been studied as a consequence of disruption of the dynamic interaction processes in diffraction. Neither the mosaic block models nor the statistical treatments of imperfections have been used. Unlike the existing extinction theory (Darwin, 1914a,b; Zachariasen, 1945; James, 1954) where intensity transfer through independent mosaics is the basic process, the present treatment permits dynamical amplitudes to change in response to disturbances in dynamical interactions by imperfections. The extinction in diffracted intensities as treated in this paper is thus caused solely by inhomogeneous strains in a single coherent domain.

We have idealized crystal imperfections by considering the example of thickness vibrations. This idealization is undoubtedly too narrow. There are many other modes required to describe real crystal

imperfections; especially the modes for which $q_t \neq 0$ cannot be neglected. Although the fundamental processes in diffraction for the $q_t \neq 0$ mode are well decribed in a similar way to the case treated in the present paper, the resultant intensities are expected to be smoother than those in the Figures. The simultaneous existence of various q_t values give uncertainty to the value of y as well as $\bar{\mathbf{k}}_t$ (final states). Therefore the quantity to be compared with experimental results should be resultant intensity integrated over the uncertainty ranges of y and $\bar{\mathbf{k}}_t$. This procedure certainly smears out the oscillatory behavior observed in the Figures, giving a curve similar to that presented in Fig. 1. The resultant intensity in the Bragg diffracted direction starts at the dynamical value, increases like sine squared of y, the imperfection parameter, and finally reaches the kinematical value. In view of the results of the present discussion, a qualitative statement in a previous paper (§5g, Kuriyama, 1969) should be modified. The diffracted intensity may exceed the kinematical value (about 1·5 times) before settling down at the kinematical value.

APPENDIX

The scattering amplitude of a crystal undergoing an ideal thickness vibration is given (Kuriyama & Miyakawa, 1969) by

$$S(\mathbf{k}',\mathbf{k}) = (2\omega_k)^{-1}\delta(\omega_k - \omega_{k'})\delta(\mathbf{k}_t + \mathbf{K}_t - \mathbf{k}'_t)$$
$$\times[\sum_i S^{(0)}(i) + \sum_{(i \neq j)} S^{(1)}(i,j)], \quad (A1)$$
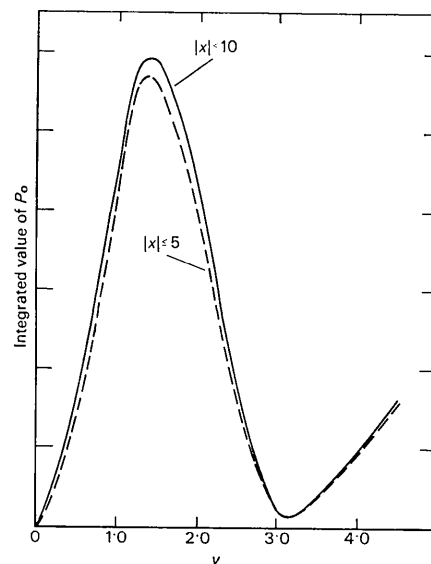

Fig. 4. Integrated values of P_0 (in the transmitted direction) as a function of the degree of crystal imperfection. Two different ranges of x are chosen.

where

$$S^{(0)}(i) = [1 + 2iLU(\mathbf{H})](\alpha_i + K_z + '_z)F_{\mathbf{K}}(\mathbf{k}_t; i)$$
$$\times \exp [iL(\alpha_i + K_z - k'_z)] \quad (A2a)$$

and

$$S^{(1)}(i,j) = iLV(\mathbf{H})F_{\mathbf{H}-\mathbf{K}}(\mathbf{k}_t; i) \exp [iL(\alpha_j + K_z - k'_z)] . \quad (A2b)$$

The symbols used in the above equations are

$$U(\mathbf{H}) = (\alpha_i + k_z)^{-1} v(-\mathbf{H})[\cos (\mathbf{H} . \mathbf{A}_q)$$
$$- J_0(\mathbf{H} . \mathbf{A}_q)]F_{\mathbf{H}}(\mathbf{k}_t; i) \quad (A3)$$

and

$$V(\mathbf{H}) = \sigma_{ij}(0)v(-\mathbf{H}\delta_{\mathbf{K}0} + \mathbf{H}\delta_{\mathbf{H}\mathbf{K}})[f_+(\mathbf{K}; i,j)$$
$$\{\cos (\mathbf{H} . \mathbf{A}_q) - J_0(\mathbf{H} . \mathbf{A}_q)\}$$
$$- if(\mathbf{K}; i,j) \sin (\mathbf{H} . \mathbf{A}_q)] . \quad (A4)$$

In these equations $\mathbf{K}$ takes on the values of $\mathbf{0}$ and $\mathbf{H}$ (a single Bragg reflection condition), and $F_{\mathbf{K}}(\mathbf{k}_t; i)$ are the corrected dynamical field functions in the initial state defined by (3·16) and (3·17). Other notations except $f_\pm$ have been defined in the text.

The important quantities are $f_\pm(\mathbf{K}; i,j)$ describing the degree of disruption of dynamical interactions. They are defined (Kuriyama, 1969; Kuriyama & Miyakawa, 1969) by

$$f_\pm(0; i,j) = (\beta_j + \bar{k}_z)[\Omega(\alpha_i + H_z) \pm \Omega(\beta_j + K_z)]/\Delta(\beta_j) \quad (A5)$$

$$f_\pm(\mathbf{H}; i,j) = (\beta_j + H_z + \bar{k}_z + H_z)[\Omega(\beta_i) \pm \Omega(\alpha_j)]/\Delta(\beta_i), \quad (A6)$$

where

$$\Omega(\alpha_i) = \frac{1}{\tau} [\varepsilon(\mathbf{k}) + (-1)^i R(\mathbf{k})] , \quad (A7)$$

$$\Omega(\alpha_i + H_z) = - [\varepsilon(\mathbf{k}) - (-1)^i R(\mathbf{k})] , \quad (A8)$$

$$\Delta(\alpha_i) = (2\alpha_i)(-1)^i 2R(\mathbf{k}) , \quad (A9)$$

and replacing α_i, $\varepsilon(\mathbf{k})$ and $R(k)$ by β_i, $\eta(\bar{\mathbf{k}})$ and $R(\bar{\mathbf{k}})$, respectively, gives the quantities for β_j. Here photons in the final state carry the momentum

$$\mathbf{k}' = \bar{\mathbf{k}} + \mathbf{K} \quad (A10)$$

with $\mathbf{K} = \mathbf{0}$ for the transmitted direction and $\mathbf{K} = \mathbf{H}$ for the Bragg diffracted direction. The momentum of the photons in the initial state is $\mathbf{k}$, which in general is not equal to $\bar{\mathbf{k}}$. The quantity β is the solution to the dispersion equation with the fixed $\bar{\mathbf{k}}_t$ for the final states.

In the ideal thickness vibration $\bar{\mathbf{k}}$ is equal to $\mathbf{k}$ because $\mathbf{q}_t = \mathbf{0}$. Using equations (3·12), (3·13) and (A5) to (A9), we obtain, for $i \neq j$

$$f_+(0; i,j) = -f_+(\mathbf{H}; i,j) = (-1)^i \frac{\varepsilon(\mathbf{k})}{R(\mathbf{k})} \quad (A11)$$

and

$$f_-(0; i,j) = -f_-(\mathbf{H}; i,j) = -1. \quad (A12)$$

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