Acta Cryst. (1957). **10**, 609

The Scattering of Electrons by Atoms and Crystals. I. A New Theoretical Approach

By J. M. Cowley and A. F. Moodie

Chemical Physics Section, Division of Industrial Chemistry, C.S.I.R.O., Melbourne, Australia

(*Received* 7 *February* 1957)

The scattering of electrons by three-dimensional potential fields, and, in particular, the potential fields associated with a crystal lattice, is considered in terms of the new approach to physical optics recently proposed by Cowley & Moodie. The three-dimensional potential field is approximated by a large number of closely spaced two-dimensional potential distributions. An expression is obtained for the wave function on an arbitrary plane of observation for a point source of electrons at a finite distance or at infinity (parallel irradiation). Particular cases considered are the wave function at the exit surface of a crystal, corresponding to the image produced by an ideal electron microscope, and the diffraction pattern, or angular scattering function, of a crystal.

Two methods of approximation to the general expressions are discussed. In the first the wave function on the plane of observation is expressed as the sum of the contributions of electron waves scattered 1, 2, ..., n, ... times. The contribution from singly scattered waves is shown to be equivalent to the amplitude distribution given by the usual kinematic theory of scattering.

The second method of approximation corresponds to the successive increase in the number of two-dimensional distributions by which the three-dimensional potential field is approximated.

It is shown that for the special case, in which only the incident beam and one diffracted beam have appreciable amplitude in the crystal, the present theory gives essentially the same result as the dynamical theory of Bethe.

The present theory is particularly suited to the study of the diffraction of electrons by very thin crystals and crystals containing imperfections. Its applications to matters of practical importance in this field will be considered in a future publication.

1. Introduction

In considerations of the elastic scattering, or diffraction, of electrons by the potential fields of a crystal lattice, it is customary to apply either the dynamic theory or the kinematic theory of scattering, depending on the nature of the problem in hand. In the dynamic theory, as initially stated by Bethe (1928) and developed by various workers, the Schrödinger equation is set up for an electron in the periodic potential field appropriate to a perfect crystal of infinite extent. Boundary conditions representing idealized experimental conditions are applied, and solutions are obtained to varying degrees of approximation. The kinematic theory may be considered as the zero-order approximation to the dynamic theory, valid for the range of values of the potential field and the crystal thickness for which the intensities of the diffracted electron beams are negligible compared with the intensity of the incident electron beam.

The theory of the scattering of electrons by the potential field of atoms was developed along somewhat different lines, (see, for example, Mott & Massey, 1949). The so-called first Born approximation, considered until recently to be adequate for all electron-diffraction work, is equivalent to the application of the kinematic theory to atoms. Glauber & Schomaker (1953) showed that this approximation is not always adequate, and Hoerni & Ibers (1953) (also Ibers & Hoerni (1954)) calculated atomic scattering factors from a more precise, 'dynamic' scattering theory.

More recently Hoerni (1956*a*, *b*) has developed the 'pseudo-kinematic' theory of scattering by molecules and crystals, taking into account dynamic scattering within atoms but considering diffraction by the assembly of atoms to be kinematic.

For a number of problems of interest in the fields of electron diffraction and electron microscopy, neither the kinematic nor the dynamic theory can be applied to give more than a qualitative indication of the diffraction effects which might be observed. For example, the kinematic or pseudo-kinematic theory can be applied to the interpretation of the extensive spot patterns ('cross-grating' patterns) given by single crystals, if the crystal thickness is very small (Cowley, 1953), but for thicker crystals the dynamic theory must be invoked. However, applications of the dynamic theory have been limited to cases where only two or three strong beams exist in the crystal at any one time, and it is difficult to obtain even a qualitative impression of the dynamic scattering effects in the presence of a large number of diffracted beams. Again, while the influence of imperfections of the crystal lattice on the diffraction of electrons may in some cases be treated adequately by use of the kinematic theory for thin crystals, any application of the dynamic theory to such problems would be extremely difficult.

It appears that some of the difficulties encountered in the application of the dynamic theory may be overcome by employing a new theoretical approach to the scattering of electrons by atoms and crystals, based on a new formulation of physical optics recently outlined (Cowley & Moodie, 1957*a*). In this formulation Huygens' Principle is applied to give the amplitude distribution on one plane in terms of the amplitude distribution on preceding planes. In its usual form, a small-angle approximation is made, in that a spherical wave front is approximated by a paraboloid. Such an approximation is valid under the conditions usually encountered in present-day electron diffraction and microscopy where the electron energies are of the order of 50 keV. and the maximum angles of scattering considered are of the order of a few degrees. It may be noted that the dynamical theory of Bethe (1928) was originally applied to the description of the diffraction processes of low-energy electrons (a few hundred volts) with angles of scattering up to 90°, and no essential simplification of the theory has been made for applications to low-angle scattering processes.

In the paper giving the theoretical basis of the new formulation of physical optics (Cowley & Moodie, 1957*a*), expressions are derived for the wave function on the plane of observation resulting when light (or electrons) from a source traverses an arbitrary number of optical components, the effect of each of which may be represented by multiplying the wave function on a plane perpendicular to the axis of the system by some real, complex or imaginary function. In the present paper, we show how the effect on the wave function of a three-dimensional modifying system, and, in particular, a crystal of finite thickness, can be deduced from such expressions by a limiting process. General expressions will be derived for images and diffraction patterns given by thick crystals, and methods of obtaining approximations useful for special cases will be outlined. In a future publication, some matters of practical interest in the field of electron diffraction will be discussed in the light of this new theoretical approach.

2. The scattering of electrons by a potential field

We consider a monoenergetic stream of electrons, travelling in the direction of the z axis, passing through a field of electrostatic potential, $\varphi(x, y, z)$. The wavelength of the electron beam is

$$\lambda = h/[2m\{W_0+\varphi(x, y, z)\}]^{\frac{1}{2}} = \lambda_0\{1+\varphi(x, y, z)/W_0\}^{-\frac{1}{2}},$$

where W_0 is the accelerating potential for the electron beam and λ_0 is the wavelength for $\varphi(x, y, z) = 0$.

Let us assume that in a laminar volume of the potential field of thickness Δz, centred about $z = z_1$, the potential function does not vary appreciably with z and may be written $\varphi(x, y, z_1)$.

After traversing this volume, the phase of the electron waves will be changed, relative to the wave in the absence of a field, by an amount

$$[\{1+\varphi(x, y, z_1)/W_0\}^{\frac{1}{2}}-1].2\pi\Delta z/\lambda_0 .$$

For $\varphi(x, y, z_1) \ll W_0$ this is approximately $\pi\Delta z\varphi(x, y, z_1)/\lambda_0 W_0$. The effect of this slice of the potential field on the wave function is then represented by multiplying the wave function by

$$\exp [i\pi\Delta z\varphi(x, y, z_1)/\lambda_0 W_0] .$$

If, as is the case for atoms, the potential field results from a distribution of charged particles, the electrons may undergo inelastic collisions with a probability proportional to some inelastic scattering density function, $\chi(x, y, z_1)$. The effect may be considered as an absorption of electrons, since electrons inelastically scattered will be effectively incoherent with those elastically scattered and so may be regarded as completely removed from the system. In recording the intensity distribution in the plane of observation on, for example, a photographic plate, the intensities of elastically and inelastically scattered electrons are added. However, only the contributions of elastically scattered electrons will be considered here.

The effect of passing through the slice of the potential field at $z = z_1$ is thus given by multiplying the wave function by the function

$$q(x, y, z_1) = \exp [-\varrho\chi(x, y, z_1)\Delta z+i\sigma\varphi(x, y, z_1)\Delta z] , \quad (1)$$

where

$$\sigma = \pi/\lambda_0 W_0 = 2\pi m\lambda_0/h^2, \text{ and } \varrho \text{ is a constant} .$$

For Δz sufficiently small, we may write

$$q(x, y, z_1) = 1-\varrho\chi(x, y, z_1)\Delta z+i\sigma\varphi(x, y, z_1)\Delta z . \quad (2)$$

To this approximation, the wave function leaving the slice of the potential field may be described as the incident wave minus an absorption term, plus a scattered wave 90° out of phase with the incident wave. The effect is that of an amplitude object plus a phase object.

Taking the two-dimensional Fourier transform, we have

$$Q_1(u, v) = \iint q(x, y, z_1) \exp [-i(ux+vy)]dxdy \quad (3)$$

$$= \delta(u, v)-\varrho\Delta z G_1(u, v)+i\sigma\Delta z E_1(u, v) , \quad (4)$$

where

$$G_1(u, v) = \int G(u, v, w) \exp (iwz_1)dw , \quad (5)$$

$$E_1(u, v) = \int E(u, v, w) \exp (iwz_1)dw \quad (6)$$

and $\delta(u, v)$ is a Dirac delta function representing a sharp peak at the origin, of unit weight.

In analogy with conventional diffraction usage, and for convenience in manipulation, we define the function

$$\left.\begin{aligned} F_1(0,0) &= -i/\Delta z+\sigma E_1(0,0)+i\varrho G_1(0,0)\,, \\ F_1(u,v) &= \sigma E_1(u,v)+i\varrho G_1(u,v)\,. \end{aligned}\right\} \quad (7)$$

For periodic objects, the variables u, v are replaced by the integers h, k.

We make the assumption usually made in diffraction theory that the incident radiation is perfectly coherent and either is parallel or else comes from a point source. The effects of the deviations from these ideal conditions, occurring in most experimental arrangements, will be considered in a future publication.

A three-dimensional potential field may be considered to be divided into N thin slices. The effect of the nth slice is given by multiplying the wave function by $q_n(x, y)$ on a plane at the centre of the slice. The system then consists of a plane source, given by $q_0(x, y)$, N planes on which the wave function is modified, and a plane of observation, as in Fig. 1.

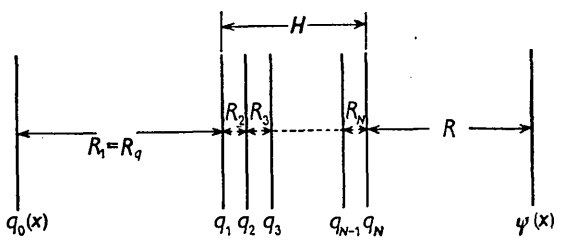

Fig. 1. The notation used for planes and distances.

The distance between the $(n-1)$th and nth plane is R_n, with $R_1 = R_q$, the distance from source to the first object plane, and $R_{N+1} = R$, the distance from the Nth plane to the plane of observation.

We consider initially functions $q_n(x)$ of one variable only. The extension to two dimensions is made readily.

The wave function $\psi(x)$ in the plane of observation is given in the new formulation of physical optics (Cowley & Moodie, 1957*a*, equation (3.4)) in terms of the Fourier transforms, $Q_n(u)$, of the functions $q_n(x)$ as

$$\begin{aligned} \psi(x) = K \exp\left(-\frac{ik_s x^2}{2R}\right) &\left[\,\right]_{N+1}\left[Q_N\left(-\frac{k_s x}{R}\right) * \left[\,\right]_{N-1} \cdots \right. \\ \cdots * \left[Q_2\left(-\frac{k_s x}{R}\right) * \right.&\left[Q_1\left(-\frac{k_s x}{R}\right) * Q_0\left(-\frac{k_s x}{R}\right) \times \right. \\ \exp\left(\frac{ik_s R_1 x^2}{2R^2}\right)\Big]_1 &\exp\left(\frac{ik_s R_2 x^2}{2R^2}\right)\Big]_2 \cdots \exp\left(\frac{ik_s R_{N-1} x^2}{2R^2}\right)\Big]_{N-1} \times \\ \exp\left(\frac{ik_s R_N x^2}{2R^2}\right)\Big]_N &* \exp\left(\frac{ik_s x^2}{2R}\right)\Big]_{N+1}, \end{aligned} \quad (8)$$

where K is a constant, $k_s = 2\pi/\lambda$, the $*$ sign denotes a convolution, and the brackets are distinguished by subscripts. The diffraction pattern or angular distribution of elastically scattered electron amplitude may be found by expressing this wave function as a function of the angle of incidence on the plane of observation and taking the limit as R becomes infinitely large. It is more convenient to avoid the limiting process by considering instead the amplitude distribution on the back focal plane of an ideal thin lens placed after the object. By this method the diffraction pattern is given as (Cowley & Moodie, 1957*a*; compare equation (4.4))†

$$\begin{aligned} \psi(\theta) = K \Big[\,\Big]_{N+1}\Big[Q_N(-2k_s\theta) * \Big[\,\Big]_{N-1} \cdots * \Big[Q_2(-2k_s\theta) \\ * \Big[Q_1(-2k_s\theta) * Q_0(-2k_s\theta) \exp(2ik_s R_1\theta^2)\Big]_1 \\ \times \exp(2ik_s R_2\theta^2)\Big]_2 \cdots \Big]_{N-1} \\ \times \exp(2ik_s R_N\theta^2)\Big]_N \exp(2ik_s R\theta^2)\Big]_{N+1}, \end{aligned} \quad (9)$$

where θ is the Bragg angle, and 2θ is the inclination to the axis of the system.

For our present purposes, we consider the potential field of the object to be subdivided into N equal slices of thickness Δz so that in the above expressions we substitute

$$R_2 = R_3 = \ldots = R_N = \Delta z\,.$$

Then, from equations (4) and (7) it follows that

$$Q_n(-2k_s\theta) = i\Delta z F_n(-2k_s\theta)\,,$$

or, for a periodic object with periodicity a in the x direction,

$$Q_n(-2k_s\theta) = i\Delta z \sum_h F_n(h).\delta\left(\theta+\frac{h\lambda}{a}\right).$$

The scattering from a three-dimensional potential field is found by evaluating the expressions (8) and (9) when N tends to infinity and Δz tends to zero in such a way that $(N-1).\Delta z = H$, the thickness of the field in the z direction.

3. The scattering of electrons by a perfect crystal

The potential distribution of a perfect crystal, considered infinite in all its dimensions, is represented by the Fourier series

$$\varphi(x,y,z) = \sum_h \sum_k \sum_l E(h,k,l) \times \exp\left\{2\pi i\left(\frac{hx}{a}+\frac{ky}{b}+\frac{lz}{c}\right)\right\}.$$

We consider a crystal infinite in the x and y directions but of finite thickness, H, in the z direction. If this crystal is divided into N slices, each of thickness Δz, the potential distribution in one of these slices is

$$\varphi(x,y,z_n) = \sum_h \sum_k E_n(h,k) \exp\left\{2\pi i\left(\frac{hx}{a}+\frac{ky}{b}\right)\right\}$$

where

$$E_n(h,k) = \sum_l E(h,k,l) \exp\left\{2\pi i\left(\frac{lz_n}{c}\right)\right\}, \quad (10)$$

† The notation in the present paper differs from that of Cowley & Moodie (1957*a*) in the numbering of the components and intervals.

and similarly for $\chi(x, y, z_n)$, $G_n(hk)$ and $G(hkl)$. In the one-dimensional equation (8) we substitute

$$Q_n\left(-\frac{k_s x}{R}\right) = i\Delta z F_n\left(-\frac{k_s x}{R}\right) = i\Delta z \sum_h F_n(h).\delta\left(x+\frac{Rh\lambda}{a}\right),$$

giving

$$\psi(x) = K(i\Delta z)^N \exp\left(-\frac{ik_s x^2}{2R}\right)\left[\ldots\left[F_N\left(-\frac{k_s x}{R}\right) * \ldots\right.\right.$$
$$\ldots * \left[F_1\left(-\frac{k_s x}{R}\right) * Q_0\left(-\frac{k_s x}{R}\right)\exp\left(\frac{ik_s R_q x^2}{2R^2}\right)\right]_1$$
$$\left.\left.\times \exp\left(\frac{ik_s \Delta z x^2}{2R^2}\right)\right]_2 \ldots \exp\left(\frac{ik_s \Delta z x^2}{2R^2}\right)\Big]_N * \exp\left(\frac{ik_s x^2}{2R}\right)\Big]_{N+1}. \quad (11)$$

For a point source at

$$x = -x_q,\ q_0(x) = \delta(x+x_q),$$

and

$$Q_0(-k_s x/R) = \exp(-ik_s x_q x/R).$$

The bracket $[_1\ldots]_1$ of (11) then becomes

$$F_1\left(-\frac{k_s x}{R}\right) * \exp\left(-\frac{ik_s x_q x}{R}\right).\exp\left(\frac{ik_s R_q x^2}{2R^2}\right)$$
$$= \sum_{h_1} F_1(h_1)\int \delta\left(X+\frac{Rh_1\lambda}{a}\right).\exp\left\{\frac{ik_s R_q(x-X)^2}{2R^2}\right\}$$
$$\times \exp\left\{-\frac{ik_s x_q(x-X)}{R}\right\} dX$$
$$= \exp\left\{ik_s\left(\frac{R_q x^2}{2R^2}-\frac{x_q x}{R}\right)\right\}.\sum_{h_1}|F_1(h_1)\exp\left\{-2\pi i\frac{h_1 x_q}{a}\right\}$$
$$\times \exp\left\{2\pi i\frac{R_q h_1 x}{Ra}\right\}.\exp\left\{\pi i\frac{R_q\lambda h_1^2}{a^2}\right\}.$$

Substituting this expression in the second bracket gives

$$\left[\ldots\right]_2 = \left[\ldots\right]_1.\exp\left\{\frac{ik_s\Delta z x^2}{2R^2}\right\} * F_2\left(-\frac{k_s x}{R}\right)$$
$$= \exp\left\{ik_s\left(\frac{(R_q+\Delta z)x^2}{2R^2}-\frac{x_q x}{R}\right)\right\}.\sum_{h_1}\sum_{h_2}F_1(h_1).F_2(h_2)$$
$$\times\exp\left\{-2\pi i\frac{(h_1+h_2)x_q}{a}\right\}.\exp\left\{2\pi i\left(\frac{R_q h_1}{Ra}+\frac{(R_q+\Delta z)h_2}{Ra}\right)x\right\}$$
$$\times\exp\left\{\frac{\pi i\lambda}{a^2}(R_q(h_1+h_2)^2+\Delta z.h_2^2)\right\}.$$

Continuing the process of substitution, we obtain

$$\left[\ldots\right]_N = \exp\left\{ik_s\left(\frac{(R_q+H)x^2}{2R^2}-\frac{x_q x}{R}\right)\right\}.\sum_{h_1}\sum_{h_2}\ldots$$
$$\ldots\sum_{h_N}F_1(h_1).F_2(h_2)\ldots F_N(h_N)$$
$$\times\exp\left\{-2\pi i(h_1+h_2+\ldots+h_N)\frac{x_q}{a}\right\}$$
$$\times\exp\left\{\frac{2\pi i}{R}[R_q h_1+(R_q+\Delta z)h_2+\ldots+(R_q+H)h_N]\frac{x}{a}\right\}$$
$$\times\exp\left\{\frac{\pi i\lambda}{a^2}[R_q(h_1+h_2+\ldots+h_N)^2+\Delta z$$
$$\times(h_2+h_3+\ldots+h_N)^2+\ldots+\Delta z.h_N^2]\right\}.$$

The wave function on the plane of observation is then

$$\psi(x) = K(i\Delta z)^N.\exp\left\{-\frac{ik_s x^2}{2R}\right\}\left[\left[\ldots\right]_N * \exp\left\{\frac{ikx^2}{2R}\right\}\right].$$

Carrying out the convolution, we have

$$\psi(x) = K_1(i\Delta z)^N$$
$$\times\exp\left\{-\frac{\pi i(x+x_q)^2}{\lambda(R+R_q+H)}\right\}\sum_{h_1}\sum_{h_2}\ldots\sum_{h_N}F_1(h_1).F_2(h_2)\ldots$$
$$\ldots F_N(h_N)\exp\left\{-2\pi i\sum_{n=1}^{N}\frac{h_n x_q}{a}\right\}$$
$$\times\exp\left\{\frac{2\pi i(x+x_q)}{a(R+R_q+H)}\sum_{n=1}^{N}[R_q+(n-1)\Delta z]h_n\right\}$$
$$\times\exp\left\{\frac{2\pi i\lambda}{(R+R_q+H)}\sum_{n=1}^{N}\sum_{m=n}^{N}p_{nm}[R_q+(n-1)\Delta z]\right.$$
$$\left.\times[R+H-(m-1)\Delta z]\frac{h_n h_m}{a^2}\right\}, \quad (12)$$

where K_1 is another constant, and $p_{nm} = 1$ for $n < m$ and $p_{nm} = \frac{1}{2}$ for $n = m$.

Generalizing this result for two dimensions, and substituting

$$s = R+R_q+H,\quad (n-1).\Delta z = z_n \text{ and } (m-1)\Delta z = z_m,$$

gives

$$\psi(x, y) = K_2\exp\left\{-\frac{\pi i}{\lambda}.\frac{[(x+x_q)^2+(y+y_q)^2]}{s}\right\}$$
$$\times\sum_{h_1}\sum_{k_1}\sum_{h_2}\sum_{k_2}\ldots\sum_{h_N}\sum_{k_N}F_1(h_1,k_1).F_2(h_2,k_2)\ldots$$
$$\ldots F_N(h_N,k_N)$$
$$\times\exp\left\{\frac{2\pi i}{s}\sum_{n=1}^{N}(R_q+z_n)\left[\frac{h_n(x+x_q)}{a}+\frac{k_n(y+y_q)}{b}\right]\right\}$$
$$\times\exp\left\{-2\pi i\sum_{n=1}^{N}\left(\frac{h_n x_q}{a}+\frac{k_n y_q}{b}\right)\right\}$$
$$\times\exp\left\{\frac{2\pi i\lambda}{s}\sum_{n=1}^{N}\sum_{m=n}^{N}p_{nm}(R_q+z_n)(R+H-z_m)\right.$$
$$\left.\times\left(\frac{h_n h_m}{a^2}+\frac{k_n k_m}{b^2}\right)\right\}.$$

The quantities R_q and R will normally be considered as parameters rather than variables in the expression for the wave function. However, in order to distinguish readily between the wave function in the general case and the wave functions for particular values of R_q and R, which will be considered later, we write for the general case

$$\psi(x, y) \equiv \psi(x, y;\ R_q, R)\ .$$

By using equation (10) and the analogous equation for G, $\psi(x, y;\ R_q, R)$ can be written in terms of $F(h, k, l)$ only, rather than the $F_n(h, k)$, by putting

$$F_n(h, k) = \sum_{l_n} F(h_n, k_n, l_n) \exp\left(\frac{2\pi i l_n z_n}{c}\right), \quad (13)$$

where

$$F(h_n, k_n, l_n) = -\frac{i}{\Delta z}.\delta(h_n, k_n) \exp\left\{-\frac{2\pi i l_n z_n}{c}\right\} + \sigma E(h_n, k_n, l_n) + i\varrho G(h_n, k_n, l_n)\ . \quad (14)$$

Then,

$$\psi(x,y;\ R_q,R) = K_2(i\Delta z)^N \exp\left\{-\frac{\pi i}{\lambda}\frac{[(x+x_q)^2+(y+y_q)^2]}{s}\right\} \times \sum_{h_1}\sum_{k_1}\sum_{l_1}\sum_{h_2}\sum_{k_2}\sum_{l_2}\cdots\sum_{h_N}\sum_{k_N}\sum_{l_N} F(h_1, k_1, l_1) \times F(h_2, k_2, l_2)\ldots F(h_N, k_N, l_N).\exp\left\{2\pi i \sum_{n=1}^{N}\frac{l_n z_n}{c}\right\} \times \exp\left\{\frac{2\pi i}{s}\sum_{n=1}^{N}(R_q+z_n)\left(\frac{h_n x}{a}+\frac{k_n y}{b}\right)\right\} \times \exp\left\{-\frac{2\pi i}{s}\sum_{n=1}^{N}(R+H-z_n)\left(\frac{h_n x_q}{a}+\frac{k_n y_q}{b}\right)\right\} \times \exp\left\{\frac{2\pi i\lambda}{s}\sum_{n=1}^{N}\sum_{m=n}^{N} p_{nm}(R_q+z_n)(R+H-z_m) \times \left(\frac{h_n h_m}{a^2}+\frac{k_n k_m}{b^2}\right)\right\}. \quad (15)$$

It may be noted that, in the absence of the final exponential term, this expression would represent the product of the functions given by projecting each slice of the crystal individually on the plane of observation. If only the cross-product terms ($n \neq m$) in the final exponential term were omitted, the expression would represent the product of all the Fourier images (Cowley & Moodie, 1957*c*) of the individual slices of the crystal formed on the plane of observation. Inclusion of these cross-product terms introduces the interaction of the effects of the various slices.

The special case of (15) of particular interest in electron microscopy is that for which R_q is very large and $R = 0$, i.e. the source is so far from the crystal that the radiation is very nearly parallel, and the electron microscope is focused on the exit face of the crystal.

If a parallel beam of electrons is incident on the crystal from a direction defined by $\alpha_x = -x_q/R_q$ and $\alpha_y = -y_q/R_q$ equation (15) for $R = 0$ becomes

$$\psi(x, y;\ \infty, 0) = k_2(i\Delta z)^N \times \exp\left\{-\frac{\pi i}{\lambda}(\alpha_x^2+\alpha_y^2)\right\}\sum_{h_1}\sum_{k_1}\sum_{l_1}\sum_{h_2}\sum_{k_2}\sum_{l_2}\cdots\sum_{h_N}\sum_{k_N}\sum_{l_N} \times F(h_1, k_1, l_1).F(h_2, k_2, l_2)\ldots F(h_N, k_N, l_N) \times \exp\left\{2\pi i\sum_{n=1}^{N}\left[\frac{h_n}{a}(x+(H-z_n)\alpha_x)+\frac{k_n}{b}(y+(H-z_n)\alpha_y)\right]\right\} \times \exp\left\{2\pi i\sum_{n=1}^{N} z_n\left[-\frac{h_n\alpha_x}{a}-\frac{k_n\alpha_x}{b} + \frac{l_n z_n}{c}-\frac{\lambda h_n^2}{2a^2}-\frac{\lambda k_n^2}{2b^2}-\sum_{m=1}^{n-1}\left(\frac{h_n h_m}{a^2}+\frac{k_n k_m}{b^2}\right)\right]\right\}. \quad (16)$$

The final exponent in this expression may be written in the form

$$-2\pi i\sum_n z_n(\zeta_n-\zeta_{n-1})\ ,$$

where

$$\zeta_n = \frac{\lambda}{2}\left\{\left(\sum_{r=1}^{n}h_r\right)^2\Big/a^2+\left(\sum_{r=1}^{n}k_r\right)^2\Big/b^2\right\} - \sum_{r=1}^{n}\frac{l_r}{c}+\sum_{r=1}^{n}\frac{h_r\alpha_x}{a}+\sum_{r=1}^{n}\frac{k_r\alpha_y}{b}\ . \quad (17)$$

The ζ_n have a particularly simple interpretation as distances in the reciprocal lattice space of the crystal. The intensities of the diffracted electrons beams from a crystal are given, in the kinematic theory of diffraction, by the intersection of the Ewald sphere, of radius $1/\lambda$, with the distribution of scattering power in the neighbourhood of the reciprocal-lattice points. To the degree of approximation used in the derivation of the expressions (8) and (9) above (Cowley & Moodie, 1957*a*), the Ewald sphere may be replaced by the paraboloid

$$w = \tfrac{1}{2}\lambda(u^2+v^2)$$

for normal incidence ($\alpha_x = \alpha_y = 0$), or by the paraboloid

$$w = \tfrac{1}{2}\lambda(u^2+v^2)-\alpha_x u-\alpha_y v$$

for α_x and $\alpha_y \neq 0$.

ζ_n, therefore, represents the distance, measured in the l-direction in reciprocal-lattice space, of the paraboloid of reflexion from the reciprocal-lattice point with coordinates

$$\sum_{r=1}^{n}h_r,\ \sum_{r=1}^{n}k_r,\ \sum_{r=1}^{n}l_r\ ,$$

corresponding to a beam which has been diffracted n times, the indices of the individual reflecting planes being $h_1k_1l_1$, $h_2k_2l_2$, ... $h_nk_nl_n$. It is analogous to the 'excitation error', ζ, of Bethe's dynamic theory of electron diffraction.

The final exponent in the expression (16) then becomes

$$-2\pi i\sum_{n=1}^{N} z_n(\zeta_n-\zeta_{n-1}) = -2\pi i\Delta z\sum_{n=1}^{N}(n-1)(\zeta_n-\zeta_{n-1}) = -2\pi i\left(H\zeta_N-\Delta z\sum_{n=1}^{N-1}\zeta_n\right).$$

Hence (16) may be written

$$\psi(x, y;\ \infty, 0) = K_2(i\Delta z)^N \times \exp\left\{-\frac{\pi i}{\lambda}(\alpha_x^2+\alpha_y^2)\right\} \sum_{h_1}\sum_{k_1}\sum_{l_1}\sum_{h_2}\sum_{k_2}\sum_{l_2}\ldots\sum_{h_N}\sum_{k_N}\sum_{l_N} \times F(h_1, k_1, l_1).F(h_2, k_2, l_2)\ldots F(h_N, k_N, l_N) \times \exp\left\{2\pi i\sum_{n=1}^{N}\left[\frac{h_n}{a}(x+(H-z_n)\alpha_x)+\frac{k_n}{b}(y+(H-z_n)\alpha_x)\right]\right\} \times \exp\left\{-2\pi i\left(H\zeta_N-\Delta z\sum_{n=1}^{N-1}\zeta_n\right)\right\}. \quad (18)$$

4. The diffraction pattern of a perfect crystal

The diffraction pattern of the crystal considered in § 3 above, in one dimension, $\psi(\theta)$, is obtained from equation (9) by putting $R_2 = R_3 = \ldots = R_N = \Delta z$, and assuming parallel incident radiation, i.e. $Q_0(-2k_s\theta) = \delta(\theta+\alpha)$; then

$$\psi(\theta) = K(i\Delta z)^N\Big[_N F_N(-2k_s\theta) * \Big[_{N-1} F_{N-1}(-2k_s\theta)*\ldots \ldots\Big[_1 F_1(-2k_s\theta) * \delta(\theta+\alpha).\exp(2ik_sR_q\theta^2)\Big]_1 \times\exp(2ik_s\Delta z\theta^2)\Big]_2\ldots\exp(2ik_s\Delta z\theta^2)\Big]_{N-1} \times\exp(2ik_s\Delta z\theta^2)\Big]_N. \quad (19)$$

The value of R_q chosen is arbitrary, and so can be taken as zero. Then the inner bracket of (19) becomes

$$[_1\ldots]_1 = F_1\{-2k_s(\theta+\alpha)\}.$$

Then

$$[_2\ldots]_2 = F_1\{-2k_s(\theta+\alpha)\}.\exp(2ik_s\Delta z\theta^2) * F_2(-2k_s\theta) = \sum_{h_1}\sum_{h_2}F_1(h_1).F_2(h_2).\delta\left(\theta+\alpha+\frac{(h_1+h_2)\lambda}{2a}\right) \times\exp\left\{2ik\Delta z\left(\theta+\frac{h_2\lambda}{2a}\right)^2\right\}.$$

Continued substitution gives

$$\psi(\theta) = K(i\Delta z)^N\exp\{2ik_s(H-\Delta z)\theta^2\} \times\sum_{h_1}\sum_{h_2}\ldots\sum_{h_N}F_1(h_1).F_2(h_2)\ldots F_N(h_N) \times\delta\left(\theta+\frac{\lambda}{2a}(h_1+h_2+\ldots+h_N)+\alpha\right) \times\exp\left\{2\pi i\sum_{n=1}^{N}z_n\left[\frac{h_n\theta}{a}+\frac{\lambda h_n^2}{2a^2}+\sum_{m=n+1}^{N}\frac{\lambda h_nh_m}{a^2}\right]\right\}. \quad (20)$$

If now we consider the amplitude, $U(h)$, of the diffracted beam in the direction

$$\theta = -\frac{\lambda}{2a}\sum_{n=1}^{N}h_n-\alpha,$$

where

$$h = \sum_{n=1}^{N}h_n,$$

the number of summations in (20) is reduced by one, and substituting for θ in the exponential term gives

$$\exp\left\{-2\pi i\sum_{n=1}^{N}z_n\left[\frac{\lambda}{2}\frac{h_n^2}{a^2}+\frac{h_n\alpha}{a}+\sum_{m=1}^{n-1}\frac{\lambda h_nh_m}{a^2}\right]\right\}.$$

Generalizing for two dimensions, and making use of equation (13) to introduce $F(h_n, k_n, l_n)$ instead of the $F_n(h_n, k_n)$, gives the amplitude $U(h, k)$ of the diffracted beam with indices

$$h = \sum_{n=1}^{N}h_n,\quad k = \sum_{n=1}^{N}k_n,$$

in a form analogous to (18) above:

$$U(h, k) = K(i\Delta z)^N\exp(2ik_sH\theta^2)\sum_{h_1}\sum_{k_1}\sum_{l_1}\sum_{h_2}\sum_{k_2}\sum_{l_2}\ldots \ldots\sum_{h_{N-1}}\sum_{k_{N-1}}\sum_{l_{N-1}}.\sum_{l}.F(h_1, k_1, l_1).F(h_2, k_2, l_2)\ldots \ldots F(h_{N-1}, k_{N-1}, l_{N-1}).F\left(h-\sum_{n=1}^{N-1}h_n,\ k-\sum_{n=1}^{N-1}k_n,\ l-\sum_{n=1}^{N-1}l_n\right) \times\exp\left\{-2\pi i\left(H\zeta-\Delta z\sum_{n=1}^{N-1}\zeta_n\right)\right\}, \quad (21)$$

where $\zeta = \zeta_N$, the distance in the l-direction of the paraboloid of reflection from the reciprocal-lattice point with coordinates h, k, l.

It may be noted that, for $\alpha_x = \alpha_y = 0$, i.e. for normal incidence, the diffraction pattern $U(h, k)$, given by (21) is the Fourier transform of the amplitude distribution at the exit face of the crystal, $\psi(x, y;\ \infty, 0)$, given by (18), apart from constant terms and terms of modulus unity. With the same reservations, it is similarly the Fourier transform of the amplitude distribution, $\psi(x, y;\ \infty, R)$, in any plane in or near the crystal.

5. More general problems and approximations

The considerations of §§ 3 and 4 above could be extended to give expressions analogous to (15), (18) and (21) for scattering from non-periodic objects such as isolated atoms, molecules or imperfect crystals, including crystals of finite extent. The summations over the integers h_n, k_n and l_n are then replaced by integrations over the reciprocal-lattice coordinates u_n, v_n and w_n. It may also be noted that the derivations of the expressions (15), (18), (21) could be carried out without making the assumption that $R_2 = R_3 = \ldots = R_N = \Delta z$. For some problems it may be more appropriate to use the distances

$$z_n = \sum_{m=2}^{n}R_m$$

and to redefine the $F_n(h_n, k_n)$ suitably.

No way has yet been found of expressing the limits of the general expressions (15), (18) and (21) for N tending to infinity and Δz tending to zero, with $(N-1).\Delta z = H$, except in terms of an infinite number of summations. It is therefore necessary to determine the limit for each particular problem considered or for approximate forms of these expressions, valid in particular cases.

Two methods of obtaining approximations to the general expressions, of use in considerations of thin crystals, will be discussed in the following sections. In the first we divide the general expressions into terms representing single scattering, double scattering, triple scattering and so on. The second involves the successive subdivision of the crystal into more and more parts, the effect of each part being approximated by a two-dimensional potential distribution.

6. Single- and multiple-scattering terms

We consider first the expression (21) giving the diffraction pattern of the crystal. From the definition (14), it can be seen that in each set of $F(h_n, k_n, l_n)$ values, the $F(0, 0, 0)$ is very much larger than any other. If therefore, the expression (21) were written out in full without summation signs, the largest individual term would be that containing $F^N(0, 0, 0)$, occurring for $h = 0$, $k = 0$ only. Next largest would be N terms for each h, k, containing $F^{N-1}(0, 0, 0).F(h, k, l)$. Then there would be $N(N-1)/2$ terms for each h, k, containing

$$F^{N-2}(0, 0, 0).\sum_{h_1}\sum_{k_1}\sum_{l_1} F(h_1, k_1, l_1).F(h-h_1, k-k_1, l-l_1),$$

and so on. These groups of individual terms then represent, respectively, the beam transmitted in the forward direction, beams diffracted once only in a direction given by the indices h, k, beams diffracted twice with the final direction given by h, k, and so on.

The zero-order approximation to the diffraction pattern is thus, from (21),

$$\begin{aligned} U_0(0, 0) &= \exp(2ik_s H\theta^2).(i\Delta z)^N.F^N(0, 0, 0) \\ &= \exp(2ik_s H\theta^2).\{1+i\sigma\Delta z E(0, 0, 0) \\ &\qquad -\varrho\Delta z G(0, 0, 0)\}^N. \end{aligned}$$

Neglecting the exponential term of modulus unity, and taking the limit as N tends to infinity and Δz tends to zero, we have

$$U_0(0, 0) = \exp\{i\sigma HE(0, 0, 0)-\varrho HG(0, 0, 0)\}. \quad (22)$$

6(a). *Single-scattering diffraction patterns*

If scattering takes place in only one slice of the crystal, say the mth, then all $h_n, k_n, l_n = 0$ except $h_m, k_m, l_m = h, k, l$. Then

$$\zeta_n = 0 \quad \text{for} \quad n < m,$$
$$\zeta_n = \zeta \quad \text{for} \quad n \geq m,$$

and

$$H\zeta_N - \Delta z.\sum_{n=1}^{N-1}\zeta_n = H\zeta-(N-m)\Delta z.\zeta = z_m.\zeta.$$

Summing the amplitude of once-scattered electrons for all slices, and neglecting terms of modulus unity, we have

$$\begin{aligned} U_1(h, k) &= (i\Delta z)^{N-1}.F^{N-1}(0, 0, 0) \\ &\quad \times \sum_m \sum_l i\Delta z F(h, k, l) \exp\{-2\pi i z_m \zeta\}. \end{aligned}$$

Taking the limit for $N \to \infty$, $\Delta z \to 0$ and $(N-1)\Delta z = H$, gives

$$\begin{aligned} U_1(h, k) &= \exp\{i\sigma HE(0, 0, 0)-\varrho HG(0, 0, 0)\} \\ &\quad \times \sum_l iF(h, k, l).\int_0^H \exp\{-2\pi i z\zeta\}.dz \\ &= \exp\{i\sigma HE(0, 0, 0)-\varrho HG(0, 0, 0)\} \\ &\quad \times \sum_l iF(h, k, l)\left(\frac{1-\exp(-2\pi i H\zeta)}{-2\pi i\zeta}\right) \\ &= \exp\{i\sigma HE(0, 0, 0)-\varrho HG(0, 0, 0)\} \\ &\quad \times \sum_l i[\sigma E(h, k, l)+i\varrho G(h, k, l)] \\ &\quad \times \exp(-\pi i H\zeta).\left(\frac{\sin\pi H\zeta}{\pi\zeta}\right). \end{aligned} \quad (23)$$

Assuming that ζ is large for all values of l except one, the intensity of the reflexion is proportional to

$$\begin{aligned} &U_1(h, k).U_1^*(h, k) = \exp\{-2\varrho HG(0, 0, 0)\}. \\ &\quad [\sigma^2E^2(h, k, l)+\varrho^2G^2(h, k, l)].(\sin^2\pi H\zeta)/(\pi\zeta)^2. \end{aligned} \quad (24)$$

The single scattering approximation is thus entirely analogous to the kinematic theory of electron diffraction. The intensity is seen to be proportional to the square of the (complex) structure factor and to the term $(\sin^2\pi H\zeta)/(\pi\zeta)^2$, which represents the effect of the Ewald sphere (or, in this case, paraboloid) cutting the region of scattering power about the reciprocal-lattice point which has the form of the square of the 'shape transform' of the crystal. Inelastic scattering reduces the intensity by an exponential factor.

Boersch (1948) has shown that under the conditions usually encountered in electron diffraction or microscopy, $\varrho G(h, k, l)$, the part of the structure factor arising from the amplitude-grating effect, is small compared with $\sigma E(h, k, l)$ and can usually be neglected. Then $F(h, k, l) = \sigma E(h, k, l)$ and is proportional to the structure factor for electrons normally used in the kinematic theory. The presence of the i before the structure factor in (23) implies that the scattered beams are 90° out of phase with the undiffracted beam. This is usually ignored in the kinematic theory, since it does not affect the intensities. It must be retained, however, when multiple scattering is considered.

6(b). *Multiple scattering in diffraction patterns*

The amplitudes of beams scattered two, three or more times are found by an extension of the method

used for single scattering. For example, for triple scattering we have all $h_n, k_n, l_n = 0$ except that $h_m, k_m, l_m = h_1, k_1, l_1, h_p, k_p, l_p = h_2, k_2, l_2$ and $h_r, k_r, l_r = h-h_1-h_2, k-k_1-k_2, l-l_1-l_2$.

Then

$$\zeta_n = \begin{cases} 0 & \text{for} \quad 0 < n < m \\ \zeta_1 & \text{for} \quad m \leq n < p \\ \zeta_2 & \text{for} \quad p \leq n < r \\ \zeta & \text{for} \quad r \leq n \leq N \end{cases}$$

and

$$\begin{aligned} H\zeta_N - \Delta z \sum_{n=1}^{N-1} \zeta_n &= H\zeta - \{(p-m)\zeta_1 + (r-p)\zeta_2 + (N-r)\zeta\} . \Delta z \\ &= z_1\zeta_1 + z_2(\zeta_2-\zeta_1) + z_3(\zeta-\zeta_2) . \end{aligned}$$

In the limit, neglecting terms of modulus unity,

$$\begin{aligned} U_3(h, k) &= \exp\{i\sigma HE(0, 0, 0) - \varrho HG(0, 0, 0)\} . (-i) \\ &\quad \times \sum_{h_1} \sum_{k_1} \sum_{l_1} \sum_{h_2} \sum_{k_2} \sum_{l_2} \sum_{l} F(h_1, k_1, l_1) . F(h_2, k_2, l_2) \\ &\quad \times F(h-h_1-h_2, k-k_1-k_2, l-l_1-l_2) \\ &\quad \times \int_0^H \int_{z_1}^H \int_{z_2}^H \exp\{-2\pi i[z_1\zeta_1 + z_2(\zeta_2-\zeta_1) \\ &\qquad + z_3(\zeta-\zeta_2)]\} dz_3 . dz_2 . dz_1 \\ &= \exp\{i\sigma HE(0, 0, 0) - \varrho HG(0, 0, 0)\} . i^3 \\ &\quad \times \sum_{h_1} \sum_{k_1} \sum_{l_1} \sum_{h_2} \sum_{k_2} \sum_{l_2} \sum_{l} F(h_1, k_1, l_1) . F(h_2, k_2, l_2) \\ &\quad \times F(h-h_1-h_2, k-k_1-k_2, l-l_1-l_2) \\ &\quad \times \frac{\exp(-2\pi_1\zeta H)}{(2\pi i)^3} \left\{ \frac{\exp(2\pi i\zeta_1 H) - 1}{\zeta_1(\zeta_1-\zeta_2)(\zeta_1-\zeta)} \right. \\ &\quad \left. + \frac{\exp(2\pi i\zeta_2 H) - 1}{\zeta_2(\zeta_2-\zeta_1)(\zeta_2-\zeta)} + \frac{\exp(2\pi i\zeta H) - 1}{\zeta(\zeta-\zeta_1)(\zeta-\zeta_2)} \right\} . \end{aligned} \tag{25}$$

Generalizing this result, for n-times scattered beams, we have

$$\begin{aligned} U_n(h, k) &= \exp\{i\sigma HE(0, 0, 0) - \varrho HG(0, 0, 0)\} . i^n \\ &\times \sum_{l} . \sum_{h_1} \sum_{k_1} \sum_{l_1} \cdots \sum_{h_{n-1}} \sum_{k_{n-1}} \sum_{l_{n-1}} . F(h_1, k_1, l_1) . F(h_2, k_2, l_2) \cdots \\ &\quad \cdots F\left(h - \sum_{r=1}^{n-1} h_r, k - \sum_{r=1}^{n-1} k_r, l - \sum_{r=1}^{n-1} l_r\right) . \frac{\exp(-2\pi i\zeta H)}{(2\pi i)^n} \\ &\times \left[\frac{\exp(2\pi i\zeta_1 H) - 1}{\zeta_1(\zeta_1-\zeta_2)\cdots(\zeta_1-\zeta_{n-1})(\zeta_1-\zeta)} \right. \\ &\quad + \sum_{m=2}^{n-1} \frac{\exp(2\pi i\zeta_m H) - 1}{\zeta_m(\zeta_m-\zeta_1)\cdots(\zeta_m-\zeta_{m-1})(\zeta_m-\zeta_{m+1})\cdots(\zeta_m-\zeta)} \\ &\quad \left. + \frac{\exp(2\pi i\zeta H) - 1}{\zeta(\zeta-\zeta_1)(\zeta-\zeta_2)\cdots(\zeta-\zeta_{n-1})} \right] . \end{aligned} \tag{26}$$

The total amplitude is given by

$$U(h, k) = \sum_{n=0}^{\infty} U_n(h, k) .$$

6(c). *Single-scattering approximation to the microscope image*

A similar series of approximations may be made to the amplitude distribution corresponding to the image seen in an ideal electron microscope, with parallel incidence and $R = 0$. The zero-order approximation to equation (18), taking $\alpha_x = \alpha_y = 0$, is

$$\begin{aligned} &\psi_0(x, y; \infty, 0) \\ &= K(i\Delta z)^N \left[\sum_h \sum_k \sum_l F(0, 0, 0) \exp\left\{2\pi i\left(\frac{hx}{a} + \frac{ky}{b}\right)\right\} \right]^N \\ &= K \exp\{i\sigma HE(0, 0, 0) - \varrho HG(0, 0, 0)\} . \end{aligned}$$

If we make the single-scattering approximation in the same way as in § 6(a) above, and consider the case of normal incidence ($\alpha_x = \alpha_y = 0$) only, we have

$$\begin{aligned} &\psi_1(x, y; \infty, 0) \\ &= K(i\Delta z)^N F^{N-1}(0, 0, 0) \sum_m \sum_{h_m} \sum_{k_m} \sum_{l_m} F(h_m, k_m, l_m) \\ &\qquad \times \exp\left\{2\pi i\left(\frac{h_m x}{a} + \frac{k_m y}{b} + \frac{l_m z}{c}\right)\right\} . \exp(-2\pi i z\zeta_0) , \end{aligned}$$

where

$$\zeta_0 = \zeta(h_m, k_m, 0) = -\tfrac{1}{2}\lambda(h_m^2/a^2 + k_m^2/b^2) .$$

The summation over h_m, k_m and l_m represents the inverse Fourier transform of the product of $F(hkl)$ and $\exp(-2\pi i z\zeta_0)$. The inverse Fourier transform of $\exp(-2\pi i z\zeta_0)$ is

$$\frac{(1+i)ab}{2\lambda z} \exp\left\{\frac{i\pi(x^2+y^2)}{\lambda z}\right\} .$$

Hence, in the limit as $N \to \infty$, $\Delta z \to 0$ and $(N-1) . \Delta z = H$,

$$\begin{aligned} \psi_1(x, y; \infty, 0) &= K \exp\{i\sigma HE(0, 0, 0) - \varrho HG(0, 0, 0)\} . \\ &\quad \times \int_0^H i \left[\{\sigma\varphi(x, y, z) + i\varrho\chi(x, y, z)\} * \frac{(1+i)ab}{2\lambda z} \right. \\ &\quad \left. \times \exp\left\{\frac{i\pi(x^2+y^2)}{\lambda z}\right\} \right] dz . \end{aligned} \tag{27}$$

If λ is very small, ζ_0 will be small, and (27) reduces to

$$\begin{aligned} \psi_1(x, y; \infty, 0) &= K_1 \exp\{i\sigma HE(0, 0, 0) - \varrho HG(0, 0, 0)\} \\ &\quad \times \int_0^H \{i\sigma\varphi(x, y, z) - \varrho\chi(x, y, z)\} dz , \end{aligned} \tag{28}$$

i.e. the image of the crystal is obtained by projection of the structure in the direction of the incident beam. It may be noted that the contribution of $\varphi(x, y, z)$ being 90° out of phase with the unscattered beam, $\psi_0(x, y; \infty, 0)$, will not affect the intensity distribution. The only image of the lattice visible will be that due to the much smaller term $\varrho\chi(x, y, z)$.

The modification of $\psi_1(x, y; \infty, 0)$ due to finite λ, is represented by the convolution in (27). The effect on the image intensity distribution is not easy to

assess, but will, in general, represent a loss of resolution.

The contributions of beams scattered twice, three and more times to the amplitude function $\psi(x,y;\infty,0)$ can be assessed by an extension of the method used for single scattering, as in the case of the diffraction pattern.

7. Successive subdivision approximations

A method of approximation to the general expressions derived above for the scattering of electrons by crystals, which may be of use for some problems, is to consider the crystal of thickness H subdivided into N crystals of thickness H/N, assuming that each subcrystal may be approximated by a two-dimensional potential distribution and that all subcrystals scatter coherently.

7(*a*). *First approximation*, $N = 1$

Neglecting the inelastic scattering term, $\chi(x, y, z)$, the crystal is approximated by a potential distribution $\varphi(x, y)$ on a plane through the crystal, where

$$\varphi(x, y) = \int_0^H \varphi(x, y, z)dz .$$

The effect on the wave function is then represented by multiplying by

$$q_1(x, y) = \exp\{i\sigma\varphi(x, y)\} .$$

For a point source, distance R_q from the object plane, the wave function on a plane of observation at a distance R is

$$\psi'(x, y;\ R_q, R) = K \sum_h \sum_k E'(h, k) \times \exp\left\{\frac{2\pi i}{(R+R_q)}[h(R_q x - Rx_q)+k(R_q y - Ry_q)]\right\} \times \exp\left\{\frac{\pi i\lambda R R_q}{(R+R_q)}\left(\frac{h^2}{a^2}+\frac{k^2}{b^2}\right)\right\} , \quad (29)$$

where $E'(h, k)$ is the Fourier transform of $\exp\{i\sigma\varphi(x, y)\}$. This expression represents the totality of Fourier images of a phase grating. The nature of these images is the subject of a separate publication (Cowley & Moodie, 1957*b*).

The diffraction pattern, in this approximation, is simply

$$U'(h, k) = K_1 . E'(h, k) ,$$

where K_1 contains constant terms and terms of modulus unity. This is equivalent to a pseudo-kinematic theory of diffraction by a crystal with the assumption that the Ewald sphere is planar. If $\sigma\varphi(x, y)$ is so small that the exponential can be approximated by $1+i\sigma\varphi(x, y)$, then $E'(h, k) = \delta(h, k) + i\sigma E(h, k)$. Ignoring the delta function and the i then gives the kinematic theory for a planar Ewald sphere.

7(*b*). $N = 2$

The scattering of the crystal is here approximated by the scattering of two planar potential distributions a distance $H/2$ apart. This is a special case of the more general problem of the successive scattering by two crystals, treated in detail elsewhere (Cowley & Moodie, 1957*d*).

In particular, the diffraction pattern for $\alpha_x = \alpha_y = 0$ is given by

$$U''(h, k) = K_1 \sum_{h_1} \sum_{k_1} E_1'(h_1, k_1) . E_2'(h-h_1, k-k_1) \times \exp\left\{-2\pi i\lambda \frac{H}{2}\left[\frac{h_1^2}{2a^2}+\frac{k_1^2}{2b^2}\right]\right\}$$

where $E_1'(h, k)$ is the Fourier transform of

$$\exp\{i\sigma\varphi_1(x, y)\} \quad \text{with} \quad \varphi_1(x, y) = \int_0^{H/2} \varphi(x, y, z)dz$$

and $E_2'(h, k)$ is the Fourier transform of

$$\exp\{i\sigma\varphi_2(x, y)\} \quad \text{with} \quad \varphi_2(x, y) = \int_{H/2}^{H} \varphi(x, y, z)dz .$$

For H very small it is evident that this reduces to the first approximation, since, without the exponential term, $U''(h, k)$ is the Fourier transform of

$$\exp\{i\sigma\varphi_1(x, y)\} . \exp\{i\sigma\varphi_2(x, y)\} = \exp\{i\sigma\varphi(x, y)\} .$$

Subdivision of the crystal into a sufficiently large number of parts will lead to the general expressions (15), (18) and (21) derived above.

8. The two-beam approximation

Although it is envisaged that the principal applications of the present theory will probably be in consideration of relatively thin crystals, it is interesting to confirm that for thick crystals it gives the same result as the dynamic theory of diffraction of Bethe in the special case most commonly treated by that theory, namely, the case where only two beams of appreciable intensity exist in the crystal, the primary beam and one diffracted beam.

We assume that the paraboloid of reflexion (equivalent to the Ewald sphere) is close to the reciprocal-lattice point h, k, l, passes through the origin, 0, 0, 0, and does not approach any other reciprocal-lattice point, i.e. $\zeta_0 = 0$, ζ_h is small, and all other ζ are so large that all terms in the general expression (21) containing them may be neglected. We then express the general expression as the sum of the contributions of beams scattered once, twice, three times and so on, as in § 6 above, scattering here referring to changes in the direction of propagation.

Only the amplitude in the h, k direction will be considered. Thus, the zero-order term $U_0(h, k)$ has no component in the h, k direction and so is omitted. The single-scattering contribution contains only one term:

that involving ζ_h with the coefficient F_h (the single index h being used in abbreviation of the index triple h, k, l). From (23),

$$U_1(h,k) = -\exp\{i\sigma HE_0 - \varrho HG_0\} iF_h \times \frac{\exp(-2\pi i\zeta_h H)}{2\pi i\zeta_h}\{\exp(2\pi i\zeta_h H)-1\}.$$

The double-scattering contribution has no term in the direction h, k containing only ζ_0 and ζ_h. The triple-scattering contribution contains only one term in the h, k direction involving ζ_0 and ζ_h only, namely that for which $\zeta_1 = \zeta_h$, $\zeta_2 = \zeta_0$, $\zeta = \zeta_h$. Thus

$$\begin{aligned}U_3(h,k) &= \exp\{i\sigma HE_0 - \varrho HG_0\}.i^3F_h.F_{\bar{h}}.F_h \\ &\times \int_0^H\int_{z_1}^H\int_{z_2}^H \exp\{2\pi i(-\zeta_h z_1+\zeta_h z_2-\zeta_h z_3)\}dz_1.dz_2.dz_3 \\ &= \exp\{i\sigma HE_0-\varrho HG_0\}.i^3.F_h^2.F_{\bar{h}}.\frac{\exp(-2\pi i\zeta_h H)}{(2\pi i\zeta_h)^3} \\ &\times [(-2+2\pi i\zeta_h H)\exp(2\pi i\zeta_h H)-(2+2\pi i\zeta_h H)].\end{aligned}$$

The next contribution is from the five-times scattered term with $\zeta_1 = \zeta_h$, $\zeta_2 = \zeta_0$, $\zeta_3 = \zeta_h$, $\zeta_4 = \zeta_0$, $\zeta = \zeta_h$. This is

$$\begin{aligned}U_5(h,k) &= \exp\{i\sigma HE_0-\varrho HG_0\}.i^5F_h^3.F_{\bar{h}}^2.\frac{\exp(-2\pi i\zeta_h H)}{(2\pi i\zeta_h)^5} \\ &\times\left[\left\{6-3(2\pi i\zeta_h H)+\frac{(2\pi i\zeta_h H)^2}{2!}\right\}\right. \\ &\left.\times\exp(2\pi i\zeta_h H)-\left\{6+3(2\pi i\zeta_h H)+\frac{(2\pi i\zeta_h H)^2}{2!}\right\}\right].\end{aligned}$$

Using the abbreviations $s = 2\pi\zeta_h$, $t^2 = F_hF_{\bar{h}}/4\pi\zeta_h^2 = F_h.F_{\bar{h}}/s^2$, the sum of all contributions to $U(h,k)$ can be expressed as

$$\begin{aligned}U(h,k) &= \exp\{i\sigma HE_0-\varrho HG_0\}it(F_h/F_{\bar{h}})^{\frac{1}{2}} \\ &\times[\{1-\exp(-isH)\}(1-2t^2+6t^4-20t^6+\ldots) \\ &+\{1+\exp(-isH)\}(1-3t^2+10t^4-35t^6+\ldots) \\ &+\{1-\exp(-isH)\}(1-4t^2+15t^4-\ldots\ldots) \\ &+\{1+\exp(-isH)\}(1-5t^2+21t^4-\ldots\ldots) \\ &+\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots)] \\ &= \exp\{i\sigma HE_0-\varrho HG_0\}it(F_h/F_{\bar{h}})^{\frac{1}{2}}. \\ &\times\left[\sum_{n=0}^{\infty}\frac{(Hst^2)^n}{n!}\sum_{m=0}^{\infty}C_m^{n+2m}(it)^{2m}\right. \\ &\left.-\exp(-isH)\sum_{n=0}^{\infty}\frac{(-Hst^2)^n}{n!}\sum_{m=0}^{\infty}C_m^{n+2m}(it)^{2m}\right]. \qquad (30)\end{aligned}$$

Summations of the form $\sum_m C_m^{n+2m}\alpha^m$ can be evaluated by use of the following relations between the binomial coefficients for which no formal derivation or reference to the literature can be given, but which may be readily verified by expansion:

$$\sum_{m=0}^{\infty}C_m^{2n+2m}\alpha^m = (1-2\alpha)^{-n-1}\sum_{m=0}^{\infty}C^{n+2m}\{\alpha(1-2\alpha)^{-1}\}^{2m},$$

$$\sum_{m=0}^{\infty}C_m^{2m}\alpha^m = (1-2\alpha)^{-1}\sum_{m=0}^{\infty}C_m^{2m}\{\alpha(1-2\alpha)^{-1}\}^{2m},$$

$$\sum_{m=0}^{\infty}C_m^{2n+1+2m}\alpha^m = \alpha\sum_{m=0}^{\infty}C_m^{2n+2+2m}\alpha^m+\sum_{m=0}^{\infty}C_n^{2n+2m}\alpha^m.$$

Thus we derive

$$\sum_m C_m^{2m}\alpha^m = (1-4\alpha)^{-\frac{1}{2}},$$

$$\begin{aligned}\sum_m C_m^{1+2m}\alpha^m &= (2\alpha)^{-1}\{(1-4\alpha)^{-\frac{1}{2}}-1\} \\ &= \{2\alpha(1-4\alpha)^{\frac{1}{2}}\}^{-1}\{1-(1-4\alpha)^{\frac{1}{2}}\},\end{aligned}$$

$$\begin{aligned}\sum_m C_m^{2+2m}\alpha^m &= (2\alpha^2)^{-1}\left\{\frac{(1-2\alpha)}{(1-4\alpha)^{\frac{1}{2}}}-1\right\} \\ &= \{4\alpha^2(1-4\alpha)^{\frac{1}{2}}\}^{-1}.\{1-(1-4\alpha)^{\frac{1}{2}}\}^2,\end{aligned}$$

and so on.

Hence

$$\begin{aligned}\sum_n\frac{(Hst^2)^n}{n!}\sum_m C_m^{n+2m}(it)^{2m+1} &= \frac{it}{(1+4t^2)^{\frac{1}{2}}}\left[1-\frac{iHst^2}{2t^2}\{1-(1+4t^2)^{\frac{1}{2}}\}\right. \\ &\left.+\frac{(Hst^2)^2}{2!4t^4}\{1-(1+4t^2)^{\frac{1}{2}}\}^2-\ldots\right] \\ &= \frac{it}{(1+4t^2)^{\frac{1}{2}}}\exp\left[-\frac{isH}{2}\{1-(1+4t^2)^{\frac{1}{2}}\}\right].\end{aligned}$$

Then

$$\begin{aligned}U(h,k) &= \exp(i\sigma HE_0-\varrho HG_0).it(F_h/F_{\bar{h}})^{\frac{1}{2}} \\ &\times(1+4t^2)^{-\frac{1}{2}}\{\exp[-\tfrac{1}{2}isH\{1-(1+4t^2)^{\frac{1}{2}}\}] \\ &\qquad-\exp[+\tfrac{1}{2}isH\{1-(1+4t^2)^{\frac{1}{2}}\}-isH]\} \\ &= \exp(i\sigma HE_0-\varrho HG_0)\left(\frac{F_h}{F_{\bar{h}}}\right)^{\frac{1}{2}}\frac{it\exp(-\frac{1}{2}isH)}{(1+4t^2)^{\frac{1}{2}}} \\ &\qquad\times\sin\frac{Hs}{2}(1+4t^2)^{\frac{1}{2}}.\end{aligned}$$

Substituting for s and t gives, finally,

$$U(h,k) = \exp\{i\sigma HE_0-\varrho HG_0\}iF_h\exp(-2\pi i\zeta_h H)\times\frac{\sin H(\pi^2\zeta_h^2+F_hF_{\bar{h}})^{\frac{1}{2}}}{2(\pi^2\zeta_h^2+F_h.F_{\bar{h}})^{\frac{1}{2}}}. \qquad (31)$$

If we neglect the component of F_h due to inelastic scattering, and assume a centre of symmetry so that $E_h = E_{\bar{h}}$, this reduces to

$$U(h,k) = \exp\{i\sigma HE_0-\varrho HG_0-2\pi i\zeta_h H\}i\sigma E_h\times\frac{\sin H(\pi^2\zeta_h^2+\sigma^2E_h^2)^{\frac{1}{2}}}{2(\pi^2\zeta_h^2+\sigma^2E_h^2)^{\frac{1}{2}}}.$$

Apart from the exponential factor, this expression is identical with that obtained from the usual dynamic

theory under the same assumptions, (see, for example, Pinsker, 1953), namely,

$$\psi_h = \frac{v_h}{k} \frac{\sin \frac{1}{2}kH\{(\theta-\theta_0)^2 \sin^2 2\theta + v_h^2/k_s^4\}^{\frac{1}{2}}}{\{(\theta-\theta_0)^2 \sin^2 2\theta + v_h^2/k_s^4\}^{\frac{1}{2}}} ,$$

since from the definitions of the various quantities it can readily be shown that

$$E_h = v_h/2k_s \quad \text{and} \quad \zeta_h\lambda = (\theta-\theta_0).\sin 2\theta .$$

9. Discussion

It may be noted that the derivation of the amplitude of the scattered wave for the two-beam case by the above method is not very much simpler than the derivation by means of Bethe's dynamic theory, even though the mathematics might be considered to follow the physical processes more closely and the nature of the approximations made may be seen more readily. Nor does the present theory, as it now stands, offer any much more convenient way of calculating amplitudes for thick crystals when more than two strong beams are present. However, the application of the theory to such problems is of secondary interest, particularly since only rarely in practice are the appropriate experimental conditions found, i.e. the crystal is relatively thick, has parallel faces and has no lattice imperfections. The formulation of the theory is such as to make it particularly suited to the discussion of relatively thin crystals, not much thicker than the limit for which the kinematic theory is valid, or crystals containing lattice imperfections, giving a large number of diffracted beams simultaneously. The results are thus of particular interest in connection with structure analysis of small single crystals based on their electron-diffraction spot patterns. The methods of structure analysis now used are based on the assumption that the intensities of the diffraction spots are correctly given by the kinematic theory. The experimental techniques of specimen preparation and examination are not as yet sufficiently developed to ensure that this assumption is always justified. It is therefore necessary to know how the intensities may be modified if the crystal thickness or degree of lattice perfection is such that some 'dynamic' scattering takes place.

Considerable interest in the nature of electron-microscope images of crystal lattices has recently been generated by the observation by Menter (1956) of features in electron micrographs which represent, in some sense, the coarser features of the lattice structure of some crystals. It seems probable that the present theory may be of use in connexion with the interpretation of such images. From the limited discussion given above it is evident that the image may be regarded as a projection of the crystal lattice on the plane of observation only in very special circumstances.

Finally, it may be emphasized that the description of the scattering of electrons by crystals may be applied more generally to the scattering by periodic or non-periodic objects of any form of radiation, including visible light, X-rays, neutrons etc., providing only that the scattering process is scalar, and that the angles of scattering involved are small.

References

Bethe, H. A. (1928). *Ann. Phys., Lpz.* **87**, 55.

Boersch, H. (1948). *Optik*, **3**, 24.

Cowley, J. M. (1953). *Acta Cryst.* **6**, 516.

Cowley, J. M. & Moodie, A. F. (1957*a*). In the Press.

Cowley, J. M. & Moodie, A. F. (1957*b*). In preparation.

Cowley, J. M. & Moodie, A. F. (1957*c*). *Proc. Phys. Soc.* B, **70**, 486.

Cowley, J. M. & Moodie, A. F. (1957*d*). In preparation.

Glauber, F. & Schomaker, V. (1953). *Phys. Rev.* **89**, 667.

Hoerni, J. A. (1956*a*). *Phys. Rev.* **102**, 1530.

Hoerni, J. A. (1956*b*). *Phys. Rev.* **102**, 1534.

Hoerni, J. A. & Ibers, J. A. (1953). *Phys. Rev.* **91**, 1182.

Ibers, J. A. & Hoerni, J. A. (1954). *Acta Cryst.* **7**, 405.

Menter, J. W. (1956). *Proc. Roy. Soc.* A, **236**, 119.

Mott, N. F. & Massey, H. S. W. (1949). *Theory of Atomic Collisions*, 2nd ed. Oxford: Clarendon Press.

Pinsker, Z. G. (1953). *Electron Diffraction*. London: Butterworths.