Electron Diffraction and Imaging Effects for Superimposed Thin Crystals

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The possibility is explored of obtaining structural information from the moiré-like patterns observed in electron micrographs of overlapping thin crystals. General expressions are derived for the amplitude distribution in any plane of observation following two thin crystals. It is shown that in certain special cases this amplitude distribution approximates to the convolution of the projections of the potential distributions of the two crystals. Structural information may be derived from such a convolution function. Methods are indicated for determining how the resolution obtainable depends on the thickness of the crystals, their separation and mutual orientation.

1. Introduction

In recent years many observations have been made of moiré-like fringe in electron microscope images of overlapping crystals. Some account of these observations and of the theory due to Farrant & Rees (1956) of their origin, has been included in the review article by Cowley & Rees (1958).

The most frequent observation is that the region in which appropriately oriented crystals overlap is crossed by a set of parallel equidistant fringes. Each crystal is then acting as a diffraction grating periodic in one direction only since it is sufficiently thick and so oriented that only one strong diffracted beam is produced. In such cases the scattering is almost certainly dynamic, rather than kinematic. The intensity distribution of the fringes then bears no direct relationship to the lattice structure and Cowley (1959) and Hashimoto, Naiki & Mannami (1958) have shown that bending or changes of thickness of the crystals can give rise to distortion, changes of spacing and stepped structures in the fringes.

When each crystal gives a number of reflections simultaneously the several sets of fringes may combine to give a pattern periodic in two dimensions. In the case of very thin crystals, the diffraction pattern of each will approximate to the Fourier transform of a projection of the potential distribution of the crystal lattice. Then the resulting two-dimensionally periodic pattern may be related to the crystal structure and could conceivably be used to obtain information about the structure. Dowell, Farrant & Rees (1956) showed that in certain limiting cases the pattern may represent the Patterson function of the crystal projection.

A more general and detailed theoretical study is required in order to explore more fully the possibilities of obtaining structural information in this way and to determine the experimental requirements for their realization. For this purpose the formulation of

the physical optics of Cowley & Moodie (1958, hereafter referred to as I) is ideally suited. Successive scattering by two crystals may be treated by a simple extension of the methods used in considering the formation of Fourier images by single planar periodic objects (Cowley & Moodie, 1957*a*, *b*, *c*).

In this paper we apply the methods of I to the case of electrons scattered successively by two thin crystals since this appears to include the practical cases of greatest immediate interest. It will be apparent, however, that the treatment applies equally well to other forms of radiation and the appropriate periodic phase objects; for example, visible light and transparent optical diffraction gratings. Only minor changes of an obvious nature, are required to include cases of amplitude gratings and the well-known geometric moiré effects.

We assume coherent radiation from a point source. The way in which the results are modified when the source is of finite extent or of imperfect coherence may be inferred by comparison with the case of the single periodic object (Cowley & Moodie, 1957*c*) or may be calculated in detail by the methods used by Cowley & Moodie (1959*a*).

2. The point source and two thin crystals

As shown by Cowley & Moodie (1957*d*), if a crystal is so thin that Fresnel diffraction effects within it may be neglected (less than a few hundred Å for 50 kV. electrons), it acts as a phase grating, modifying an incident electron wave by multiplying the wave function by

$$q(x, y) = \exp \{i\sigma\varphi(x, y)\}, \qquad (1)$$

where $\sigma = \pi/\lambda W_0$. W_0 is the accelerating potential, and

$$\varphi(x, y) = \int_{-\infty}^{\infty} \varphi(x, y, z)dz ,$$

i.e., $\varphi(x, y)$ is the projection in the direction of the electron beam, the z-direction, of the potential distribution of the crystal.

The small amplitude-grating effect given by the inelastic scattering density function $\chi(x, y)$, could be included by making $\varphi(x, y)$ complex without affecting the arguments, but will be ignored here.

The projected potential distributions of the two crystals may be represented by the Fourier series

$$\varphi_1(x, y) = \sum_{h_1} \sum_{k_1} E_1(h_1, k_1) \exp\left\{-2\pi i \left(\frac{h_1 x}{a_1} + \frac{k_1 y}{b_1}\right)\right\},$$

$$\varphi_2(x, y) = \sum_{h_2} \sum_{k_2} E_2(h_2, k_2) \exp\left\{-2\pi i \left(\frac{h_2 x}{a_2} + \frac{k_2 y}{b_2}\right)\right\}. \quad (2)$$

The implied assumption that the a and b axes for both crystals are parallel to axes of coordinates does not involve any loss of generality since whatever the shape or orientation of the unit cells, the structures may be described in terms of lattice axes parallel to the axes of coordinates with any required degree of accuracy by making the a and b sufficiently large.

The function $q_1(x, y)$, representing the effects of the distribution $\varphi_1(x, y)$ in the wave function, is likewise periodic and can be written

$$q_1(x, y) = \sum_{h_1} \sum_{k_1} E_1'(h_1, k_1) \exp\left\{-2\pi i \left(\frac{h_1 x}{a_1} + \frac{k_1 y}{b_1}\right)\right\}, \quad (3)$$

and similarly $q_2(x, y)$ has Fourier coefficients $E_2'(h_2, k_2)$. Because of the ease of manipulation of δ-functions, we choose to work with the Fourier transforms of these functions:

$$Q_1(\xi, \eta) = \mathfrak{F}q_1(x, y)$$

$$= \sum_{h_1} \sum_{k_1} E_1'(h_1, k_1) . \delta\left(\xi - \frac{2\pi h_1}{a_1}, \eta - \frac{2\pi k_1}{b_1}\right). \quad (4)$$

We consider the system shown in Fig. 1, where the distance from the source to the first crystal is R_1, the distance between crystals is R_2 and the plane of observation is a distance R from the second crystal.
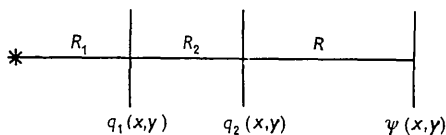

Fig. 1. The disposition of components and the notation used.

From the general expression, equation (3, 4) of I, it follows that the amplitude distribution on the plane of observation is then given by

$$\psi(x, y) = K \left[Q_2\left(-\frac{kx}{R}, -\frac{ky}{R}\right) * \left[Q_1\left(-\frac{kx}{R}, -\frac{ky}{R}\right) \right.\right.$$

$$\left. * Q_0\left(-\frac{kx}{R}, -\frac{ky}{R}\right) \exp\left\{\frac{ikR_1(x^2+y^2)}{2R^2}\right\} \right]$$

$$\left. \times \exp\left\{\frac{ikR_2(x^2+y^2)}{2R^2}\right\} * \exp\left\{\frac{ik(x^2+y^2)}{2R}\right\} \right]. \quad (5)$$

For a point source, represented by a δ-function at the origin of coordinates, we have

$$Q_0\left(-\frac{kx}{R}, -\frac{ky}{R}\right) = 1.$$

Substituting the expressions for Q_1 and Q_2 from (4) and evaluating the convolutions in (5) then gives

$$\psi(x, y) = K' \sum_{h_1} \sum_{k_1} \sum_{h_2} \sum_{k_2} E_1'(h_1, k_1) . E_2'(h_2, k_2)$$

$$\times \exp\left\{-\frac{\pi i \lambda R_1(R+R_2)}{R+R_1+R_2}\left(\frac{h_1^2}{a_1^2} + \frac{k_1^2}{b_1^2}\right)\right\}$$

$$\times \exp\left\{-\frac{\pi i \lambda R(R_1+R_2)}{(R+R_1+R_2)}\left(\frac{h_2^2}{a_2^2} + \frac{k_2^2}{b_2^2}\right)\right\}$$

$$\times \exp\left\{-\frac{2\pi i \lambda R R_1}{R+R_1+R_2}\left(\frac{h_1 h_2}{a_1 a_2} + \frac{k_1 k_2}{b_1 b_2}\right)\right\}$$

$$\times \exp\left\{-\frac{2\pi i}{R+R_1+R_2}\left[(R_1+R_2)\left(\frac{h_2 x}{a_2} + \frac{k_2 y}{b_2}\right)\right.\right.$$

$$\left.\left. + R_1\left(\frac{h_1 x}{a_1} + \frac{k_1 y}{b_1}\right)\right]\right\}. \quad (6)$$

The first three exponential terms of this expression may be regarded as 'focussing' terms, similar to those occurring in the theory of Fourier image formation by single crystals (Cowley & Moodie, 1957a). The condition that the first of these terms should be unity is

$$\frac{R_1(R_2+R)}{R+R_1+R_2} = \frac{2na_1^2}{\lambda} = \frac{2mb_1^2}{\lambda}, \quad (7)$$

where n and m are integers. This is the condition that an 'in focus' magnified or demagnified image of the first crystal should appear on the plane of observation. The second term similarly relates to the 'focussing' of a Fourier image of the second crystal on the plane of observation.

The third exponential term contains the interaction of waves scattered by both crystals and introduces an additional condition for focus

$$\frac{RR_1}{R+R_1+R_2} = \frac{2pa_1a_2}{\lambda} = \frac{2qb_1b_2}{\lambda},$$

where p and q are integers.

There is thus a total of six conditions for focus which cannot, in general, be satisfied exactly for any values of the distances R_1, R_2 and R except for the particular case $R = R_2 = 0$. In principle, by choosing the integers m, n, p, q, etc. sufficiently large the conditions may be satisfied to any desired degree of approximation but the distances involved may then prove impracticable. Exact focus may be obtained readily when the unit-cell dimensions are simply related, e.g., if $a_1 = a_2$, $b_1 = b_2$ and $Na^2 = Mb^2$, N and M being integers, or if $Pa_1 = Qa_2 = Rb_1 = Sb_2$, where P, Q, R and S are integers.

A case of particular interest is that for which $R = 0$

i.e., when, for example, an electron microscope is focussed on the plane of the second crystal. The equation (6) then reduces to

$$\psi(x, y) = K' \sum_{h_1} \sum_{k_1} \sum_{h_2} \sum_{k_2} E_1'(h_1, k_1) . E_2'(h_2, k_2)$$

$$\times \exp\left\{-\frac{\pi i \lambda R_1 R_2}{(R_1 + R_2)}\left(\frac{h_1^2}{a_1^2} + \frac{k_1^2}{b_1^2}\right)\right\}$$

$$\times \exp\left\{-2\pi i\left[\frac{R_1}{R_1 + R_2}\left(\frac{h_1 x}{a_1} + \frac{k_1 y}{b_1}\right)\right.\right.$$

$$\left.\left. + \left(\frac{h_2 x}{a_2} + \frac{k_2 y}{b_2}\right)\right]\right\} . \quad (8)$$

Only one focussing condition, namely (7), then remains. This is the condition that the second crystal lies in the plane of an in-focus Fourier image of the first crystal.

If all focussing conditions are satisfied, the expressions (6) and (8) represent the product of two transmission functions, appropriately scaled, of periodic phase objects. The function $\psi(x, y)$ then has a modulus equal to a constant. The intensity distribution is uniform, with no contrast due to the crystal lattice structures. Appreciable contrast may be generated in two ways: by restricting the aperture of the observing system, or by going out of focus. The out-of-focus images will in general have intensity distributions which are very complicated and difficult to interpret. It can be shown that in the limiting case of very small defect of focus the intensity distribution may be interpreted readily but then no moiré-like effects can be observed (Cowley & Moodie, to be published).

3. Effect of a limited aperture in the observing system

For convenience we now consider equation (8) for which $R = 0$. If we include the focussing term involving h_1^2 and k_1^2 in $E_1'(h_1, k_1)$ and include the magnification term $R_1/(R_1 + R_2)$ in the a_1 and b_1, we get the multiplication of two Fourier series which can be written in vector form as

$$\psi(x, y) = K' \sum_{h} \sum_{h'} G(h) . G'(h') \exp\{2\pi i(h + h') . r\} \quad (9)$$

where $h = ha^* + kb^*$, $h' = h'a'^* + k'b'^*$, and a^*, b^* and a'^*, b'^* are the unit-cell axes of the reciprocal lattices of the two functions and correspond to the real unit-cell axes a, b and a', b' which, in general, are neither equal nor parallel.

The general equation (6) can be reduced to the same form if all focussing term are unity.

The unit-cell origins of the two lattices will coincide, to a given approximation, only at a set of points which may be widely spaced relative to the unit-cell dimensions. These points will be disposed periodically and so will define a superlattice with unit-cell vectors

$$\begin{aligned} A &= pa + qb = p'a' + q'b', \\ B &= ra + sb = r'a' + s'b', \end{aligned} \quad (10)$$

where p, q, r, s, p', q', r' and s' are integers.

The vectors in two-dimensional reciprocal, or Fourier transform space, corresponding to A and B are then A^* and B^* defined by the relations $A^* = (B \times C)/|A \times B|$ and $B^* = (C \times A)/|A \times B|$, where the C axis is taken to be of unit length and in the direction of $A \times B$.

By using the analogous relations between a, b and a^*, b^* we then obtain

$$A^* = (sa^* - rb^*)/(ps - rq) = (s'a'^* - r'b'^*)/(p's' - r'q') ,$$
$$B^* = (-qa^* + pb^*)/(ps - rq)$$
$$= (-q'a'^* + p'b'^*)/(p's' - r'q') . \quad (11)$$

It is possible to choose indices H, K such that

$$H = HA^* + KB^* = h + h' . \quad (12)$$

The vector so defined then gives the position of the point in reciprocal space corresponding to a twice-diffracted beam.

Except in the case of phthalocyanines and a few other crystals with strongly diffracting planes having spacings of 10 Å or more, even the grosser features of crystal lattice structure have not yet been resolved by electron microscopes. The aberrations of the objective lens prevent the coherent recombination of the diffracted beams with the undiffracted beam. The effect is approximately that of placing an aperture in the back-focal plane of an ideal objective lens to remove all of the diffraction pattern except for a small region around the central spot. None of the singly diffracted beams may pass through this aperture but some doubly diffracted beams may do so.

Hence we consider that only those electron beams corresponding to points near the reciprocal lattice origin contribute to the image. We limit H to an area around the origin not greater than that of the reciprocal lattice unit cell of either crystal, i.e.,

$$\begin{aligned} |H.a^*| &< \tfrac{1}{2}|a^*|^2; \quad |H.a'^*| < \tfrac{1}{2}|a'^*|^2; \\ |H.b^*| &< \tfrac{1}{2}|b^*|^2; \quad |H.b'^*| < \tfrac{1}{2}|b'^*|^2. \end{aligned} \quad (13)$$

It can readily be seen that, if the reciprocal lattice of one crystal is laid down with its origin displaced to coincide with any point of the reciprocal lattice of the other crystal, not more than one reciprocal lattice point from the first crystal will fall within the region defined by (13). Hence for this region there is a one-to-one correspondence between the possible values of h, h' and H.

The double summation in (9) then reduces to a single summation

$$\psi'(x, y) = \sum_{h} G(h) . G'(h') \exp\{2\pi i(H.r)\} , \quad (14)$$

where h' and H may be expressed in terms of h.

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This equation has a particularly interesting interpretation when the dimensions of the Fourier images of the two crystals are so related that the value of h' associated with h is $\bar{\mathbf{h}}$, i.e., when $h' = -h$, $k' = -k$.

Then $\mathbf{H} = \mathbf{h} + \mathbf{h}' = h(\mathbf{a}^* - \mathbf{a}'^*) + k(\mathbf{b}^* - \mathbf{b}'^*)$, so that

$$\left.\begin{array}{ll} H = h\,, & A^* = \mathbf{a}^* - \mathbf{a}'^*\,, \\ K = k\,, & B^* = \mathbf{b}^* - \mathbf{b}'^*\,, \end{array}\right\} \quad (15)$$

and equation (14) becomes

$$\psi'(x, y) = \sum_{\mathbf{H}} G(H, K).G'(-H, -K) \exp\{2\pi i(\mathbf{H}.\mathbf{r})\}\,. \quad (16)$$

This represents the convolution of the two functions describing the Fourier images of the two crystals, related to the periodic lattice defined by the vectors A and B. In particular, if the focussing term of the equation (8) is unity, the convolution becomes

$$\psi'(x, y) = q_1(X, Y) * q_2(-X, -Y)\,, \quad (17)$$

where the coordinates X and Y are used to indicate that there is a change of scale, and the relative lengths and orientations of the axes have been changed.

In the particular case that the two crystals are identical, $\psi'(x, y)$ becomes the suitably scaled Patterson function of $q_1(x, y) = \exp\{i\sigma\varphi(x, y)\}$. It has been shown by Cowley & Moodie (1959b) that the Patterson function becomes identical with the kinematic Patterson function, $P(x, y) = \sigma^2 \varphi(x, y) * \varphi(-x, -y)$, except for a constant term, in the limiting case of zero thickness and that for the range of thickness considered here it is related to the kinematic Patterson function in a relatively simple way. Similarly it may be argued that the convolution (17) represents an approximation to the kinematic convolution $\varphi_1(x, y) * \varphi_2(-x, -y)$ and deviates from this kinematic form in a qualitatively predictable way with increasing thickness.

The conditions under which such magnified convolution functions may be observed are obtained from the equations (13) by inserting the restrictions (15) to give

$$|H\{|\mathbf{a}^*|^2 - (\mathbf{a}^*.\mathbf{a}'^*)\} + K\{(\mathbf{a}^*.\mathbf{b}^*) - (\mathbf{a}^*.\mathbf{b}'^*)\}| < \tfrac{1}{2}|\mathbf{a}^*|^2$$
$$|H\{(\mathbf{a}^*.\mathbf{b}^*) - (\mathbf{a}'^*.\mathbf{b}^*)\} + K\{|\mathbf{b}^*|^2 - (b'^*.b^*)\}| < \tfrac{1}{2}|b^*|^2$$

and so on.

Applying the relationships between reciprocal lattice vectors in equation (11) leads to four equations of the type

$$\left| H\left\{ 1 - \left(\frac{p's - q'r}{ps - qr}\right) - \frac{(\mathbf{a}^*.\mathbf{b}^*)}{|\mathbf{a}^*|^2}\left(\frac{pr' - rp'}{ps - qr}\right)\right\} \right.$$
$$\left. + K\left\{\frac{(\mathbf{a}^*.\mathbf{b}^*)}{|\mathbf{a}^*|^2}\left(1 - \frac{ps' - rq'}{ps - qr}\right) - \frac{sq' - qs'}{ps - qr}\right\}\right| < \tfrac{1}{2}\,. \quad (18)$$

In particular, if the unit cells are rectangular so that terms of the type $(\mathbf{a}^*.\mathbf{b}^*)$ are zero, we get

$$\left| H\left(1 - \frac{p's - q'r}{ps - qr}\right) - K\left(\frac{q's - s'q}{ps - qr}\right)\right| < \tfrac{1}{2}\,,$$
$$\left| -H\left(\frac{pr' - rp'}{ps - qr}\right) + K\left(1 - \frac{ps' - q'r}{ps - qr}\right)\right| < \tfrac{1}{2}\,, \quad (19)$$

and analogous expressions with $(p's' - q'r')$ in the denominators. These expressions limit the range of H and K values, and so determine the number of terms of the Fourier series (16) which are included. In order that the convolution function should be resolved as well as possible, therefore, it is necessary that equations (19) should be satisfied by $H_{\max}$ and $K_{\max}$ which are the maximum values of the indices of spots in the diffraction pattern of either crystal for which the spot intensities are appreciable.

An indication of the number of terms of the series (16) obtained for any particular set of constants, $p, q, r, s, p', q', r', s'$, or of the relative magnitudes of these constants required for a given resolution in the convolution image, may be obtained by considering the limitation of the diffraction pattern in the directions of the axes.

For example, when

$$K = 0, \quad |H| < \tfrac{1}{2}\left|\frac{ps - qr}{(p - p')s - (r - r')q}\right| \quad (20a)$$

and

$$|H| < \tfrac{1}{2}\left|\frac{ps - qr}{rp' - pr'}\right| \quad (20b)$$

and when

$$H = 0, \quad |K| < \tfrac{1}{2}\left|\frac{ps - qr}{(s - s')p - (q - q')r}\right| \quad (20c)$$

and

$$|K| < \tfrac{1}{2}\left|\frac{ps - qr}{q's - s'q}\right| \quad (20d)$$

and similar inequalities hold when the primed and unprimed constants are interchanged.

4. Examples and applications

(a) Identical crystals superimposed with rotation

We consider first the case of identical crystals with rectangular unit cells, so thin that when they are placed in contact we may take $R_2 = 0$. This is a good approximation if the crystal thickness is less than the spacing between Fourier images for a sinusoidal grating with periodicity $d_{\min}$ corresponding to an outer spot of the diffraction pattern

$$\text{i.e.,} \quad R_2 < d_{\min}^2.\lambda\,.$$

For example, if $d = 2$ Å, $\lambda = 0.05$ Å the distance between crystal centres must be less than 80 Å.

If one of these crystals is rotated with respect to the other through a small angle, the origins of unit cells of the two crystals will coincide at points of a 'superlattice' defined by the axes

$$\mathbf{A} = p\mathbf{a}+q\mathbf{b}$$
$$\mathbf{B} = r\mathbf{a}+s\mathbf{b},$$

wherever integers p, q, r, s, can be found such that

$$\frac{pq}{rs} = -b^2/a^2.$$

If an electron microscope of limited aperture is focussed on a Fourier image plane of the crystals (the $R = 0$ case being included as the zero-order Fourier image) the amplitude in the plane of observation is given by $q_1(X, Y) * q_1(-X, -Y)$, which, for very thin crystals, represents the Patterson function of the potential distribution function, magnified by a factor of $|\mathbf{A}|/|\mathbf{a}| = |\mathbf{B}|/|\mathbf{b}| = (p^2a^2+q^2b^2)^{\frac{1}{2}}/a$. For example, if $p = 100$, $q = 1$, $r = -1$, $s = 100$ (and $p' = 100$, $q' = -1$, $r' = 1$, $s' = 100$), the Patterson function of the crystal projection will be observed with a unit cell approximately 100 times as large as the crystal lattice unit cell. The number of possible terms in the summation of (16) is indicated by the equations (20), from which we obtain $|H| < 50$, $|K| < 50$.

(b) Parallel identical crystals held apart

If the lattice axes of the crystals are exactly parallel but the crystals are separated by a distance R_2 such that one crystal lies on a Fourier image plane of the other, then $q_1(x, y)$ and $q_2(x, y)$ will be identical except for a scale factor. The superlattice unit cell is defined by the vectors $\mathbf{A} = p\mathbf{a} = (p+1)\mathbf{a}'$ and $\mathbf{B} = p\mathbf{b} = (p+1)\mathbf{b}'$ i.e., $s = p$, $q = r = q' = r' = 0$, $p' = s' = p+1$.

Equation (16) becomes

$$\psi'(x, y) = \sum_{\mathbf{H}} G(H, K).G(-H, -K)$$
$$\times \exp\left\{2\pi i\left(\frac{Hx}{pa} + \frac{Ky}{pb}\right)\right\},$$

which approximates to the Patterson function of the crystal potential distribution with unit-cell dimensions magnified p times. The condition that one crystal lies on a Fourier image plane of the other gives

$$\frac{p+1}{p} = \frac{R_1+R_2}{R_1} = \frac{R_2\lambda}{2na^2}.$$

Hence

$$p = R_1/R_2 = 2na^2/(R_2\lambda-2na^2).$$

For example, for $a = 10$ Å, $\lambda = 0.05$ Å and $R_1 = 0.1$ cm., $n = 5$ gives $R_2 = 2 \times 10^{-4}$ cm., and the magnification, $p = 500$.

If now one of the crystals is rotated through a small angle with respect to the other a combination of the effects (a) and (b) will be observed and by suitable adjustment of the parameters convolution functions with various magnifications may be obtained. In particular for R_1 infinite so that the crystals are illuminated by parallel irradiation, the Fourier image of one in the plane of the other will have unit magnification and consequently only the effect of the mutual rotation of the crystals remains. Magnified convolution functions are then observed exactly as in (a).

(c) Isomorphous crystals

Isomorphous replacement of one of the component atoms by another type of atom in a crystal structure frequently leads to very small differences in the unit-cell dimensions and the atomic positions. When two crystals which differ in this way are superimposed, the difference in unit-cell dimensions may be sufficient to produce a convolution function image visible in an electron microscope. In any case a convolution function image may be produced by a suitable rotation of one crystal or a separation of the crystals. If the potential distribution functions are represented by $\varphi_1(x, y)$ and $\varphi_2(x, y) = \varphi_1(x, y)+\varphi''(x, y)$, the convolution function has the limiting form

$$[\varphi_1(x, y) * \varphi_1(-x, -y)] + [\varphi_1(x, y) * \varphi''(-x, -y)]. \quad (21)$$

The first term represents the Patterson function image given when both crystals have the structure $\varphi_1(x, y)$. If this image is recorded and subtracted from the image (21) the difference gives the second term of (21). This second term may give an indication of the crystal structure. For example, if the crystals differ only in the replacement of a single light atom by a heavy atom, $\varphi''(x, y)$ will contain only one peak per unit cell and the second term of (21) will reduce to $\varphi_1(x, y)$, modified by a broadening of the peaks. Under these circumstances an image of the crystal structure can be deduced directly from electron microscope observations.

(d) Crystals of known and unknown structure

In general, if the structure of a crystal is unknown except for the unit-cell dimensions, it should be possible to choose a crystal of known structure such that when the crystal of unknown structure is placed suitably with respect to it, an electron microscope image of the convolution function, $\varphi_1(x,y) * \varphi_2(-x,-y)$ will be observable. This convolution function may then be analysed by the method of image-seeking frequently used in structure analysis by X-ray and electron diffraction methods for the interpretation of Patterson maps (Lipson & Cochran, 1953). The atomic positions of the known crystal structure may be used as the set of image-seeking points to obtain an approximate structure for the unknown crystal.

5. Discussion

The finite size of the electron sources used in electron microscopy is not a serious limiting factor in the resolution obtainable in the convolution images. This resolution depends on the resolution of the individual

Fourier images of the two crystals. In our paper dealing with the effects of finite source size in the Fourier images of a grating (Cowley & Moodie, 1957c) it is shown that the least resolvable distance in a Fourier image, referred to the scale of the crystal lattice is approximately $\Delta = R\sigma/(R+Rq)$ where σ is the source diameter, R_q is the distance from source to grating and R is the distance from grating to the plane of observation. Since in the present application it is not necessary to work with Fourier images of appreciable magnification, the ratio of R_q to R can be made large. In particular, if the electron microscope is focussed on the second crystal there is virtually no restriction on source size if the first crystal is considered sufficiently close so that one can take $R_2 = 0$. If the crystals are separated by a distance of the order of a micron, and the convolution image is produced by rotation or difference of unit-cell dimensions rather than by obtaining a magnified Fourier image, the sources commonly used are adequate. For example, with $R = R_2 = 1\mu$, $R_1 = Rq = 20$ cm. and $\sigma = 20\mu$, the least resolvable distance, referred to the scale of the crystal lattice is 1 Å.

If the source is brought close to the crystals in order to obtain a magnified image of one crystal on the other, as in 4(b) above, a smaller source must be used. For example, in the case cited in 4(b), where $R_1 = 0.1$ cm., $R_2 = 2 \times 10^{-4}$ cm., a resolution of 1 Å is given by a source of diameter 500 Å.

We have considered in detail only the relatively simple cases for which at least two of the three focussing terms in the equation (6) are equal to unity. In general equation (6) can not be reduced to the form (9) or expressed as a convolution. The interpretation of patterns obtained with appreciable defect of focus would therefore be very difficult.

The theoretical treatment here applied to the case of two thin crystals can be extended without difficulty to deal with the cases of three, four or more thin crystals, or, in fact, arbitrary assemblies of periodic or non-periodic objects including perfect thick crystals (Cowley & Moodie, 1957d). We defer discussion of these matters except to notice the interesting possibility that with three crystals suitably related the magnified image formed from the central part of the diffraction pattern may take the form

$$\psi'(x, y) = q_1(x, y) * q_2(x, y) * q_3(x, y) .$$

This could occur, for example, if the first two crystals were identical and gave a pattern of doubly diffracted spots which almost coincides with the spots of the primary diffraction pattern of the third crystal. The magnified image would then approximate to an image of the third crystal lattice, since in the Patterson function the origin peak is usually much higher than any other. However, it should be noted that the restrictions on the distances between, and relative rotations of, the three crystals would be much more severe than in the case of two crystals, and the image intensity would be very much less. The use of three crystals in this way is, therefore, scarcely a practical proposition.

The experiments of Dowell, Farrant & Rees (1956) and others confirm our conclusions that, for the two-crystal case, the electron optical requirements may be met with conventional electron microscopes. The principal experimental difficulties in the observation of the convolution images are the preparation and manipulation of suitable crystals. The crystal thickness must be very small, of the order of a hundred Ångströms, and the diameter of the crystals must be sufficient to allow at least one, and preferably many unit cells of the convolution image to be observed i.e., thousands of Ångströms. Unless one relies on the chance observation of two crystals lying together with a suitable relative orientation, manipulation of one crystal is required with an accuracy of a fraction of a micron in translation and a fraction of a degree in rotation.

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