4.3. DIFFUSE SCATTERING IN ELECTRON DIFFRACTION

$$I(\mathbf{u} + \mathbf{h}) = \sum_j |C_h^j|^2 \left\{ A_1^j \sum_g \sum_{g \neq g'} F_1(\mathbf{u}, \mathbf{g}, \mathbf{g'}) C_g^j C_{g'}^{j*} \right.$$

$$\left. + \sum_n \sum_g A_{ng}^j F_n(\mathbf{u}, \mathbf{g}) |C_g^j|^2 \right\}, \qquad (4.3.4.10)$$

where $F_n$ are normalized scattering factors for $n$th-order multiple diffuse scattering and $A_n^j$ are multiple-scattering coefficients which include absorption.

When the thickness is increased, the variation of $F_n(\mathbf{u}, \mathbf{g})$ with angle becomes slower, and an expression for intensity of the channelling pattern is obtained (Gjønnes & Taftø, 1976):

$$I(\mathbf{u} + \mathbf{h}) = \sum_j \sum_g \sum_n |C_h^j|^2 |C_g^j|^2 A_n^j$$

$$= \sum_j |C_h^j|^2 \sum_n A_n^j \rightarrow |C_h^j|^2 / \mu^j(\mathbf{u}). \qquad (4.3.4.11)$$

Another approach is the use of a modified diffusion equation (Ohtsuki *et al.*, 1976).

These expressions seem to reproduce the development of the general background with thickness over a wide range of thicknesses. It may thus appear that the contribution to the diffuse background from known sources can be treated adequately – and that such a procedure must be included together with adequate filtering of the inelastic component in order to improve the quantitative interpretation of diffuse scattering.

### 4.3.5. Multislice calculations for diffraction and imaging

The description of dynamical diffraction in terms of the progression of a wave through successive thin slices of a crystal (Chapter 5.2) forms the basis for the multislice method for the calculation of electron-diffraction patterns and electron-microscope images [see Section 4.3.6.1 in *IT* C (1999)]. This method can be applied directly to the calculations of diffuse scattering in electron diffraction due to thermal motion and positional disorder and for calculating the images of defects in crystals.

It is essentially an amplitude calculation based on the formulation of equation (4.3.4.1) [or (4.3.4.2)] for first-order diffuse scattering. The Bragg scattering in the first part of the crystal is calculated using a standard multislice method for the set of beams **h**. In the $n$th slice of the crystal, a diffuse-scattering amplitude $\Psi_d(\mathbf{u})$ is convoluted with the incident set of Bragg beams. For each **u**, propagation of the set of beams $\mathbf{u} + \mathbf{h}$ is then calculated through the remaining slices of the crystal. The intensities for the exit wave at the set of points $\mathbf{u} + \mathbf{h}$ are then calculated by adding either amplitudes or intensities. Amplitudes are added if there is correlation between the defects in successive slices. Intensities are added if there are no such correlations. The process is repeated for all **u** values to obtain a complete mapping of the diffuse scattering.

Calculations have been made in this way, for example, for short-range order in alloys (Fisher, 1969) and also for TDS on the assumption of both correlated and uncorrelated atomic motions (Doyle, 1969). The effects of the correlations were shown to be small.

This computing method is not practical for electron-microscope images in which individual defects are to be imaged. The perturbations of the exit wavefunction due to individual defects (vacancies, replaced atoms, displaced atoms) or small groups of defects may then be calculated with arbitrary accuracy by use of the 'periodic continuation' form of the multislice computer programs in which an artificial, large, superlattice unit cell is assumed [Section

4.3.6.1 in *IT* C (1999)]. The corresponding images and micro-diffraction patterns from the individual defects or clusters may then be calculated (Fields & Cowley, 1978). A more recent discussion of the image calculations, particularly in relation to thermal diffuse scattering, is given by Cowley (1988).

In order to calculate the diffuse-scattering distributions from disordered systems or from a crystal with atoms in thermal motion by the multislice method with periodic continuation, it would be necessary to calculate for a number of different defect configurations sufficiently large to provide an adequate representation of the statistics of the disordered system. However, it has been shown by Cowley & Fields (1979) that, if the single-diffuse-scattering approximation is made, the perturbations of the exit wave due to individual defects are characteristic of the defect type and of the slice number and may be added, so that a considerable simplification of the computing process is possible. Methods for calculating diffuse scattering in electron-diffraction patterns using the multislice approach are described by Tanaka & Cowley (1987) and Cowley (1989). Loane *et al.* (1991) introduced the concept of 'frozen phonons' for multislice calculations of thermal scattering.

### 4.3.6. Qualitative interpretation of diffuse scattering of electrons

Quantitative interpretation of the intensity of diffuse scattering by calculation of *e.g.* short-range-order parameters has been the exception. Most studies have been directed to qualitative features and their variation with composition, treatment *etc.* Many features in the scattering which pass unrecognized in extensive X-ray or neutron investigations will be observed readily with electrons, frequently inviting other ways of interpretation.

Most such studies have been concerned with substitutional disorder, but the extensive investigations of thermal streaks by Honjo and co-workers should be mentioned (Honjo *et al.*, 1964). Diffuse spots and streaks from disorder have been observed from a wide range of substances. The most frequent may be streaks due to planar faults, one of the most common objects studied by electron microscopy. Diffraction patterns are usually sufficient to determine the orientation and the fault vector; the positions and distribution of faults are more easily seen by dark-field microscopy, whereas the detailed atomic arrangement is best studied by high-resolution imaging of the structure [Section 4.3.8 in *IT* C (1999)].

This combination of diffraction and different imaging techniques cannot be applied in the same way to the study of the essentially three-dimensional substitutional local order. Considerable effort has therefore been made to interpret the details of diffuse scattering, leaving the determination of the short-range-order (SRO) parameters usually to X-ray or neutron studies.

Frequently, characteristic shapes or splitting of the diffuse spots from *e.g.* binary alloys are observed. They reflect order extending over many atomic distances, and have been assumed to arise from forces other than the near-neighbour pair forces invoked in the theory of local order. A relationship between the diffuse-scattering distribution and the Fourier transform of the effective atom-pair-interaction potential is given by the ordering theory of Clapp & Moss (1968). An interpretation in terms of long-range forces carried by the conduction electrons was proposed by Krivoglaz (1969). Extensive studies of alloy systems (Ohshima & Watanabe, 1973) show that the separations, $m$, observed in split diffuse spots from many alloys follow the predicted variation with the electron/atom ratio $e/a$:

$$m = \left[ \frac{12}{\pi} (e/a) \right]^{1/3} t - \sqrt{2},$$

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