High-resolution electron-microscope images of crystals with correlated atomic displacements

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Dedicated to Professor A. F. Moodie on the occasion of his 75th birthday

Abstract

A coordinate-space multislice description of the scattering of high-energy electrons is constructed from consecutions of differential operators acting upon atomic potentials. It is used to find expressions for the intensity distribution in high-resolution electron-microscope images of crystals whose atoms are periodically displaced relative to a reference lattice according to a modulation wave. Both static correlated displacements, such as occur in modulated structures, and time-dependent correlated displacements, as are generated by phonons, are considered. Two aspects of the image are examined in detail; its translational symmetry and its dependence upon the correlations between the atomic displacements. It is shown that the intensity distribution due to scattering from static correlated displacements has the translational symmetry of the modulated structure in that projection, as determined by the component of the modulation wavevector perpendicular to the incident beam, whereas that due to scattering from phonons has the translational symmetry of the reference lattice in that projection. The former is a consequence of higher-order Laue-zone interactions. The intensity distribution due to scattering from static displacements depends upon the absolute phase of the displacement at each scattering atomic site whereas that due to scattering from phonons depends only upon the relative phase of the displacements between different scattering sites, both within the same atomic column parallel to the beam and in adjacent columns. In both cases, the influence of the component of the correlation wavevector parallel to the incident beam is different to that perpendicular to the beam; the former affects the intensity mostly at the atomic sites whilst the latter affects the intensity mostly between the atomic sites. It is also observed that, as a consequence of the periodic nature of the polarization-vector function, the interference terms are small, both relative to the non-interference term and in an absolute sense, particularly for phonon scattering. For this reason, the contribution to the image due to scattering from correlated atomic displacements will have greater and sharper atomic contrast than that due to scattering from the reference structure without displacements. In addition, this component of the intensity distribution will not exhibit strong contrast reversal when the objective-lens defocus is changed.

1. Introduction

Crystal structures are conveniently described by reference to an idealized lattice with three-dimensional space-group symmetry. The actual structure can then be characterized in terms of displacements of atoms from their reference lattice site. As a consequence of interatomic forces, atomic displacements tend to be correlated with those of their neighbours over some range. Phonons and modulated structures are examples of long-range correlated atomic displacements. In each case, the atoms of the structure are periodically displaced from their reference site according to a modulation wave. In the first case, the wave is propagating, in the second it is not. Either way, the configuration of atoms at any instant will not, in general, have the three-dimensional space-group symmetry of the reference lattice. What then is the translational symmetry of the high-resolution electron-microscope image of such a crystal? This is the principal question addressed in this paper.

It is important in general to understand how the wave function of an incident high-energy electron is influenced by its interaction with such correlated atomic displacements and how this in turn affects the electron diffraction pattern and image of a crystal. This problem has been tackled for the case of phonons in a variety of ways, with varying degrees of approximation both in the scattering process and in the crystal potential. (An overview of these methods is given by Fanidis *et al.*, 1992). In this paper, it is the specific purpose to examine the spatial periodicity of high-resolution electron-microscope (HREM) images of crystals with both static and dynamic correlated atomic displacements and to investigate the extent to which the correlations between displaced atoms affect the intensity distribution of this image.

To this end, a coordinate-space multislice description of electron scattering is constructed in which the exit wave function is expressed as sums of noncommuting 'products' of differential operators and atomic potentials. The contribution of scattering from each atomic potential in the crystal is thus exhibited explicitly, so that the fomulation lends itself to an assessment of the effect upon the image of a displaced atom at a given site and of correlations between displaced atoms. Expressions can be constructed for the intensity distribution in the lattice image due to dynamical scattering from displaced as well as non-displaced atoms, or, in other words, due to multiple 'diffuse' as well as multiple 'Bragg' scattering, and are written out explicitly for the case of single and double-diffuse scattering. They are, however, designed only to provide physical insight and not for numerical convenience.

The analysis is set out as follows. Firstly, a conventional and general description of the potential of a crystal with correlated atomic displacements, either static or dynamic, is defined in §2. Then, the independent task of constructing a dynamical description of the forward scattering of fast electrons is dealt with in §3. Here the multislice solution is recast into an unconventional coordinate-space representation that does not involve the integral operation of convolution. This formulation is then used in §4 to find unabridged expressions for the intensity distribution at the image plane due to scattering from a crystal with static or dynamic correlated atomic displacements (as defined in §2). From these expressions, the translational symmetry of the image in each case is determined (§4) and the influence of the relative phase of atomic displacements on the image considered (§5). A few observations are also made with respect to the contrast in the intensity distribution due to scattering from correlated displacements.

2. The lattice potential

Consider a crystal structure which is periodically deformed relative to a reference structure with three-dimensional space-group symmetry, according to a modulation wave, m, with wavevector, $\mathbf{q}$. Define the reference structure by the set of points $\{\overline{\mathbf{R}}_{ij} : i = 1, \ldots, n$ atoms, $j = 1, \ldots$ unit cells$\}$ with translation vector $\mathbf{R}_0$, where $\overline{\mathbf{R}}_{ij}$ is the reference position of atom i in unit cell j, and each reference atomic site in a given unit cell is labelled identically to that site in every other cell, so that $\overline{\mathbf{R}}_{ij} + \mathbf{R}_0 = \overline{\mathbf{R}}_{ij'}$.

The potential, at instant t (t may represent time or be constant), of this crystal with atomic displacements $\mathbf{h}_{ij}^{\mathbf{q}m}(t)$ from its reference lattice sites $\overline{\mathbf{R}}_{ij}$ can be given by

$$V^{\mathbf{q}m}(\mathbf{R}, t) = \sum_i \sum_j V_i(\mathbf{R}) * \delta[\mathbf{R} - \overline{\mathbf{R}}_{ij} - \mathbf{h}_{ij}^{\mathbf{q}m}(t)], \quad (1)$$

where $\mathbf{R} = (x, y, z)$ is taken from an origin that can be defined arbitrarily and $V_i(\mathbf{R})$ is the potential contributed by each atom i, independent of its cell.

As a consequence of Bloch's theorem, the atom i in each cell has a displacement or polarization vector function, $\mathbf{h}_{ij}^{\mathbf{q}m}(t) = h_{ij}^{\mathbf{q}m}(t)\hat{\mathbf{h}}_i^{\mathbf{q}m}$, with the same direction, $\hat{\mathbf{h}}_i^{\mathbf{q}m}$, and same amplitude, $h_{ij}^{\mathbf{q}m}(t) = h_i^{\mathbf{q}m}(\theta_{ij} + \omega_{qm}t)$, up to a cell-dependent phase, $\theta_{ij} = \overline{\mathbf{R}}_{ij} \cdot \mathbf{q}$ (for example, Ziman, 1986). It is thus only this cell-dependent phase that causes $V^{\mathbf{q}m}(\mathbf{R}, t)$ not to have the translational symmetry of the reference lattice at any instant t. Note that the amplitude, $h_i^{\mathbf{q}m}(\theta_{ij} + \omega_{qm}t)$, can take any periodic form and that $\mathbf{q}$ may be either commensurate or incommensurate with the reference lattice. In the latter case, the structure at any instant, t, will of course not have three-dimensional translational symmetry.

Provided the atomic displacements are small relative to the lattice constants of the reference lattice, the potential may be expanded as a Taylor series in atomic displacements thus separating out the terms depending upon t.

$$V^{\mathbf{q}m}(\mathbf{R}, t) = \sum_i \sum_j V_i(\mathbf{R} - \overline{\mathbf{R}}_{ij})$$

$$+ \sum_i \sum_j \sum_{a=1}^{\infty} (1/a!)[\mathbf{h}_{ij}^{\mathbf{q}m}(t) \cdot \nabla]^a V_i(\mathbf{R} - \overline{\mathbf{R}}_{ij})$$

$$= b(\mathbf{R}) + d^{\mathbf{q}m}(\mathbf{R}, t). \quad (2)$$

Here $b(\mathbf{R})$ is the zeroth-order term which is just the potential of the reference lattice and thus, by definition, has the translation vector of that lattice. $d^{\mathbf{q}m}(\mathbf{R}, t)$ is the sum of the higher-order terms. Although each of these terms consists of cell- and t-independent partial derivatives of the function, $V_i(\mathbf{R})$, located at the reference lattice sites, $\overline{\mathbf{R}}_{ij}$, they are modulated by powers of the cell-*dependent* amplitudes, $h_{ij}^{\mathbf{q}m}(t)$, and their sum will thus have the translational symmetry of the actual crystal as determined by $\mathbf{q}$, at any given instant, t.

This potential describes in a general way a crystal with correlated atomic displacements. If t is a constant, then it defines a crystal with a modulated structure. If t represents time, then it can define a crystal with a phonon mode m and wavevector $\mathbf{q}$.

It is a common and useful approach in diffraction physics, when estimating the contribution of phonon scattering to diffracted intensities, to take the time average of this potential *before* scattering from it. The second term ($a = 2$) of the above potential then becomes the well known Debye–Waller factor. However, this is an approximation that will not be made here since it leads to erroneous conclusions about translational symmetry, the subject of the present work.

In order to consider the intensity distribution in a high-resolution electron-microscope image of a crystal with this potential, a coordinate-space multislice

formulation of electron scattering is first constructed in the following section.

3. Coordinate-space multislice description of electron scattering

The elastic scattering of an electron, represented by the wave function $\Psi(\mathbf{R})$ and a potential $V(\mathbf{R})$, is described by a Klein–Gordan equation in the form,

$$[\nabla^2 + k^2 + 2k\sigma V]\Psi = 0, \tag{3}$$

where the interaction constant $\sigma = 2\pi me\lambda/h^2$, m is the relativistic mass of the electron, k is the scalar wave number with magnitude $2\pi/\lambda$ and λ is the relativistic wavelength,

$$\lambda = h[2m_0|e|W(1 + |e|W/2m_0c^2)]^{-1/2}$$

with W the accelerating voltage and e and m_0 the charge and rest mass of the electron, respectively.

Define the incident beam to be parallel to the z direction. For high-energy electrons ($W \geq 10^2$ keV), the influence of the specimen potential on the z component of the electron wavefunction is negligible. It is thus appropriate to extract the principal z dependence from the wave function *via* the substitution $\Psi(\mathbf{r}, z) = \psi(\mathbf{r}, z)\exp ikz$, where $\mathbf{r} = (x, y)$, and to neglect $\partial^2\psi/\partial z^2$ to give an equation describing forward scattering,

$$\partial\psi/\partial z = (i/2k)(\nabla_\mathbf{r}^2 + 2k\sigma V)\psi, \tag{4}$$

where

$$\nabla_\mathbf{r}^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} = \Delta.$$

This has the form of a two-dimensional 'time'-dependent Schrödinger equation.

The finite difference form (Feynman, 1948) of this equation was derived by Cowley & Moodie (1957) using an argument intuitively based in physical optics,

$$\psi_{n+1}(\mathbf{r})$$
$$= \exp(ikr^2/2\varepsilon) * \left\{ \exp\left[i\sigma \int_{z_n}^{z_n+\varepsilon} V(\mathbf{r}, z)\, dz \right] \psi_n(\mathbf{r}) \right\}, \tag{5}$$

where

$$\int_{z_n}^{z_n+\varepsilon} V(\mathbf{r}, z)\, dz = \varepsilon V(\mathbf{r}, z_n) = \varepsilon V(\mathbf{r}, z_{n+1}) = \varepsilon V_{n+1}(\mathbf{r}).$$

The index n refers to the nth slice, of thickness ε, of a crystal partitioned by N two-dimensional phase objects, $\varepsilon V_n(\mathbf{r})$, separated by vacuum. Equation (5) describes the wave function at the exit face of the $(n + 1)$th slice, ψ_{n+1}, in terms of the wave function at the exit face of the nth slice, ψ_n. As ψ_n impinges on the $(n + 1)$th slice, it is perturbed by the $(n + 1)$th projected potential, suffering

a phase change before propagating through the vacuum *via* Fresnel diffraction to the exit face of the $(n + 1)$th slice.

In order to elucidate the impact of the modulated potential, (2), on the wave function, (5), it is helpful to construct a series expansion in coordinate space of ψ in terms of the projected potentials of each slice and their differentials. To this end, the convolution in (5) is rewritten in differential form† (Stuart, 1987):

$$\psi_{n+1}(\mathbf{r}) = \int_{-\infty}^{\infty} \exp(ikr_1^2/2\varepsilon)$$
$$\times \{\exp[i\sigma\varepsilon V_{n+1}(\mathbf{r} - \mathbf{r}_1)]\psi_n(\mathbf{r} - \mathbf{r}_1)\}\, d\mathbf{r}_1$$
$$= \int_{-\infty}^{\infty} \exp(ikr_1^2/2\varepsilon)\exp(-\mathbf{r}_1 \cdot \nabla_\mathbf{r})$$
$$\times \{\exp[i\sigma\varepsilon V_{n+1}(\mathbf{r})]\psi_n(\mathbf{r})\}\, d\mathbf{r}_1$$
$$= \exp[(i\varepsilon/2k)\Delta]\{\exp[i\sigma\varepsilon V_{n+1}(\mathbf{r})]\psi_n(\mathbf{r})\}. \tag{6}$$

In this representation, the kinetic energy (differential) term describes the diffusion of the wave function as it propagates across the slice, whilst the potential term [unchanged from (5)] describes the scattering by the slice.

[It is noted in passing that equation (6) is an approximation to the equation

$$\psi_{n+1}(\mathbf{r}) = \psi(\mathbf{r}, z_n + \varepsilon)$$
$$= \exp\{(i/2k)[\Delta + 2k\sigma V_{n+1}(\mathbf{r})]\varepsilon\}\psi(\mathbf{r}, z_n), \tag{7}$$

which is the solution of the Schrödinger equation, (4), for a slice for which the potential is independent of z. This approximation neglects half the second-order and all the higher-order mixed scattering/propagation terms, however, the error incurred vanishes with slice thickness.]

Iterating (6), with the incident wave function set equal to unity, gives

$$\psi(\mathbf{r}) = \left[\exp(i\ell\varepsilon\Delta)\exp i\ell\varepsilon U_n(\mathbf{r})\big(\ldots\right.$$
$$\ldots \{\exp(i\ell\varepsilon\Delta)\exp i\ell\varepsilon U_2(\mathbf{r})$$
$$\times [\exp(i\ell\varepsilon\Delta)\exp i\ell\varepsilon U_1(\mathbf{r})]\} \ldots \big)\big], \tag{8}$$

where $\ell = 1/2k$ and $U_n = (2me/\hbar^2)V_n$ is a modified potential.

This gives an exact solution for the wave function in the limit $\varepsilon \to 0$, $N \to \infty$, $N\varepsilon \to D$, where D is the crystal thickness. Expanding each of the exponentials in (8) in Taylor's series gives

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† Stuart (1987) has given a rigorous account of the differential representation of convolution, which can be summarized as $f(\mathbf{r}) * g(\mathbf{r}) = \bar{f}[(i/2\pi)\nabla_\mathbf{r}] \cdot g(\mathbf{r})$, where $\bar{f}$ denotes the Fourier transform of the function f, defined as $\bar{f}(\mathbf{u}) = \int_{-\infty}^{\infty} f(\mathbf{r})\exp(2\pi i\mathbf{u} \cdot \mathbf{r})\, d\mathbf{r}$.

$$\psi(\mathbf{r}) = \sum_{\alpha_1=0}\sum_{\beta_1=0}\cdots\sum_{\alpha_N=0}\sum_{\beta_N=0}\frac{(i\ell\varepsilon)^{\alpha_1+\beta_1+\ldots+\alpha_N+\beta_N}}{\alpha_1!\beta_1!\ldots\alpha_N!\beta_N!}$$
$$\times\left[\Delta^{\alpha_N}\left(U_N^{\beta_N}(\mathbf{r})\Delta^{\alpha_{N-1}}\{U_{N-1}^{\beta_{N-1}}(\mathbf{r})\ldots\right.\right.$$
$$\left.\left.\ldots\Delta^{\alpha_2}[U_2^{\beta_2}(\mathbf{r})\Delta^{\alpha_1}U_1^{\beta_1}(\mathbf{r})]\ldots\}\right)\right]. \tag{9}$$

This describes the wave function explicitly as a consecution of scattering and diffusion processes that act simultaneously in the limit $\varepsilon\to0$, $N\to\infty$, $N\varepsilon\to D$. Note that α_n and β_n are integers only, the subscript n being used simply as a distinguishing label and not to represent a dependence upon n. Hereafter, for brevity, the brackets will not be written explicitly. It should then be remembered that each Δ operates on everything to its right.

This is the wave function at the exit face of the specimen. The wave function at the image plane, $\varphi(\mathbf{r})$ after perturbation by lens aberrations, can be approximated by

$$\varphi(\mathbf{r}) = s(\mathbf{r}) * \psi(\mathbf{r}), \tag{10}$$

where $s(\mathbf{r})$ is the Fourier transform of the transfer function $S(\mathbf{u}) = \exp i\chi(\mathbf{u})$ of the microscope. Using the differential form of convolution, (10) becomes

$$\varphi(\mathbf{r}) = \exp\{i\chi[(i/2\pi)\nabla_{\mathbf{r}}]\}\psi(\mathbf{r}). \tag{11}$$

This is equivalent to a modulation of the wave function on the back focal plane of the objective lens by $\exp i\chi(\mathbf{u})$. Using a typical form for $\chi(\mathbf{u})$ gives

$$\exp i\chi[(i/2\pi)\nabla_{\mathbf{r}}] = \exp\frac{i}{2}\left[-f\frac{\lambda}{2\pi}\nabla_{\mathbf{r}}^2 + \frac{C_s}{2}\left(\frac{\lambda}{2\pi}\right)^3(\nabla_{\mathbf{r}}^2)^2\right]$$
$$= 1 - if\ell\Delta + \left[-\tfrac{1}{2}(f\ell)^2 + i2C_s\ell^3\right]\Delta^2$$
$$+ \left[(i/6)(f\ell)^3 + 2fC_s\ell^4\right]\Delta^3 + \ldots$$
$$= S(\Delta), \tag{12}$$

where f is the defect of focus and C_s is the spherical aberration coefficient of the objective lens. Thus the wave function at the image plane, $\varphi(\mathbf{r})$, is equal to the wave function at the exit face, $\psi(\mathbf{r})$, operated on by a polynomial function of Δ, $S(\Delta)$,

$$\varphi(\mathbf{r}) = S(\Delta)\psi(\mathbf{r}). \tag{13}$$

It can be seen that the effect of $S(\Delta)$ on the series expansion of $\psi(\mathbf{r})$ in (9) is to raise the power of the left-hand differential operator.

4. Translational symmetry of HREM images of crystals with correlated atomic displacements

4.1. *Static displacements – modulated structures*

Now consider the wave function due to scattering from the configuration of atoms in (2) at a given value of t. So that the argument is not disguised behind a labyrinth of indices, consider the simplest case of one atom

per unit cell. (The argument remains unchanged with multiatom cells.) Partitioning the crystal with this potential into slices one atomic layer thick, that is, one unit cell thick, the modified potential of slice s projected perpendicular to z will then be

$$U_s^{\mathbf{q}m}(\mathbf{r}, t) = \sum_j U[\mathbf{r} - \bar{\mathbf{r}}_j - \mathbf{h}_{js}^{\mathbf{q}m}(t)]$$
$$= \sum_j U(\mathbf{r} - \bar{\mathbf{r}}_j)$$
$$+ \sum_j\sum_{a=1}^{\infty}(1/a!)[h_{js}^{\mathbf{q}m}(t)\hat{\mathbf{h}}^{\mathbf{q}m}\cdot\nabla_{\mathbf{r}}]^a U(\mathbf{r} - \bar{\mathbf{r}}_j)$$
$$= b_s(\mathbf{r}) + d_s^{\mathbf{q}m}(\mathbf{r}, t), \tag{14}$$

where $b_s(\mathbf{r})$ is the reference potential and $d_s^{\mathbf{q}m}(\mathbf{r}, t)$ is the 'displacement' potential of slice s. In (14), the indices js label each unit cell in the crystal; j in the (x, y) plane and s along the z direction, so that the atomic site in unit cell js of the reference lattice has position vector $\bar{\mathbf{R}}_{js} = (\bar{\mathbf{r}}_j, \bar{z}_s)$. For simplicity, it is assumed that the atom in the jth unit cell has the same reference position, $\bar{\mathbf{r}}_j$, in each slice; that is, $\bar{\mathbf{r}}_j$ refers to the jth column of atoms parallel to the beam. (This assumption is readily generalized and does not alter the argument.)

The component of atomic displacements parallel to the incident beam is averaged into the projected potential of the slice so that the direction, $\hat{\mathbf{h}}^{\mathbf{q}m}$, of the displacement vector, $\mathbf{h}_{js}^{\mathbf{q}m}(t)$, is now confined to the (x, y) plane. (The error incurred is negligible. Displacements parallel to the incident beam have an intrinsically different effect on the transmitted electron to those displaced in the plane perpendicular to the beam. Parallel displacements indirectly influence the propagator, albeit weakly, but do not alter the shape of the atomic potential, whereas perpendicular displacements effectively change the shape of the atomic potential but do not influence the propagator.) The phase of the displacement vector retains its z dependence and this is made explicit with the slice index, s, so that

$$\mathbf{h}_{js}^{\mathbf{q}m}(t) = h_{js}^{\mathbf{q}m}(t)\hat{\mathbf{h}}^{\mathbf{q}m} = h^{\mathbf{q}m}(\theta_{js} + \omega_{\mathbf{q}m}t)\hat{\mathbf{h}}^{\mathbf{q}m} \tag{15}$$

with

$$\theta_{js} = \bar{\mathbf{R}}_{js}\cdot\mathbf{q} = \bar{\mathbf{r}}_j\cdot\mathbf{q}_{xy} + \bar{z}_s q_z.$$

By substituting the potential (14) into the series expansion of ψ, (9), and operating with $S(\Delta)$, it is evident that the wave function at the image plane consists of configuration (t)-independent and -dependent parts,

$$\varphi(\mathbf{r}, t) = \varphi_b(\mathbf{r}) + \varphi_d^{\mathbf{q}m}(\mathbf{r}, t). \tag{16}$$

$\varphi_b(\mathbf{r})$ is the configuration-independent wave function at the image plane due to dynamical scattering from the reference lattice potential, $b(\mathbf{R})$, (2), and has the translation vector of the reference lattice in this projection since its terms consist exclusively of the reference slice potentials, $b_s(\mathbf{r})$, and their derivatives, that is,

$$\varphi_b(\mathbf{r}) = S(\Delta) \sum_{\substack{\alpha_1,\dots,\alpha_N \\ \beta_1,\dots,\beta_N}} \frac{(i\ell\varepsilon)^{\alpha_1+\beta_1+\dots+\alpha_N+\beta_N}}{\alpha_1!\beta_1!\dots\alpha_N!\beta_N!}$$
$$\times \Delta^{\alpha_N}[b_N(\mathbf{r})]^{\beta_N}\dots\Delta^{\alpha_1}[b_1(\mathbf{r})]^{\beta_1}, \qquad (17)$$

where it must be remembered that each Δ operates on everything to its right.

$\varphi_d^{\mathbf{q}m}(\mathbf{r}, t)$ is the correction term to $\varphi_b(\mathbf{r})$ due to scattering from the tth configuration of atomic displacements from the reference lattice sites.

In the corresponding electron diffraction pattern, $\varphi_b(\mathbf{r})$ is the component of the wave function that generates the periodic Bragg intensity peaks whilst $\varphi_d^{\mathbf{q}m}(\mathbf{r}, t)$ generates the diffuse intensity or superlattice reflections (depending on $\mathbf{q}$) between the Bragg peaks.

All those scattering processes that consist of scattering from non-displaced atoms *only* will contribute to $\varphi_b(\mathbf{r})$. These processes are sometimes referred to as 'Bragg scattering'. All other scattering processes, namely those that consist of scattering from one or more displaced atoms, as well as scattering from the remaining non-displaced atoms, contribute to $\varphi_d^{\mathbf{q}m}(\mathbf{r}, t)$. Processes in this category are sometimes referred to as 'diffuse scattering'.

The terms of $\varphi_d^{\mathbf{q}m}(\mathbf{r}, t)$ may be collated into series according to the order of the 'diffuse' scattering process,

$$\varphi_d^{\mathbf{q}m}(\mathbf{r}, t) = \varphi_{d(1)}^{\mathbf{q}m}(\mathbf{r}, t) + \varphi_{d(2)}^{\mathbf{q}m}(\mathbf{r}, t) + \varphi_{d(3)}^{\mathbf{q}m}(\mathbf{r}, t) + \dots,$$
$$(18)$$

where $\varphi_{d(l)}^{\mathbf{q}m}(\mathbf{r}, t)$ is the contribution to $\varphi_d^{\mathbf{q}m}(\mathbf{r}, t)$ due to dynamical scattering from the 'displacement' potential, $d_s^{\mathbf{q}m}(\mathbf{r}, t)$, at l different slices (or depths), s, and dynamical scattering from the reference potential, $b_n(\mathbf{r})$, in the other $N - l$ slices (see the mathematical formulation that follows below). $\varphi_{d(1)}^{\mathbf{q}m}(\mathbf{r}, t)$, $\varphi_{d(2)}^{\mathbf{q}m}(\mathbf{r}, t)$ *etc.* will be referred to as the components of the wave function due to 'single-diffuse scattering', 'double-diffuse scattering' *etc.*, respectively. (It is emphasized that these components each describe dynamical scattering from multiple slices, with the labels 'single', 'double' *etc.* referring only to the number of these slices at which dynamical *diffuse* scattering takes place.) Explicitly,

$$\varphi_{d(1)}^{\mathbf{q}m}(\mathbf{r}, t) = \sum_{s=1}^{N} S(\Delta) D_s(\mathbf{r}, t), \qquad (19)$$

where

$$D_1(\mathbf{r}, t) = \sum_{\substack{\alpha_1,\dots,\alpha_N \\ \beta_1,\dots,\beta_N}} \frac{(i\ell\varepsilon)^{\alpha_1+\beta_1+\dots+\alpha_N+\beta_N}}{\alpha_1!\beta_1!\dots\alpha_N!\beta_N!} \Big\{ \Delta^{\alpha_N}[b_N(\mathbf{r})]^{\beta_N}\dots$$
$$\dots\Delta^{\alpha_2}[b_2(\mathbf{r})]^{\beta_2}\Delta^{\alpha_1}[d_1^{\mathbf{q}m}(\mathbf{r}, t)]^{\beta_1}\Big\}$$
$$\vdots$$

$$D_s(\mathbf{r}, t) = \sum_{\substack{\alpha_1,\dots,\alpha_N \\ \beta_1,\dots,\beta_N}} \frac{(i\ell\varepsilon)^{\alpha_1+\beta_1+\dots+\alpha_N+\beta_N}}{\alpha_1!\beta_1!\dots\alpha_N!\beta_N!}$$
$$\times \{\Delta^{\alpha_N}[b_N(\mathbf{r})]^{\beta_N}\dots\Delta^{\alpha_{s+1}}[b_{s+1}(\mathbf{r})]^{\beta_{s+1}}$$
$$\Delta^{\alpha_s}[d_s^{\mathbf{q}m}(\mathbf{r}, t)]^{\beta_s}\Delta^{\alpha_{s-1}}[b_{s-1}(\mathbf{r})]^{\beta_{s-1}}\dots$$
$$\dots\Delta^{\alpha_1}[b_1(\mathbf{r})]^{\beta_1}\}$$
$$\vdots$$

$$D_N(\mathbf{r}, t) = \sum_{\substack{\alpha_1,\dots,\alpha_N \\ \beta_1,\dots,\beta_N}} \frac{(i\ell\varepsilon)^{\alpha_1+\beta_1+\dots+\alpha_N+\beta_N}}{\alpha_1!\beta_1!\dots\alpha_N!\beta_N!}$$
$$\times \{\Delta^{\alpha_N}[d_N^{\mathbf{q}m}(\mathbf{r}, t)]^{\beta_N}\Delta^{\alpha_{N-1}}[b_{N-1}(\mathbf{r})]^{\beta_{N-1}}\dots$$
$$\dots\Delta^{\alpha_1}[b_1(\mathbf{r})]^{\beta_1}\} \qquad (20)$$

and

$$\varphi_{d(2)}^{\mathbf{q}m}(\mathbf{r}, t) = \sum_{s=1}^{N-1} \sum_{s'=s+1}^{N} S(\Delta) D_{ss'}(\mathbf{r}, t),$$

where, in analogy to $D_s(\mathbf{r}, t)$,

$$D_{ss'}(\mathbf{r}, t) = \sum_{\substack{\alpha_1,\dots,\alpha_N \\ \beta_1,\dots,\beta_N}} \frac{(i\ell\varepsilon)^{\alpha_1+\beta_1+\dots+\alpha_N+\beta_N}}{\alpha_1!\beta_1!\dots\alpha_N!\beta_N!}$$
$$\times \Big\{ \Delta^{\alpha_N}[b_N(\mathbf{r})]^{\beta_N}\dots\Delta^{\alpha_{s'+1}}[b_{s'+1}(\mathbf{r})]^{\beta_{s'+1}}$$
$$\Delta^{\alpha_{s'}}[d_{s'}^{\mathbf{q}m}(\mathbf{r}, t)]^{\beta_{s'}}\Delta^{\alpha_{s'-1}}[b_{s'-1}(\mathbf{r})]^{\beta_{s'-1}}\dots$$
$$\dots\Delta^{\alpha_{s+1}}[b_{s+1}(\mathbf{r})]^{\beta_{s+1}}\Delta^{\alpha_s}[d_s^{\mathbf{q}m}(\mathbf{r}, t)]^{\beta_s}$$
$$\Delta^{\alpha_{s-1}}[b_{s-1}(\mathbf{r})]^{\beta_{s-1}}\dots\Delta^{\alpha_1}[b_1(\mathbf{r})]^{\beta_1}\Big\}$$

so that, for example,

$$D_{1N}(\mathbf{r}, t) = \sum_{\substack{\alpha_1,\dots,\alpha_N \\ \beta_1,\dots,\beta_N}} \frac{(i\ell\varepsilon)^{\alpha_1+\beta_1+\dots\alpha_N+\beta_N}}{\alpha_1!\beta_1!\dots\alpha_N!\beta_N!}$$
$$\times \Big\{ \Delta^{\alpha_N}[d_N^{\mathbf{q}m}(\mathbf{r}, t)]^{\beta_N}\Delta^{\alpha_{N-1}}[b_{N-1}(\mathbf{r})]^{\beta_{N-1}}\dots$$
$$\dots\Delta^{\alpha_2}[b_2(\mathbf{r})]^{\beta_2}\Delta^{\alpha_1}[d_1^{\mathbf{q}m}(\mathbf{r}, t)]^{\beta_1}\Big\}.$$

The higher-order terms, $l > 2$, may be similarly constructed.

It is desirable to separate the t-dependent and t-independent components of the wave function and this paragraph describes the manipulations that achieve this. The t-dependent amplitude, $h_{js}^{\mathbf{q}m}(t)$, is first detached from the operator, $\hat{\mathbf{h}}^{\mathbf{q}m} \cdot \nabla$, in the term $[d_s^{\mathbf{q}m}(\mathbf{r}, t)]^{\beta_s}$ (the displacement potential of the slice, s, one atomic layer thick, raised to the power, β_s) as follows:

Recall that

$$[d_s^{\mathbf{q}m}(\mathbf{r}, t)]^{\beta_s} = \left\{ \sum_j \sum_a (1/a!)[h_{js}^{\mathbf{q}m}(t)\hat{\mathbf{h}}^{\mathbf{q}m} \cdot \nabla]^a U(\mathbf{r} - \bar{\mathbf{r}}_j) \right\}^{\beta_s}.$$
$$(21)$$

Since $U(\mathbf{r} - \bar{\mathbf{r}}_j)$ is a very narrow function localized about the point $\bar{\mathbf{r}}_j$,

$$[\hat{\mathbf{h}}^{\mathbf{q}m} \cdot \nabla U(\mathbf{r} - \bar{\mathbf{r}}_j)][\hat{\mathbf{h}}^{\mathbf{q}m} \cdot \nabla U(\mathbf{r} - \bar{\mathbf{r}}_{j'})] \cong 0$$

and, hence,

$$[d_s^{\mathbf{q}m}(\mathbf{r}, t)]^{\beta_s} \approx \sum_j \left\{ \sum_a (1/a!)[h_{js}^{\mathbf{q}m}(t)\hat{\mathbf{h}}^{\mathbf{q}m} \cdot \nabla]^a U(\mathbf{r} - \bar{\mathbf{r}}_j) \right\}^{\beta_s} \tag{22}$$

is an excellent approximation. The t dependence can then be separated explicitly by using the multinomial theorem for large n,

$$(x_1 + x_2 + \ldots + x_n)^{\beta} = \sum_{\gamma_1 + \gamma_2 + \ldots + \gamma_n = \beta} \frac{\beta!}{\gamma_1! \gamma_2! \ldots \gamma_n!} x_1^{\gamma_1} x_2^{\gamma_2} \ldots x_n^{\gamma_n},$$

to give

$$[d_s^{\mathbf{q}m}(\mathbf{r}, t)]^{\beta_s} \cong \sum_j \left\{ \sum_{a=1}^n (1/a!)[h_{js}^{\mathbf{q}m}(t)\hat{\mathbf{h}}^{\mathbf{q}m} \cdot \nabla]^a U(\mathbf{r} - \bar{\mathbf{r}}_j) \right\}^{\beta_s}$$
$$= \sum_j \left\{ \sum_{\gamma_1 + \ldots + \gamma_n = \beta_s} [h_{js}^{\mathbf{q}m}(t)]^{\gamma_1 + 2\gamma_2 + \ldots + n\gamma_n} \right.$$
$$\times \beta_s! / (1!)^{\gamma_1} \gamma_1! \ldots (n!)^{\gamma_n} \gamma_n!$$
$$\times [(\hat{\mathbf{h}}^{\mathbf{q}m} \cdot \nabla)^1 U(\mathbf{r} - \bar{\mathbf{r}}_j)]^{\gamma_1} \ldots$$
$$\left. \ldots [(\hat{\mathbf{h}}^{\mathbf{q}m} \cdot \nabla)^n U(\mathbf{r} - \bar{\mathbf{r}}_j)]^{\gamma_n} \right\}, \tag{23}$$

permitting the t dependence to be separated explicitly in the wavefunction

$$\varphi_{d(1)}^{\mathbf{q}m}(\mathbf{r}, t) = \sum_j \sum_{s=1}^N \sum_{\beta_s} \sum_{\gamma_1 + \ldots + \gamma_n = \beta_s} [h_{js}^{\mathbf{q}m}(t)]^{\gamma_1 + 2\gamma_2 + \ldots + n\gamma_n}$$
$$\times \{S(\Delta)W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})\}, \tag{24}$$

where

$$W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r}) = \frac{(i\ell\varepsilon)^{\beta_s}}{1!^{\gamma_1}\gamma_1! \ldots n!^{\gamma_n}\gamma_n!} \sum_{\substack{\alpha_1, \ldots, \alpha_N \\ \beta_1, \ldots, \beta_N \\ \text{except } \beta_s}} \frac{(i\ell\varepsilon)^{\alpha_1 + \beta_1 + \ldots + \alpha_N + \beta_N}}{\alpha_1! \beta_1! \ldots \alpha_N! \beta_N!}$$
$$\times \left(\Delta^{\alpha_N}[b_N(\mathbf{r})]^{\beta_N} \ldots \Delta^{\alpha_{s+1}}[b_{s+1}(\mathbf{r})]^{\beta_{s+1}} \Delta^{\alpha_s} \right.$$
$$\left\{ [(\hat{\mathbf{h}}^{\mathbf{q}m} \cdot \nabla)^1 U(\mathbf{r} - \bar{\mathbf{r}}_j)]^{\gamma_1} \ldots \right.$$
$$\left. \ldots [(\hat{\mathbf{h}}^{\mathbf{q}m} \cdot \nabla)^n U(\mathbf{r} - \bar{\mathbf{r}}_j)]^{\gamma_n} \right\}$$
$$\left. \Delta^{\alpha_{s-1}}[b_{s-1}(\mathbf{r})]^{\beta_{s-1}} \ldots \Delta^{\alpha_1}[b_1(\mathbf{r})]^{\beta_1} \right)$$

and the γ_i, labelling $W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})$, is shorthand for the set of values $\{\gamma_1, \gamma_2, \ldots, \gamma_n\}$.

Similarly,

$$\varphi_{d(2)}^{\mathbf{q}m}(\mathbf{r}, t) = \sum_{j,j'} \sum_{s=1}^{N-1} \sum_{s'=s+1}^N \sum_{\beta_s, \beta_{s'}} \sum_{\substack{\gamma_1 + \ldots + \gamma_n = \beta_s \\ \gamma_1' + \ldots + \gamma_n' = \beta_{s'}}} [h_{js}^{\mathbf{q}m}(t)]^{\gamma_1 + 2\gamma_2 + \ldots + n\gamma_n}$$
$$\times [h_{j's'}^{\mathbf{q}m}(t)]^{\gamma_1' + 2\gamma_2' + \ldots + n\gamma_n'} \{S(\Delta)W_{js\beta_s\gamma_i,j's'\beta_{s'}\gamma_i'}^{\mathbf{q}m}(\mathbf{r})\}, \tag{25}$$

where

$$W_{js\beta_s\gamma_i,j's'\beta_{s'}\gamma_i'}^{\mathbf{q}m}(\mathbf{r}) = \frac{(i\ell\varepsilon)^{\beta_s + \beta_{s'}}}{(1!^{\gamma_1}\gamma_1! \ldots n!^{\gamma_n}\gamma_n!)(1!^{\gamma_1'}\gamma_1'! \ldots n!^{\gamma_n'}\gamma_n'!)}$$
$$\times \sum_{\substack{\alpha_1, \ldots, \alpha_N \\ \beta_1, \ldots, \beta_N \\ \text{except } \beta_s, \beta_{s'}}} \frac{(i\ell\varepsilon)^{\alpha_1 + \beta_1 + \ldots + \alpha_N + \beta_N}}{\alpha_1! \beta_1! \ldots \alpha_N! \beta_N!}$$
$$\times \left(\Delta^{\alpha_N}[b_N(\mathbf{r})]^{\beta_N} \ldots \Delta^{\alpha_{s'+1}}[b_{s'+1}(\mathbf{r})]^{\beta_{s'+1}} \right.$$
$$\Delta^{\alpha_{s'}} \left\{ [(\hat{\mathbf{h}}^{\mathbf{q}m} \cdot \nabla)^1 U(\mathbf{r} - \bar{\mathbf{r}}_{j'})]^{\gamma_i'} \ldots \right.$$
$$\left. \ldots [(\hat{\mathbf{h}}^{\mathbf{q}m} \cdot \nabla)^n U(\mathbf{r} - \bar{\mathbf{r}}_{j'})]^{\gamma_n'} \right\} \Delta^{\alpha_{s'-1}}$$
$$[b_{s'-1}(\mathbf{r})]^{\beta_{s'-1}} \ldots \Delta^{\alpha_{s+1}}[b_{s+1}(\mathbf{r})]^{\beta_{s+1}}$$
$$\Delta^{\alpha_s} \left\{ [(\hat{\mathbf{h}}^{\mathbf{q}m} \cdot \nabla)^1 U(\mathbf{r} - \bar{\mathbf{r}}_j)]^{\gamma_1} \ldots \right.$$
$$\left. \ldots [(\hat{\mathbf{h}}^{\mathbf{q}m} \cdot \nabla)^n U(\mathbf{r} - \bar{\mathbf{r}}_j)]^{\gamma_n} \right\}$$
$$\left. \Delta^{\alpha_{s-1}}[b_{s-1}(\mathbf{r})]^{\beta_{s-1}} \ldots \Delta^{\alpha_1}[b_1(\mathbf{r})]^{\beta_1} \right).$$

In the above, $W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})$ is the 'shape' of the 'wavelet' contributed by the γ_ith component of β_s-order scattering from displaced atom j in slice s (including scattering, to all orders, from all other non-displaced atoms in the crystal). $[h_{js}^{\mathbf{q}m}(t)]^{\gamma_1 + 2\gamma_2 + \ldots + n\gamma_n}$ is the 'amplitude' of this 'wavelet'. Note that $W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})$ is necessarily localized in the vicinity of $\bar{\mathbf{r}}_j$ because of the narrowness of $U(\mathbf{r} - \bar{\mathbf{r}}_j)$.

Similarly, $W_{js\beta_s\gamma_i,j's'\beta_{s'}\gamma_i'}^{\mathbf{q}m}(\mathbf{r})$ is the shape of the wavelet contributed by the γ_ith component of β_s-order scattering from displaced atom j in slice s and then the γ_ith component of $\beta_{s'}$-order scattering from displaced atom j' in slice s' (including scattering, to all orders, from all other nondisplaced atoms in the crystal).

$$[h_{js}^{\mathbf{q}m}(t)]^{\gamma_1 + 2\gamma_2 + \ldots + n\gamma_n}[h_{j's'}^{\mathbf{q}m}(t)]^{\gamma_1' + 2\gamma_2' + \ldots + n\gamma_n'}$$

is the amplitude of this wavelet. $W_{js\beta_s\gamma_i,j's'\beta_{s'}\gamma_i'}^{\mathbf{q}m}(\mathbf{r})$ is necessarily localized in the vicinity of $\bar{\mathbf{r}}_j$ and $\bar{\mathbf{r}}_{j'}$ because of the narrowness of $U(\mathbf{r} - \bar{\mathbf{r}}_j)$ and $U(\mathbf{r} - \bar{\mathbf{r}}_{j'})$, and will tend to zero with the distance between j and j'.

The wavelet, $W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})$, due to the γ_ith component of β_s-order scattering at site js has the same shape as that due to scattering at site Js, $W_{Js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})$. This is because they are only differential functions of $U(\mathbf{r})$ and are therefore cell, j, and configuration, t, independent. [This remains true after operation by the lens, $S(\Delta)$.] However, the contribution that each of these wavelets makes to the wave function, $\varphi_d^{\mathbf{q}m}(\mathbf{r}, t)$, is weighted by the cell-*dependent* displacement amplitudes, $[h_{js}^{\mathbf{q}m}(t)]^{\gamma_1 + 2\gamma_2 + \ldots + n\gamma_n}$ and $[h_{Js}^{\mathbf{q}m}(t)]^{\gamma_1 + 2\gamma_2 + \ldots + n\gamma_n}$, respectively,

so that $\varphi_{d(1)}^{\mathbf{q}m}(\mathbf{r}, t)$, and hence $\varphi_d^{\mathbf{q}m}(\mathbf{r}, t)$, will not have the translational symmetry of the reference lattice. In other words, for a given order of scattering, the shape of the wavelet generated by scattering from a displaced atom at site js (and hence localized about the position $\bar{\mathbf{r}}_j$) is the same as that generated by scattering from a displaced atom at site Js (and hence localized about the position $\bar{\mathbf{r}}_J$). However, the amplitude of these wavelets depends upon the size of the atomic displacement $h_{js}^{\mathbf{q}m}(t)$ and $h_{Js}^{\mathbf{q}m}(t)$, respectively, at the site at which the diffuse scattering occurs.

Similarly, for double-diffuse scattering, the wavelet, $W_{js\beta_s\gamma_i, j's'\beta_{s'}\gamma'_i}^{\mathbf{q}m}(\mathbf{r})$, has the same shape as $W_{Js\beta_s\gamma_i, J's'\beta_{s'}\gamma'_i}^{\mathbf{q}m}(\mathbf{r})$. However, the contribution that each of these wavelets makes to the wave function, $\varphi_d^{\mathbf{q}m}(\mathbf{r}, t)$, is weighted by the cell-*dependent* displacement amplitudes,

$$[h_{js}^{\mathbf{q}m}(t)]^{\gamma_1+2\gamma_2+...+n\gamma_n}[h_{j's'}^{\mathbf{q}m}(t)]^{\gamma'_1+2\gamma'_2+...+n\gamma'_n}$$

and

$$[h_{Js}^{\mathbf{q}m}(t)]^{\gamma_1+2\gamma_2+...+n\gamma_n}[h_{J's'}^{\mathbf{q}m}(t)]^{\gamma'_1+2\gamma'_2+...+n\gamma'_n},$$

respectively. So, the wavefunction, $\varphi_{d(2)}^{\mathbf{q}m}(\mathbf{r}, t)$, and hence $\varphi_d^{\mathbf{q}m}(\mathbf{r}, t)$, will not have the translational symmetry of the reference lattice.

To determine what the translational symmetry of the final image is, consider the intensity distribution at the image plane, due to scattering from the configuration labelled by t, $V^{\mathbf{q}m}(\mathbf{R}, t)$,

$$\begin{aligned}
I^{\mathbf{q}m}(\mathbf{r}, t) &= |\varphi^{\mathbf{q}m}(\mathbf{r}, t)|^2 \\
&= |\varphi_b(\mathbf{r}) + \varphi_d^{\mathbf{q}m}(\mathbf{r}, t)|^2 \\
&= |\varphi_b(\mathbf{r})|^2 + \{|\varphi_d^{\mathbf{q}m}(\mathbf{r}, t)|^2 + 2[\Re\varphi_b(\mathbf{r})\Re\varphi_d^{\mathbf{q}m}(\mathbf{r}, t) \\
&\quad + \Im\varphi_b(\mathbf{r})\Im\varphi_d^{\mathbf{q}m}(\mathbf{r}, t)]\} \\
&= I_b(\mathbf{r}) + I_d^{\mathbf{q}m}(\mathbf{r}, t).
\end{aligned} \qquad (26)$$

$I_b(\mathbf{r}) = |\varphi_b(\mathbf{r})|^2$ is the intensity distribution due to scattering from the reference lattice and has the translation vector of the reference lattice in this projection. $I_d^{\mathbf{q}m}(\mathbf{r}, t)$ is the intensity distribution due to diffuse scattering from the configuration of atomic displacements labelled by t. In order to consider the translational symmetry of $I_d^{\mathbf{q}m}(\mathbf{r}, t)$, and hence $I^{\mathbf{q}m}(\mathbf{r}, t)$, assume, in the first instance, that the electron scatters diffusely only once in its passage through the crystal, that is, $\varphi_d^{\mathbf{q}m}(\mathbf{r}, t) \cong \varphi_{d(1)}^{\mathbf{q}m}(\mathbf{r}, t)$ so that $I_d^{\mathbf{q}m}(\mathbf{r}, t) \cong I_{d(1)}^{\mathbf{q}m}(\mathbf{r}, t)$. [This is a good approximation for crystals less than a few hundred ångströms thick (Hall & Hirsch, 1965; Doyle, 1971).] Then, substituting (24) into (26) gives an explicit expression for $I_d^{\mathbf{q}m}(\mathbf{r}, t)$:

$$\begin{aligned}
I_d^{\mathbf{q}m}(\mathbf{r}, t) &\approx I_{d(1)}^{\mathbf{q}m}(\mathbf{r}, t) \\
&= |\varphi_{d(1)}^{\mathbf{q}m}(\mathbf{r}, t)|^2 + 2[\Re\varphi_b(\mathbf{r})\Re\varphi_{d(1)}^{\mathbf{q}m}(\mathbf{r}, t) \\
&\quad + \Im\varphi_b(\mathbf{r})\Im\varphi_{d(1)}^{\mathbf{q}m}(\mathbf{r}, t)] \\
&= \sum_j \sum_{s=1}^N \sum_{\beta_s} \sum_{\gamma_1+...+\gamma_n=\beta_s} [h_{js}^{\mathbf{q}m}(t)]^{2\gamma}\{|S(\Delta)W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})|^2\} \\
&\quad + \sum_{\substack{j,J \\ j \neq J \text{ when } s=S, \\ \beta_s=\beta_S \text{ and } \gamma=\Gamma}} \sum_{s,S=1}^N \sum_{\beta_s,\beta_S} \sum_{\substack{\gamma_1+...+\gamma_n=\beta_s \\ \Gamma_1+...+\Gamma_n=\beta_S}} [h_{js}^{\mathbf{q}m}(t)]^\gamma [h_{JS}^{\mathbf{q}m}(t)]^\Gamma \\
&\quad \times \{[S(\Delta)W_{jS\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})][S(\Delta)W_{JS\beta_S\Gamma_i}^{\mathbf{q}m}(\mathbf{r})]^*\} \\
&\quad + 2\sum_j \sum_{s=1}^N \sum_{\beta_s} \sum_{\gamma_1+...+\gamma_n=\beta_s} [h_{js}^{\mathbf{q}m}(t)]^\gamma \\
&\quad \times \{\Re\varphi_b(\mathbf{r})\Re[S(\Delta)W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})] \\
&\quad + \Im\varphi_b(\mathbf{r})\Im[S(\Delta)W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})]\},
\end{aligned} \qquad (27)$$

where γ and Γ are shorthand for $\gamma_1 + 2\gamma_2 + \ldots + n\gamma_n$ and $\Gamma_1 + 2\Gamma_2 + \ldots + n\Gamma_n$, respectively. The expressions in the curly brackets of (27) are cell- and t-independent waveforms which are functions of $U(\mathbf{r})$, localized at the given reference lattice site, labelled by j, in the plane projected perpendicular to the electron beam $\bar{\mathbf{r}}_j$. These functions are modulated according to $\mathbf{q}$ by cell-dependent amplitudes,

$$[h_{js}^{\mathbf{q}m}(t)]^\gamma = [h^{\mathbf{q}m}(\bar{\mathbf{r}}_j \cdot \mathbf{q}_{xy} + \bar{z}_s q_z + \omega_{\mathbf{q}m}t)]^\gamma,$$

so that the contribution to the intensity distribution, $I_{d(1)}^{\mathbf{q}m}(\mathbf{r}, t)$ at $\mathbf{r}$ from the cell-independent functions localized at $\bar{\mathbf{r}}_j$ is given different weight to the contribution from the same functions localized at $\bar{\mathbf{r}}_{j'}$. The *difference* between these weights for diffuse scattering at a given depth, s, depends only on $\mathbf{q}_{xy}$, the component of the wavevector in the plane perpendicular to the beam, and does not depend on q_z, the component parallel to the beam. (The weights and hence the intensity distribution do of course depend on q_z, however, their dependence is independent of atomic column, j, so that q_z influences the intensity distribution with the periodicity of the reference lattice.)

The difference between weights ensures that the intensity distribution in the image does not have the translational symmetry of the reference lattice but will be modulated relative to the reference lattice with a 'wavevector' that depends in some way upon $\mathbf{q}_{xy}$. In order to consider the translational symmetry of the intensity distribution in the image, consider the standard case of a cosine form for the displacement vector function,

$$h_{js}^{\mathbf{q}m}(t) \propto \cos(\bar{\mathbf{r}}_j \cdot \mathbf{q}_{xy} + \bar{z}_s q_z + \omega_{\mathbf{q}m}t) = \cos(\theta_{js} + \omega_{\mathbf{q}m}t),$$

then

$$[h_{js}^{\mathbf{q}m}(t)]^{2\gamma}$$

$$= 2^{1-2\gamma}\left[\frac{1}{2}\binom{2\gamma}{\gamma} + \sum_{k=0}^{\gamma-1}\binom{2\gamma}{k}\right.$$

$$\left. \times \cos 2(\gamma - k)(\theta_{js} + \omega_{\mathbf{q}m}t)\right]$$

$$[h_{js}^{\mathbf{q}m}(t)]^{\gamma}[h_{JS}^{qm}(t)]^{\Gamma}$$

$$= 2^{1-\gamma-\Gamma}\left[\frac{1}{2}\binom{\gamma}{\frac{1}{2}\gamma}\binom{\Gamma}{\frac{1}{2}\Gamma} + \binom{\Gamma}{\frac{1}{2}\Gamma}\sum_{k=0}^{\frac{1}{2}\gamma-1}\binom{\gamma}{k}\right.$$

$$\times \cos(\gamma - 2k)(\theta_{js} + \omega_{\mathbf{q}m}t)$$

$$+ \binom{\gamma}{\frac{1}{2}\gamma}\sum_{k=0}^{\frac{1}{2}\Gamma-1}\binom{\Gamma}{k}\cos(\Gamma - 2k)(\theta_{JS} + \omega_{\mathbf{q}m}t)$$

$$+ \sum_{k=0}^{\frac{1}{2}\gamma-1}\sum_{k'=0}^{\frac{1}{2}\Gamma-1}\binom{\gamma}{k}\binom{\Gamma}{k'}\{\cos[(\gamma - 2k)\theta_{js}$$

$$+ (\Gamma - 2k')\theta_{JS} + (\gamma - 2k + \Gamma - 2k')\omega_{\mathbf{q}m}t]$$

$$+ \cos[(\gamma - 2k)\theta_{js} - (\Gamma - 2k')\theta_{JS}$$

$$\left. + (\gamma - 2k - \Gamma + 2k')\omega_{\mathbf{q}m}t]\}\right]$$

$$[h_{js}^{\mathbf{q}m}(t)]^{\gamma}$$

$$= 2^{1-\gamma}\left[\frac{1}{2}\binom{\gamma}{\frac{1}{2}\gamma} + \sum_{k=0}^{\frac{1}{2}\gamma-1}\binom{\gamma}{k}\right.$$

$$\left. \times \cos(\gamma - 2k)(\theta_{js} + \omega_{\mathbf{q}m}t)\right]. \tag{28}$$

[As an aside, note here that the standard 'damping function', describing the damping of correlations between atomic displacements with distance, has deliberately been left out of the discussion since it does not alter the argument but does add to the visual complexity of the equations. (Only the *form* of the displacement amplitude is considered, hence the proportional sign.) The damping function perpendicular to the beam is insignificant to the argument because, as will be described later, the intensity distribution in the HREM image only depends upon the correlations between atoms in the same or adjacent columns. The range over which the displacements are correlated perpendicular to the beam is therefore not important for the issues in this paper. The damping function parallel to the beam will have an effect on the quantitative intensity distribution of the image but does not affect the translational symmetry discussed in this paper.]

The translational symmetry of the image depends upon the nature of the dependence of $I_{d(1)}^{\mathbf{q}m}(\mathbf{r}, t)$ on $\mathbf{q}_{xy}$. From (28), it can be seen that different sets of terms in (27) contribute an intensity distribution with a different

dependence on $\mathbf{q}_{xy}$. The second term (the interference between the single-diffuse scattered wavelets) and the large third term (the interference term between the wave function due to 'Bragg' scattering and the wave function due to single-diffuse scattering) both consist of terms that contribute an intensity distribution that is modulated according to a 'wavevector' that is some integer multiple of $\mathbf{q}_{xy}$ (28). Whilst the large first term [the incoherent (*i.e.* interference-free) sum of the single-diffuse scattered wavelets] comprises terms that each contribute an intensity distribution that is modulated with a 'wavevector' that is some integer multiple of $2\mathbf{q}_{xy}$. Since the smallest effective 'wavevector' contributed by any set of terms in (27) is $\mathbf{q}_{xy}$, the 'wavevector' of the intensity distribution of the image as a whole, $I^{\mathbf{q}m}(\mathbf{r}, t)$, is $\mathbf{q}_{xy}$. In other words, the translational symmetry of the image is such that the only translations that leave the image invariant are those lattice translations, $\mathbf{p}$, of the reference structure satisfying $\mathbf{p} \cdot \mathbf{q}_{xy} = 2\pi m$ with m an integer. Thus, the two-dimensional intensity distribution $I^{\mathbf{q}m}(\mathbf{r}, t)$ has the same translational symmetry as the modulated structure projected onto the plane perpendicular to the beam, as determined by $\mathbf{q}_{xy}$. That is, the intensity distribution at the image plane due to scattering from the configuration of atoms $V^{\mathbf{q}m}(\mathbf{r}, z, t)$ has the translational symmetry of $V^{\mathbf{q}m}(\mathbf{r}, t)$. The argument follows similarly for double and higher-order diffuse scattering terms, all of which contribute an intensity distribution that is modulated with a 'wavevector' that is some integer multiple of $\mathbf{q}_{xy}$.

If a crystal has static correlated atomic displacements from its periodic lattice sites, then each incident electron will experience the same configuration, $V^{\mathbf{q}m}(\mathbf{r}, z, t_0)$, and will give rise to the same intensity distribution, $I^{\mathbf{q}m}(\mathbf{r}, t_0)$, for all of the exposure time. This gives the intuitive result that *the high-resolution electron-microscope image of a crystal with static correlated atomic displacements from its reference lattice sites will have the translational symmetry of the modulated structure in the plane perpendicular to the incident beam, as determined by $\mathbf{q}_{xy}$, the component of the modulation wavevector in that plane.*

It is important to note that this result derives entirely from upper-layer interactions, that is, dynamical scattering into or out of higher-order Laue-zone (HOLZ) reflections (regardless of whether the HOLZ reflections are included explicitly within the objective aperture or not). Put another way, this result is a consequence of the fact that the differential operators do not commute, so that $W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r}) \neq W_{js'\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})$ and the wave function arising from scattering from a displaced atom depends on the depth of that atom in the atomic column parallel to the beam. If this were not the case, that is, if the projection approximation were valid $[W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r}) = W_{js'\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})]$, then the phase-dependent weights, $h_{js}^{\mathbf{q}m}$, in (27) could be summed over depth, s, independently of the wavelets, $W_{js\beta_s\gamma}^{\mathbf{q}m}(\mathbf{r})$, with the result that all terms in (27) but the

second term would lose their phase dependence. In addition, the second term, the term describing the interference between waves scattered diffusely from different sites in the crystal, would depend only upon the *relative* phase between the diffusely scattering sites, so that the image as a whole would have the translation vector of the reference lattice. It is therefore only the upper layer interactions that convey information on the translational symmetry of the modulated structure to the intensity distribution.

Care must therefore be taken if making approximations in the calculation of the image of a crystal with a modulated structure, as many approximations lead to the calculation of images with incorrect translational symmetry. An obvious example is the random-phase approximation, which ignores the correlations between displaced atoms and so would generate a calculated image with the translational symmetry of the reference lattice. Another example is that just described, the projection approximation, which would also generate an image with the translational symmetry of the reference lattice. No diffuse scattering or superlattice reflections would therefore be present in the Fourier transform of images calculated using these approximations. The diffuse scattering or superlattice reflections that register in the Fourier transform of the experimental image are thus, in a sense, a measure of the invalidity of these and other approximations (see §4.3).

4.2. Time-dependent displacements – phonons

Now consider an electron impinging on a crystal with time-dependent correlated atomic displacements. The interaction time of the electron with the lattice is negligible compared with the period of oscillation, τ_i, of the atoms. Thus each electron effectively interacts with a static lattice with an instantaneous configuration, $V^{\mathbf{q}m}(\mathbf{R}, t)$, labelled by t. In other words, since the phonon energy ($< 10^{-1}$ eV) is negligible compared with the energy of the incident electron (10^2 keV), energy exchange between the electron and the lattice can be neglected. [Wang (1992) has shown explicitly that this is a valid approximation up to temperatures just above room temperature.] In this approximation then, the fnal intensity distribution recorded on the photographic plate due to scattering from phonon mode $\mathbf{q}m$, after exposure time $T \gg \tau_i$, can be treated as the sum of the individual intensity distributions due to elastic scattering from each of these instantaneous configurations. That is,

$$I^{\mathbf{q}m}(\mathbf{r}) = \langle I^{\mathbf{q}m}(\mathbf{r}, t) \rangle$$
$$= I_b(\mathbf{r}) + \langle I_d^{\mathbf{q}m}(\mathbf{r}, t) \rangle$$
$$= I_b(\mathbf{r}) + \lim_{T \to \infty} (1/T) \int_0^T I^{\mathbf{q}m}(\mathbf{r}, t)\, \mathrm{d}t. \quad (29)$$

Taking the time average of $I_{d(1)}^{\mathbf{q}m}(\mathbf{r}, t)$ in (27) gives

$$I_{d(1)}^{\mathbf{q}m}(\mathbf{r}, t) = \sum_j \sum_{s=1}^N \sum_{\beta_s} \sum_{\gamma_1 + \ldots + \gamma_n = \beta_s} \langle [h_{js}^{\mathbf{q}m}(t)]^{2\gamma} \rangle$$
$$\times \{ |S(\Delta) W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})|^2 \}$$
$$+ \underbrace{\sum_{j,J}}_{\substack{j \neq J \text{ when } s=S, \\ \beta_s = \beta_S \text{ and } \gamma = \Gamma}} \sum_{s,S=1}^N \sum_{\beta_s,\beta_S} \sum_{\substack{\gamma_1 + \ldots + \gamma_n = \beta_s \\ \Gamma_1 + \ldots + \Gamma_n = \beta_S}} \langle [h_{js}^{\mathbf{q}m}(t)]^{\gamma} [h_{JS}^{\mathbf{q}m}(t)]^{\Gamma} \rangle$$
$$\times \{ [S(\Delta) W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})][S(\Delta) W_{JS\beta_S\Gamma_i}^{\mathbf{q}m}(\mathbf{r})]^* \}$$
$$+ 2 \sum_j \sum_{s=1}^N \sum_{\beta_s} \sum_{\gamma_1 + \ldots + \gamma_n = \beta_s} \langle [h_{js}^{\mathbf{q}m}(t)]^{\gamma} \rangle$$
$$\times \{ \Re\varphi_b(\mathbf{r}) \Re[S(\Delta) W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})]$$
$$+ \Im\varphi_b(\mathbf{r}) \Im[S(\Delta) W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})] \}, \quad (30)$$

where the time averages for the standard case, $h_{js}^{\mathbf{q}m}(t) \propto \cos(\theta_{js} + \omega_{\mathbf{q}m}t)$, are given by

$$\langle [h_{js}^{\mathbf{q}m}(t)]^{2\gamma} \rangle = 2^{-2\gamma} \binom{2\gamma}{\gamma}$$

$$\langle [h_{js}^{\mathbf{q}m}(t)]^{\gamma} [h_{JS}^{\mathbf{q}m}(t)]^{\Gamma} \rangle = 2^{2-\gamma-\Gamma} \left[\frac{1}{4} \binom{\gamma}{\frac{1}{2}\gamma} \binom{\Gamma}{\frac{1}{2}\Gamma} \right.$$
$$+ \sum_{k=0}^{\frac{1}{2}\gamma-1} \binom{\gamma}{k} \binom{\Gamma}{\frac{\gamma+\Gamma}{2} - k}$$
$$\left. \times \cos(\gamma - 2k)(\theta_{js} - \theta_{JS}) \right]$$

$$\langle [h_{js}^{\mathbf{q}m}(t)]^{\gamma} \rangle = 2^{-\gamma} \binom{\gamma}{\frac{1}{2}\gamma},$$

and where $\theta_{js} - \theta_{JS} = (\overline{\mathbf{R}}_{js} - \overline{\mathbf{R}}_{JS}) \cdot \mathbf{q}$ is the relative phase of the atoms in cells js and JS.

It is immediately evident that the first and third terms in (30) have the periodicity of the reference lattice when it is noted that the time averages are constants that depend only on the integers $\gamma = \gamma_1 + 2\gamma_2 + \ldots + n\gamma_n$ and the order of the expansion of the phase-object transmission function in the diffusely scattering slice, β_s (since $\gamma_1 + \gamma_2 + \ldots + \gamma_n = \beta_s$). They are completely independent of j and s and hence the unit cell since the cell-dependent phase of the displacement amplitude vanishes in the time averaging. Classically, this is a statement that the starting position of each atom becomes irrelevant in the final intensity distribution of the image, which is taken over an exposure time many times the period of oscillation of the atoms. Thus the functions $|S(\Delta) W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})|^2$ and

$$\{ \Re\varphi_b(\mathbf{r}) \Re[S(\Delta) W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})] + \Im\varphi_b(\mathbf{r}) \Im[S(\Delta) W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})] \}$$

contribute to the intensity distribution with weights

$$2^{-2\gamma} \binom{2\gamma}{\gamma} \quad \text{and} \quad 2^{-\gamma} \binom{\gamma}{\frac{1}{2}\gamma},$$

respectively, independent of the unit cell, j, at which they are centred. The first and last terms in (30) therefore have the translational symmetry of the reference lattice in the plane perpendicular to the beam.

The second term of (30) represents the interference between the diffusely scattered wavelets generated from different atomic sites in the crystal. It can be seen that, in general, the time average, $\langle [h_{js}^{\mathbf{q}m}(t)]^\gamma [h_{JS}^{\mathbf{q}m}(t)]^\Gamma \rangle$, yields a constant that depends not only on β_s and β_S but also on the relative phase of the atomic displacements at the diffusely scattering sites, $\theta_{js} - \theta_{JS} = (\bar{\mathbf{R}}_{js} - \bar{\mathbf{R}}_{JS}) \cdot \mathbf{q}$.

Since the choice of origin is arbitrary, $\theta_{js} - \theta_{JS}$ and hence $\langle [h_{js}^{\mathbf{q}m}(t)]^\gamma [h_{JS}^{\mathbf{q}m}(t)]^\Gamma \rangle$ do not depend on the absolute positions of the columns j and J. Explicitly, if l is an integer,

$$
\begin{aligned}
\theta_{js} - \theta_{JS} &= (\bar{\mathbf{r}}_j - \bar{\mathbf{r}}_J) \cdot \mathbf{q}_{xy} + (z_s - z_S)q_z \\
&= [(\bar{\mathbf{r}}_j + \bar{\mathbf{r}}_0) - (\bar{\mathbf{r}}_J + \bar{\mathbf{r}}_0)] \cdot \mathbf{q}_{xy} + (z_s - z_S)q_z \\
&= (\bar{\mathbf{r}}_{j+l} - \bar{\mathbf{r}}_{J+l}) \cdot \mathbf{q}_{xy} + (z_s - z_S)q_z \\
&= \theta_{j+l,s} - \theta_{J+l,S}
\end{aligned}
\tag{31}
$$

so that

$$
\langle [h_{js}^{\mathbf{q}m}(t)]^\gamma [h_{JS}^{\mathbf{q}m}(t)]^\Gamma \rangle = \langle [h_{j+l,s}^{\mathbf{q}m}(t)]^\gamma [h_{J+l,S}^{\mathbf{q}m}(t)]^\Gamma \rangle
$$

Thus, the second term in (30) has the periodicity of the reference lattice in that projection since the term, in curly brackets, due to interference between the diffusely scattered waves generated at site js and at site JS is multiplied by the same constant, $\langle [h_{js}^{\mathbf{q}m}(t)]^\gamma [h_{JS}^{\mathbf{q}m}(t)]^\Gamma \rangle$, as the term due to interference between waves generated at sites $j + l, s$ and $J + l, S$, $\langle [h_{j+l,s}^{\mathbf{q}m}(t)]^\gamma [h_{J+l,S}^{\mathbf{q}m}(t)]^\Gamma \rangle$. Thus the lattice image of a crystal with time-dependent phase-correlated atomic displacements from the reference lattice due to mode $\mathbf{q}m$ has the translation vector of the reference lattice in the plane perpendicular to the incident beam under the approximation of single-diffuse scattering.

The argument may be continued for double-diffuse scattering. The contribution to the image by double-diffuse scattering, $\langle I_{d(2)}^{\mathbf{q}m}(\mathbf{r}, t) \rangle$, is given by

$$
\begin{aligned}
\langle I_{d(2)}^{\mathbf{q}m}(\mathbf{r}, t) \rangle = \sum_{\substack{j,j'}} \sum_{s=1}^{N-1} \sum_{s'=s+1}^{N} \sum_{\beta_s, \beta_{s'}} \sum_{\substack{\gamma_1+\ldots+\gamma_n=\beta_s \\ \gamma_1'+\ldots+\gamma_n'=\beta_{s'}}} \langle [h_{js}^{\mathbf{q}m}(t)^\gamma h_{j's'}^{\mathbf{q}m}(t)^{\gamma'}]^2 \rangle \\
\times |S(\Delta)W_{js\beta_s\gamma, j'\beta_{s'}\gamma'}^{\mathbf{q}m}(\mathbf{r})|^2 \\
+ \sum_{\substack{j,j' \\ J,J'}} \sum_{s=1}^{N-1} \sum_{s'=s+1}^{N} \sum_{S=1}^{N-1} \sum_{S'=S+1}^{N} \sum_{\substack{\beta_s, \beta_{s'} \\ \beta_S, \beta_{S'}}} \sum_{\substack{\gamma_1+\ldots+\gamma_n=\beta_s \\ \gamma_1'+\ldots+\gamma_n'=\beta_{s'} \\ \Gamma_1+\ldots+\Gamma_n=\beta_S \\ \Gamma_1'+\ldots+\Gamma_n'=\beta_{S'}}} \\
\times \langle h_{js}^{\mathbf{q}m}(t)^\gamma h_{j's'}^{\mathbf{q}m}(t)^{\gamma'} h_{JS}^{\mathbf{q}m}(t)^\Gamma h_{J'S'}^{\mathbf{q}m}(t)^{\Gamma'} \rangle \\
\times [S(\Delta)W_{js\beta_s\gamma, j'\beta_{s'}\gamma'}^{\mathbf{q}m}(\mathbf{r})]
\end{aligned}
$$

$$
\begin{aligned}
\times [S(\Delta)W_{JS\beta_S\Gamma, J'S'\beta_{S'}\Gamma'}^{\mathbf{q}m}(\mathbf{r})]^* \\
+ \sum_{\substack{j,j' \\ J}} \sum_{s=1}^{N-1} \sum_{s'=s+1}^{N} \sum_{S=1}^{N-1} \sum_{S'=S+1}^{N} \sum_{\substack{\beta_s, \beta_{s'} \\ \beta_S}} \sum_{\substack{\gamma_1+\ldots+\gamma_n=\beta_s \\ \gamma_1'+\ldots+\gamma_n'=\beta_{s'} \\ \Gamma_1+\ldots+\Gamma_n=\beta_S}} \\
\times \langle h_{js}^{\mathbf{q}m}(t)^\gamma h_{j's'}^{\mathbf{q}m}(t)^{\gamma'} h_{JS}^{\mathbf{q}m}(t)^\Gamma \rangle \\
\times 2\{\Re[S(\Delta)W_{js\beta_s\gamma, j's'\beta_{s'}\gamma'}^{\mathbf{q}m}(\mathbf{r})] \\
\times \Re[S(\Delta)W_{JS\beta_S\Gamma}^{\mathbf{q}m}(\mathbf{r})] \\
+ \Im[S(\Delta)W_{js\beta_s\gamma, j's'\beta_{s'}\gamma'}^{\mathbf{q}m}(\mathbf{r})]\Im[S(\Delta)W_{JS\beta_S\Gamma}^{\mathbf{q}m}(\mathbf{r})]\} \\
+ \sum_{\substack{j,j'}} \sum_{s=1}^{N-1} \sum_{s'=s+1}^{N} \sum_{\beta_s, \beta_{s'}} \sum_{\substack{\gamma_1+\ldots+\gamma_n=\beta_s \\ \gamma_1'+\ldots+\gamma_n'=\beta_{s'}}} \\
\times \langle (h_{js}^{\mathbf{q}m}(t)^\gamma h_{j's'}^{\mathbf{q}m}(t)^{\gamma'}) \rangle \\
\times 2\{\Re\varphi_b(\mathbf{r})\Re[S(\Delta)W_{js\beta_s\gamma, j's'\beta_{s'}\gamma'}^{\mathbf{q}m}(\mathbf{r})] \\
+ \Im\varphi_b(\mathbf{r})\Im[S(\Delta)W_{js\beta_s\gamma, j's'\beta_{s'}\gamma'}^{\mathbf{q}m}(\mathbf{r})]\},
\end{aligned}
\tag{32}
$$

where the time averages, for the standard case, $h_{js}^{\mathbf{q}m}(t) \propto \cos(\theta_{js} + \omega_{\mathbf{q}m}t)$, are given by

$$
\begin{aligned}
& \langle [h_{js}^{\mathbf{q}m}(t)]^{2\gamma} [h_{j's'}^{\mathbf{q}m}(t)]^{2\gamma'} \rangle \\
&= 2^{2(1-\gamma-\gamma')} \left[\frac{1}{4} \binom{2\gamma}{\gamma} \binom{2\gamma'}{\gamma'} + \sum_{k=0}^{\gamma-1} \binom{2\gamma}{k} \right. \\
& \left. \times \binom{2\gamma'}{\gamma+\gamma'-k} \cos 2(\gamma-k)(\theta_{js} - \theta_{j's'}) \right]
\end{aligned}
$$

$$
\begin{aligned}
& \langle h_{js}^{\mathbf{q}m}(t)^\gamma h_{j's'}^{\mathbf{q}m}(t)^{\gamma'} h_{JS}^{\mathbf{q}m}(t)^\Gamma h_{J'S'}^{\mathbf{q}m}(t)^{\Gamma'} \rangle \\
&= 2^{2-\gamma-\gamma'-\Gamma-\Gamma'} \left\{ \frac{1}{4} \binom{\gamma}{\frac{1}{2}\gamma} \binom{\gamma'}{\frac{1}{2}\gamma'} \binom{\Gamma}{\frac{1}{2}\Gamma} \binom{\Gamma'}{\frac{1}{2}\Gamma'} \right. \\
& + \binom{\Gamma}{\frac{1}{2}\Gamma} \binom{\Gamma'}{\frac{1}{2}\Gamma'} \sum_{k=0}^{\frac{1}{2}\gamma-1} \binom{\gamma}{k} \binom{\gamma'}{\frac{\gamma+\gamma'}{2}-k} \\
& \times \cos(\gamma-2k)(\theta_{js} - \theta_{j's'}) + \binom{\gamma'}{\frac{1}{2}\gamma'} \binom{\Gamma'}{\frac{1}{2}\Gamma'} \\
& \times \sum_{k=0}^{\frac{1}{2}\gamma-1} \binom{\gamma}{k} \binom{\Gamma}{\frac{\gamma+\Gamma}{2}-k} \cos(\gamma-2k)(\theta_{js} - \theta_{JS}) \\
& + \binom{\gamma'}{\frac{1}{2}\gamma'} \binom{\Gamma}{\frac{1}{2}\Gamma} \sum_{k=0}^{\frac{1}{2}\gamma-1} \binom{\gamma}{k} \binom{\Gamma'}{\frac{\gamma'+\Gamma'}{2}-k} \\
& \times \cos(\gamma-2k)(\theta_{js} - \theta_{J'S'}) + \binom{\gamma}{\frac{1}{2}\gamma} \binom{\Gamma'}{\frac{1}{2}\Gamma'} \\
& \times \sum_{k=0}^{\frac{1}{2}\gamma'-1} \binom{\gamma'}{k} \binom{\Gamma}{\frac{\gamma'+\Gamma}{2}-k} \cos(\gamma'-2k)(\theta_{j's'} - \theta_{JS})
\end{aligned}
$$

$$+\binom{\gamma}{\frac{1}{2}\gamma}\binom{\Gamma}{\frac{1}{2}\Gamma}\sum_{k=0}^{\frac{1}{2}\gamma'-1}\binom{\gamma'}{k}\binom{\Gamma'}{\frac{\gamma'+\Gamma'}{2}-k}$$

$$\times\cos(\gamma'-2k)(\theta_{j's'}-\theta_{J'S'})+\binom{\gamma}{\frac{1}{2}\gamma}\binom{\gamma'}{\frac{1}{2}\gamma'}$$

$$\times\sum_{k=0}^{\frac{1}{2}\Gamma-1}\binom{\Gamma}{k}\binom{\Gamma'}{\frac{\Gamma+\Gamma'}{2}-k}\cos(\Gamma-2k)(\theta_{JS}-\theta_{J'S'})$$

$$+\binom{\Gamma'}{\frac{1}{2}\Gamma'}\sum_{k=0}^{\frac{1}{2}\gamma-1}\sum_{k'=0}^{\frac{1}{2}\gamma'-1}\sum_{m=0}^{1}\binom{\gamma}{k}\binom{\gamma'}{k'}$$

$$\times\binom{\Gamma}{\frac{\gamma+(-1)^m\gamma'+\Gamma}{2}-[k+(-1)^m k']}$$

$$\times\cos[(\gamma-2k)(\theta_{js}-\theta_{JS})$$

$$+(-1)^m(\gamma'-2k')(\theta_{j's'}-\theta_{JS})]$$

$$+\binom{\Gamma}{\frac{1}{2}\Gamma}\sum_{k=0}^{\frac{1}{2}\gamma-1}\sum_{k'=0}^{\frac{1}{2}\gamma'-1}\sum_{m=0}^{1}\binom{\gamma}{k}\binom{\gamma'}{k'}$$

$$\times\binom{\Gamma'}{\frac{\gamma+(-1)^m\gamma'+\Gamma'}{2}-[k+(-1)^m k'])}$$

$$\times\cos[(\gamma-2k)(\theta_{js}-\theta_{J'S'})$$

$$+(-1)^m(\gamma'-2k')(\theta_{j's'}-\theta_{J'S'})]$$

$$+\binom{\gamma'}{\frac{1}{2}\gamma'}\sum_{k=0}^{\frac{1}{2}\gamma-1}\sum_{k'=0}^{\frac{1}{2}\Gamma-1}\sum_{m=0}^{1}\binom{\gamma}{k}\binom{\Gamma}{k'}$$

$$\times\binom{\Gamma'}{\frac{\gamma+(-1)^m\Gamma+\Gamma'}{2}-[k+(-1)^m k']}$$

$$\times\cos[(\gamma-2k)(\theta_{js}-\theta_{J'S'})$$

$$+(-1)^m(\Gamma-2k')(\theta_{JS}-\theta_{J'S'})]$$

$$+\binom{\gamma}{\frac{1}{2}\gamma}\sum_{k=0}^{\frac{1}{2}\gamma'-1}\sum_{k'=0}^{\frac{1}{2}\Gamma-1}\sum_{m=0}^{1}\binom{\gamma'}{k}\binom{\Gamma}{k'}$$

$$\times\binom{\Gamma'}{\frac{\gamma'+(-1)^m\Gamma+\Gamma'}{2}-[k+(-1)^m k']}$$

$$\times\cos[(\gamma'-2k)(\theta_{j's'}-\theta_{J'S'})$$

$$+(-1)^m(\Gamma-2k')(\theta_{JS}-\theta_{J'S'})]$$

$$+\sum_{k=0}^{\frac{1}{2}\gamma-1}\sum_{k'=0}^{\frac{1}{2}\gamma'-1}\sum_{k''=0}^{\frac{1}{2}\Gamma-1}\sum_{l,m=0}^{1}\binom{\gamma}{k}\binom{\gamma'}{k'}\binom{\Gamma}{k''}$$

$$\times\binom{\Gamma'}{\frac{\gamma+(-1)^l\gamma'+(-1)^m\Gamma+\Gamma'}{2}-[k+(-1)^l k'+(-1)^m k'']}$$

$$\times\cos[(\gamma-2k)(\theta_{js}-\theta_{J'S'})+(-1)^l(\gamma'-2k')$$

$$\times(\theta_{j's'}-\theta_{J'S'})+(-1)^m(\Gamma-2k'')(\theta_{JS}-\theta_{J'S'})]\Big\}$$

$$\langle[h_{js}^{\mathbf{q}m}(t)]^{\gamma}[h_{j's'}^{\mathbf{q}m}(t)]^{\gamma'}[h_{JS}^{\mathbf{q}m}(t)]^{\Gamma}\rangle$$

$$=2^{2-\gamma-\gamma'-\Gamma}\Bigg\{\frac{1}{4}\binom{\gamma}{\frac{1}{2}\gamma}\binom{\gamma'}{\frac{1}{2}\gamma'}\binom{\Gamma}{\frac{1}{2}\Gamma}$$

$$+\binom{\gamma'}{\frac{1}{2}\gamma'}\sum_{k=0}^{\frac{1}{2}\gamma-1}\binom{\gamma}{k}\binom{\Gamma}{\frac{\gamma+\Gamma}{2}-k}$$

$$+\binom{\Gamma}{\frac{1}{2}\Gamma}\sum_{k=0}^{\frac{1}{2}\gamma'-1}\binom{\gamma'}{k}\binom{\gamma}{\frac{\gamma+\gamma'}{2}-k}$$

$$\times\cos(\gamma'-2k)(\theta_{j's'}-\theta_{js})$$

$$+\binom{\gamma}{\frac{1}{2}\gamma}\sum_{k=0}^{\frac{1}{2}\Gamma-1}\binom{\Gamma}{k}\binom{\gamma'}{\frac{\gamma'+\Gamma}{2}-k}$$

$$\times\cos(\Gamma-2k)(\theta_{JS}-\theta_{j's'})$$

$$+\sum_{k=0}^{\frac{1}{2}\gamma-1}\sum_{k'=0}^{\frac{1}{2}\gamma'-1}\binom{\gamma}{k}\binom{\gamma'}{k'}\Bigg[\binom{\Gamma}{\frac{\gamma+\gamma'+\Gamma}{2}-k-k'}$$

$$\times\cos[(\gamma-2k)(\theta_{js}-\theta_{JS})+(\gamma'-2k')(\theta_{j's'}-\theta_{JS})]$$

$$+\binom{\Gamma}{\frac{\gamma-\gamma'+\Gamma}{2}-k+k'}$$

$$\times\cos[(\gamma-2k)(\theta_{js}-\theta_{JS})$$

$$-(\gamma'-2k')(\theta_{j's'}-\theta_{JS})]\Bigg]\Bigg\}$$

$$\langle[h_{js}^{\mathbf{q}m}(t)]^{\gamma}[h_{j's'}^{\mathbf{q}m}(t)]^{\gamma'}\rangle$$

$$=2^{2-\gamma-\gamma'}\Bigg[\frac{1}{4}\binom{\gamma}{\frac{1}{2}\gamma}\binom{\gamma'}{\frac{1}{2}\gamma'}+\sum_{k=0}^{\frac{1}{2}\gamma-1}\binom{\gamma}{k}$$

$$\times\binom{\gamma'}{\frac{\gamma+\gamma'}{2}-k}\cos(\gamma-2k)(\theta_{js}-\theta_{j's'})\Bigg].$$

The first of the four terms in (32) is the incoherent sum (*i.e.* without interference) of the double-diffuse scattered wavelets generated at the pair of sites ($js, j's'$) and depends on the relative phase of these two sites. The second term represents the interference between double-diffuse scattered waves generated at different pairs of sites, ($js, j's'$) and ($JS, J'S'$), and depends on the relative phases within each pair (for example $\theta_{js} - \theta_{j's'}$) and between the pairs (for example $\theta_{js} - \theta_{JS}$). The third term represents the interference between a double and a single-diffuse scattered wave and depends on the relative phases within the pair and between each site within the pair and the single-diffuse scattering site. (It is noted that this term vanishes with the time average, under the approximations $a = 1$, β_s, $\beta_{s'} = 1$ and the assumption that the displacements take the standard cosine form.) The last term represents the interference between the wave function due to scattering solely from the reference lattice and each double-diffuse scattered wave and

depends on the relative phase of the two sites at which the double-diffuse scattered wave is generated. Just as with single-diffuse scattering, the contribution to the intensity from double-diffuse scattering depends on the *relative* phases of the diffusely scattering sites. Since these relative phases are translationally invariant, the intensity distribution contributed by the double-diffuse scattered waves also has the periodicity of the reference lattice in that projection. The argument follows similarly for higher-order diffuse scattering terms. Thus, the contribution to the image of a crystal with time-dependent atomic displacements due to the lattice mode $\mathbf{q}m$, $I^{\mathbf{q}m}(\mathbf{r})$, has the translational symmetry of the reference lattice in the plane perpendicular to the incident beam, that is, $I^{\mathbf{q}m}(\mathbf{r}) = I^{\mathbf{q}m}(\mathbf{r} + \mathbf{r}_0)$, where $\mathbf{R}_0 = (\mathbf{r}_0, z_0)$.

Since different lattice modes may be considered uncorrelated, the final intensity of the lattice image is the incoherent sum of the contributions from each of the different phonons (Doyle, 1969), that is,

$$I(\mathbf{r}) = \sum_{\mathbf{q}m} I^{\mathbf{q}m}(\mathbf{r}). \qquad (33)$$

Each $I^{\mathbf{q}m}(\mathbf{r})$ has the periodicity of the reference lattice so $I(\mathbf{r})$ must have the periodicity of the reference lattice, that is, *the high-resolution electron microscope image of a crystal with time-dependent correlated atomic displacements from its reference-lattice sites will have the translational symmetry of the reference lattice in the projection perpendicular to the incident beam.*

4.3. *Comparison of images with static and time-dependent correlated displacements*

It has been demonstrated in the sections above that the periodicity of the HREM image of a crystal with static atomic displacements, as in a modulated structure, is different to that with time-dependent displacements, as generated by phonons. The former has the periodicity determined by the modulation wave in that projection whereas the latter has the periodicity of the reference lattice. This is the intuitive idea behind the extremely useful experimental method described by Van Tendeloo & Amelinckx (1986) for distinguishing whether structured diffuse scattering in electron diffraction patterns is temporal or spatial in origin from the Fourier transform of the corresponding high-resolution image. If the displacements are static, then the HREM image will have the translational symmetry of the modulated structure in that projection and so the transform will exhibit structured diffuse scattering with the same geometry as the electron diffraction pattern. If the displacements are time dependent, then the HREM image will have the translational symmetry of the reference lattice so the Fourier transforms will exhibit Bragg peaks only. In performing this experiment, care must be taken to:

(*a*) Ensure the lattice image is taken with minimal beam convergence to avoid convolution effects. The degree of beam convergence and its effect on the structure of diffuse scattering is of course readily checked by taking a diffraction pattern using the same illumination conditions as applied to the lattice image.

(*b*) Use the largest possible objective aperture to maximize the number of diffusely scattered electrons contributing to the image.

(*c*) Take the optical diffraction pattern from approximately the same region of crystal (as recorded in the lattice image) as the selected-area diffraction pattern was taken from.

5. Dependence of HREM image on phase correlations between atomic displacements

The expressions constructed above were formulated in order to analyse the translational symmetry of the image of a crystal with correlated atomic displacements. All of the terms that depend upon the correlations between the atomic displacements were included in these expressions, however small, so that the argument would be made without approximation. Some of the phase-correlation terms are, however, very small and it is the purpose of this section to consider the nature and extent of their influence on the image using the formulation constructed above.

5.1. *Static displacements – modulated structures*

The intensity distribution in high-resolution images of modulated structures depends on the wavevector defining the correlations between the displaced atoms. The intensity distribution due to diffuse scattering, (27), depends upon the absolute phase of the displacement at each atomic site. As a result, the total intensity distribution is modulated according to the wavevector of the modulated structure in that projection, as discussed in §4.1. However, those terms that have a dependence on phase in the intensity distribution are small. Such terms consist of wavelets (in the curly brackets), all of which have the same shape, independent of column, j, that are multiplied by a weight that has a sinusoidal dependence upon the absolute phase of the displaced atom(s) at the site(s) at which the diffuse scattering has taken place. Since these weights will be as often positive as negative along the depth of a column and since $W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r}) \approx W_{js+1\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})$ [although $W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r}) \neq W_{js'\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})$ in general], the sum over all the weighted wavelets along the column will be small.

Although these terms are small, they are nevertheless significant enough for the modulation of the image intensity distribution they generate to be readily detectable experimentally. For example, as mentioned in §4.3, the intensity modulation is usually sufficiently strong to give rise to diffuse scattering or superlattice

reflections corresponding to $\mathbf{q}_{xy}$ in the Fourier transform of the image (Van Tendeloo & Amelinckx, 1986), even for thin crystals (there is a thin-crystal example in Etheridge, 1996).

5.2. Time-dependent displacements – phonons

For the case of phonons, the final intensity distribution in the high-resolution image can be treated, as in §4.2, as a normalized sum of the intensity distributions due to each of the 'static' modulated structures that represent the dynamic structure at every point in its cycle. As a consequence of this sum, information on the phase of atomic displacements is lost from the large single-diffuse scattering 'non-interference' term [first term of (30)] because it gives the contribution from each atom, summed over every point of its individual cycle, independent of the position of other atoms. [By 'non-interference' terms it is meant the incoherently summed terms; the first terms of (30) and (32).] In other smaller terms, some information on the phase of atomic displacements is still retained, for example, those terms describing the interference between the wavefunctions of electrons that have been scattered diffusely at different sites in the crystal, either one or more times [for example, the second term of (30) and the last three terms of (32)], or those terms describing the contribution from direct multiple diffuse scattering [e.g. the first term of (32)], where the electron wavefunction is tagged by the phase of each of the displaced atoms from which it has been diffusely scattered. The nature and significance of the phase dependence of each of these terms is considered one by one below.

5.2.1. *Single-diffuse scattering.* The contribution to the total intensity distribution made by phase-correlation terms in the single-diffuse scattering expression (30) are first considered. The function $W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})$ is sharply localized about the column j, so that in a high-resolution image $S(\Delta)W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})$ is unlikely to extend much beyond adjacent atomic columns. Thus, the major contribution in the summation over j and J in the phase-dependent term of (30) (the second half of the second term) will be that due to interference between wavelets generated by diffuse scattering at different depths in the same column, that is $j = J$. A small contribution will also be made by wavelets generated by diffuse scattering in adjacent columns but all other terms will be negligible. The single-diffuse scattering intensity therefore depends on the relative phase between all pairs of atoms within a single atomic column and, to a lesser extent, in adjacent columns, but does not depend on any other pairs.

The size of the phase-dependent term of (30) (the second half of the second term) is in any case small, since $\cos(\gamma - 2k)(\theta_{js} - \theta_{JS})$ will be just as often positive as negative across the range of values of $\theta_{js} - \theta_{JS}$, so that

the sum over the length of the columns (*i.e.* over s, S) will be small. Therefore, the phase dependence of the single-diffuse scattered intensity is small but non-zero.

5.2.2. *Double-diffuse scattering.*

(*a*) *First and fourth terms.* Now consider the contribution of double-diffuse scattering, (32). The first and fourth terms correspond to the electron that has scattered diffusely at depth s in column j and then later at depth s' in column j' in its passage through the crystal, generating the wavelet $W_{js\beta_s\gamma,j's'\beta_{s'}\gamma'}^{\mathbf{q}m}(\mathbf{r})$. The first term represents the incoherent sum of these wavelets whilst the fourth represents its interference with the wavefunction generated by scattering from the reference lattice. $W_{js\beta_s\gamma,j's'\beta_{s'}\gamma'}^{\mathbf{q}m}(\mathbf{r})$ will be localized about the positions j and j', having maximum amplitude when $j = j'$, and rapidly tending to zero the further j is from j'. The phase-dependent component of these terms will therefore depend predominantly on the relative phase between atoms in the same atomic column, $j = j'$, and, to a much lesser extent, on atoms in adjacent columns.

As with single-diffuse scattering, the size of these phase-dependent terms (the second part of each of these terms) is small, since they both have a cosine dependence on $\theta_{js} - \theta_{j's'}$ and will be just as often positive as negative across the range of values of $\theta_{js} - \theta_{j's'}$, so that the sum over the length of the columns (*i.e.* over s, s') will be small. That is, the phase-dependent part of the first and fourth terms will be small (but again non-zero).

(*b*) *Second term.* The second term in (32) arises from the interference between different pairs of double-diffuse scattered wavelets, $js, j's'$ and $JS, J'S'$. Since these wavelets will only have a significant amplitude when $j = j'$ or j, j' are nearest neighbours and $J = J'$ or J, J' are nearest neighbours, respectively, their product will only have a large amplitude when $j = j' = J = J'$, with smaller contributions for the cases when j and j' are adjacent or equal to each other, at the same time as J and J' are adjacent or equal to each other, at the same time as at least one of j and j' is adjacent or equal to J or J'. In other words, this term depends predominantly on the relative phases between pairs of atoms in the same column and to a much lesser extent on the relative phases between pairs of atoms in adjacent columns.

The phase-dependent component of the amplitude (32) multiplying the second term will again be small because there are many depth-dependent components in the second term that are positive as there will be negative, so that the overall contribution of the components with a depth dependence will be small.

(*c*) *Third term.* The third term in (32) arises from the interference between single-diffuse scattered wavelets from sites JS and double-diffuse scattered wavelets from the pair of sites, $js, j's'$. Since the single-diffuse scattered wavelets are localized about site J and the double-diffuse scattered wavelets will only have significant

amplitude when $j = j'$ or j, j' are nearest neighbours, their product will only have a large amplitude when $j = j' = J$, with smaller contributions for the cases when j and j' are adjacent or equal to each other, at the same time as J is adjacent or equal to j or j'. So, like the other terms, this term depends predominantly on the relative phases between pairs of atoms in the same column and to a much lesser extent on the relative phases between pairs of atoms in adjacent columns.

The phase-dependent component of the amplitude (32) multiplying the third term will again be small because there are as many depth-dependent components in the second term that are positive as there will be negative, so that the overall contribution of the components with a depth dependence will be small.

(*d*) *Summary*. In summary, the intensity distribution due to scattering from a phonon mode depends in a small way upon the relative phase between pairs of displaced atoms, provided the pairs are either within the same atomic column or in adjacent columns. The strongest dependence is on pairs in the same column.

The terms that depend upon correlations between atoms in a plane perpendicular to the beam will influence the intensity between the reference lattice positions whilst the terms that depend upon correlations between atoms within the same atomic column parallel to the beam will mostly influence the intensity at the reference-lattice positions. The phase-correlation terms therefore contribute in a specific way to the intensity distribution and do not simply contribute a featureless background that does not affect the image contrast. It can also be seen from the discussion above that this is the case even for single-diffuse scattering. [This is contrary to the suggestion of Wang (1992), although Wang's qualitative argument vividly describes the inherent difference between the dependence of images and diffraction patterns on the correlations between atomic displacements.]

It can be seen by inspection that an image of a crystal with time-dependent correlated displacements has a smaller, as well as different, dependence on the phase correlations between displacements than the image of a crystal with equivalent static correlated displacements.

For the case of phonon scattering, the smallness of these phase-correlation terms means that, to a crude approximation, a random-phase model can be used for a qualitative assessment of the contribution of thermal diffuse scattering to the image [Cowley (1988) has shown this is exactly true for single-diffuse scattering under a type of column approximation where the wave function is independent of the depth in the column of a displaced atom, assuming a weak-phase-object approximation for the diffusely scattering slice and ignoring multiple scattering from a single lattice wave], however, for quantitative work, phase correlations should be included.

6. Contrast in the intensity distribution due to scattering from correlated atomic displacements

There has been some discussion about the effect of phonon scattering on contrast in the intensity distribution in a HREM image. [For example Cowley (1988), Rez (1993) and Wang (1992). Each of these references addresses the question using different formulations and different degrees of approximation.] This discussion has also had some bearing on the interpretation of HAADF–STEM (high-angle annular dark field–scanning transmission electron microscope) images, since phonon scattering can make a significant contribution to such images. Whilst it is not the principal aim of the present work, it is worthwhile using the different formulation set up here to consider the resolution and contrast in the intensity distribution generated by scattering from correlated atomic displacements. For brevity, the argument that follows is made for the case of scattering from dynamic displacements (phonons) only. It can readily be seen that an analogous argument applies to scattering from static displacements (modulated structures) but the effects are much weaker.

Firstly, for comparison, consider the intensity distribution due to scattering from a lattice only, without atomic displacements. In analogy with $I_d^{qm}(\mathbf{r}, t)$, (30), $I_b(\mathbf{r})$ can be written as

$$
\begin{aligned}
I_b(\mathbf{r}) &= |\varphi_b(\mathbf{r})|^2 \\
&= \sum_j \sum_{\beta_s} \left\{ |S(\Delta) W_{j\beta_s}(\mathbf{r})|^2 \right\} \\
&\quad + \underbrace{\sum_{j,J}}_{j \neq J, \text{ when } \beta_s = \beta_S} \sum_{\beta_s, \beta_S} \left\{ [S(\Delta) W_{j\beta_s}(\mathbf{r})][S(\Delta) W_{J\beta_S}(\mathbf{r})]^* \right\},
\end{aligned}
$$
(34)

where s is arbitrary and

$$
\begin{aligned}
W_{j\beta_s}(\mathbf{r}) &= (i\ell\varepsilon)^{\beta_s} \sum_{\substack{\alpha_1,\dots,\alpha_N \\ \beta_1,\dots,\beta_N \\ \text{except } \beta_s}} \frac{(i\ell\varepsilon)^{\alpha_1 + \beta_1 + \dots + \alpha_N + \beta_N}}{\alpha_1! \beta_1! \dots \alpha_N! \beta_N!} \\
&\quad \times \{ \Delta^{\alpha_N} [b_N(\mathbf{r})]^{\beta_N} \dots \Delta^{\alpha_s} [U(\mathbf{r} - \bar{\mathbf{r}}_j)]^{\beta_s} \dots \\
&\quad \dots \Delta^{\alpha_1} [b_1(\mathbf{r})]^{\beta_1} \}.
\end{aligned}
$$

As with $I_{d(1)}^{qm}(\mathbf{r}, t)$, (30), $I_b(\mathbf{r})$ has been separated here into an incoherently summed contribution (the sum of the square of each wavelet), which is the first term, plus the remaining sum over the terms describing the interference between different wavelets, which is the second term. Similarly to $I_{d(1)}^{qm}(\mathbf{r}, t)$, the component of the potential, $U(\mathbf{r} - \bar{\mathbf{r}}_j)$, and hence the wavelet $W_{j\beta_s}(\mathbf{r})$, is highly localized about the position $\bar{\mathbf{r}}_j$, so the first term tends to give intensity peaks localized about the reference lattice sites whilst the second term gives intensity peaks in between the reference lattice sites. In this formulation, it is the second term, the interference term,

that generates two of the well established characteristics of lattice resolving images, namely:

(i) the possibility of obtaining significant intensity peaks in between atomic sites;

(ii) the possibility of reversal of contrast with change in the focal length of the objective lens.

This section considers briefly the extent to which these characteristics are present in the intensity distribution arising from scattering from the correlated displacements, $I_d^{\mathbf{q}m}(\mathbf{r}, t)$, or, in other words, how large the interference term is, both relative to the incoherently summed term as well as in an absolute sense.

6.1. *Relative magnitude of the interference term – atomic contrast*

In the image due only to scattering from the reference lattice, $I_b(\mathbf{r})$, (34), the incoherently summed term and the interference term contribute with the same 'weight', that is, there are no multiplicative factors outside the curly brackets. However, in the contribution to the image due to scattering from the displacement potential, $I_d^{\mathbf{q}m}(\mathbf{r}, t)$, (30) and (32), the incoherently summed term and the interference term contribute with different weights, which depend in different ways upon the amplitudes of the atomic displacements in each case. Consider the relative magnitude of these factors for the case of $I_{d(1)}^{\mathbf{q}m}(\mathbf{r}, t)$,

$$\langle [h_{js}^{\mathbf{q}m}(t)]^{2\gamma} \rangle = 2^{-2\gamma} \begin{pmatrix} 2\gamma \\ \gamma \end{pmatrix} \leq \tfrac{1}{2} \quad \text{for all } \gamma$$

$$\langle [h_{js}^{\mathbf{q}m}(t)]^{\gamma} [h_{JS}^{\mathbf{q}m}(t)]^{\Gamma} \rangle$$

$$= 2^{2-\gamma-\Gamma} \left[\frac{1}{4} \begin{pmatrix} \gamma \\ \tfrac{1}{2}\gamma \end{pmatrix} \begin{pmatrix} \Gamma \\ \tfrac{1}{2}\Gamma \end{pmatrix} + \sum_{k=0}^{\frac{1}{2}\gamma-1} \begin{pmatrix} \gamma \\ k \end{pmatrix} \begin{pmatrix} \Gamma \\ \frac{\gamma+\Gamma}{2} - k \end{pmatrix} \right.$$

$$\left. \times \cos(\gamma - 2k)(\theta_{js} - \theta_{JS}) \right]$$

$$= \begin{cases} 0 & \text{for either } \gamma \text{ or } \Gamma \text{ odd} \\[2mm] 2^{2-\gamma-\Gamma} \sum\limits_{k=0}^{\frac{1}{2}\gamma-1} \begin{pmatrix} \gamma \\ k \end{pmatrix} \begin{pmatrix} \Gamma \\ \frac{\gamma+\Gamma}{2} - k \end{pmatrix} \\[2mm] \quad \times \cos(\gamma - 2k)(\theta_{js} - \theta_{JS}) & \text{for } \gamma \text{ and } \Gamma \text{ odd} \\[2mm] \leq 2^{2-\gamma-\Gamma} \left[\tfrac{1}{16} + \sum\limits_{k=0}^{\frac{1}{2}\gamma-1} \begin{pmatrix} \gamma \\ k \end{pmatrix} \begin{pmatrix} \Gamma \\ \frac{\gamma+\Gamma}{2} - k \end{pmatrix} \right. \\[2mm] \left. \quad \times \cos(\gamma - 2k)(\theta_{js} - \theta_{JS}) \right] & \text{for } \gamma \text{ and } \Gamma \text{ even} \end{cases}$$

$$\langle [h_{js}^{\mathbf{q}m}(t)]^{\gamma} \rangle = 2^{-\gamma} \begin{pmatrix} \gamma \\ \tfrac{1}{2}\gamma \end{pmatrix} = \begin{cases} 0 & \text{for } \gamma \text{ odd} \\ \leq \tfrac{1}{2} & \text{for } \gamma \text{ even} \end{cases} \quad (35)$$

Also, for a given *even* γ,

$$2^{-\gamma} \begin{pmatrix} \gamma \\ \tfrac{1}{2}\gamma \end{pmatrix} \leq 2^{1/2-2\gamma} \begin{pmatrix} 2\gamma \\ \gamma \end{pmatrix},$$

that is, $\langle [h_{js}^{\mathbf{q}m}(t)]^{2\gamma} \rangle \leq 2^{1/2} \langle [h_{js}^{\mathbf{q}m}(t)]^{\gamma} \rangle$.

The factors multiplying the incoherently summed terms are significantly greater overall than those multiplying the interference terms. This means that the *relative* intensity between the incoherently summed term and the interference term is greater for $I_d^{\mathbf{q}m}(\mathbf{r}, t)$ than it is for $I_b(\mathbf{r})$. That is, for $I_d^{\mathbf{q}m}(\mathbf{r}, t)$, the intensity between atomic sites will be considerably diminished relative to that at the atomic sites. The 'first characteristic' is therefore a less significant effect in $I_d^{\mathbf{q}m}(\mathbf{r}, t)$ than $I_b(\mathbf{r})$. As a consequence, the intensity distribution due to scattering from the displacement potential will have sharper atomic contrast than that due to scattering from the reference potential without displacements. The argument can be followed similarly for double-diffuse scattering.

It was demonstrated by Cowley (1988), within the phase-object approximation, that scattering from a displacement potential approximated by $\sum_j h_j(t) \nabla_{\mathbf{r}} U(\mathbf{r} - \bar{\mathbf{r}}_j)$ gives enhanced resolution ('sharper atom images') relative to scattering from the non-displaced atomic potential because of the narrower peaks of $\nabla_{\mathbf{r}} U(\mathbf{r} - \bar{\mathbf{r}}_j)$ compared with $U(\mathbf{r} - \bar{\mathbf{r}}_j)$. The suppression of the interference terms in dynamical scattering from the full displacement potential described here represents an additional resolution-enhancing effect with different physical origins, deriving not from the shape of the scattering function but from the periodic nature of the displacement amplitudes.

6.2. *Absolute magnitude of interference term – contrast reversal*

It can be seen from (35) that the absolute value of the interference term between two diffuse scattered wavelets is very small. Half of the wavelets in curly brackets are multiplied by a zero factor (corresponding to either of the gammas being odd). A further quarter are only multiplied by the sinusoidally dependent term (which when summed along a column gives a very small contribution, as described in §5).

Similarly, the interference term between a diffuse scattered wavelet and the reference wavefunction is small since half of the factors are zero (corresponding to gamma odd). In fact, if the weak-phase-object approximation is made for the transmission function of the diffusely scattering atomic layer, that is $\beta_s = 1$, and only single scattering from a given displacement potential is considered, $a = 1$, then this term vanishes completely.

The smallness of the two interference terms in (30) (the second and third terms) means that the contribution to the image from scattering from a phonon mode is dominated by the incoherently summed first term. Since $S(\Delta)$ and $W_{js\beta_s\gamma_i}^{\mathbf{q}m}(\mathbf{r})$ are contained within the modulus of

this term, the contribution to the image due to scattering from the dynamic atomic displacements will not exhibit strong contrast reversal with change in defect of focus or thickness.

The argument has been spelt out explicitly here for single-diffuse scattering but can be readily extended to double-diffuse scattering.

6.3. *Summary*

In summary, as a consequence of the periodic nature of the displacement amplitude, the contribution to a HREM image from the interference terms owing to dynamical scattering from correlated atomic displacements is both small relative to the incoherently summed term and small in absolute terms. The former means that the intensity between the atomic sites will tend to be depressed relative to that at the atomic sites, enhancing the image contrast, and the latter means that contrast-reversal effects will be weaker. The argument has been set out only for time-dependent atomic displacements. Analogous arguments can be made for the scattering from modulated structures, although the interference terms in this case are larger and so the effects are not as strong as for scattering from phonons.

7. Conclusions

A coordinate-space multislice formulation of electron scattering has been constructed that describes dynamical scattering as sums of non-commuting products of kinetic and potential energy terms, without the integral operation of convolution. It is used to analyse high-resolution electron-microscope images of crystals with correlated atomic displacements that are either static (as generated, for example, in modulated structures) or dynamic (as generated by phonons) and, in particular, to determine the translational symmetry and the dependence upon the correlations between atomic displacements of these images.

From the formulation, expressions can be constructed that describe multiple scattering from displaced and nondisplaced atoms. These expressions comprise a component describing multiple scattering from a perfect reference potential without displacements and a component describing multiple scattering from a displacement potential. Expressions are given explicitly for the contribution to the latter component from single- and double-diffuse scattering events embedded between multiple Bragg scattering events.

It is shown that the intensity distribution due to scattering from correlated atomic displacements depends upon the wavevector defining the correlations between these displacements, although the dependence is greater and different in nature for static displacements than for dynamic displacements. Both the components of the wavevector perpendicular, $\mathbf{q}_{xy}$, and parallel, $\mathbf{q}_z$, to the incident beam influence the intensity distribution but their influence manifests itself in different ways, with $\mathbf{q}_{xy}$ mostly affecting the intensity distribution between the reference sites and $\mathbf{q}_z$ mostly affecting it at the reference sites.

The lattice image of static correlated atomic displacements depends upon the *absolute* phase of the displacement at each atomic site, whilst that of time-dependent displacements depends only upon the *relative* phase of the displacements at different atomic sites. For this reason, the lattice image of a crystal with static correlated displacements, as in a modulated structure, has the translational symmetry of the modulated structure in that projection, as determined by the component of the modulation wavevector perpendicular to the incident beam, whereas the lattice image of a crystal with dynamic correlated displacements, as generated by phonons, has the translational symmetry of the crystal's reference structure in that projection (the reference structure being that from which the displacements are measured and which determines the three-dimensional space-group symmetry of the crystal). The Fourier transform of the lattice image of a modulated structure will therefore exhibit structured diffuse scattering with a wavevector determined by the modulation wavevector of the crystal in that projection whereas the transform of the lattice image of a crystal with phonon-induced displacements will exhibit Bragg peaks only. This means that it can be possible to distinguish whether structured diffuse scattering in electron diffraction patterns is due to static or time-dependent displacements by determining whether or not structured diffuse scattering of the same geometry is present in the Fourier transform of the corresponding high-resolution image [an experimental method described by Van Tendeloo & Amelinckx (1986)].

In order to be able to detect the effect of correlated atomic displacements on the translational symmetry of an image, it is not necessary to be able to resolve the <0.1 Å displacements directly. The displacement amplitude of an atom has little effect on the *position* or shape of a peak in the intensity distribution but does affect the *amplitude* of the peak. The question of being able to 'resolve' the displacements is therefore irrelevant. Their dominant effect is to modulate the intensity of image peaks, rather than to shift the position of the peak. Therefore, in order to detect their effect on the crystal's translational symmetry, it is only necessary to be able to detect the intensity modulation, it is not necessary to detect a 0.1 Å shift in atomic spacing. This is well within the capabilities of a microscope resolving 2 Å.

For the case of static displacements, the longitudinal component of the modulation wavevector (that parallel to the incident beam) influences the intensity distribution with the periodicity of the reference lattice whereas the transverse component (that perpendicular to the

incident beam) influences the intensity distribution with the periodicity of the modulated structure in that projection (and fractions thereof). This influence is communicated solely through scattering into or out of higher-order Laue-zone (HOLZ) reflections (regardless of whether the HOLZ reflections are included explicitly within the objective aperture or not). For this reason, the intensity distribution is modulated according to the transverse component of the wavevector of the crystal's modulation.

For static displacements, the dependence of the intensity distribution upon the modulation wavevector is small but not so small that the modulation of the intensity generated by its transverse component cannot be readily detected in the Fourier transform of the image.

Some approximations to the calculation of scattering from static correlated displacements can yield calculated images with the incorrect translational symmetry. For example, approximations that assume the phase of atomic displacements is random or approximations that do not include HOLZ interactions and so ignore the fact that the intensity contributed by a diffusely scattered wave depends upon the depth(s) at which the diffuse scattering took place. Either of these will lead to calculated images with the translational symmetry of the reference lattice. The fact that diffuse scattering is detectable in the Fourier transforms of experimental images with modulated structures is a measure both of the significance of the correlations between displaced atoms and of the importance of the depth at which the diffuse scattering takes place (HOLZ interactions).

For dynamic displacements, the dependence of the intensity distribution upon the phonon wavevector is small but there is nevertheless information contained within the image about the relative phase of the displacements of atoms within the same atomic column parallel to the beam and in adjacent columns perpendicular to the beam (although the extent of the dependence of the latter is much smaller and depends upon the imaging conditions, the crystal projection and the amplitude of the atomic displacements). The transverse component of the wavevector will mostly influence the intensity distribution between the reference lattice sites whereas the longitudual component will mostly influence the intensity distribution at the reference lattice positions. The phase correlation terms therefore add structure to the image contrast and do not simply contribute a homogeneous background.

When scattering from the reference potential, the contribution due to the interference term is given the same 'weight' as that due to the incoherently summed term (the sum of the square of the scattered wavelets). On the other hand, when scattering from the displacement potential (whether static or dynamic), the contri-

bution due to the interference term is suppressed relative to that contributed by the incoherently summed term, that is, the intensity between the reference sites is suppressed relative to the intensity peaks localized about the reference sites. The atomic contrast in the image generated by scattering from the displacement potential is therefore enhanced relative to that generated by scattering from the reference lattice. This improvement in contrast is in addition to that due to scattering from the 'sharper' peaks of the displacement potential, as described by Cowley (1988). This effect is much stronger for scattering from phonons than scattering from modulated structures.

As a consequence of the periodic nature of the displacement amplitude, the absolute value of the interference terms due to scattering from the displacement potential is also small. This means that the contribution to the image from scattering from the displacement potential is dominated by the incoherently summed term and will therefore not exhibit strong reversal of contrast with change in defocus or crystal thickness.

A quantitative treatment of the intensity distribution will of course require inclusion of the interference terms but its qualitative behaviour may be considered roughly as incoherent. The latter is more strongly the case with phonon scattering than with scattering from a modulated structure.

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