

least two images in the first and second defocus bands in order to detect the points at which they are not complementary. The addition of these two images will give the direction in which the atomic columns should be moved in order to improve the accuracy of the input structure. Obviously, these conclusions are still valid for more general structures and planar interfaces. The important parameters are the distances between projected atomic columns. In all cases for which these distances are smaller than the Scherzer resolution limit the effect of the transfer function will result in a large distortion of the image. The determination of the selected defoci for imaging tunnels or atoms has to be studied in a specific case prior to the interface structure determination. It should, however, be concluded that there is an urgent need for developing a practical algorithm allowing deconvolution of the effect of the transfer function and restoration of an aberration-free image with a resolution limit close to the information limit.

References

- ANTERROCHES, C. D' & BOURRET, A. (1984). *Philos. Mag. A*, **49**, 783-807.
- BOURRET, A. & BACMANN, J. J. (1985). *Surf. Sci.* **162**, 495-509.
- BOURRET, A. & BACMANN, J. J. (1987). *Rev. Phys. Appl.* **22**, 563-568.
- BOURRET, A., BILLARD, L. & PETIT, M. (1985). *Inst. Phys. Conf. Ser.* **76**, 23-28.
- BOURRET, A. & PENISSON, J. M. (1987). *JEOL News*, **25E**, 2-7.
- BOURRET, A., THIBAUT-DESSEAUX, J., D'ANTERROCHES, C., PENISSON, J. M. & DE CRECY, A. (1982). *J. Microsc. (Oxford)*, **129**, 337-345.
- COENE, W., VAN DYCK, D., VAN TENDELOO, G. & VAN LANDUYT, J. (1985). *Philos. Mag. A*, **52**, 127-143.
- COWLEY, J. M. (1981). *Diffraction Physics*, 2nd ed. Amsterdam: North-Holland.
- DESSEAUX, J., RENAULT, A. & BOURRET, A. (1977). *Philos. Mag.* **35**, 357-372.
- HEGGIE, H. & JONES R. (1987). *Inst. Phys. Conf. Ser.* **87**, 367-374.
- KRAKOW, W., WETZEL, J. T. & SMITH, D. A. (1986). *Philos. Mag. A*, **53**, 739.
- LYNCH, D. F. (1971). *Acta Cryst.* **A27**, 399-407.
- NOWICKI, T. N., PENISSON, J. M. & BISCONDI, M. (1988). Proc. Conf. Interface Science and Engineering, Lake Placid, New York, 1987. To be published in *J. Phys. (Paris)*.
- PAPON, A. M., PETIT, M., SILVESTRE, G. & BACMANN, J. J. (1983). *J. Microsc. Spectrosc. Electron.* **8**, 135-146.
- PENISSON, J. M., NOWICKI, T. & BISCONDI, M. (1988). Submitted to *Philos. Mag.*
- RADI, G. (1970). *Acta Cryst.* **A26**, 41-56.
- SKARNULIS, A. J. (1982). *J. Microsc. (Oxford)*, **127**, 39-46.
- SPENCE, J. C. H. (1981). *Experimental High Resolution Electron Microscopy*. Oxford: Clarendon Press.
- TANAKA, N. & COWLEY, J. M. (1987). *Acta Cryst.* **A43**, 337-346.
- TERSOFF, J. (1986). *Phys. Rev. Lett.* **56**, 632-635.
- VAN DYCK, D. & COENE, W. (1987). *Acta Cryst.* **A43**, C248.
- VAN DYCK, D., VAN TENDELOO, G. & AMELINCKX, S. (1982). *Ultramicroscopy*, **10**, 263-280.

Acta Cryst. (1988). **A44**, 847-853

Electron Microscopy of Crystals with Time-Dependent Perturbations

BY J. M. COWLEY

Department of Physics, Arizona State University, Tempe, Arizona 85287-1504, USA

(Received 15 February 1988; accepted 20 April 1988)

Abstract

The electron microscope image intensity of a thin crystal is described as a time average of the image of a crystal perturbed by time-dependent fluctuations corresponding to thermal motion of the atoms or low-energy electronic excitations. For very thin crystals the phase-object approximation indicates that images having atomic resolution may be obtained from the inelastically scattered electrons. It is shown that the use of suitable approximations allows estimates to be made of the contribution of the inelastically scattered electrons to the high-resolution images of thicker crystals. The resolution of images formed by inelastically scattered electrons is not affected by the non-localization of the inelastic scattering process.

1. Introduction

Recent trends towards the more quantitative uses of electron microscopy have emphasized the need for a more complete assessment of the formation of images by the electrons inelastically scattered from the specimen (Cowley & Smith, 1987). Existing treatments such as those of Misell & Atkins (1973) and Kohl & Rose (1985) appear adequate for most moderate-resolution imaging but some uncertainties exist regarding the possibility that inelastically scattered electrons may contribute to the atomic-scale detail of high-resolution images of crystals.

For the scattering by phonons, or the thermal motions of atoms, it is usually considered adequate to apply a Debye-Waller factor to the atomic scattering amplitudes so that the potential distribution in

real space is convoluted by a smearing function. Then for thick crystals an absorption function with non-zero Fourier coefficients is applied. The thermal diffuse scattering occurs at high angles in the diffraction pattern and for medium-resolution bright-field imaging it may be assumed that most, or all, of the scattering is excluded by the objective aperture. For high-resolution imaging, however, the objective aperture is large enough to include an appreciable proportion of the thermal diffuse scattering and the question arises as to how this scattering will contribute to the images.

For the excitations of plasmons or single-electron excitations, involving energy losses in the range of 5 to 30 eV, the inelastic scattering is assumed to occur only in the small-angle range of 0.1 to 1 mrad. The corresponding absorption coefficients for the elastic scattering are dominated by the zero-order Fourier coefficient although for single-electron excitations some small amplitudes for non-zero Fourier coefficients have been calculated (Doyle, 1970). Imaging with the small-angle inelastic scattering is not expected to give any high-resolution detail in the weak-phase-object approximation (WPOA) because of the non-localized nature of the excitations. However, elastic scattering of the inelastically scattered electrons or inelastic scattering of the elastically scattered electrons can in principle give resolution comparable to that for elastic scattering alone. In practice the chromatic aberration for conventional transmission electron microscopy (TEM) (but not for scanning transmission electron microscopy, STEM) is a complicating factor if the range of energies used to form the image is not restricted to 1 or 2 eV by an energy filter for 100–200 keV electron beams. For higher voltages the chromatic aberration effect is correspondingly less, so that for 1 MeV, for example, with chromatic aberration constant $C_c = 2$ mm, the chromatic aberration effect will not appreciably degrade a resolution of 3 Å unless the energy range exceeds about 20 eV. Hence the contribution of the inelastic scattering to high-resolution images may be increasingly important as the resolution is improved and higher accelerating voltages are used.

It is the purpose of this paper to present a formulation of the problem of incorporating the various forms of inelastic scattering into the computed high-resolution images of crystals in such a way that simple approximations may be made to allow estimates of the magnitudes of the effects to be expected. Firstly a phase-object approximation will be made to treat a very thin sample, or a single slice for use in a multi-slice treatment. This allows the WPOA effects to be seen and shows the nature of the complications which arise from dynamical diffraction effects. Then the means for including the diffuse scattering in multi-slice formulations will be reviewed and simplifying approximations will be suggested so that the magni-

tude of effects to be expected for thicker crystals can be estimated. The formulation follows the principles of Van Hove (1954) as previously applied to a range of diffraction problems (Cowley, 1981). The inelastic scattering is taken into account by considering that the potential distribution of the sample is a function of time. The changes of energy involved are assumed to have a negligible effect on the interaction constant and the Fresnel diffraction processes within the sample but are taken into account in the imaging process by assuming the transfer function to be modified by the appropriate change of the objective lens defocus, Δf .

2. General formulation

The wave function at the exit face of a sample may be written as $\psi(\mathbf{r}, t)$, where \mathbf{r} is a two-dimensional vector, coordinates x, y . The image intensity is then

$$I(\mathbf{r}) = \langle |\psi(\mathbf{r}, t) * t(\mathbf{r})|^2 \rangle \quad (1)$$

where the angular brackets $\langle \rangle$ represent a time average; thus

$$I(\mathbf{r}) \equiv \langle I(\mathbf{r}, t) \rangle = \lim_{T \rightarrow \infty} T^{-1} \int_0^T I(\mathbf{r}, t) dt.$$

In (1), $t(\mathbf{r})$ is the complex spread function given by Fourier transform of the transfer function $T(\mathbf{u})$ where \mathbf{u} is the two-dimensional reciprocal-space vector.

The time dependence of $\psi(\mathbf{r}, t)$ can be represented as time-dependent deviations from an average function $\psi_0(\mathbf{r}) \equiv \langle \psi(\mathbf{r}, t) \rangle$.

Then

$$\psi(\mathbf{r}, t) = \psi_0(\mathbf{r}) + \Delta\psi(\mathbf{r}, t) \quad (2)$$

and

$$\langle \Delta\psi(\mathbf{r}, t) \rangle = 0.$$

Equation (1) becomes

$$I(\mathbf{r}) = |\psi_0(\mathbf{r}) * t_0(\mathbf{r})|^2 + \sum_m \langle |\Delta\psi_m(\mathbf{r}, t) * t_m(\mathbf{r})|^2 \rangle \quad (3)$$

where $t_0(\mathbf{r})$ is the spread function corresponding to zero energy loss and the index m refers to distinguishable energy loss values for which the transfer functions $T_m(\mathbf{u})$ and spread functions $t_m(\mathbf{r})$ are appreciably different. In the multi-slice formulation for the many-beam dynamical diffraction of electrons (Cowley & Moodie, 1957), the exit wave function is expressed in terms of the transmission function $q_n(\mathbf{r}, t)$ for each of N slices of crystal taken perpendicular to the incident-beam direction, and the propagation function $p_n(\mathbf{r})$ for propagation of the wave between the center of one slice and center of the next. With a convenient notation,

$$\psi(\mathbf{r}, t) = \psi_i(\mathbf{r}) \cdot \mathbf{q}_0^N(\mathbf{r}, t) \quad (4)$$

where $\mathbf{q}_0^N(\mathbf{r}, t)$ represents all the processes of multiplication by $q_n(\mathbf{r}, t)$ with n from 0 to N and convolution by the propagation functions $p_n(\mathbf{r})$, and $\psi_i(\mathbf{r})$ is the incident wave function, which for convenience, is taken as unity. Then

$$\psi(\mathbf{r}, t) = \mathbf{q}_0^{n-1}[q_n(\mathbf{r}, t) * p_n(\mathbf{r})]\mathbf{q}_{n+1}^N. \quad (5)$$

The inelastic scattering may be considered to arise from the time-dependent variations of $q_n(\mathbf{r}, t)$:

$$q_n(\mathbf{r}, t) = \langle q_n(\mathbf{r}, t) \rangle + \Delta q_n(\mathbf{r}, t). \quad (6)$$

Then the exit wave function may be expressed as a series in powers of the small quantities $\Delta q_n(\mathbf{r}, t)$:

$$\begin{aligned} \psi(\mathbf{r}, t) = & \langle \mathbf{q}_0^{n-1}(\mathbf{r}, t) \rangle [\langle q_n(\mathbf{r}, t) \rangle * p_n(\mathbf{r})] \\ & \times \langle q_{n+1}^N(\mathbf{r}, t) \rangle + \sum_n \langle \mathbf{q}_0^{n-1}(\mathbf{r}, t) \rangle \\ & \times [\Delta q_n(\mathbf{r}, t) * p_n(\mathbf{r})] \langle \mathbf{q}_{n+1}^N(\mathbf{r}, t) \rangle \\ & + \sum_{m>n} \langle \mathbf{q}_0^{m-1}(\mathbf{r}, t) \rangle [\Delta q_m(\mathbf{r}, t) * p_m(\mathbf{r})] \\ & \times \langle \mathbf{q}_{m+1}^N(\mathbf{r}, t) \rangle + \sum_{\rho>m>n} \dots \quad (7) \end{aligned}$$

The first term of (7) is equal to $\langle \psi(\mathbf{r}, t) \rangle = \psi_0(\mathbf{r})$ if all other terms average to zero. The second term, the first-order inelastic scattering, averages to zero by definition. The averages of the further terms may contain non-zero components if there is correlation between the perturbations in different slices, *i.e.* if

$$\langle \Delta q_n(\mathbf{r}, t) \Delta q_m(\mathbf{r}, t) \rangle \neq 0.$$

For thermal diffuse scattering (TDS), it seems probable that, as in the case of the calculation of diffraction pattern intensities (Doyle, 1969), these components will, in general, be very small and can be neglected. Then (7) may be written as

$$\psi(\mathbf{r}, t) = \psi_0(\mathbf{r}, t) + \psi_1(\mathbf{r}, t) + \psi_2(\mathbf{r}, t) + \dots \quad (8)$$

and

$$\begin{aligned} I(\mathbf{r}) = & |\psi_0(\mathbf{r}) * t(\mathbf{r})|^2 + \langle |\psi_1(\mathbf{r}, t) * t(\mathbf{r})|^2 \rangle \\ & + \langle |\psi_2(\mathbf{r}, t) * t(\mathbf{r})|^2 \rangle + \dots \quad (9) \end{aligned}$$

It is customary to limit considerations of diffuse scattering in electron diffraction to the first two of these terms, *i.e.* to neglect double-diffuse scattering and higher-order terms.

3. Single-slice formulation

As the basis for consideration of the general expression (7) and in order to illustrate the nature of the effects to be expected, it is appropriate to consider the form of the $\langle q_n(\mathbf{r}, t) \rangle$ and $\Delta q_n(\mathbf{r}, t)$ terms and the diffraction and imaging of a very thin crystal. The

transmission function for a single slice of crystal is

$$\begin{aligned} q(\mathbf{r}, t) = & \exp[-i\sigma\varphi(\mathbf{r}, t)] \\ = & \exp[-i\sigma\langle\varphi(\mathbf{r}, t)\rangle] \exp[-i\sigma\Delta\varphi(\mathbf{r}, t)], \quad (10) \end{aligned}$$

where $\varphi(\mathbf{r}, t)$ is the projection in the direction of the incident beam, the z axis, of the potential distribution of the crystal:

$$\varphi(\mathbf{r}, t) = \int \varphi(x, y, z, t) dz. \quad (11)$$

The interaction constant σ is equal to $\pi/\lambda E$ and, by definition,

$$\langle \Delta\varphi(\mathbf{r}, t) \rangle = 0. \quad (12)$$

The terms in (6) are then

$$\begin{aligned} \langle q(\mathbf{r}, t) \rangle = & \exp[-i\sigma\langle\varphi(\mathbf{r}, t)\rangle] \\ & \times [1 - \frac{1}{2}\sigma^2\langle\Delta\varphi^2(\mathbf{r}, t)\rangle + \dots] \\ = & \exp[-i\sigma\langle\varphi(\mathbf{r}, t)\rangle - \mu(\mathbf{r})] \end{aligned}$$

where

$$\mu(\mathbf{r}) = \frac{1}{2}\sigma^2\langle\Delta\varphi^2(\mathbf{r}, t)\rangle \quad (13)$$

and

$$\begin{aligned} \Delta q(\mathbf{r}, t) = & \exp[-i\sigma\langle\varphi(\mathbf{r}, t)\rangle] [-i\sigma\Delta\varphi(\mathbf{r}, t) \\ & - \frac{1}{2}\sigma^2\Delta\varphi^2(\mathbf{r}, t) + \dots] \\ = & \langle q(\mathbf{r}, t) \rangle \{-i\sigma\Delta\varphi(\mathbf{r}, t) \\ & - \frac{1}{2}\sigma^2[\Delta\varphi^2(\mathbf{r}, t) - \langle\Delta\varphi^2(\mathbf{r}, t)\rangle] + \dots\}. \quad (14) \end{aligned}$$

Thus, to at least the second order in $\Delta\varphi(\mathbf{r}, t)$, $\langle q(\mathbf{r}, t) \rangle$ can be expressed as the transmission function of the time-averaged periodic structure with an added absorption function. In the diffraction patterns from thin crystals, the sharp Bragg reflections are given by Fourier transformation of (13). The time averaging, $\langle \varphi(\mathbf{r}, t) \rangle$, introduces the Debye-Waller factor for thermal motions and a corresponding factor for other inelastic processes. There is also a loss of energy represented by the absorption function $\mu(\mathbf{r})$. The deviation from the average transmission function, (14), is given by a modulation of the time-averaged transmission function by the time-dependent deviation function. In the diffraction pattern, this term gives the diffuse scattering, which is similar to the kinematical diffuse scattering convoluted by the amplitude distribution for the sharp Bragg reflections (Cowley & Fields, 1979).

If a very thin crystal, equivalent to a single slice of a thicker crystal, is imaged, the observed image intensity is

$$\begin{aligned} I(\mathbf{r}) = & \langle |q(\mathbf{r}, t) * t(\mathbf{r})|^2 \rangle \\ = & \langle |q(\mathbf{r}, t) * t(\mathbf{r})|^2 \rangle + \sum_m \langle |\Delta q_m(\mathbf{r}, t) * t_m(\mathbf{r})|^2 \rangle. \quad (15) \end{aligned}$$

It is customary to consider only the first term of (15), sometimes including the absorption function. The second term, arising from the imaging of the diffuse background of the diffraction pattern, is usually neglected. The second term will, in fact, tend to zero for poor resolution because $\Delta q(\mathbf{r}, t)$ is equally often positive and negative and the convolution with a sufficiently broad $t(\mathbf{r})$ function will average $\Delta q(\mathbf{r}, t)$ to zero. On the other hand, for very high resolution, the second term will cancel out the effect of absorption in the first term and no absorption function should apply. In the limit that $t(\mathbf{r})$ becomes a δ function, *i.e.* for an ideally perfect microscope with no aberrations or defocus, the contrast is zero, as expected. For present-day high-resolution imaging conditions, the situation is intermediate between these two extreme cases.

High-resolution images of crystals are usually obtained with the incident beam in an axial direction. Then, ideally, the projection $\varphi(\mathbf{r})$ contains non-overlapping peaks due to individual rows of atoms, and the function $\Delta\varphi(\mathbf{r}, t)$ occurring in (13) and (14) may usually be written

$$\Delta\varphi(\mathbf{r}, t) = \sum_i \varepsilon_i(t) \Delta\varphi_i(\mathbf{r} - \mathbf{r}_i) \quad (16)$$

where $\Delta\varphi_i(\mathbf{r})$ is some time-independent function characteristic of the row of atoms and the time dependence comes in a multiplicative constant $\varepsilon_i(t)$. If, in the high-resolution image, the individual atom rows are resolved, it may be assumed that the functions $\Delta\varphi_{im}(\mathbf{r}) * t_m(\mathbf{r})$ do not overlap, so that the last term of (15) becomes

$$\sum_m \sum_i \{ \exp[-i\sigma\langle\varphi(\mathbf{r}, t)\rangle] (-i\sigma) \times [\varepsilon_{im}(t) \Delta\varphi_{mi}(\mathbf{r} - \mathbf{r}_i)] * t(\mathbf{r}) \}^2 \dots \quad (17)$$

In the WPOA, the first-order term is

$$\sigma^2 \sum_m \sum_i \langle \varepsilon_{im}^2(t) \rangle |\Delta\varphi_{mi}(\mathbf{r} - \mathbf{r}_i) * t_m(\mathbf{r})|^2. \quad (18)$$

The next term is

$$\sigma^3 \sum_m \sum_i \langle \varepsilon_{im}^2(t) \rangle 2 \operatorname{Re} \{ [\Delta\varphi_{mi}(\mathbf{r} - \mathbf{r}_i) * t_m(\mathbf{r})] \times [\langle\varphi(\mathbf{r}, t)\rangle \Delta\varphi_{mi}(\mathbf{r} - \mathbf{r}_i) * t_m(\mathbf{r})] \}. \quad (19)$$

The last bracket of this is dominated by whichever of the two functions, $\langle\varphi(\mathbf{r}, t)\rangle$ or $\Delta\varphi_m(\mathbf{r} - \mathbf{r}_i)$, is the narrower. In fact, from the full POA expression, (17), it is seen that if $\langle\varphi(\mathbf{r}, t)\rangle$ varies more slowly than $\Delta\varphi_{mi}(\mathbf{r} - \mathbf{r}_i)$, the influence of the average projected potential will be small and the total contribution of the diffuse scattering to the image will be given to a reasonable approximation by (18). Thus the diffuse scattering by itself will produce an image of atomic resolution, provided that the chromatic aberration effects, introduced by the range of $t_m(\mathbf{r})$ functions, are not too great.

On the other hand, if the $\Delta\varphi_{mi}(\mathbf{r} - \mathbf{r}_i)$ functions vary much more slowly than $\langle\varphi(\mathbf{r}, t)\rangle$ and are essentially constant, equal to $\varphi_{mi}(\mathbf{r}_i)$ over interatomic distances, (17) will reduce to

$$\sigma^2 \sum_m \sum_i \langle \varepsilon_{im}^2(\mathbf{r}) \rangle |\Delta\varphi_m(\mathbf{r}_i)|^2 \times |\exp[-i\sigma\langle\varphi(\mathbf{r}, t)\rangle * t(\mathbf{r})]|^2 \quad (20)$$

and the contribution of the diffuse scattering to the image will be proportional to that for the average structure.

4. Thermal diffuse scattering

For the time-dependent displacement, $\varepsilon_i(t)$, of individual atoms,

$$\varphi(\mathbf{r}, t) = \sum_i \varphi_i[\mathbf{r} - \mathbf{r}_i - \varepsilon_i(t)]$$

so that

$$\Delta\varphi(\mathbf{r}, t) = \sum_i \varepsilon_i(t) \operatorname{grad} \varphi_i(\mathbf{r} - \mathbf{r}_i). \quad (21)$$

The thermal diffuse scattering in the diffraction pattern for a single atom would be

$$\langle \varepsilon_i^2(t) \rangle u^2 |\Phi(\mathbf{u})|^2,$$

but for an assembly of atoms this distribution is modulated by the effects of the correlations of atom movements (Cowley, 1981). The absorption function to be applied to the sharp Bragg reflection is

$$\mu(\mathbf{r}) = \frac{1}{2} \sigma^2 \sum_i \langle \varepsilon_i^2(t) \rangle |\operatorname{grad} \varphi_i(\mathbf{r} - \mathbf{r}_i)|^2. \quad (22)$$

From (18), the WPOA contribution to the high-resolution image intensity, since $t_m(\mathbf{r}) = t_0(\mathbf{r})$, is

$$\sigma^2 \sum_i \langle \varepsilon_i^2(t) \rangle |\operatorname{grad} \varphi(\mathbf{r} - \mathbf{r}_i) * t(\mathbf{r})|^2. \quad (23)$$

Since $\operatorname{grad} \varphi(\mathbf{r})$ is antisymmetric, convolution with the symmetric function $t(\mathbf{r})$ will reduce this contribution unless $t(\mathbf{r})$ is a narrower function than $\varphi_i(\mathbf{r})$. However, since $\operatorname{grad} \varphi(\mathbf{r})$ gives a narrower peak than $\langle\varphi(\mathbf{r}, t)\rangle$ the thermal diffuse scattering will give a contribution to the image having better resolution (sharper atom images) than the elastic Bragg reflection image, in both the WPOA and the POA [following (20)].

5. Plasmons and single-electron excitations

For the excitations of outer-shell or conduction-band electrons, the ground state for even the most loosely bound electrons is strongly modulated by the periodic atomic distribution. The excited state may be considered as modulated by the periodic structure and also by a plasmon wave, $\exp(i\mathbf{q} \cdot \mathbf{r})$. Following Sturm (1982), we may write the fluctuation of the potential distribution as

$$\Delta\varphi(\mathbf{r}) = \sum_{\mathbf{h}} \Delta\Phi(\mathbf{h}) \exp[i(\mathbf{q} + 2\pi\mathbf{h}) \cdot \mathbf{r}]. \quad (24)$$

The time dependence enters through the frequency of the plasmon excitations. Averaging over time gives zero for the first-order terms and $[\Delta\varphi(\mathbf{r})]^2$ for the second-order terms, as before. For metals having conduction electrons well separated in energy from the core electrons (such as Al) the magnitudes of the $\Delta\Phi(\mathbf{h})$ for $\mathbf{h} \neq 0$ may be small and decreasing rapidly with $|\mathbf{h}|$. For other crystals the values of the $\Delta\Phi(\mathbf{h})$ may be larger, corresponding to the greater concentration of the electrons around the atoms. The $\Delta\varphi_{mi}(\mathbf{r}-\mathbf{r}_i)$ distributions correspond to the outer shells of electrons around the atoms and so are less sharply peaked than the $\langle\varphi(\mathbf{r}, t)\rangle$. Because the $\Delta\varphi_{mi}(\mathbf{r}-\mathbf{r}_i)$ distributions are centrosymmetrical, the convolution with $t(\mathbf{r})$ in (18) will not reduce magnitudes as in the case of thermal motions. The contributions to the image in the WPOA have somewhat poorer resolution than for the elastic scattering in the absence of chromatic aberration effects. The effects of chromatic aberration, represented by the summation over m in (18), provide the main limitation of resolution for current electron microscopes unless an energy filter with a narrow window is used.

It may be noted that the argument leading to (18) does not imply crystallinity of the specimen so that, in the WPOA, the resolution of images of any samples by inelastically scattered electrons is limited almost entirely by the chromatic aberration effects.

For the POA, when the higher-order terms dominate the contrast, the resolution of the images is the same as for the elastic scattering, degraded by the chromatic aberration effects. The presence of the $\exp[-i\sigma\langle\varphi(\mathbf{r}, t)\rangle]$ term in (17) and in (20) is consistent with the familiar concept that, except for very thin crystals, the imaging of inelastically scattered electrons is dominated by combined inelastic plus elastic scattering, which can give much the same image resolution as for elastic scattering only (Ajika, Hashimoto, Yamaguchi & Endoh, 1985).

6. Approximations for the multi-slice formulation

By substituting the expression for $\Delta q(\mathbf{r}, t)$ from (14) in the general multi-slice formula (9), with $\psi(\mathbf{r}, t)$ given by (7), it is possible, in principle, to calculate the contribution of the inelastic scattering to the image for the general many-beam transmission case. The first term of (9) represents a single multi-slice calculation for N slices using the time-averaged potential function and the appropriate absorption function. The second term of (9), as in the diffraction pattern calculations (Doyle, 1969), involves N sets of multi-slice calculations; one for the single diffuse scattering taking place in each of the N slices. Each set of multi-slice calculations must be made for a number M of directions of diffuse scattering between the Bragg reflections sufficiently large to represent the

continuously varying function in two dimensions in reciprocal space. If we take M as 100, for example, the number of N -slice calculations which must be made is $100N$. For the third term of (9) the number of N -slice calculations to be made would be $100N^2$. Thus for even the first approximation to the inelastic contribution it is scarcely feasible to follow this scheme, especially when it is realized that, for high-resolution images of moderately thick crystals (200 Å or more), N should be of the order of 100.

The calculations problem can be greatly simplified for special, although commonly occurring, cases by using further approximations analogous to those used for the POA. For an incident beam in a principal axial direction of a reasonably close-packed structure, it is possible to make a column approximation which is valid for much greater thicknesses than for arbitrary orientations. Marks (1986) and Kirkland, Loane & Silcox (1987) have shown that a channelling phenomenon occurs which confines the electrons to single rows of atoms through the crystal. It has been confirmed by calculations for metal structures by Tanaka & Cowley (1987) that a column approximation with a single line of atoms as a column may apply for thicknesses of the order of 200 Å in the axial orientation whereas thicknesses of 50 Å would normally be considered excessive (see also Coene, Van Dyck, Van Tendeloo & Van Landuyt, 1985). Calculations made for metal structures containing defects (vacancies, substitute atoms) suggest that each column of atoms through the crystal may be considered to be imaged separately. Then $\psi_1(\mathbf{r}, t)$ in (8) may be written as

$$\sum_m \sum_n \sum_i \langle q_0^{n-1}(\mathbf{r}, t) \rangle \{ \exp[-i\sigma\langle\varphi_n(\mathbf{r}, t)\rangle] \times [-i\sigma\epsilon_{inm}(t)\Delta\varphi_{inm}(\mathbf{r}-\mathbf{r}_i)] * p_n(\mathbf{r}) \} \cdot \langle q_N^{n+1}(\mathbf{r}, t) \rangle, \quad (25)$$

and, if the individual columns of atoms give well resolved peaks in the image,

$$\begin{aligned} & \langle |\Delta\psi_1(\mathbf{r}, t) * t(\mathbf{r})|^2 \rangle \\ &= \sum_m \sum_n \sum_i \langle |[\psi_0(\mathbf{r})(-i\sigma)\epsilon_{inm}(t) \\ & \quad \times \Delta\varphi_{inm}(\mathbf{r}-\mathbf{r}_i)] * t_m(\mathbf{r})|^2 \rangle, \end{aligned} \quad (26)$$

since it is a consequence of the column approximation that the position of an atom, or a displacement of an atom within the column, does not affect the result, so that each deviation $\Delta q_n(\mathbf{r}, t)$ may be considered to occur at the exit face of the crystal. Then, if we consider all slices to be identical,

$$\begin{aligned} & \langle |\psi_1(\mathbf{r}, t) * t(\mathbf{r})|^2 \rangle \\ &= N\sigma^2 \sum_m \sum_i \langle \epsilon_{im}^2(t) \rangle \{ \psi_0(\mathbf{r})\Delta\varphi_{im}(\mathbf{r}-\mathbf{r}_i) \} * t_m(\mathbf{r}) \}^2. \end{aligned} \quad (27)$$

Thus, in the case of thermal motion, to calculate the first-order contribution of the TDS to the image the exit wave function is calculated for the time-averaged crystal; this wave function is multiplied by $\text{grad } \varphi_i(\mathbf{r} - \mathbf{r}_i)$ for each projected atom position, the spread function is applied, and the intensity is calculated. The image intensity contribution for each atom peak position is scaled according to the mean square displacement of the atoms, and is linearly proportional to the crystal thickness.

The additional simplification of (18), applicable for a thin phase object, will not apply in this case because $|\psi_0(\mathbf{r})|^2 \neq 1$. The TDS contribution, (27), will partly compensate for the loss of intensity in the image for the averaged structure arising from the presence of the absorption function. To the level of approximation used, this compensation should be complete in the limit of very high resolution. Thus it appears possible that in practical calculations the scaling of the terms involved can be checked by calculating the total intensity, $\int I(\mathbf{r}) \, d\mathbf{r}$, at the exit face of the crystal.

For electronic excitations, similar considerations apply, except that there may well be correlations in the $\Delta\varphi_{im}$ for successive slices. Then, over the range of correlation, the amplitudes rather than the intensities are added so that for a correlation range of M slices, (27) will contain the factor NM in place of N (Doyle, 1971). In the approximation that $\Delta\varphi_{im}$ varies more slowly than $\psi_0(\mathbf{r})$, the contribution of (27) to the image is identical with that for the elastic scattering except for the chromatic aberration effects which may be best evaluated from a known chromatic aberration constant and a measured energy-loss spectrum.

Concluding remarks

The effects of inelastic scattering on high-resolution images of crystals may be taken into consideration by: (a) using the time-averaged potential distribution for the elastic scattering; (b) applying an absorption function to the elastic scattering calculation; and (c) adding the high-resolution image produced by the inelastically scattered electrons included in the objective aperture, incorporating the chromatic aberration effects due to the spread of energies when appropriate. For thermal diffuse scattering, the time-averaged potential distribution is found by applying an appropriate Debye-Waller factor to the structure amplitudes used to calculate the potential distribution. The absorption functions which are available for use (Doyle, 1970; Radi, 1970) have been calculated on the basis that all thermal diffuse scattering is excluded from the image and the image is formed from the sharp Bragg elastic reflections only. For the present case of high-resolution imaging, some thermal diffuse scattering is included in the bright-field aper-

ture and contributes to the image. In our formulation we have separated out the effect of the absorption function applied to the elastic imaging and the contribution of the diffuse scattering to the image, which takes the form of a high-resolution image with atomic peaks which are sharper than those for the elastic scattering. The limitation of this contribution from the diffuse scattering by the objective aperture is included through the application of the transfer function of the objective lens in the imaging process.

The contribution of the thermal diffuse scattering to the image cancels out, to some extent, the effect of the absorption function. For an ideally perfect microscope, with $t(\mathbf{r}) = \delta(\mathbf{r})$, this cancellation would be complete. An estimate of the extent of this calculation in practice may be obtained by estimating the fraction of the thermal diffuse scattering intensity which passes through the objective aperture. Calculations of this sort, made for Si and Pb crystals at room temperature for a 400 keV microscope having a resolution of about 1.6 Å, suggest that a fraction of 10 to 20% of the diffuse scattering will be included in the aperture for very thin crystals. The fraction will increase for improved resolution, thicker crystals and higher temperatures.

In most multi-slice calculations now made, the absorption functions are not included. Calculations made by Pirouz (1979) suggest that for the case of a Cu₃Au alloy with 100 keV electrons and a resolution of approximately 3 Å, the effects of the absorption function produce gross modifications of the relative heights or positions of the intensity maxima in the image for thicknesses exceeding 200 Å. For improved resolutions, the effects should be greater for a given thickness and modifications should become apparent for smaller thicknesses, especially if quantitative interpretation of image intensities is attempted.

To a first rough approximation, the contribution of the thermal diffuse scattering to the image may be taken into account by reducing the magnitude of the absorption function by an appropriate small factor.

For the inelastic scattering by excitation of the crystal electrons, the modification of the potential distribution by time averaging is not normally considered. The absorption functions are usually considered to have no non-zero Fourier coefficients, at least for plasmon excitations; however, for the quantitative assessment of elastic images a reassessment of this assumption is clearly necessary in order that the known periodic modulations of the initial and final states should be taken into consideration, especially for materials other than ideal metals (Sturm, 1982).

In the absence of chromatic aberration effects, the contributions of the diffuse scattering to the image in these cases would very largely compensate for the effects of the absorption function, since most of the

diffuse scattering is included within the objective aperture. However, the loss of resolution due to chromatic aberration effects is of importance for most present-day high-resolution electron microscopy, so that for quantitative image interpretation, the elastic and inelastic images should be calculated separately with appropriate absorption functions.

It may be noted that, for the conditions of high-resolution imaging which we have discussed, the image intensities are not affected by the correlations of the perturbations of the potential in neighboring atoms. The non-localization of the excitation of the crystal in an inelastic scattering process can influence the elastic scattering only through such correlation effects. The effects of the non-localization can thus appear in the diffraction patterns but do not affect the high-resolution images.

This work was supported by NSF grant DMR 8510059.

References

- AJIKI, N., HASHIMOTO, H., YAMAGUCHI, K. & ENDOH, H. (1985). *Jpn J. Appl. Phys.* **24**, L41-44.
- COENE, W., VAN DYCK, D., VAN TENDELOO, G. & VAN LANDUYT, J. (1985). *Philos. Mag. A*, **52**, 127-143.
- COWLEY, J. M. (1981). *Diffraction Physics*, 2nd ed. Amsterdam: North Holland.
- COWLEY, J. M. & FIELDS, P. M. (1979). *Acta Cryst.* **A35**, 28-37.
- COWLEY, J. M. & MOODIE, A. F. (1957). *Acta Cryst.* **10**, 609-619.
- COWLEY, J. M. & SMITH, D. J. (1987). *Acta Cryst.* **A43**, 737-751.
- DOYLE, P. A. (1969). *Acta Cryst.* **A25**, 569-577.
- DOYLE, P. A. (1970). *Acta Cryst.* **A26**, 133-139.
- DOYLE, P. A. (1971). *Acta Cryst.* **A27**, 109-116.
- KIRKLAND, E. J., LOANE, R. F. & SILCOX, J. (1987). *Ultramicroscopy*, **23**, 77-96.
- KOHL, H. & ROSE, H. (1985). *Adv. Electron. Electron Phys.* **65**, 173-227.
- MARKS, L. D. (1986). *Ultramicroscopy*, **18**, 33-38.
- MISELL, D. L. & ATKINS, A. J. (1973). *J. Phys. A*, **6**, 218-235.
- PIROUZ (1979). *Optik (Stuttgart)*, **54**, 69-74.
- RADI, G. (1970). *Acta Cryst.* **A26**, 41-54.
- STURM, K. (1982). *Adv. Phys.* **31**, 1-64.
- TANAKA, N. & COWLEY, J. M. (1987). *Acta Cryst.* **A43**, 337-346.
- VAN HOVE, L. (1954). *Phys. Rev.* **95**, 249-262.

Acta Cryst. (1988). **A44**, 853-857

Effect of RHEED Resonances on Secondary and Auger Electron Emission of Pt(111) Surfaces

BY H. MARTEN AND G. MEYER-EHMSEN

Fachbereich Physik, Universität Osnabrück, D-4500 Osnabrück, Federal Republic of Germany

(Received 15 December 1987; accepted 25 April 1988)

Abstract

The secondary electron emission, the Auger electron emission and the elastically reflected intensity have been measured simultaneously in a reflection high-energy electron diffraction (RHEED) beam rocking experiment with Pt(111). Secondary and Auger yield depend similarly on incident angle. In particular, both quantities exhibit maxima at primary beam orientations around the resonance maxima of the specular beam intensity. The corresponding primary wave field arising by diffraction in the crystal has been calculated using the dynamical theory of RHEED. It turns out that, in contrast to previous suppositions, at the resonances there is a minimum of primary electron density at the atomic sites. It is shown that the behaviour of both yields is rather a consequence of total primary electron intensity near the surface, which is enhanced at the resonances.

1. Introduction

Owing to inelastic interactions a fast electron beam impinging on a solid can produce different kinds of secondary radiation such as X-rays and Auger elec-

trons (AE) or secondary electrons (SE). These processes are widely used for analysis of small samples, for instance, in the scanning electron microscope.

Owing to diffraction effects of the electron beam in crystals its spatial density distribution is modulated on an atomic scale. The probability of localized inelastic interactions therefore depends on the diffraction conditions fulfilled by the incident beam. On the one hand, this leads to anomalous transmission effects in thin crystals (Honjo & Mihama, 1954; Altenhein & Molière, 1954), which are also well known from X-ray transmission (Borrmann, 1941). On the other hand, an influence of the diffraction condition on the yield of characteristic X-ray emission has been observed for thin metal crystals (Duncumb, 1962; Hall, 1966), as well as for solid samples of ZnS (Miyake, Hayakawa & Miida, 1968). Theoretical interpretations on the basis of the dynamical theory have been given by Hirsch, Howie & Whelan (1962) for the transmission case and by Miyake, Hayakawa & Miida (1968) for the case of reflection. Similarly, the effect of Bragg diffraction of the incident beam on Auger yields of solid samples has been observed (e.g. Morin, 1985).