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Image formation in the electron microscope II. The application of transfer theory to a consideration of inelastic electron scattering

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Abstract. The application of transfer theory to image formation by the inelastic component of the transmitted electron beam is discussed. The analysis includes the effects of chromatic aberration on the final image. In particular, it is shown that the chromatic aberration effect due to the inelastic component may be effectively cancelled by underfocus of the objective lens.

In relation to the formation of image by the inelastically scattered electrons, the coherence and the localization of the inelastically scattered wave is discussed.

1. Introduction

In this paper an attempt is made to formulate the wave theory of image formation for inelastically scattered electrons. The basic approach represents an extension of transfer theory (Lenz 1965), as applied to elastic electron scattering, to a consideration of the inelastic component of the transmitted electron beam. To date the main interest in the inelastic component has been its effect on the elastic component of the scattered electron beam. Following the suggestion of Slater (1937) inelastic electron scattering is included as an absorption effect in the elastic scattering formalism by the use of a complex potential. In this way the effects of the various inelastic processes on the Fourier coefficients of the crystal lattice potential have been calculated. The quantum mechanical derivation of the complex potential is given by Yoshioka (1957) who specifically treated atomic excitations. More recently calculations have been made on the contribution to the imaginary part of the Fourier coefficients by plasmon excitation (eg Radi 1970), phonon excitations (thermal diffuse scattering eg Humphreys and Hirsch 1968). In relation to the image formed by the inelastically scattered electrons, a significant amount of theoretical work exists on the preservation of diffraction contrast by small angle inelastic scattering (Fujimoto and Kainuma 1963, Fukuhara 1963, Howie 1963, 1970, Cundy *et al.* 1969, Humphreys and Whelan 1969). This theoretical work and experimental results on the preservation of image contrast by the inelastic component will be considered in the context of the present method for the calculation of the image intensity (§ 2). § 3 will consider in detail the calculation of the inelastically scattered wave for inclusion into the image intensity integral; of particular relevance will be the consideration of the localization of the inelastic scattering processes. In § 4 the possibility of coherence of the inelastic component within a given energy loss band is included into the analysis of § 2. The effect of chromatic aberration on the image formed by the inelastic component is discussed in § 5. Formally the effects of the spatial and chromatic incoherence of the incident electron beam may be included into the calculation of the inelastic image intensity (§ 6).

The only previous calculations on image formation by the inelastic component, including chromatic aberration have been made within the geometrical approximation

(Haine 1957, Sahashi 1969, Crick and Misell 1971, Nagata and Hama 1971). In these papers, the complete incoherence of the inelastically scattered wave is assumed and diffraction effects at the objective aperture are neglected. However, the conclusions of the work of Crick and Misell (1971) on the chromatic aberration due to the inelastic component are consistent with the present analysis (see § 5).

2. Image formation by the inelastic component. Coherence interval ΔK is zero

The incident electron beam is described by a plane wave $\exp(i\boldsymbol{K}_0 \cdot \boldsymbol{r})$, where $\boldsymbol{K}_0$ is the incident wavevector of modulus $2\pi/\lambda_0$; the incident beam is spatially coherent ($\boldsymbol{K}_0$ is constant) and monochromatic (K_0 is constant). The scattered electron wave is $\psi_0(\boldsymbol{K}, K, \boldsymbol{r}_0)$, where $\boldsymbol{K}$ and K refer to the angular and energy characteristics of the inelastically scattered wave immediately after the object. It is assumed that the scattered electron wave carries information on the electron scattering properties of the object at $\boldsymbol{r}_0$. The potential distribution at $\boldsymbol{r}_0$ may be an averaged effect due to a delocalization of the electrons in the solid (see § 3 for a discussion on this problem). In order to calculate the image intensity, it is necessary to decide on the phase relation between electrons with different $\boldsymbol{K}$ and K. It is most unlikely that the inelastic wave exhibits coherence with the unscattered and elastic waves (eg Howie 1963). If it is assumed that electrons which have been scattered inelastically the same number of times are spatially coherent, then the dependence of ψ_0 on $\boldsymbol{K}$ may be omitted, provided that ψ_0 is separated into terms, each representing a given number of inelastic interactions, that is

$$|\psi_0(K, \boldsymbol{r}_0)|^2 = \sum_{n=1}^{\infty} |\psi_n(K, \boldsymbol{r}_0)|^2 \tag{1}$$

and the image wavefunctions corresponding to different n are superimposed incoherently. A further reason for the incoherent superposition relates to the complete lack of experimental evidence for the coherence of electrons with a given K but with different n. The absence of any phase relationship between electrons which have made a different number of inelastic scattering events is discussed by Howie (1963). The only remaining problem is the coherence of electrons with a given n but with differing K. There is a significant amount of experimental evidence on the preservation of diffraction contrast by inelastic electron scattering in single crystal specimens (Kamiya and Uyeda 1961). Of the more recent experimental work on single crystal specimens, there is evidence for the preservation of diffraction contrast by plasmon scattering (Watanabe and Uyeda 1962, Castaing *et al.* 1966a,b,c, Cundy *et al.* 1969, Colliex and Jouffrey 1970), one electron excitations (Cundy *et al.* 1969), phonon excitations (quasielastic scattering, weak preservation, eg Castaing *et al.* 1966b,c, Cundy *et al.* 1967), x ray absorption processes (Colliex and Jouffrey 1970). The requirement for contrast preservation (in a two-beam dynamical theory) is that the inelastically scattered wave (wavevector $\boldsymbol{q}_n$) should be coherent with the diffracted wave, $\boldsymbol{q}_n + 2\pi\boldsymbol{g}$ ($\boldsymbol{g}$ is the reciprocal lattice vector). However, there is no direct evidence for electrons which have loss energy $E(K)$ being coherent with electrons having lost energy $E + \Delta E(K + \Delta K)$. The fact that diffraction contrast due to the inelastic scattering is weaker than that produced by the elastic scattering may be due to this incoherent superposition of electrons with different K. Also it is not often stated whether the objective lens was refocused for these filtered images (or equivalently the high tension voltage raised); the effect of the chromatic aberration on the inelastic image could

give rise to a loss in resolution (equivalent to a decrease in contrast, see also § 5), particularly for images formed from electrons which have excited core electrons (x ray absorption processes, see eg Colliex and Jouffrey 1970). Contrast preservation in the selected energy images of amorphous specimens and polycrystalline specimens (eg Watanabe and Uyeda 1962, Castaing *et al.* 1965, Crewe 1970, Colliex and Jouffrey 1970) and notably for a carbonaceous specimen (Castaing *et al.* 1965) and for a biological specimen (Stroud *et al.* 1969) does not necessarily represent strong evidence for the phase coherence of the inelastic component. The results for amorphous specimens may be explained on the basis of scattering contrast; because inelastic electron scattering is very peaked in the small-angle region, image contrast is expected to be inferior to that contrast obtained for the elastic component.

On the basis of the above discussion, it will be initially assumed that electrons with different K are incoherent. The modification to this analysis for partial coherence will be given in § 4. The image intensity $j_i(\boldsymbol{r}_i) = |\psi_i(\boldsymbol{r}_i)|^2$ is calculated from

$$j_i(\boldsymbol{r}_i) = \sum_{n=1}^{\infty} \int_0^{K_0} \int\int \psi_n(K, \boldsymbol{r}_0)\psi_n^*(K, \boldsymbol{r}_0')G\left(K, \frac{\boldsymbol{r}_i}{M} - \boldsymbol{r}_0\right)G^*\left(K, \frac{\boldsymbol{r}_i}{M} - \boldsymbol{r}_0'\right) \times \mathrm{d}\boldsymbol{r}_0\, \mathrm{d}\boldsymbol{r}_0'\, \mathrm{d}K \tag{2}$$

where G is dependent only on the electron optical properties of the microscope for spatially and chromatically coherent illumination (see Lenz 1965, Misell 1971, to be referred to as I). The upper limit for the K integral is K_0, representing electrons which have lost no energy.

The equation (2) for $j_i(\boldsymbol{r}_i)$ may be rewritten in terms of the Fourier transforms of ψ_n and G as

$$j_i(\boldsymbol{r}_i) = \sum_{n=1}^{\infty} \int_0^{\infty} \int\int S_n(E, \boldsymbol{\nu})S_n^*(E, \boldsymbol{\nu}')T(E, \boldsymbol{\nu})T^*(E, \boldsymbol{\nu}') \times \exp\left(-2\pi\mathrm{i}(\boldsymbol{\nu}-\boldsymbol{\nu}') \cdot \frac{\boldsymbol{r}_i}{M}\right) \mathrm{d}\boldsymbol{\nu}\, \mathrm{d}\boldsymbol{\nu}'\, \mathrm{d}E \tag{3}$$

where

$$S_n(E, \boldsymbol{\nu}) = \int \psi_n(K, \boldsymbol{r}_0) \exp(2\pi\mathrm{i}\boldsymbol{\nu} \cdot \boldsymbol{r}_0)\, \mathrm{d}\boldsymbol{r}_0$$

$$T(E, \boldsymbol{\nu}) = \int G(K, \boldsymbol{r}_0) \exp(2\pi\mathrm{i}\boldsymbol{\nu} \cdot \boldsymbol{r}_0)\, \mathrm{d}\boldsymbol{r}_0. \tag{4}$$

The integral over K has been replaced by an integral over E; energy loss processes correspond to $E > 0$ and for fast electrons (20–100 keV), the probability of energy gain is considered negligible. The upper limit on the E integral, E_0 has been replaced by an infinite limit.

$T(E, \boldsymbol{\nu})$ is the wave aberration function including the effects of the spherical aberration, chromatic aberration and defocusing of the objective lens (equation (41) of I). $S_n(E, \boldsymbol{\nu})$ represents the scattered wave in the back focal plane of the objective lens for the n times inelastically scattered electron. Equation (3) is transformed from the variable $\boldsymbol{\nu}$ to real angular coordinates $\boldsymbol{\theta}$ (in the back focal plane), where

$$\boldsymbol{\theta} = \frac{2\pi\boldsymbol{\nu}}{K} \tag{5}$$

to give the following equation for $j_i(\boldsymbol{r}_i)$:

$$j_i(\boldsymbol{r}_i) = \sum_{n=1}^{\infty} \int_0^{\infty} \left(\frac{K}{2\pi}\right)^2 \int\int \Phi_n(E, \boldsymbol{\theta}) \Phi_n^*(E, \boldsymbol{\theta}') H(E, \boldsymbol{\theta}) H^*(E, \boldsymbol{\theta}') \times \exp\left(-\frac{\mathrm{i}K}{M}(\boldsymbol{\theta}-\boldsymbol{\theta}') \cdot \boldsymbol{r}_i\right) \mathrm{d}\boldsymbol{\theta}\, \mathrm{d}\boldsymbol{\theta}'\, \mathrm{d}E. \tag{6}$$

$|\Phi_n(E, \boldsymbol{\theta})|^2$ represents the intensity distribution of an electron scattered n times inelastically in the specimen. $H(E, \boldsymbol{\theta})$ is given by (see I)

$$H(E, \boldsymbol{\theta}) = \frac{1}{M} \exp\{-\mathrm{i}K\gamma(E, \boldsymbol{\theta})\} D(\boldsymbol{\theta})$$

with

$$\gamma(E, \boldsymbol{\theta}) = \frac{C_s \theta^4}{4} + \frac{C_c E \theta^2}{2E_0} + \frac{\Delta f \theta^2}{2}. \tag{7}$$

The variation of K with the energy loss is not negligible as was the case in considering the thermal energy distribution. However, the variation of K with E, which is approximately $E/2E_0$, affects only the exponential phase factors in equation (6) and to a good approximation, K may be replaced by K_0. Strictly this approximation is not necessary, since the integration over K can be evaluated numerically but in order to derive analytic results, in relation to the effect of chromatic aberration on the inelastic image (see § 5), this simplification is made.

The replacement in equation (6) of K by K_0 leads to the following equation:

$$j_i(\boldsymbol{r}_i) = \left(\frac{K_0}{2\pi M}\right)^2 \sum_{n=1}^{\infty} \int_0^{\infty} \int\int \Phi_n(E, \boldsymbol{\theta}) \Phi_n^*(E, \boldsymbol{\theta}') \exp\{-\mathrm{i}K_0\gamma(E, \boldsymbol{\theta})\} D(\boldsymbol{\theta}) \times \exp\{\mathrm{i}K_0\gamma(E, \boldsymbol{\theta}')\} D(\boldsymbol{\theta}') \exp\left(-\frac{\mathrm{i}K_0}{M}(\boldsymbol{\theta}-\boldsymbol{\theta}') \cdot \boldsymbol{r}_i\right) \mathrm{d}\boldsymbol{\theta}\, \mathrm{d}\boldsymbol{\theta}'\, \mathrm{d}E. \tag{8}$$

In order to calculate the $\Phi_n(E, \boldsymbol{\theta})$ consideration of the angular-energy distributions and of the localization of inelastic electron scattering must be made.

It is noted that in equation (8) the assumption has been made that electrons with the same E and n are coherent. However for a given resultant energy loss E, the inelastically scattered electron may lose energy by different processes and by different amounts. E is a sum of individual terms $E_1, E_2, E_3, \ldots E_n$. More exactly equation (8) should not only contain a summation over n but a further summation of all possible sets $(E_1, E_2, \ldots E_n) = E$ and the integration over E is omitted.

3. Angular-energy distributions and the localization of inelastic electron scattering

3.1. *Angular-energy distributions*

In this section the problem of calculating the angular-energy distributions for n inelastic events is considered. As stated in § 2, electrons which have been inelastically scattered a different number of times are incoherent and the scattered wave $\Psi_I(E, \boldsymbol{\theta})$ can be calculated by the incoherent superposition of the Φ_n for each E value, that is

$$|\Psi_I(E, \boldsymbol{\theta})|^2 = \sum_{n=1}^{\infty} |\Phi_n(E, \boldsymbol{\theta})|^2 = \sum_{n=1}^{\infty} P_n |\phi_n(E, \boldsymbol{\theta})|^2. \tag{9}$$

P_n is the Poisson distribution $(t/\Lambda_I)^n \exp(-t/\Lambda_I)/n!$ for independent scattering processes (eg Howie 1963); t is the specimen thickness and Λ_I is the mean free path for inelastic electron scattering. If the inelastic processes leading to different n are incoherent, then the $\Phi_n(E, \boldsymbol{\theta})$ are given by

$$\Phi_n(E, \boldsymbol{\theta})\Phi_n^*(E, \boldsymbol{\theta}) = P_n D_n(E, \boldsymbol{\theta}) \simeq P_n Q_n(\boldsymbol{\theta}) f_n(E) \tag{10}$$

where $Q_n(\boldsymbol{\theta})$ is the angular distribution after n inelastic events with energy distribution $f_n(E)$ calculated by a simple folding procedure (see eg Crick and Misell 1971). In equation (10) the dispersion relationship, that is, the dependence of E on θ, has been neglected; this approximation is justified further below. The inelastic wave $\Phi_n(E, \boldsymbol{\theta})$ can be written as

$$\Phi_n(E, \boldsymbol{\theta}) = P_n^{1/2}\{D_n(E, \boldsymbol{\theta})\}^{1/2} \exp\{i\mu_n(E, \boldsymbol{\theta})\}. \tag{11}$$

The phase term μ_n is assumed to be independent of θ, consistent with the assumption of coherence for a given E over all θ (provided θ is small, see Howie 1963, Cundy *et al.* 1969). If μ_n is a function of E only and electrons with different E are incoherent (μ_n for different E values are not correlated), then the $\exp(i\mu_n)$ factors cancel for a given E, since the phase factor occurs as a product $\Phi_n(E, \boldsymbol{\theta})\Phi_n^*(E, \boldsymbol{\theta}')$ in equation (8).

It is strictly unnecessary to assume that $\Phi_n(E, \boldsymbol{\theta})$ is a separable function of E and $\boldsymbol{\theta}$ as in equation (10); the dielectric formulation for inelastic scattering in the volume of the specimen (neglecting surface excitations and retardation effects, see review by Daniels *et al.* 1970) gives the following expression for $|\phi_1(E, \boldsymbol{\theta})|^2$ (Hubbard 1955, Raether 1965)

$$\phi_1(E, \boldsymbol{\theta})\phi_1^*(E, \boldsymbol{\theta}) = -\frac{1}{2\pi^2 a_H E_0 \sigma_I} \frac{1}{\theta^2 + \theta_E^2} \operatorname{Im}\left(\frac{1}{\epsilon(q_1, E)}\right). \tag{12}$$

$\sigma_I = \Lambda_I^{-1}$, $\theta_E = E/2E_0$ and $\epsilon(q_1, E)$ is the wavevector, frequency dependent dielectric constant; $q_1 = (2\pi/\lambda_0)(\theta^2 + \theta_E^2)^{1/2}$ is $|\boldsymbol{q}_1|$. The term $\phi_2(E, \boldsymbol{\theta})$ may then be calculated by a convolution of $|\phi_1|^2$ with $|\phi_1|^2$ over E and $\boldsymbol{\theta}$, that is

$$\phi_2(E, \boldsymbol{\theta})\phi_2^*(E, \boldsymbol{\theta}) = C^2 \int_0^E \int_{\boldsymbol{\theta}'} \frac{1}{\boldsymbol{\theta}'^2 + \theta_{E'}^2} \frac{1}{(\boldsymbol{\theta} - \boldsymbol{\theta}')^2 + \theta_{E-E'}^2} \times \operatorname{Im}\left(\frac{1}{\epsilon(\boldsymbol{\theta}', E')}\right) \operatorname{Im}\left(\frac{1}{\epsilon(\boldsymbol{\theta} - \boldsymbol{\theta}', E - E')}\right) d\boldsymbol{\theta}'\, dE' \tag{13}$$

neglecting elastic electron scattering which may be included by a further convolution (eg Misell 1970); $C = -(2\pi^2 a_H E_0 \sigma_I)^{-1}$. Equation (13) represents a double integration since the $\boldsymbol{\theta}$ integration can be simplified by an integration over the azimuthal angle of scattering for a cylindrically symmetric angular term (see eg Daniels *et al.* 1970). A knowledge of $\operatorname{Im}\{1/\epsilon(\boldsymbol{\theta}, E)\}$ is not readily available and two further simplifications to equation (13) are usually made (Daniels *et al.* 1970), namely $\epsilon(\boldsymbol{\theta}, E) \simeq \epsilon(0, E)$ over the $\boldsymbol{\theta}$ integral and θ_E is constant (or negligible) over the E integral. These two approximations enable the $\boldsymbol{\theta}$ and E integrations to be separated and this then is the equivalent of the formulation of $\Phi_n(E, \boldsymbol{\theta})$ given above (equations (10) and (11)). These approximations appear to be totally justified by the results of Daniels *et al.* (1970) on the calculation of $\epsilon(0, E)$ from electron energy loss data.

The expression for $\Phi_n(E, \boldsymbol{\theta})\Phi_n^*(E, \boldsymbol{\theta}')$ in equation (8) is then

$$\Phi_n(E, \boldsymbol{\theta})\Phi_n^*(E, \boldsymbol{\theta}') = P_n\phi_n(\boldsymbol{\theta})\phi_n^*(\boldsymbol{\theta}')f_n(E) \tag{14}$$

where ϕ is a function of $\boldsymbol{\theta}$ only, namely $Q_n(\boldsymbol{\theta})^{1/2}$.

Hence the $\Phi_n(E, \boldsymbol{\theta})$ are calculated on the basis of the following assumptions, which are not inconsistent with previous theoretical and experimental work: (i) inelastically scattered electrons are incoherent with the main beam and the elastic scattering. This justifies the separate treatment of the inelastic image. (ii) electrons with differing n are incoherent even for the same E. (iii) Φ_n may be written as a separable function of $\boldsymbol{\theta}$ and E. (iv) inelastic electrons with the same E and n are coherent over all $\boldsymbol{\theta}$. This is strictly valid only for small q (Howie 1963, Metherell 1967, Cundy *et al.* 1969). A further assumption, which is unlikely to be valid, is made to simplify the analysis: (v) electrons inelastically scattered with same resultant E and a given n are coherent. An electron incident on the specimen can excite various processes leading to the same E, where the energy distribution $f_n(E)$ is calculated from

$$f_n(E) = \int_0^E f_{n-1}(E')f_1(E-E')\,\mathrm{d}E'.$$

For $n > 1$, E can result from the set $(E_1, E_2,\ldots E_n)$. If electrons only with the same set are coherent then

$$\Phi_n(E_1, E_2,\ldots E_n, \boldsymbol{\theta})\Phi_n(E_1, E_2,\ldots E_n, \boldsymbol{\theta}') = P_n\phi_n(\boldsymbol{\theta})\phi_n(\boldsymbol{\theta}')f_1(E_1)\ldots f_1(E_n+E_{n-1}). \tag{15}$$

The coherence of inelastic electrons over the complete energy distribution has not been considered in the literature. The experimental work using energy selecting electron microscopes has concentrated on the formation of images within a narrow energy loss band ± 1 eV (eg Watanabe and Uyeda 1962, Castaing *et al.* 1966a,b,c) and in energy analysing electron microscopes, the effective energy loss interval is about 2 eV (eg Cundy *et al.* 1967, 1969). It would be of interest to examine these inelastic images using either a larger energy gate or decreasing the resolution of the electron spectrometer. For the crystalline specimens examined, the energy loss profiles are of a discrete character, for example, Al (Watanabe and Uyeda 1962), Si (Castaing *et al.* 1966a), electrons with different n are quite well separated and it is difficult to make an assessment of the coherence of electrons with the same E but different n. Of particular interest is the coherence of the inelastic electron scattering in organic and biological materials, where the energy halfwidth of $f_1(E)$ is 15–20 eV (Marton *et al.* 1955, Swanson and Powell 1963, Johnson and Rymer 1967, Jäger 1969).

The angular distributions $\phi_n(\boldsymbol{\theta})$ include the modifications on the inelastic scattering due to elastic electron scattering. In an amorphous specimen these distributions may be calculated by a convolution procedure involving intensity distributions (Crick and Misell 1971) or for a crystalline specimen an n beam calculation is used (eg Metherell 1967, Doyle 1971). However, the localization of inelastic electron scattering must be considered before information is available on $\Phi_n(E, \boldsymbol{\theta})$ in a complete form.

3.2. *Localization of inelastic scattering*

The parameter which describes the screening of the electrons in the solid from the incident electron is $1/\epsilon(q, E)$ (Hubbard 1955, Nozières and Pines 1959, Raether 1965). The screening is a measure of the localization of the inelastic scattering.

One electron excitations including core excitations, and thermal diffuse scattering (which contributes only a small amount to the scattering within the normal objective aperture) are localized phenomenon (eg Cundy *et al.* 1969, Howie 1970). The plasmon excitation in metallic specimens is a delocalized phenomenon over a radius of about 20 Å (eg Cundy 1968, Cundy *et al.* 1968). For the plasmon excitation in organic and biological materials, the localization can be considered from the behaviour of $1/\epsilon(0, E)$; a small value for this reciprocal implies a localized excitation. In particular, the behaviour of $\mathrm{Im}\{1/\epsilon(0, E)\}$ and $\mathrm{Re}\{1/\epsilon(0, E)\}$ for metallic (nontransition metals) and amorphous specimens is considered in the region of the plasmon energy E_p. $\mathrm{Re}\{1/\epsilon(0, E)\}$ is nearly zero in the region $E \simeq E_\mathrm{p}$ (Daniels *et al.* 1970) and $\mathrm{Im}\{1/\epsilon(0, E)\}$ is a direct measure of the energy loss profile, obtained under single scattering conditions at $\theta \simeq 0$. The energy loss profile for the nontransition metals can be described by a Drude free electron model (Daniels *et al.* 1970) and a similar profile accurately describes the 20–25 eV plasmon loss in amorphous carbon (Burge and Misell 1968), organic materials (Swanson and Powell 1963, Jäger 1969, Misell and Crick 1969) and materials of biological significance (Marton *et al.* 1955, Johnson and Rymer 1967). Hence

$$-\mathrm{Im}\left(\frac{1}{\epsilon(0, E)}\right) = \frac{E_\mathrm{p}^2 \tau E}{(E_\mathrm{p}^2 - E^2)^2 \tau^2 + E^2}. \tag{16}$$

The parameter τ is related to the energy halfwidth of the profile by $\tau \simeq 1/E_{1/2}$. For $E \simeq E_\mathrm{p}$, $-\mathrm{Im}(1/\epsilon) \simeq E_\mathrm{p}/E_{1/2}$, that is, the screening is related directly to the halfwidth of the energy loss profile for plasmon excitation. In the case of Al, $E_\mathrm{p} = 15$ eV and $E_{1/2} = 0{\cdot}8$ eV (Raether 1965) and $-\mathrm{Im}\{1/\epsilon(0, E_\mathrm{p})\} = 18$; for amorphous carbon $E_\mathrm{p} \simeq 20$–25 eV and $E_{1/2} = 15$–20 eV and $-\mathrm{Im}\{1/\epsilon(0, E_\mathrm{p})\} \simeq 1{\cdot}5$. Both results are consistent with optical data. This relation between the screening and the halfwidth $E_{1/2}$ is not just a result of using the Lorentzian above, but a similar result can be derived for gaussian and maxwellian profiles, provided that the sum rule (Nozières and Pines 1959)

$$\int_0^\infty \mathrm{Im}\left(\frac{1}{\epsilon(0, E)}\right) E \, \mathrm{d}E = -\frac{\pi E_\mathrm{p}^2}{2}$$

is satisfied

The implication of these calculations is that the plasmon excitation in organic and biological materials is a localized phenomenon and $\Phi_n(E, \boldsymbol{\theta})$ is characteristic of the specimen structure on a short range scale.

4. Image formation by the inelastic component. Coherence interval ΔK is finite

If the inelastically scattered electrons with a given n are coherent within an energy band between E and $E+\Delta E$ (K and $K+\Delta K$), then the wavefunctions $\psi_n(K, \boldsymbol{r}_0)$ are superimposed coherently within this interval. Electrons within different energy bands are superimposed incoherently. If it is assumed that the inelastic wave in the back focal plane of the objective lens $\Phi_n(E, \boldsymbol{\theta})$ is a separable function of $\boldsymbol{\theta}$ and E (see § 3.1), then the equation for the image intensity is

$$j_\mathrm{i}(\boldsymbol{r}_\mathrm{i}) = \left(\frac{K_0}{2\pi M}\right)^2 \sum_{n=1}^{\infty} P_n \sum_{p\Delta E=0}^{p\Delta E = E_0} f_n'(E) \int\int \phi_n(\boldsymbol{\theta}) \phi_n^*(\boldsymbol{\theta}') D(\boldsymbol{\theta}) D(\boldsymbol{\theta}')$$
$$\times \exp[-\mathrm{i}K_0\{\gamma(E, \boldsymbol{\theta}) - \gamma(E, \boldsymbol{\theta}')\}] \exp\left(-\frac{\mathrm{i}K_0}{M}(\boldsymbol{\theta} - \boldsymbol{\theta}') \cdot \boldsymbol{r}_\mathrm{i}\right) \mathrm{d}\boldsymbol{\theta} \, \mathrm{d}\boldsymbol{\theta}' \tag{17}$$

where

$$f_n'(E) = \int_{p\Delta E}^{(p+1)\Delta E} f_n(E)\,\mathrm{d}E.$$

The constant phase factor in ϕ_n for a given n and within the interval E, $E+\Delta E$ is irrelevant because electrons outside a given ΔE are superimposed incoherently.

If the scattering by the specimen exhibits cylindrical symmetry then equation (17) simplifies (for normal bright field electron microscopy) to give

$$j_i(r_i) = \left(\frac{K_0}{M}\right)^2 \sum_{n=1}^{\infty} P_n \sum_{p\Delta E=0}^{p\Delta E=E_0} f_n'(E) \int_0^{\alpha}\int_0^{\alpha} \phi_n(\theta)\phi_n(\theta') \times \exp[-\mathrm{i}K_0\{\gamma(E,\theta)-\gamma(E,\theta')\}]\mathrm{J}_0\left(\frac{K_0 r_i \theta}{M}\right)\mathrm{J}_0\left(\frac{K_0 r_i \theta'}{M}\right)\theta\,\mathrm{d}\theta\,\theta'\,\mathrm{d}\theta' \qquad (18)$$

where $\phi_n(\theta)\phi_n{}^*(\theta') \equiv \phi_n(\theta)\phi_n(\theta')$ for a constant phase factor independent of θ and $\phi_n(\theta)$ is taken as a real quantity.

5. The effect of chromatic aberration on the inelastic image

In order to investigate the effect of the energy loss distribution on the image formed by the inelastic component, the integration over E in equation (8) is considered. Using equation (14) equation (8) is rewritten as

$$j_i(\boldsymbol{r}_i) = \left(\frac{K_0}{2\pi M}\right)^2 \sum_{n=1}^{\infty} P_n \int\int L_n(\boldsymbol{\theta},\boldsymbol{\theta}')\phi_n(\boldsymbol{\theta})\phi_n{}^*(\boldsymbol{\theta}') \exp[-\mathrm{i}K_0\{\chi(\boldsymbol{\theta})-\chi(\boldsymbol{\theta}')\}] \times D(\boldsymbol{\theta})D(\boldsymbol{\theta}')\exp\left(-\frac{\mathrm{i}K_0}{M}(\boldsymbol{\theta}-\boldsymbol{\theta}')\cdot\boldsymbol{r}_i\right)\mathrm{d}\boldsymbol{\theta}\,\mathrm{d}\boldsymbol{\theta}' \qquad (19)$$

where

$$L_n(\boldsymbol{\theta},\boldsymbol{\theta}') = \int_0^{\infty} \exp\left(-\frac{\mathrm{i}K_0 C_c E}{2E_0}(\theta^2-\theta'^2)\right) f_n(E)\,\mathrm{d}E. \qquad (20)$$

$\mathrm{i}K_0\chi(\boldsymbol{\theta})$ is the phase shift term including only the spherical aberration and defocusing terms (see equation (7)).

The evaluation of $L_n(\boldsymbol{\theta},\boldsymbol{\theta}')$ requires a model for the distributions $f_n(E)$. The $f_n(E)$ are calculated by a repetitive folding of $f_1(E)$; $f_1(E)$ is well approximated by a Lorentzian (equation (16)) for carbon, organic and biological materials (curve fitting based on the results of Marton *et al.* 1955, Swanson and Powell 1963, Johnson and Rymer 1967, Jäger 1967; method given by Burge and Misell 1968) and for the nontransition metals (eg Raether 1965). However, with the lorentzian lineshape for $f_1(E)$, it is not possible to evaluate the $f_n(E)$ analytically and in general the integration (20) can only be evaluated numerically except for $n = 1$. As an approximation to $f_1(E)$, the symmetrical gaussian curve $(b/\pi)^{1/2}\exp\{-b(E-E_p)^2\}$ is used in equation (20), that is

$$L_n(\boldsymbol{\theta},\boldsymbol{\theta}') = \left(\frac{b}{n\pi}\right)^{1/2} \exp\left(-\frac{\mathrm{i}K_0 C_c n E_p}{2E_0}(\theta^2-\theta'^2)\right)\int_{-nE_p}^{\infty} \exp\left(-\frac{\mathrm{i}K_0 C_c E}{2E_0}(\theta^2-\theta'^2)\right) \times \exp\left(-\frac{bE^2}{n}\right)\mathrm{d}E. \qquad (21)$$

Except for the lower limit $-nE_p$ on the E integral, this integral is the Fourier transform of a Gaussian; for $E < -nE_p$, the exponential factor is negligibly small and the lower limit may be replaced by a negative infinite limit, that is

$$L_n(\boldsymbol{\theta}, \boldsymbol{\theta}') = \exp\left(-\frac{iK_0C_cnE_p}{2E_0}(\theta^2-\theta'^2)\right)\exp\left\{-\left(\frac{K_0C_c}{2E_0}\right)^2\frac{n(\theta^2-\theta'^2)^2}{4b}\right\}. \quad (22)$$

The optimum conditions for image formation by the inelastic component, neglecting for the present the elastic contribution to the image intensity, can be considered on the basis of equation (22). The θ dependence of the oscillatory term is the same as that of the defocusing term in $\chi(\boldsymbol{\theta})$, that is, $-iK_0\Delta f(\theta^2-\theta'^2)/2$. With $n = 1$, corresponding to a main peak at E_p in the energy loss distribution and assuming that the multiple peaks nE_p are not significant, the maximum value of the oscillatory exponent in equation (22) is calculated to be $-91{\cdot}5$ rad for $E_0 = 20$ keV ($E_p = 25$ eV, $\alpha = 0{\cdot}01$ rad, $C_c = 0{\cdot}2$ cm) and $-10{\cdot}6$ rad for $E_0 = 100$ keV ($\alpha = 0{\cdot}005$ rad). These phase shifts are an order of magnitude greater than the corresponding spherical aberration phase shifts ($-3{\cdot}66$ rad and $-0{\cdot}53$ rad respectively, $C_s = 0{\cdot}2$ cm). These large phase shifts can be exactly cancelled by underfocus of the objective lens; for $E_0 = 20$ keV, $\Delta f = -2{\cdot}5$ μm and for $E_0 = 100$ keV, $\Delta f = -0{\cdot}5$ μm. This partial cancellation of the chromatic aberration defect is expected because the gaussian image plane has been displaced by underfocusing to focus electrons with energy E_0-E_p. For a specific n, the corresponding defocus necessary to cancel that particular oscillatory term is $\Delta f = -C_cnE_p/E_0$ and this corresponds to focusing of electrons with energy E_0-nE_p. Thus depending on the specimen thickness, an optimum defocusing can be calculated in order to minimize the effect of chromatic aberration on the inelastic image. This underfocusing will produce a large chromatic aberration effect on the elastic image; the equivalent phase shift is $+iK_0C_cnE_p(\theta^2-\theta'^2)/2E_0$ introduced into the elastic wave (see § 4 of I). This point is discussed further below, where a comparison of the inelastic intensity $|\phi_n(\boldsymbol{\theta})|^2$ and the elastic intensity $|\Psi_E(\boldsymbol{\theta})|^2$ is made.

The cancellation of the phase term in equation (22) leaves as the main chromatic aberration effect the exponential decay term

$$L_n(\boldsymbol{\theta}, \boldsymbol{\theta}') \sim \exp\left\{-\left(\frac{K_0C_c}{2E_0}\right)^2\frac{n(\theta^2-\theta'^2)^2}{4b}\right\}. \quad (23)$$

If $n = 1$, then the θ dependence of the exponential factor with $b = 0{\cdot}007$ eV^{-2} (corresponding to the distribution $f_1(E)$ for carbon) is

	$\theta = 0{\cdot}002$ rad	$\theta = 0{\cdot}003$ rad	$\theta = 0{\cdot}004$ rad
$E_0 = 20$ keV	0·47	0·02	0·00
$E_0 = 100$ eV	0·84	0·28	0·07.

Thus the $L_1(\boldsymbol{\theta})$ term is negligible for $\theta \simeq \alpha$, unless $\theta \simeq \theta'$. For α in the range 0·0025–0·02 rad, the contribution from $L_1(\boldsymbol{\theta}, \boldsymbol{\theta}')$ to the $\boldsymbol{\theta}$ and $\boldsymbol{\theta}'$ integrands in equation (19) will only be significant for small arguments and a limited number of angular values where $\theta \simeq \theta'$. The conclusion is that for a broad $f_1(E)$ the chromatic aberration effect is not very dependent on the aperture size. This is consistent with the recent calculations of Misell and Crick (1971) on the effect of chromatic aberration on the image of a 'biological structure'; using the incoherent approximation, it was found

that the convolution effect of the chromatic aberration on the inelastic image was a slow varying function of α. It is noted that the above conclusions were arrived at by using a gaussian model for the energy loss distributions and in general it is not possible to cancel completely the exponential phase term by a single value for the underfocus. In the case where $f_1(E)$ is discrete (eg Al, $b = 3{\cdot}0$ eV^{-2}), the exponential factor decreases less rapidly with θ increasing, but there is still a significant attenuation effect. The behaviour of $L_n(\boldsymbol{\theta}, \boldsymbol{\theta}')$ explains why for even a uniform illumination of the objective aperture, that is, $|\phi_n(\boldsymbol{\theta})|^2$ is constant for $0 \leqslant \theta \leqslant \alpha$, that the chromatic defect is overestimated by the equation

$$r_c = \frac{C_c g(E)\alpha}{E_0}. \tag{24}$$

$g(E)$ is a measure of the energy loss, for example, the most probable loss (Cosslett 1956), the energy halfwidth of the loss distribution (Cosslett 1969, where a factor of $\frac{1}{2}$ is introduced into equation (24) for focusing on the most probable energy loss), and for materials exhibiting multiple plasmon losses, $g(E) = (t/\Lambda_I)E_p$ or $2{\cdot}35\, E_p(t/\Lambda_I)^{1/2}$ (Hirsch and Humphreys 1968); experimental estimates of $g(E)$ have

Table 1. The ratio of the inelastic to elastic electron scattering, $h_i(\alpha)/h_e(\alpha)$, for a specimen of thickness t Å and objective aperture semi-angle α rad†

Carbon (amorphous)	$E_0 = 20$ keV	$\Lambda_E = 323$ Å	$\Lambda_I = 223$ Å (171 Å)
t (Å)	$\alpha = 0{\cdot}01$ rad	$\alpha = 0{\cdot}02$ rad	$\alpha = 0{\cdot}04$ rad
50	18·0 (24·1)	6·5 (8·8)	2·9 (3·9)
100	19·2 (26·3)	7·2 (10·0)	3·4 (4·7)
200	21·9 (31·9)	8·9 (13·2)	4·6 (6·9)
300	25·4 (40·0)	11·2 (18·1)	6·3 (10·4)
400	30·3 (52·1)	14·4 (25·8)	8·8 (16·1)
500	36·9 (70·8)	19·0 (38·0)	12·5 (25·6)
Carbon (amorphous)	$E_0 = 100$ keV	$\Lambda_E = 1314$ Å	$\Lambda_I = 775$ Å (583 Å)
t (Å)	$\alpha = 0{\cdot}005$ rad	$\alpha = 0{\cdot}01$ rad	$\alpha = 0{\cdot}02$ rad
50	17·0 (22·7)	5·9 (8·0)	2·7 (3·7)
100	17·3 (23·4)	6·1 (8·3)	2·9 (3·9)
200	18·1 (24·8)	6·5 (9·0)	3·1 (4·3)
300	18·9 (26·4)	6·9 (9·8)	3·4 (4·8)
400	19·7 (28·1)	7·4 (10·7)	3·7 (5·4)
500	20·6 (30·0)	7·9 (11·7)	4·1 (6·1)
Aluminium	$E_0 = 20$ keV	$\Lambda_E = 163$ Å	$\Lambda_I = 263$ Å (90 Å)
t (Å)	$\alpha = 0{\cdot}01$ rad	$\alpha = 0{\cdot}02$ rad	$\alpha = 0{\cdot}04$ rad
50	4·5 (15·0)	1·9 (6·4)	1·2 (4·1)
100	4·8 (19·2)	2·1 (9·0)	1·4 (6·1)
200	5·7 (33·7)	2·8 (18·5)	2·1 (14·1)
300	6·8 (66·4)	3·8 (41·9)	3·0 (35·1)
400	8·2 (145)	5·1 (103)	4·3 (92·8)
500	10·2 (346)	7·0 (270)	6·2 (257)

Table 1 (cont.)

Aluminium	E_0 = 100 keV	Λ_E = 659 Å	Λ_I = 985 Å (308 Å)
t (Å)	α = 0·005 rad	α = 0·01 rad	α = 0·02 rad
50	3·7 (12·4)	1·6 (5·3)	1·0 (3·5)
100	3·8 (13·3)	1·6 (5·8)	1·1 (3·9)
200	4·0 (15·5)	1·8 (7·1)	1·2 (4·8)
300	4·2 (18·2)	1·9 (8·6)	1·3 (6·1)
400	4·4 (21·5)	2·1 (10·6)	1·5 (7·7)
500	4·7 (25·7)	2·3 (13·2)	1·6 (9·8)
Gold	E_0 = 20 keV	Λ_E = 15 Å	Λ_I = 182 Å (68 Å)
t (Å)	α = 0·01 rad	α = 0·02 rad	α = 0·04 rad
50	1·2 (3·9)	0·59 (2·0)	0·39 (1·3)
100	1·1 (4·4)	0·76 (3·4)	0·71 (3·2)
150	1·9 (10·0)	1·3 (8·0)	1·2 (7·8)
200	3·1 (21·3)	2·1 (17·7)	2·0 (17·3)
250	4·4 (42·9)	3·1 (37·8)	2·9 (37·2)
Gold	E_0 = 100 keV	Λ_E = 59 Å	Λ_I = 629 Å (232 Å)
t (Å)	α = 0·005 rad	α = 0·01 rad	α = 0·02 rad
50	1·5 (4·4)	0·57 (1·6)	0·27 (0·79)
100	1·2 (3·8)	0·55 (1·6)	0·33 (1·0)
150	0·82 (2·7)	0·49 (1·6)	0·41 (1·4)
200	0·58 (2·0)	0·44 (1·6)	0·56 (2·0)
250	0·50 (1·8)	0·45 (1·7)	0·78 (3·1)

† The scattering model for the inelastic electron scattering is a composite model; figures in parentheses correspond to a free atom calculation for $h_i(\alpha)$. The mean free path value for inelastic electron scattering is Λ_I and that for elastic scattering is Λ_E.

been given by Verdier (1968). It is suggested that the maximum values for θ, α, in equation (24) should be replaced by the value of θ for which the exponential factor decreases to (say) 0·5 of its value at $\theta = 0$; for carbon $\theta = 0{\cdot}002$ rad at 20 keV and $\theta = 0{\cdot}0028$ rad at 100 keV. For nonuniform illumination of the objective aperture, Cundy *et al.* (1968) replace α in equation by θ_m, the mean angle of scattering; again this leads to an overestimate of r_c.

Assuming that the inelastic component carries useful structural information on the specimen, then the decision on whether to defocus the objective lens by the large amounts given above will depend on the magnitude of the elastic and inelastic contributions transmitted by the objective aperture, that is

$$\int_0^{2\pi}\int_0^{\alpha} |\Psi_E(\boldsymbol{\theta})|^2 \, d\boldsymbol{\theta} = h_e(\alpha)$$

$$\sum_{n=1}^{\infty} P_n \int_0^{2\pi}\int_0^{\alpha} |\phi_n(\boldsymbol{\theta})|^2 \, d\boldsymbol{\theta} = h_i(\alpha).$$

$h_e(\alpha)$ and $h_i(\alpha)$ are respectively the fractions of the incident electron beam that are scattered within the objective aperture. For carbon, aluminium and gold $|\Psi_E(\boldsymbol{\theta})|^2$ has been calculated from the free atom formulation; for the solid state, particularly the

crystalline state, this model can only give a reasonable estimate for $h_e(\alpha)$. The term $|\phi_n(\boldsymbol{\theta})|^2$ is calculated using two models, namely, the free atom theory or more realistically a composite model, Bohm–Pines/Ferrell/free atom expression (see Misell and Crick 1969 for details and references). In table 1 the values for h_i/h_e for several film thicknesses t Å of carbon, aluminium and gold and for several objective aperture sizes are given. The main figures correspond to h_i/h_e for the composite inelastic scattering model; the figures in parentheses correspond to the free atom calculations.

6. The effect of the spatial and chromatic incoherence of the electron source on the inelastic image

The effect of the spatial incoherence and the chromatic incoherence of the incident electron beam can be included into the calculation of the inelastic image. In order to simplify the analysis, spatial and chromatic incoherence will be treated separately within the framework of the transfer theory.

6.1. *Spatial incoherence*

The incident electron beam in monochromatic (K_0 = constant) and the spatial coherence is represented by the distribution of $\boldsymbol{K}_0$, $F(\boldsymbol{K}_0)$. The inelastic wave for a particular n can be written as

$$\psi_n(\boldsymbol{K}_0, K, \boldsymbol{r}_0) = \psi_n(K, \boldsymbol{r}_0)\exp(\mathrm{i}\boldsymbol{K}_0 \cdot \boldsymbol{r}_0)$$

since the electron scattering properties of the specimen are not dependent on the incident wavevector $\boldsymbol{K}_0$. The function $F(\boldsymbol{K}_0)$ represents the angular distribution of the incident electron beam (see Lenz 1965, § 3 of I). The wavefunctions ψ_n with different $\boldsymbol{K}_0$ and K are superimposed incoherently (weighted with the distribution function $F(\boldsymbol{K}_0)$); equation (2) for the image intensity is then

$$j_i(\boldsymbol{r}_i) = \sum_{n=1}^{\infty} \int_0^{K_0} \int\int\int \psi_n(K, \boldsymbol{r}_0)\psi_n^*(K, \boldsymbol{r}_0')G\left(K, \frac{\boldsymbol{r}_i}{M} - \boldsymbol{r}_0\right)G^*\left(K, \frac{\boldsymbol{r}_i}{M} - \boldsymbol{r}_0'\right) \times \exp\{\mathrm{i}\boldsymbol{K}_0 \cdot (\boldsymbol{r}_0 - \boldsymbol{r}_0')\}F(\boldsymbol{K}_0)\,\mathrm{d}\boldsymbol{r}_0\,\mathrm{d}\boldsymbol{r}_0'\,\mathrm{d}\boldsymbol{K}_0\,\mathrm{d}K. \tag{25}$$

Using the definition equation (4) for the Fourier transforms of ψ_n and G, the equation for $j_i(\boldsymbol{r}_i)$ becomes

$$j_i(\boldsymbol{r}_i) = \sum_{n=1}^{\infty} \int_0^{\infty} \int\int\int S_n\left(E, \frac{\boldsymbol{K}_0}{2\pi} + \boldsymbol{\nu}\right)S_n^*\left(E, \frac{\boldsymbol{K}_0}{2\pi} + \boldsymbol{\nu}'\right) T(E, \boldsymbol{\nu})T^*(E, \boldsymbol{\nu}') \times \exp\left(-2\pi\mathrm{i}(\boldsymbol{\nu} - \boldsymbol{\nu}') \cdot \frac{\boldsymbol{r}_i}{M}\right)F(\boldsymbol{K}_0)\,\mathrm{d}\boldsymbol{\nu}\,\mathrm{d}\boldsymbol{\nu}'\,\mathrm{d}\boldsymbol{K}_0\,\mathrm{d}E. \tag{26}$$

Equation (26) is transformed to real angular coordinates in the back focal plane with

$$\boldsymbol{\theta} = \frac{2\pi\boldsymbol{\nu}}{K} \qquad \text{and} \qquad \boldsymbol{\theta}_c = \frac{\boldsymbol{K}_0}{K} \tag{27}$$

and $\boldsymbol{\theta}_c$ defines the angular coordinate of the incident electron beam. With this transformation, equation (26) becomes

$$j_i(\boldsymbol{r}_i) = \sum_{n=1}^{\infty} \int_0^{\infty} \left(\frac{K}{2\pi}\right)^2 \int\int\int \Phi_n(E, \boldsymbol{\theta} + \boldsymbol{\theta}_c)\Phi_n^*(E, \boldsymbol{\theta}' + \boldsymbol{\theta}_c)H(E, \boldsymbol{\theta})H^*(E, \boldsymbol{\theta}') \times \exp\left(-\frac{\mathrm{i}K}{M}(\boldsymbol{\theta} - \boldsymbol{\theta}') \cdot \boldsymbol{r}_i\right)I_0(\boldsymbol{\theta}_c)\,\mathrm{d}\boldsymbol{\theta}\,\mathrm{d}\boldsymbol{\theta}'\,\mathrm{d}\boldsymbol{\theta}_c\,\mathrm{d}E \tag{28}$$

or with $K \simeq K_0$

$$j_i(\boldsymbol{r}_i) = \left(\frac{K_0}{2\pi M}\right)^2 \sum_{n=1}^{\infty} P_n \int_0^{\infty} \int\int\int \phi_n(\boldsymbol{\theta}+\boldsymbol{\theta}_c)\phi_n^*(\boldsymbol{\theta}'+\boldsymbol{\theta}_c) I_0(\boldsymbol{\theta}_c) D(\boldsymbol{\theta}) D(\boldsymbol{\theta}') \times \exp[-\mathrm{i}K_0\{\gamma(E,\boldsymbol{\theta})-\gamma(E,\boldsymbol{\theta}')\}] \exp\left(-\frac{\mathrm{i}K_0}{M}(\boldsymbol{\theta}-\boldsymbol{\theta}')\,.\,\boldsymbol{r}_i\right) f_n(E)\,\mathrm{d}\boldsymbol{\theta}\,\mathrm{d}\boldsymbol{\theta}'\,\mathrm{d}\boldsymbol{\theta}_c\,\mathrm{d}E \tag{29}$$

where $\Phi_n(E, \boldsymbol{\theta}+\boldsymbol{\theta}_c)\Phi_n^*(E, \boldsymbol{\theta}'+\boldsymbol{\theta}_c)$ has been taken as $P_n\phi_n(\boldsymbol{\theta}+\boldsymbol{\theta}_c)\phi_n^*(\boldsymbol{\theta}'+\boldsymbol{\theta}_c)f_n(E)$ (see § 3.1).

Evidently equation (29) is not very amenable to numerical evaluation since it contains, at the least, a triple angular integration (in the cylindrically symmetric case). The main comment on the inclusion of the spatial coherence of the electron source is that $I_0(\boldsymbol{\theta}_c)$ has a comparable angular halfwidth to $|\phi_n(\boldsymbol{\theta})|^2$ for inelastic electron scattering. Thus $I_0(\boldsymbol{\theta}_c)$ introduces into the inelastic scattering a further degree of incoherence; it is possible that in this case an incoherent approach would be a simpler and adequate solution to the problem (Crick and Misell 1971).

6.2. *Chromatic incoherence*

The incident electron wave is spatially coherent ($\boldsymbol{K}_0$ is constant) and the energy distribution of the incident electron beam is represented by $F(K_0)$. Electrons with a different K_0 are assumed to be incoherent. The electron is scattered inelastically in the specimen, where the transmitted electron has an energy represented by K_1; K_1-K_0 represents the energy loss of the scattered electron. Only electrons with the same K_0 and K_1 are coherent for a given n; electrons with a different K_0 or K_1 are superimposed incoherently, that is

$$j_i(\boldsymbol{r}_i) = \sum_{n=1}^{\infty} \int_0^{\infty}\int_0^{\infty}\int\int \psi_n(K_1-K_0+K_0', \boldsymbol{r}_0)\psi_n^*(K_1-K_0+K_0', \boldsymbol{r}_0') \times G\left(K_1, \frac{\boldsymbol{r}_i}{M}-\boldsymbol{r}_0\right) G^*\left(K_1, \frac{\boldsymbol{r}_i}{M}-\boldsymbol{r}_0'\right) F(K_0)\,\mathrm{d}\boldsymbol{r}_0\,\mathrm{d}\boldsymbol{r}_0'\,\mathrm{d}K_0\,\mathrm{d}K_1. \tag{30}$$

K_0' represents the most probable energy of the incident electron beam and K_0' corresponds to $E = 0$. Equation (30) may be transformed to

$$j_i(\boldsymbol{r}_i) = \sum_{n=1}^{\infty} \int_{-\infty}^{+\infty}\int_{-\infty}^{E_1}\int\int S_n(E_1-E, \boldsymbol{\nu})S_n^*(E_1-E, \boldsymbol{\nu}')T(E_1, \boldsymbol{\nu})T^*(E_1, \boldsymbol{\nu}') \times \exp\left(-2\pi\mathrm{i}(\boldsymbol{\nu}-\boldsymbol{\nu}')\,.\,\frac{\boldsymbol{r}_i}{M}\right) N(E)\,\mathrm{d}\boldsymbol{\nu}\,\mathrm{d}\boldsymbol{\nu}'\,\mathrm{d}E\,\mathrm{d}E_1. \tag{31}$$

The thermal energy distribution $N(E)$ is centred on $E = 0$ (see § 4 of I).

In $\boldsymbol{\theta}$ coordinates the final equation for the image intensity $j_i(\boldsymbol{r}_i)$ is

$$j_i(\boldsymbol{r}_i) = \left(\frac{K_0}{2\pi M}\right)^2 \sum_{n=1}^{\infty} P_n \int_{-\infty}^{+\infty}\int_{-\infty}^{E_1}\int\int \phi_n(\boldsymbol{\theta})\phi_n^*(\boldsymbol{\theta}') \exp[-\mathrm{i}K_0\{\gamma(E_1,\boldsymbol{\theta})-\gamma(E_1,\boldsymbol{\theta}')\}] \times D(\boldsymbol{\theta})D(\boldsymbol{\theta}')\exp\left(-\frac{\mathrm{i}K_0}{M}(\boldsymbol{\theta}-\boldsymbol{\theta}')\,.\,\boldsymbol{r}_i\right) f_n(E_1-E)N(E)\,\mathrm{d}\boldsymbol{\theta}\,\mathrm{d}\boldsymbol{\theta}'\,\mathrm{d}E\,\mathrm{d}E_1. \tag{32}$$

In practice the lower limits on the E integrations may be replaced by approximately

-1 eV, since $N(E)$ decreases rapidly to zero for large arguments and $f_n(E) = 0$ for $E \leqslant 0$. The chromatic incoherence of the source introduces a further incoherence into the inelastic component of the transmitted electron beam and the incoherent approximation may be a valid approach.

The effect of partial chromatic coherence of the incident electron beam may be included into the above analysis (see § 4).

7. Conclusion

A treatment of image formation by the inelastic component of the transmitted electron beam has been given. The effects of the energy distribution and chromatic aberration on the inelastic image have been considered. If the inelastic component forms an acceptable image, then the effect of chromatic aberration may be partially cancelled by underfocusing of the objective lens. It is also noted that the chromatic defect is not very dependent on the objective aperture size. The use of the semiangle α in the equation $r_c = C_c g(E)\alpha/E_0$, even for uniform illumination of the objective aperture, is an overestimate of the chromatic aberration. This conclusion is consistent with recent experimental estimates on the chromatic aberration for biological specimens (Nagata and Hama 1971). The value of minimizing the effect of the chromatic aberration on the inelastic component will depend on the structural information carried by the inelastic component and the ratio of inelastic to elastic scattering transmitted by the objective aperture. The former point is related to the localization of the inelastic electron scattering; the present work indicates that in biological materials the localization may be about 2–5 Å. On the latter point, the ratio $h_i(\alpha)/h_e(\alpha)$ is usually significantly greater than unity; even for gold, where the inelastic mean free path is about ten times larger than the elastic mean free path, h_i/h_e can be significantly greater than unity (see table 1). (The inelastic mean free path calculated from the plasmon excitation is probably too large, since one electron excitations are expected to be a predominant mechanism for inelastic electron scattering in the transition metals.)

The degree of coherence of the inelastic wave has been discussed in respect of the available experimental and theoretical evidence; the only decisive conclusion is that electrons with the same energy loss E and same n exhibit phase coherence. The possibility of coherence of the inelastic wave over a finite energy interval has been included into the calculation of the image intensity. Because of the incoherence of of the inelastic wave with respect to the unscattered and elastic waves, the intensity distribution in the image plane due to the inelastic component can be considered independently of the elastic–unscattered image. The essential incoherence of the inelastic component indicates that phase contrast effects with the inelastic image are unlikely.

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