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Image transfer with spatial coherence for aberration corrected transmission electron microscopes

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ABSTRACT

The formula of spatial coherence involving an aberration up to six-fold astigmatism is derived for aberration-corrected transmission electron microscopy. Transfer functions for linear imaging are calculated using the newly derived formula with several residual aberrations. Depending on the symmetry and origin of an aberration, the calculated transfer function shows characteristic symmetries. The aberrations that originate from the field's components, having uniformity along the *z* direction, namely, the n-fold astigmatism, show rotational symmetric damping of the coherence. The aberrations that originate from the field's derivatives with respect to *z*, such as coma, star, and three lobe, show non-rotational symmetric damping. It is confirmed that the odd-symmetric wave aberrations have influences on the attenuation of an image via spatial coherence. Examples of image simulations of haemoglobin and Si [211] are shown by using the spatial coherence for an aberration-corrected electron microscope.

1. Introduction

In transmission electron microscopy (TEM), the effect of partially coherent illumination can be expressed as a loss of information in a reciprocal space. The partial coherence involves a spatial term and temporal term, which respectively result from the brightness of the source and energy fluctuation of the imaging electrons. Here, we note that the current fluctuation of an imaging device might be converted to that of electron energy. The behaviours of the partial coherence were effectively formulated by Frank [1] and Wade and Frank [2], respectively, for spatial coherence and both spatial and temporal coherence of a weak phase object (e.g., thin amorphous specimen). The effects of both spatial coherence and temporal coherence are given as the envelope functions enfolding the contrast transfer function. Thus, regarding coherence under partially coherent illumination, the imaging of weak phase objects can be treated using the linear transfer theory. As for crystal specimens, Ishizuka [3] developed an algorithm for partially coherent imaging in which the interference between the diffracted waves is not weak enough to ignore. In the crystal image simulation of Ishizuka's method, coherences that are calculated for each pair of the imaging waves are involved in the convolution formula in the reciprocal plane; therefore, explicit formulation of attenuation of the Fourier component of the image is not given.

The transmission cross coefficient (TCC) is the basic starting point for calculating the partial coherence effect. TCC was originally defined for light optics [4] and has been analogously used in electron microscopy to develop coherence theories [1–3]. For electron microscopy, coherence theories including the brightness, electron energy distribution and resulting TCC have been consolidated in detail by Hawkes and Kasper [5]. TCC is the expression of the degree of coherence of two arbitrary waves in the form of the double integration of the image intensity over the electron source intensity and energy distribution. Linear imaging employs the TCCs between the transmitted wave and all scattered waves, while general imaging employs the TCCs of the pairs of any waves inside the imaging aperture. As the integrand in the TCC, the image component formed by two waves emitted from a certain point of an electron source is calculated using the phase disturbance introduced at each reciprocal point, which is defined by the wave aberration function. For a microscope without an aberration corrector, the wave aberration function for the partial coherence is presumed to include only the third-order spherical aberration (C_s) and defocus (df). Since the contrast transfer function of an electron microscope was given by Scherzer [6], C_s had been the main aberration that restricts the resolution of an electron microscope, until the aberration correction was established. Particularly, not much attention has been paid to the odd-symmetric wave aberrations because of their invisibility in the power spectrum,

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although they cause phase modulation that deforms the images of objects. The importance of odd-symmetric aberrations was examined by Zemlin [7], who introduced the measurement method of aberrations up to the second order, including coma and three-fold astigmatism. This method has been used for the measurement of residual aberrations since the birth of the aberration-corrected TEM [8].

Today, the aberration correction technique has been well established [9] and the residual aberrations are of wide variety [10], having magnitudes comparable to the corrected C_s values, which are approximately several micron metres or less. Therefore, for the calculation of spatial coherence in TCC, all of the influential aberrations should be taken into account. In this study, we incorporate all of the possible aberrations in the calculation of the spatial coherence term in TCC and derive the general formula to calculate the linear transfer function for weak phase objects. We also show examples of the transfer functions in some typical conditions. The temporal coherence effect on the aberration-corrected microscope will not be discussed detailed in this paper, assuming the validity of the conventional treatment [1–3], in which the attenuation function due to the defocus spread is multiplied. The temporal coherence should be carefully reconsidered in future works, taking into account the strongly excited correction devices in aberration-corrected microscopes.

2. Transmission cross coefficient (TCC) for general imaging

In this study, we focus our interest on the spatial coherence in TCC; for convenience, we will omit the temporal coherence term, which is the exponential decay function. In Section 5 (TEM image simulation), the conventional temporal coherence function will be used in the calculations. In Sections 2, 3 and 4, the source is treated as if it is quasi-monochromatic. For a quasi-monochromatic energy source, the decay function due to the temporal coherence can be regarded as a constant with one value, the image spectrum I($\boldsymbol{k}$) is given as follows;

$$\mathrm{I}(k) = \int \mathrm{TCC}\left(k + k', k'\right)\mathrm{F}\left(k + k'\right)\mathrm{F}^*\left(k'\right)\mathrm{d}k' \tag{1}$$

$$\mathrm{TCC}\left(k'', k'\right) = \int \mathrm{s}(\mathbf{q})\mathrm{t}\left(q + k''\right)\mathrm{t}^*\left(q + k'\right)\mathrm{d}q \tag{2}$$

The function $\mathrm{s}(\boldsymbol{q})$ is the normalised intensity distribution of the effective source, which is considered as an assembly of incoherent point sources. $\mathrm{F}(\boldsymbol{k})$ is the Fourier transform of the exit wave from the object, while $\mathrm{t}(\boldsymbol{k})$ is the pupil function, expressed as

$$\mathrm{t}(k) = \mathrm{a}(k)\exp(2\pi i\chi(k)/\lambda) \tag{3}$$

where

$$\mathrm{a}(\boldsymbol{k}) = \begin{cases} 1, & \text{if } \boldsymbol{k} \notin \text{objective lens aperture} \\ 0, & \text{if } \boldsymbol{k} \notin \text{objective lens aperture} \end{cases} \tag{4}$$

The wave aberration $\chi(\boldsymbol{k})$, which is a real value function and considered in units of length, represents the distance from the Gaussian sphere to the wave front. The $\chi(\boldsymbol{k})$ for microscopes without C_s correctors is dominantly determined by C_s and d_f. However, for the C_s-corrected microscopes, wave aberration $\chi(\boldsymbol{k})$ should include the aberration coefficients up to six-fold astigmatism to reproduce the experimental high resolution images.

$$\begin{aligned}\chi(\boldsymbol{k}) = \mathrm{Re}\{&\mathbf{O}_2|\omega|^2/2 + \mathbf{A}_2\omega^{*2}/2 + \mathbf{P}_3\omega\omega^{*2}/3 + \mathbf{A}_3\omega^{*3}/3 \\ &+ \mathbf{O}_4|\omega|^4/4 + \mathbf{Q}_4\omega\omega^{*3}/4 \\ &+ \mathbf{A}_4\omega^{*4}/4 + \mathbf{P}_5\omega^2\omega^{*3}/5 + \mathbf{R}_5\omega\omega^{*4}/5 + \mathbf{A}_5\omega^{*5}/5 \\ &+ \mathbf{O}_6|\omega|^6/6 + \mathbf{A}_6\omega^{*6}/6\}\end{aligned} \tag{5}$$

Here, we use Sawada's notation of the aberration coefficients [11] in Eq. (5), which is listed in the Appendix. $\boldsymbol{\omega}$ is the complex angle, which is related to the spatial frequency vector $\boldsymbol{k}$ by

$$\omega = \boldsymbol{k}\lambda \tag{6}$$

$$\boldsymbol{k} = r_k\exp\left(i\theta_k\right) = k_x + ik_y \tag{7}$$

where λ is the wavelength of the electron. The polar coordinate expression of $\boldsymbol{k}$ and $\boldsymbol{\omega}$ is convenient when tilt-induced wave aberration caused by misalignment needs to be evaluated [12], while Cartesian coordinates are suitable when intrinsic geometrical aberrations need to be expressed as derivatives of the wave aberrations.

The partial coherence integration in Eq. (2) can be solved analytically under the following conditions:

(a) the incoherent source has a Gaussian distribution of size q_0:

$$\mathrm{s}(\boldsymbol{q}) = \left(1/\left(\pi q_0{}^2\right)\right)\exp\left(-\left(|q|^2/q_0{}^2\right)\right) \tag{8}$$

(b) the wave aberration can be approximated by using the first-order term of a Taylor series:

$$\begin{aligned}\chi(\boldsymbol{k} + \boldsymbol{q}) &\approx \chi(\boldsymbol{k}) + \boldsymbol{q}\cdot\nabla\chi(\boldsymbol{k}) \\ &= \chi(\boldsymbol{k}) + \left(q_x, q_y\right)\cdot\left(\partial\chi(\mathbf{k})/\partial k_x, \partial\chi(\mathbf{k})/\partial k_y\right)\end{aligned} \tag{9}$$

(c) the aperture term $\mathrm{a}(\boldsymbol{k}+\boldsymbol{q})$ can be removed from the TCC expression because the source size $|\boldsymbol{q}|$ in electron microscopy is negligibly small compared to the OL aperture and imaging frequency $|\boldsymbol{k}|$.

Then, TCC becomes [1,3]

$$\mathrm{TCC}\left(\boldsymbol{k}'', \boldsymbol{k}'\right) = \exp\left(i\gamma\left(\boldsymbol{k}''\right) - i\gamma\left(\boldsymbol{k}'\right)\right)\int s(\boldsymbol{q})\exp\left\{2\pi i\boldsymbol{q}\cdot\left(\nabla\chi\left(\boldsymbol{k}''\right) - \nabla\chi\left(\boldsymbol{k}'\right)\right)/\lambda\right\}d\boldsymbol{q} \tag{10 a}$$

$$= \exp\left(i\gamma\left(\boldsymbol{k}''\right) - i\gamma\left(\boldsymbol{k}'\right)\right)\exp\left\{-\pi^2 q_0{}^2\left(1/\lambda^2\right)\left|\nabla\chi\left(k''\right) - \nabla\chi\left(k'\right)\right|^2\right\} \tag{10 b}$$

where $\gamma(\boldsymbol{k})$ is the phase shift caused by the wave aberration, i.e.,

$$\gamma(\boldsymbol{k}) = (2\pi/\lambda)\chi(\boldsymbol{k}) \tag{11}$$

Eq. (10a and b) were originally derived from the wave aberration with only C_s and d_f [1]. Even with the more general $\chi(\boldsymbol{k})$ given by Eq. (5), the same procedure can be applied to obtain the same result as shown above.

Eq. (10b) can be modified by replacing the derivative of the wave aberration with the geometrical aberration $\mathbf{G}(\boldsymbol{k})$, which is given by

$$\mathbf{G}(\boldsymbol{k}) = \nabla\chi(\boldsymbol{k})/\lambda \tag{12}$$

$$\begin{aligned}&= \mathrm{O}_2\omega + \mathbf{A}_2\omega^* + \left(2\mathbf{P}_3\omega\omega^* + \mathbf{P}_3{}^*\omega^2\right)/3 + \mathbf{A}_3\omega^{*2} + \mathbf{O}_4\omega^*\omega^2 \\ &\quad + \left(3\mathbf{Q}_4\omega^{*2}\omega + \mathbf{Q}_4{}^*\omega^3\right)/4 + \mathbf{A}_4\omega^{*3} + \left(3\mathbf{P}_5\omega^2\omega^{*2} + 3\mathbf{P}_5{}^*\omega\omega^{*3}\right)/5 \\ &\quad + \left(4\mathbf{R}_5\omega\omega^{*3} + \mathbf{R}_5{}^*\omega^4\right)/5 + \mathbf{A}_5\omega^{*4} + \mathbf{O}_6\omega^{*2}\omega^3 + \mathbf{A}_6\omega^{*5}\end{aligned} \tag{13}$$

The integral of Eq. (10a) coincides with the Fourier transform of