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# Recording low and high spatial frequencies in exit wave reconstructions ☆

S.J. Haigh [a,*], B. Jiang [b], D. Alloyeau [c], C. Kisielowski [c], A.I. Kirkland [a]

[a] Department of Materials, University of Oxford, Parks Road, Oxford, OX1 3PH, UK
[b] FEI Company, NE Dawson Creek Drive, Hillsborow, OR 97124, USA
[c] National Center for Electron Microscopy and Joint Center for Artificial Photosynthesis, Lawrence Berkeley National Laboratory, One Cyclotron Road, Berkeley, CA 94720, USA

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ABSTRACT

Aberration corrected Transmission Electron Microscope (TEM) images can currently resolve information at significantly better than 0.1 nm. Aberration corrected imaging conditions seek to optimize the transfer of high-resolution information but in doing so they prevent the transfer of low spatial frequency information. To recover low spatial frequency information, aberration corrected images must be acquired at a large defocus which compromises high spatial frequency information transfer. In this paper we present two *a posteriori* solutions to this problem in which the information bandwidth in an exit wave reconstruction is increased. In the first we reconstruct the electron exit wavefunction from two focal series datasets, with different, uniform focal steps, experimentally demonstrating that the width of the transfer interval can be extended from 0.2 $nm^{-1}$ (~5 nm) to better than 10 $nm^{-1}$ (0.1 nm). In the second we outline the use of a focal series recorded with a non-uniform focal step to recover a wider range of spatial frequencies without the need for a large number of images. Using simulated data we show that using this non-uniform focal step the spatial frequency interval for a five image data set may be increased to between 0.25 $nm^{-1}$ (4 nm) and 8.3 $nm^{-1}$ (0.12 nm) compared to between 0.74 $nm^{-1}$ (1.4 nm) and 8.3 $nm^{-1}$ (0.12 nm) for the standard focal series geometry.



## 1. Introduction

Modern transmission electron microscopes have interpretable resolution limits better than 0.1 nm at intermediate accelerating voltages [1–6] due to the development of corrected electron optics suitable for TEM imaging with plane wave illumination [3–13]. Optimal aberration corrected imaging conditions use values of the correctable third order spherical aberration ($C_3$) and adjustable defocus ($C_1$) to achieve a point resolution for phase-contrast imaging that is limited by the uncorrectable fifth order spherical aberration ($C_5$) or first order chromatic aberration ($C_c$) [14–18]. For high resolution phase contrast imaging these parameters provide a wide interval of spatial frequencies that are transmitted without phase oscillation but at the expense of information transfer at intermediate and low spatial frequencies [17].

For uniform small unit cell structures, the absence of low spatial frequency transfer ($<1$ $nm^{-1}$) in an image is of little concern and can even improve the qualitative appearance of images by reducing transfer of low spatial frequency components in amorphous damage or contamination layers. In contrast, many complex systems (including proteins and other biological macro-molecules) have characteristic feature sizes greater than 1 nm. For phase contrast imaging of these objects, it is normal practice to record images at high defocus values that maximize transfer of low spatial frequencies. However, this improvement in low spatial frequency transfer introduces phase oscillations at high spatial frequencies which adversely affect the interpretability of high resolution information.

Physical phase plates offer an alternative route to increase the low resolution phase contrast of suitable specimens and do not directly rely on the control of objective lens aberrations. In this approach a phase-shifting device is placed in the back focal plane of the objective lens (or a suitable conjugate plane) to introduce a $\pi/2$ relative phase shift between scattered and unscattered electrons. A variety of physical phase plate geometries have been demonstrated including thin discs with a central aperture [19,20], Foucault plates (or knife edges) [21], electrostatic systems [22–24] and magnetized rings which produce a phase difference using the Aharonov–Bohm effect [8,25]. Physical phase plates offer the



* Corresponding author. Present address: School of Materials, University of Manchester, Grosvenor Street, Manchester M1 7HS, UK. Tel.: +44 161 3063618; fax: +44 161 3063586.

*E-mail addresses:* sarah.haigh@manchester.ac.uk (S.J. Haigh), angus.kirkland@materials.ox.ac.uk (A.I. Kirkland).

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potential for optimal phase transfer over a wide range of spatial frequencies [26] particularly when combined with aberration corrected optics [24]. However, there are still considerable practical challenges to be overcome including effects due to charging [25] and long term stability [23]. In this paper we propose an alternative way of recovering high spatial frequencies without diminishing contrast at lower spatial frequencies using a modified through focus series exit wavefunction restoration scheme. We explore the spatial frequencies recovered in exit wavefunctions from typical experimental data sets acquired using bimodal or non-uniform focal step geometries to maximize the interval of spatial frequencies for different instruments. Specifically we present results recorded using a spherical aberration corrected microscope operated at 200 kV and a microscope with both spherical and chromatic aberration correction operated at 80 kV.

## 2. Phase contrast imaging theory

The Phase Contrast Transfer Function (PCTF) is frequently used to describe the contrast of images of weak phase objects in the TEM and is given by sin ($\chi(k)$), which contains the phase change ($\chi(k)$) due to the wave aberration function. Considering only spherically symmetric aberrations to third order $\chi(k)$ is given by

$$\chi(k) = \frac{1}{2}\pi C_1 \lambda k^2 + \frac{1}{4}\pi C_3 \lambda^3 k^4$$

where $k$ is the spatial frequency, $\lambda$ the electron wavelength, $C_1$, the first order objective lens defocus, and $C_3$, the 3rd order spherical aberration of the objective lens. The partial coherence of the electron wave limits contrast transfer in TEM, and can be described within a linear imaging approximation by envelope functions for the partial temporal coherence $E_t(k)$ and partial spatial coherence $E_s(k)$ by

$$E_s(k) = \exp\left\{-\left(\frac{\beta}{2\lambda}\right)^2 \left(\frac{\partial\chi(k)}{\partial k}\right)^2\right\}$$

$$E_t(k) = \exp\left\{-\frac{1}{2}(\pi\Delta\lambda)^2 k^4\right\}$$

where $\beta$ is the semi-angle characterizing the root mean squared spread in the Gaussian beam profile distribution (beam divergence) [27]. The focal spread, $\Delta$, is calculated from the intrinsic energy spread of the electron source $\delta E$ and the instabilities of the current, $\delta I$, and voltage, $\delta V$, supplies

$$\Delta = C_c\left\{\left(\frac{2\delta I}{I}\right)^2 + \left(\frac{2\delta E}{E}\right)^2 \left(\frac{\delta V}{V}\right)^2\right\}$$

Conventionally an information limit is defined by the spatial frequency where the product $E_s(k)E_t(k)$ falls below the recorded noise level which is assumed here to be 10% (i.e. $E_s(k)$ $E_t(k)$sin ($\chi(k)$)=0.1).where fifth order spherical aberration is negligible, Lentzen [16] has derived optimal values for residual values of the 3rd order spherical aberration $C_{3,opt}$ and first order defocus $C_{1,opt}$ which provide a compromise between the transfer of high frequency phase contrast and image delocalization for an information limit, $k_{inf}$, as

$$C_{3,o}pt = \frac{64}{27}\frac{1}{\lambda^{3}k_{inf}^4}$$

$$C_{1,o}pt = -\frac{16}{9}\frac{1}{\lambda k_{inf}^2}$$

Importantly, a negative value of $C_3$ (with a corresponding positive $C_1$) gives enhanced contrast where non-linear components are present in the image [16]. However, we only consider linear imaging in this study and have therefore chosen a positive value for $C_3$ in all our calculations.

## 3. Experimental methods

In this paper we present experimental exit wavefunction restorations recorded using two different aberration corrected microscopes; the first a medium voltage instrument with 3rd order spherical aberration correction (Microscope 1: a JEOL 2200MCO FEG (S)TEM operated at 200 kV with a focal spread of 3.8 nm) and the second a low voltage instrument with both 3rd order spherical and 1st order chromatic aberration correction (Microscope 2: TEAM 1 operated at 80 kV with a low beam convergence (~40 μrad) and a focal spread of 2 nm) [1,9].

Exit wavefunction restorations were calculated using a linear Wiener filter approach [28] implemented within the Semper image processing software [29] and also using the FTSR commercial software package [30]. Prior to exit wavefunction restoration the images in all data sets were aligned using a phase correlation function/phase contrast index function to correct for specimen drift and to determine the precise focal step variations between images [31,32]. The modulation transfer function for the camera was determined independently and compensated as part of the restoration process [33,34].

## 4. Results and discussion

To determine the optimal imaging conditions for our focal series we have initially examined phase contrast transfer functions at different values of defocus and spherical aberration for Microscope 1 (JEOL 2200MCO FEG TEM at 200 kV). Fig. 1(a) and (b) compares information transfer for this microscope with an uncorrected value of $C_3$ to the case where the spherical aberration has been corrected to an optimized value, $C_{3,opt}$ =10 μm. For uncorrected $C_3$=0.45 mm, at the Scherzer defocus [14] ($C_1 = (\lambda C_3)^{0.5} = -34$ nm) Microscope 1 has a point resolution (the upper limit of the first broad passband in the PCTF) of 4.5 nm$^{-1}$ (0.22 nm) and an interval of oscillation free phase transfer between the point resolution and 0.57 nm$^{-1}$ (1.74 nm). The interval of oscillation free transfer for the same instrument under aberration corrected imaging conditions ($C_{3,opt}$ =10 μm, $C_{1,opt}$=−5 nm) is shifted to higher spatial frequencies with the point resolution improved to 8.3 nm$^{-1}$ (0.12 nm). However, the lower spatial frequency limit for oscillation free transfer is also increased from 0.57 nm$^{-1}$ (1.74 nm) to 1.6 nm$^{-1}$ (0.62 nm) as shown in Fig. 1(b). The significant advantage of the aberration corrected imaging condition is that the PCTF is free from contrast oscillations at all spatial frequencies. In contrast, the PCTFs shown in Fig. 1(a), (c) and (d) contain oscillations at high spatial frequencies which introduce image delocalization (Fig. 2). However the transfer of low and medium spatial frequencies is also reduced under aberration corrected conditions. For uncorrected instruments large defocus values can be used to improve low spatial frequency transfer at the expense of introducing significant oscillations in the PCTF at high spatial frequencies (Fig. 1(c), (d)). We also note that recent work has shown that high resolution at low voltage is limited by partial temporal coherence and benefits significantly from chromatic aberration correction [35–37] which is not considered in the calculations presented here.

Restoring the exit wavefunction from a series of TEM images with differing defocus values provides local structural information in the phase of the exit wavefunction [38–44] and the primary