Finding phase information in the darkness

S.J.Haigh¹, A.I. Kirkland¹

¹ Department of Materials, University of Oxford, Parks Road, OX1 3PH, UK.
sarah.haigh@materials.ox.ac.uk

Abstract. High resolution local structure information is revealed when the complex exit wavefunction is restored from a series of transmission electron microscope (TEM) images. Aberration corrected image data sets allow high spatial frequency information to be restored but this is often at the expense of lower spatial frequencies. In this paper we present a new approach to exit wavefunction restoration using a novel five image focal series with non-uniform focal steps. This data set extends the spatial frequency range over which information can be successfully restored. We present simulation studies comparing this approach to a conventional focal series and demonstrate the benefit of including low spatial frequencies in the restoration. We propose that this approach will be important for the study of large unit cell materials or those with long range ordering.

1. Introduction

High resolution Transmission Electron Microscopy (TEM) is an established technique for atomic scale imaging of materials. With the recent development of electron optical elements capable of direct aberration correction, current instruments are able to routinely record “aberration corrected” images of inorganic materials with a resolution better than 0.1 nm [1-3]. Fig. 1 compares the contrast transfer function (CTF) for the JEOL 2200MCO (S)TEM [4] at the Scherzer defocus condition, \( f_{\text{Sch}} = -\sqrt{C_s/\lambda} \), with and without 3rd order spherical aberration correction. The CTF is defined here as \( E(k) \sin (\chi(k)) \), where \( k \) is the spatial frequency, \( E(k) \) the envelope function resulting from spatial and temporal coherence effects and \( \chi(k) \) is the phase change due to the wave aberration function assuming all aberrations above 3rd order are negligible. Adjustment of the 3rd order spherical aberration to 10 \( \mu \)m [5] clearly eliminates oscillations in the CTF at high spatial frequencies as shown in figure 1. In Fig. 1(a) and (b) the interpretable limit of 10% transfer is improved from 0.22 nm to 0.14 nm when the spherical aberration is corrected. However, a disadvantage of aberration corrected imaging is that the transfer at low spatial frequencies is significantly reduced. At the Scherzer defocus condition the lower limit of 10% transfer, for an aberration corrected microscope, is 0.63 nm whereas for a conventional \( C_s \) the limit is 1.74 nm. This does not affect imaging of small unit cell crystalline materials although it reduces the contrast of long range features such as specimen bending and contamination. However, for large unit cell structures such as macromolecules this loss of long range information is critical.

For imaging large organic structures, a large defocus (>200 nm) is often used in order to improve the transfer of low spatial frequencies but this has the disadvantage of introducing rapid oscillations to the CTF at high spatial frequencies as shown in Fig. 1c. These phase reversals can be partially removed using ‘CTF correction’ [6,7] however, the accuracy of defocus determination limits interpretability for resolvable distances smaller than 0.5 nm. Slightly better resolution is achievable by ‘single particle reconstruction’ [8] but requires averaging many thousands of images.
2. Extending the low spatial frequency transfer in exit wavefunction restoration.

Restoring the exit wavefunction from a series of TEM images with differing defocus values reveals local structural information in the phase of the exit wavefunction. Due to the need for multiple images this technique is not generally suitable for radiation sensitive samples. However, for materials where stability is related to the total electron dose, exit wavefunction restoration using a Wiener restoring filter optimally suppresses noise by a factor of \(\sqrt{N}\) where \(N\) is the number of images in the data set [9]. Thus for an equivalent total electron dose, the noise in the restored exit wavefunction will be reduced in comparison to a single image.

A Wiener filter approach to exit wavefunction restoration [9] requires a minimum of 3 images, although for inorganic materials a typical experimental data set consists of 10-20 images centered about the Scherzer defocus with a small uniform focal increment [10,11]. For radiation sensitive materials the optimal number of images is determined by experimental considerations, such as the minimum detectable signal in a single image, the noise characteristics of the detector used and the minimum reproducible exposure time, as well as by the specimen’s total dose tolerance. In practice a five image data set has been found to be an experimentally feasible compromise for beam sensitive specimens.

A five image dataset with a focal step of 5 nm centered about the Scherzer defocus is limited to a long range (low spatial frequency) resolution of \(\sim1\text{nm}\). In this paper we propose using an alternative five image dataset for exit wave restoration with non-uniform focal increments. The defoci of the novel data set were chosen as -5 nm, -10 nm, -15 nm, -54 nm and -200 nm (for the microscope used). These focal increments do not compromise the transfer of high spatial frequencies but the inclusion of images at larger defoci optimizes transfer of long range information, extending the lower 10% transfer limit to 4 nm. The overlap of the consecutive CTF’s has been optimised such that the transfer does

![Fig. 1. Contrast transfer functions for the JEOL 2200MCO (S)TEM (200kV) with a focal spread of 3.8 nm at different values of defocus (C₁) and spherical aberration (C₃). For uncorrected C₃ the Scherzer defocus condition (a) transfers phase information without contrast reversals for resolutions between 1.74 nm and 0.22 nm. The equivalent range for a corrected value of C₃ at the Scherzer defocus (b) lies between 0.63 nm and 0.14 nm and at a large underfocus (200nm) (c) lies between 4 nm and 0.72 nm.](image)
not fall below 60% of the maximum for any spatial frequency (Fig. 2).

![Fig. 2. Comparison of CTF's for the JEOL 2200MCO (S)TEM with a focal spread of 3.8 nm and a third order spherical aberration of 10 µm at various defoci as marked in the legend. The real space distance (1/k) in nm is indicated on the horizontal axis.](image)

**Fig. 2.** Comparison of CTF’s for the JEOL 2200MCO (S)TEM with a focal spread of 3.8 nm and a third order spherical aberration of 10 µm at various defoci as marked in the legend. The real space distance (1/k) in nm is indicated on the horizontal axis.

![Fig. 3. Modulus (left) and phase (right) of the exit wavefunctions restored from five simulated images of an 18.5 nm thick sample of copper phthalocyanine with an electron dose of 100 electrons Å⁻². The restoration in (a) used a standard focal series of equally spaced defocus values and (b) used a focal series with unequal focal steps to extend the spatial frequency range of the restoration. The original exit wavefunction used to simulate the input TEM images is shown inset.](image)

**Fig. 3.** Modulus (left) and phase (right) of the exit wavefunctions restored from five simulated images of an 18.5 nm thick sample of copper phthalocyanine with an electron dose of 100 electrons Å⁻². The restoration in (a) used a standard focal series of equally spaced defocus values and (b) used a focal series with unequal focal steps to extend the spatial frequency range of the restoration. The original exit wavefunction used to simulate the input TEM images is shown inset.
3. Simulation studies.
To evaluate this new focal series geometry for low spatial frequency transfer image simulations have been performed using the multislice method [12] for the α-copper phthalocyanine (CuPc) structure [13]. Specimen thicknesses of 3.7 nm, 8 nm, 18.5 nm and 37 nm were investigated at various electron doses appropriate to the imaging of beam sensitive materials (10, 25 and 100 electrons Å$^{-2}$). These simulated images were then used as input to the exit wavefunction restoration procedure. The exit wave recovered from the extended focal series geometry (defoci of -5nm, -10nm,-15nm, -54nm and -200nm) was subsequently compared to that recovered from the traditional focal series (defoci of -5nm, -10nm,-15nm, -20nm and -25nm). The comparison for a sample thickness of 18.5nm and a moderate electron dose (100 electrons Å$^{-2}$) is shown in Fig. 3. Using the new extended focal series approach the phase of the restored exit wavefunction closely resembles that of the original exit wavefunction, whereas that restored from the standard focal series approach is clearly missing low spatial frequency information.

4. Conclusions
In this paper we have presented a new geometry for exit wavefunction restoration which improves the transfer of low spatial frequencies to the exit wavefunction. This approach extends the spatial frequency range for a five image data set to between 4 nm and 0.09 nm (compared to between 1nm and 0.09 nm for a standard focal series). The benefit of this approach for a large unit cell test material has been demonstrated using simulations.

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