Two-dimensional Object Functions and Three-dimensional Illumination Functions: their Validity, Interaction and Utility

L. Jones, P. D. Nellist
Department of Materials, University of Oxford, Parks Road Oxford OX1 3PH, UK
E-mail: lewys.jones@materials.ox.ac.uk

Abstract. In high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) the data is generally interpreted as the convolution of the sample’s sharply-peaked object function with the intensity of the real-valued point-spread function (PSF) of the illumination. As a single image is itself necessarily two-dimensional (2D) it is typically assumed that the object function and PSF can also be accurately described as 2D; this is the so-called 2D-object imaging assumption. Here the validity of these two-assumptions is evaluated using experimental HAADF STEM focal-series. It is found that the contrast contained in each image of the focal-series is accurately described by the convolution of a focus-invariant 2D object with a 2D optical-transfer function (OTF) which describes the illumination at the focus value used for imaging.

1. Introduction
The detector geometry in the high-angle annular dark-field (HAADF) imaging mode in the scanning transmission electron microscopy (STEM) causes the recorded signal to be regarded as being incoherent. Because of this, it is the intensity distribution of the focussed STEM probe that most closely dictates both the recorded contrast and resolution. This is highlighted by the so-called two-dimensional (2D) object function model used to describe the imaging of on-axis crystalline samples:

\[ I(r) = |P(r)|^2 \otimes O(r) \]  

(1)

In this model the image data, \( I \), that is recorded as a function of real-space probe position, \( r \), is described by a convolution between the intensity (modulus squared) of the 2D real-space probe-function, \( P(r) \), and a 2D representation of the object, \( O(r) \), where the object the sample is represented by a collection of sharply-peaked points at the atomic column positions and is sparse otherwise.

While this model describes the contrast in individual HAADF images, it is becoming increasingly common to record through-focus image series for either optical-sectioning [1] or image reconstruction [2] type experiments. We now have to consider the third-dimension (either real-space z-direction or equivalently defocus), and whether it remains reasonable to assume that the probe and/or object behaves two-dimensionally. The first of these is easy to address as from a consideration of the geometric optics of the highly-convergent STEM probe we can immediately see that this varies greatly with defocus [3], such that \( P(r,z) \).

1 To whom any correspondence should be addressed.
The two remaining scenarios to consider are then:

\[
S(r, z) = \begin{cases} 
|P(r, z)|^2 \otimes O(r) & \text{Object behaves 2D} \\
|P(r, z)|^2 \otimes O(r, z) & \text{Object behaves 3D}
\end{cases}
\]  

(2)

The first of these assumes that the object function can be treated as being independent of defocus, and the second that focus-dependent information appears in the object function. To determine which of these two regimes operates we need then to evaluate the “apparent thickness” of the object function and focal-series allows us to do this.

2. Method

To interpret the z-response of the sample we must first understand the defocus-response of the illumination. Fortunately the 3D form of the STEM probe is well understood and can be readily simulated. Probe simulations were performed in the MatLab™ programming environment matching the experimental conditions (200kV acceleration and a 22 mrad objective aperture).

Next, to capture the entire profile of the contrast response a HAADF-STEM focal-series should be recorded with a total defocus-range such that the first and last images are just out of focus (this can equivalently be achieved with fixed optics and a piezo scanning stage). Here focal-series were recorded from [100] oriented MgO smoke cubes, where sample thickness can be inferred from the cube width. Within this image-stack then the laterally resolved contrast would begin at near-zero, peak when in-focus and then deteriorate again. To quantitatively measure the contrast transfer performance of the images in the series the common technique of Fourier transform (FT) inspection is useful. With lattice-resolved images of crystalline samples, transforming each image in the series into reciprocal-space allows the periodic image components (laterally resolved contrast) to be observed as FT-spot. To quantify the contrast we can then simply integrate the modulus in the area immediately adjacent to each FT-spot and track this through defocus. This can be repeated for every observable spot in the FT and after fitting of say a Gaussian profile curve, a defocus –spread, or –persistence is measured from this fit. Interpolating this defocus-spread across all reciprocal space and finding the minima then gives the ultimate depth-sensitivity exhibited by the experimental data [4].

3. Results and Discussion

First the expected contrast transfer performance of the simulated illumination:

Figure 1. Simulated OTFs for two aberration conditions. Left: aberration free, and right: the same but with +50 µm spherical aberration (C3,0). Horizontal axis shows reciprocal nanometres (spatial frequency) while vertical axis shows real-space defocus.
In the perfect (diffraction limited only) probe the band of peak information transfer is flat with respect to defocus and centred about zero defocus. Upon the introduction of positive spherical aberration this both shifts and sweeps towards negative defocus (familiar to many operators $C_s$ must be balanced with defocus). In this investigation only the defocus width of the maximal transfer band is needed and can be measured with again the same Gaussian fitting. Further examples of such simulations, including discussion of the OTF shapes stemming from other common non-round aberrations can be found in [5].

After transforming the experimental data-cube to Fourier-space and thresholding (to say a 10% intensity level) only those voxels with usable contrast are revealed, Figure 2.

![Figure 2](image2.png)

**Figure 2.** Side-on 3D-visualisation of the lattice contrast in an experimental focal-series. Vertical rods correspond to the intensity iso-surface encapsulating FT-spot intensities. Inset shows the viewing direction of the side-on stack.

Using this representation the locations of FT-spot rods can be readily identified and analysed.

![Figure 3](image3.png)

**Figure 3.** Integrated intensities (blue dots) from the immediate area around an example [200] FT-spot (3 pixel radius) across a 30-frame experimental focal-series. Sample was $\approx$83 nm thick. Dashed line indicates the fitted Gaussian profile through the data used to measure the defocus spread / persistence. Note that at the start of the series, the FT-spot intensity is near zero indicating no resolved image contrast.
Now the ratio between the FT-spot defocus-spread (contrast-persistence) and the OTF $z$-response gives the ‘normalised thickness’ this is the contribution to the defocus-spread in the convolution in equation (2). Plotting this as a function of the real (measured) thickness we get Figure 4.

![Figure 4](image)

**Figure 4.** Plot of the calculated ‘normalised thickness’ versus the measured real thickness (red squares). The dashed line represents the 100% datum where experimental objects behave perfectly 2D. Additional results from different voltages and objective aperture sizes are shown (circles and diamonds) indicating the generality of the result.

Now we find that for a wide range of sample thicknesses all appear approximately 2D. The reader may verify this by comparing again the contrast-persistence in Figure 3 of around 15 nm with both the true sample thickness of ≈83 nm, and the OTF profile in Figure 1 (also ≈15 nm).

4. Conclusions

- With increasing interest in focal-series for optical-sectioning or image reconstruction the 2D object-function assumption and imaging model merits evaluation.
- Experimental focal-series facilitate such an investigation, measuring the defocus-persistence of FT-spots (an indicator of resolved contrast).
- Comparison with simulated probe-OTFs allows the additional $z$-blurring from the sample to be investigated equivalent to the apparent sample thickness in the convolution model.
- We find that for the thickness range investigated the MgO samples appear two-dimensional and we confirm the validity of the 2D object function assumption for simply projecting crystals.
- This conclusion then suggests that it may be possible to retrieve the 2D sample object function from the over-determined 3D focal-series, and an image reconstructed free from the effects of residual illumination aberrations.

5. References

[1] G. Behan, E.C. Cosgriff, A.I. Kirkland, P.D. Nellist, *Phil. Trans. A* **367** (2009).
[2] L. Jones, P.D. Nellist, *Microscopy and Microanalysis* **18** (2012).
[3] A.R. Lupini, N. de Jonge, *Microscopy and Microanalysis* **17** (2011).
[4] L. Jones, P.D. Nellist, “Testing the Accuracy of the Two-dimensional Object Model in HAADF STEM”, *Micron* (in review) (2013).
[5] L. Jones, P.D. Nellist, “Three Dimensional Optical Transfer Functions in the Aberration Corrected Scanning Transmission Electron Microscope”, *Journal of Microscopy* (in submission) (2013).

Acknowledgments

The authors thanks Dr Armand Béché for help with the experiential imaging. This research was supported by the EU-FP7 Grant Agreement 312483 (ESTEEM2) and the UK EPSRC.