# A three-dimensional investigation of the STEM-probe / sample interaction by annular dark-field focal series

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If we assume that because of electron channeling in the scanning transmission electron microscope (STEM) thin samples can be reliably approximated as two-dimensional (2D) object functions, then recorded high-angle annular dark field (HAADF) images can be considered as the convolution of the illumination's point spread function with this 2D object function [1]. A STEM HAADF focal-series then represents a convolution of the object with each of a differently defocused probe and when inspecting such data, the effects of uncorrected lens aberrations can become apparent. Additionally, previous work has also showed that defocused atomic-resolution HAADF images can reveal information at spatial frequencies higher than those of a Scherzer focus image [1,2]. Unfortunately, at large defocuses lower spatial frequencies are transferred less strongly. If a focal series of STEM images is recorded then all spatial frequencies are transferred at their peak intensity in one image or another.

Recording focal-series then offers us a means to evaluate the properties of the STEM illumination itself. The simplest example of this might be, for example, diagnosing a two-fold astigmatism. In this case, spatial frequencies in one direction are transferred more strongly on one side of focus than the other. In a complimentary fashion their perpendicular counterparts are transferred more strongly with an equal defocus of the opposite sense. Again in a focal series data set all the transferred information is available.

Recently we have developed a new technique of STEM focal-series restoration (FSR) which offers the potential for lens aberration correction in software, rather than hardware. The result of this is both improved image resolution and signal-noise performance compared to uncorrected images. The reconstruction algorithm methodology is to analyse the focal-series data cube as a series of Fourier transforms (FTs). The integrated intensity of spots, which correspond to sample lattice-spacings, is then inspected. The defocus at which this intensity comes to a maximum then represents the focus at which that spatial frequency was transferred most strongly. Performing this for every visible FT spot then defines a surface of maximum contrast as shown in Figure 1. The simplest method of restoring an image from the focal-series is to extract the complex data corresponding to this maximum-contrast surface. This creates a reconstructed FT whose inverse Fourier transform then yields a single real-space image. More involved methods currently being evaluated could include a weighted average of the images either side of this maximal-surface or, possibly, a full 3D deconvolution of an experimentally determined probe-form from the data volume; extracting all the information transferred and making use of every electron of sample dose. Initial analysis was limited to the detection of two-fold astigmatism; however, recent refinements have made it possible to detect more subtle remnant aberrations such as four-fold astigmatism.

In addition to the image-reconstruction described above, within the focal-series data, the intensity of the FT spots across the focus range can be used to calculate the depth-resolution for each lateral spatial frequency. This can equivalently be thought of as the 3D optical transfer function (OTF) of the STEM probe. Figure 2 shows an experimentally determined STEM OTF and the equivalent predicted from theory [3]. The horizontal extreme of the OTF gives the maximum lateral resolution achievable by the instrument, the incline at the center gives the convergence angle of the condenser aperture and the vertical peak gives the greatest possible depth resolution. An analysis of this depth-resolution as a function of sample thickness is currently underway to determine the validity of the reconstruction's initial 2D object-function assumption and as a means to explore the range of thicknesses over which electron channeling theory is valid. Investigating the relationship between electron-channeling (2D object behavior) and optical-sectioning (3D object behavior) is expected to yield important clarification to these essential STEM topics [4].

## *References*

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[4] Data courtesy of Dr. P Wang, U. Oxford. Work financially supported by the EPSRC (DTA).

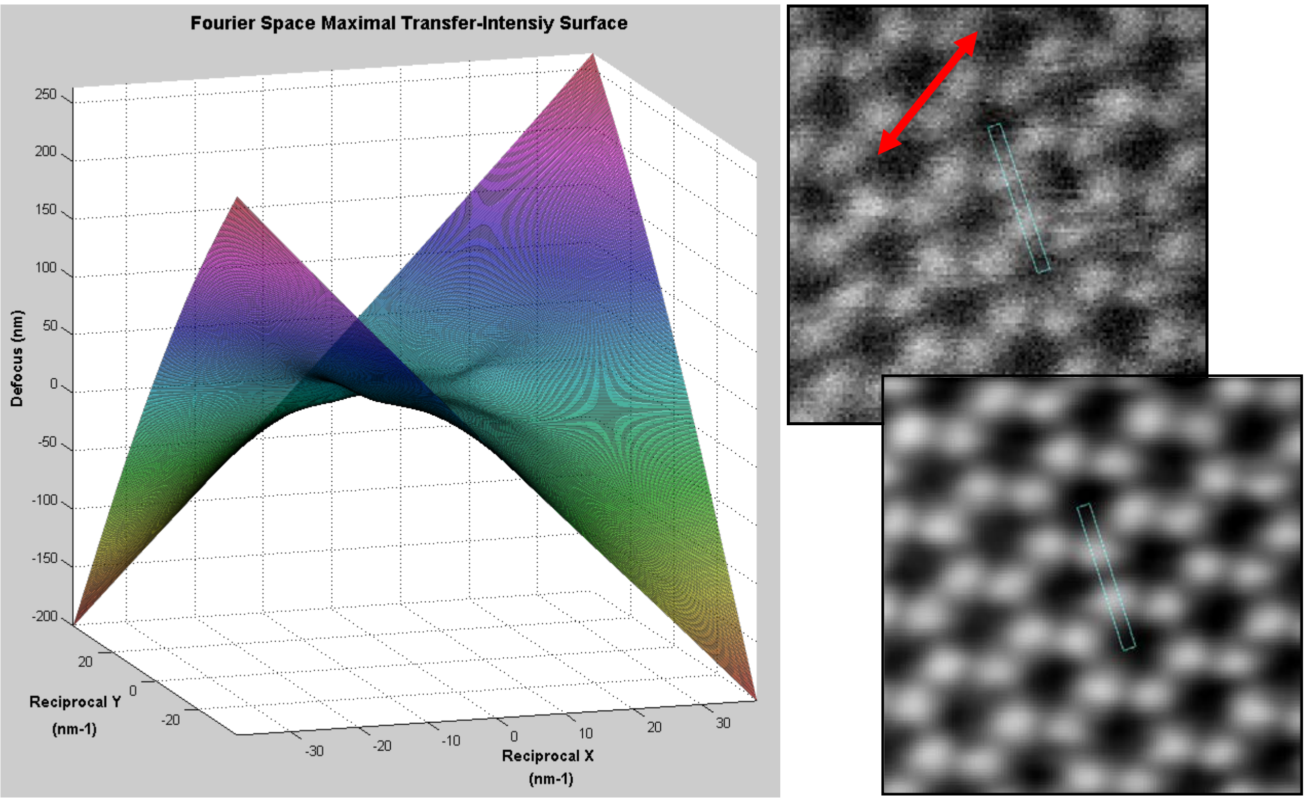

***Figure 1.*** *Left: Experimentally determined maximum-intensity defocus surface indicating the presence of two-fold astigmatism (orientation ≈45°). Right: enlargements from example HAADF raw data ($MoS_2$) and the same area after restoration, red arrow indicates same astigmatism orientation. Images show a field of view of 15Å.*

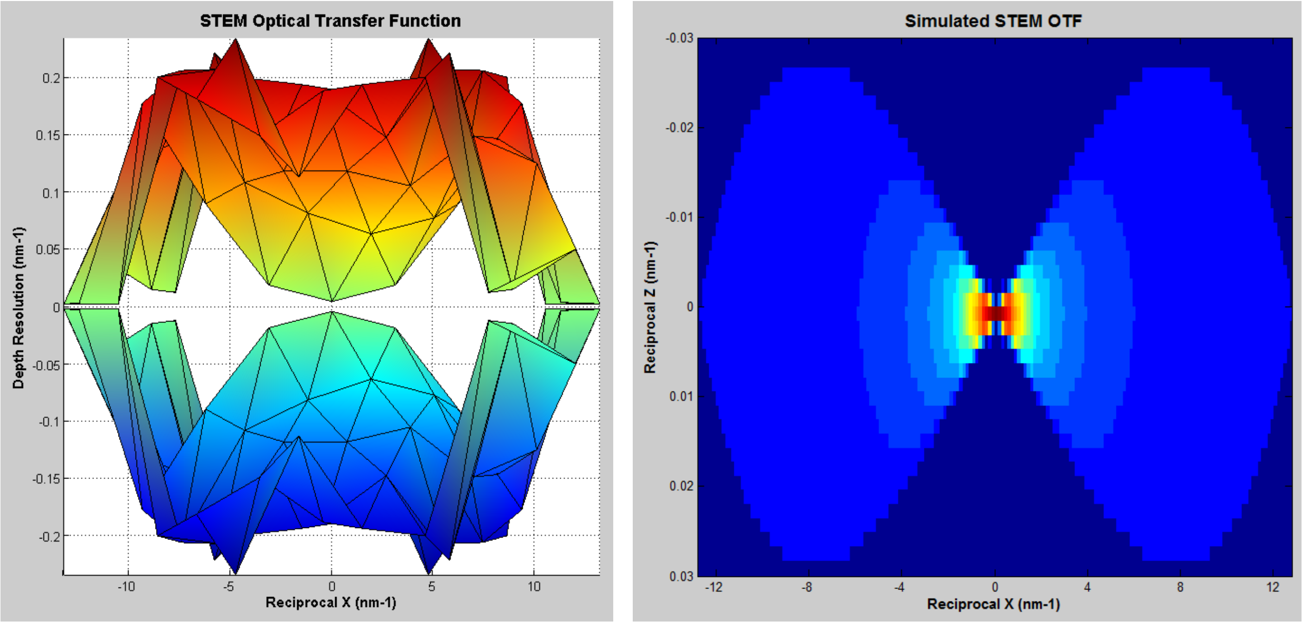

***Figure 2.*** *Left: View through toroidal surface of an experimentally determined STEM three-dimensional optical transfer function (OTF). Right: A two-dimensional section (x-z plane) through the analogous simulated OTF.*