Quantitative Measurement of Residual Aberrations in the Aberration-corrected STEM

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Background

In the scanning transmission electron microscope (STEM), aberration corrected optics allow to use larger probe forming apertures. This can improve both the lateral-resolution and vertical-sensitivity of the instrument whilst also making more current available for analytical work. However using larger apertures may leave the user exposed to small remnant higher-order aberrations. It has also been shown, that large defocii can be used to detect high-spatial-frequency information in a sample but with the loss of the lower spatial frequencies [1,2]. Recently we have developed a method to measure residual probe aberrations from focal-series and to restore a single image compensating for these defects producing a higher-resolution and SNR image.

Measuring Remnant Aberrations

To perform any sort of reconstruction first the raw image data must be recorded. As the effect of remnant probe aberrations is to cause the image contrast to peak at slightly different defocii for different spatial frequencies, we must record a HAADF STEM focal-series as our staring point. Then every one of these images will have an equivalent Fourier transform, Figure 1.

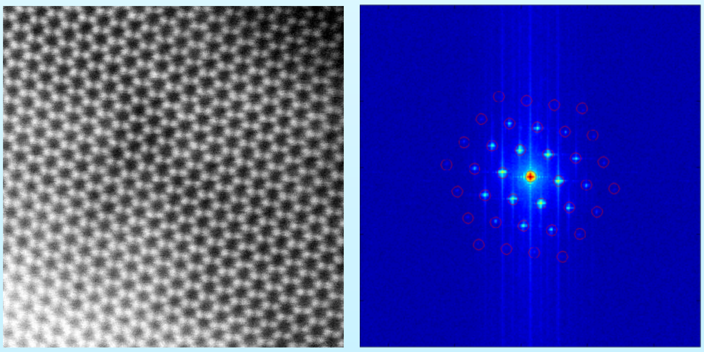

Figure 1. Median HAADF-STEM image from focal series and its accompanying Fourier transform

This new data-cube of Fourier transforms is then examined for bright spots corresponding to the crystal spacings in the sample. These spots come to a peak intensity at measurably different defocus values. Plotting the defocus of the peak information-transfer as a function of each 2D spatial-frequency then gives the below defocus versus spatial-frequency surface, Figure 2:

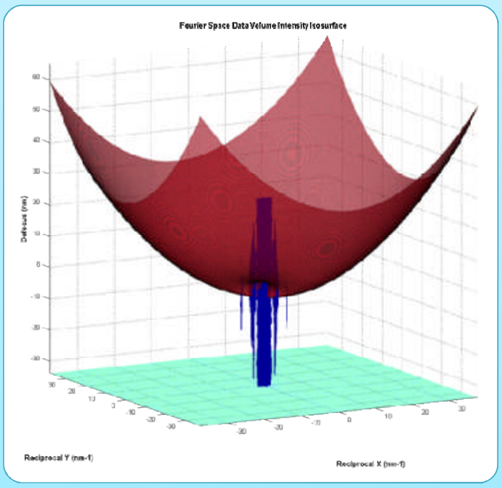

Figure 2. Representation of Fourier-space data cube. Blue vertical rods correspond to an iso-surface of intensity 1000 times brighter than the FT background and highlight the persistence of FT spots over several nanometres of defocus. Fitted surface shows the determined spherical aberration and 2-fold astigmatism present in the data volume.

Calculating Aberration Coefficients & Reconstructing an Aberration Free Image

Studying the shape of the contrast-defocus surface opposite, we can then fit aberration coefficients in a one of the conventional notations. In this way STEM focal series can be used directly an another form of aberration measurement. The example data set here was dominated by two-fold astigmatism, $C_{1,2}$, and spherical aberration, $C_{3,0}$.

Once the defocus needed for peak contrast transfer is know an image can be reconstructed by simply populating a blank 2D Fourier transform with the corresponding values from the Fourier-space data cube. Once populated this can be filtered if required (say to remove single pixel-shot noise) and then it's inverse calculated. What is returned is a 2D real-space image with all spatial frequencies transferred as strongly as possible.

Sample Material & Image Quantification

The material image was a thin flake of MoS_2, and the expected structure is shown overlaid on Figure 3 below. The stacking with molybdenum above sulphur and viva-versa then leaves all the atomic columns of roughly the same brightness. This material has for many years been widely used as a lubricant but has recently attracted interest as a potential transistor material once exfoliated down to a single layer[3].

The raw image from the centre of the data cube (defocus = 0nm) is shown below and the reconstruction to its right. Line profiles were taken from each and image performance quantified. The reconstructed has a 9.9% improved resolution, 73.8% larger inter-peak dip and a 205% increased SNR.

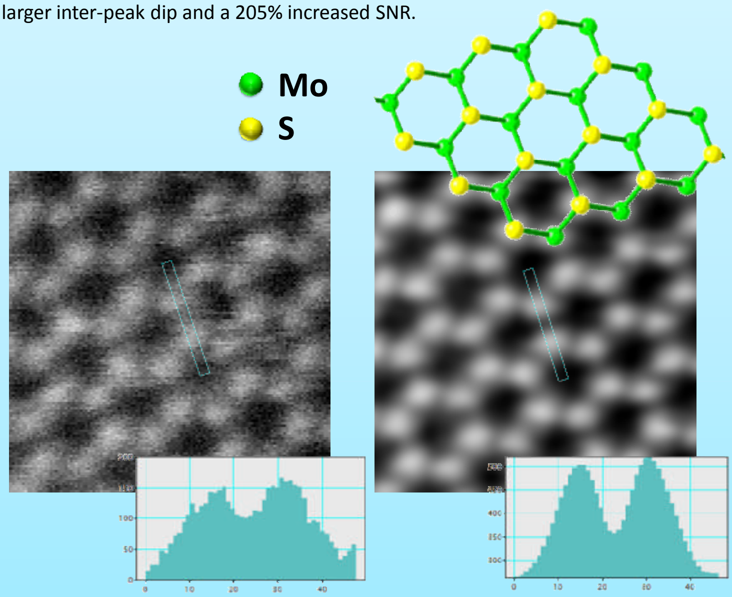

Figure 3. Enlargements and atom-pair line-profiles from the raw data (left) and reconstructed image (right). Field of view is ≈ 12Å.

Conclusions

- Atomic resolution ADF-STEM images recorded over a range of focus can exhibit some remnant astigmatism or other aberrations.
- Analysing the images in frequency space allows these aberrations to be identified and parameterised.
- The measured aberrations can be used to reconstruct an aberration compensated image with improved resolution and SNR.

References

1) Loane, R., Xu, P. & Silcox, J. *Ultramicroscopy* **40**, (1992).
2) Nellist, P.D. & Pennycook, S.J. *Physical Review Letters* **81**, (1998).
3) Schwierz, F. *Nature Nanotechnology* **6**, 135-136 (2011).

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Further Information

Copies of this poster, the associated manuscript and author contact details are available online at: www.lewysjones.com