Getting the Best from an Imperfect Detector:

an Alternative Normalisation Procedure for Quantitative ADF STEM

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Background

Quantitative annular dark-field scanning transmission electron microscopy (ADF STEM), where the recorded image data is normalised into units of 'fractional beam-current' is becoming increasingly popular [1]. This normalisation simply requires the vacuum or 'D.C.' level to be subtracted, followed by division by the detector sensitivity (Equation 1). This technique is extremely useful as it allows the direct comparison of experimental and simulated data as now both are expressed on identical absolute scales [2]. For the normalised results to be reliable it is essential to have an accurate measure of the detector's efficiency, but these can vary significantly [3]. At present the detector efficiency profile, such as in Figure 1 d), must be fed into the image simulations This greatly reduces simulation software options available and can make it difficult to validate results between different codes. Additionally, any simulated library values are detector specific and must be recalculated if the microscope setup is changed. This requirement is at best time-consuming and at worst can cause discrepancies when collaborating with, or verifying the findings of, others. Here we present an alternative 'electron-flux weighted' method to calculate the effective detector sensitivity. This allows simulated library values to be calculated that are only dependent on the detector's inner and outer angles.

$$I_{norm} = \frac{I_{exp} - I_{vac}}{I_{det} - I_{vac}}$$

Equation 1. General form of the quantitative ADF normalisation equation where vacuum level is subtracted before detector sensitivity normalisation.

Origins of Detector Asymmetry

In the ideal case ADF detectors would be perfect, they would be totally circular and have a homogenous sensitivity response. In reality, detectors are observed to be highly asymmetric with some regions being half as efficient as others in detecting electrons [3]. In the majority of cases this asymmetry is caused by the 'shadow' of the hole through the centre of the detector. This hole is sometimes lined with an inactive liner-tube but in all cases causes the region of the detector on its far side to behave less efficiently. Averaging this behaviour azimuthally hides much of this but still many detectors show a reduced efficiency at low angles right where the electron flux is highest.

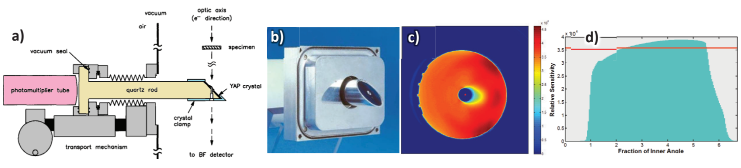

Figure 1. Origins of detector asymmetry; a) schematic of an example ADF detector geometry, b) photograph of the Fischione model-3000 ADF detector, c) the measured detector response and d) the azimuthally-averaged radial sensitivity profile in arbitrary counts units.

The Electron Flux Weighted (EFW) Method

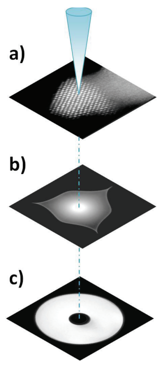

Figure 2. The three data inputs needed for the EFW normalisation method: a) the experimental ADF image, b) the electron-flux distribution (either experimental, deduced from camera-length series or image simulation), and c) the detector sensitivity scan.

Using the EFW method the user simulates reference data assuming a 'perfect' detector (uniform sensitivity). The detector is then only defined by its inner and outer angles (i.e. camera-length) and is reproducible across different instruments. The experimental data is normalised using an 'effective sensitivity' for the detector. This describes the average efficiency expressed as a single number. Unlike previous quantitative ADF work [1], we now use three inputs to analyse the data (Figure 1). To calculate the effective sensitivity we first perform an experimental detector sensitivity scan such as Figure 1 c) from an FEI Titan or Figure 3 b) from a JEOL ARM. Next a two-dimensional flux-distribution is recorded using a CCD as close as possible to the same plane as the ADF detector, Figure 2 b). If this is unavailable the same data can be obtained from a simulated PACBED pattern or from camera-length series.

Next, the flux-map is converted to cylindrical-coordinates and averaged azimuthally to yield the radial scattering profile, Figure 3 a). Using this, the section of the profile between the detector inner and outer angles is fitted to a power-law. We now have a mathematical description of the flux shape and bounds and can generate a mask for the detector sensitivity map, Figure 3 d). This mask is normalised so that it integrates to one before being multiplied across the detector efficiency scan, Figure 3 e). Integrating the 'flux-weighted detector map' we find the effective detector sensitivity suitable for use in Equation 1. Accounting for the detector asymmetry in this way means that the reference library of simulations can be performed for a 'perfect' detector and can be shared with anyone using the same inner and outer angles – the operator must simply record their own flux-map and efficiency scan. Being able to share simulation data in this way greatly improves productivity and aids collaboration with other labs.

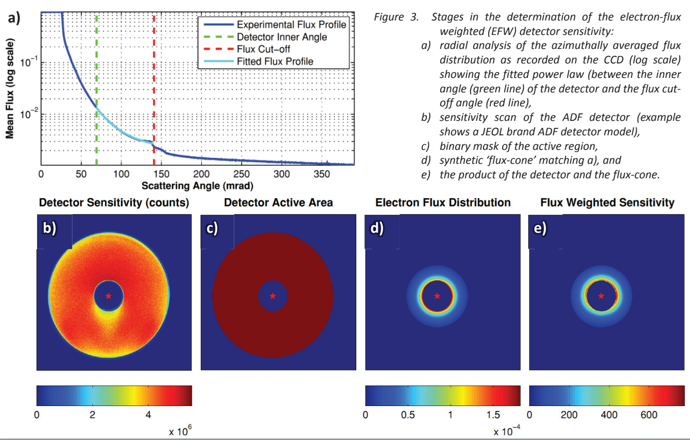

Figure 3. Stages in the determination of the electron-flux weighted (EFW) detector sensitivity:
a) radial analysis of the azimuthally averaged flux distribution as recorded on the CCD (log scale) showing the fitted power law (between the inner angle (green line) of the detector and the flux cut-off angle (red line),
b) sensitivity scan of the ADF detector (example shows a JEOL brand ADF detector model),
c) binary mask of the active region,
d) synthetic 'flux-cone' matching a), and
e) the product of the detector and the flux-cone.

Experimental Testing & Validation

As with all new methods, extensive testing is required. To benchmark this new method we compare it with the current literature practice [1]. To achieve this an experimental data-set was recorded from a platinum wedge in a [110] orientation, Figure 4 a). This was analysed using the in-house developed ***'Absolute Integrator'*** software which performs the image peak-finding, data normalisation and atomic-column integration tasks. Both the conventional and our new EFW method are available as choices in the software and both approaches were followed for this comparison. In parallel with the experimental analysis image simulations were performed using the two approaches of imperfect and perfect detector simulations respectively [4]. These two experimental quantifications with their accompanying simulations were used to count the number of atoms in every visible atomic-column in the image. To provide an independent count of the number of atoms in each column a ICL / Gaussian mixture model fit was used [5].

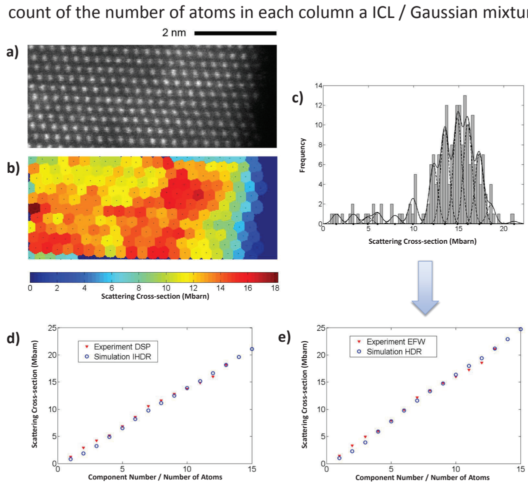

Figure 4. Experimental validation of the proposed EFW detector sensitivity calibration method: a) experimental ADF image of a pure platinum wedge sample in [110] orientation, b) visualisation of the quantitative scattering cross-sections extracted from the experimental data, c) histogram representation of the values from b) with fitted Gaussian mixture model overlaid, d) & e) comparison between the experimental (red) and simulated (blue) values for the DSP (flat-detector / weighted-simulation) and EFW (weighted-detector / flat-simulation) methods respectively.

Figure 4 c) shows the fitted GMM used for the atom counting procedure which were used to compare the quality of the quantification results using both the current, Figure 4 d) method and the proposed EFW method, Figure 4 e). Both methods yield identically accurate quantification performance confirming that the EFW method is valid. Additional tests are also presented in the accompanying full length paper [6].

Conclusions

The current practice for ADF quantification requires time-consuming image simulations to be performed for every different detector and microscope used. These simulations often have to be performed using particular codes which can make comparative studies over time or between groups more difficult or lead to discrepancies. Here we introduce a new 'electron-flux weighted' (EFW) method which allows simulated library data to be calculated, using any simulation code of choice, that is only dependent on the detector's inner and outer angles.

Tests of this method show that it is precisely as accurate as the current practice while also offering significant time savings. This new EFW method is available as an option within the ***'Absolute Integrator'*** ADF quantification software which is available free of charge for academic / non-commercial use from **www.lewysjones.com** .

References

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