Depth sectioning using electron energy loss spectroscopy

A J D’Alfonso\textsuperscript{1}, E C Cosgriff\textsuperscript{2}, S D Findlay\textsuperscript{1}, A I Kirkland\textsuperscript{2}, P D Nellist\textsuperscript{2}, M P Oxley\textsuperscript{3} and L J Allen\textsuperscript{1}

\textsuperscript{1} School of Physics, University of Melbourne, Victoria 3010, Australia
\textsuperscript{2} Department of Materials, University of Oxford, Oxford OX1 3PH, United Kingdom
\textsuperscript{3} Materials Science and Technology Division, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, TN 37831, USA

E-mail: adalf@phys.unimelb.edu.au

Abstract. The continued development of electron probe aberration correctors for scanning transmission electron microscopy has enabled finer electron probes, allowing atomic resolution column-by-column electron energy loss spectroscopy. Finer electron probes have also led to a decrease in the probe depth of focus, facilitating optical slicing or depth sectioning of samples. The inclusion of post specimen aberration corrected image forming lenses allows for scanning confocal electron microscopy with further improved depth resolution and selectivity. We show that in both scanning transmission electron microscopy and scanning confocal electron microscopy geometries, by performing a three dimensional raster scan through a specimen and detecting electrons scattered with a characteristic energy loss, it will be possible to determine the location of isolated impurities embedded within the bulk.

1. Introduction
The development of scanning transmission electron microscopy (STEM) aberration-correcting electron optics resulted in sub-Ångström probes\textsuperscript{[1]}. Smaller probe size increases the transverse resolution of STEM while simultaneously reducing the depth of focus. Atomic resolution STEM imaging offers the tantalising possibility to depth section samples and so to extract three dimensional atomic locations\textsuperscript{[2, 3, 4]}.

Depth sectioning involves recording a series of through focus images which are then used to reconstruct a three dimensional model of the specimen. Depth sectioning in STEM using high angle annular dark field (HAADF) imaging has been experimentally realised\textsuperscript{[4]}. As the contrast in HAADF imaging scales approximately with the square of the target’s atomic number, an impurity must have a significantly larger atomic number than the supporting crystal for it to be detectable. Using electron energy loss spectroscopy (EELS) as an adjunct to the HAADF experiment should alleviate this condition and locate impurities with a similar atomic number to their surroundings inside the specimen\textsuperscript{[5]}.

Scanning confocal electron microscopy (SCEM), an electron microscope analogue of confocal scanning optical microscopy, can be used in conjunction with aberration correction to further reduce the probe depth of focus. SCEM differs from conventional STEM through the inclusion of an imaging lens post specimen. If no specimen is present, this lens creates a real-space image of the focussed probe at the image plane. Including a pinhole aperture in the image.
plane further reduces out of focus contributions. Nellist et al. [6] recently achieved the confocal arrangement of an aberration corrected JEOL 2200MCO, operating with a transverse full width at half maximum of 1.7 Å and an experimental depth resolution of 35 Å.

The schematic of the depth sectioning process using STEM and SCEM is shown in Fig. 1. The intention is that changing the probe defocus shifts the depth of the beam waist (which contains the significant proportion of the probe intensity), thus imaging a specific depth inside the crystal. Ideally a direct correlation of depth with defocus would make image interpretability straight forward, however the channelling conditions of the crystal and the coherent nature of the inelastic scattering complicate matters somewhat. HAADF imaging is effectively incoherent due to the large detectors used, allowing the direct correlation of the probe intensity to the recorded signal. EELS imaging is typically not totally incoherent. However, just like HAADF, in the limit that the EELS detector subtends a large solid angle, partial coherence effects average out. Large detectors allow a local interpretation, correlating probe intensity with signal strength. Energy filtered SCEM is similar as the coherent optics post specimen will make the coherent nature of the inelastic scattering manifest strongly in the image. This will affect the extent to which we correlate probe position and image intensity with image source. Using large probe forming and imaging apertures with a small pinhole aperture at the detector ensures that the significant image contributions come from inelastic waves which originate at the focal point of the imaging system.

Models of STEM EELS and energy filtered SCEM must take into account the delocalisation of the transition potential. The extent of the transition potential delocalisation can be viewed (albeit carefully) as being a function of the binding energy and the orbital occupied by the bound electron. The transition potentials \( H_{n0} \) for the ionization of an Al K-shell electron \( l = 1, m = 0 \) to the \( l' = 1, m' = 0 \) and \( l' = 1, m' = 1 \) continuum states are shown in Fig. 2. The spatial extent of these transition potentials and how they couple with the probe determine a significant proportion of the features observed in simulations.

2. Case study Al in \( \langle 001 \rangle \) zone axis GaAs
Our simulations used a real-space multisillice formulation, including elastic scattering both pre and post the inelastic scattering event. The inelastic electron wave function resulting from the ionization interaction is

\[
\psi_n(r_\perp, z) \propto H_{n0}(r_\perp)\psi_0(r_\perp, z),
\]

where the elastic wave function \( \psi_0 \) at the depth \( z \) is multiplied by the projected transition potential \( H_{n0} \) for transitions to the \( n \)th excited state [7]. The 100 keV STEM probe forming aperture semi-angle was 30 mrad. The 100 keV SCEM probe and image forming aperture semi-angles were also set to 30 mrad allowing a direct comparison. The only aberration in all lenses is...
Figure 2. Al K-shell transition potentials to final continuum states (a) \( l' = 1, m' = 0 \) and (b) \( l' = 1, m' = 1 \) at 1 eV above ionization threshold. 200 keV incident energy assumed.

was controllable defocus which is used to effect the depth sectioning in the simulations. The 204 Å thick GaAs crystal was oriented along the \( \langle 100 \rangle \) zone axis. A single Al dopant is embedded 101 Å deep, substitutionally displacing a Ga atom on column. Figure 3 shows a series of Al K-shell STEM EELS and energy filtered SCEM line scans as a function of probe position and defocus (over-focus positive) with negative defocus implying the probe is focused into the crystal. A total of 25 different final states 1 eV above ionisation threshold were used in calculations. Each mesh plot in Fig. 3 has a solid black line indicating the depth of the impurity and a corresponding 2D greyscale plot to clearly show the important features of the through focus images. The 2D greyscale plots are symmetric; the mesh plots only show half of the scan.

Comparing the results for three different STEM detector sizes, Figs. 3(a)–(c), a few features warrant comment. The STEM images all exhibit strong channelling effects, manifest in the small peak occurring at zero defocus. This is a consequence of the probe coupling to the dynamical Ga column 1s state at the entrance surface of the specimen and subsequently channelling along the column to the depth of the impurity. The effect of 1s channelling is most pronounced for the smallest 10 mrad detector. The pre-focussing effect of the atomic column, the peak occurring for a larger defocus value than the impurity depth, is also seen in all of the STEM images. Interestingly, this is most prominent for the largest, 50 mrad detector. By contrast, the SCEM image, Fig. 3(d), has no pre-focussing effect: the peak signal in the SCEM image is located at the impurity depth and is symmetric for the probe defocused both before and after the impurity.

The other notable feature is the absence of volcanoes for the 50 mrad STEM detector and the SCEM images. The image formed by the smallest STEM detector, which is the most likely to demonstrate the partial coherence of the inelastic waves, returns the most delocalised image and shows the largest volcanoes. The intermediate STEM detector size of 30 mrad gives moderate image delocalisation and a smaller volcano. This is unexpected as we are operating in a regime where for a single atom a local interpretation is valid (the collector and probe forming apertures span the same semi-angle) and volcanoes should not be present [8]. The dynamical evolution of the probe inside the specimen makes image interpretation difficult.

The SCEM geometry has the narrowest depth of focus. This is a favorable consequence of the pinhole aperture in the image plane. By comparison, in the equivalent STEM geometry, where the collection semi-angle is 30 mrad, the depth resolution is quite poor. Imposing the pinhole aperture increases the depth resolution of the image at the expense of signal intensity.

3. Conclusion
We have shown that it is possible to depth section samples using core-loss spectroscopy in STEM when aligned along a zone axis. The image depth of field depends on the probe size, the depth of focus and the detector collection angle. The latter point is subtle but states that the degree of partial coherence of the inelastic electron is crucial to the shapes of STEM images both as a function of probe position and defocus. Increasing the detector collection angle localises the
Figure 3. Al K-shell EELS line scans as a function of defocus for a STEM detector collection semi-angle of (a) 10 mrad, (b) 30 mrad and (c) 50 mrad, and in (d) a symmetric SCEM geometry image. The Al impurity is placed 101 Å deep, substitutionally displacing a Ga atom on the column.

signal toward the dopant depth and removes the volcano structure. Finally, we have shown that energy filtered SCEM allows greater depth resolution than STEM and will possibly allow 3D specimen mapping.

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