Three-dimensional imaging using aberration-corrected scanning transmission and confocal electron microscopy

E C Cosgriff¹, A J D’Alfonso², L J Allen², S D Findlay², A I Kirkland¹ and P D Nellist¹

¹Department of Materials, University of Oxford, Oxford OX1 3PH, United Kingdom
²School of Physics, University of Melbourne, Victoria 3010, Australia
E-mail: eireann.cosgriff@materials.ox.ac.uk

Abstract. A reduction in the focal depth of field as a result of the installation of aberration correctors in scanning transmission electron microscopy, allows three-dimensional information to be retrieved by optical depth sectioning. A three-dimensional representation of the specimen is achieved by recording a series of images over a range of focal values. Optical depth sectioning in zone-axis crystals is explored computationally using a Bloch wave analysis to explain the form of the electron intensity in the crystal as a function of depth. We find that the intensity maximum deviates from that of the expected defocus value due to pre-focusing by the atomic column and also due to channelling pendellosung. The possibility of performing bright-field imaging in a double corrected two lens system in a confocal arrangement is also investigated computationally. The method offers some advantages over depth sectioning using conventional transmission electron microscopy.

1. Introduction

The recent dramatic improvement in the resolution of scanning transmission electron microscopy (STEM) images is due mainly to the increase of the numerical aperture of the objective lens made possible by the development of spherical aberration correctors. The lateral resolution is inversely proportional to the aperture size, while the depth of field is inversely proportional to the square of the aperture size. Expressing the depth of field as the full-width at half-maximum (FWHM) of the STEM probe intensity along the optic axis plotted as a function of axial position \( z \) it can be written as

\[
\Delta z = 1.7 \frac{\lambda}{\alpha^2},
\]

where \( \lambda \) is the electron wavelength and \( \alpha \) is the aperture semi-angle. For a 300 kV probe and a 22 mrad semi-angle aperture, the depth of field is 7.0 nm. Taking advantage of this reduction in the depth of field, a three-dimensional representation of the specimen can be achieved by recording a series of images over a range of focal values. Depth resolutions of a few nanometers have recently been demonstrated for heavy atoms dispersed in lighter supports [1].

In light optics it is known that it is possible to reduce the depth of field further by the use of a confocal geometry, shown in Fig. 1(a). In electron optics this can be achieved using a second aberration-corrected lens system placed post-specimen. The Oxford JEOL 2200MCO
has been fitted with both pre-specimen and post-specimen spherical aberration correctors and recently the establishment of a confocal regime was demonstrated [2]. An atomic-scale electron probe formed using aberration-corrected optics is scanned across and through the sample in a three-dimensional raster. Contributions from scattering away from the focal plane of the probe are reduced by the application of a second post-specimen aberration-corrected system confocal to the illumination optics, which images the confocal plane onto an aperture in the detector plane. Here we assume that only zero-loss electrons contribute to the image (possible through the use of an energy filter) and hence neglect the effect of any chromatic aberrations present in the post-specimen lens. There are two possible imaging modes available for the configuration, involving elastic and inelastically scattered electrons.

A question of interest for depth sectioning in conventional STEM and scanning confocal electron microscopy is whether these results can be obtained for stronger scattering matrices such as crystal aligned along a zone-axis. In general for STEM, dynamical diffraction effects are important and channelling of the probe is advantageous for image interpretation. Whereas for optical sectioning the probe should converge to a small cross over and broaden again. A situation which seems irreconcilable with channelling conditions.

2. Depth sectioning in zone-axis crystals

Bloch wave calculations were performed for a 20 nm thick \(\langle 110 \rangle\) GaAs crystal (Fig. 1(b)). We assume an aberration-free 300 kV probe and 22 mrad probe-forming aperture. We also include an optical absorption potential [3] with Debye-Waller factors taken from Ref. [4]. The intensity of the centre of the probe at zero thickness and zero defocus is normalised to unity. Figure 2 is normalised in this way.

We begin by considering depth sectioning for the case of shallow sectioning. The intensity immediately below the probe was calculated as a function of depth for two probe positions, directly above an As column and a point away from all projected columns (see Fig. 1(b)), when the probe is underfocused by 5.0 nm. These plots are shown in Fig. 2(a). We see that when the probe is located away from an atomic column (solid line), the intensity-depth graph shows a profile that would be expected for a probe propagating in vacuum. It peaks at 5.0 nm depth, and has a FWHM of 7.0 nm along the optic axis. However, when the probe is moved over to the As column, the intensity maximises prematurely at a depth of only 3.0 nm (rather than the intended 5.0 nm). The depth FWHM of the first peak reduces to 4.0 nm.

Bloch wave analysis shows that there is an angle-dependent phase-shift which pre-focuses the beam by about 2.0 nm along the atomic column: the atomic column is acting as a lens [5]. This effect arises from all of the excited states, including high-angle ones. For deeper sectioning, where
channelling conditions are present, more complicated behaviour is observed. Figure 2(b) shows the intensity along the As column (dotted line) when the probe is underfocused by 13.0 nm and the intensity one might expect from a free space probe underfocused by 11.0 nm, thus taking into account the 2.0 nm pre-focusing. Here, the 1s state is important and the interference between this and the non-1s states superimposes a depth oscillation on the pre-focused peak when on the As column. We see that, for this case, the interference has resulted in the development of two peaks either side of the main pre-focusing.

3. Scanning confocal electron microscopy
To explore the likely image contrast observed when depth sectioning in the bright-field scanning confocal electron microscopy configuration, multislice calculations were performed using parameters based on those demonstrated for the Oxford machine [2]: an incident energy of 200 kV and nominal aperture sizes of 30 mrad, assuming an aberration-free probe. We looked at trying to locate a buried impurity atom inside 30 nm of ⟨110⟩ GaAs. Figure 3(a) shows the confocal signal for pure GaAs and the confocal signal when the substitutional In atom (in the Ga position) is located at three different depths. A negative value for the probe defocus indicates an underfocused probe and zero indicates the probe is focused on the surface. Thus negative values correspond to depths within the sample. The post-specimen lens is always focused on the same plane as that of the illumination optics. Immediately clear is that the confocal arrangement gives a very good estimate of the sample thickness. The intensity drops quite sharply at both surfaces of the crystal. We can also see reductions in the intensity at the approximate depths of the impurity atom (5 nm, 10 nm and 15 nm). The In atom at 5 nm and 10 nm causes the signal to decrease slightly at that depth, while the In atom located in the midplane of the crystal serves to flatten the central peak.

How well does this compare to conventional transmission electron microscopy? Figure 3(b) shows the equivalent case for plane wave illumination and a 30 mrad imaging lens aperture. The In atom at any depth would be very hard to locate using this kind bright-field imaging as the signal is ambiguous. Unlike the confocal system, the change in signal from the impurity atom is not localised to its depth.
Figure 3. (a) Intensity as a function of probe defocus for 30 nm thick ⟨110⟩ GaAs (solid line) and 30 nm thick ⟨110⟩ GaAs with a single In atom at depths of 5 nm, 10 nm and 15 nm using a 30 mrad confocal system. (b) As per (a) excepting illumination is by conventional transmission electron microscopy.

4. Conclusions
We have shown that the intensity as a function of depth in an atomic column, when illuminated by an underfocused STEM probe, is dependent upon two main processes. The potential of the atomic column acts like a lens to pre-focus the probe. This process involves all states and is not purely due to channelling. Superimposed on this effect is the shorter period oscillation in intensity due to the channelling 1s state. The interference of the 1s state with the rest of the wavefunction creates the form of the intensity we see along the column. In certain cases this acts to move the peak intensity away from the intended depth, or gives rise to depths in the sample at which it is impossible to form a maximum in the illuminating probe. Our preliminary study of scanning confocal electron microscopy has suggested that an accurate determination of sample thickness can be obtained with the method. It also offers some advantages over conventional electron microscopy for the depth determination of impurity atoms as the change in signal is localised to the impurity depth.

Acknowledgments
E C Cosgriff and P D Nellist acknowledge financial support from Trinity College Dublin and the Irish Higher Education Authority (HEA) through the Programme for Research in Third Level Institutions (PRTLI). A J D’Alfonso acknowledges support of the Australian Microscopy and Microanalysis Society and the assistance of the Postgraduate Overseas Research Experience Scholarship scheme. L J Allen acknowledges support by the Australian Research Council. A I Kirkland and P D Nellist acknowledge funding from The University of Oxford, JEOL Ltd and the EPSRC.

References
[1] Borisevich A Y, Lupini A R and Pennycook S J 2006 Proc. Natl. Acad. Sci. 103 3044-3048
[2] Nellist P D, Behan G, Kirkland A I and Hetherington C J D 2006 Appl. Phys. Lett. 89 124105
[3] Bird D M and King Q A 1990 Acta Cryst. A46 202-208
[4] Boothroyd C B 2002 J. Electron Microsoc. 51 S279-S287
[5] Cosgriff E C and Nellist P D 2007 Ultramicroscopy 107 626-634