

## Atomic-Resolution EELS in Aberration-Corrected STEM

S. D. Findlay,\* M. P. Oxley,\* L. J. Allen,\* A. R. Lupini,\*\* and S. J. Pennycook,\*\*

\* School of Physics, University of Melbourne, Victoria 3010, Australia.

\*\* Condensed Matter Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831-6030 USA.

Core loss electron energy loss spectroscopy (EELS) has become a powerful tool for determining structure/property relationships at interfaces and grain boundaries, and for probing local stoichiometry, impurity segregation and electronic structure. When acquired simultaneously with an atomic-resolution *Z*-contrast image in STEM, atomic-column resolution has been demonstrated [1,2]. Two issues are important in this context, firstly, the “localization” of the ionization interaction, and secondly, the role of dynamical diffraction. Quantum mechanical calculations of delocalization have been inconsistent, some predicting significant delocalization [3,4] and others strong localization [5,6]. Here we simulate STEM images of single atoms, demonstrating that the “width” of the STEM image is limited by the “width” of the probe rather than by the nature of the ionization interaction, for the current generation of aberration correctors. We also report a comparison of experiment with theoretical simulations to show empirically that the ionization interaction is indeed sufficiently “localized” and, furthermore, that under strong channeling conditions atomic-column resolution is feasible, though interpretation issues arise.

Simulations of inelastic STEM images use nonlocal mixed dynamic form factors (MDFF) [7]. Figure 1 shows Ti *L*-shell and O *K*-shell EELS STEM line scans of isolated Ti and O atoms respectively, for an accelerating voltage of 100 kV, a detector semi-angle of 20 mrad and a 40 eV energy window above threshold. Three probes are used. The first is aberration-free, with a  $1.0 \text{ \AA}^{-1}$  cutoff. The second is aberration-balanced, with  $C_s = -0.05 \text{ mm}$ ,  $C_5 = 63 \text{ mm}$ ,  $\Delta f = 62 \text{ \AA}$ , and a  $0.539 \text{ \AA}^{-1}$  cutoff. The third has  $C_s = 0.5 \text{ mm}$  and Scherzer conditions. It is seen that the full width at half maximum (FWHM) of the image is only slightly broadened from the probe FWHM (see caption), indicating little ionization delocalization. The dip in the middle of the line scans for the finer probes has been explained elsewhere in the literature [4].

Figure 2 shows experimental line scans for *Z*-contrast and Ti *L*-shell EELS in SrTiO<sub>3</sub>, scaled by their average value for convenience of display. The ADF signal clearly resolves the structure of SrTiO<sub>3</sub> (cubic, at room temperature, with side length 3.9 Å). The bright peaks are the Sr columns, the weaker peaks are the Ti/O columns. Image simulations bear this out; in no parameter set investigated was the ratio between the columns reversed. Taking the peaks of the *Z*-contrast signal as indicative of these columns, it may be seen that the EELS signal does indeed give peaks corresponding to the Ti/O column positions. The fluctuation in the signals, and the distinct variation between the two adjacent cells, may be attributed to physical instability in the microscope and the presence of amorphous surface layers.

Figure 3 shows the Ti *L*-shell EELS STEM image for each of the probes described. In each case the crystal thickness is 200 Å. The experimental data is also shown for comparison. It is clear that in this case the atomic column resolution is maintained in the presence of dynamical diffraction.

## References:

- [1] N. D. Browning, M. F. Chisholm and S. J. Pennycook, *Nature* 366 (1993) 143.  
 [2] G. Duscher, N. D. Browning and S. J. Pennycook, *Phys. Stat. Sol. (a)* 166 (1998) 327.  
 [3] D. A. Muller and J. Silcox, *Ultramicroscopy* 59 (1995) 195.  
 [4] H. Kohl and H. Rose, *Adv. Imaging and Electron Phys.* 65 (1985) 173.  
 [5] B. Rafferty and S. J. Pennycook, *Ultramicroscopy* 78 (1999) 141.  
 [6] O. F. Holbrook and D. M. Bird, *Proc. MSA* 147 (1995) 175.  
 [7] V. W. Maslen and C. J. Rossouw, *Phil. Mag. A*49 (1984) 735, C. J. Rossouw and V. W. Maslen, *Phil. Mag. A*49 (1984) 743, L. J. Allen and T. W. Josefsson, *Phys. Rev. B* 52 (1995) 3184.  
 [8] This work was supported by the USDOE under contract DE-AC05-00OR22725 managed by UT-Battelle, LLC.

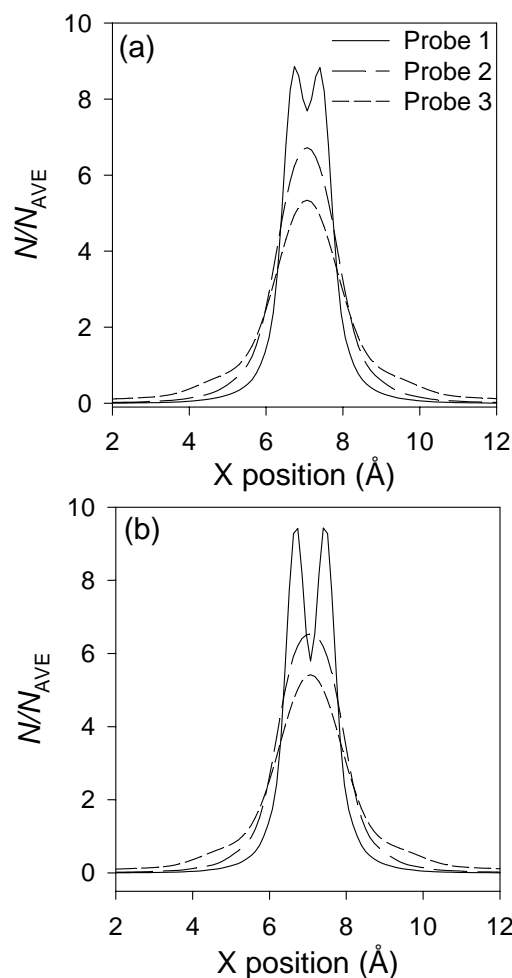


FIG. 1: Calculations of a) Ti *L*-shell EELS STEM image of a single, isolated Ti atom; b) O *K*-shell EELS STEM image of a single, isolated O atom. Probe 1 has a FWHM of 0.5 Å, probe 2 a FWHM of 1.0 Å, and probe 3 a FWHM of 1.3 Å.

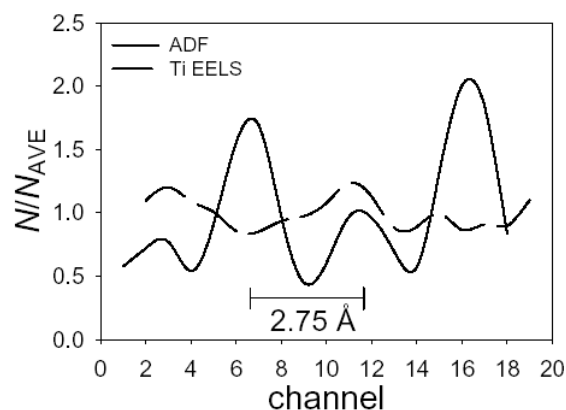


FIG. 2: Experimental data plots showing the ADF (Z-contrast) and Ti *L*-shell EELS results.

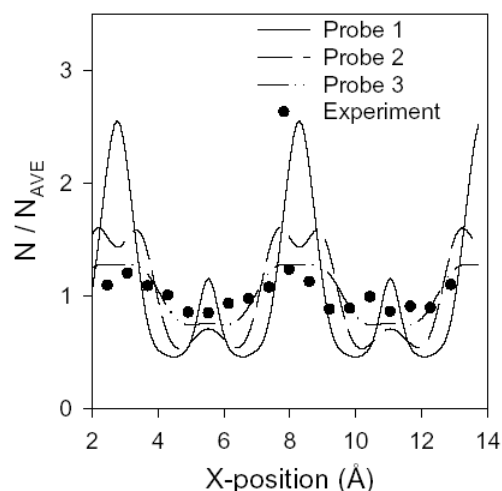


FIG. 3: Calculations of Ti *L*-shell EELS image in a full nonlocal calculation for a perfect crystal of thickness 200 Å for each of the probes described previously. The experimental data are included for comparison.