Workshop on Advanced Topics in Transmission Electron Microscopy

Demonstration: Image simulations using μSTEM

**μSTEM**

Executable and manual available at http://tcmp.ph.unimelb.edu.au/mustem/muSTEM.html

**Viewing output**

μSTEM outputs binary files with file name form *width*×*height*\_*description*.bin.

**Digital Micrograph**

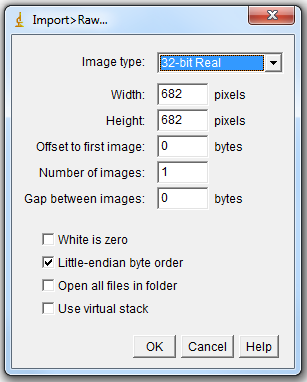
To view in Digital Micrograph:

* File → Import data…
* Select the “binary” radial button and tick “swap data bytes”
* Select “Real 4 byte” in the data type drop-down menu  
  (“Real 8 byte” if the double precision version of the code is used rather than the single precision version)
* Enter width and height in the width and height fields

**Image-J**

To view in ImageJ:

* File → Import → Raw…
* Select “32-bit Real” in the data type drop-down menu  
  (“64-bit Real” if the double precision version of the code is used rather than the single precision version)
* Enter width and height in the width and height fields
* Untick the “Little-endian” checkbox



**Program control file**

How the program executes is determined by the file *user\_input.txt*. If the first line reads:

* *interactive*: all inputs taken from the keyboard, but not saved
* *record*: all inputs taken from the keyboard, and saved to the text file specified by second line
* *record overwrite*: all inputs taken from the keyboard, and saved to the text file specified by second line. The existing text file is overwritten.
* *play*: all inputs taken from the text file specified by the second line
* *play all*: Plays all the text files listed in *user\_input.txt*

**Input structure file**

The crystal structure is taken from an \*.xtl file, which is in plain text format. Examples are included in the distribution. A detailed description is given in the manual.

**Examples and exercises**

Please work through the following activities.

STEM HAADF

**Getting started**: Play control file “STEM\_STO\_HAADF\_driver.txt” and view the image.

**Things to try**:

1. Vary some of the following parameters in the control file and check whether the qualitative and quantitative changes in the output images are consistent with your expectations:
   1. Thickness (Angstrom units, a string of the form 50:200:50 or 50,100,200 gives a thickness series)
   2. Aperture cut-off (mrad units; in particular, try using 2 mrad: what happens and why?)
   3. Probe defocus (Angstrom units, a string of the form -100:100:25 or -100,-50,0 gives a defocus series)
   4. Detector range (mrad units; control file calls this TDS inner and outer angle)

**Things to think about**:

1. The column peaks in your simulations are likely narrower than those you usually see in experiment. This is because these simulations are missing something (that can easily be added later). Can you tell what?
2. If you extract a suitable line scan you will likely find that small peaks are present at the oxygen columns. Do you expect this to be true in the experiment?

STEM PACBED

**Getting started**: Play control file “STEM\_Si110\_PACBED\_driver.txt” and view the output.

**Things to try**:

1. A Si110 PACBED recorded on MCEM’s Tecnai F20 microscope is included in the directory (*F20\_Si110\_PACBED.tif*) by comparison with the simulations what can we say about the crystal thickness?
2. Try changing the lens aberrations (Cs coefficient, mm units; C5 coefficient, mm units; defocus, Angstrom units). What affect does this have on the PACBED pattern?

**Things to think about**:

1. Is there an optimum aperture size for PACBED? What happens if the aperture is “too small”? Try running the simulation for a 24 mrad aperture at 300 keV, how easy is it to tell thicknesses apart now?

STEM CBED

**Getting started**: Play control file “STEM\_Al110\_CBED\_driver.txt” and view the output.

**Things to try**:

1. Simulate CBED patterns for a range of different thicknesses. To what precision do you think thickness could be gauged by comparison with such simulations?

**Things to think about**:

1. Find the supercell pixel size and tiling size in the control file, and compare with the values used when calculating HAADF images. What do you notice? What motivated these choices of values? (Hint: read the notes on sampling on the final page of this worksheet.)
2. Compare the use of PACBED and CBED for thickness estimation. What considerations might lead you to select one technique over the other?

STEM EDX

**Getting started**: Play control file “STEM\_STO\_EDX\_SrK\_driver.txt” and view the output.

Note: HAADF images will also be output. The EDX output has “Sr\_K\_shell” in the file name.

**Things to try**:

1. By editing the control file, run the calculation again to generate images for the Ti-K and the O-K peaks, and use the “Color mix” functionality of Digital Micrograph or the “Stacks to RGB” functionality of ImageJ to form a color composite map of the different elements present.

STEM ABF

**Getting started**: Play control file “STEM\_Al3Li\_ABF\_driver.txt” and compare the resultant STEM ABF image to the projected structure in the .xtl file.

**Things to try**:

1. Generate a STEM ABF image for SrTiO3 by suitable modification of the control file “STEM\_STO\_HAADF\_ driver.txt”.
2. The Li column contrast is weak. Are we really seeing lithium itself or instead some artefact from dynamical scattering? Explore this by making a pseudo-structure by removing the Li atoms. (Hint: you will need to edit an .xtl file.)

**Things to think about**:

1. Do you expect the ABF contrast to increase or decrease if the accelerating voltage was changed? This non-trivial question can be answered using simulation, but, for a “fair” comparison, what else (amongst the microscope parameters) might you want to change?

HRTEM

**Getting started**: Play control file “TEM\_Si3N4\_HRTEM\_driver.txt” and view the output.

Note: The diffraction pattern is output as a smaller array

**Things to try**:

1. The calculation run is for a pre-aberration-corrected instrument. Can you identify the structure from the image? Try the following negative-Cs imaging parameters [Jia et al. Microscopy and Microanalysis 10 (2004) 174–184]: *C*s = –0.031 mm, aperture = 20 mrad, defocus = 90 Å. Is the resultant image more intuitive?
2. For your preferred set of imaging parameters, run simulations for a series of increasingly thick samples. How does the image contrast change? Compare with HAADF STEM.
3. View the Diffraction pattern (you will need to adjust contrast to see other diffraction spots alongside the central spot). Does this diffraction pattern resemble anything you’ve ever seen experimentally? How might the simulation be made to look more similar to experiment?

**Homework**

All the examples above have been based on elastic wavefunction calculations using thermally smeared elastic potentials and absorptive potentials for inelastic thermal scattering. These calculations are conveniently fast and generally adequate for elastic imaging (e.g. ABF, PACBED and HRTEM). However, for thicker samples the positive contribution of thermally scattered electrons to HAADF images and the contribution from electrons which undergo thermal scattering before causing ionization events for EDX imaging cannot be ignored for quantitative work.

Better models for such imaging are the so-called “frozen phonon” model and the quantum excitation of phonons (QEP) model. μSTEM can perform calculations in the latter. Re-run some of the above calculations using the QEP model by recording new control files and selecting “perform QEP calculations” rather than “perform elastic wave function calculations” at the appropriate boxed menu entry. With this option selected, you will ultimately be prompted to enter “number of phase grates calculated” and “number of Monte Carlo calculated”. Consult the manual for more details, but for testing purposes 20 and 10 would be reasonable choices. You may also need to increase the number of pixels in the supercell and the tiling of the unit cell within the supercell – QEP calculations have more stringent requirements for convergence, and the calculation array needs to include scattering angles spanning the full range of the HAADF detector.

These calculations will take significantly longer than the corresponding absorptive potential calculations, but you should seek to convince yourself that the two approaches give quantitatively different results for, say, HAADF imaging for samples thicker than about 100 Å.

**A few comments on sampling**

One important aspect of μSTEM calculations has, by using existing control files, been sidestepped above: the question of sampling. A few comments are offered here. Please consult the μSTEM manual for further details.

Use of Fourier transforms enforces periodic boundary conditions, but there is no periodic array of STEM probes in the experiment. Calculations therefore use a “supercell”, a 2D tiling of unit cells sufficiently large that the spurious repeat has no adverse consequences.

The number of pixels in each dimension should be a power of two for efficient Fourier transforms. Larger values allow better sampling, but make for slower calculations. 512 or 1024 are typical values for STEM imaging, but 2048 might be necessary for CBED where fine sampling in both real and reciprocal space may be required. Suitable numbers for the tiling of the input unit cell within the supercell along each dimension depend on the unit cell size. Supercells in the approximate range 20–30 Å on a side are advised for STEM image simulations, though this should be closer to 100 Å if a CBED pattern with high reciprocal space resolution is sought.

For converged numerical calculations, both real space and reciprocal-space must be adequately sampled. Real space sampling size Δ*x* is the supercell length *Lx* divided by the number of pixels in that direction *Nx*, i.e. Δ*x*=*Lx*/*Nx*. *Lx* should not be made too small because of the false periodicity problem, but making *Nx* very large makes calculations slow. Δ*x* should be somewhere near the 0.02–0.05 Å range for QEP calculations; slightly larger values suffice for absorptive calculations. Reciprocal space sampling size Δ*qx*=1/*Lx*, or, in scattering angle units, Δ*βx*=Δ*qx*/*K*, where *K* is the wavevector of the fast electron (*K*=39.875 Å–1 at 200 keV). For technical reasons, the calculation includes scattering angles only up to *Nx*.Δ*βx*/3. Therefore, if Δ*βx* is too small the calculation may not include all the necessary high angle scattering (essential for HAADF simulations in QEP). If Δ*βx* is too large relative to the size of the probe-forming aperture angle then one is not sampling angles very well. One will often want Δ*βx* to be small for CBED calculations where fine reciprocal space sampling is necessary to see the features of interest.