StatSTEM tutorial

Quantitative (S)TEM by using StatSTEM

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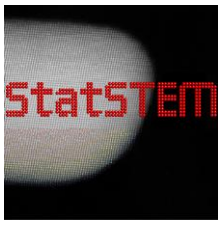

1. Introduction

This lab session is intended to introduce the StatSTEM software and its capabilities in the field of quantitative (scanning) transmission electron microscopy ((S)TEM).

The StatSTEM software is an open source code written in MATLAB. The code and a manual can be found on the StatSTEM website: https://github.com/quantitativeTEM/StatSTEM under the releases button. In this lab session, a compiled version will be used. More details on the algorithm can be found in: Ultramicroscopy 171 (2016) 104-116.

In the instructions that follow, **bold letters** refer to a button or menu in the program while *italic letters* refer to a panel within the program. In the top left corner different TOOLS are available which will be active for the shown image:

Please ask for help if the instructions are not clear enough!

Launch StatSTEM by double clicking on its icon on the desktop.

2. A general overview on StatSTEM its functionalities

2.1 The colormap

In this tutorial, we will go briefly through the different functionalities that StatSTEM has to offer.

Click on **Load** and open the image "ExamplePtIr.txt". StatSTEM will ask for the pixel size, which is 0.1211Å.

A grayscale image of a normalised Pt/Ir particle is loaded. The grayscale of the image can be modified by adding a colorbar to the image:

By right-clicking on the inserted colorbar, the colormap of the image can be changed. Activate for example the jet colormap: **Standard Colormaps→jet**

Since most electron microscopy images are shown in grayscale, this colormap is set as the standard colormap in StatSTEM. Change back to the grayscale colormap: **Standard Colormaps→gray**

Sometimes the most interesting features of an image are only present within a specific range of pixel values. Edit the range of pixel values by right-clicking on the colorbar and change the range: **Open Colormap Editor**

Restore the colormap range by right-clicking on the colorbar: **Reset Range Colors**

When the range of colors is optimised, you can further highlight different features in the image by right-clicking on the colorbar: **Interactive Colormap Shift**
By using this function you can drag a color in the colorbar to a different value.

Restore the colormap by right-clicking on the colorbar: **Standard Colormaps→gray**

Can you think of any features in an image that can be highlighted by using the colorbar?

2.2 Starting values

Continue using the image "ExamplePtIr.txt".

Before modelling an image as a superposition of Gaussian peaks, initial column positions should be given. For this purpose, StatSTEM offers a peak finding routine and the possibility to give manually starting values. Furthermore, starting positions of the columns can be loaded from an external file.

We start by using the peak finding routine: **Peak Finder Routine→Tune parameters**

The peak finder routine searches for local maxima in your image. Since noise is always present in the experimental images, different filters can be used. The parameters of these filters can be tuned in the new window that has been opened. Standard, a Gaussian filter is used with a width of 2 pixels. Also a threshold value can be given. The pixel values should have higher values to be found as an atomic column position.

Add a filter and modify the parameters of the filters and the threshold value to see how it affects the image.

Restore the parameters to the initial values by: **Reset values**
A threshold value of 0 and a Gaussian filter with a width of 2 pixels will be used.

Test the peak finder routine: **Test peak finder**

Can you explain why it finds peaks everywhere in the image, even where there are no atomic columns?

Set the threshold value to 0.02 and test the peak finder routine again: **Test peak finder**

After suitable parameters and filters have been selected, the parameters can be exported to StatSTEM: **Store and close**

In StatSTEM, execute the peak finder routine: **Peak Finder Routine →Execute routine**

As can be seen, the peak finder routine did not find initial values for all column positions. Manually add positions for the columns that you can visually see: **Add/Remove Peaks→ Add**

The last option is to load coordinates from an external file. Load the column positions from the file "CoordinatesPtIr.txt": **Import peak locations → Load file**

Are the loaded column positions similar as compared to your initial values? If not, can you think of a reason that explains the difference?

2.3 Modelling the image

After initial starting values are given for the column positions, the image can be modelled as a superposition of Gaussian peaks. Go to the next tab *Fit Model*.

In this panel different options are available:

- Fit a constant background
- Model the column intensities by Gaussian peaks having all the same width, all a different width, or a fixed given width
- Test for convergence
- Parallel computing

For the moment use the standard values: fit a constant background, use the same width for all the Gaussian peaks and don't use test conditions. Start the fitting procedure: **Run fitting routine**

The fitting procedure may take a while. Once it is finished, the fitted model will be shown. Compare the fitted model to the original image by selecting the shown image in the right top corner: **Select image → Observation** and **Select image → Model**

Show the model: **Select image → Model** and make both the input and fitted coordinates visible: **Image Options → Input coordinates**

Is there much difference between the initial starting positions and the fitted positions?
A large difference between the input and fitted positions indicates that the initial values were most likely not accurate enough and might need an update.

2.4 Atom counting

After the image is modelled, structure parameters can be extracted. One of the structure parameters has already been discussed: the coordinates of the atomic columns. Another structure parameter is the so-called scattering cross-section (SCS). The SCS is the total scattered intensity by an atomic column and scales in HAADF STEM imaging with both the thickness and the chemical composition of the atomic column. The SCS can be obtained by integrating the total intensity scattered by an atomic column, which is done in StatSTEM by using the volumes of the Gaussian peaks. Here, both the height and width of each Gaussian peak is used to calculate the volume. In StatSTEM, the SCSs of all the different columns can be seen in a histogram: **Select Image → Histogram SCS**

How should the histogram of the SCSs look in the case that no image noise and distortions are present?

Despite the noise that is present in the measured values of the SCSs, we still want to perform atom counting. Visually, it is impossible to identify the number of atoms that should be associated with each measured value of the SCS from the histogram. Therefore a statistics- and a simulation-based method will be discussed. Go to the next panel: *Analysis*.

2.4.1 Atom counting – Statistics-based method

The first method to perform atom counting is by using a statistics-based method in which the SCSs are modelled by a Gaussian mixture model. It is, however, unknown how thick the particle is and how many Gaussian components are present in the histogram. In order to decide how many Gaussian components are present in the histogram, the ICL criterion will be used. This criterion will show a local minimum at the correct number of Gaussian components in the histogram.

In order to use the ICL criterion to count the number of atoms, the SCSs in StatSTEM will be modelled by a Gaussian mixture model for an increasing number of Gaussian components. Open the options: **Atom counting – Statistical → Show options**

In the standard settings, the maximum number of Gaussian components is 50. For the moment this is fine, as we don't expect that the column thickness of the particle is larger than 20 atoms. Therefore, you can also change this number to reduce the computation time: **Atom counting – Statistical → Pre-analysis options → Max. components → 20**

How can we determine an appropriate value for the maximum number of Gaussian components?

Start the fitting of the Gaussian mixture model and the ICL procedure: **Atom counting – Statistical → Run ICL**

When the procedure is finished, the user is asked to click on the local minimum. Select a minimum which you think is suitable. After selecting the minimum the experimental image is shown together with the counting results.

Open the ICL image: **Select image → ICL**
Select another minimum and see how the counting results change: **Atom counting – statistical → Post-analysis options → Select new ICL minimum**

Open the histogram: **Select image → Histogram SCS**
In this image, the Gaussian mixture model is shown together with the individual components. The colors of the individual components are directly related to the colors of the atom counts in the experimental image.

Open the graph with the SCSs as a function of thickness: **Select image → SCS vs. thickness**
Here, the locations of the different Gaussian mixture model components are shown. Often the component number is directly related to the number of atoms. Select the local minimum 16 in the

ICL, if not yet selected: **Post-analysis options → Select new ICL minimum** and go back to the graph **Select image → SCS vs. thickness**

From this graph it can be seen that the SCSs increase monotonically with thickness. It should be noted that this increase is not a linear increase due to the channeling of the electrons along the columns.

2.4.2 Atom Counting – Simulation-based method

Another method to count the number of atoms is by comparing with image simulations. By using, for example, the MULTEM software simulated images can be acquired as a function of specimen thickness. The SCSs of the columns in the simulated image as a function of thickness serve as a library for atom counting. In this method, the experimentally measured SCSs are directly compared to the library values.

Load the library "LibraryPtIr.txt": **Atom counting – Simulation → Load library values**

In the graph showing the SCSs as a function of thickness, **Select image → SCS vs. thickness**, the loaded library is shown together with the experimental locations of the Gaussian components in the mixture model. Do the library values match with the experimental values?

Select another minimum in the ICL and find a match between the library and the experimental values: **Post-analysis options → Select new ICL minimum**

What minimum did you find? What is the percentage of atomic columns counted with single atom sensitivity (see the message panel)?

The library values can also be used in StatSTEM to perform atom counting: **Atom counting – Simulation → Match with simulations**

In the experimental image, compare the counting results from the simulation-based method and the statistics-based method: **Image Options → Atom Counts** and **Image Options → Lib Counts**

Are there any differences? How can you explain the differences?

2.5 Strain mapping

Another feature of StatSTEM is atomically resolved strain mapping. Go to the analysis panel: *Analysis*.

For strain mapping, a necessary input to StatSTEM is the projected unit cell: **Strain mapping → Projected unit cell**

In this new window one can design its own projected unit cell. For Pt in the 110 direction there is already a database file "DatabaseStrain/Pt110.txt". Load this file: **Load file**

Export the file to StatSTEM: **Export to StatSTEM**

Open the strain mapping options: **Strain mapping → Show options**

Deselect the atom counting results: **Image Options → Atom Counts**

First a reference coordinate should be given, in which no strain of a minimum amount of strain is expected. Standard the most central coordinate is chosen: **Strain mapping → Reference coordinate → Most central → Show**
If this coordinate is not suitable, another coordinate can be chosen manually: **Strain mapping → Reference coordinate → User defined → Select** and select a coordinate in the image.

In this particle, the most central atomic column experiences most likely less strain as compared to the others: **Strain mapping → Reference coordinate → Most central → Show**

Next, the direction of the lattice parameters should be found. This procedure requires an accurate value for the pixel size of the image! An automatic procedure is available to find the a-direction of the unit cell in the image: **Strain mapping → Direction a lattice → Automatic → Show**

Otherwise, the a-direction can be given manually. In the procedure of finding the a-direction, the given lattice parameter can be updated by a fitting procedure. Standard, a region of 3 unit cells around the selected reference coordinate is used to update the value of the lattice parameter a by a fitting procedure. If this is unwanted, this option can be deselected. For now, leave this option turned on.

After selecting a reference coordinate and finding the a-direction, a displacement map can be made: **Strain mapping → Make displacement map**

A warning will be given that not all points are found. This is caused by the loose atoms that are not packed to the Pt/Ir particle. Accept the warning and manually select a region around the particle which neglects the loose atoms: **Strain mapping → Show options** next select **Strain mapping → General options → Select columns in image**

Create again a displacement map: **Strain mapping → Make displacement map**

In this displacement map, the fitted coordinates are compared to the expected ideal lattice positions when no strain is present in the sample.

The red arrows indicate the displacement of the atomic column positions with respect to expected atomic columns of the ideal lattice. The displacement of the atomic columns at the edge of the particle is largest, why?

After the displacement map is created, a strain map can be created by using a first derivative of the displacement vectors: **Strain mapping → Make strain map**

Standard, the ε_{xx} map is shown. Look also at the other strain maps: **Image Options → ε_{xy}** , **Image Options → ε_{yy}** and **Image Options → ω_{xy}**. In which atomic columns is most strain present? Is this in agreement with the displacement map?

2.6 Store and close image

When you are finished analyzing an image, you can store the results in a MATLAB file: **Save**
After saving the results, close the image in StatSTEM by right-clicking on the image tab: *ExamplePtIr* → **Close image**

Afterwards you can reopen the saved file in StatSTEM or MATLAB to modify or extract some of the results