

HRTEM image simulation of arbitrary [uvw] direction

Pierre Stadelmann
jems-jongny
Chemin Rouge 15
CH-1805 Jongny
Switzerland
jems.jongny@gmail.com

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1 Introduction

jems uses 2 methods in order to do HRTEM image simulation:

1. Multislice method.
2. Bloch-wave method.

These 2 methods are described in sections 2 and 3). Both methods uses the very same scheme to define the diffraction geometry of dynamical calculations (Fig. 1). The specimen orientation is specified by an arbitrary zone axis direction (given by its $[uvw]$ indices) and a small mis-orientation given by the vector $\vec{\chi}$ from the center of the Laue circle to the origin.

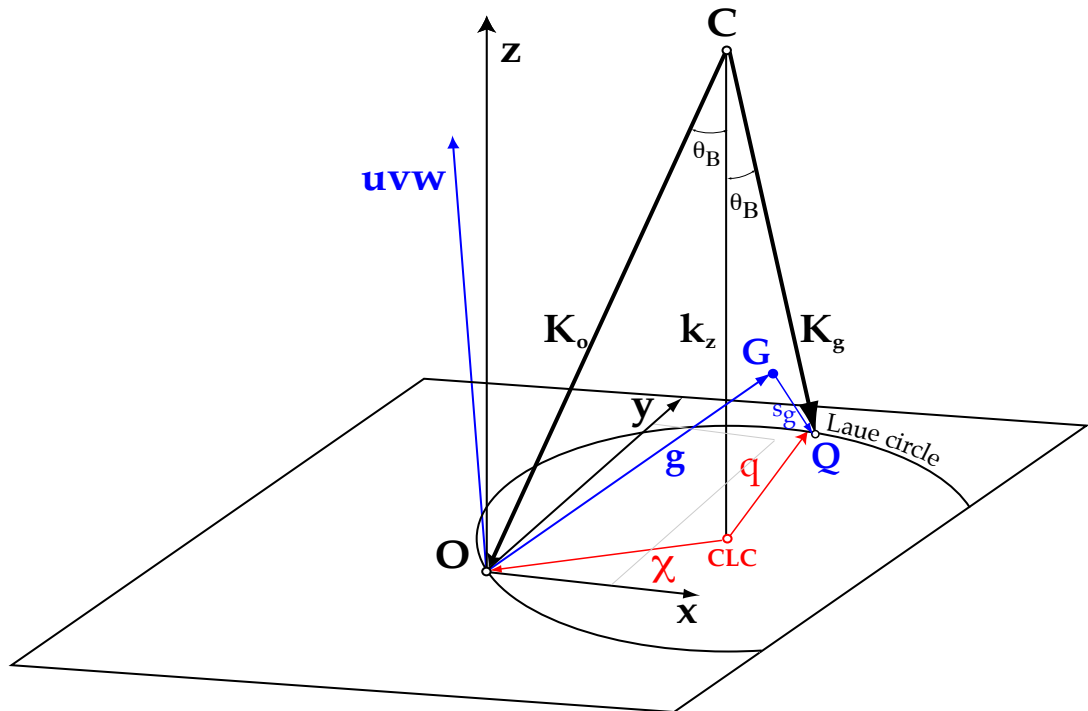


Figure 1: Diffraction geometry is given by the $[uvw]$ zone axis indices and the Center of Laue Circle (CLC) given $\vec{\chi}$.

The wave-vector \vec{K}_0 of the incident wave-function is decomposed in a component \vec{k}_z parallel to the optical axis and $\vec{\chi}$ perpendicular to it. The scattering vectors \vec{q}

are described as $\vec{g} + \vec{s}_g$ where \vec{g} and \vec{s}_g are a reciprocal vector and the deviation from exact Bragg conditions¹.

¹ $\vec{g} + \vec{s}_g = -\vec{\chi} + \vec{q}$

2 Multislice method

The Multislice method is best adapted to perform image simulation of large unit cell crystals under simple $[uvw]$ orientations. It assumes periodic continuation, i.e. the cell or super-cell is periodic perpendicularly to the incident electron beam direction. Using this method it is usually necessary to cut the crystal in thinner slices (of the order of $0.1 \sim 0.2$ nm thick) and due to the periodic assumption to do some transformation the unit cell.

For crystals with small lattice parameters and arbitrary $[uvw]$ indices it requires also to transform the unit cell $(\vec{a}, \vec{b}, \vec{c})$ into a larger cell $(\vec{a}', \vec{b}', \vec{c}')$ that is orthogonal in the $[uvw]$ direction, i.e. $\vec{c}' \perp \vec{a}'$ and $\vec{c}' \perp \vec{b}'$.

CeO_2 (space-group $Fm - 3m$, lattice parameter $a = 0.545$ nm, Ce at $(0,0,0)$, O at $(\frac{1}{4}, \frac{1}{4}, \frac{1}{4})$ Fig. 2) will be chosen to demonstrate the approximations and methods.

The projection of CeO_2 observed in direction $[110]$ is shown in Fig. 3. Note that Ce lattice planes alternate with O planes.

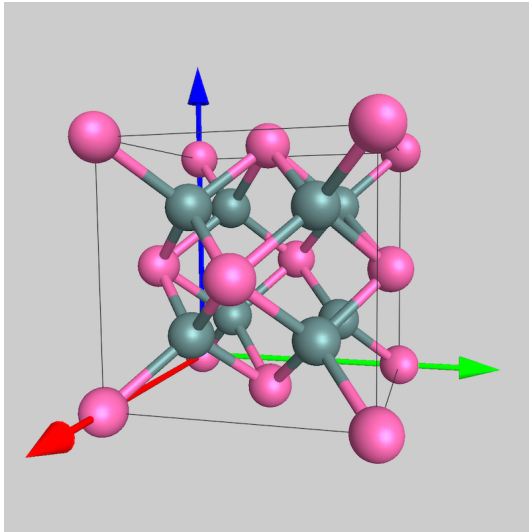


Figure 2: CeO_2

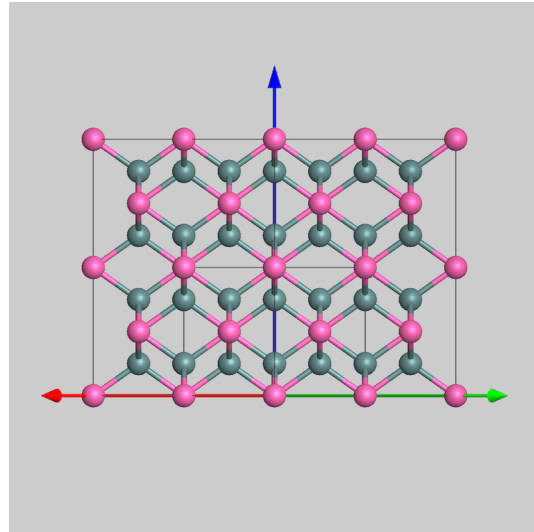


Figure 3: CeO_2 projected in $[uvw] = [110]$ direction.

Fig. 4 shows the amplitude and phase of the (002) , (004) and (-111) reflections as a function of crystal thickness. At a thickness of ~ 5 nm the phase of (004)

reflection is opposite to the phase of the (040) and (-111) ones. One should expect an inversion of HRTEM images contrast at this thickness.

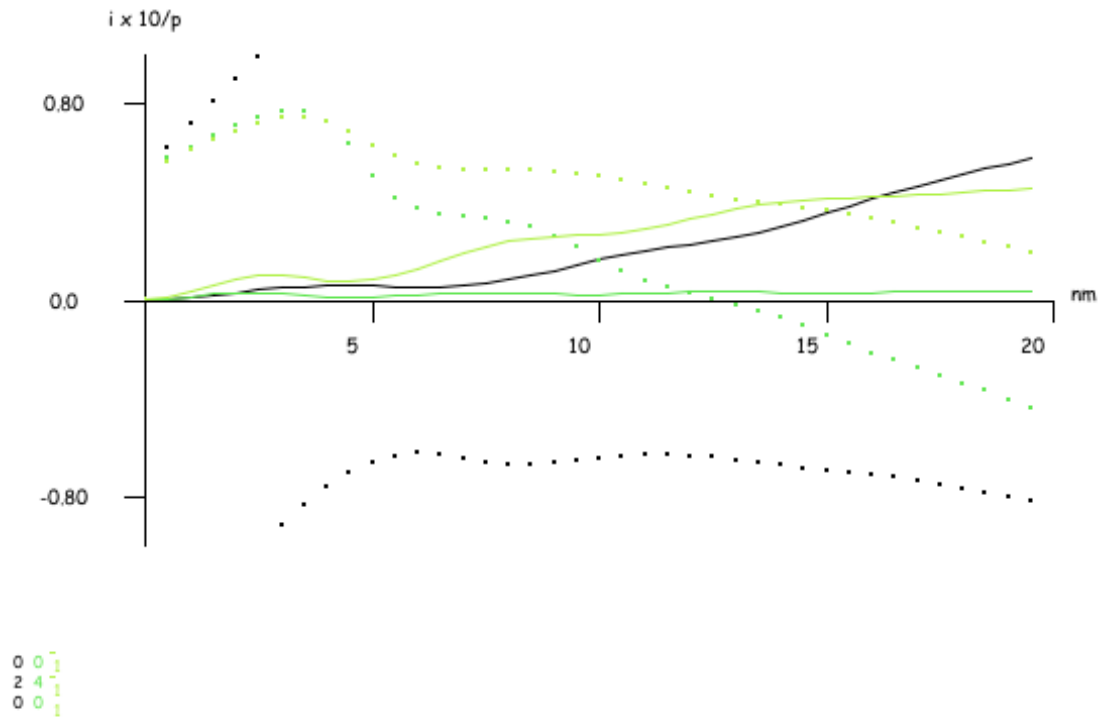
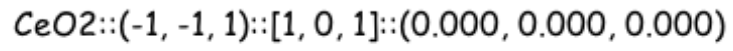


Figure 4: Dynamical diffraction behaviour of the (002), (004) and (-111) reflections.

2.1 Simple approximation

The simplest approximation consists of:

1. projecting the potential of the whole unit-cell in the selected $[uvw]$ direction.
2. preparing slices made of a fraction of the projected potential.

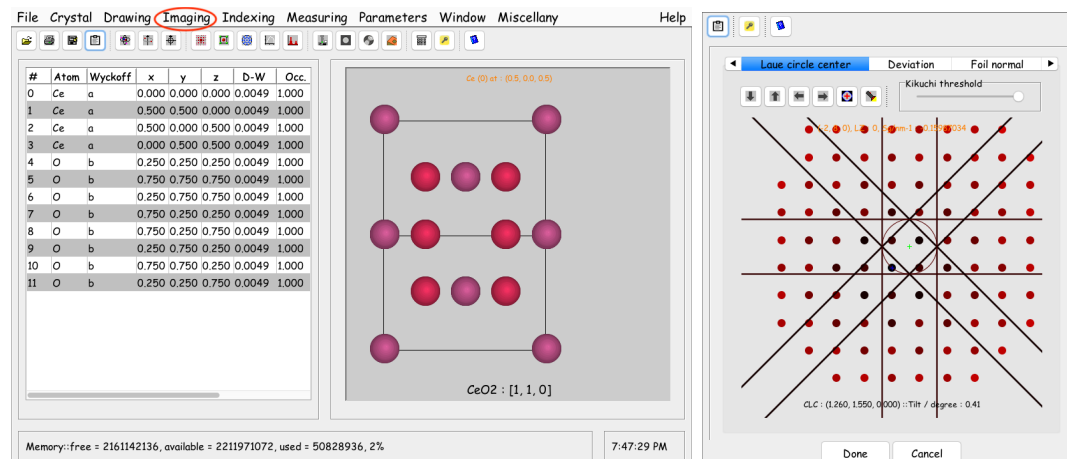




Figure 5: jems main window. Bloch-wave and Figure 6: *Specimen* dialog box to set the Center of Laue Circle (grab the center cross and move it).

This approximation neglects the 3-D aspect of the crystal and ignores any high-order Laue zones effects. The procedure is done as follow:

1. Start jems (.
2. Load the CeO_2 crystal structure (if not set as default crystal) using **[File]→[Open]** or  and select the $[110]$ direction (Fig. 5) using the popup menu of the projection drawing or the *Specimen* dialogue (Fig. 6).
3. Define the *center of the Laue circle* using the *Specimen* dialogue (Fig. 6).
4. Select **[Imaging]→[Multislice]** .
5. In **HRTEM** tab select **Illumination** tab, **Imaging** tab and **Iteration** tab to set the imaging and diffraction conditions (Fig. 7, Fig. 8, Fig. 9, Fig. 10).

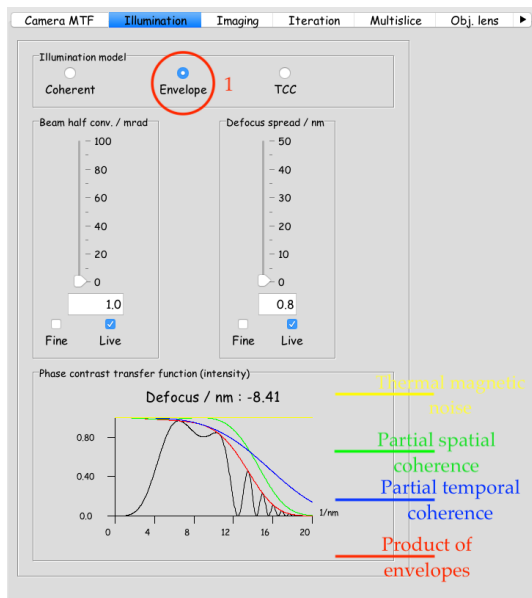


Figure 7: Illumination parameters: partial coherence envelopes.

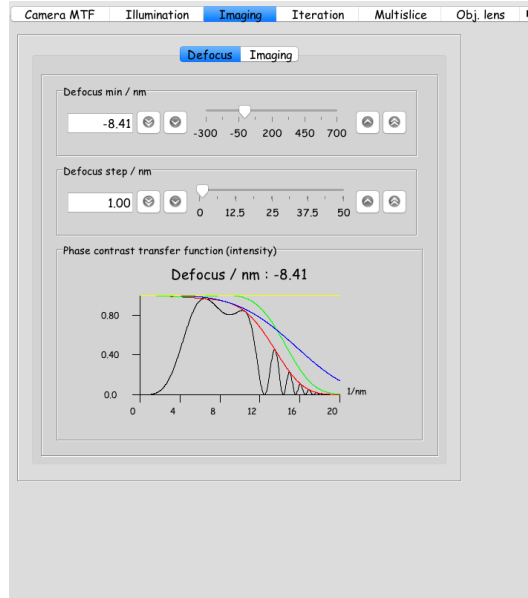


Figure 8: Defocus parameters: defocus of 1st image and defocus step between 2 images.

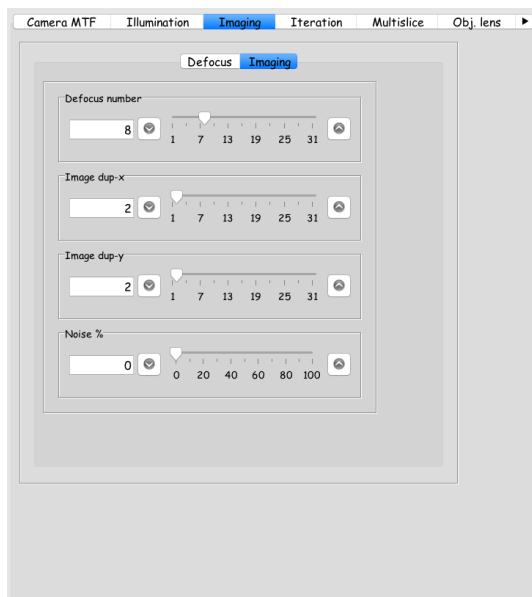


Figure 9: Imaging parameters: defocus series size 8, each image shows 2 x 2 unit cells.

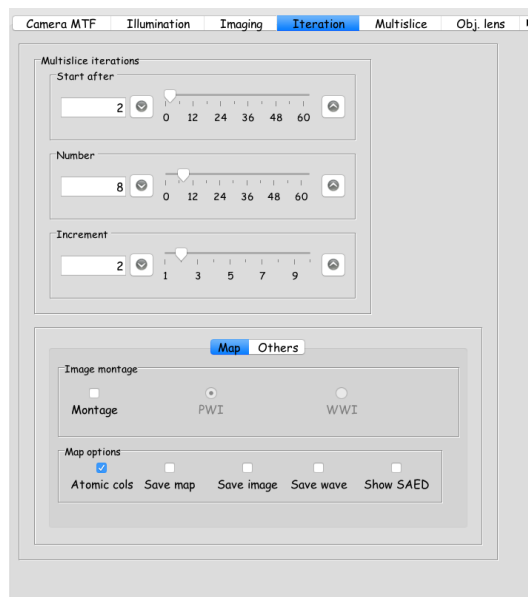


Figure 10: Iteration parameters: 1st wave-function after 2 unit cells, 8 wave-functions separated by 2 unit cells.

In most cases check box **Envelope**, that uses the partial coherence envelopes approximation, should be selected (Fig. 7). The better TCC (**T**ransmission **C**ross **C**oefficients) illumination model is more time consuming and should be not selected for *real time* image simulation of large unit cell crystals.

When the simulation parameters are set, start the calculation i.e. **Start**².

A typical image map is shown in the **Map** tab (Fig. 11). The crystal thickness increases from bottom to top while the defocus increases from left to right. The defocus and thickness are displayed in the cursor box. The image map is also displayed in the **Map** tab of the result tabbed pane. A large image map is also shown in a separate window (Fig. 12).

When check box **Atomic columns** is selected, the cursor shows the atomic column positions. A mouse click on any position identify the atom kind.

²This button is only activated when the **Map** tab is selected.

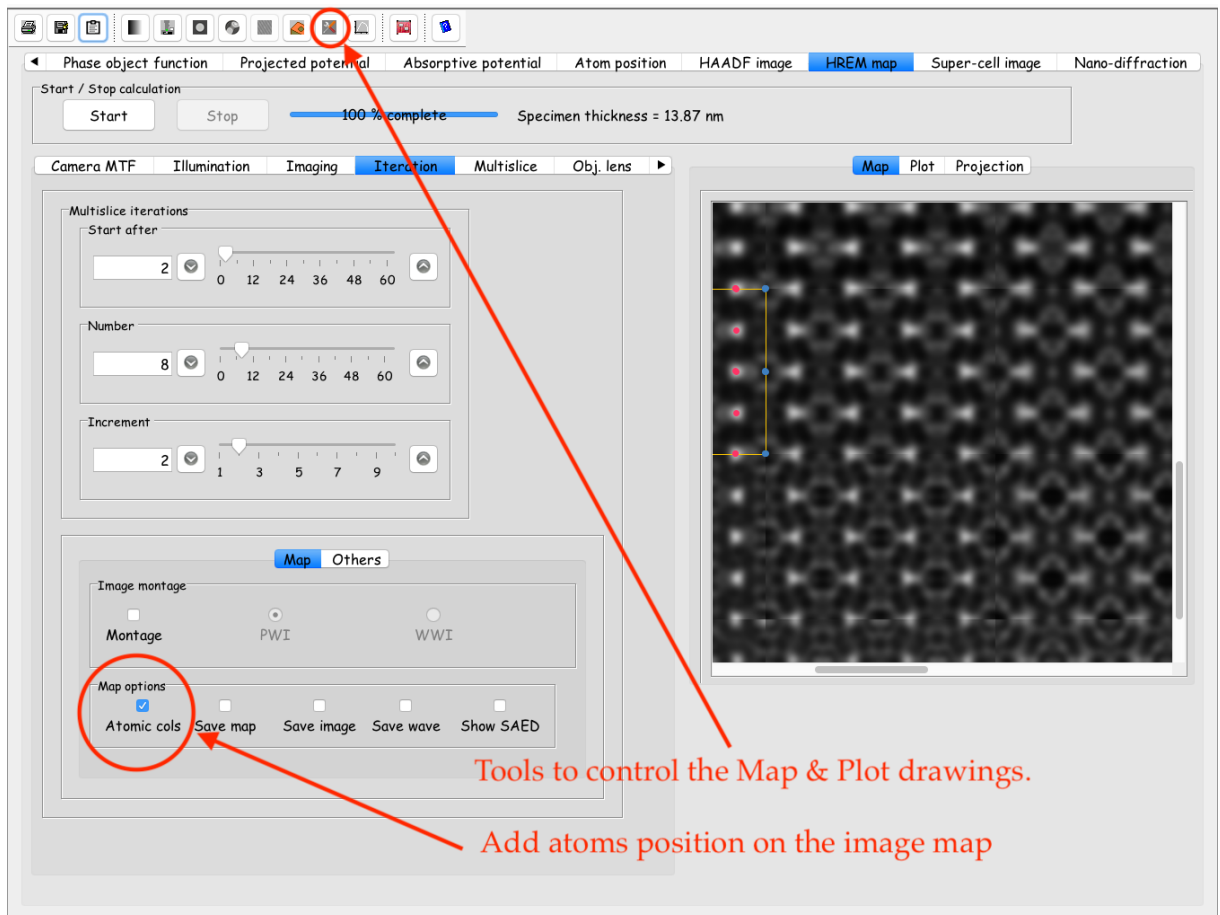


Figure 11: HRTEM images: simple approximation.

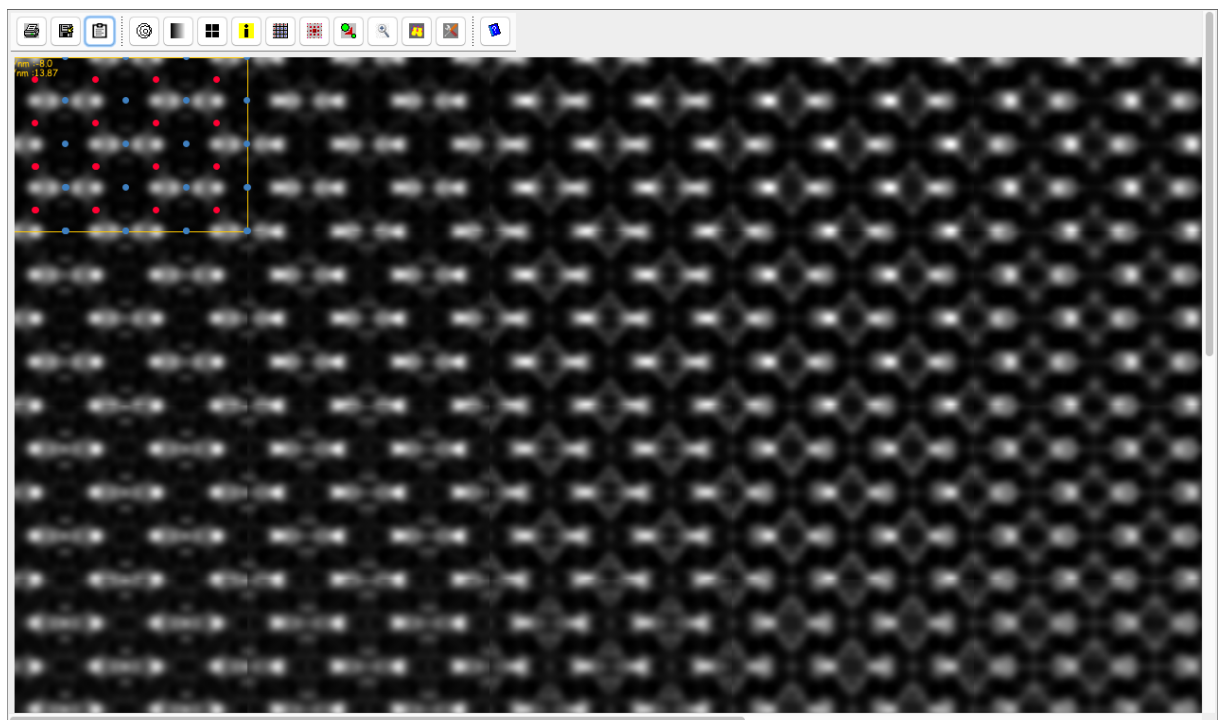


Figure 12: HRTEM images map with superimposed atomic position cursor.

2.1.1 Montage image

When check box **Montage** is selected the image map shows the projected potential, 1st column, the wave-function intensity 2nd column (radio button **PWI**), and the defocus series (Fig. 13) or the real and imaginary parts of the wave-function (radio button **wwi**) and defocus series (Fig. 14).

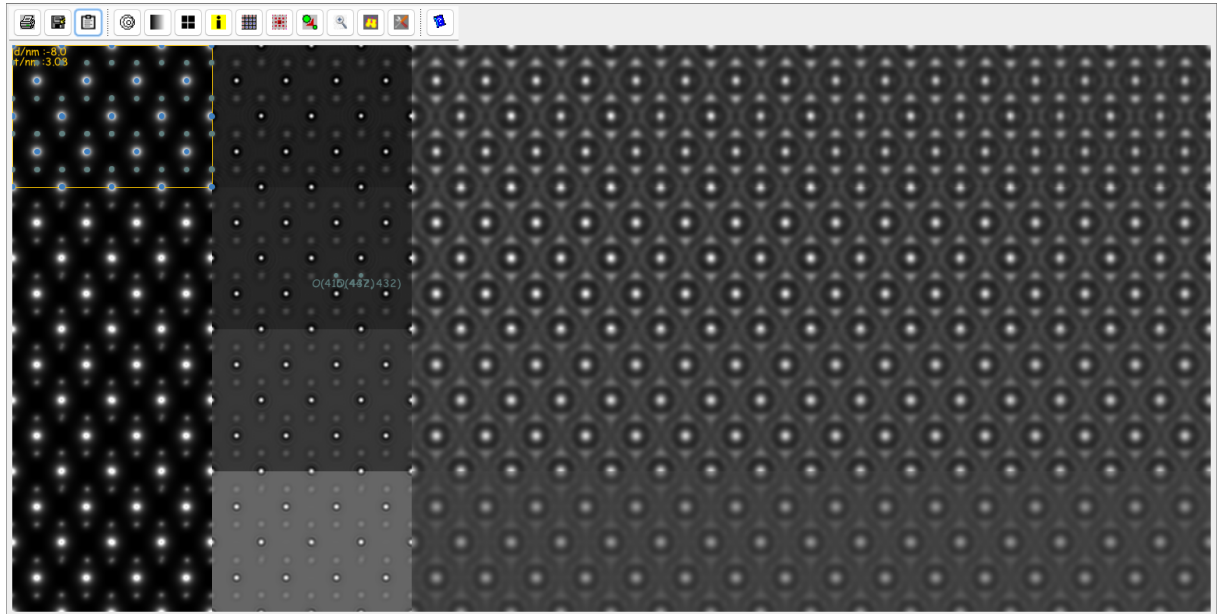


Figure 13: Montage image: projected potential, wave-function intensity and defocus series.

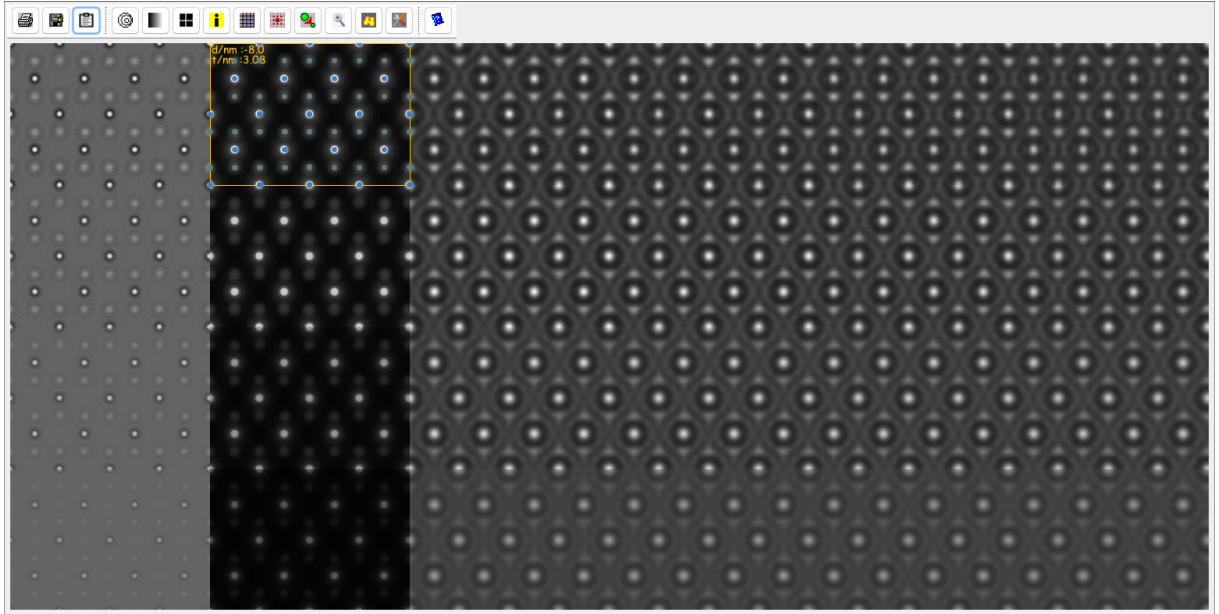



Figure 14: Montage image: wave-function real part, wave-function imaginary part and defocus series.

2.1.2 Seeing aberrations effects on the simulated image

When the check box **Plot** of the **Iteration Others** tab is selected, the **Multi-slicer** window can plot, in the **Plot** tab of the result pane, the reflections and image as a function of thickness. Effects on the image of a change of defocus, C_s , C_5 , astigmatism (2-fold and 3-fold), 2nd order coma, drift, vibration, ..., can be observed interactively. After selecting check box **Plot** it is necessary to redo the calculation.

To vary the thickness or contrast settings use the sliders activated by  (Fig. 11). To change the defocus select the **Imaging Defocus** tab, to change the coherence select the **Illumination** tab, to introduce astigmatism select the **Obj. lens** tab (Fig. 15, and Fig. 16), etc.

In order to introduce effects of aberrations up to order 8, it is necessary to save the wave-function and to activate the HRTEM imager (**[Imaging]** → **[HRTEM imager]**), see section 2.4.

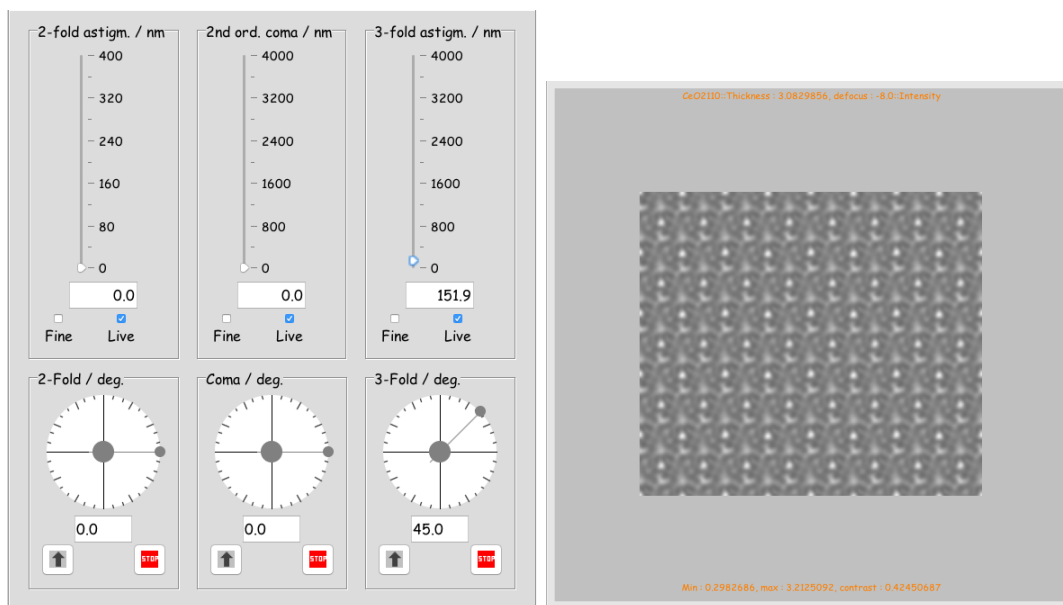


Figure 15: The **Obj. lens** tab controls 2-, 3-fold astigmatism and 2nd order coma. Figure 16: Effect of 3-fold astigmatism on the image contrast (Titan 80-300, negative C_s).

2.2 Using an orthogonal cell

In order to get images of CeO_2 in the $[110]$ direction where HOLZ effects are taken into account it is necessary to define a periodic orthogonal cell with \vec{c} parallel to the optical axes (Fig. 23). The large periodic cell can then be sliced into thinner sub-slices.

This slicing technique can be applied to any $[uvw]$ direction, but the super-cell is very often pretty large as show in Fig. 17 or Fig. 18. Since the procedure is tedious, one can use several approximations or a Bloch-wave approach (section 3). The simplest approximation is to project the potential of CeO_2 at once and to take a fraction of the projected potential for each sub-slice. This is the default jems procedure, that ignores **H**igh-**O**rders **L**auve **Z**ones effects (these effects are usually negligible when the total crystal thickness is less than ~ 20 nm). It is equivalent to perform a Bloch-wave calculation including only reflections of the ZOLZ (**Z**eroth **O**rders **L**auve **Z**one).

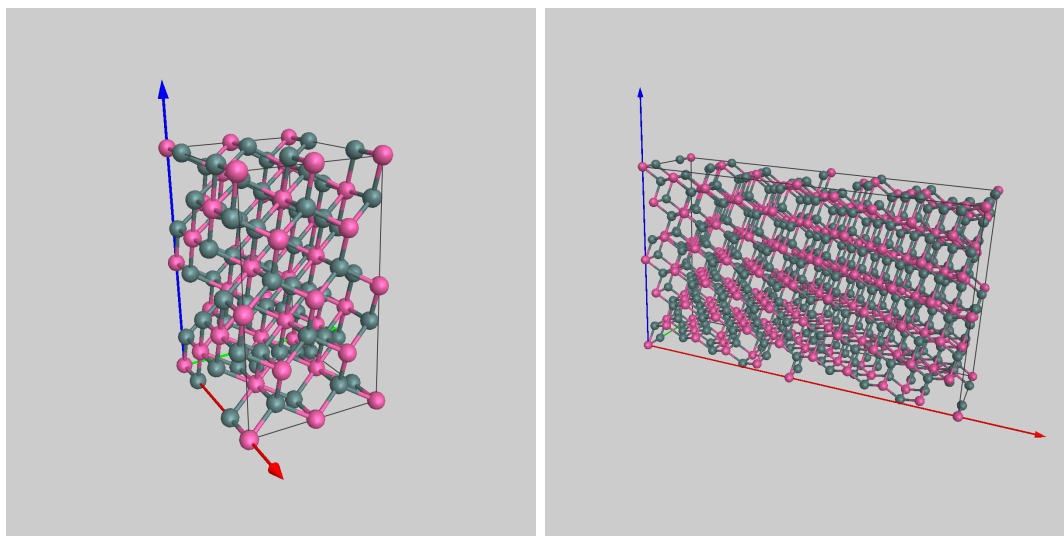



Figure 17: Orthogonal CeO_2 in $[121]$ direction ready for slicing in order to perform multislice calculations. Figure 18: Orthogonal CeO_2 in $[213]$ direction ready for slicing in order to perform multislice calculations.

2.2.1 Defining the periodic orthogonal cell

An periodic orthogonal cell is defined such that its [001] direction is parallel to any [uvw] direction of the original unit cell. To define the periodic orthogonal do the following:

- Start jems and load the CeO_2 unit cell.
- Activate dialogue [Crystal]→[Make orthogonal] (Fig. 19).
- Use the *specimen dialog* dialogue () to define the [UVW] zone axis direction of the original unit cell that will define parallel to the [001] direction of the orthogonal unit cell.
- The lattice parameters, atoms position, the [uvw] directions of the original unit cell parallel to the [100], [010] and [001] directions of the orthogonal cell as well as the ratio of the volume of the cells is provided (Fig. 20).

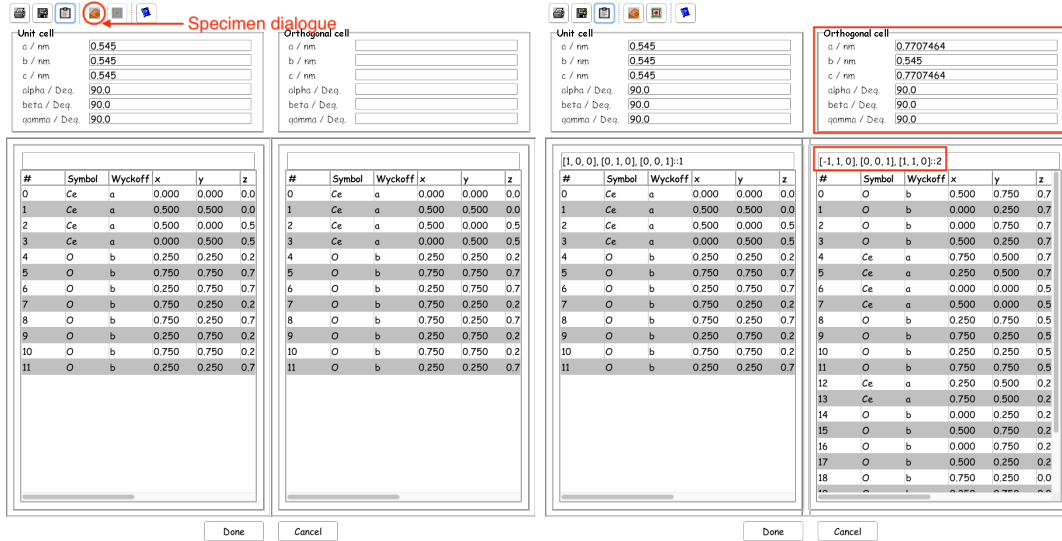



Figure 19: *Make orthogonal* dialogue dialog. Figure 20: The newly defined orthogonal cell with its [001] direction parallel to [110] of CeO_2 .

Finally close the *Make orthogonal* dialogue (**Done**) and save the new orthogonal cell ([File]→[Save] or ).

2.2.2 Multislice tab: calculating the projected potential

The 2-D projected potential and 3-D potential can be calculated using 3 different methods:

- Method 1 (radio button Direct) calculates the 2-D projected potential or 3-D potential of the different atom species and patch it at the atom position. This method uses bilinear or bi-cubic interpolation (both in 2-D or 3-D) in order to patch the atomic potential at the atom position ($0 \leq (x, y, z) < 1$). It is the fastest method, but can introduce artefacts (due to interpolation).
- Method 2 (radio button Analytic) calculates the Fourier transform of 2-D projected potential or 3-D potential of the different atom species and convolves them with the phase image generate using only the atoms position. An inverse Fourier transform provides the 2-D or 3-D potential. It is slightly slower but avoid introducing artefacts in the 2-D or 3-D potential.
- Method 3 (radio button Fourier) generates the 2-D or 3-D potential in Fourier space. The structure factors of the crystal are calculated on a 2-D or 3-D grid (defined by reciprocal space) and an inverse Fourier transform provides the potential. It is the most precise method but also the slower since structure factors are calculated on many Laue zone planes. It should be used when all the image simulation parameters have been defined properly.

The 3-D potential is then integrated over the different sub-slices (of identical thickness). Note that the sub-slices should not be too thin in order to keep the approximation of the Fresnel diffraction (distance of propagation of the order of 50 to 100 wavelengths).

The **Multislice** tab contains the necessary options to introduce HOLZ reflections effects (by slicing the 3-D potential of the unit cell into n sub-slices ($1 \leq n < 256$) (check box Include). Since generating the projected potential of the sub-slices can be time consuming there are also options to load or save the generated phase-object images, check box Load phase-object and check box Save phase-object (Fig. 21). The interpolation method as well as the *Frozen lattice* approximation (random displacement of the atoms around their crystallographic position, i.e. Debye-Waller temperature factor) are selected in the **Frozen lattice** tab (Fig. 22).



Figure 21: Multislice tab: introducing HOLZ reflections.

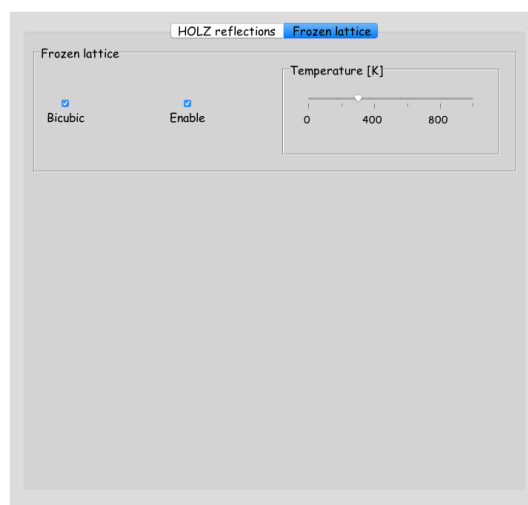


Figure 22: Multislice tab: introducing phonon scattering using the *Frozen lattice* approximation, i.e. Einstein model.

2.2.3 Image simulation with HOLZ reflections

Using the **Multislice** tab calculate 4 sub-slices (Fig. 24). The 3-D potential is first calculated on a (128 x 128 x 64) 3-D grid and then integrated as 4 potential 2-D images (Fig. 25). Finally the HRTEM images quite similar to what is obtained with the simplest procedure (Fig. 26).

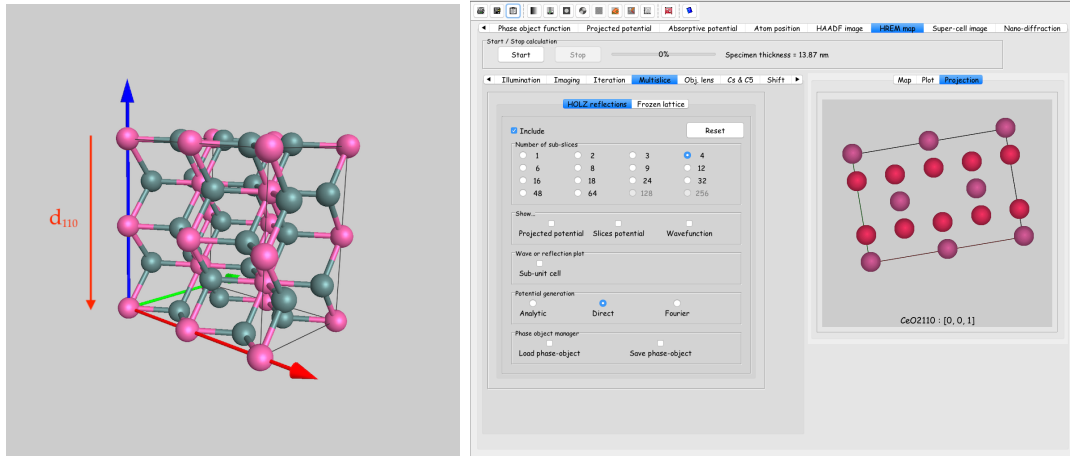


Figure 23: Orthogonal CeO_2 cell, Figure 24: Slicing the cell into 4 sub-slices [110] parallel to optical axis. using the **Multislice** tab.

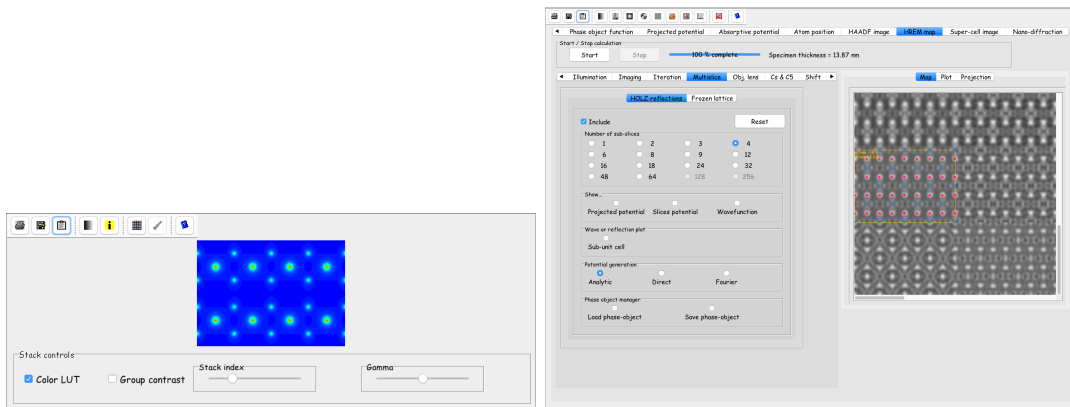


Figure 25: 3-D potential (using a 128 x 128 x 64) 3-D grid. Figure 26: HRTEM image simulation using 4 sub-slices.

2.3 User defined slices

A side view of the propagation of the incident electron wave ([110] direction) is shown in Fig. 27. Due to the large d_{110} it is necessary to cut the structure in slices in order to make use of the multislice method. Cutting this $2 \times 2 \times 2$ CeO_2 cell will produce images of a small cubic particle observed in the [110] direction not of bulk CeO_2 . It is consequently required to slice the periodic orthogonal cell shown on Fig. 28.

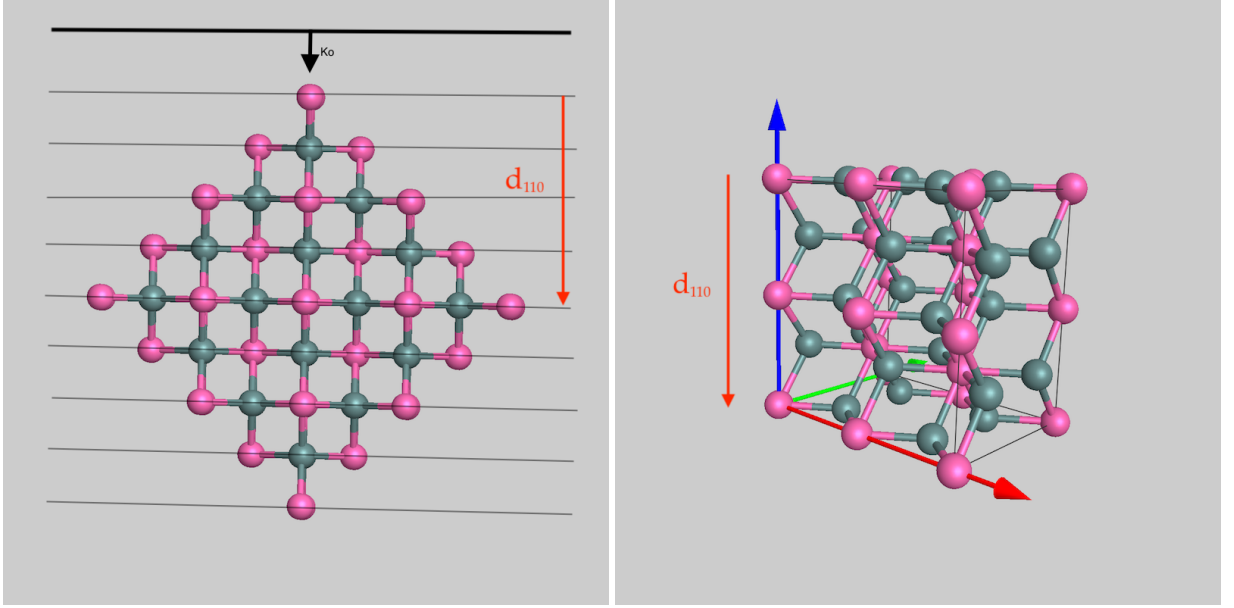


Figure 27: Electron wave of wave vector K_o propagating in CeO_2 oriented in [110] direction ($2 \times 2 \times 2$ unit cells). Figure 28: CeO_2 made orthogonal (1 unit cell), ready for slicing. The blue arrow (O_z axis) indicates the original CeO_2 [110] direction, anti-parallel to \vec{K}_o .

The slicing is performed with `[Miscellany]→[To slices]` (Fig. 29). In order to define slices as square as possible (not required, but nearly square images are simpler to manipulate) one has to repeat the orthogonal unit cell in the \vec{a} and \vec{b} directions (here 2 and 3 times respectively). The sub-slice thickness or number of sub-slices should be selected before slicing (`[Slice]`).

The 4 slices (thickness 0.192 nm) in Fig. 30, Fig. 31, Fig. 32, Fig. 33. Note that for CeO_2 in [110] direction slices 1 and 3 as well as slices 2 and 4 are identical. The final *super cell* is shown in Fig. 34.

#	Symbol	Wyckoff	x	y	z	Debye-Waller	Occupancy	Absorption	Charge	AFF
0	O	b	0.500	0.750	0.750	0.0049	1.000	0.0299	Def	0
1	O	b	0.000	0.250	0.750	0.0049	1.000	0.0299	Def	0
2	O	b	0.000	0.750	0.750	0.0049	1.000	0.0299	Def	0
3	O	b	0.500	0.250	0.750	0.0049	1.000	0.0299	Def	0
4	Ce	a	0.750	0.500	0.750	0.0049	1.000	0.041	Def	0
5	Ce	a	0.250	0.500	0.750	0.0049	1.000	0.041	Def	0
6	Ce	a	0.000	0.000	0.500	0.0049	1.000	0.041	Def	0
7	Ce	a	0.500	0.000	0.500	0.0049	1.000	0.041	Def	0
8	O	b	0.250	0.750	0.500	0.0049	1.000	0.0299	Def	0
9	O	b	0.750	0.250	0.500	0.0049	1.000	0.0299	Def	0
10	O	b	0.250	0.250	0.500	0.0049	1.000	0.0299	Def	0
11	O	b	0.750	0.750	0.500	0.0049	1.000	0.0299	Def	0
12	Ce	a	0.250	0.500	0.250	0.0049	1.000	0.041	Def	0
13	Ce	a	0.750	0.500	0.250	0.0049	1.000	0.041	Def	0
14	O	b	0.000	0.250	0.250	0.0049	1.000	0.0299	Def	0
15	O	b	0.500	0.750	0.250	0.0049	1.000	0.0299	Def	0
16	O	b	0.000	0.750	0.250	0.0049	1.000	0.0299	Def	0
17	O	b	0.500	0.250	0.250	0.0049	1.000	0.0299	Def	0
18	O	b	0.750	0.250	0.000	0.0049	1.000	0.0299	Def	0
19	O	b	0.250	0.750	0.000	0.0049	1.000	0.0299	Def	0
20	O	b	0.250	0.250	0.000	0.0049	1.000	0.0299	Def	0
21	O	b	0.750	0.750	0.000	0.0049	1.000	0.0299	Def	0
22	Ce	a	0.500	0.000	0.000	0.0049	1.000	0.041	Def	0
23	Ce	a	0.000	0.000	0.000	0.0049	1.000	0.041	Def	0
24	O	b	1.500	0.750	0.750	0.0049	1.000	0.0299	Def	0
25	O	b	1.000	0.250	0.750	0.0049	1.000	0.0299	Def	0
26	O	b	1.000	0.750	0.000	0.0049	1.000	0.0299	Def	0
27	O	b	1.500	0.250	0.750	0.0049	1.000	0.0299	Def	0
28	Ce	a	1.750	0.500	0.750	0.0049	1.000	0.041	Def	0
29	Ce	a	1.250	0.500	0.750	0.0049	1.000	0.041	Def	0
30	Ce	a	1.000	0.000	0.500	0.0049	1.000	0.041	Def	0
31	Ce	a	1.500	0.000	0.500	0.0049	1.000	0.041	Def	0
32	O	b	1.250	0.750	0.500	0.0049	1.000	0.0299	Def	0
33	O	b	1.750	0.250	0.500	0.0049	1.000	0.0299	Def	0
34	O	b	1.250	0.250	0.500	0.0049	1.000	0.0299	Def	0
35	O	b	1.750	0.750	0.500	0.0049	1.000	0.0299	Def	0
36	Ce	a	1.250	0.500	0.250	0.0049	1.000	0.041	Def	0
37	Ce	a	1.750	0.500	0.250	0.0049	1.000	0.041	Def	0
38	O	b	1.000	0.250	0.250	0.0049	1.000	0.0299	Def	0
39	O	b	1.500	0.750	0.250	0.0049	1.000	0.0299	Def	0
40	O	b	1.000	0.750	0.250	0.0049	1.000	0.0299	Def	0
41	O	b	1.500	0.250	0.250	0.0049	1.000	0.0299	Def	0
42	O	b	1.750	0.250	0.000	0.0049	1.000	0.0299	Def	0
43	O	b	1.250	0.750	0.000	0.0049	1.000	0.0299	Def	0
44	O	b	1.250	0.250	0.000	0.0049	1.000	0.0299	Def	0

Duplicate x: -25, 17, -9, -1, 2

Duplicate y: -25, 17, -9, -1, 3

Duplicate z: -25, -17, -9, -1, 1

Slice

Oxy [001] Oyz [101] Oyz [100]

Slice thick. / nm: 1, 0.8, 0.6, 0.4, 0.2, 0, 0.1927

Sub-slices number: 120, 80, 60, 40, 20, 0, 4

Show slices

Figure 29: To slices dialogue.

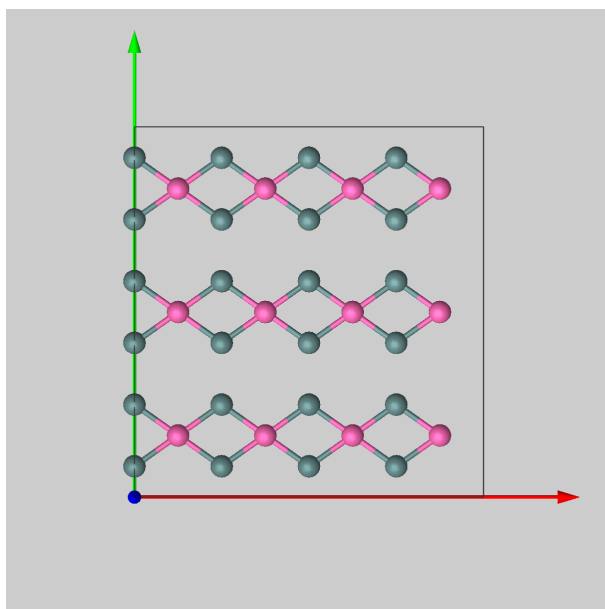


Figure 30: Slice 1.

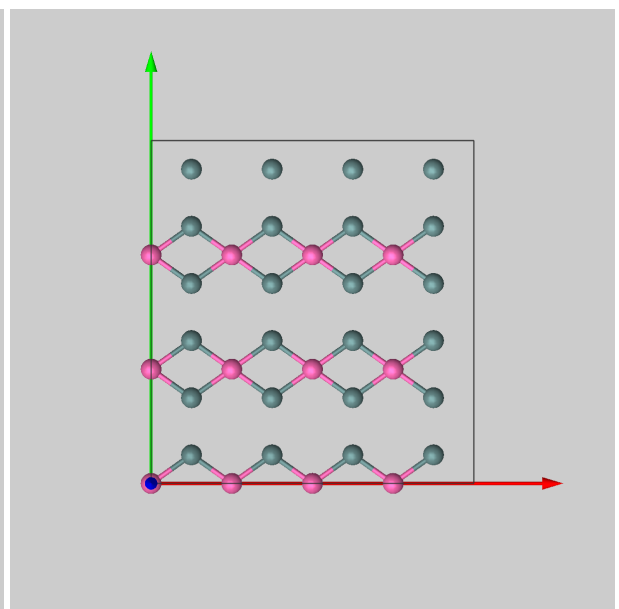


Figure 31: Slice 2.

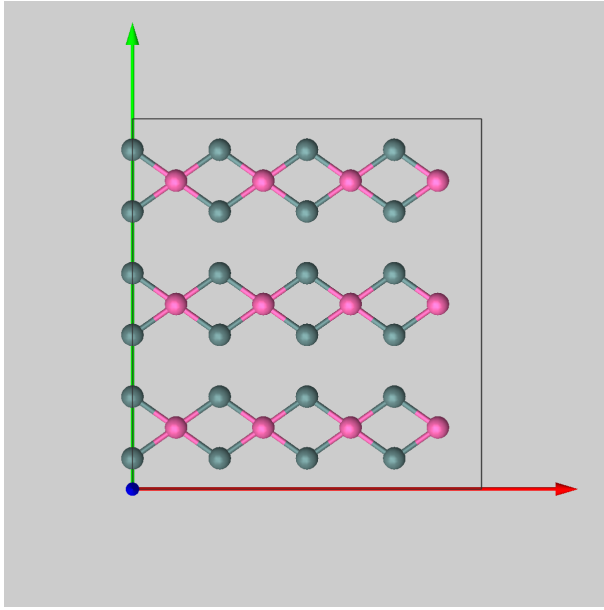


Figure 32: Slice 3.

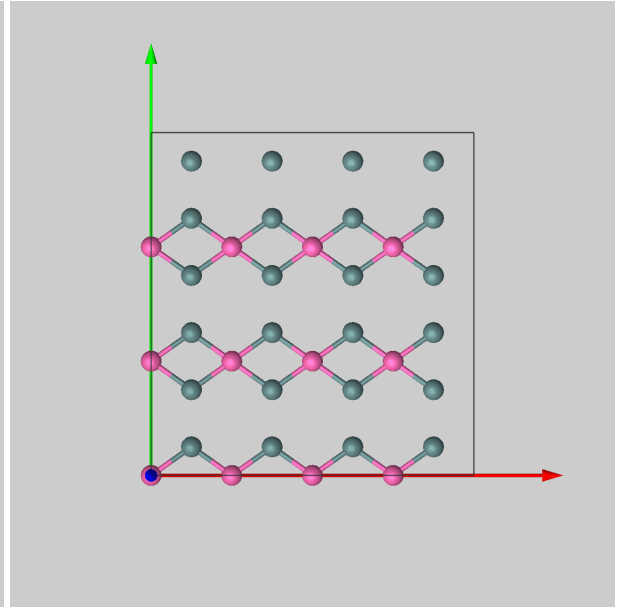


Figure 33: Slice 4.

Calculations using user defined slices are handled by the **Super-cell image** tab of the multislice window. The procedure is as follow:

- Start jems and load the first slice **[File]**→**[Open]** (Fig. 30).
- Open the multislice window and select the **Super-cell image** tab (Fig. 36).
- Select the **Cell** tab and using the either the **(+)** or **(++)** buttons load the 3 other slices (Fig. 36).
- Using the popup menu of the table (Fig. 37) duplicate the stack of slices. The whole stack is duplicated every time **[Duplicate]** is selected. The total crystal thickness is displayed. Any slice can be suppressed in order to introduce stacking faults.
- Start the calculation (**(Start)**).

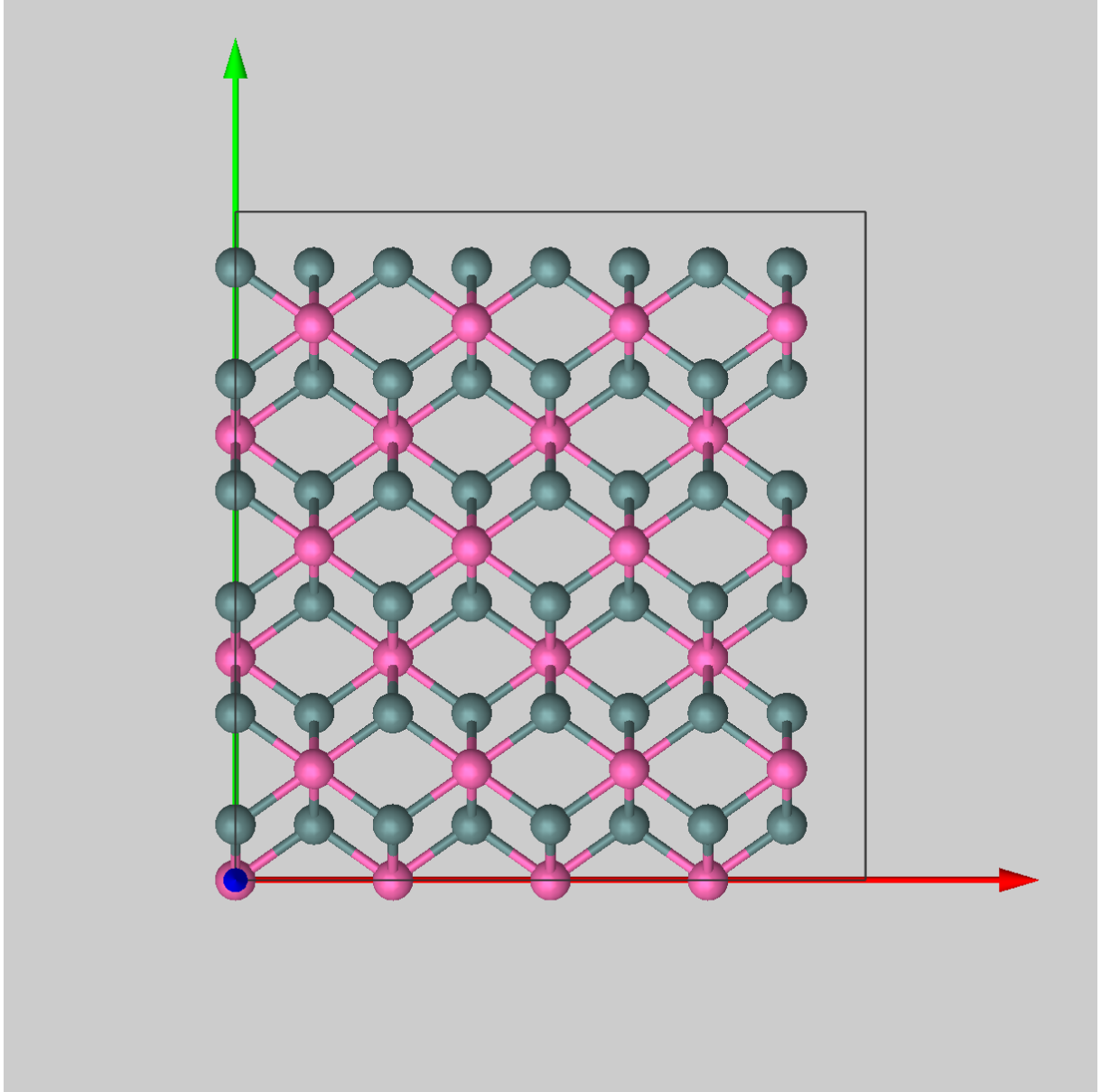


Figure 34: Stacked slices.

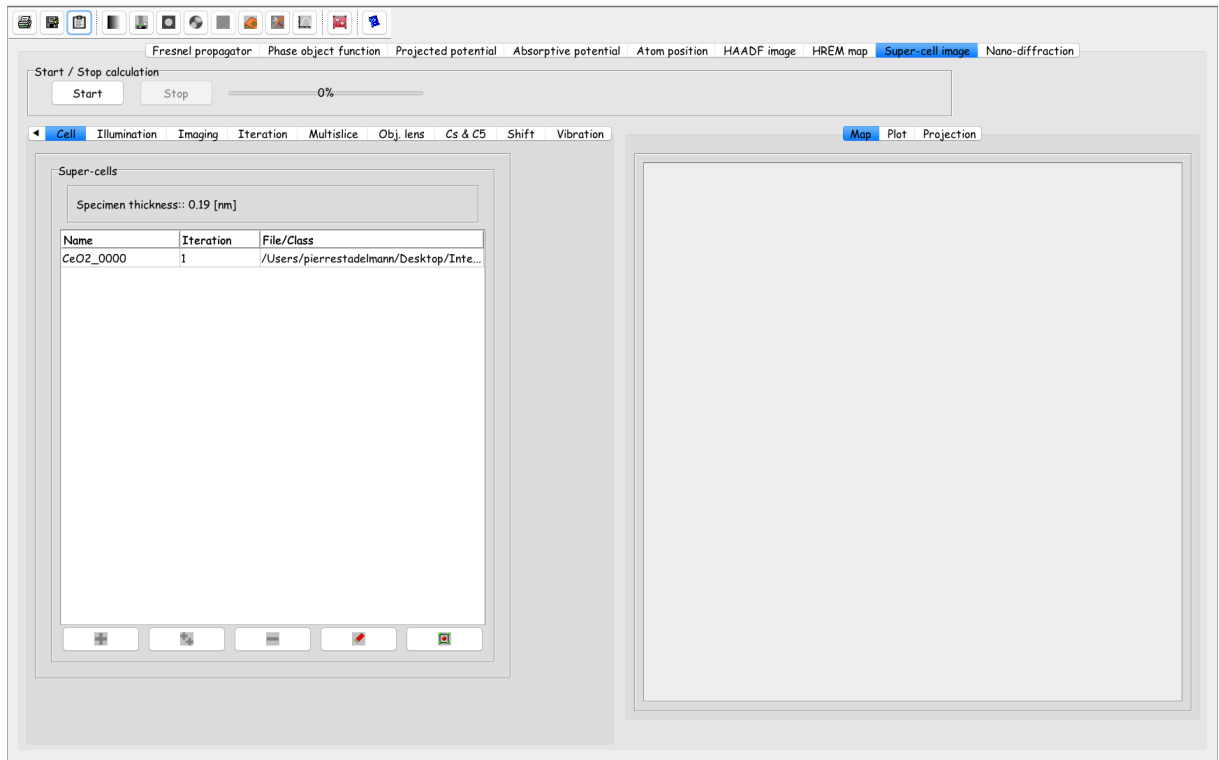


Figure 35: Multislice image calculations using user defined slices.

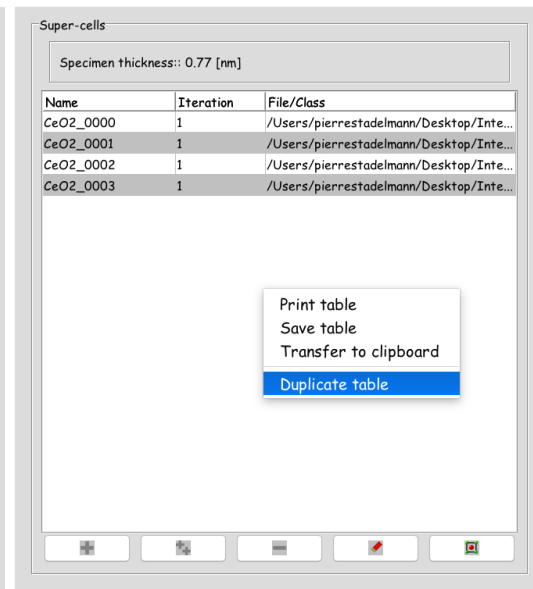
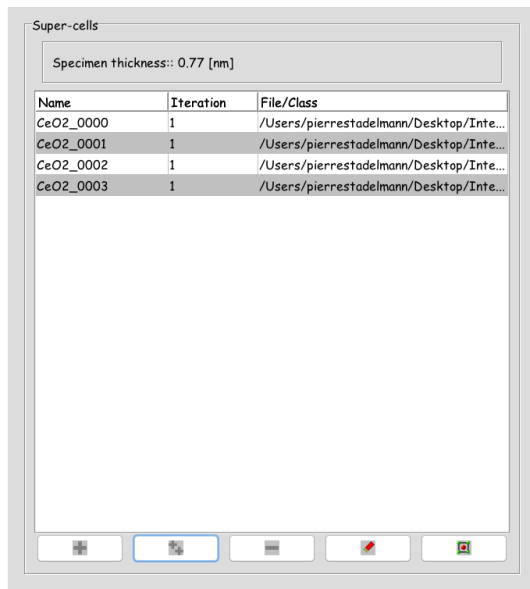


Figure 36: Stack of the 4 sub-slices.

Figure 37: Popup menu to duplicate the stack of super-cells.

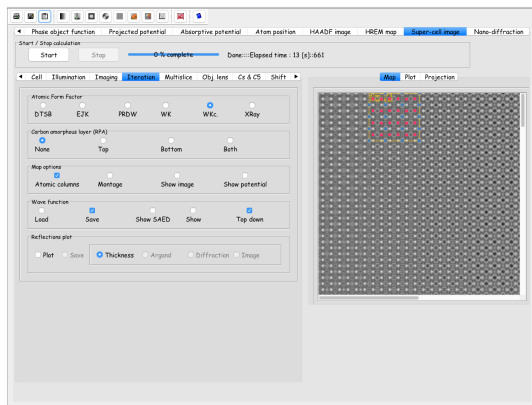


Figure 38: Super-cell imaging, **Iteration** tab.

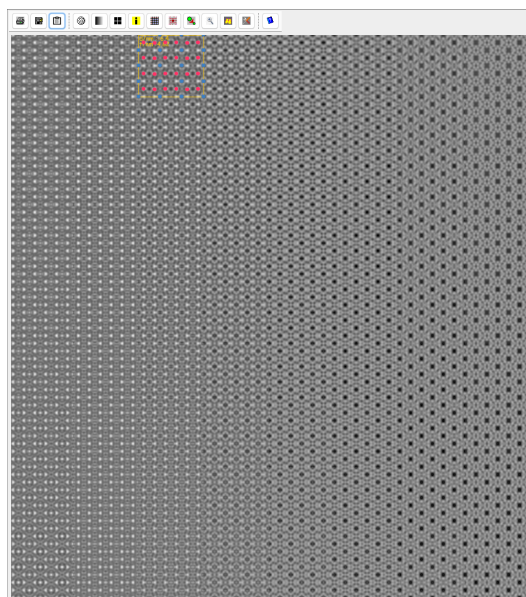


Figure 39: HRTEM images map.

2.4 Interactive image simulation using a multislice wave-function

The wave-functions obtained by the multislice procedure can be saved (check box **Save wave**) and used later in order to introduce, in real time, effects of tilted illumination, badly centred objective aperture, high-order aberrations, etc (Fig. 40). After selecting the save option (check box **Save wave**) redo the calculation. You will be asked to save the wave-functions.

Once the wave-functions are saved, close the *Multislicer* window and activate the HRTEM imager (**[Imaging]**→**[HRTEM imager]**). Load one of the saved wave-functions. The HRTEM imager window is shown in Fig. 41. The HRTEM image is shown on the right tabbed pane.

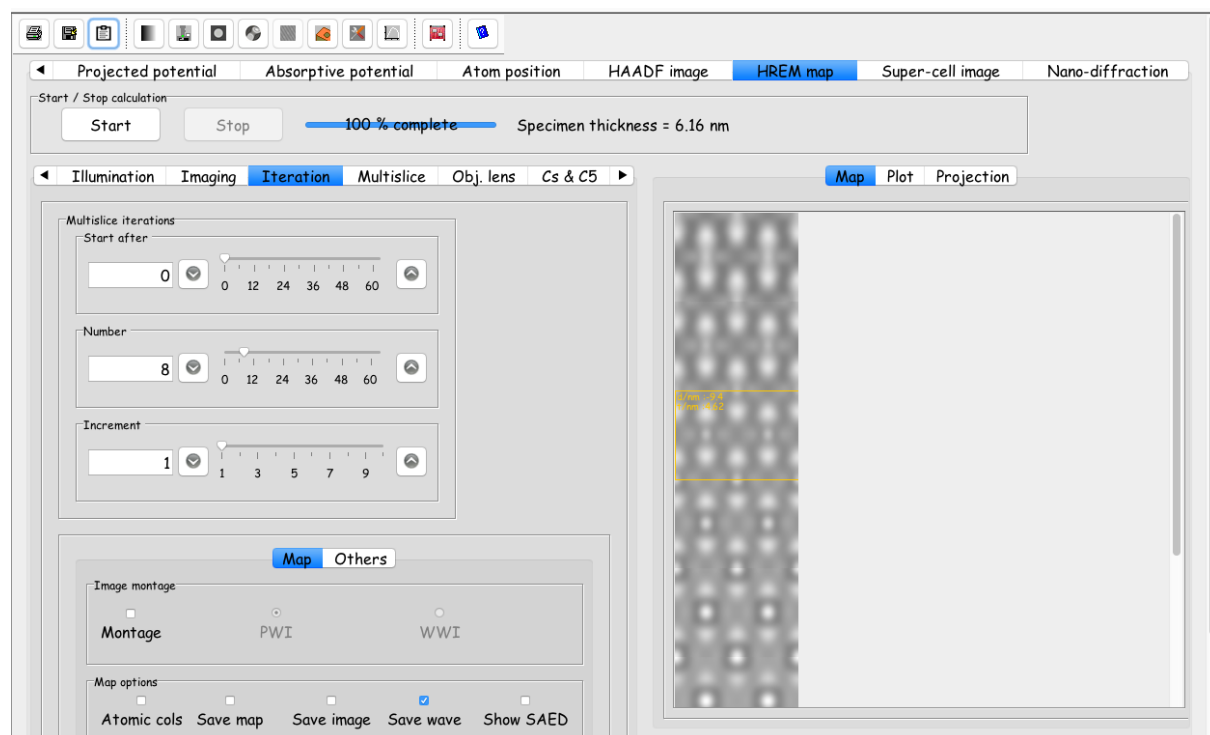


Figure 40: Saving HRTEM wave-functions.

The **Imaging** tab contains controls to duplicate the image (and wave-function³), to modify the brightness, contrast and gamma of the image and to add *Poisson*

³The geometry of the wave-function is not set according to the geometry of the images.

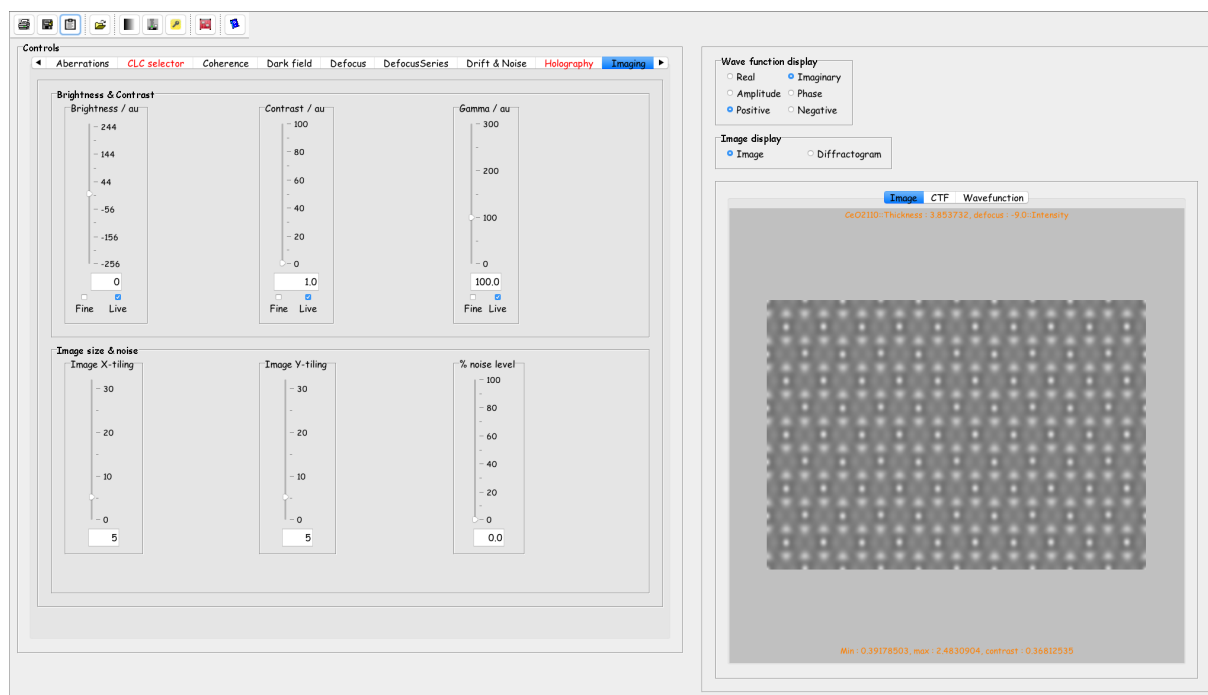


Figure 41: *HRTEM imager* window window allows to introduce wave-front aberrations up to order 8 (**Aberrations** tab). The imaging parameters are grouped in several tabs (**Coherence** tab: to set the illumination coherence , **Dark field** tab: to set the illumination tilt, etc).

noise to the image. The HRTEM imager also displays the contrast transfer function (CTF) and the wave-function (Figs. 42 and 43).

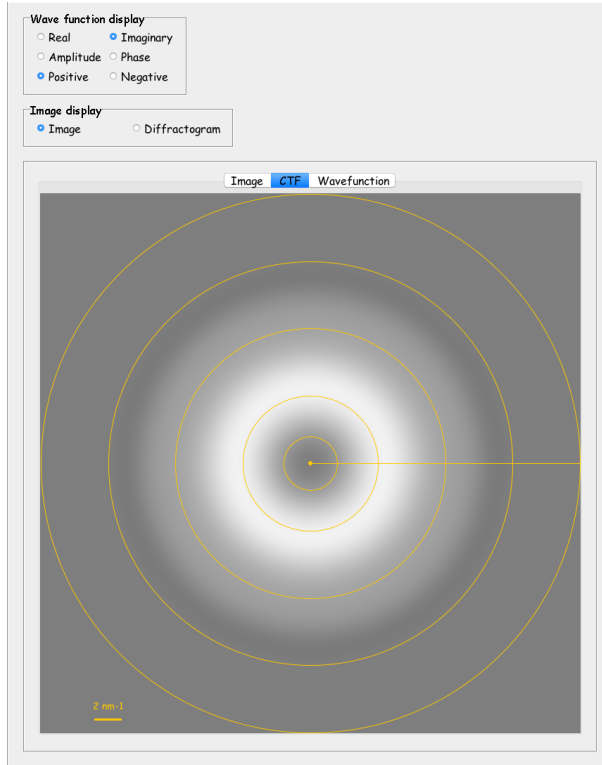


Figure 42: Contrast transfer function.

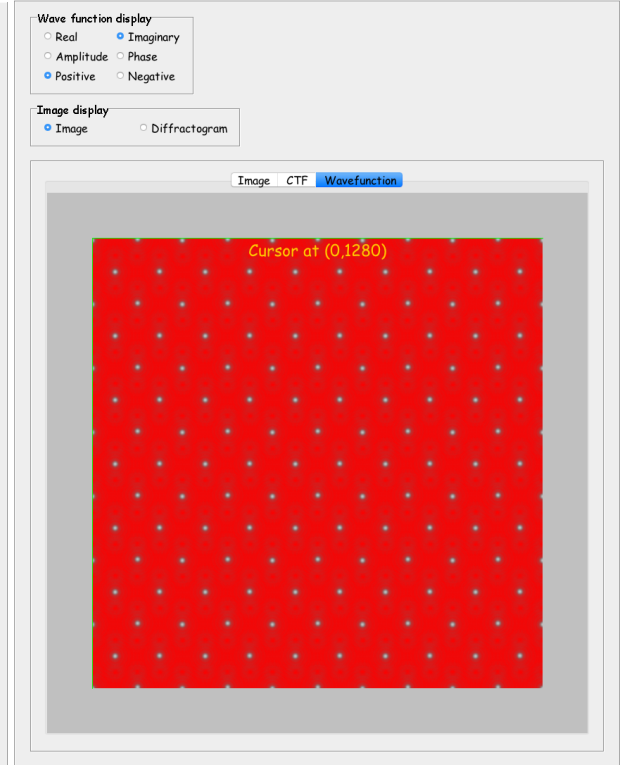


Figure 43: Wave-function.

As an example, any illumination tilt has a large impact on the HRTEM image contrast. Fig. 44 shows a tilted imaging condition optical axis shifted from transmitted beam position, Fig. 45 shows a tilted CTF, Fig. 46 shows the simulated HRTEM image (to be compared to Fig. 47). The optical axis position is controlled using the green arrow.

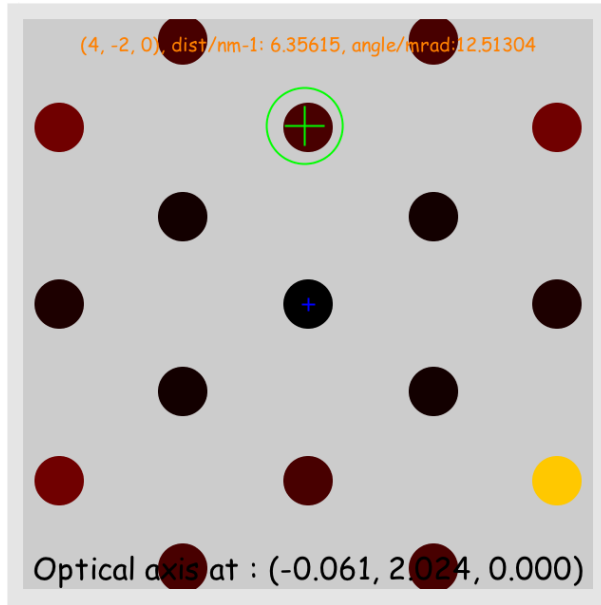


Figure 44: Dark field control.

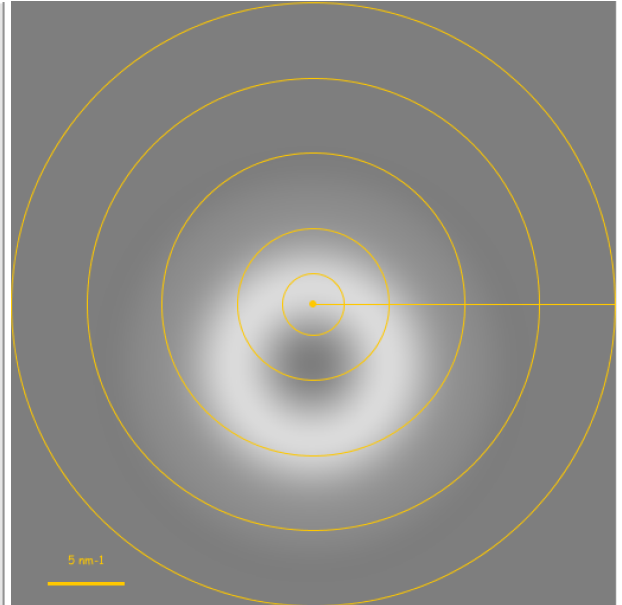


Figure 45: CTF under dark field condition.



Figure 46: HRTEM image under dark field condition.

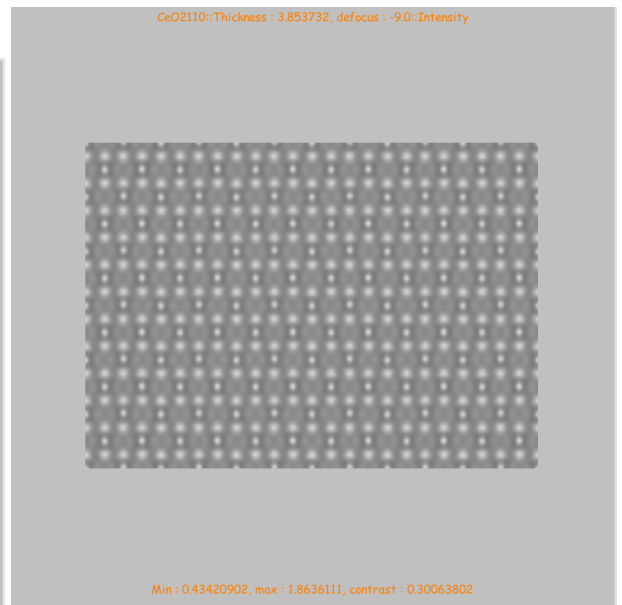



Figure 47: HRTEM image, aligned illumination.

3 Bloch-wave method

A much simpler way to do HRTEM image simulation for a particular [uvw] direction is to use the **Bloch-wave** method that does not require to slice the structure in order to introduce **High Order Laue Zone** effects. Moreover, when small unit cell crystals (like CeO_2) are imaged it is possible, using this method, to do interactive dynamical calculations and observe the effect of crystal thickness or crystal tilt (beside the illumination or objective aperture mis-alignments, of aberrations (up to order 8) or coherence) in (almost) real time.

Fig. 48 shows the *Blochwaver* window that controls dynamical calculation using the Bloch-wave method. By default the **HREM map** tab is selected and the  contains sliders to set the camera length and the number of reflections. Most of the tabs of the **HREM map** tab behaves similarly to those of the *Multislicer* window, only the **Iteration** tab contains slightly different controls.

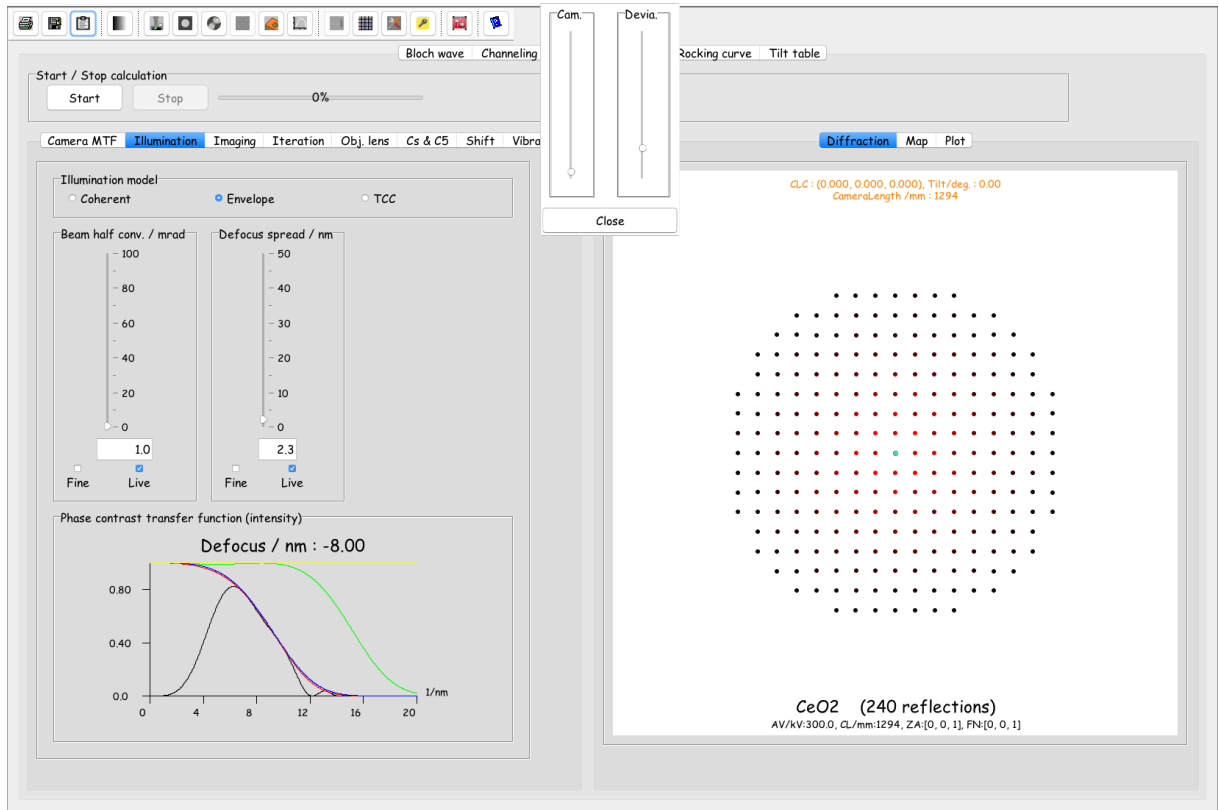


Figure 48: *Bloch waver* window allows to perform dynamical diffraction calculations and to simulate HRTEM images using the Bloch-wave method.

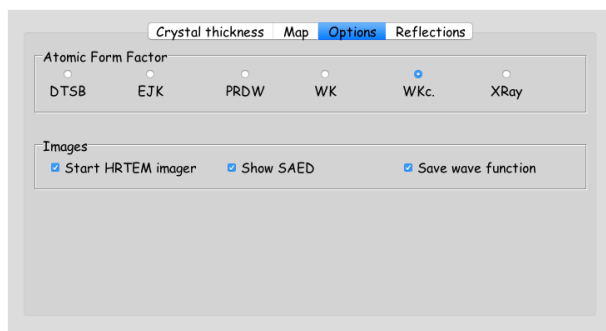


Figure 49: The **Iteration Options** tab allows to save the wave-functions and to start the *HRTEM imager* window.

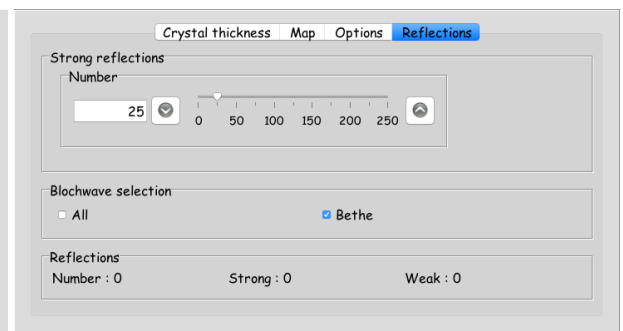


Figure 50: The **Iteration Reflections** tab controls the dynamical diffraction calculation.

The **Iteration Reflections** tab (Fig. 50) selects the reflections strong enough to enter the dynamical calculations, the weak reflections are taken into account by a Bethe perturbation of the strong reflections. Note that the Bloch-wave calculation time goes as n^3 where n is the number of strong reflections.

When the check box **Start HRTEM imager** is selected, the **HRTEM imager** window pops up and allows to change interactively the crystal thickness and orientation in addition to the what is possible using multislice wave-functions (Fig. 51). This tab is only activated when imaging wave-functions calculated by the Bloch-wave method.

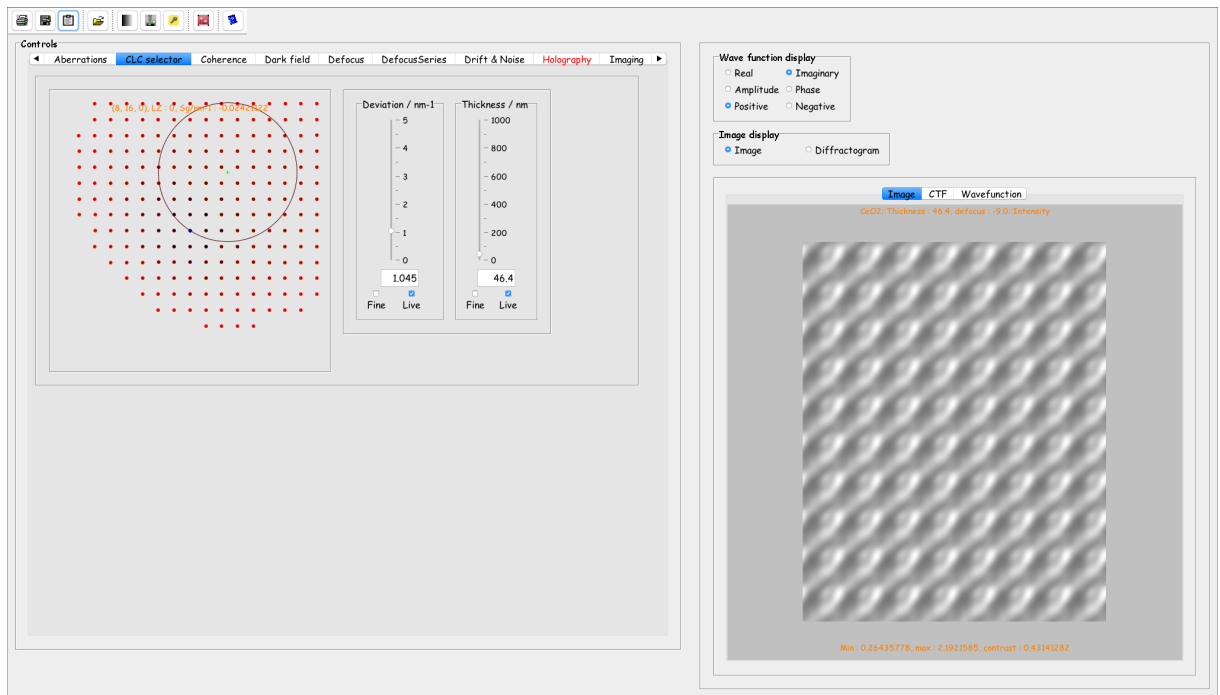


Figure 51: The **HRTEM imager CLC** tab contains controls to tilt the crystal and to vary its thickness.

Thickness and orientation changes can also be observed on the wave-function (Fig. 52).

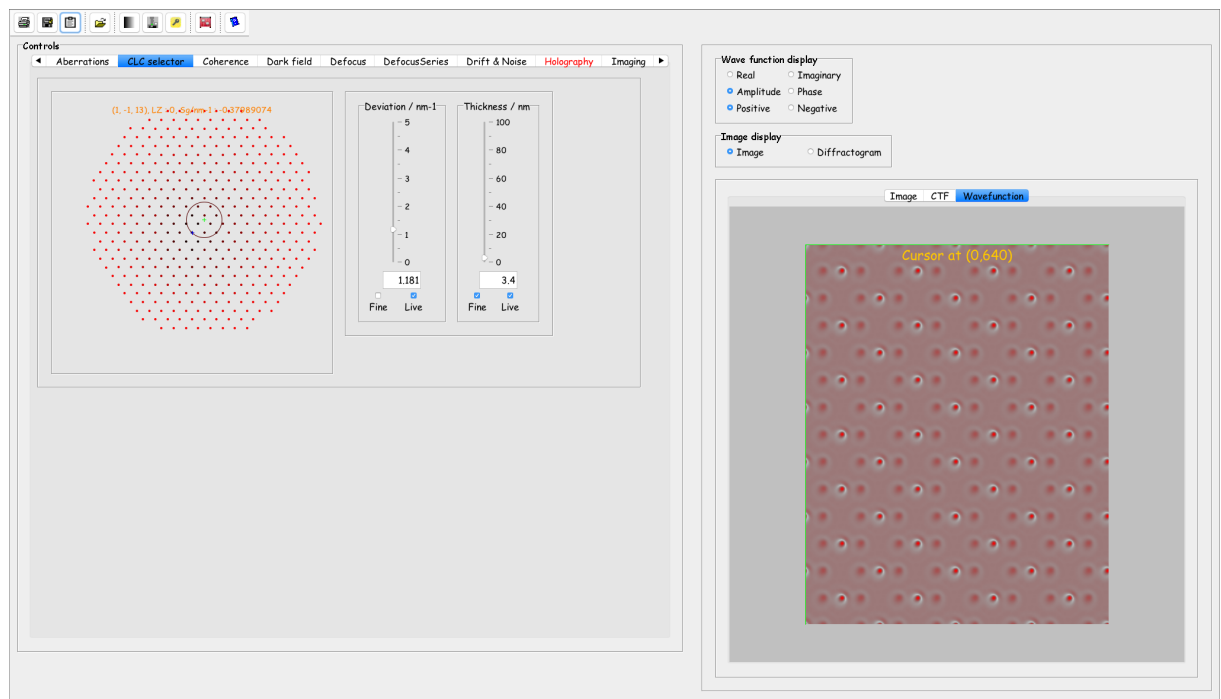


Figure 52: The **HRTEM imager CLC** tab contains controls to tilt the crystal and to vary its thickness. Effects on the wave-function are imaged.