Q-CBED USING JEMS

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1. Introduction

Convergent Electron Beam Diffraction (CBED) is a very sensitive Transmission Electron Microscopy (TEM) technique to perform measurements of specimen thickness. Not only specimen thickness can be assessed by CBED but also the proper specimen orientation (i.e. the (hkl) indices of the center of the Laue circle or CLC). A variant of CBED, Quantitative-CBED, is able to give precise complex value to the crystal structure factors, to evaluate the accelerating voltage, the Debye-Waller temperature factor or site occupancy of the atoms, etc. Q-CBED makes use of the standard X-Ray structure refinement approach where a model structure is inferred from the experimentally measured intensity of a large number of diffracted beams and its many parameters adjusted using a least-square fit procedure that compares the experimental diffracted beams intensity to calculated one.

In a single CBED pattern, the approach is basically the same except that a limited number of diffracted beams is recorded under hundreds or even thousands of different dynamical electron diffraction conditions. At present jems can analyze and fit a calculated linescan profile to an experimentally measured one. An extension of the procedure towards fitting several profiles or an portion of the CBED pattern is considered.

The overall process is accomplished using at least 2 diffraction patterns:

(1) A low magnification CBED pattern is used to determine approximately the diffraction conditions, namely the zone axis [u,v,w] indices, the (hkl) indices of center of Laue circle, the reflection (hkl) indices and their number.

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(2) A large magnification diffraction pattern is then precisely aligned using mouse controlled sliders and shifts. The alignment makes use principally of Kikuchi lines, ZOLZ (Zero Order deficiency lines) or HOLZ (High Order Laue Zone) deficiency lines and other CBED pattern details like spot diameter, camera length, accelerating voltage, etc.

The experimental profile is subsequently compared to simulated profiles (using the full dynamical electron diffraction theory) and the diffraction parameters and/or the structural parameters (i.e. Debye-Waller, occupancy, structure factors, ...) varied either manually or using a *Levenberg-Marquardt* algorithm. This procedure is described in the next sections.

2. Finding approximately the diffraction conditions

The first step consists in defining a model structure, in this document the CBED patterns are from **Al** [1]. The model structure can be loaded from the following folder which also collects images and diffraction conditions:

```
..\jemsData\ThicknessMeasurement
```

Figure 1 shows where and how to selected the Al structure.

The simple geometric comparison of the experimental CBED pattern with simulated diffraction patterns (kinematical approximation) is available as menu item **HOLZ pattern** of menu **Indexing** (Figure 2). It allows to figure out with a relatively good accuracy the experimental diffraction conditions.[2]

The various controls are organized in a *tabbed pane* where:

- **Align** allows rotating (rotation alignment) or using either the mouse (blue cross) or the arrow keys (up, down, left and right) moving the calculated pattern (this pane must be selected in order to have the keys action shifting the pattern center, i.e. the transmitted beam position, when not selected the CLC is moved).

[1] 100 kV accelerating voltage.

[2] Images can be loaded *As is*, *Interpolated* or *Subsampled*. This allows to load images acquired with large CCD cameras (see dialogue **Parameters - Preferences**, tab **Imaging**).

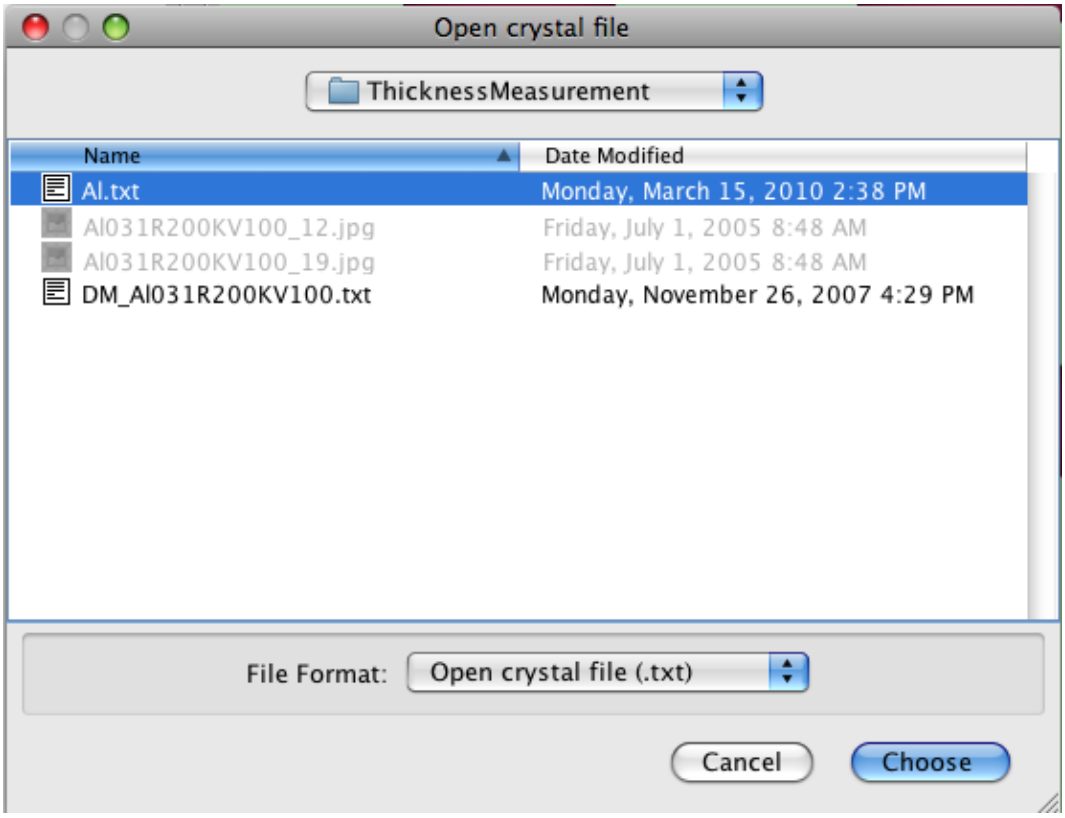

FIGURE 1. Loading the Al crystal structure.

- **Crystal** allows selecting the [uvw] zone axis indices, the number of Laue zones to display and moving the center of the Laue circle (CLC) using either the mouse (after selecting the green cross) or the arrow keys. These parameters can also be set using the specimen dialogue.

- **Diffraction** contains the controls for changing the acceptance angle of the objective lens, the camera length, the convergence of the illumination, the deviation, the accelerating voltage and to zoom the drawing when needed. It is possible to keep constant some of these controls in order to **not change** them when opening other boxes/dialogues [3].

- **Lattice** allows to deform the crystal lattice.

[3]The acceptance angle and other parameters are mostly set by default to low values in order to draw the SAED pattern as fast as possible.

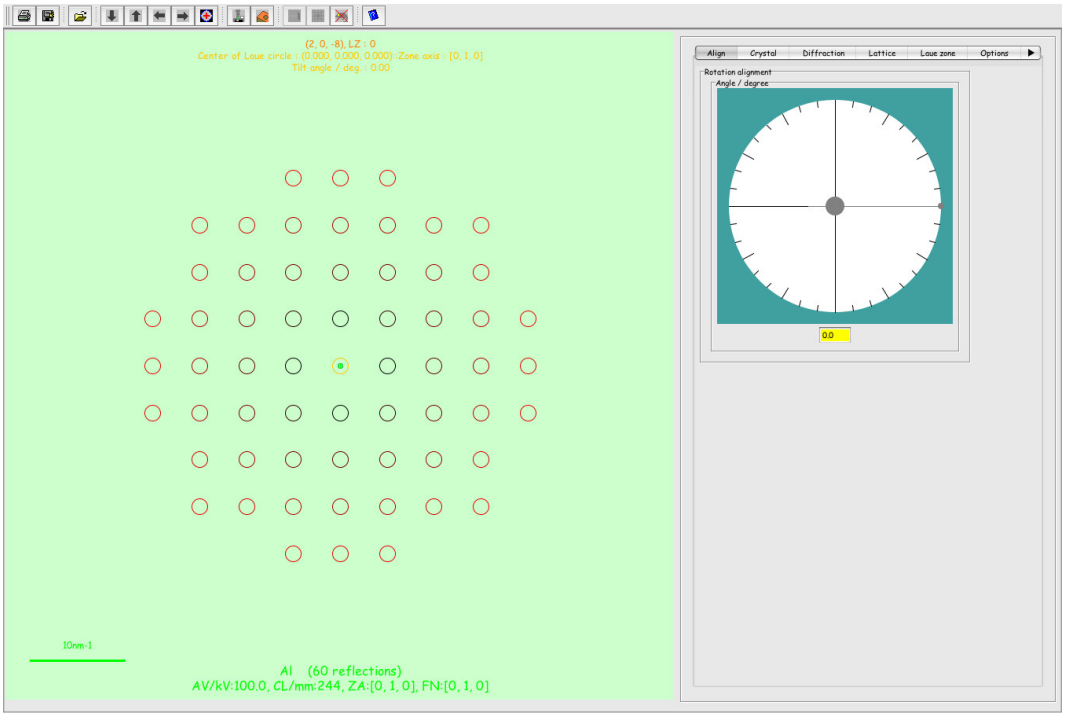

FIGURE 2. The HOLZ indexing box allowing to compare the geometry of experimental and kinematically calculated diffraction patterns.

- **Laue zone** has radio buttons to boost the intensity of HOLZ lines when the **LZ colors** of the **Options** tab is selected.

- **Options** contains check boxes to decorate the simple SAED kinematical drawing with HOLZ lines, Kikuchi lines, double diffraction spots, ..., and Zero Order Laue Zone deficiency lines which very useful to find the diffraction conditions.

- **Peak list** will provide a list of the (x, y) coordinates of the experimental SAED spots when the experimental pattern is processed using the **Find peak** kernel of the **Processing** tab.

- **Processing** contains image processing options and features to improve (hopefully) the visibility of spots/lines of the experimental diffraction pattern. A pretty useful one is **Colorize** (see Figure 3).

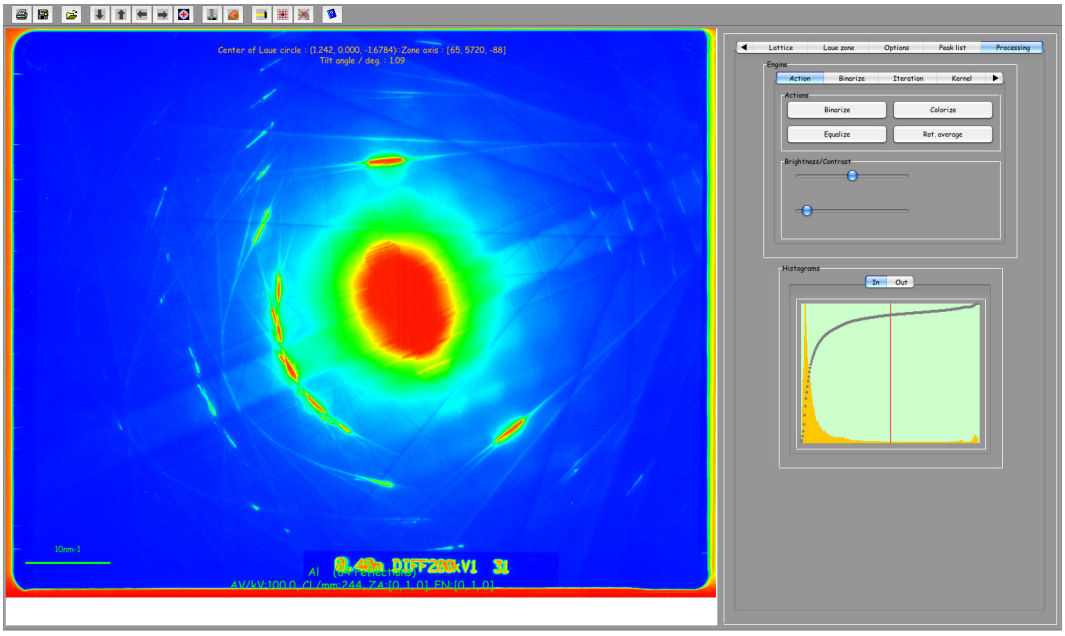

FIGURE 3. A colorizedCBED pattern (Al near [031]).

Figure 4 shows the window as it appears after loading the SAED pattern. The pattern has been colorized (the default [0,0,1] orientation is selected).

Using the *Circles*, *HOLZ*, *Kikuchi*, etc options of the *Options* tab, the calculated pattern is decorated with lines, etc that help determine the diffraction conditions. The crystal is tilted under mouse control or using the arrow keys until a good match with the experimental pattern is obtained. Indeed any experimentally obtained diffraction condition is of great help, i.e. accelerating voltage, camera length, shift of HOLZ lines position, ... This kinematical procedure does not consider specimen thickness.

The matching conditions are finally saved using the *Dead mouse* tool button (Figure 6). The diffraction condition are saved on file:

`..\jemsData\ThicknessMeasurement\Al031R200KV100.txt`

3. CBED PROFILE SELECTION

With the diffraction condition approximately defined, the large camera length CBED pattern can be loaded into the thickness measurement box (Figure 7). It

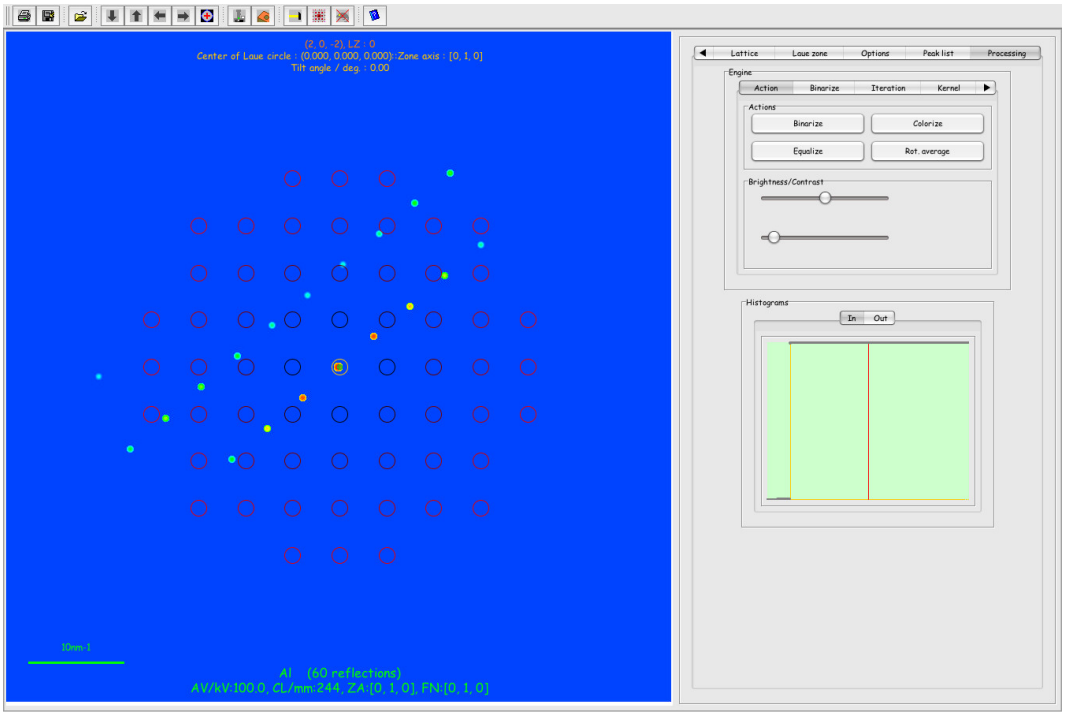

FIGURE 4. A low magnification SAED pattern with superimposed ZOLZ lines.

might be necessary to set the center of the CBED pattern, to rotate it, to rotate, scale and tune the CLC. The controls that allows this tuming operate as in the *HOLZ index box*. Figure 8 shows the kinematical CBED pattern after adjustment of the camera length, beam convergence, center of Laue circle and center of the diffraction pattern. These diffraction conditions should again be saved using the *DeadMouse dialogue* in order to very quickly be able to set them again.

The next operation is to get a profile from the CBED pattern. This is obtained by selecting the **Align** tab and the **Profile mask** radio button. A rectangular box is drawn on the CBED pattern, the blue part correspond to the (000) spot and the red parts to the (200). Have the mouse clicked on the green cross and drag the rectangle. The rectangle can be placed in between the reflections or centered in one of the reflections. Use the left square to broaden the rectangle (mouse click on the square and drag). A mouse click on the little yellow disk allows to rotate the rectangle. Mouse click and drag the middle square to reduce the height of the rectangle until a satisfactory profile is obtained. Figure 9 shows the selection of a profile (rocking-curve). [4]

[4]Do not select select more than 20-30 reflections when starting a new fit. Adjust the reflections number using the **deviation** slider of the **Diffraction** tab.

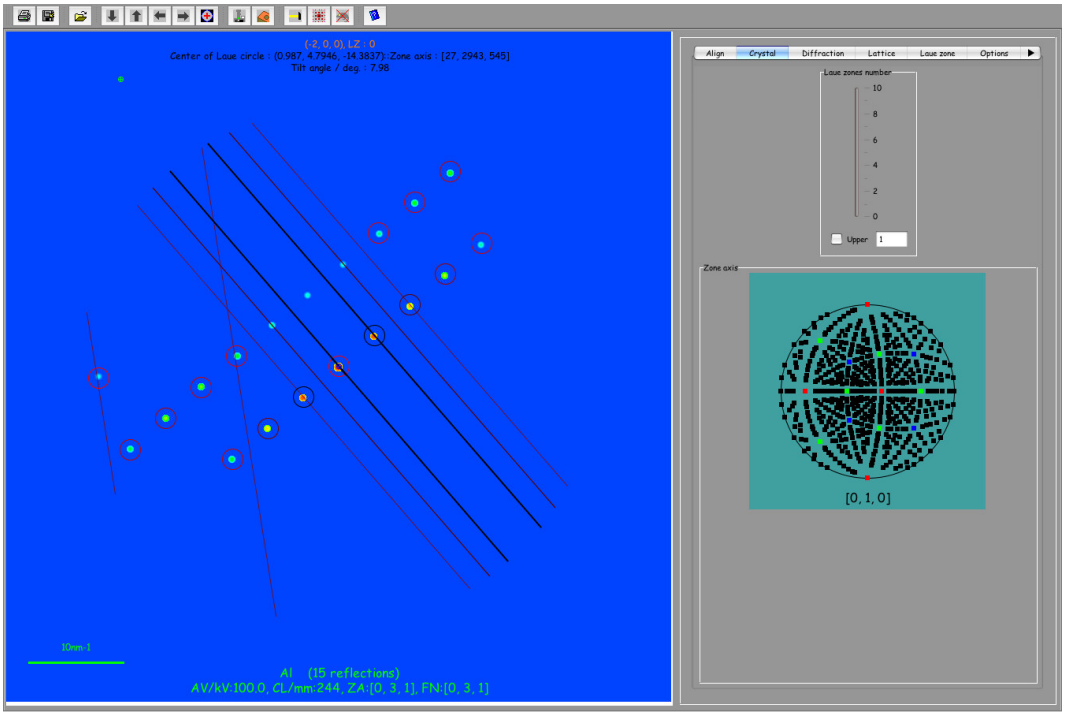

FIGURE 5. A low magnification SAED pattern with superimposed ZOLZ lines.

4. Specimen thickness measurement using the two-beams dynamical electron diffraction theory

When a suitable profile is selected use the **hammer** tool to process the profile. Figure 10 shows the experimental profile, the two-beams dynamical profile (rocking curve) and the difference (absolute value) together with the deviation error defined by the profile.

The upper part of the **Profile Processing Box** contains check boxes and radio buttons to tune the fitting procedure (Figure 11):

- **Delta** ...

By default, a 2-beams dynamical calculation is performed and a first guess of the specimen thickness usually obtained. When the fit is poor, it is necessary to set manually the specimen thickness. For that purpose several controls are gathered in different tabs. The **Controls** tab groups sliders to scale the deviation, the fog (constant background) and the specimen thickness. The **Scale** control is dimmed since by default the scale of the calculated profile is adjusted in order to minimize

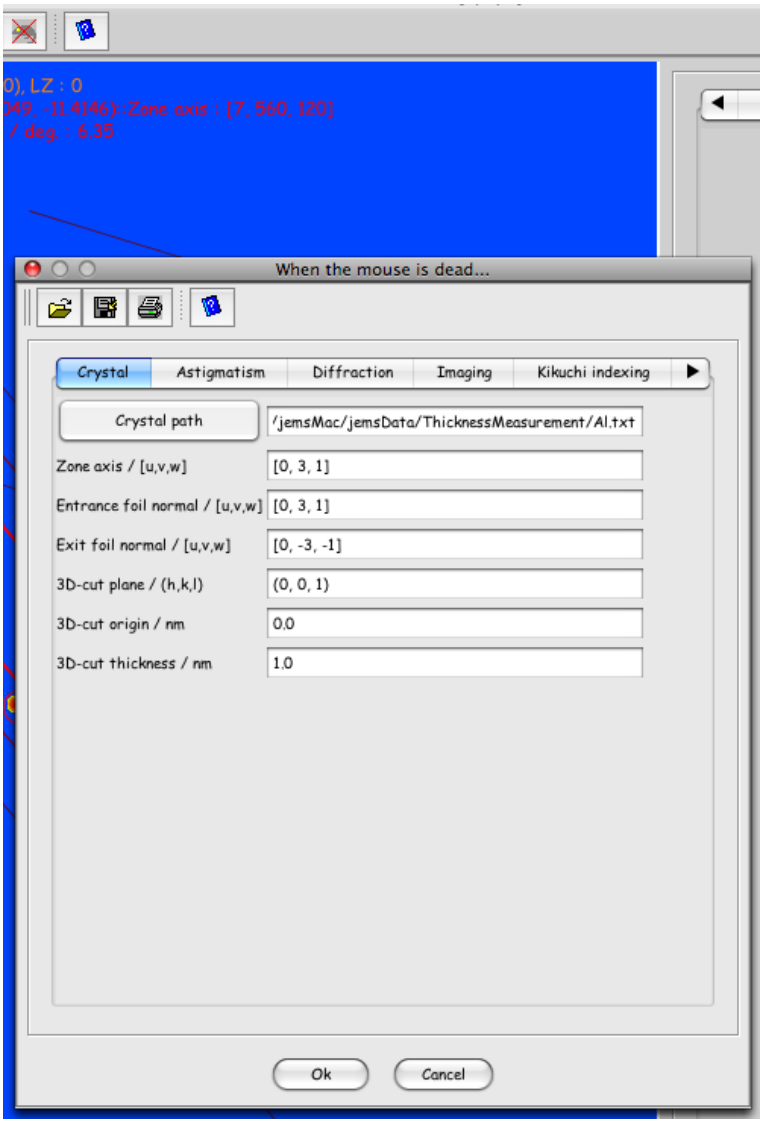

FIGURE 6. The *DeadMouse* dialogue keeps the value of the diffraction conditions.

the difference experimental-calculated profile. Since the profile is extremely sensitive to small changes of the specimen thickness, the effect of setting a larger or smaller thickness affects immediately the simulated profile and the residuals.

It is also possible to adjust the CLC (hkl) indices and to shift the position of the transmitted beam. This can account, together with the deviation for non uniform spacing of the profile samples. After proper manual adjustment of these parameters (when needed), a least-square fit can be performed (two-beams dynamical

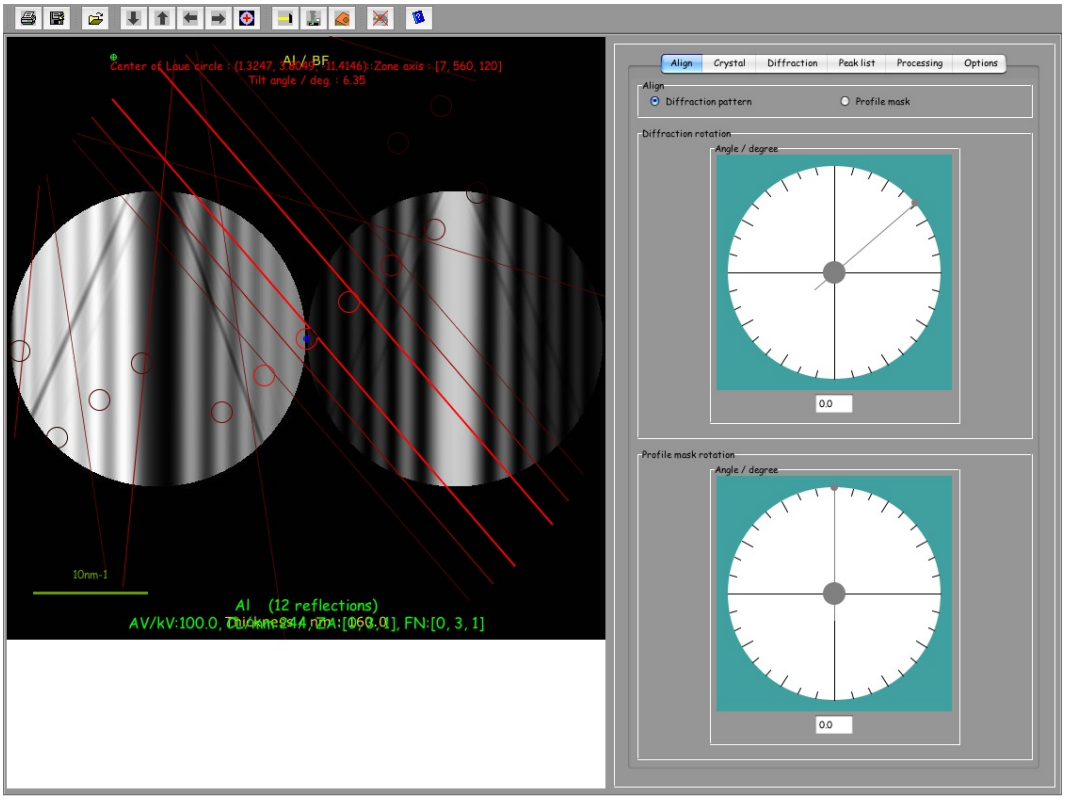

FIGURE 7. The CBED pattern is loaded in thickness measurement box.

calculation). Figure 13 shows the calculated two-beams rocking curve. The fit sets the starting values of the diffraction parameters of the many-beams dynamical calculation.

5. Q-CBED USING MANY-BEAMS DYNAMICAL ELECTRON DIFFRACTION THEORY

The many-beams rocking curve fit controls are placed in **Many beams** tab (Figure 14).

The **Fitting** tab allow to control the number of strong reflections introduced in the dynamical calculation, the **Bethe** check box introduces the weak reflections as a perturbation of the structure factor of the strong reflections. The **Show progress** check box when selected allows to draw during the profiles as the fit is progressing. A manual control of several diffraction parameters is available in tabs

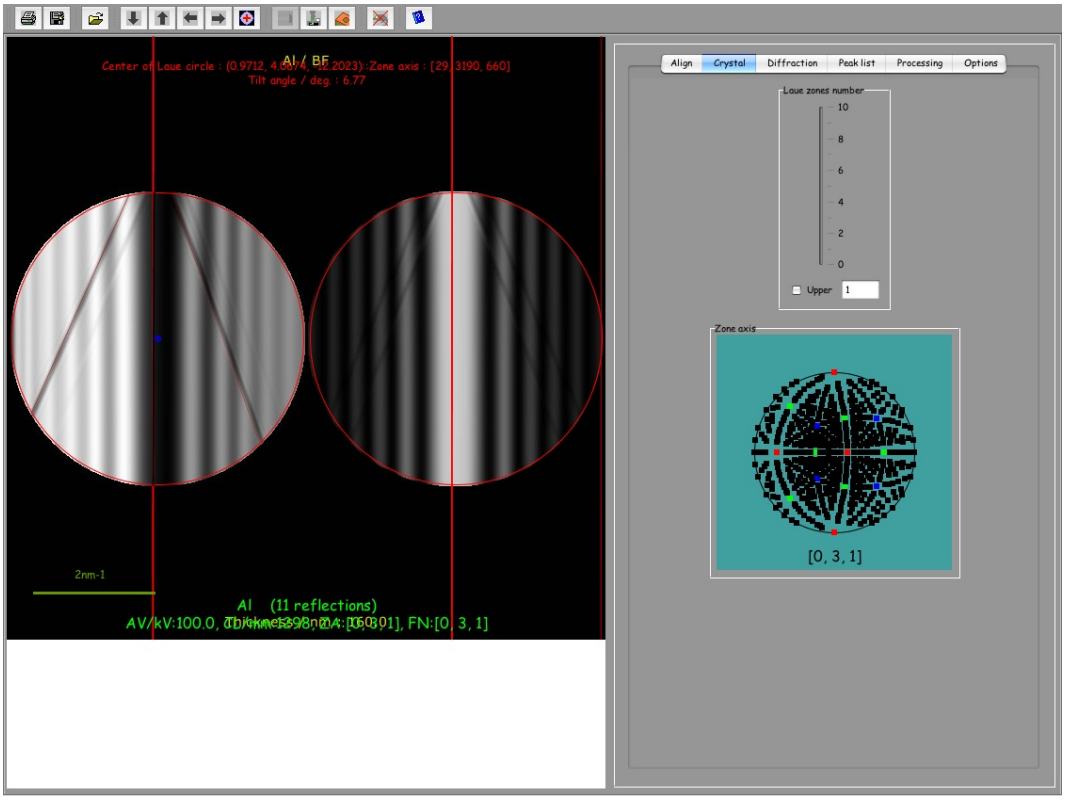

FIGURE 8. The CBED diffraction conditions are tuned.

CLC, **Coherence**, **Controls** [5].

The **Fit** parameters are grouped the 3 tabs:

(1) **Parameters** tab groups the fitting of the microscope parameters.

(2) **DW & Occ. &Abs** tab allows fitting the Debye-Waller, site occupancy and absorption of the different atoms (grouped by orbit).

(3) **Struct. fact.** allows to fit the complex structure factor of one, several or all reflections participating to the dynamical rocking-curve calculation.

[5]By default the illumination is assumed incoherent, i.e. the intensity of overlapping CBED disks is added.

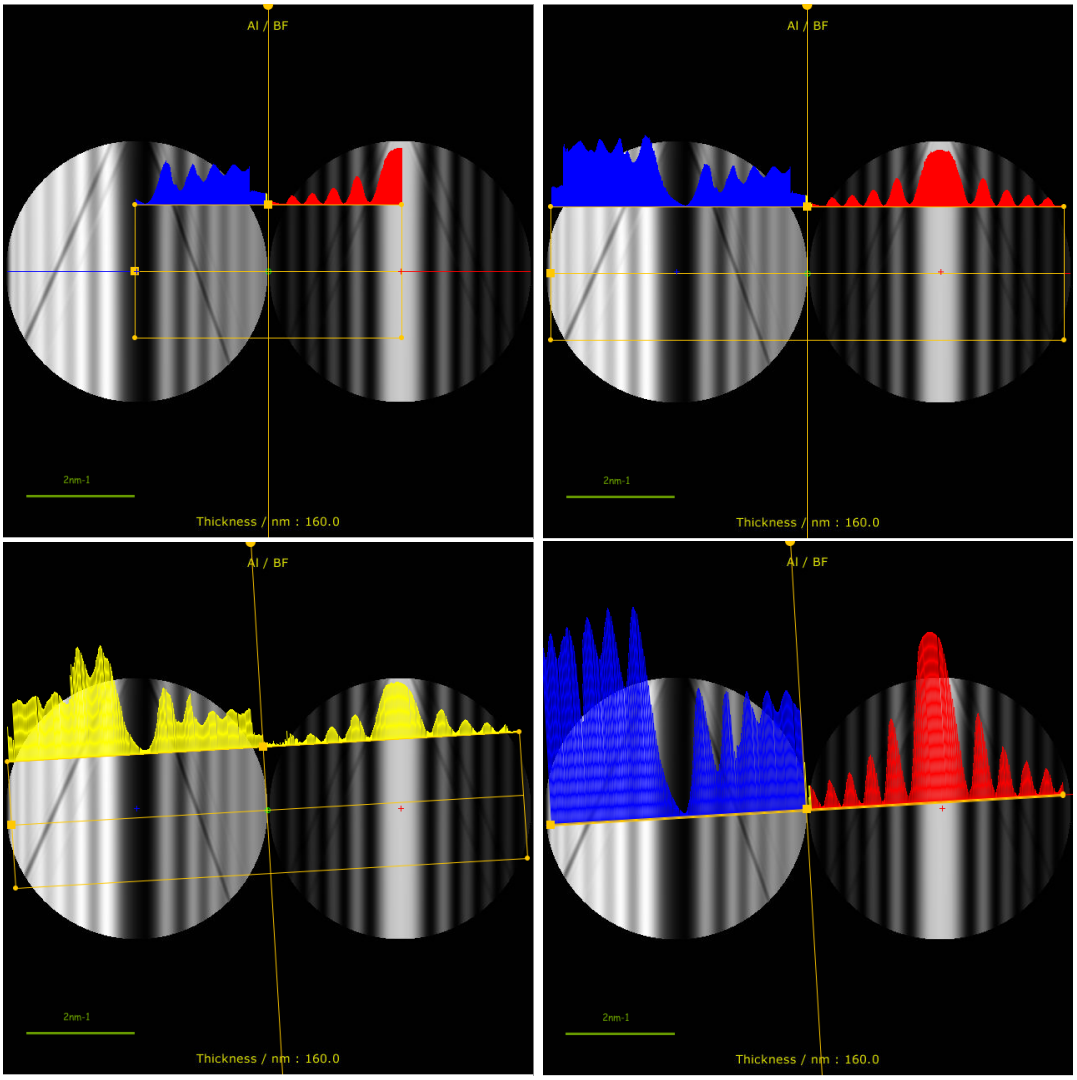

FIGURE 9. The selection of a profile across the (000) and (200) disks.

A good strategy is usually to first perform the fit of the microscope parameters, than the Debye-Waller temperature factor and finally the structure factors. The following figures illustrate the many-beams fit. Figure 15 shows the rocking curves after fitting the microscope parameters, Figure 16 a new fit with the Debye-Waller temperature factor, and finally Figure 17 after another fit where several structure factors are adjusted.

The rocking curves and fitted parameters are available in a table (Figure 18) where the CLC of a particular rocking curve position, the experimental and calculated

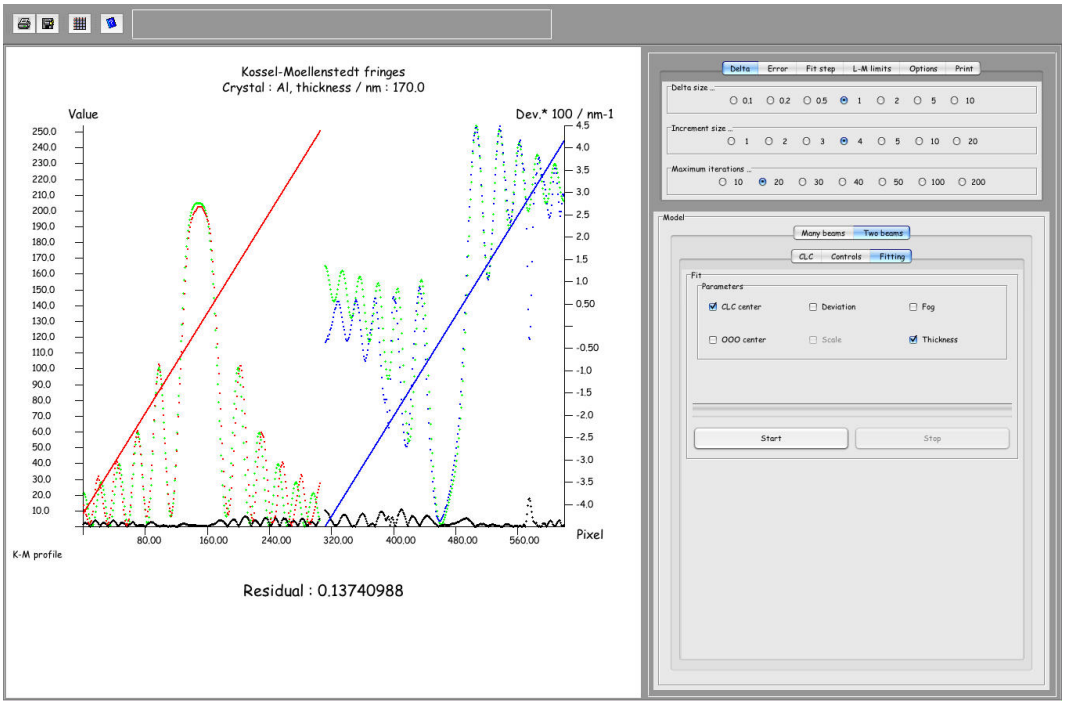

FIGURE 10. The profile processing box showing the 2-beams dynamical profile, the experimental profile and their difference.

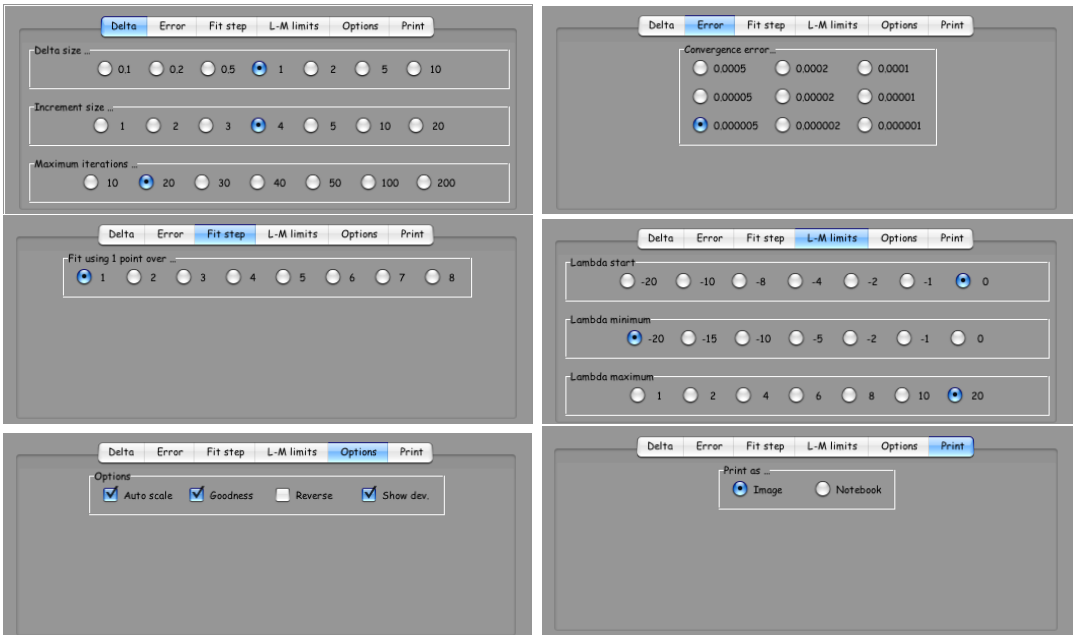

FIGURE 11. Fitting procedure controls and options.

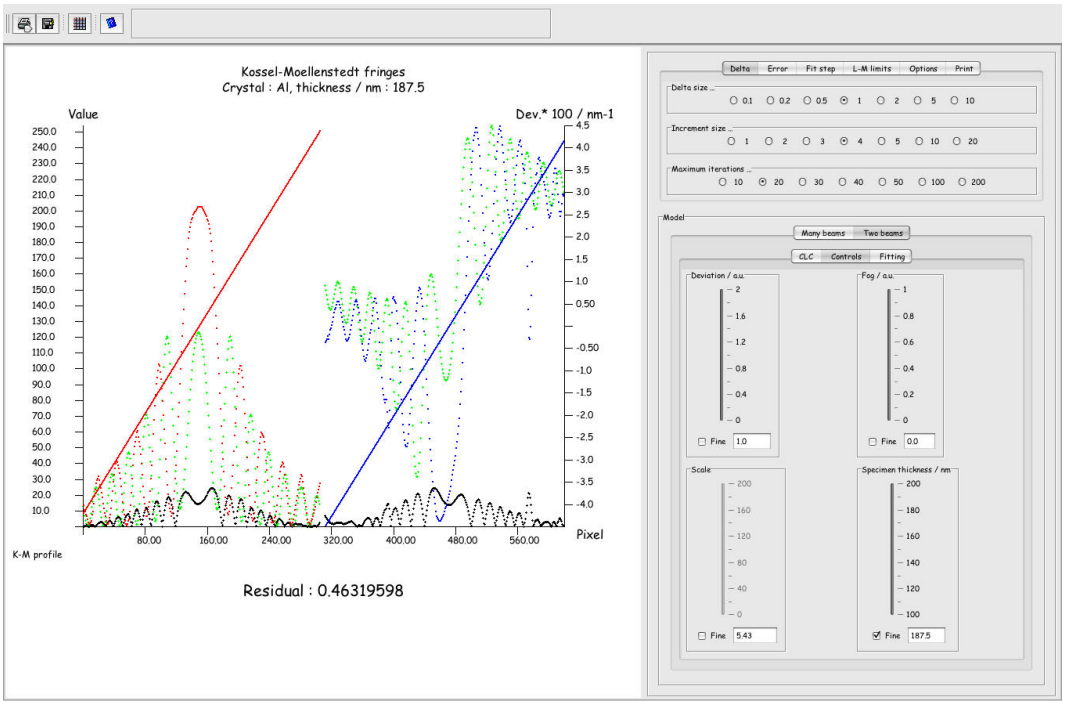

FIGURE 12. The thickness can be adjusted manually to get a better match experimental-simulated pattern.

values are tabulated. The table can be printed or saved in the form of a (very) simplified *Mathematica* notebook (Figure 19).

6. REMARKS

- The fitting of structure factors using the rocking curve profile requires energy filtered CBED patterns.

- There is no absolute strategy to fit a rocking curve. Though it is always possible to adjust almost all diffraction conditions and parameters as well atomic parameters, it is always better to determine very accurately using known specimen the accelerating voltage.

- Some parameters should be fitted with great care since they might have opposed effects on the fit (for example thickness and Debye-Waller).

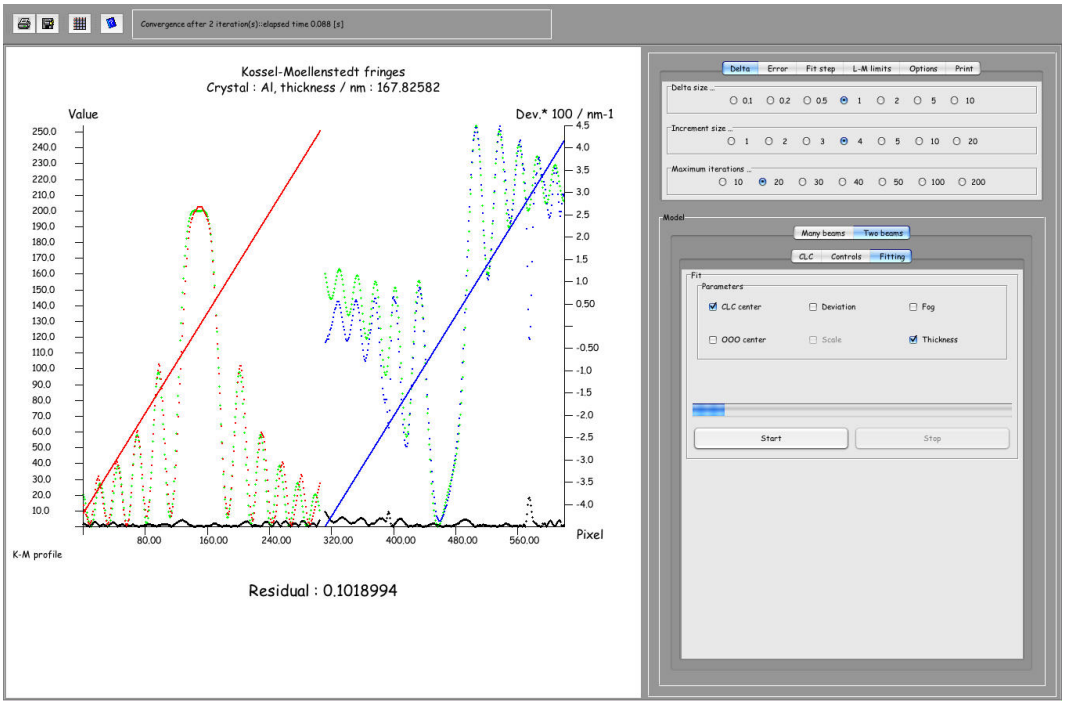

FIGURE 13. The best two-beams dynamical rocking curve calculation.

7. Introducing the fitted structure factors in the CBED calculation

The structure factors of any CBED calculation are generally calculated using different sources of atomic scattering amplitudes. jems offers the possibility to do CBED calculations with Q-CBED fitted structure factors. Particular fitted structure factors, either in complex form **(real, imaginary)** ([Volt], [Volt]) or Argand form **(amplitude, phase)** ([Volt], [Degree]) can be specified in an editable list provided by the *Vhkl editor* as shown in Figure 20.

When checked the structure factor can be modified either by the user or directly by jems. In Figure 21 the phase of the four (2,0,0) reflections has been set to 45 degrees (the real and complex part are automatically updated when the amplitude and phase are given as well as the amplitude and phase are updated when the real or imaginary part of a structure factor is given). Deselecting any check box makes the associated structure factor non editable and the CBED calculation will use it.

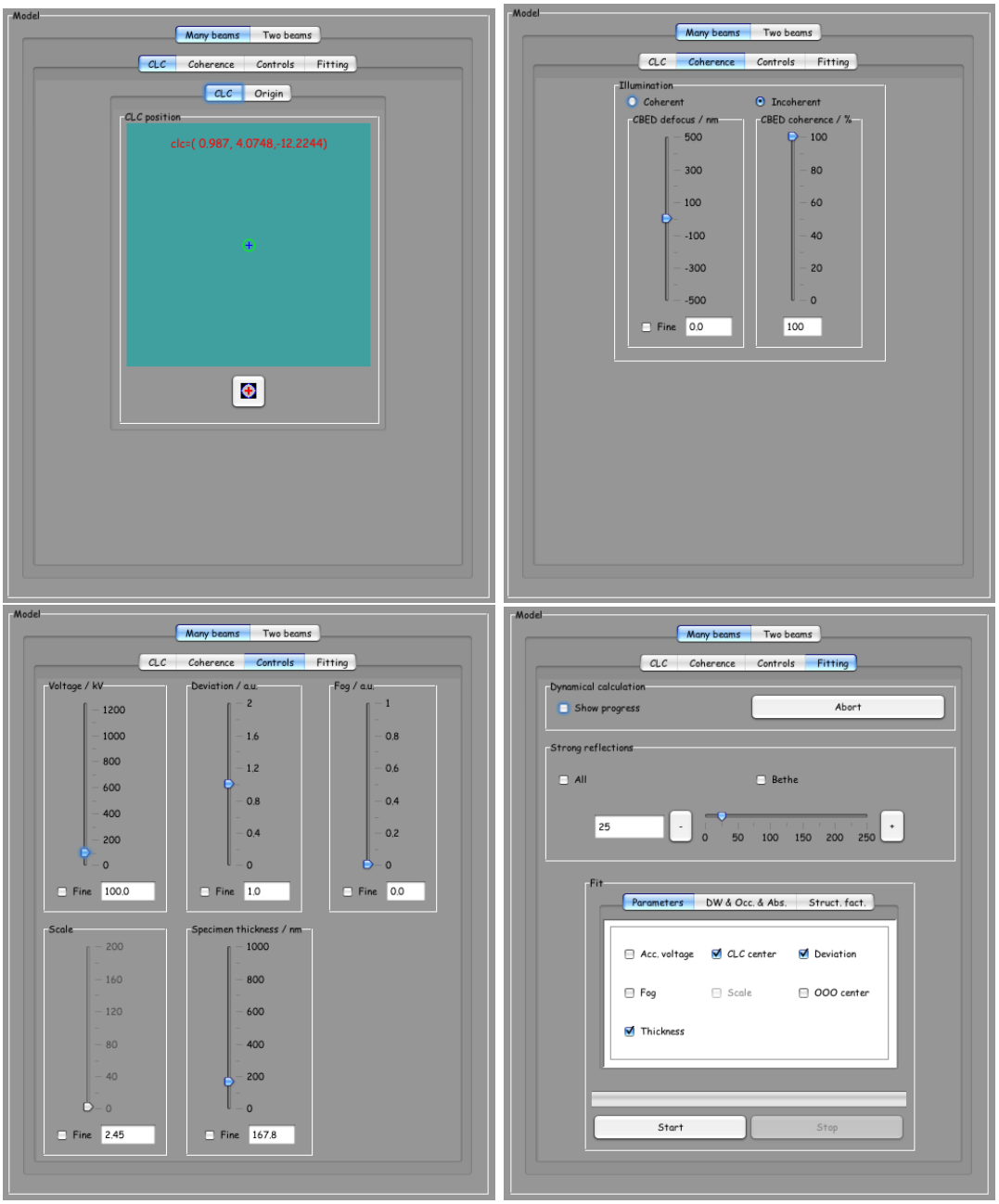

FIGURE 14. The many beams rocking curve calculation controls.

Figures 22 and 23 show CBED patterns calculated with the modified and standard four (200) structure factors.

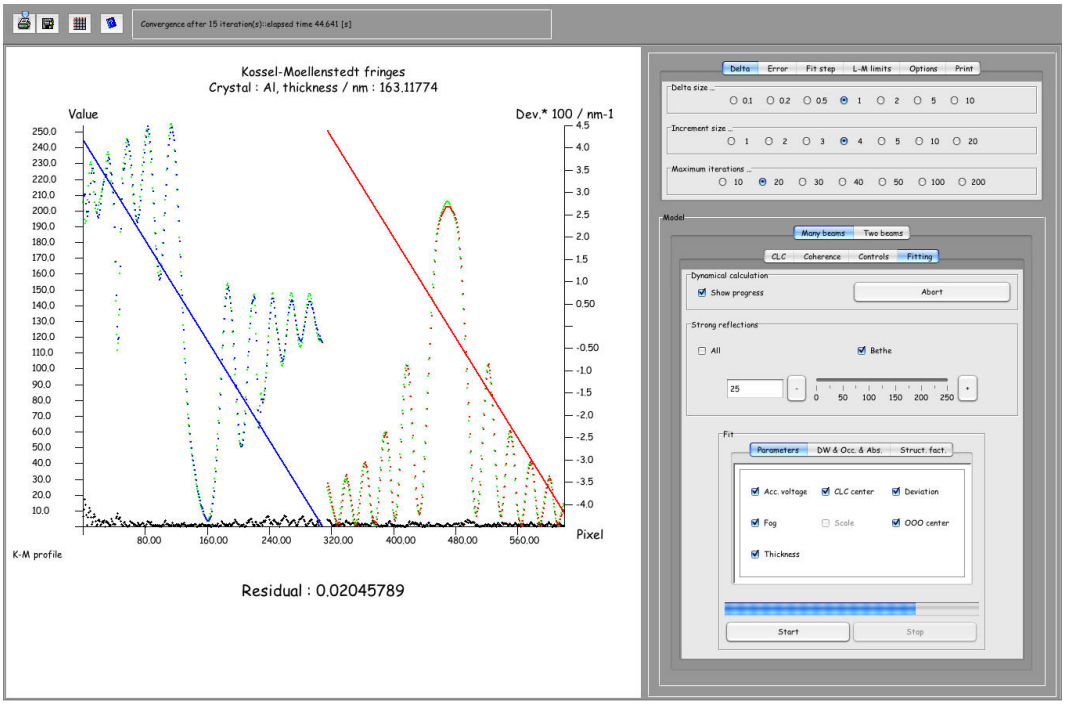

FIGURE 15. Rocking curves and residual after the fit of the microscope parameters.

7.1. **Saving and loading the deselected structure factors.** The ***Load*** and ***Save*** buttons at the bottom of the form are for loading structure factors and saving the modified structure factors (the standard structure factors are not loaded nor saved). Only *deselected* structure factors are either loaded or saved on the file. The same file can contain several sets of user specified structure factors as shown below. The load process can be interrupted before reading any SF set. A match between the deselected reflections of the form and the reflections of the file is performed. It is thus possible to store on one file reflections belonging to different zone axis, only the reflections of the current zone axis will be loaded.

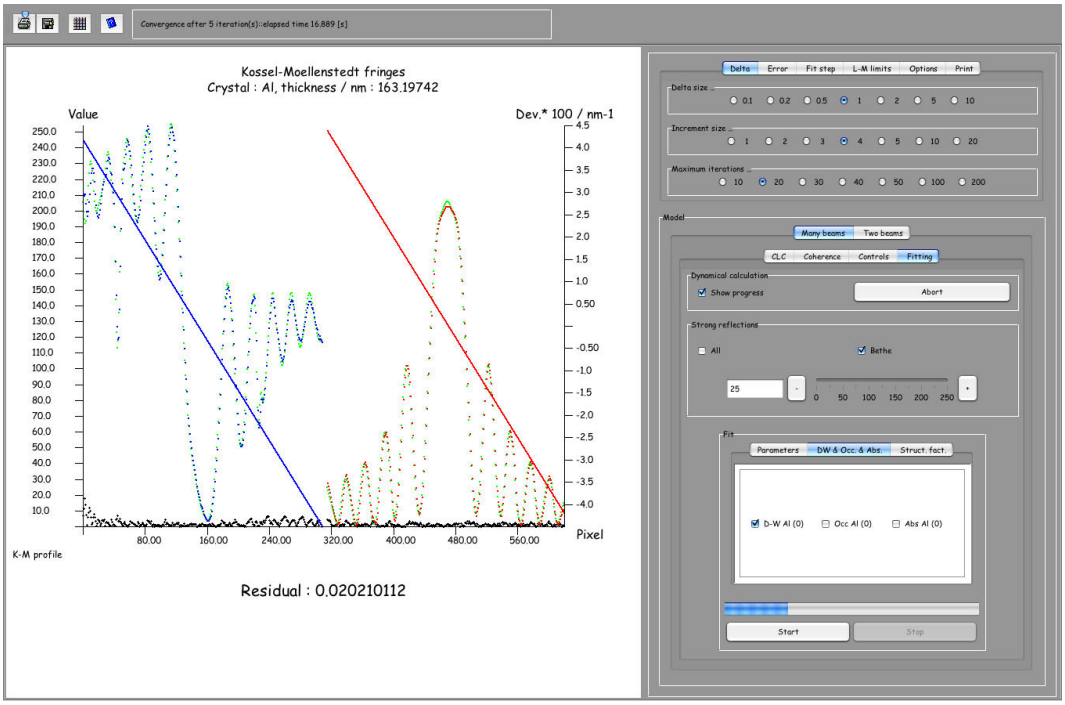

FIGURE 16. Rocking curves and residual after the fit of the microscope parameters and Debye-Waller temperature factor.

```
;Vhkl set::/Users/pierrestadelmann/Desktop/jemsNetBeans/jemsData/Experimental/VhklSet
(2, 0, 0)  0.000000  6.88172  6.88172  90.000
(-2, 0, 0)  0.000000  6.88172  6.88172  90.000
(0, 2, 0)  0.000000  6.88172  6.88172  90.000
(0, -2, 0)  0.000000  6.88172  6.88172  90.000
;Vhkl set::/Users/pierrestadelmann/Desktop/jemsNetBeans/jemsData/Experimental/VhklSet
(2, 0, 0)  4.86611  4.86611  6.88172  45.000
(-2, 0, 0)  4.86611  4.86611  6.88172  45.000
(0, 2, 0)  4.86611  4.86611  6.88172  45.000
(0, -2, 0)  4.86611  4.86611  6.88172  45.000
```

Deselected structure factors saved on a file.

Lines starting with ";" are comments, other lines contain the (hkl) indices of the deselected reflections, the real and imaginary parts of their structure factor [Volt], and their structure factor as amplitude [Volt] and phase [Degree]. Note that the relativistic correction is included.

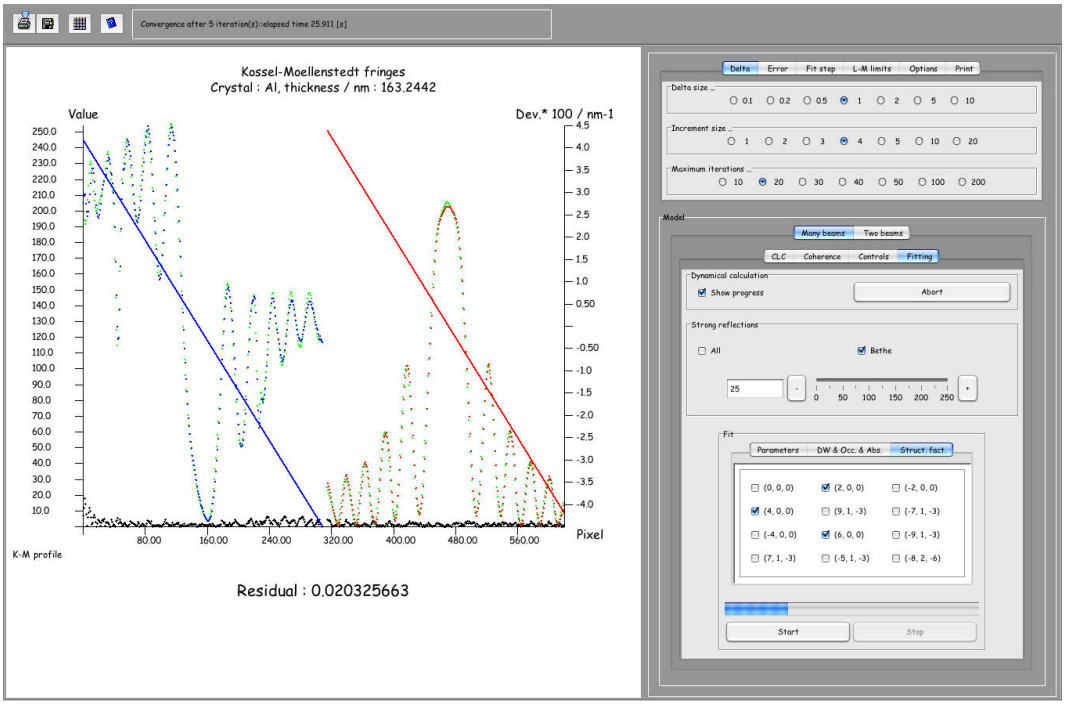

FIGURE 17. Rocking curves and residual after the fit of the microscope parameters, Debye-Waller temperature factor and several structure factors.

When selected the reflections are automatically updated. The update can occur any time and it is necessary to deselect them immediately after entering their (r, i) or (a, p) values.

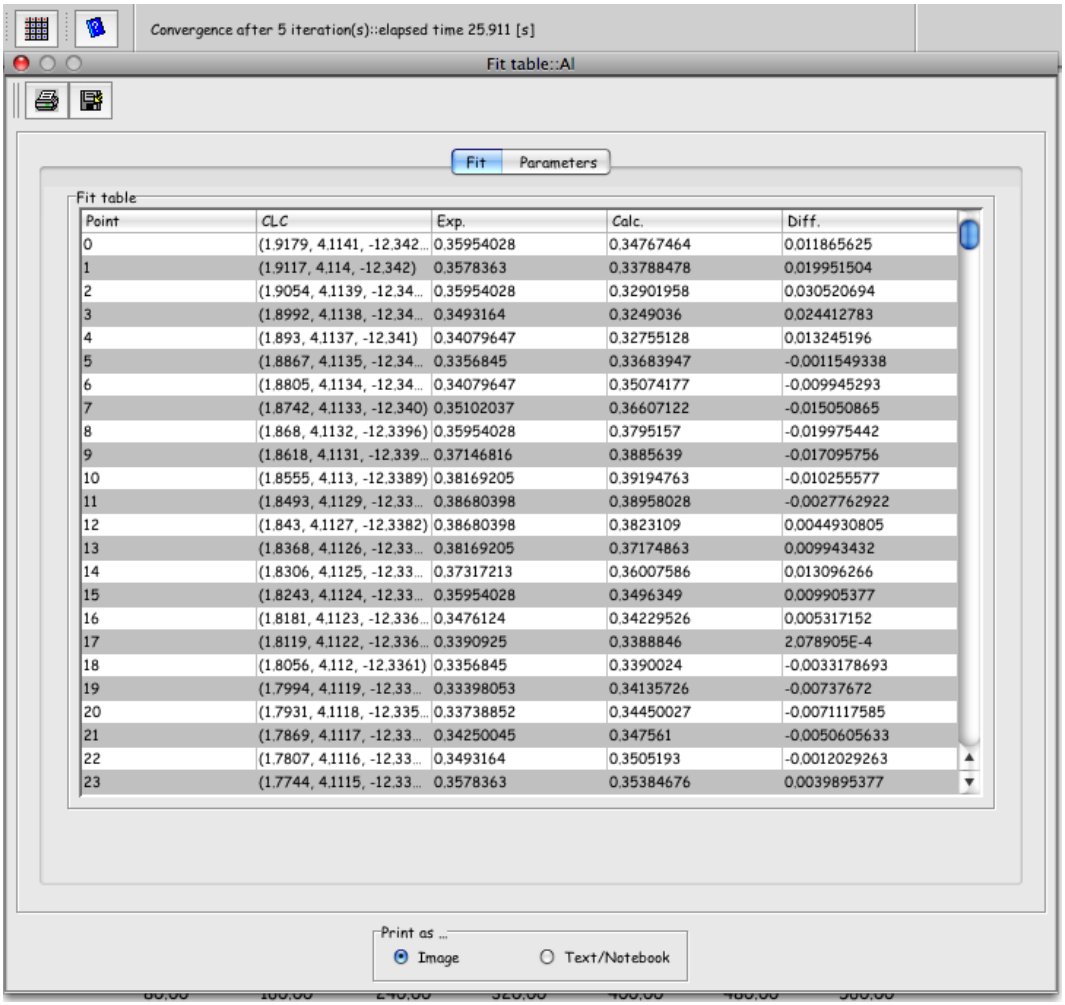

FIGURE 18. The fitted parameters are available in table form.

```
{610,  0.01193,  0.00979, 0.00214,-0.04064, {2, 0, 0}},
{611,  0.01363,  0.01353, 0.0001,-0.04092, {2, 0, 0}},
{612,  0.01704,  0.01711,-0.00007,-0.04121, {2, 0, 0}},
{613,  0.01874,  0.02042,-0.00168,-0.04149, {2, 0, 0}},
{614,  0.02386,  0.02351, 0.00035,-0.04177, {2, 0, 0}}};

kme = km /. {i_, e_, f_ , r_ , d_, h_} -> {i, e};
pre = {{0, Max@(Transpose [kme][[1]])},{0,Max@(Transpose [kme][[2]])}};
kmePlot = ListPlot [kme, PlotRange->pre, PlotLabel->"Exp", PlotJoined->True,    Plo
kmf = km /. {i_, e_, f_ , r_ , d_, h_} -> {i, f};
prf = {{0, Max@(Transpose [kmf][[1]])},{0,Max@(Transpose [kmf][[2]])}};
kmfPlot = ListPlot [kmf, PlotRange->prf, PlotLabel->"Fit", PlotJoined->True,    Plo
kmr = km /. {i_, e_, f_ , r_ , d_, h_} -> {i, Abs [r]};
prr = {{0, Max@(Transpose [kmr][[1]])},{0,Max@(Transpose [kmr][[2]])}};
kmrPlot = ListPlot [kmr, PlotRange->prr, PlotLabel->"|Res|", PlotJoined->True,  Plo
kmd = km /. {i_, e_, f_ , r_ , d_, h_} -> {i, Abs [d]};
prd = {{0, Max@(Transpose [kmd][[1]])},{0,Max@(Transpose [kmd][[2]])}};
kmdPlot = ListPlot [kmd, PlotRange->prd, PlotLabel->"|Dev|", PlotJoined->True,  Plo
Show [{kmePlot, kmfPlot,kmrPlot, kmdPlot}, PlotLabel-> "Exp - Fit - |Res| - |Dev|"]
```

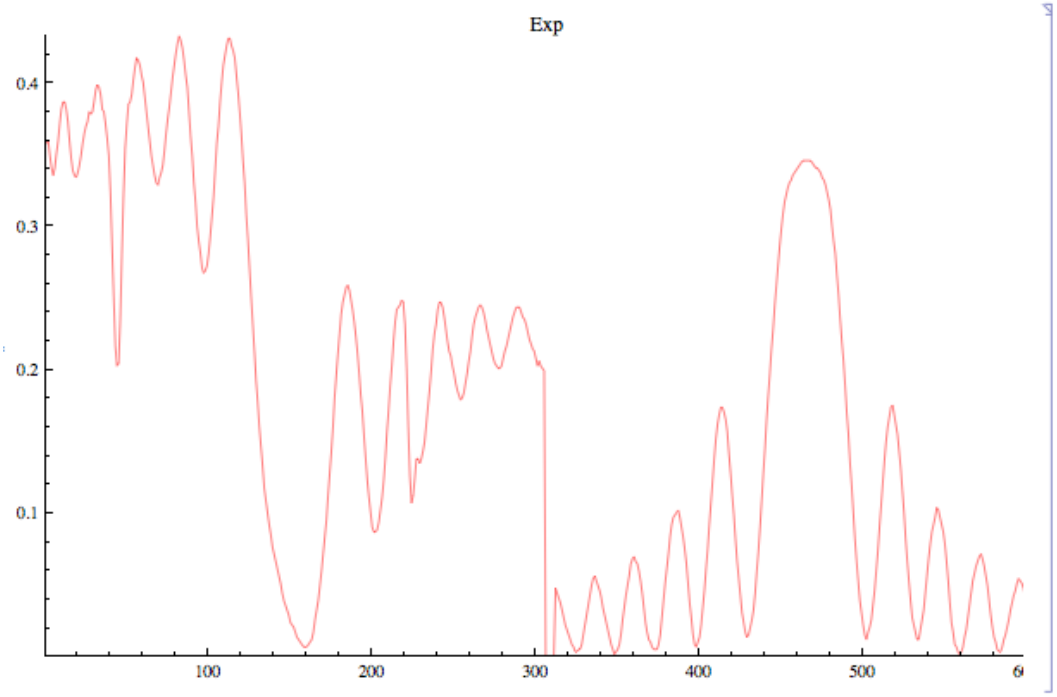

FIGURE 19. Rocking curves plotted using *Mathematica*.

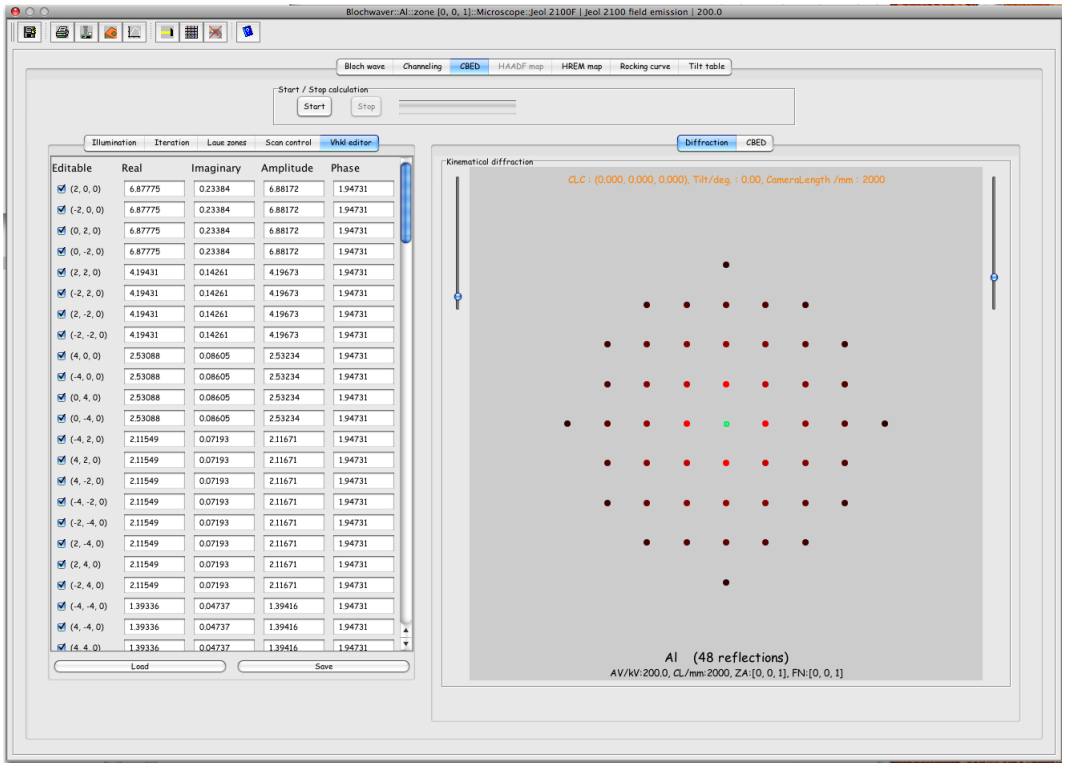

FIGURE 20. The form allowing to enter particular structure factors values in the CBED calculations.

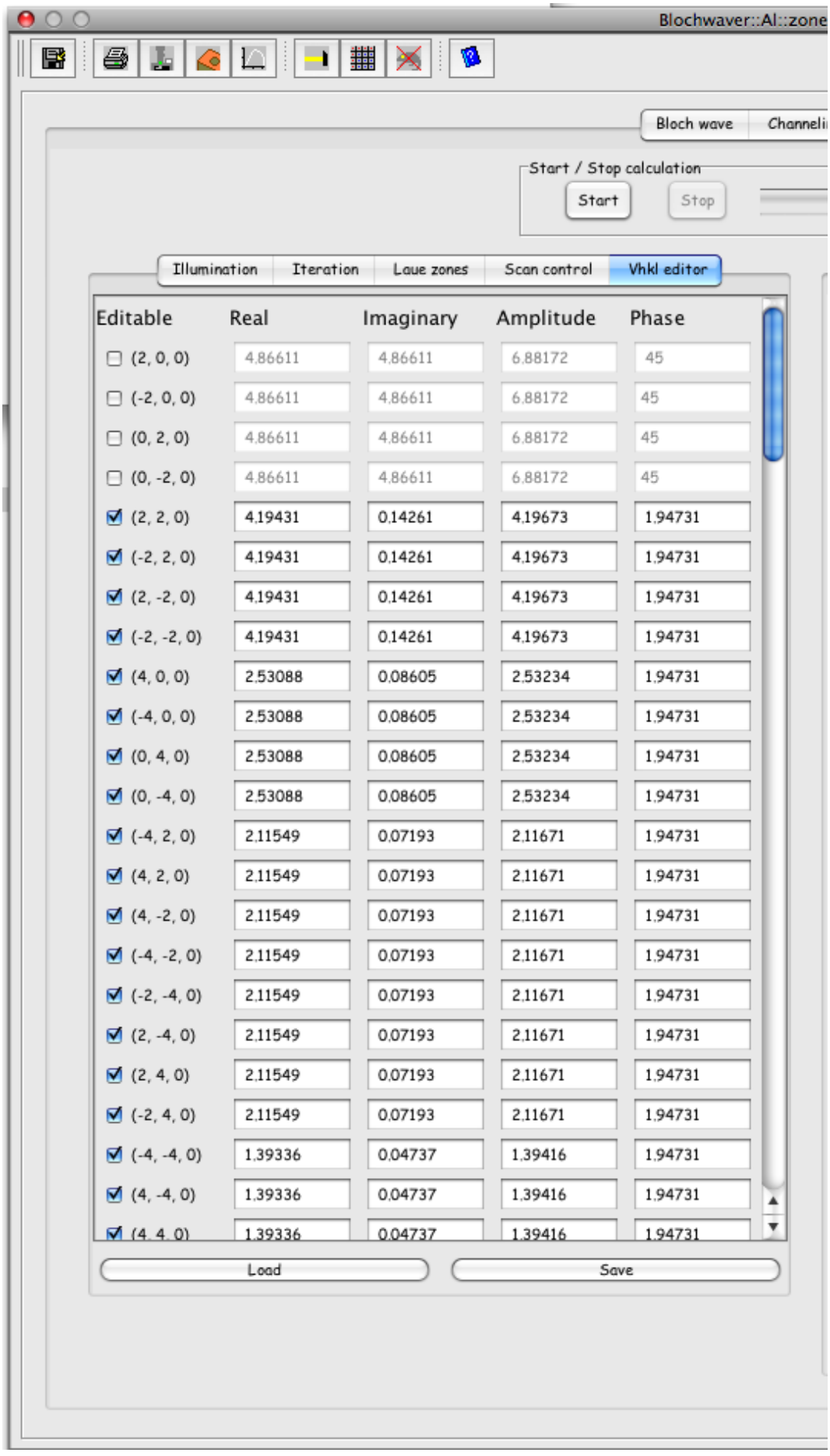

FIGURE 21. Un-selecting the reflections makes the CBED calculations use them instead of the computer generated ones.

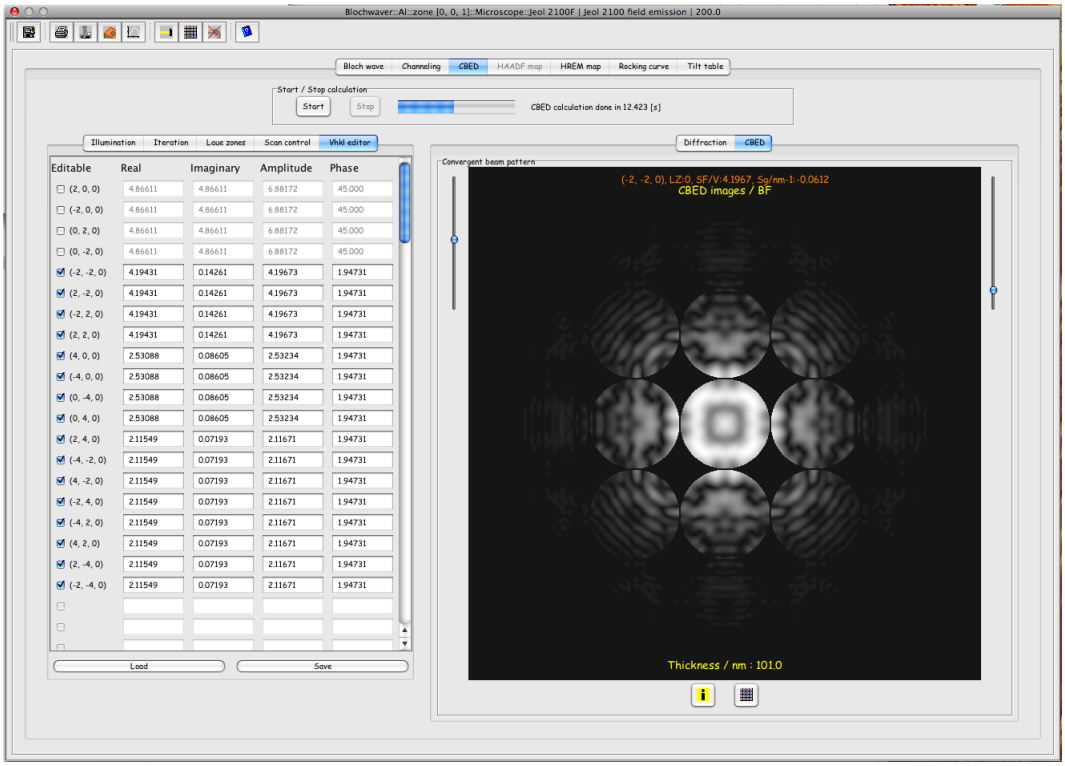

FIGURE 22. CBED pattern calculated with edited (200) structure factors.

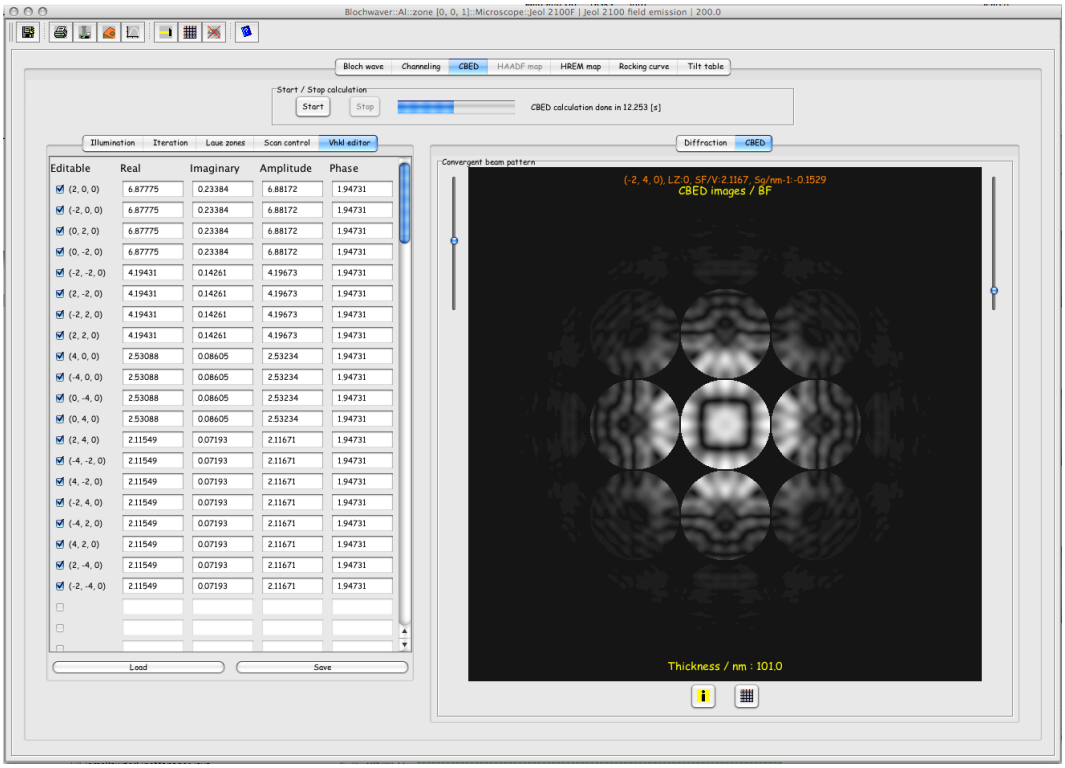

FIGURE 23. CBED pattern calculated with standard (200) structure factors.