

***LANDYNE***

# **User Manual**

**SMART**

Simulation and Measurement with Accuracy on Rocking curve  
for crystal Thickness determination

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## 1. Introduction

Electron diffraction is readily available on any transmission electron microscope (TEM), where it can be further combined with other complementary techniques such as imaging and/or spectroscopy. Two common techniques of electron diffraction, selected-area electron diffraction (SAED) and convergent-beam electron diffraction (CBED). In SAED, a parallel beam interacts with sample. An aperture is used to define the area from which the diffraction pattern is to be recorded from a thin sample. SAED diffraction patterns are either simple spot patterns corresponding to single-crystal diffraction or ring patterns corresponding to powder diffraction from multiple crystals with variable orientation. In CBED, the incident electron beam is a cone of incident rays impinging on the sample over a range of angles. As a result, a diffraction spot will appear as a disc in the back focal plane. Such disc contains information from higher-order Laue zones (HOLZ) and the interfering strips between the incident and diffraction beams.

Accurate data in electron diffraction experiments can be used to deduce reliable crystallographic information. At the lowest level of data collection it should be possible to deduce the crystal system, its Bravais lattice, and the unit cell dimensions. In the case of high-quality data one can uniquely identify the space group of a crystal structure. It is also possible to have accurate information about a particular polytype or about the non-stoichiometry of the specimen. CBED yields information about specimen thickness, unit-cell parameters (accuracy of approx. 0.01%), crystal system, and 3D crystal symmetry (point group and space group). In this content, we will discuss the determination of specimen thickness by the CBED technique.

SMART is designed for simulation and measurement in high accuracy of rocking curve for crystal thickness. This is a new component in the Landyne suite, which has a total of 14 components, each focusing one topic in the application for materials science. The Landyne suite can be used as a tool for research works and a teaching aid on a TEM course.

SMART is mainly coded in Java and partially coded for PC with Microsoft Windows only. The features of SAKI are listed here,

- A user-friendly graphic design and a nice integration of the simulated and experimental parts.
- A template is provided for creating new crystal structures, which was developed in the Landyne suite.
- Drag and drop action for loading experimental diffraction patterns (.jpg and .tif).
- SMART can be used for the simulation of two-beam CBED pattern, including the absorption effect.
- Computer assisting the measurement of crystal thickness using the experimental two-beam CBED pattern.
- Comparison of the simulated and experimental patterns.
- Allow selecting a region of interest on the simulated and experimental CBED patterns to .tif, .jpg, and .gif formats.
- Prepare images with figure labels ready for publication.
- It provides a periodic table of elements with a molecular mass calculator.
- It provides a table of the number and symbols of the 230 space group.

## 2. Theory background

### 2.1 Simulation of two-beam CBED pattern

The details on two-beam electron diffraction theory can be referred to in many books on transmission electron microscopy, e.g., electron microscopy of thin crystals by Hirsch *et al.* (1967) and electron microdiffraction by Spence and Zou (1992).

The main use of the two-beam theory in CBED comes from its ability to provide a quick estimate of sample thickness. The two-beam theory in the Bloch many-beam theory is listed in Eq. (1) if only one Fourier coefficient  $V_g$  is retained. Suppose absorption is also neglected here and will be discussed in the next section.

$$\begin{pmatrix} -2K_n\gamma & U_{-g} \\ U_g & 2KS_g - 2K_n\gamma \end{pmatrix} \begin{pmatrix} C_o \\ C_g \end{pmatrix} = 0 \quad (1)$$

The values of  $\gamma$  are found by setting the determinant of the structure matrix equal to zero, i.e.,  $|A|=0$ . With the solution of this equation, we obtain  $I_g$  as a function of sample thickness  $t$ , structure factor  $U_g$ , accelerating voltage, and excitation error  $S_g$ :

$$I_g = \frac{|U_g|^2 \sin^2 \left( \frac{\pi t}{K_n} \sqrt{K^2 S_g^2 + |U_g|^2} \right)}{K^2 S_g^2 + |U_g|^2} \quad (2)$$

$$I_0 = 1 - I_g \quad (3)$$

The intensity minima of Eq. (2) occur at  $S_g$  values given by

$$S_g^2 = \frac{n^2}{t^2 (K/K_n)^2} - \frac{|U_g|^2}{K^2} \quad (4)$$

The factor  $K/K_n$  in Eq (4) is often neglected in the literature. If so, then the thickness measured is effective crystal thickness,  $t^{\text{eff}} = t(K/K_n) = t/\cos\theta$ , where  $\theta$  is the angle between the beam direction and the surface normal.

The above discussion assumes an ideal two-beam case in which all other beams have zero intensities. In reality, there are always weak beams present. One way to include these weak-beam effects on two-beam diffraction is to use the perturbation method of Bethe in the book by Spence and Zou (1992).

### 2.2 Anomalous absorption and phenomenological treatment

The effect of absorption has become known as the anomalous transmission effect by analogy with the Borrmann effect in X-ray diffraction. In CBED, this anomalous transmission effect is revealed by the intensity asymmetry around the Bragg condition in the transmitted (zero-order) disk, instead of the symmetry which we expect from the two-beam intensity expression of Eq (2). The two-beam theory has been applied to electron diffraction with absorption, and comparisons were made with the experiment in the work of Hasimoto *et al.* (1962).

The concept of an optical potential  $V(\mathbf{r}) = V^c(\mathbf{r}) + iV'(\mathbf{r})$  is used to take account of the absorption effect, with  $V^c(\mathbf{r})$  the real crystal potential (describing the interaction of the incident electron with the crystal electrons and the nuclei) and  $V'(\mathbf{r})$  a second real potential which accounts for depletion of the elastic wavefield by inelastic scattering. The term ‘absorption’ is used to describe this depletion since the probability that the electron is scattered back into the original state is very small.

### 2.3 Analysis of two-beam CBED pattern for crystal thickness

The measurement of foil thickness is becoming increasingly important in electron microscopy due to the need for quantitative measurement of dislocation density, point defect concentration interparticle spacing, etc. The other application of sample thickness is frequently used for X-ray microanalysis.

The popular CBED method (Kelly *et al.* 1975; Allen 1981) to use the formula below,

$$\left(\frac{s_i}{n_i}\right)^2 + \left(\frac{1}{n_i}\right)^2 \left(\frac{1}{\xi_g}\right)^2 = \left(\frac{1}{t}\right)^2 \quad (5)$$

With  $s_i$  the excitation error at the  $i$ th minimum and  $t$  the effective specimen thickness along the beam direction. A plot of  $(s_i/n_i)^2$  against  $(1/n_i)^2$ , therefore, gives  $(1/t)^2$  as the intercept, and hence the thickness. Since the rocking curve is symmetrical the required values of  $\Delta X$  were obtained by taking half the distance between corresponding minima on either side of the Bragg condition. The distance  $X$  was obtained by measuring the distance between the edges of the CBED disks corresponding to twice the Bragg angle.

However, an important problem arises of how to determine the first value of  $n$  ( $i=1$ ). For a sample thickness  $t < \xi_g$ , we require  $n_1=1$ ; for  $\xi_g < t < 2\xi_g$ ,  $n_1=2$ , and so on. Thus  $n_1$  is the first integer larger than  $t/\xi_g$ . In practice, one tries several initial values until a good straight line fit is obtained for a reasonable value of  $\xi_g$ . (A table of values yielded the data plotted in Fig. 4.2. We note that  $i=4$  corresponds to the first minimum either inside or outside the Bragg condition.)

There are a number of difficulties with this method; however, for small unit-cell crystals, an accuracy of 2% in thickness has been claimed. The most important limitation of the method results from the excitation of other beams, which become increasingly important as the reciprocal lattice becomes dense. Many-beam effects are minimized by using a large reciprocal lattice vector and a small unit cell. The problem of finding  $n_1$  can also be resolved by conducting thickness measurements at several thickness.

## 2.4 Least-square line fitting

Fitting a straight line is a statistical procedure to construct a straight line that best fits the series of data points. The line best fits a set of scattered data points: some points that lie below and above the line are equal, and the line will pass through as many points as probable. One of the most accurate ways to find the best fit is the least squares method.

The steps to be followed in the least squares method are list below:

1. Calculate the mean of the x values and the mean of the y-values.
2. Calculate the slope of the line of best fit from the below formula:

$$b = \frac{\sum(X - \bar{X})(Y - \bar{Y})}{\sum(X - \bar{X})^2} \quad (6)$$

3. Compute the y-intercept of the line from the formula:

$$a = \bar{Y} - b\bar{X} \quad (7)$$

4. Use the obtained slope and the intercept to frame the equation.

## 2.5 Other methods for the measurement of specimen thickness

Generally, the specimen thickness is measured using the two-beam CBED method. However, fringes in the CBED disk are degraded as the dislocation density in the matrix increases by the mechanical deformation. The fine dispersion of precipitate in the matrix hinders the clear identification of fringes. Other methods for specimen thickness are mentioned here for cross-checking the specimen thickness or on the specimen where the CBED method is not applicable.

Thin foil thickness can be measured through electron energy loss spectroscopy (EELS). Low loss spectra, including zero-loss and plasmon peaks, are used for the calculation. According to the previous report (Egerton and Cheng 1987), this method has an accuracy of less than 2 nm (10%) in the thickness range of 10 to 150 nm. Although this method needs additional information such as convergence and collection angles, the strong benefit of applying it regardless of material state promotes its use. Three different methods (log-ratio method, Bethe sum rule, and Kramers-Kronig sum rule) are available for the measurement of thickness using an EELS spectrum (Egerton and Cheng 1987).

In principle it is simple to calculate the thickness of a foil at any one point by counting the number of thickness fringes from the edge of the crystal. In bright-field micrographs, white fringes occur at  $t/\xi(g,\omega)=0,1,2$ , etc, while dark fringes occur at  $t/\xi(g,\omega)=1/2,3/2,5/2$ , etc., where  $t$  is the thickness of the foil and  $\xi(g,\omega)$  is the effective extinction distance given by:

$\xi(g,\omega) = \xi(g)/(1+\omega^2)^{1/2}$ ,  $\xi(g)$  is the true extinction distance at the exact Bragg angle, and  $\omega$  is the deviation from the Bragg angle.

A method for determining the thickness of foils from the diffraction pattern was firstly described by Pinsker (1953). The angular width,  $2\psi$ , of the zero order Laue zone is related to the thickness of the foil  $t$ , through the length of the spikes in reciprocal space and the curvature of the reflecting sphere as follows:

$$2\psi = 2 \left( \frac{2\lambda}{t} \right)^{1/2}$$

This method is limited to foils in which area is less than one extinction distance thick. At greater thickness, the length of the spikes in reciprocal space is determined by the extinction distance rather than the overall thickness.

### 3. The graphic user interface of SMART

#### 3.1 Main graphic interface

The main graphic interface of SMART is a frame with a drop-down menu, a toolbar, and a display panel, as shown in Figure 1. The frame of SMART can display simulated and experimental rocking curves or CBED patterns in a single one or the combination mode of selected two. Various tools can be enacted from the menu or toolbar, e.g., a caliper for measuring the scale bar, calculators for the wavelength and extinction distance. Region of interest is for output. A periodic table of elements and a list of the space groups are provided.

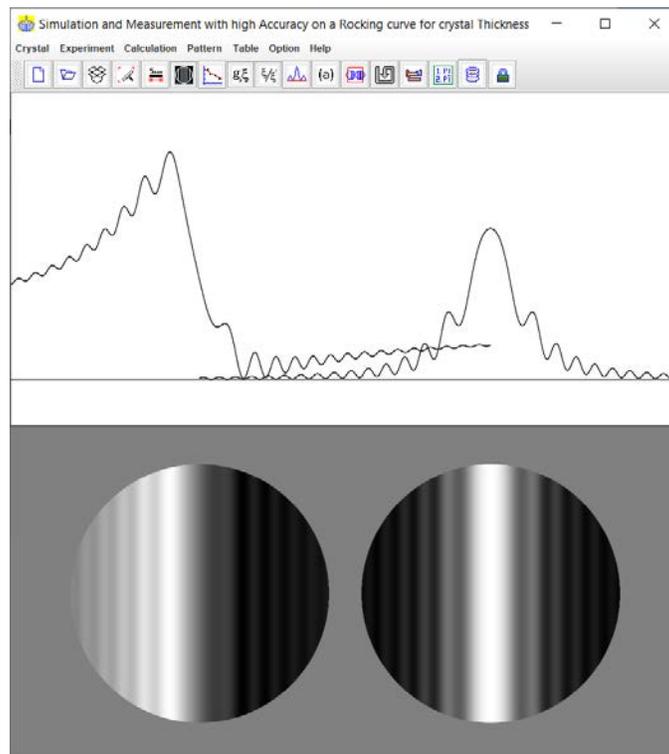


Figure 1. The graphic user interface of SMART. The rocking curve and CBED pattern of Aluminum under the condition of two-beam and  $g(200)$ .

#### 3.2 Control panel for simulation

The two-beam CBED simulation can be carried out step by step as listed below referring to the control panel in Figure 2.

Step 1. Two-beam dynamical diffraction theory is used for simulation. An absorption effect is an option, and the value of absorption can be adjusted. The wavelength can be set up through a HT &  $\lambda$  values dialogue panel. The filename is displayed after a crystal file is loaded. The default crystal file is Aluminum.

Step 2. Parameters for simulation can set up through a  $g$  &  $\xi$  values dialogue panel – calculated from the structure file and the diffraction index (hkl) and transfer the control panel. The reciprocal length  $g$  and the extinction distance  $\xi$ , and the specimen thickness  $t$  can also be filled directly on the control panel.

Step 3. Simulation is executed by the simulate button. The calculated rocking curve and simulated CBED pattern can be displayed separately or together. The background color for the CBED disks can be adjusted.

Step 4. The display of the calculated rocking curve and simulated CBED pattern can be further adjusted to experimental conditions, e.g. the contrast, the camera length, tilt angle on the Bragg conditions, and the convergent angle.

Step 5. In Full image mode, the disk position and radius need to be marked separately for disk o and disk g.

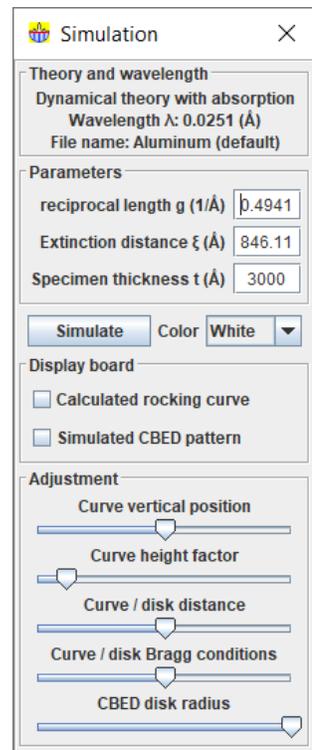


Figure 2. Control panel for simulation.

### 3.3 Control panel for measurement

The measurement of experimental two-beam CBED pattern can be carried out step by step as listed below referring to the control panel in Figure 3.

Step 1. The filename is displayed after an experimental CBED pattern is loaded. The experimental pattern can be loaded using the drop-down menu for a file or drag & drop operation on the menu bar. Scale info can be renewed through the calibration for scale bar dialogue panel.

Step 2. In Full image mode, the disk position and radius need to be marked separately for disk o and disk g.

Step 3. The rocking curve and CBED pattern ready for measurement are transferred by the transfer button. The experimental rocking curve and simulated CBED pattern can be displayed separately or together. The color for the marks on the local minima can be adjusted.

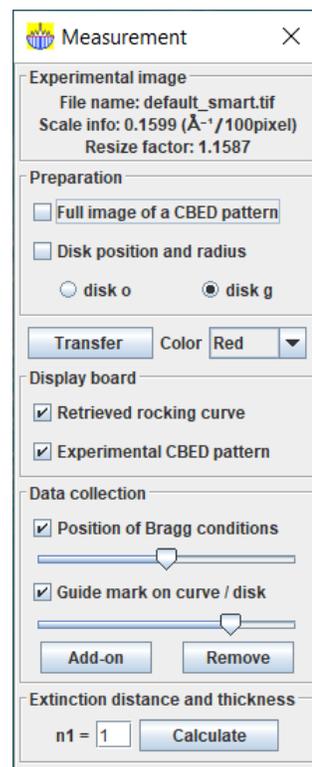


Figure 3. Control panel for measurement.

Step 4. On the experimental rocking curve and simulated CBED pattern, find the Bragg diffraction position for the reciprocal length of  $g$  and collect at least three positions of the local minima on the disk  $g$ .

Step 5. Selection of  $n_1$  (default value =1) and calculate the extinction distance and specimen thickness by the calculate button. A straight line should be obtained from the least-square fit panel, see Figure 4. If not, increase the value of  $n_1$  and calculate again until a straight line is obtained.

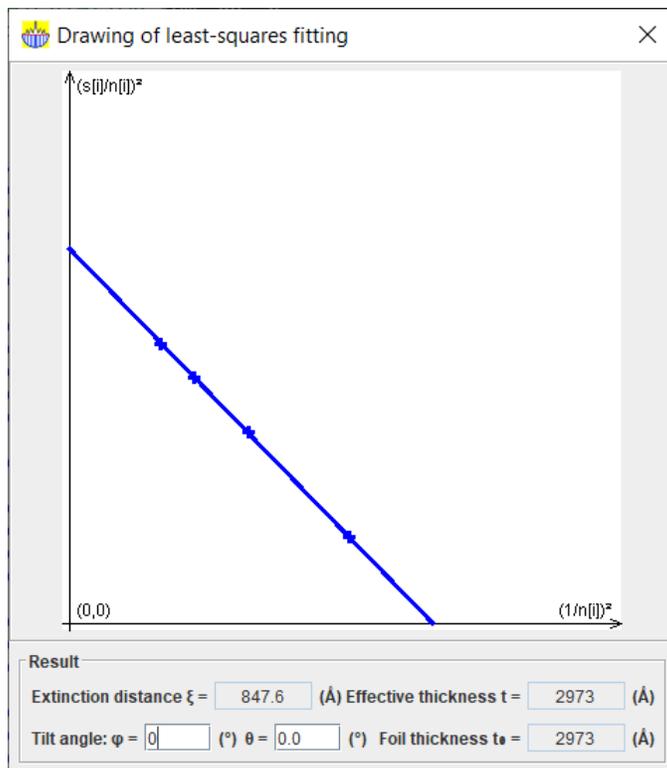


Figure 4. The drawing panel and result table for sample thickness measurement.

### 3.4 Combination of simulation and measurement

SMART provides a unique way to combine the simulated and experimental rocking curve and CBED pattern to measure specimen thickness. The display board options in simulation and measurement panels allow the display of any pair of them. It allows to confirm the measured result or to estimate the specimen thickness when less than two local minima positions are displayed in the experimental CBED pattern.

## 4. Other topics

### 4.1 Preparation of new data file

Landyne suite provides a unified **New Crystal Structure File** dialogue in all components, as shown in Figure 5. The dialogue window provides an automatic assistant for the user and makes

sure to meet the requirement of the file format. The template is embedded with the 230 space groups in the Hermann-Mauguin notation, which list in the international table for crystallography. However, only b unique axis will be used in a monoclinic system. Two origin choices can be accepted as input parameters, but choice-2 will be converted to choice-1. To save the data structure, click the **Save** button; or make a new one click the **New** button.

Figure 5. Snap-shot of the crystal data template.

The crystal file can be also converted from a previous data for modification or from a crystallographic information file (CIF). If a data file in an alternative setting of space group for triclinic, monoclinic and orthorhombic systems is used, please click on the alternative settings (1~74). A tool for the transformation of files in the nonconventional notations to the standard notations is provided.

#### ***4.2 Region of interest and figure label for an image***

Figure 6 shows a tool-of-region of interest. The rectangle area can be defined with the top-left corner and then the width and height. The point per inch can be defined in the low part of the dialogue panel. Figure 7 shows a tool for adding a figure label, with adjustable on the font size and the position for convenience. The ROI area with a figure label can be saved in .tif, .jpg, or .gif formats.

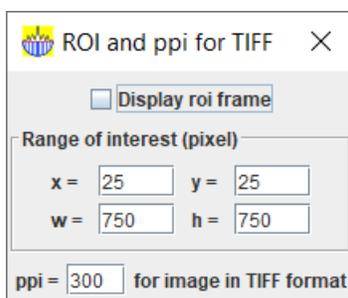


Figure 6. Snap-shot of the region of interest and ppi for TIFF.

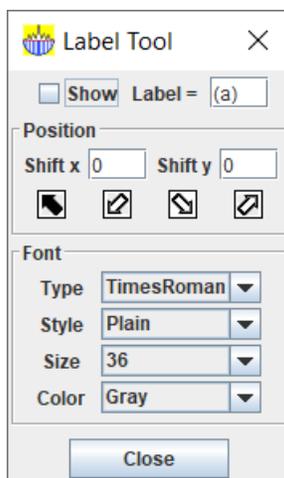


Figure 7. Snap-shot of the tool for adding a figure label.

### ***4.3 Periodic table of elements and list of space group***

Landyne suite provides a unified periodic table of elements (PTELS) and a list of space groups (SG), is available for convenience in the preparation of a new structure file. The PTELS and SG can be activated from the drop-down menu or the graphic menu bar. The same PTELS can also be accessed from the Landyne launcher. The SG shows the number and symbol, including the non-standard settings in a separate list.

## **5. Application examples**

### ***5.1 Simulation of rocking curves and two-beam CBED patterns***

The basic steps for simulation of the two-beam rocking curve and CBED pattern are listed in Section 3.1. We need to provide the input data for simulation. Input data on the microscope, the value of the high voltage needs to update the wavelength, as shown in Figure 8. Input data on the specimen, the crystal file is loaded, and two-beam diffraction  $g(hkl)$  is selected, as shown in Figure 9. The absorption coefficients are simplified by assuming  $\xi_g/\xi'_0 = \xi_g/\xi'_g$ , as shown in Figure 10.

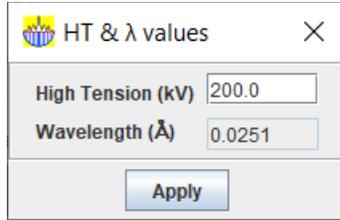


Figure 8. The dialogue panel for high voltage and wavelength.

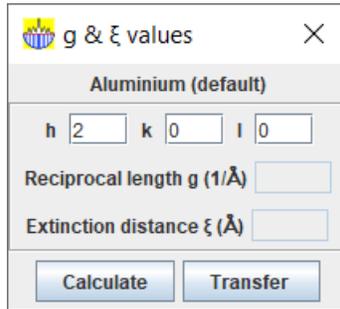


Figure 9. The dialogue panel for  $g(hkl)$ , the reciprocal spacing, and the extinction distance.

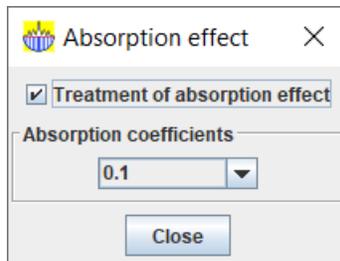


Figure 10. The dialogue panel for absorption effect and the parameter.

Once these parameters are ready, the simulation can be executed, and one of the simulated results was shown in Figure 1. The only rocking curve after adjustment is shown in Figure 11.

### 5.2 Measurement of the crystal thickness and the extinction distance

The basic steps for the measurement of two-beam CBED pattern for crystal thickness were listed in Section 3.2. We will use the simulated two-beam CBED pattern as an example to determine the thickness. The two-beam CBED pattern is loaded, and then the positions and radiuses of the o disk and the g disk are defined. Figure 12 shows the halfway of the process; only the position and radius of the o disk are defined. Once the CBED disk is transferred, the positions of the Bragg condition and the local minima can be easily defined, as shown in Figure 13. A plot on the coordinate axes of  $(1/n[i])^2$ ,  $(s[i]/n[i])^2$  will be displayed after selection of the  $n[1]$  value, as shown in Figure 4. If the data shows a straight line within the errors, the least-square fitting straight line allows deducing the extinction distance and the crystal thickness. After trying  $n_1 = 1, 2, 3, 4$ , a straight line is obtained when  $n_1 = 4$ . The deduced extinction distance is comparable to the calculated value in the simulation, thus we obtained that measured thickness, which is agreement with the simulated value (3000 Å).

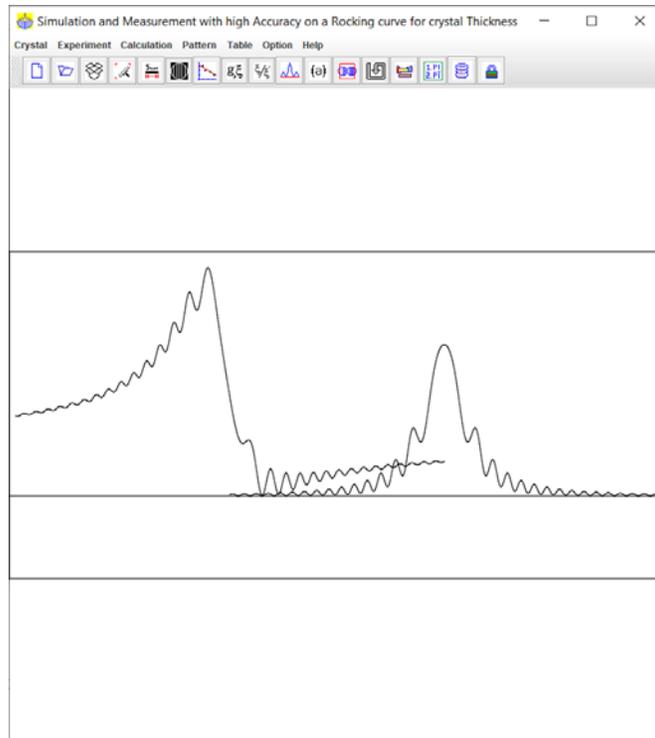


Figure 11. The rocking curve of Aluminum under the condition of two-beam and  $g(200)$ .

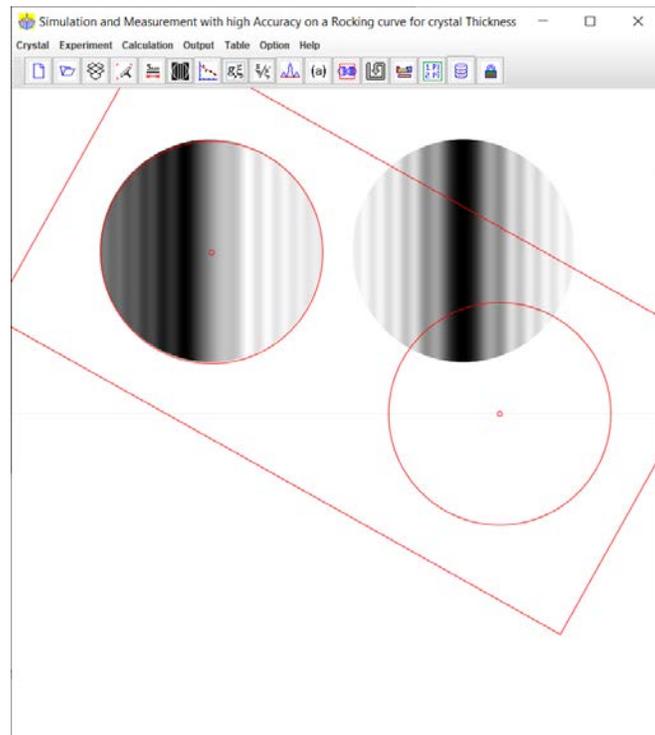


Figure 12. The halfway of the process; only the position and radius of the o disk are defined.

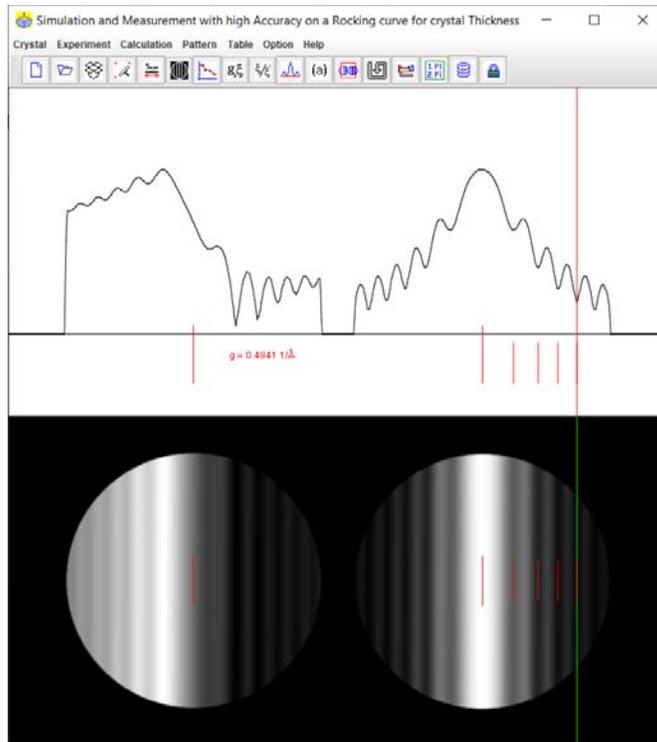


Figure 13. The experimental rocking line and CBED disk. Local minima are marked.

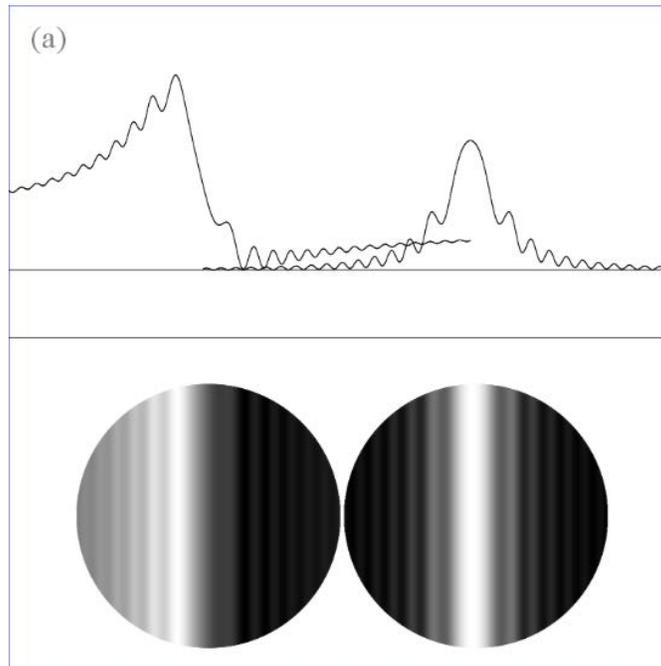


Figure 14. The output image in region of interest with a figure label.

### 5.3 Output image in the region of interest (roi) and a figure label

The simulated rocking curve and CEED pattern can be selected with the tools for a region of interest in Figure 6 and added with a figure label in Figure 7. The image within the roi can be saved in TIFF and JPEG format, as shown in Figure 14.

## **6. Installation of the Landyne suite**

The SMART is one of the key components in the Landyne software suite, which can be download from the Landyne website:

<https://landyne.com>

Decompress the installation file landyne.z7 in a folder, for example, c:\landyne5\. It includes a set of sub-folders, the documents folder contains user manuals for all software, including this one, and the programs folder contains executable files. For convenience, it is recommended to create a shortcut to landyne.exe and move it to the desktop. To start the launcher by double clicking on landyne.exe.

The Landyne suite requires Java Runtime Environment above JRE 1.8.0. The image TIFF format needs the support of Java Advance Image, for example, JAI-1\_1\_3-LIB.zip , which can be downloaded online, e.g.,

<https://www.unl.edu/ncmn-enif/xzli/download/>.

After decompression, copy jai\_core.jar and jai\_codec.jar to the JRE folder of the Java system of the microcomputer, for example, c:\program files\java\jre1.8.0\_31\lib\ext\.

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