LANDYNE

User Manual

Simulation of HOLZ lines and

a CBED whole pattern

Calculation under kinematical theory and dynamical first-order approximation

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1. Introduction

HOLZ4 is a component of the Landyne suite, developed by Dr. X.-Z. Li for the simulation of electron diffraction and crystallographic analysis. HOLZ4 is an enhancement of the earlier JECP/HOLZ (Li, 2005). It serves as both a teaching aid for students learning fundamental crystallography and a tool for researchers conducting TEM analysis.

In convergent-beam electron-diffraction (CBED) patterns at a zone axis orientation, the bright-field disc is often intersected by a set of fine dark lines known as higher-order Laue zone (HOLZ) lines. These lines result from high-angle scattering, causing higher-order Laue reciprocal planes to intersect the Ewald sphere. HOLZ4 is an interactive program that simulates these HOLZ lines using kinematical approximation and a first-order dynamic correction.

The main improvements in HOLZ4 include:

- GUI Enhancements: Improved display panel and calculation dialog.
- Intensity Threshold Adjustments: Easy adjustments for intensity thresholds.
- Index Display: Updated display system to show indices on the entire CBED pattern and on HOLZ lines.
- Crystal Constraint: Introduced for tuning lattice parameters.
- I/O System Improvements: Enhanced input/output system for data and patterns.
- Integration: Software integrated with an elemental periodic table and a space group table.
- Figure Preparation Tools: Provided figure label tool and region of interest (ROI) feature for preparing publication-ready figures.
- File Saving Formats: Integrated interface for saving files in TIFF, PNG, JPEG, and GIF formats.

These improvements make HOLZ4 a robust tool for both educational purposes and advanced electron diffraction analysis.

2. Software design and features

The features of HOLZ4 are briefly listed as:

1. Window Frame with Panel: Displays simulated HOLZ lines and the entire CBED pattern.
2. Operational Panel and Dialog Windows: Initializes input parameters for the calculations.
3. Computer Assistant for Data Preparation: Facilitates the creation of input structure data files.
4. Region of Interest and Label Tool: Prepares figures for publication.

HOLZ4 is written and compiled in openJDK21, with further code optimization, including obfuscation, for the compiled class files.

Both HOLZ lines on the bright-field disc and whole-pattern modes are integrated into HOLZ4. Figure 1 shows a snapshot of the HOLZ4 panel in the bright-field disc mode, and Figure 2 shows

a snapshot in the CBED whole-pattern mode. The examples are simulated using aluminum at 200 kV. The interface includes a dropdown menu and a graphical menu bar, allowing the indices of the calculated HOLZ lines and the CBED whole pattern to be displayed.
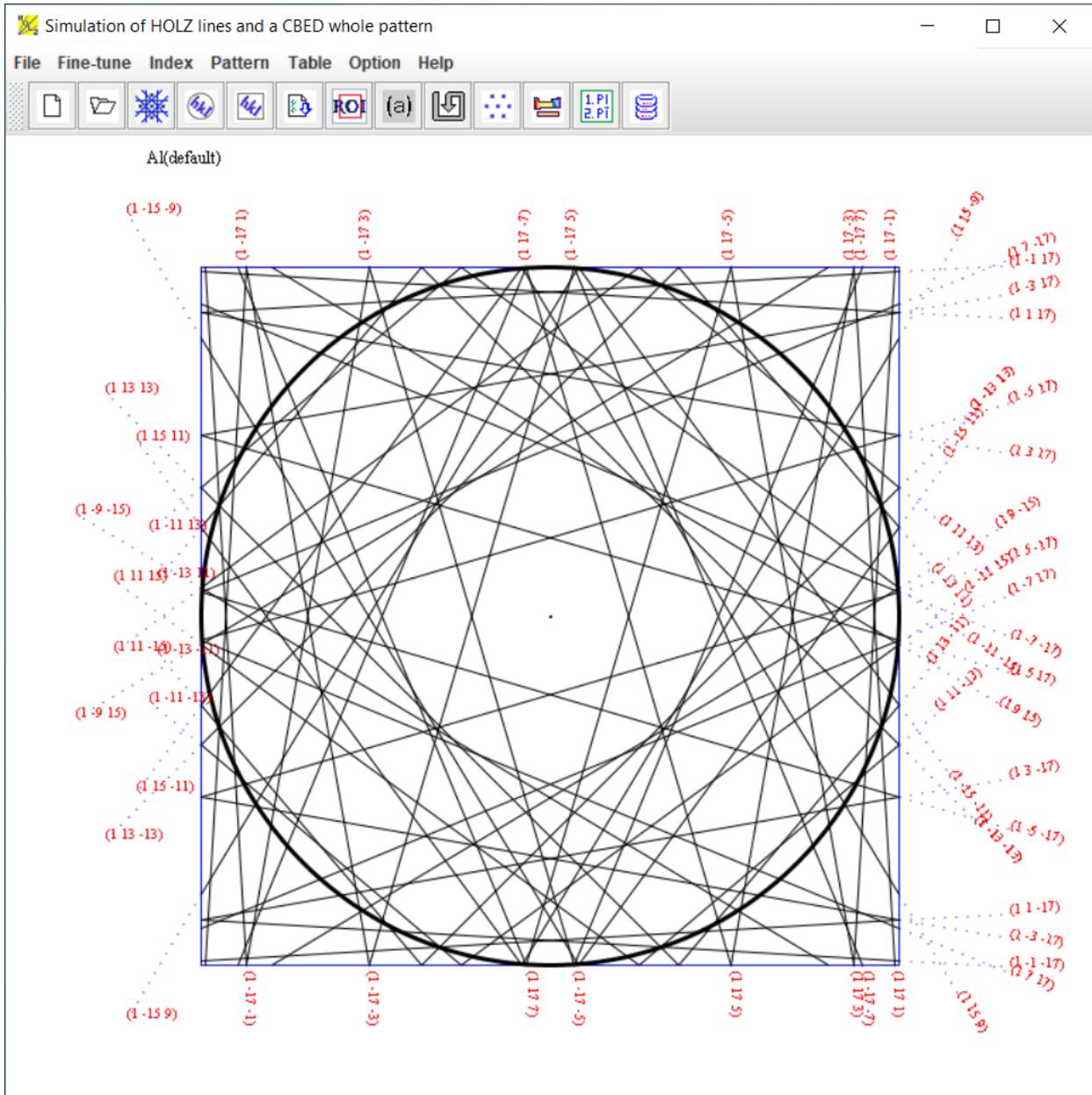

Figure 1. GUI of the HOLZ4 in HOLZ lines in bright-field disc mode. The simulation is done with an Al crystal structure along [100] zone axis and 200 kV.

The other parameters for HOLZ lines and CBED whole pattern are TEM high voltage and the zone axis of the crystal sample, converge angle and tilt away from the true zone axis. The appearance of the pattern can also be adjusted. The index panel provides various position options for optimal indexing of HOLZ lines and a CBED whole pattern. Two types of the appearance of HOLZ lines in straight lines or curves are available.
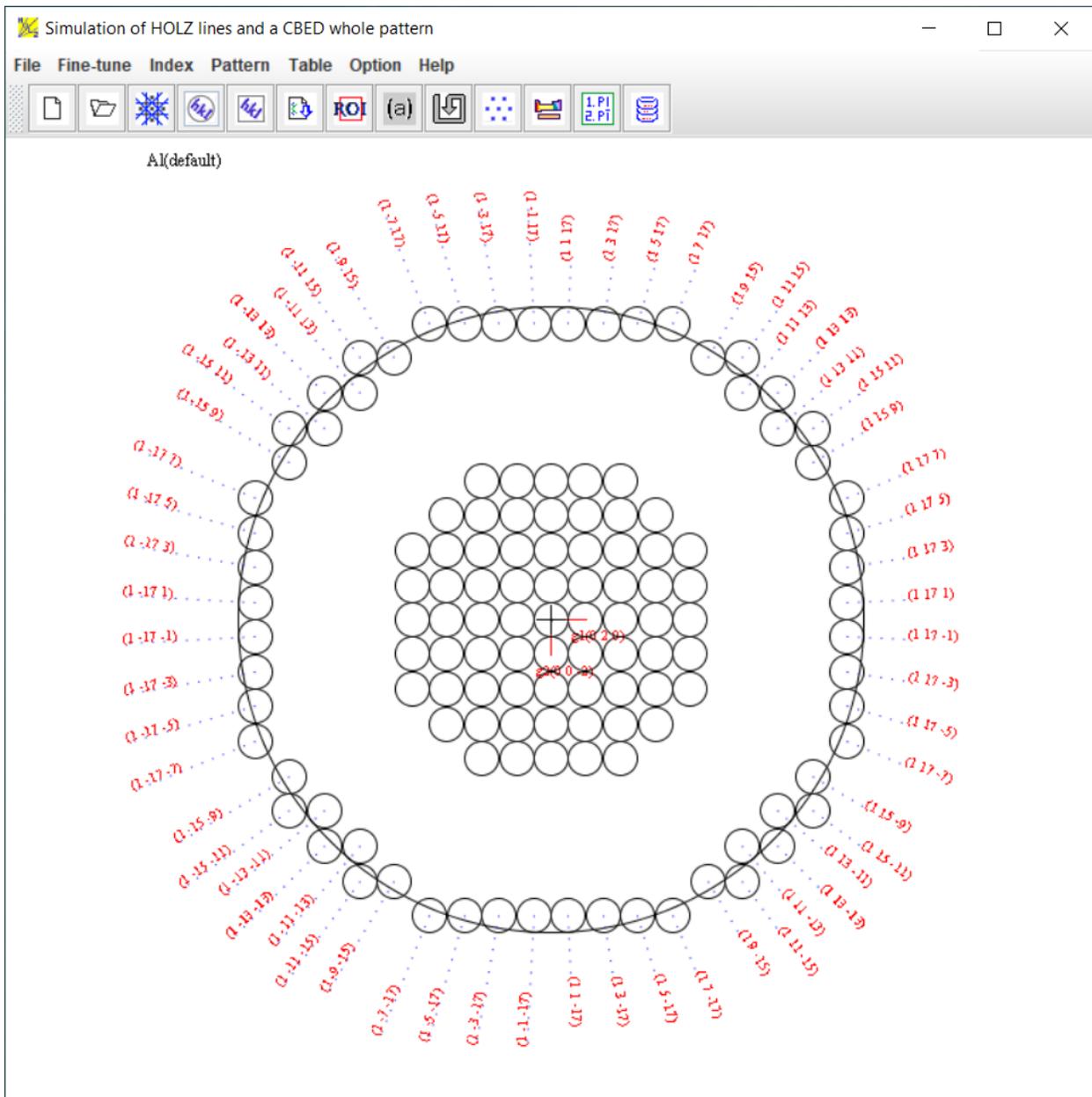
Figure 2. GUI of the HOLZ3 in the CBED whole-pattern mode. The simulation is done with an Al crystal structure along [100] zone axis and 200 kV.

Figure 3 shows a set of the dialogs for the HOLZ4 software, (a) HOLZ simulation, (b) index of HOLZ lines, (c) index of CBED whole pattern, (d) the HOLZ/CBED display option, and (e) transformation of index for a hexagonal system.

For simulation, the lattice parameters can be loaded on the drop-down menu for the calculation. The parameters can be changed slightly on the calculation panel, and the calculated pattern will be updated immediately.
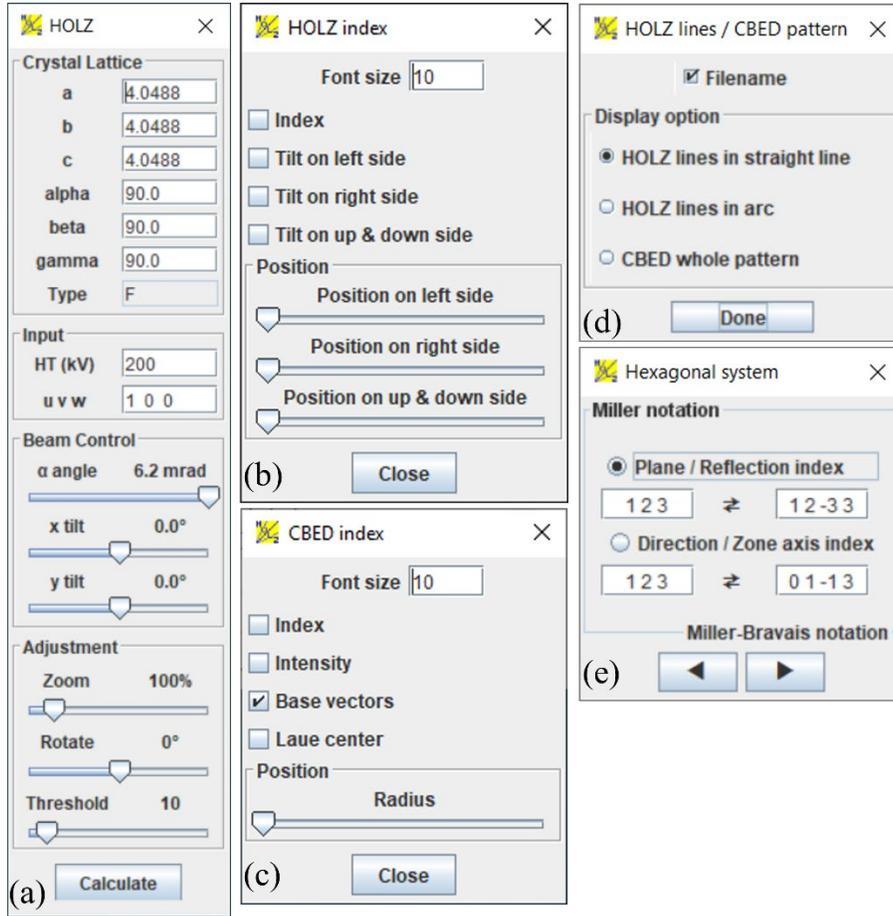
Figure 3. The control dialogs for the HOLZ4 software, (a) HOLZ simulation, (b) index of HOLZ lines, (c) index of CBED whole pattern, (d) the display HOLZ/CBED option, and (e) transformation of index for a hexagonal system.

3. Formulas for calculating the positions of HOLZ lines

3.1 Formulas of the HOLZ lines under the kinematical theory

A HOLZ line in the kinematical approximation is the Bragg condition's locus for a HOLZ reflection **g**. The incident beam **k** is described as k_n along **-z** and k_t in **(x, y)** plane. We may think of the HOLZ line as a function of K_t, a vector that originates in the center of the zone axis and extends to the point of interest in the bright-field disc. The trajectory is described by the following two equations (Spencer and Zuo, 1992):

$$g_x k_x + g_y k_y - g_z k_z + \frac{g^2}{2} = 0 \qquad \text{......(1)}$$

$$k_z = \sqrt{(k^2 - k_x^2 - k_y^2)} \qquad \text{......(2)}$$

Here

$$g^2 = g_x^2 + g_y^2 + g_z^2.$$

If we use a paraboloid equation, $k_z = k - \dfrac{k_x^2 + k_y^2}{2k}$ as an approximation of the sphere equation, $k_z = \sqrt{(k^2 - k_x^2 - k_y^2)}$, we end up with an equation for the HOLZ line trajectory (Li, 2007):

$$(k_x + \frac{g_x}{g_z}k)^2 + (k_y + \frac{g_y}{g_z}k)^2 = k^2 + (\frac{g}{g_z}k)^2 - \frac{g^2}{g_z}k \quad ......(3)$$

$$(k_x + g_x)^2 + (k_y + g_y)^2 = r^2 = k^2 - (k_z - g_z)^2 \quad ......(4)$$

3.2 Formulas of the HOLZ lines under a first-order dynamical correction

The first-order dynamical correction was developed for simplicity in the calculation (Bithell and Stobbs, 1989; Lin *et al.*, 1989; Zuo, 1992). In the first-order dynamical correction, it is assumed that only weak interactions occur between HOLZ reflections. The position of a HOLZ line in the central disc can be approximated as the intersection between the zero-layer dispersion surface (k1=k0+γ(1), in which γ(1) is the distance of the topmost excited branch of the dispersion surface from the sphere at the zone axis itself), and a plane-wave sphere centered on the HOLZ reflection.

When the incident beam is far away from a zone axis, the first branch of the dispersion surface can be approximately considered a sphere with a radius of $k_1 = k_0 + \gamma^{(1)}$. Thus, the dynamically corrected HOLZ line equation is derived as (Li, 2007):

$$g_x k_x + g_y k_y - g_z k_z + \frac{g^2}{2} + g\Delta g = 0 \quad ......(5)$$

Here,

$$g\Delta g = (k\gamma^{(1)} + \frac{\gamma^{(1)2}}{2})$$

When the incident beam is near or at a zone axis, the first branch of the dispersion surface can be approximately considered a flat plane with a distance of $k_1 = k_0 + \gamma^{(1)}$ to the origin of the reciprocal lattice. Thus, the dynamically corrected HOLZ line equation is derived as (Li, 2007)

$$(k_x + g_x)^2 + (k_y + g_y)^2 = r^2 = k^2 - (k_z + \gamma^{(1)} - g_z)^2 \quad ......(6)$$

4.   Installation and operation instruction

HOLZ4 is coded in Java 8 and partially coded for PC with Microsoft Windows only. A recent version of openJDK21 or later version must be installed on a PC with an Microsoft Windows 10 version or later version to run the software.

The Landyne software suite, landyn6, including HOLZ4, is packed in .z7 and .zip forms. The compressed file is available at

https://www.unl.edu/ncmn-enif/xzli/computer-programs
https://landyne.com

For the latest updates and news about HOLZ4 and other Landyne components, please visit the above website. Decompress the landyne6.z7 or landyne6.zip in a selected directory and use the Landyne launcher, landyne6.exe.

The basic steps for using the HOLZ4,

 i) A crystal data file can be read in the menu bar and modified in the fields of the lattice parameters in the HOLZ calculation dialog.
 ii) Select the HOLZ reflections' graphic styles, the HOLZ lines (straight or curved lines) from the menu bar Pattern.
 iii) HOLZ lines can be calculated using an interactively changing voltage, beam direction, lattice parameters, and convergent angle.
 iv) There are dialogs to index the HOLZ lines and a CBED whole pattern.
 v) A pattern in ROI, together with the figure label, can be saved to a file in TIFF, PNG, JPEG, and GIF format.

Although only lattice parameters are involved in the HOLZ4, we usually use the crystal structure file for the Landyne suite as the input file. A new crystal structure data file can be prepared using the **New Crystal Structure File** dialogue window (for more details, see SVAT). The dialogue window provides an automatic assistant for the user and makes sure to meet the requirements of the file format. The template is embedded with the 230 space groups in the Hermann-Mauguin notation, which are listed in the International Table for Crystallography. To save the data structure, click the **Save** button, or make a new one, click the **New** button.

The crystal file can also be converted from 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.

5. Application examples

5.1 Simulation of HOLZ lines under kinematical theory and a first-order dynamical correction

The dynamical interaction produces an important complication for HOLZ simulation. Where the dynamical interaction is strong, the position of the line is not that predicted by Bragg's law. The

line is shifted. It should be noted that the shift can be remedied: Bragg's law is still valid but for a value of the electron wavelength (i.e., microscope voltage), which is not the true value.

The default setting in the HOLZ4 is simulation under kinematical theory. Furthermore, fine-tuning can be carried out using the improved formulas (i) including mean crystal mean potential and (ii) including first-order dynamical correction for HOLZ line shift. Figure 4 shows the simulated HOZL lines in CBED patterns of silicon at [100] zone axis and 100 kV using (a) kinematical theory and (b) a first-order dynamical correction. The two patterns show the shift of reflection lines after adopting the first-order dynamical correction. Figure 4(b) shows better agreement with the experimental pattern under the same conditions in Figure 1 in the paper by Eades (1992).
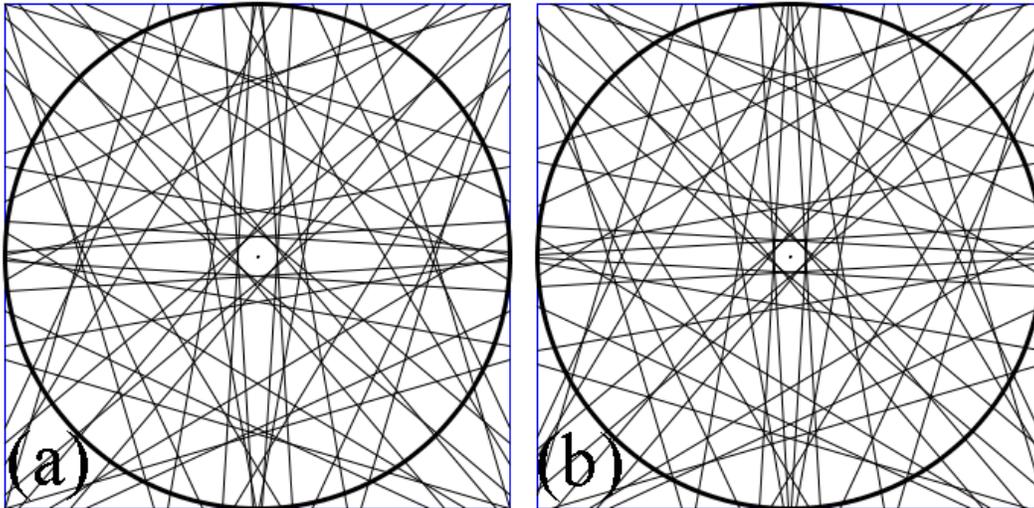

Figure 4. The simulated HOLZ lines on the bright-field disc of a CBED pattern of silicon at [100] zone axis and 100 kV using (a) kinematical theory and (b) a first-order dynamical correction.

5.2 Position of HOLZ lines and the determination of lattice parameters

The HOLZ lines are valuable for the determination of the local lattice parameter of the crystal in the volume traversed by the electron beam. The principle is to measure a single line – HOLZ line – with respect to a nearby reference point. Normally, we are looking for a small change in the lattice parameter, or what is equivalent, the strain in the crystal. Nowadays, it is more usual, indeed it is almost universal, to simulate the pattern of all the lines in the HOLZ disc with a computer program. The advantage of this technique, in addition to its convenience, is that it allows us to obtain the best fit while varying all the unit cell parameters.

Figure 5 shows simulated HOLZ lines in bright-field CBED disc with a Si-type variant structure for comparison, (a) $a = b = c = 0.54294$ nm, (b) $a = 0.54335$ nm, $b = c = 0.54294$ nm, (c) $a = 0.5441$ nm, $b = 0.54294$ nm, $c = 0.5441$ nm. Both experimental bright-field CBED disc and simulated HOLZ lines were shown in Figure 6 in the paper by Eades (1992). The same conditions were used for the simulation here.
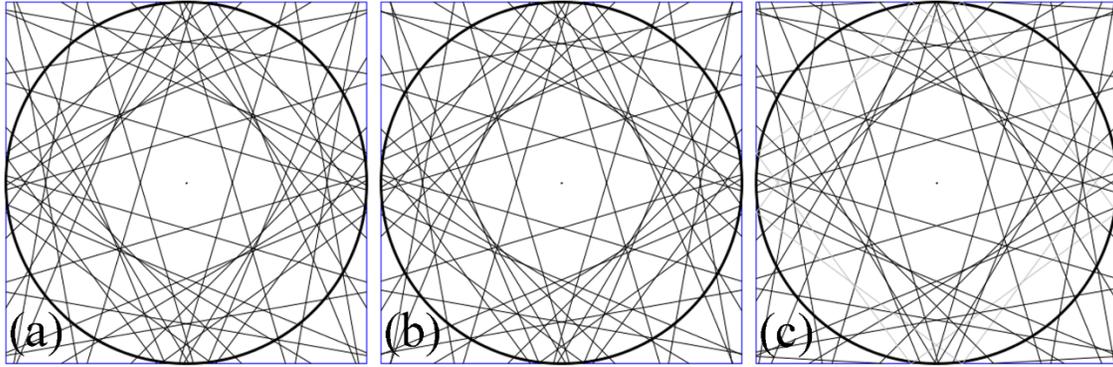
Figure 5. Simulated HOLZ lines in bright-field CBED disc with a Si-type variant structure for comparison, (a) a = b = c = 0.54294 nm, (b) a = 0.54335 nm, b = c = 0.54294 nm, (c) a = 0.5441 nm, b = 0.54294 nm, c = 0.5441 nm.

5.3 Comparison of experimental and simulated HOLZ lines

Figure 6 shows two experimental CBED bright-field discs taken from a Si sample. The zone axes were determined by the software HOLZ3, (a)[5 5 3] and (b) [3 3 1], both with a slight tilt away from the true zone axis. Figures 7 and 8 are the simulated HOLZ lines using the parameters in the adjacent panels. It is clear that the lines in the calculated patterns match well to all the strong HOLZ lines in the experimental CBED bright-field discs.
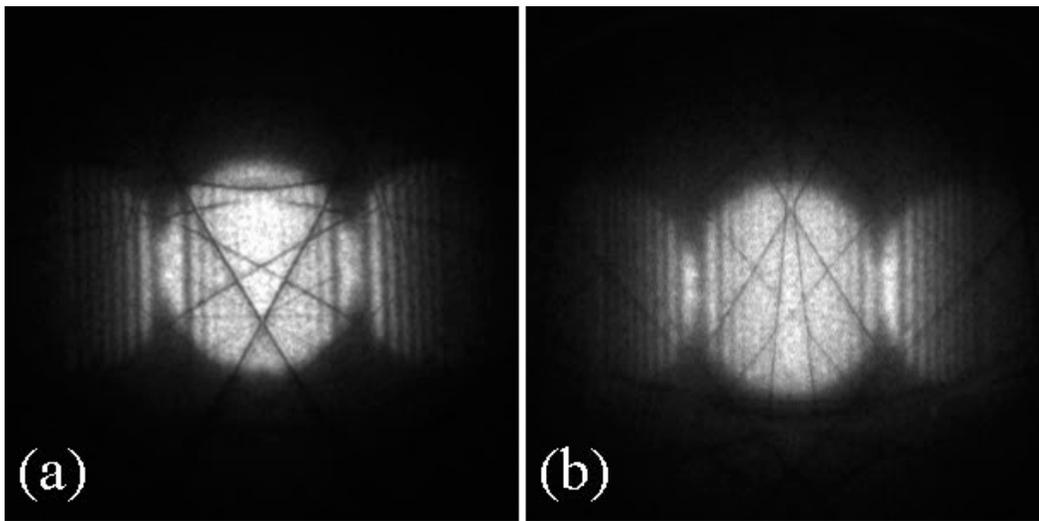

Figure 6. Two CBED bright-field discs were taken from a Si sample with the zone axes (a) [553] and (b) [331], both with a slight tilt away from the true zone axes.
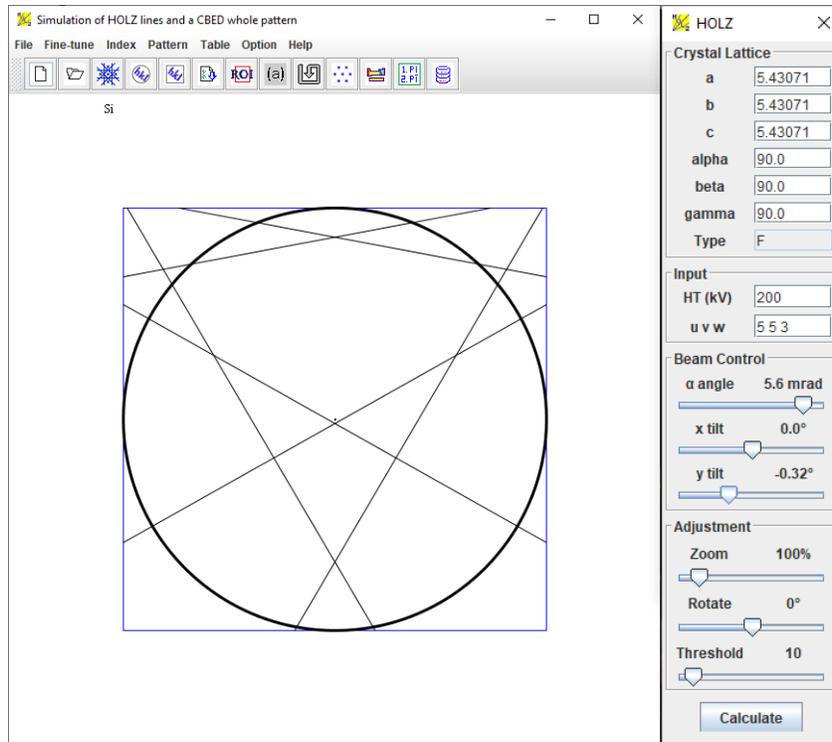
Figure 7. The calculated HOLZ lines in the bright-field disc with the parameters in the adjacent panel. The zone axis is [5 5 3], and a tilt angle away from the zone axis is -0.32º.
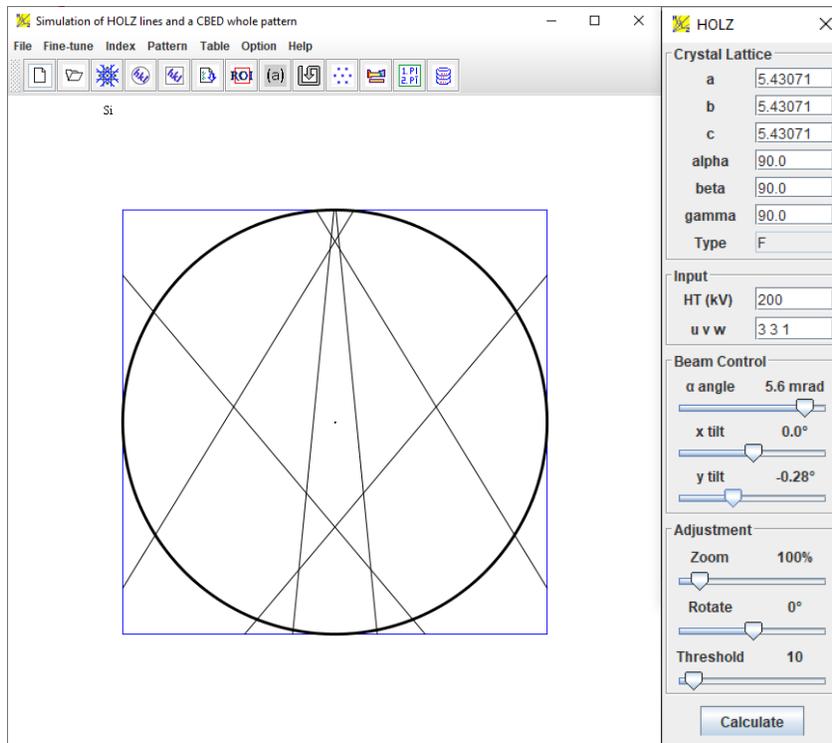

Figure 8. The calculated HOLZ lines in the bright-field disc with the parameters in the adjacent panel. The zone axis is [3 3 1], and a tilt angle away from the zone axis is -0.34º.

6. License and Feedback

Suggestions and comments are welcome; please send them to Dr. X.-Z. Li (jlandyne@gmail.com). Licenses are available from LANDYNE. Without a validated license, this program works in a demo mode.

7. References

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