LANDYNE 蓝带软件

User Manual

Unit-cell determination of crystal phase

in TEM experiments

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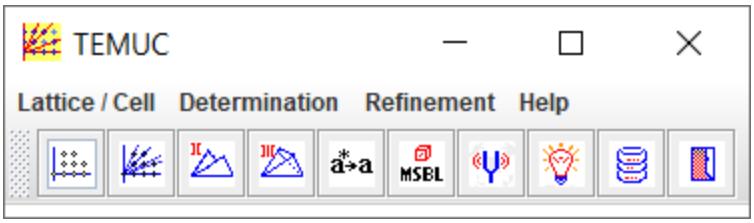

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1. Introduction

Determination of the unit-cell for unknown crystalline phases is an essential requirement for materials characterization and the first step of ab initio structure determination. Electron diffraction technique as a counterpart of X-ray and neutron diffraction techniques has been extensively used in material characterization and structure determination. Lattice parameters from selected-area electron diffraction have accuracy of approximate 5%.

A selected area electron diffraction (SAED) pattern is a two-dimensional section of a three-dimensional reciprocal lattice. A series of reciprocal planes can be obtained by rotating a double tilt holder. Due to the feature of the SAED technique, it is natural to determine the unit-cell for unknown crystalline phases by the method of reciprocal lattice reconstruction from an electron diffraction tilt series. A simple reconstruction method was shown in the book by Vanishtein (1964), i.e., a three-dimensional (3D) lattice can be constructed from an electron diffraction tilt series. This method is troublesome in the application to crystalline phases belonging to monoclinic or triclinic systems. Kuo (1978) applied the concept of the Niggli cell and cell reduction technique on the unit-cell determination in electron diffraction experiments. Similar techniques have been widely used in X-ray crystallography (e.g., Santoro & Mighell, 1970; Gruber, 1973). A general 3D reciprocal lattice reconstruction method was discussed by Fraundorf (1981), and a program with a visual 3D reciprocal space for this purpose was developed by Zou *et al*. (2004).

We developed a set of computer programs for the determination of unit-cell in electron diffraction experiments (Li, 2005), including a modified cell reduction method. The program set has been greatly updated for improvement. The current version is described in the manual, which is a practical tool for the determination of the unit cell of the crystalline phase in TEM, including 1) reciprocal lattice reconstruction approach, 2) the cell reduction approach, and 3) the lattice refinement.

2. Theory background

The reconstruction of a reciprocal lattice from a tilt series of SAED patterns is a simple geometry. There is a shared reflection/vector among the tilt series of SAED patterns, the other reflection/vector in each SAED pattern, which is perpendicular to the shared reflection/vector, is used to construct the 2D lattice, and then the 3D lattice in combination with the shared reflection/vector. The shared reflection/vector is usually the shortest one in the all reflections/vectors in the SAED patterns.

Niggli-cell-reduction theory was firstly used for unit cell determination in x-ray crystallography and later extended in electron diffraction experiments. In 1928, Niggli proved that a crystal lattice could be characterized by a unique choice of a reduced cell, and there are 44 primitive Niggli reduced cells corresponding to the 14 Bravais cells (Niggli, 1928). The importance of the Niggli cell is due to its uniqueness and the possibility that it can be used for determining the Bravais type of the lattice (Gruber, 1973). There are two steps in the determination of the unit-cell: the determination of a reduced direct primitive cell and the

transformation to a conventional cell. Clegg (1981) proposed a procedure to overcome the problem by directly comparing the reduced cell to 44 forms of the Niggli cell. A least-squares refinement procedure is needed to generate optimum lattice parameters.

Two SAED patterns with a common reflection/vector and a tilt angle between them can be used to build a cell for cell reduction and then to determine the unit cell (Li 2005). These reflection/vectors should be the shorter ones in all reflections/vectors in the SAED patterns. Three SAED patterns with common reflection/vectors among each pair of patterns can be used to build a cell for cell reduction and then to determine the unit cell (Yang *et al*. 2017).

3. Design of TEMUC

TEMUC provides tools for (i) a conventional lattice reconstruction, (ii) unit cell determination with cell reduction, and (iii) lattice parameter refinement. The tools can be activated with a dropdown menu or the menu bar and used in various orders according to the task.

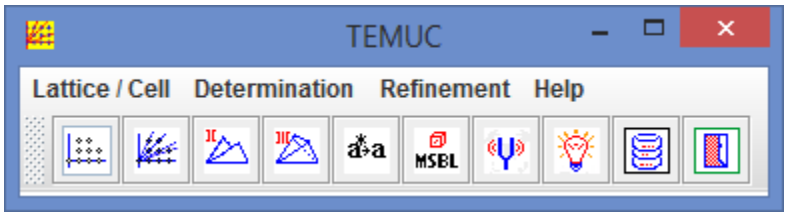

Figure 1. The menu of the TEMUC software.

The normal procedure for Bravais lattice/unit cell determination is (i) reciprocal lattice/cell, (ii) direct lattice/cell, (iii) cell reduction if necessary, (iv) lattice/cell refinement.

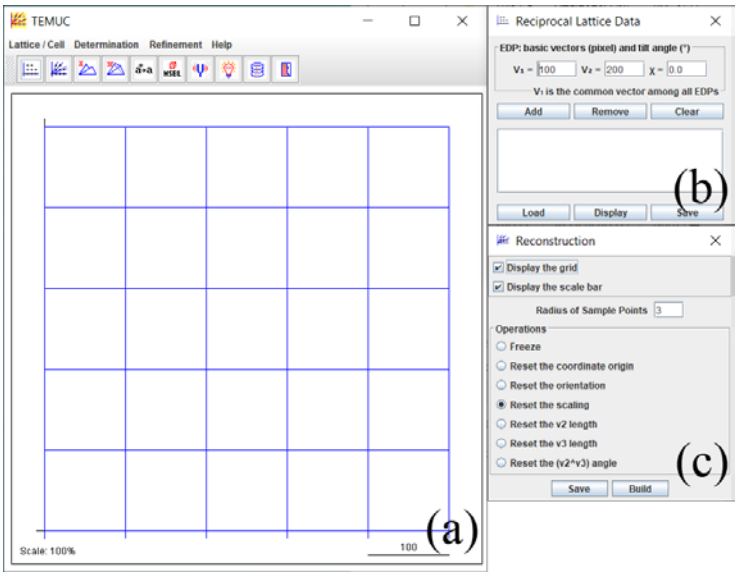

Figure 2. The GUI of the conventional method in the TEMUC software.

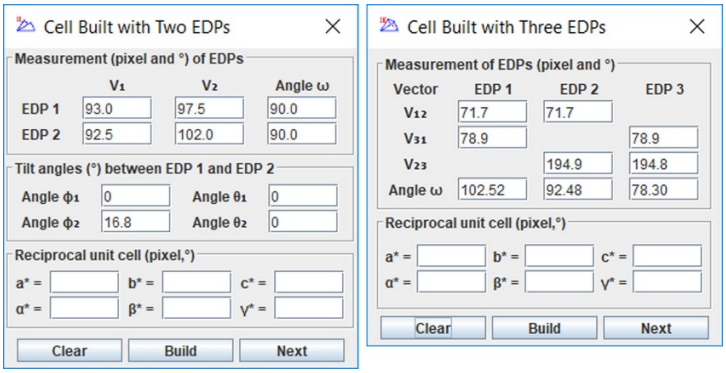

Figure 3. The GUI of the reduced-cell methods in the TEMUC software.

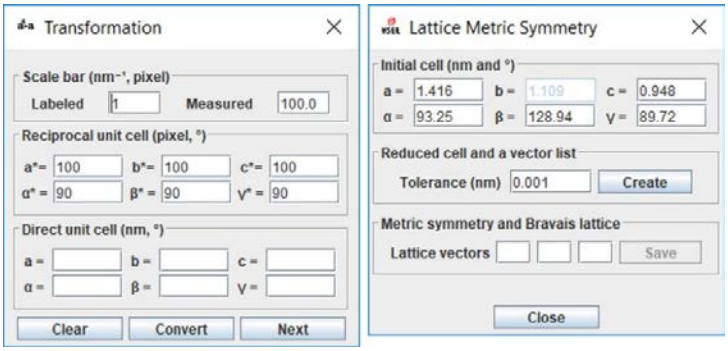

Figure 4. The GUI of the transformation method in the TEMUC software.

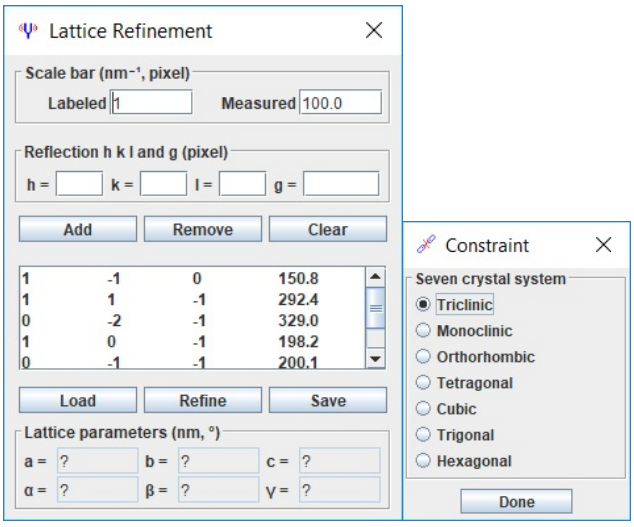

Figure 5. The GUI of the lattice refinement method in the TEMUC software.

The dialogs for the reciprocal lattice reconstruction are shown in Figure 2. The experimental data of the SAED patterns can be inputted in the Reciprocal Lattice Data dialog and displayed in the TEMUC panel. The data analysis is carried out via the Reconstruction dialog. The experimental data can be saved and reloaded.

Two dialogs for building the reciprocal cells to be reduced are shown in Figure 3. One is for the cell built from two SAED patterns plus the tilt angle between them (Li 2005), and the other is for the cell built from three the SAED patterns, in which each pair shares a common reflection/vector (Yang *et al*. 2017).

Figure 4 shows the dialog for the conversion from reciprocal lattice/unit cell to direct lattice/unit cell and the dialog for a cell reduction / Bravais lattice (metric lattice). Figure 5 shows the dialog for lattice refinement from the experimental data of the SAED patterns. The experimental data can be saved and reloaded.

4. Usage of TEMUC

4. 1 A conventional method of the lattice reconstruction

The reciprocal length of reflection/vector and the angle between two vectors can be precisely measured with QSAED software.

The vector 1 should be inputted as the common vector and vector 2 as the other vector; the angle χ is the tilt angles for the SAED patterns. The data set can be saved to a file and reloaded for a demo in a classroom. A demo data can also be loaded by clicking the light bulb icon in the menu bar.

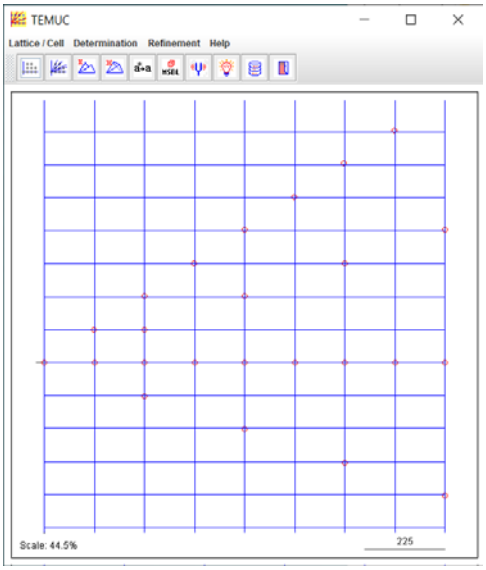

Figure 6. The application of the convention method in the TEMUC software.

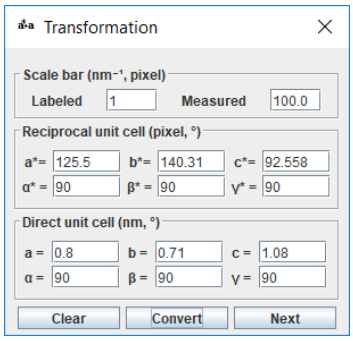

Figure 7. The application of the transformation method in the TEMUC software.

When the data is ready, the graphics of the data will be display in the panel by clicking the Display button. Adjust the net with each operation to match the experimental data; for example, the demo data in Figure 6. Rotate the data set to make the short vector parallel to the horizontal axis.

When the net is matched with experimental data, the reciprocal/direct unit cell can be built, as shown in Figure 7.

4.2 Cell reduction method of unit cell determination

Cell reduction method is (i) to get the cell from the experimental data; (ii) to get the Niggli cell by reduction; (iii) to transfer the Niggli cell to Bravais lattice/unit cell.

Figure 8 shows a method to get the cell from two SAED patterns (Li, 2015) and a method to get the cell from three SAED patterns (Yang *et al*. 2017). In the first method, the vector 1 should be inputted as the common vector and vector 2 as the other vector. Unlike the two reflections/vectors in a SAED pattern used in the conventional method, the angle ω is not restricted to be 90º. In the second method, the common vectors in SAED patterns are marked. It is worthy of pointing out that the derived cell should be the smallest one in the reciprocal space. Here is the two-dimensional analog in Figure 9. The cells spanned by V_1 and V_2 or V_1 and V_3 or V_1 and V_4 or V_2 and V_3 are the smallest cell. On the other hand, the cells spanned by V_2 and V_4 or V_3 and V_4 are not the smallest cell.

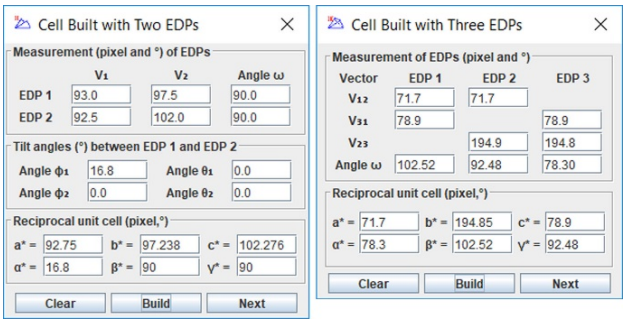

Figure 8. The application of the reduced-cell method in the TEMUC software.

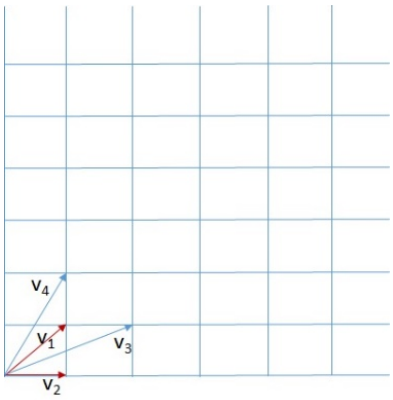

Figure 9. The two-dimensional analogy for the reduced cell.

Cell reduction and metric symmetry will be made in Figure 4. The algorithm proposed by Clegg (1981) was used in the tool. The reduced cell parameters and the vector list will be saved to a file. If there are higher metric symmetry (angle between two vectors is near 90º), the labels of the vectors related to the higher metric symmetry can be inputted the lattice vector, the possible Bravais lattice/unit cell can be saved in a file.

4.3 Unit cell refinement

The parameters of the unit cell should be refined if a list of reflections is available. Each reflection with its spacing measured in the experiment can be indexed by using the lattice parameters determined in previous work. The data set can be saved to a file, and then reloaded for the demonstration in a classroom. A constraint for the refinement needs to be selected, and then the final refined lattice parameters can be obtained.

5. Installation and license

TEMUC can be used as a stand-alone computer program or as one component in the Landyne software suite. The Landyne suite are available in

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

https://landyne.com

Decompress the compressed files to a specified fold e.g., c:\landyne4\. Make sure the latest Java Runtime Environment has been installed on the computer (PC) beforehand. Double click the .exe file to start the software.

A license is available from Landyne (jlandyne@gmail.com). Without the license, the software can be used in a demo mode. The sample data can be loaded from the light-bulb icon in the menu bar.

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