

Multislice Implementation for Inclined Illumination and Convergent-Beam Electron Diffraction

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The multislice formula has been widely used to calculate dynamical diffraction intensities for HREM image simulation. Since the multislice formula is a kind of numerical integration, there are some continuing discussions on its accuracy. Moreover, there are some open questions on an applicability of the multislice formula when used to calculate diffraction intensities in higher order Laue zones (HOLZ) or in the case of an inclined illumination. In this report, the multislice formula applicable for inclined illumination is thoroughly reviewed. A classification of reflections into Laue zones is discussed in terms of an excitation error measured along an incident beam direction. Based on the analysis of the formula an applicability of a simple multislice algorithm for simulating coherent two-dimensional convergent-beam electron diffraction (CBED) is discussed. An example of a CBED pattern obtained from (011) GaAs by intentionally tilting the beam from [011] zone is calculated and compared with an experimental result.

1. Introduction

Since an interaction of an incident electron with a material is generally large, a dynamical treatment is required for a quantitative work. A multislice method is the most efficient numerical procedure for dynamical calculation [1], especially when the FFT technique is invoked [2]. Therefore, the multislice method has been widely used for HREM image simulations of a large unit cell specimen or even of a defect structure. However, the matrix (Bethe) formula has been favored for a diffraction intensity calculation. This may be because a derivation of the matrix formula is mathematically exact, although its application on a physical problem requires some approximations.

The multislice formula was implemented by Goodman and Moodie [3] as a numerical recipe of the physical optics approach that is proposed by Cowley and Moodie [4] for a dynamical diffraction theory. Later the same formula was derived from an integral form of the Schrödinger equation [2]. Since the multislice formula uses a finite thickness of slices, there are some continuing discussions on its accuracy. Moreover, there are some open questions on an applicability of the multislice formula when used to calculate diffraction intensities of higher order Laue zones (HOLZ) [5-7] or applied to the case of an inclined illumination [8].

In a recent paper [8], the authors have developed a new multislice formula by considering back-scattering effects. They have claimed that the new formula will give results that are more accurate on the calculation of HOLZ reflections or be applicable to the case where an incident beam inclined with a large angle with respect to a surface normal. It may be worthwhile to review a paper that treats an inclined illumination published more than 15 years ago [9]. Some physical implications of an inclined illumination will be discussed. An applicability of a simple algorithm for simulating coherent two-dimensional convergent-beam electron diffraction (CBED) will be studied based on the analysis of the formula for an inclined illumination.

2. Inclined Illumination

At first, we examine a relation between the direction of incident electron beam and a sample orientation. There are following four cases (see Fig. 1) in terms of a specimen surface normal

and a zone axis. a) Both the specimen surface normal and the zone axis are parallel to the incident beam direction; b) and c) the specimen surface normal and the zone axis are respectively parallel to the incident beam direction; and d) none of them is parallel to the incident beam direction. Most of the articles that treat the multislice formula assume the first case. Here, the inclined illumination corresponds to (c) to (d). For the electron microscopy the cases of (a) and (c) are important, because they are projections along a low index axes in a real space. In a diffraction experiment, however, an incident beam may be intentionally tilted away from a zone axis.

As an extension of a multislice formula for a normal illumination, even for an inclined illumination a specimen may be divided into slices perpendicular to the incident beam direction. However, some slices at top and bottom parts are not periodic perpendicular to the incident beam direction. Moreover, a large artificial period is required in many cases. A length of the artificial period becomes not acceptable for an infinitesimal tilt. It may be noted that no reflections produced by the artificial periodicity should give rise any intensity, because a period in a reciprocal space is determined by a real crystal lattice not by the artificial lattice. Therefore, this is an inefficient approach to calculate scattering intensities.

3. Multislice formula for an inclined illumination

According to the multislice formula derived for an inclined illumination [9], the following points differ from a classical multislice formula:

- 1) The specimen is divided into slices parallel to an entrance surface. In the case of an inclined illumination, each slice is not perpendicular to the incident electrons.
- 2) A direction of projecting a potential is parallel to an incident electron beam direction, which may not be perpendicular to the slice. Then, diffraction amplitudes converge to kinematical structure factors at the limit of a thin specimen.
- 3) A propagator corresponding to an exact spherical-wave is used instead of a hyperbola that is only valid for a small angle approximation. An excitation error is measured along the surface normal of the entrance surface (see Fig. 1), which agrees with a result derived from a matrix formula.
- 4) A classical interaction constant will be modified by (k / k_z) , where k_z is a wave vector component perpendicular to an entrance surface. Thus, the wave number k in the interaction constant is replaced by an effective wave number k_z .

The above features well reflect a physical scattering situation. If a potential is projected perpendicular to the slice, a slice thickness should be very thin to take into account a lateral shift along the beam direction. This is analogous to replace an oblique straight line by a zigzag line. For an inclined illumination, a slice thickness should be smaller than that for a normal illumination. This is because an effective thickness of the slice is measured along the incident beam direction and also because an interaction constant becomes strong for an inclined illumination.

It may be noted that there is no distinction among all layer line reflections in the multislice formula, since all of them show the same excitation error. In the multislice formula, thus, there are only two indices for each reflection in stead of three Miller indices to identify a reflection. The third index is assigned to a reflection closest to the Ewald sphere.

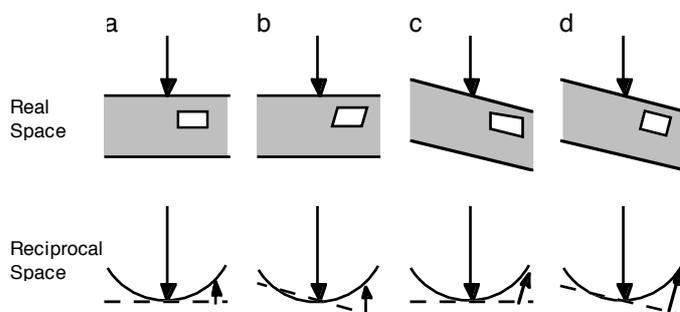


Fig. 1 Relations between an incident electron beam and a sample orientation. Four cases are classified by a relation of an incident beam direction with an entrance surface normal and zone axis.

The symmetry of the two-dimensional diffraction during multislice iteration may differ from an observed one outside of the sample. As an example, we assume the case where an electron beam parallel to a [111] zone axis impinges a (100) surface of a cubic crystal. Since each slice parallel to the incidence surface has a square unit cell, the diffraction lattice in the multislice calculation is square. According to a boundary condition at the exit surface, however, this square reciprocal lattice will transform to a hexagonal lattice outside of the sample when observed at a plane perpendicular to the [111] zone. From the symmetry during calculation, it is clear that the [111] zone diffraction in this case will not show a six-fold symmetry (6mm), but only a mirror symmetry (m). Thus, the multislice formula for an inclined illumination can take account of a surface effect as shown in Fig. 2.

4. Inclined Illumination and HOLZ

When both the specimen surface normal and the zone axis are parallel to the incident beam direction, a classification of reflections into HOLZs is straightforward. Here, reflections on the zone plane belong to the ZOLZ (zero-order Laue zone), and reflections on each reciprocal plane parallel to the zone classified into the corresponding HOLZ. It may be noted that ZOLZ reflections are mostly explained by a projection approximation. Contrary to this, HOLZ reflections depend on an arrangement of atoms along the beam direction, and thus the projection approximation can not handle the HOLZ reflections.

In the case of an inclined illumination, however, the classification of reflections into HOLZs is not so obvious. The ZOLZ and HOLZ reflections locate closely to the Ewald sphere, since an excitation error should be small for a strongly excited reflection. Thus, an excitation error may be used to classify reflections. Since an excitation error appears within a complex exponential, it should be evaluated with modulo of 2π . It becomes then clear that the excitation error measured along the beam direction is used to classify reflections into Laue zones. Thus, there are two excitation errors that determine an excitation of reflection and classify the reflections into Laue zones, respectively.

5. CBED Simulation scheme

A CBED pattern is obtained when incident electrons cover a range of incident directions. If each direction may be treated separately, then an ordinary scheme may be used to calculate diffraction intensities for each direction. This approach using a multislice formula was employed to simulate diffraction intensities on a line within a CBED disk [10].

From a similarity of physical optics and electron microscope optics, a simple procedure to simulate a coherent micro-diffraction pattern in STEM has been developed [11]. Here, a probe wave function is formed on the entrance surface of the specimen. Then, the multislice formula is used to propagate the probe through a specimen. This approach may be used to simulate a coherent high-resolution CBED pattern in CTEM. However, an applicability of the multislice algorithm has not been discussed in terms of an inclined illumination.

It is clear that the above mentioned four points for the inclined illumination should be considered, since a range of incident direction is inclined in CBED. We will study these points according to their order:

1) The first point is easily satisfied, since an incident surface is same for all illumination direction.

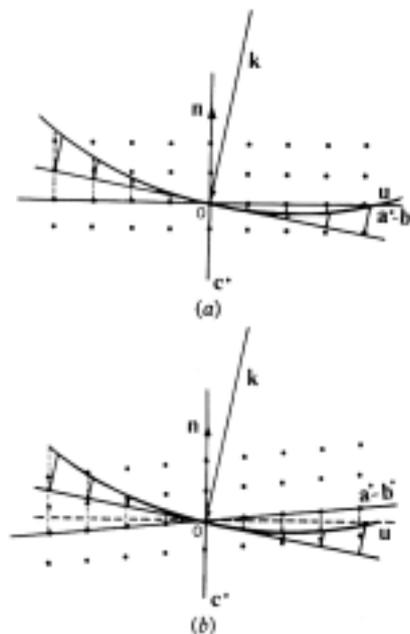


Fig. 2 Two definitions of excitation errors. One is measured along the surface normal (dotted lines) and the other the beam direction (solid lines). (a) and (b) illustrate an orthogonal and a non-orthogonal systems respectively.

2) It is demonstrated that the effect of a potential re-projection is negligible in the case of a sample with medium scattering power, when a range of incident direction is not so large [10]. This effect is easily examined for each sample, if necessary, by comparing the results obtained with and without a potential re-projection.

3) This is the most important factor, since it controls an excitation of reflection. It is shown that an exact excitation error will be assigned for each incident beam direction and its reflections, when the spherical wave propagator is used.

4) A change of the interaction constant is determined by k_z that is simply related to an inclination angle. This effect will be small, if a range of incident direction is not so large.

From the above discussion, it is verified that the multislice formula can be used to simulate a propagation of a probe forming wave function through a sample and thus a coherent CBED pattern. The validity of this approach depends on a convergence angle and a scattering power of constituent atom.

6. Results and Discussion - Inclined CBED pattern

The new multislice formula may be applicable to simulate a CBED pattern observed with a large inclined angle as shown in Fig. 3, which is obtained from a (011) GaAs [10]. Here, an incident beam is intentionally tilted from the [011] zone axis to a direction where the Bragg condition is fulfilled for the $\bar{2}00$, $\bar{1}\bar{1}, 1, \bar{1}$ and $9\bar{1}\bar{1}$ reflections. A detailed feature of 200 reflection and its simulation are shown in Fig. 3 (b) and (c), respectively. In the simulation, a reciprocal space up to 5 \AA^{-1} is sampled by $2k \times 2k$ points. It should be noted that these reflections satisfying the Bragg condition belong to the ZOLZ. Therefore, a good agreement is obtained even when a projection approximation is employed for this calculation. This kind of calculation may be used to evaluate structure by a quantitative comparison with an energy filtered CBED pattern recorded with a high quality CCD camera.

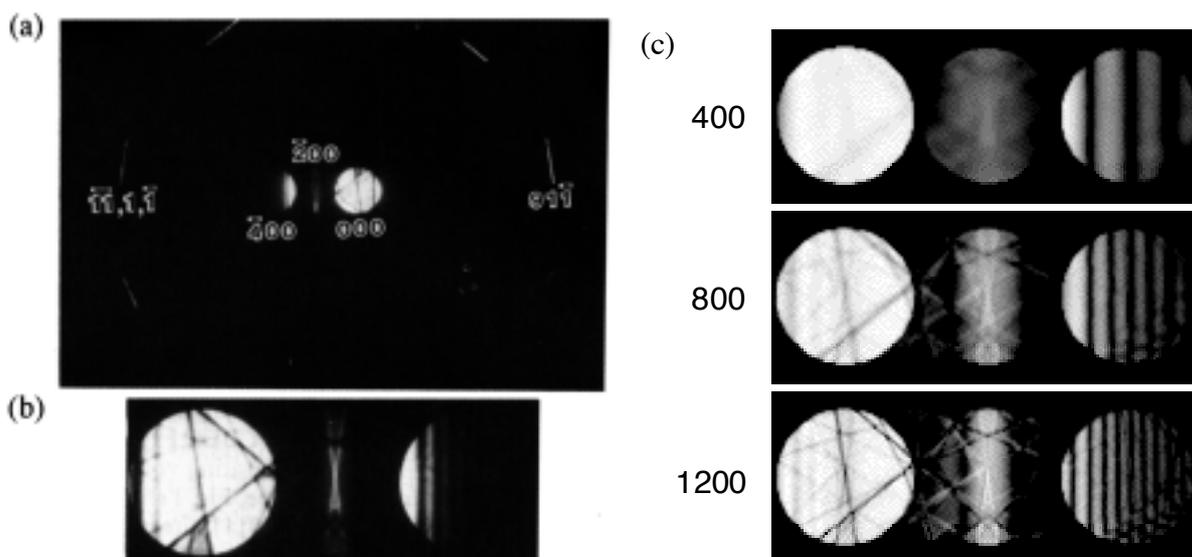


Fig. 3 (a) A CBED pattern obtained from a (011) GaAs with a large inclined angle from the [011] zone. (b) Detailed feature of the 200 diffraction disk at 1000 Å (c) Simulated CBED disks for different specimen thickness as indicated in Å.

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