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# New approach for the dynamical simulation of CBED patterns in heavily strained specimens

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## Abstract

A new method for the dynamical simulation of convergent beam electron diffraction (CBED) patterns is proposed. In this method, the three-dimensional stationary Schrödinger equation is replaced by a two-dimensional time-dependent equation, in which the direction of propagation of the electron beam, variable $z$, stands as a time. We demonstrate that this approach is particularly well-suited for the calculation of the diffracted intensities in the case of a $z$-dependent crystal potential. The corresponding software has been developed and implemented for simulating CBED patterns of various specimens, from perfect crystals to heavily strained cross-sectional specimens. Evidence is given for the remarkable agreement between simulated and experimental patterns.

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## 1. Introduction

Convergent beam electron diffraction (CBED) is widely recognized as a powerful technique to determine the crystal structure (lattice parameters, crystal symmetry, charge density distribution, etc.). Most interestingly, such information can be obtained at the nanometer scale, which makes the technique particularly valuable to investigate local variations in the structure of the studied specimens. In the last decade, both high spatial resolution and remarkable sensitivity to small variations of the lattice parameters have promoted CBED to one of the most appropriate techniques to investigate mechanical strains in new electronics devices [1–7].

While valuable information can be directly retrieved from the experimental patterns, accurate determination of most of the crystal parameters requires comparison with calculated patterns. The usual methods for the simulation of CBED patterns, perfectly suited for perfect crystals [8] and for specific defects [9], proved to be unable to reproduce the complex features observed in CBED patterns of many strained crystals [6,10–12]. Much effort has then been recently devoted to the development of reliable methods to calculate the diffracted intensity distribution in imperfect crystals. Chuvilin and Kaiser [13] developed an original multislice approach to calculate CBED patterns, which should be well adapted to the simulation of imperfect crystals. In our group, we followed a different approach, based on the unified theory for electron diffraction proposed by Gratias and Portier [14]. In this approach, the $z$ direction, represented by the zone axis, is distinguished owing to the small angle approximation for high energy electrons, and the three-dimensional stationary Schrödinger equation is replaced by a two-dimensional $z$-dependent equation, in which $z$ plays the role of a time. Such an equation can then be solved using time-dependent perturbation theory. We demonstrated in a previous paper the interest of this description to calculate the diffracted intensity in non homogeneously strained specimens as epitaxial layers undergoing surface relaxation effects [15]. Remarkable agreement between experimental and simulated HOLZ line intensity profiles was thus obtained for different lines in all the studied regions.

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In the present paper, we describe in details the method that we developed to calculate CBED patterns as well as its implementation using the home-made TDDT software (named after the time-dependent dynamical theory that is being used). The TDDT-calculated central disks of perfect, faulted and strained crystals are compared with experimental ones as well as with CBED central disks calculated by a classical Bloch-wave method [8,16]. The reached agreement validates the use of a time-dependent perturbation theory to take into account the $z$-dependent terms of the crystal potential.

## 2. Basis of the approach

As underlined by Gratias and Portier in Ref. [14], the time-dependent quantum mechanics formalism can be used to describe the interaction of a high energy electron beam with a crystal potential in a TEM, by replacing time by the variable $z$, which coincides with the direction of propagation of the incident beam. In this small angle scattering approximation, the three-dimensional stationary Schrödinger equation will be replaced by a two-dimensional $z$-dependent equation in the following way.

### 2.1. Two-dimensional z-dependent Schrödinger equation

The relativistic Schrödinger equation for electrons elastically scattered by a potential $U$ can be written [17] as

$$(\vec{\nabla}^2 + k^2)\Phi(\vec{r}) = U(\vec{r})\Phi(\vec{r}), \tag{1}$$

where $k$ is the modulus of the electron wave vector and $U = -(2m/\hbar^2)eV$, in which $V$ is the actual crystal potential. In practice, let us choose as the $z$-direction the zone axis that is closer to the direction of the incident electron beam. The $z$-component of the wave vector will be noted $k_z$ and the wave function $\Phi$ will then be expressed by

$$\Phi(\vec{r}) = \mathrm{e}^{\mathrm{i}k_z z}\Psi(\vec{\rho}, z) \quad \text{with } \vec{\rho} = (x, y). \tag{2}$$

The small angle approximation allows us to replace Eq. (1) by the following one:

$$\mathrm{i}\frac{\partial\Psi(\vec{\rho}, z)}{\partial z} = H(\vec{r})\Psi(\vec{\rho}, z)$$

$$\text{with} \quad 2k_z H(\vec{r}) = -\vec{\nabla}^2_{xy} - \vec{\chi}^2 + U(\vec{r}), \tag{3}$$

in which $\vec{\chi}$ represents the radial component of the wave vector in the $(x, y)$ plane and $\vec{\nabla}^2_{x,y} = \partial^2/\partial x^2 + \partial^2/\partial y^2$ is the radial kinetic energy operator.

As this is an evolution equation, a solution can be written as

$$\Psi(\vec{\rho}, z) = \mathcal{U}(z, z_0)\Psi(\vec{\rho}, z_0), \tag{4}$$

where $\mathcal{U}(z, z_0)$ is the causal evolution operator,

$$\mathrm{i}\frac{\partial\mathcal{U}(z, z_0)}{\partial z} = H(z)\mathcal{U}(z, z_0). \tag{5}$$

Note that in many cases, this equation cannot be solved exactly. Approximate solutions are then obtained using time-dependent perturbation theory.

### 2.2. z-Dependence of the crystal potential

In the perfect crystal, the periodic potential is usually developed in a Fourier series and can thus be written as

$$U(\vec{r}) = \sum_{\vec{g}} U_{\vec{g}} \exp(\mathrm{i}\vec{g}\cdot\vec{r}),$$

$$U_{\vec{g}} = \frac{1}{V_\mathrm{c}}\int_{cell} \mathrm{d}\vec{r}\, U(\vec{r}) \exp(-\mathrm{i}\vec{g}\cdot\vec{r}), \tag{6}$$

where $\vec{g}$ stands for a reciprocal lattice vector and $V_\mathrm{c}$ is the volume of the unit cell. To take into account the particularities of CBED, and more precisely to separate the contributions of the zero-order Laue zone (ZOLZ) from the ones of the higher-order Laue zones (HOLZ), the reciprocal lattice vectors will be written $\vec{g} = (\vec{G}, -ng_z)$, where $\vec{G}$ belongs to the zero-order Laue zone, $g_z$ is the distance between two neighboring Laue zones and $n$ the order of the zone (i.e. $n = 1$ for the first-order Laue zone). Using these notations, first proposed by Vincent et al. [18], the crystal potential can then be developed as

$$U(\vec{r}) = U^{(0)}(\vec{\rho}) + \sum_{n\neq 0} \exp(-\mathrm{i}ng_z z)U^{(n)}(\vec{\rho}),$$

$$\text{in which} \quad U^{(n)}(\vec{\rho}) = \sum_{\vec{G}} U^{(n)}_{(\vec{G}, -ng_z)} \exp(\mathrm{i}\vec{G}\cdot\vec{\rho}). \tag{7}$$

In this expression, the ZOLZ potential, $U^{(0)}(\vec{\rho})$, corresponds to the well-known projected potential while the other components $U^{(n)}(\vec{\rho})$ are known as the higher-order conditional projected potentials [18].

Using the development of $U(\vec{r})$ in Eq. (7), the Hamiltonian $H(\vec{r})$ given in Eq. (3) can be written as

$$\begin{aligned} 2k_z H(\vec{r}) &= -\vec{\nabla}^2_{xy} - \vec{\chi}^2 + U^{(0)}(\vec{\rho}) + \sum_{n\neq 0} \exp(-\mathrm{i}ng_z z)U^{(n)}(\vec{\rho}) \\ &= 2k_z H^{(0)}(\vec{\rho}) + \sum_{n\neq 0} \exp(-\mathrm{i}ng_z z)U^{(n)}(\vec{\rho}). \end{aligned} \tag{8}$$

The $H(\vec{r})$ Hamiltonian is thus clearly separated into a $z$-independent and a $z$-dependent part. Eq. (8) can then be solved in two steps, the $z$-dependent term being introduced as a perturbation of the $z$-independent operator $H^{(0)}$ [14]. As remarked by Bird [19], $H^{(0)}$ corresponds to the ZOLZ diffraction. We write

$$2k_z H(\vec{r}) = 2k_z H^{0=\mathrm{ZOLZ}}(\vec{\rho}) + U^{\mathrm{HOLZ}}(\vec{\rho}, z). \tag{9}$$

## 3. Presentation of the method

### 3.1. The ZOLZ diffraction

As $H^{(0)}$ is $z$-independent, Eq. (5) can be solved exactly (see for instance Ref. [14]) and its corresponding evolution