Treatment of Inelastic Scattering and Backscattering in the Multislice Scheme

from:

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1 Introduction

This project focuses on the examination of backscattering of electrons by crystals, as observed in transmission electron microscopy (TEM), with computational methods. A newly developed approach which will be briefly sketched in section 2.1, was implemented and tested for the single backscattering approximation. The obtained results will be presented in section 3.

The motivation for this work lies in the necessity of developing new theoretical methods to describe electron scattering in order to be able to describe and interpret recent experimental developments. In order to not damage the sample, electron microscopes are nowadays frequently run at low acceleration voltages. The resulting low energetic electron beams will show significant amounts of backscattering, inelastic effects can be expected as well - together an unsurpassable obstacle for an accurate theoretical description using methods available today. Another example can be found in measurements utilizing electron vortex beams [1], which may open access to an easy experimental measurement of magnetic properties of materials down to atomic resolution. Since existing approaches are either incapable of describing such sophisticated beam shapes as used in these experiments or can not reach a sufficient accuracy, the method used and proven to be feasible here is likely to help the theoretical understanding of future experiments.

2 Theory

2.1 General non-relativistic Multislice Method

To describe the non-relativistic scattering of electrons in crystals generally, including backscattering as well as inelastic scattering, a combined approach using the back scattering mechanism developed by Chen and Van Dyck [2] and the ansatz for inelastic scattering by Yoshioka [3] can be used.

In Yoshioka’s approach, the electronic wavefunction is defined by the Schrödinger equation

\[
\left[ \nabla^2 + 4\pi^2 K_n^2 \right] \psi_n(r) = \frac{2m_e}{\hbar^2} \sum_m H_{nm}(r) \psi_m(r) \tag{1}
\]

where \( \psi_n(r) \) are the electronic wavefunctions corresponding to the energy \( E_n = E_0 - \epsilon_n \) with \( E_0 \) as energy of the initial electron beam and \( \epsilon_n \) as energy of the crystal eigenstate \( a_n(r_1, ..., r_M) \). With the energies \( E_n, K_n \) can be computed

\[
K_n^2 = \frac{2m_0}{\hbar^2} E_n \tag{2}
\]
$H_{nm}(r)$ is defined as

$$H_{nm}(r) = \int dr_1...dr_M a_n^*(r_1,...,r_M) H_{int}(r_1,...,r_M) a_m(r_1,...,r_M)$$

(3)

where $H_{int}(r_1,...,r_M)$ is the interaction Hamiltonian. Since the diagonal elements of $H_{nm}$ simply correspond to the crystal potential $U(r)$, eq.1 can be rewritten as

$$\left[\nabla^2 + 4\pi^2(K^2_n + U(r))\right] \psi_n(r) = \frac{2m_e}{\hbar^2} \sum_{m \neq n} H_{nm}(r) \psi_m(r)$$

(4)

In the multislice scheme, the potential is partitioned into a series of slices along the $z$ axis, parallel to the surface. The potential of each slice reads

$$U_j(b) = \frac{1}{\epsilon} \int_{(j-1)\epsilon}^{j\epsilon} dz U(r)$$

(5)

where $b = (x, y)$ and $\epsilon$ denotes the slice thickness. Introducing the wavevector operator $\hat{k}^{(n)}(r)$

$$\hat{k}^{(n)}(r) = \sqrt{\frac{\Delta_b}{4\pi^2} + K^2_n + U(r)}$$

(6)

eq.4 can be rewritten as

$$[\nabla^2 + (2\pi \hat{k}^{(n)}(r))^2] \psi_n(r) = \frac{2m_e}{\hbar^2} \sum_{m \neq n} H_{nm}(r) \psi_m(r)$$

(7)

For a single slice, one has to substitute $U(r) \rightarrow U_j(b)$. The general solution can be written as a sum of a homogenous and inhomogenous solution to this inhomogenous differential equation. The homogenous part is a superposition of a forward and a backward propagating plane wave multiplied with a slowly varying function, $\Phi_n^{(f,j)}$ and $\Phi_n^{(b,j)}$:

$$\psi_n^{(j,h)}(b,z) = e^{2\pi i \hat{k}^{(n)}(r)_z} \Phi_n^{(f,j)}(b,z - \epsilon) + e^{-2\pi i \hat{k}^{(n)}(r)_z} \Phi_n^{(b,j)}(b,z - \epsilon)$$

(8)

The inhomogenous solution can be found using the Green’s function method [4]. One obtains:

$$\psi_n^{(j,i)}(b,z) = \frac{m_e}{2\pi^2 \hbar^2} \int dr' \sum_{m \neq n} H_{nm}(r') \psi_m(r') \int du \frac{e^{2\pi i u(r-r')}}{4\pi^2(K^2_n + U(r) - u^2)}$$

(9)

$$= \frac{m_e}{2\pi^2 \hbar^2} \int dr' \sum_{m \neq n} H_{nm}(r') \psi_m(r') G_0(r - r')$$

(10)
The general solution to eq.7 is thus

$$\psi_n^{(j)}(b, z) = \psi_{n^{(h)}}(b, z) + \psi_{n^{(j)}}(b, z)$$

$$= e^{2\pi i \hat{k}_{j-1}^{(n)}} \Phi_n^{(f)}(b, z - \epsilon) + e^{-2\pi i \hat{k}_{j}^{(n)}} \Phi_n^{(b)}(b, z - \epsilon) + \frac{m_e}{2\pi^2 \hbar^2} \int dr' \sum_{m \neq n} H_{nm}(r') \psi_m(r') G_0(r - r')$$

Using the boundary condition that wavefunctions between two slices should be smoothly connected, i.e.

$$\psi_n^{(j-1)}|_{z=(j-1)\epsilon} = \psi_n^{(j)}|_{z=(j-1)\epsilon}$$

$$\frac{\partial}{\partial z} \psi_n^{(j-1)}|_{z=(j-1)\epsilon} = \frac{\partial}{\partial z} \psi_n^{(j)}|_{z=(j-1)\epsilon}$$

These boundaries yield $2n \cdot N$ coupled equations for $N$ slices and $n - 1$ excitations considered. The boundaries lead to

$$e^{2\pi i \hat{k}_{j-1}^{(n)}} \Phi_n^{(f)} - e^{-2\pi i \hat{k}_{j-1}^{(n)}} \Phi_n^{(b)} = \left[ \frac{m_e}{2\pi^2 \hbar^2} \int dr' \sum_{m \neq n} H_{nm}(r') \psi_m(r') G_0(r - r') \right]_{z=(j-1)\epsilon}$$

$$2\pi i \hat{k}_j^{(n)} \left( e^{2\pi i \hat{k}_{j-1}^{(n)}} \Phi_n^{(f)} - e^{-2\pi i \hat{k}_{j-1}^{(n)}} \Phi_n^{(b)} \right) = 2\pi i \hat{k}_j^{(n)} \left( \Phi_n^{(f)} - \Phi_n^{(b)} \right) - \left[ \frac{m_e}{2\pi^2 \hbar^2} \int dr' \sum_{m \neq n} H_{nm}(r') \psi_m(r') G_0(r - r') \right]_{z=(j-1)\epsilon}$$

Introducing

$$\hat{F}_{j-1} = \frac{1}{2} \left( 1 + \hat{k}_{j-1}^{-1} \hat{k}_j \right)$$

and

$$\hat{B}_{j-1} = \frac{1}{2} \left( 1 - \hat{k}_{j-1}^{-1} \hat{k}_j \right)$$

1

4
One can relate $\Phi^{(i)}_{j-1}$ to $\Phi^{(i)}_j$. For $\Phi^{(j,f)}_n$ one finds

$$
\Phi^{(j,f)}_n = \left[ \frac{m_e}{2\pi^2 \hbar^2} \int dr' \sum_{m \neq n} H_{nm}(r') \psi_m(r') G_0(r - r') \right]_{z=j} \\
- \frac{\langle \vec{b}^{(n)} \rangle}{2\pi i} \left[ \frac{m_e}{2\pi^2 \hbar^2} \partial_z \int dr' \sum_{m \neq n} H_{nm}(r') \psi_m(r') G_0(r - r') \right]_{z=j} \\
= \tilde{F}_{j,j-1} e^{2\pi i \vec{k}^{(n)}_{j-1} r} \Phi^{(j,f-1)}_n + \tilde{B}_{j,j-1} e^{-2\pi i \vec{k}^{(n)}_{j-1} r} \Phi^{(b,j-1)}_n \\
- \frac{1}{2} \left( \frac{m_e}{2\pi^2 \hbar^2} \int dr' \sum_{m \neq n} H_{nm}(r') \psi_m(r') G_0(r - r') \right)_{z=(j-1)\epsilon} \\
+ \frac{\langle \vec{k}^{(n)} \rangle}{2\pi i} \left[ \frac{m_e}{2\pi^2 \hbar^2} \partial_z \int dr' \sum_{m \neq n} H_{nm}(r') \psi_m(r') G_0(r - r') \right]_{z=(j-1)\epsilon} \\
(19)
$$

For the backwards propagating beam, one gets similarly

$$
\Phi^{(j,b)}_n = \left[ \frac{m_e}{2\pi^2 \hbar^2} \int dr' \sum_{m \neq n} H_{nm}(r') \psi_m(r') G_0(r - r') \right]_{z=j} \\
+ \frac{\langle \vec{k}^{(n)} \rangle}{2\pi i} \left[ \frac{m_e}{2\pi^2 \hbar^2} \partial_z \int dr' \sum_{m \neq n} H_{nm}(r') \psi_m(r') G_0(r - r') \right]_{z=j} \\
= \tilde{B}_{j,j-1} e^{2\pi i \vec{k}^{(n)}_{j-1} r} \Phi^{(j,-1)}_n + \tilde{F}_{j,j-1} e^{-2\pi i \vec{k}^{(n)}_{j-1} r} \Phi^{(b,j-1)}_n \\
- \frac{1}{2} \left( \frac{m_e}{2\pi^2 \hbar^2} \int dr' \sum_{m \neq n} H_{nm}(r') \psi_m(r') G_0(r - r') \right)_{z=(j-1)\epsilon} \\
- \frac{\langle \vec{k}^{(n)} \rangle}{2\pi i} \left[ \frac{m_e}{2\pi^2 \hbar^2} \partial_z \int dr' \sum_{m \neq n} H_{nm}(r') \psi_m(r') G_0(r - r') \right]_{z=(j-1)\epsilon} \\
(20)
$$

Unfortunately, these equations are coupled to all other excited states of the $(j - 1)$th and $j$th slice. Therefore, only a self-consistent solution can be found. To proceed searching for another solution, one has to substitute eq.12 into these equations. Since eq.12 contains an inelastic part as well, one can resubstitute this equation basically any number of times, where each resubstitution corresponds to an inelastic scattering event in this slice. In the following, it is assumed that the beam is inelastically scattered only once in each slice. Hence, one substitutes

$$
\psi^j_n(b, z) = e^{2\pi i \vec{k}^{(m)}_{j} z} \Phi^{(j,m)}_n(b, z - \epsilon) + e^{-2\pi i \vec{k}^{(m)}_{j} z} \Phi^{(b,m)}_n(b, z - \epsilon) \\
(21)
$$
into eqs. 19, 20. It is possible to write eqs. 19, 20 more concise for all $n$ in a matrix equation.

$$\hat{P}^{(j)} \cdot \Phi^{(j)} = \hat{F}^{(j-1)} \cdot \Phi^{(j-1)}$$

(22)

Or explicitly

$$
\begin{pmatrix}
1 & 0 & \hat{P}_{01}^{(j,f)} & \hat{P}_{01}^{(j,b)} & \cdots & \hat{P}_{0n}^{(j,f)} & \hat{P}_{0n}^{(j,b)} \\
0 & 1 & \hat{P}_{01}^{(j,f)} & \hat{P}_{01}^{(j,b)} & \cdots & \hat{P}_{1n}^{(j,f)} & \hat{P}_{1n}^{(j,b)} \\
\hat{P}_{j,f}^{(1)} & \hat{P}_{j,f}^{(2)} & 1 & 0 & \cdots & \hat{P}_{1n}^{(j,f)} & \hat{P}_{1n}^{(j,b)} \\
\hat{P}_{j,f}^{(1)} & \hat{P}_{j,f}^{(2)} & 0 & 1 & \cdots & \hat{P}_{1n}^{(j,f)} & \hat{P}_{1n}^{(j,b)} \\
\vdots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\
\hat{P}_{n0}^{(j,f)} & \hat{P}_{n0}^{(j,b)} & \hat{P}_{n1}^{(j,f)} & \hat{P}_{n1}^{(j,b)} & \cdots & 1 & 0 \\
\hat{P}_{n0}^{(j,f)} & \hat{P}_{n0}^{(j,b)} & \hat{P}_{n1}^{(j,f)} & \hat{P}_{n1}^{(j,b)} & \cdots & 0 & 1 \\
\end{pmatrix}
\begin{pmatrix}
\Phi^{(f,0)} \\
\Phi^{(b,0)} \\
\Phi^{(f,1)} \\
\Phi^{(b,1)} \\
\Phi^{(f,n)} \\
\Phi^{(b,n)} \\
\end{pmatrix} =
$$

(23)

where the integral operators $\hat{P}_{nm}$ are defined as

$$\hat{P}_{nm}^{(i,f)} = -\frac{me}{2\pi^2\hbar^2} \left[ \int dr H_{nm} G_0(r-r')e^{2\pi ik_{i,m}} + \frac{(k_{i,m})^{-1}}{2\pi i} \int dr' H_{nm} G_0(r-r')e^{2\pi ik_{i,m}} \right]$$

(24)

and

$$\hat{P}_{nm}^{(i,b)} = -\frac{me}{2\pi^2\hbar^2} \left[ \int dr' H_{nm} G_0(r-r')e^{-2\pi ik_{i,m}} - \frac{(k_{i,m})^{-1}}{2\pi i} \int dr' H_{nm} G_0(r-r')e^{-2\pi ik_{i,m}} \right]$$

(25)
Given that the inverse of $\mathbf{P}^{(j)}$ exists, a formal solution can be written as:

$$\Phi^{(j)} = (\mathbf{P}^{(j)})^{-1} \cdot \tilde{\mathbf{F}}^{(j-1)} \cdot \Phi^{(j-1)}$$  \hspace{1cm} (26)

$$= \tilde{\mathbf{S}}^{(j-1)} \cdot \Phi^{(j-1)}$$  \hspace{1cm} (27)

introducing the slice transition matrix operator $\mathbf{S}^{(j-1)}$. One possible approximation to find $(\mathbf{P}^{(j)})^{-1}$ is to assume that the inelastic contribution to the system kinetics are small in comparison to the elastic part. Writing $\mathbf{P}^{(j)} = 1 + \tilde{\mathbf{P}}^{(j)}$, where $\tilde{\mathbf{P}}^{(j)}$ are the off-diagonal elements of $\mathbf{P}^{(j)}$, one can approximate:

$$(\tilde{\mathbf{P}}^{(j)})^{-1} = (1 + \tilde{\mathbf{P}}^{(j)})^{-1}$$  \hspace{1cm} (28)

$$\approx 1 - \tilde{\mathbf{P}}^{(j)}$$  \hspace{1cm} (29)

in analogy with $\frac{1}{1+x} \approx 1 - x$ for small $x$.

The computation and derivation of the inverse operator becomes much simpler, if the assumption that the beam is scattered inelastically only once is made. All elements $\tilde{P}^{(ij)}_{nm}$ of the matrices $\tilde{\mathbf{P}}^{(j)}$ and $\tilde{\mathbf{F}}^{(j-1)}$ with $m \neq 0$ will vanish so that $(\tilde{\mathbf{P}}^{(j)})^{-1}$ simply reads

$$\left( \begin{array}{ccccccc}
1 & 0 & 0 & 0 & \ldots & 0 & 0 \\
0 & 1 & 0 & 0 & \ldots & 0 & 0 \\
-\tilde{P}^{(j)f} & -\tilde{P}^{(j)b} & 1 & 0 & \ldots & 0 & 0 \\
-\tilde{P}^{(j)f} & -\tilde{P}^{(j)b} & 0 & 1 & \ldots & 0 & 0 \\
\vdots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\
-\tilde{P}^{(i)f} & -\tilde{P}^{(i)b} & 0 & 0 & \ldots & 1 & 0 \\
-\tilde{P}^{(n)f} & -\tilde{P}^{(n)b} & 0 & 0 & \ldots & 0 & 1 \\
\end{array} \right)$$  \hspace{1cm} (30)

Note that this expression is exact; it coincides with the approximation in eq.29 which is a first order approximation as well. If all matrix elements of $\tilde{\mathbf{P}}^{(j)}$ were taken into account, the approximation in eq.29 would differ from the exact inverse.
The slice transition matrix operator $\hat{S}^{(j-1)}$ can be written as

$$
\hat{S}^{(j-1)} = \begin{pmatrix}
\hat{F}^{(0)}_{j,j-1} e^{2\pi i \hat{k}^{(0)}_{j,j-1}} & \hat{B}^{(0)}_{j,j-1} e^{-2\pi i \hat{k}^{(0)}_{j,j-1}} & 0 & 0 & \ldots & 0 & 0 \\
\hat{B}^{(0)}_{j,j-1} e^{2\pi i \hat{k}^{(0)}_{j,j-1}} & \hat{F}^{(0)}_{j,j-1} e^{-2\pi i \hat{k}^{(0)}_{j,j-1}} & 0 & 0 & \ldots & 0 & 0 \\
\hat{P}^{(j-1,f)}_{10} - \hat{C}^{(f)}_{10} & \hat{P}^{(j-1,b)}_{10} - \hat{C}^{(b)}_{10} & \hat{F}^{(1)}_{j,j-1} e^{2\pi i \hat{k}^{(1)}_{j,j-1}} & \hat{B}^{(1)}_{j,j-1} e^{-2\pi i \hat{k}^{(1)}_{j,j-1}} & 0 & 0 & \ldots & 0 & 0 \\
\hat{P}^{(j-1,f)}_{10} - \hat{C}^{(f)}_{10} & \hat{P}^{(j-1,b)}_{10} - \hat{C}^{(b)}_{10} & \hat{F}^{(1)}_{j,j-1} e^{2\pi i \hat{k}^{(1)}_{j,j-1}} & \hat{B}^{(1)}_{j,j-1} e^{-2\pi i \hat{k}^{(1)}_{j,j-1}} & 0 & 0 & \ldots & 0 & 0 \\
\vdots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \vdots \\
\hat{P}^{(j-1,f)}_{n0} - \hat{C}^{(f)}_{n0} & \hat{P}^{(j-1,b)}_{n0} - \hat{C}^{(b)}_{n0} & \hat{F}^{(n)}_{j,j-1} e^{2\pi i \hat{k}^{(n)}_{j,j-1}} & \hat{B}^{(n)}_{j,j-1} e^{-2\pi i \hat{k}^{(n)}_{j,j-1}} & \hat{C}^{(f)}_{n0} & \hat{C}^{(b)}_{n0} & \hat{F}^{(n)}_{j,j-1} e^{2\pi i \hat{k}^{(n)}_{j,j-1}} & \hat{B}^{(n)}_{j,j-1} e^{-2\pi i \hat{k}^{(n)}_{j,j-1}}
\end{pmatrix}
$$

(31)

Where

$$
\hat{C}^{(f)}_{nm} = \hat{P}^{(j,f)}_{nm} \hat{F}^{(0)}_{j,j-1} e^{2\pi i \hat{k}^{(0)}_{j,j-1}} + \hat{P}^{(j,b)}_{nm} \hat{B}^{(0)}_{j,j-1} e^{-2\pi i \hat{k}^{(0)}_{j,j-1}}
$$

(32)

and

$$
\hat{C}^{(b)}_{nm} = \hat{P}^{(j,f)}_{nm} \hat{B}^{(0)}_{j,j-1} e^{-2\pi i \hat{k}^{(0)}_{j,j-1}} + \hat{P}^{(j,b)}_{nm} \hat{F}^{(0)}_{j,j-1} e^{2\pi i \hat{k}^{(0)}_{j,j-1}}
$$

(33)

### 2.2 Computational Scheme

A calculation using the general formula eq.22, would require immense computational resources due to the nature of the objects in the equation. Furthermore, a self-consistent solution has to be sought since the coupling of forward and backward propagating wave of each state with the waves from each other state does not allow any other procedure to obtain a solution.

In the framework of the single inelastic scattering approximation, the computation becomes a lot easier. At first, many matrix elements are zero and multiplication with the inverse matrix does not lead to multiple sums over integral operators, but rather simple expressions in the first two columns of $\hat{S}^{(j-1)}$. Furthermore, most of the couplings disappear, the two waves of the zeroth state are coupled to each other only and all other waves just to the wave of the same state travelling into the opposite direction and the zeroth states waves. A solution to this system can be found by a similar ansatz as in the elastic case: One uses the block diagonal form of $\hat{S}^{(j-1)}$ to define elastic forward and backward propagation matrix for the $n$th state. Additionally to this elastic parts, $\hat{S}^{(j-1)}$ has inelastic
contributions in the first two columns. Each elastic propagation matrix reads

$$\mathbf{S}^{(j-1,n)}_{el} = \begin{pmatrix} \mathbf{F}_{j,j-1}^{(n)} e^{2\pi i k_{j-1}^{(n)} e} & \mathbf{B}_{j,j-1}^{(n)} e^{-2\pi i k_{j-1}^{(n)} e} \\ \mathbf{B}_{j,j-1}^{(n)} e^{2\pi i k_{j-1}^{(n)} e} & \mathbf{F}_{j,j-1}^{(n)} e^{-2\pi i k_{j-1}^{(n)} e} \end{pmatrix}$$

(34)

$$\mathbf{S}^{(f,n)} = \mathbf{S}^{(b,n)}_{j-1} + \mathbf{S}^{(b,cont)}_{j-1}$$

(35)

$$\begin{pmatrix} s_{11}^{12} & 0 \\ s_{21}^{12} & 0 \end{pmatrix} + \begin{pmatrix} 0 & s_{j-1}^{12} \\ 0 & s_{j-1}^{22} \end{pmatrix}$$

(36)

The computation starts with the calculation of $\Phi_0^{(f)}$ at every slice. This step corresponds to the elastic propagation of the incident electron beam of whom prior to the computation only the forward traveling wavefunction at the zeroth slice, $\Phi_0^{(f)}$, is known. This function can be propagated using only $\mathbf{S}^{(f,0)}_{j-1}$ yielding $\Phi_N^{(f)}$ to zeroth order of backscatter events. Additionally, at every slice, a back scattered part $\Phi_i^{(b,cont)}$ is obtained. Starting from the $(N-1)$th slice, where $\Phi_{N-1}^{(b,cont)} = \Phi_{N-1}^{(b)}$ since $\Phi_N^{(b)} = 0$, the backscattered wave can be propagated backwards using the inverse of $\mathbf{S}_{j-1}^{(b,0)}$. After having propagated from the $(i+1)$th slice to the $i$th slice, one has to add $\Phi_i^{(b,cont)}$ to the backward propagated wave. Finally, when reaching the zeroth slice, one has calculated $\Phi_0^{(b)}$ to first order. Due to the matrix elements $\tilde{s}_{j-1}^{12}$, second order forward propagating elements occur; a forward propagation analogously to the zeroth order propagation can be started. This propagation cycle is sketched in fig.1.

Each respective propagation produces elements of the next order, so that the multiple backscattered contributions to $\Phi_N^{(f)}$ and $\Phi_0^{(b)}$ can be calculated similarly. A cutoff can be chosen freely by setting $\tilde{s}_{j-1}^{mn}(n \neq m)$ to 0 in the last desired propagation.

After having obtained a converged elastic wavefunction, the inelastic contributions of $\Phi_0^{(f)}$ to each $\Phi_n^{(f)}$ at every slice are computed. Now all information needed for the elastic propagations of $\Phi_n$ through $\mathbf{S}_{j-1}^{(i,n)}$ are gathered and the next propagation can be stared, where one can choose arbitrarily whether one starts with the backward or forward propagation. Both wavefunctions ($\Phi_N^{(f)}$ and $\Phi_N^{(b)}$) have the boundary that they must vanish at the respectively first slice of the propagation. In the first propagation cycle, one adds the inelastic contributions to the propagated part at each slice; in the following cycles, only the contributions obtained through the previous cycle are considered. When all wavefunctions have been obtained, the problem is solved.
Figure 1: Schematic view of the elastic propagation. Each row corresponds to one slice, each column to one backscattering event.

2.3 Implementation of the elastic Propagator

The matrix elements of the slice transition operator matrix in eq.34 all contain either $\tilde{F}_{j,j-1}$ or $\tilde{B}_{j,j-1}$ operators, which are connected to the backscattering of the electron beam, and the exponential $e^{2\pi i \tilde{k}_{j-1}^n}$ which resembles the elastic forward propagator without considering backscattering. The implementation of $\tilde{F}_{j,j-1}$ and $\tilde{B}_{j,j-1}$ are relatively straightforward once the inverse of $\tilde{k}_{j-1}^n$ has been found. We propose similar to eq.29 to approximate the inverse as

$$(\tilde{k}_{j-1}^n)^{-1} = \left( K_n \sqrt{1 + \frac{1}{K_n^2} \left( \frac{\Delta_b}{4\pi^2} + U_j(r) \right)} \right)^{-1} \approx \frac{1}{K_n} \sqrt{1 - \frac{1}{K_n^2} \left( \frac{\Delta_b}{4\pi^2} + U_j(r) \right)}$$

(37)

As the exponential, it has to be expanded before it can be implemented. The expansion of a square root $\sqrt{1+x}$ around $x=0$ simply reads

$$\sqrt{1+x} = \sum_{n=0}^{\infty} \frac{c_n}{n!} x^n$$

(38)
where
\[ \frac{c_n}{n!} = (-1)^{n-1} \frac{(2n-3)!}{2^{2n-2}n!(n-2)!} \] (39)

In this case, one has to substitute
\[ x \to -\frac{1}{K_n^2} \left( \frac{\Delta \beta}{4\pi^2} + U_j(r) \right) \]

For the original, not inverted, operator, only the sign of the substitution changes, the expansion remains the same.

Our approach to expand the exponential makes use of spherical Bessel functions of the first and second kind, \( j_n(z) \) and \( y_n(z) \), respectively

\[ \cos(\sqrt{z^2 - 2zt}) = \frac{\cos(z)}{z} + \sum_{n=1}^{\infty} \frac{t^n}{n!} j_{n-1}(z) \] (40)
\[ \sin(\sqrt{z^2 - 2zt}) = \frac{\sin(z)}{z} + \sum_{n=1}^{\infty} \frac{t^n}{n!} y_{n-1}(z) \] (41)

which can be expressed in terms of spherical Hankel functions:

\[ e^{iz\sqrt{z^2 - 2zt}} = \cos(\sqrt{z^2 - 2zt}) + i \sin(\sqrt{z^2 - 2zt}) \]
\[ = e^{iz} + \sum_{n=1}^{\infty} \frac{t^n}{n!} h_{n-1}^{(+)}(z) \] (42)

Expanding the Hankel functions as

\[ h_{n}^{(+)}(z) = e^{iz} \sum_{k=0}^{n} \frac{i^{n-k-1}(n+k)!}{2^k k!(n-k)!(n-k+1)!} \] (43)

leads to

\[ e^{iz\sqrt{1-\frac{2t}{z}}} = e^{iz} + \sum_{n=1}^{\infty} \frac{t^n}{n!} \sum_{k=0}^{n-1} e^{iz} \frac{i^{n-k-1}(n-1+k)!}{2^k k!(n-1-k)!(n-k+1)!} z^k \] (44)

Now one needs to identify \( z \) and \( t \) so that the exponential \( e^{2\pi i k_{j-1}} \) can be traced back to the equation above. This is achieved by setting

\[ z \to 2\pi K_0 \epsilon \]
\[ t \to -\epsilon \left( \frac{\Delta}{4\pi K_0} + \sigma U(r) \right) = -z \left( \frac{\Delta}{8\pi^2 K_0^2} + \frac{\sigma U(r)}{2\pi K_0} \right) \]
The expansion of the exponential thus reads

\[ e^{2\pi i K_0 \epsilon \sqrt{1 + \frac{\Delta}{4\pi^2 K_0^2} + \frac{z}{\pi K_0}}} = 1 + \sum_{n=1}^{\infty} \sum_{k=0}^{n-1} i^{k-n} (n - 1 + k)! \sum_{k=0}^{\infty} \left( \frac{\Delta}{8\pi^2 K_0^2} + \frac{\sigma U(r)}{2\pi K_0} \right)^n \]

This equation is quite practical since the coefficients \( \alpha_n \) can be determined beforehand and a recursive computation of higher order terms can be done. In practice, the phase factor of \( e^{2\pi i K_0 \epsilon} \) can be omitted.

### 3 Results

In this project, only single elastic backscattering is considered, i.e. \( \bar{s}_{12} = 0 \) in eq.34. The obtained results were compared to analogical computations using a conventional multislice algorithm (CMS) [5] and the approach of Cai et al. (RSMS) [6]. The system examined was a bcc-iron slab of varying thickness in beam direction and 16 x 16 unit cells in the two lateral directions. The energy of the incoming electron beam was chosen to be 200 kV. Each unit cell was divided into 20 slices each of them having a grid of 42 x 42 points.

The obtained wavefunctions of the forward traveling beam and their Fourier transforms are displayed in appendix A and B for different numbers of unit cells in beam direction (1, 5, 35 and 70). The plotted differences between the CMS wavefunctions and the other wavefunctions were obtained by subtracting the respective other wavefunction from the CMS function. Interestingly, RSMS and FEM predict different deviations from the multislice result: Where RSMS points towards stronger peaks, FEM indicates more diffuse peaks (smaller height with broader FWHM). It appears to be reasonable that the differences between the three methods are most pronounced around atomic columns as here the wavefunctions amplitude is largest. The smaller amplitude of the FEM wavefunction at these points can be associated with the likelihood of backscattering events which is only included in this method.

In fig.2 the backwards scattered wavefunctions for two different acceleration voltages are displayed. The choice of parameters from the left figure coincides with those from appendix A. A comparison of the amplitudes of backwards scattered wavefunction and the difference amplitude shows that these two amplitudes are indeed of the same order of magnitude strengthening the considerations made above. As
expected, the wavefunction with the higher acceleration voltage shows a significantly lower amount of backscattering.

Generally speaking, for an accurate description of backscattering phenomena a finer mesh of slices has to be used than the computation of the forward scattered wavefunction would require. In fig.3 the backward scattered wavefunctions of two identical calculations except for a doubled number of slices per unit cell in the right figure are displayed. Clearly, the computation with 10 slices per unit cell is not convergent. A glance at the Fourier transformed wavefunctions (fig.4) reveals that both calculated patterns show the same central structure. The non-converged calculation possesses in addition to this structure an intense pattern for large scattering angles. The occurrence of this pattern may be due to spurious self-interaction between adjacent super cells - an effect which has to be more pronounced for backscattering, where large scattering angles occur frequently.

At last, the prediction of the wavefunction’s squared amplitude by the CMS, RSMS and FEM method have been compared. The respective wavefunctions were sampled in the systems center, the point where the electron beam enters the crystal and where a row of atoms is underneath so that a comparatively large deviation between the three methods can be expected. The obtained amplitudes are shown in fig.5. It should be noted that the wavefunction’s relative amplitude (with respect to the CMS amplitude) fluctuates quite a bit. Fig.5 shows that the FEM amplitude is larger than 1 at a thickness of 25 unit cells, a comparison with the figures from appendix A shows that at 35 unit cells thickness the relative amplitude drops again below 1. It is not known whether the amplitude converges or can be described by some asymptotic function after a certain thickness.
Calculations including higher order backscattering showed a similar behaviour as non-converged computations with too few slices per unit cell - large angle scattering contributions diverged due to self-interaction. As a result, the general wavefunction, which sums over all orders of backscattering, was no longer normalised; a physical interpretation of the data could not be done.

4 Conclusion

In this report a general formalism to describe electron scattering in the multislice scheme has been proposed. Unfortunately, its wide applicability and high accuracy come at the cost of computationally very expensive calculations. In order to not exceed the framework of this project, only elastic scattering was implemented in a simulation software. The predicted wavefunctions show nonnegligible deviations from less sophisticated approaches mainly due to the inclusion of backscattering processes in the formalism.

It was found that the correct description of backscattering processes requires a finer mesh of potential slices than forward scattering needs. This rise in computational cost is unfortunate but has to be accepted if low beam electron energies or grazing beam techniques (RHEED) in TEM shall be described. Higher order backscattering could not be implemented successfully, the obtained wavefunctions diverged. It is suspected that this effect is due to self-interaction of adjacent supercells as it is likely to occur for high angle electron scattering. To test this hypothesis one should perform simulations including a larger super cell (with respect

Figure 3: Backscattered wavefunction of a sample with a thickness of 5 unit cells. The acceleration voltage was 200 kV. left: 10 slices per unit cell, right: 21 slices per unit cell.
Figure 4: Fourier transform of the backscattered wavefunction of a sample with a thickness of 5 unit cells. The acceleration voltage was 200 kV. left: 10 slices per unit cell, right: 21 slices per unit cell.

Figure 5: Squared amplitudes of the wavefunction (left) and the relative amplitudes with respect to the CMS amplitude (right).

to the two spatial dimensions orthogonal to the beam direction).

Besides the application of the code to more realistic experimental problems, future projects could include the theoretical examination of inclined illumination which is of relevance in some experimental techniques as mentioned before or a more detailed study of higher order backscattering. The most challenging extension of the current work would be the implementation of inelastic scattering combined with backscattering. For this purpose, suitable approximations of eq.22 have to be found, e.g., single inelastic scattering possibly combined with single backscattering or inelastic forward scattering approximation. In any case the resulting code will be able to provide the most accurate description of electron scattering among the theoretical methods available today.
A Wavefunctions of the forward scattered Beam

Figure 6: Real space wavefunctions of the electron beam after passing through a bcc-iron slab of various thicknesses (given in unit cells) as predicted by CMS and the difference of these wavefunctions to the corresponding wavefunctions predicted by RSMS and FEM.
B Fourier-Transformed Wavefunctions of the forward scattered Beam

Figure 7: Momentum space wavefunctions of the electron beam after passing through a bcc-iron slab of various thicknesses (given in unit cells) as predicted by CMS and the difference of these wavefunctions to the corresponding wavefunctions predicted by RSMS and FEM.
References


