Calculations of HRTEM and STEM images

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Calculations of HRTEM and STEM images and their comparison to experimental micrographs require:

1. Models for elastic and/or inelastic scattering of high energy electrons by crystals.
   - Phase object (and/or amplitude object).
   - Weak phase object.

2. Model for the microscope (Abbe image formation):
   - Illumination system (coherence).
     - Coherent illumination.
     - Partially coherent illumination (transmission cross-coefficients).
     - Incoherent illumination (STEM).
   - Image formation system (aberrations).
     - Spherical aberration (3, 5, ...).
     - Astigmatism, coma, ...

3. Model for the detector.
   - MTF of CCD camera (Modulation Transfer Function).
   - Annular detector.
And a computer program...

Mastering the theory is **mandatory** to perform meaningful simulations!
There are many excellent books on the dynamical electron scattering and image formation in electron microscopy. In particular:

1. Electron Microscopy of Thin Crystals by Hirsch et al. ([3]).
2. Diffraction physics by Cowley ([1]).
3. Transmission Electron Microscopy by Reimer ([6]).

and many excellent (and readable...) papers or reviews, in particular:

- P.G. Self, *Practical Computation of Amplitudes and Phases in Electron Diffraction* ([9]).
- K. Ishizuka, *A new theoretical and practical approach to the multislice method* ([5]).
- S. Pennycook, *Scanning Transmission Electron Microscopy* ([8]).
1. Scattering center.
2. Diffraction (scattering + interference).
3. Dynamical theory.
4. Image formation.
   ▶ Coherent illumination (HRTEM).
   ▶ Incoherent illumination (STEM).
Scattering center: atomic scattering amplitude

\[ s = \frac{\sin \theta}{\lambda} \]
2 Scattering centers

**Fig.** Two scatterers a large distance apart.

**Fig.** Two scatterers a small distance apart.
Single scattering: kinematical theory
Elastic scattering theory:

- Energy conservation ($\Delta E = 0$).
- Momentum transfer ($q \geq 0$).
- Scattering of high energy electrons (i.e. electrons accelerated by potential $\geq 50$ kV):
  - $\implies$ electrons scattered at small angles (few degrees).
- Strong electron-matter interaction ($10^4$ times larger X-rays):
  - $\implies$ very thin crystals only can be imaged.
  - $\implies$ Relaxed Bragg condition (reciprocal rod - many reflections).

$\vec{k}_i$: incident wavevector

$\vec{k}_g$: scattered wavevector

$\vec{g}$: reciprocal lattice vector

$\vec{s}_g$: deviation from exact Bragg condition

1. Energy conservation: $|\vec{k}_g| = |\vec{k}_i|$.

2. Momentum transfer: $\vec{k}_i + \vec{g} + \vec{s}_g = \vec{k}_g$. 

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Bragg law

With energy conservation and momentum transfer $= \vec{g}$ ($s_g = 0$):

$$|\vec{k}_i + \vec{g}| = |\vec{k}_g|$$

$$k_i^2 + 2 \times k_i \times g \times \cos(\vec{k}_i, \vec{g}) + g^2 = k_g^2$$

$$2k_i \times \cos(\vec{k}_i, \vec{g}) = -g$$

$$2k_i \times \cos(90^\circ - \theta_B) = -g$$

$$\frac{2}{\lambda} \times \sin(\theta_B) = g = \frac{1}{d_g}$$

$\implies$ Bragg law:

$$2 \times d_{hkl} \times \sin(\theta_B) = \lambda$$
Electron diffraction: Ewald sphere picture

- Ewald sphere: large R
- hkl node: relrod

Figure: Two beams at Bragg condition: $\vec{s}_g = 0$

Relrod $\rightarrow$ relaxed Bragg condition $\rightarrow$ many reflections
\[ \theta_B(400) = 15.87 \times 10^{-3} \text{rad} \approx 0.9 \text{ deg}. \]
Reflections intensity

1. Kinematical or single scattering approximation $\implies$ weak phase object.
2. Dynamical or multiple scattering approximation.

The structure factor gives the scattering strength of the $(h,k,l)$ planes.

$$F_{hkl} = \sum_{i=\text{atoms}} f_i(s_{hkl}) e^{(2\pi i (hx_i + ky_i + lz_i))} e^{-DW_i s_{hkl}^2 \text{Occ}_i}$$

where:

- $f_i(s_{hkl})$: atomic scattering amplitude.
- $(x_i, y_i, z_i)$: fractional coordinates of atom $i$ ($0 \leq x_i < 1$).
- $s_{hkl} = \frac{\sin(\theta_B)}{\lambda} = \frac{1}{2d_{hkl}}$.
- DW: Debye-Waller temperature factor (important for inelastic scattering).
- Occ: atom site occupancy.

Diffracted intensity (single scattering approximation): $I_{hkl} = |F_{hkl}|^2$

$\implies$ precise description of the crystal structure (ICSD, AMS, ...).
Multiple scattering (dynamical theory)
Dynamical theory?

Figure: \[ I_h = \sum_i I_{g_i} I_{g_i-h} (Vainstein[11]). \]

Figure: Dynamical SAED.
Dynamical theory of elastic scattering of high energy electron

We aim to understand in details multiple elastic scattering of electrons by crystals.

- High energy electron (eE).
- Periodic interaction potential $V(\vec{r})$.
- Time independent flux of incident electrons.

The fundamental equation of electron elastic scattering by a potential $V_v [\text{Volt}]$ (positive inside a crystal) in the approximation of a stationary flux of electrons of a given energy $eE$ is the Schrödinger equation ([4]):

$$\Delta \Phi(\vec{r}) + \frac{2me}{\hbar^2} [E + V_v(\vec{r})] \Phi(\vec{r}) = 0$$

With a change of notation its is written as:

$$[\Delta + 4\pi^2 K_i^2] \Phi(\vec{r}) = -4\pi^2 V_v(\vec{r}) \Phi(\vec{r})$$

Where the wavevector $|\vec{K}_i|$ of the incident electrons is given by:

$$|K_i| = \frac{\sqrt{2meE}}{\hbar}$$

and

$$m = \gamma m_0$$
**Constants**

\[ c = \text{speed of light in vacuum.} \]
\[ e = \text{electron charge.} \]
\[ E = \text{accelerating voltage (} \geq 50 \text{kV).} \]
\[ \gamma = 1 + \frac{e^2 E^2}{2m_0 c^2} \text{ (relativistic mass correction).} \]
\[ m_0 = \text{rest mass of the electron (} \approx 511 \text{ keV).} \]
\[ V = \frac{2m_e}{\hbar^2} V_v(x, y, z) \text{ (scattering potential [} nm^{-2} \text{]).} \]

<table>
<thead>
<tr>
<th>( E [kV] )</th>
<th>( \gamma )</th>
<th>( \lambda [pm] )</th>
<th>( \frac{\nu}{c} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>1.098</td>
<td>5.362</td>
<td>0.412</td>
</tr>
<tr>
<td>100</td>
<td>1.119</td>
<td>3.706</td>
<td>0.548</td>
</tr>
<tr>
<td>200</td>
<td>1.391</td>
<td>2.511</td>
<td>0.695</td>
</tr>
<tr>
<td>500</td>
<td>1.978</td>
<td>1.423</td>
<td>0.862</td>
</tr>
<tr>
<td>1000</td>
<td>2.957</td>
<td>0.873</td>
<td>0.941</td>
</tr>
</tbody>
</table>

*Relativistic mass correction, wavelength and speed of the electron*
At 100 \( kV \), Bragg angles for several reflections of Al are given in the next table.

<table>
<thead>
<tr>
<th>(hkl)</th>
<th>Bragg angle [mrad]</th>
<th>Bragg angle [deg.]</th>
<th>(hkl) spacing nm(^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1,1,1)</td>
<td>7.91</td>
<td>0.453</td>
<td>4.276</td>
</tr>
<tr>
<td>(2,0,0)</td>
<td>9.14</td>
<td>0.523</td>
<td>4.938</td>
</tr>
<tr>
<td>(2,2,0)</td>
<td>12.92</td>
<td>0.740</td>
<td>6.983</td>
</tr>
<tr>
<td>(1,1,3)</td>
<td>15.15</td>
<td>0.868</td>
<td>8.189</td>
</tr>
<tr>
<td>(2,2,2)</td>
<td>15.83</td>
<td>0.906</td>
<td>8.553</td>
</tr>
<tr>
<td>(4,0,0)</td>
<td>18.28</td>
<td>1.047</td>
<td>9.876</td>
</tr>
</tbody>
</table>

**Bragg angles for some Al reflections at 100 \( kV \).**

Notice that the Bragg angles are pretty small (of the order a few \(^{\circ}\)) and that consequently the small angle approximation is quite good.
Vacuum solution: plane wave

When scattering potential = 0 (vacuum) $\implies$ solution of fundamental equation:

plane wave that could be defined either as:

$$\Phi(\vec{r}) = e^{i\vec{K}_v \cdot \vec{r}}$$

or as:

$$\Phi(\vec{r}) = e^{-i\vec{K}_v \cdot \vec{r}}$$

We will use $\Phi(\vec{r}) = e^{i\vec{K}_v \cdot \vec{r}}$ since it is compatible with the $O_{xyz}$ basis selected in jems where the wavevector $\vec{K}_v$ is anti-parallel to $O_z$.

This choice makes the phase of the wave increase with the direction of propagation.

Notice that the wave-vector $\vec{K}_v$ is perpendicular to the wave-front.
C: center of the Ewald sphere and CLC: C projection onto the zero order Laue zone (Center of Laue Circle).
Diffraction geometry: definition

- Electron microscope favors $O_z$ direction $\implies$ i.e. destroys the space isotropy.
- 3-D space described a coordinate system $(\rho; z)$ where $\rho = \{x, y\}$ coordinates of on plane $O_{xy}$ perpendicular to $O_z$.
- $O_z$ anti-parallel to optical axis of electron microscope.
- Incident electron beam $\vec{K}_i$ anti-parallel to $O_z$ axis and parallel optical axis.
- Crystal zone axis $\vec{u} = [u, v, w]$ parallel or nearly parallel to $O_z$.
- High order Laue zone reflections $\vec{g} = (h, k, l)$ satisfy $\vec{g} \cdot \vec{u} = h u + k v + l w = 1, 2, 3, \ldots$.
- Laue circle $\implies$ intersection of Ewald sphere and ZOLZ (zero order Laue zone, i.e. Laue zone containing the origin $O$ of reciprocal space).
- Excitation error $\implies$ vector from reciprocal node $G$ to Ewald sphere parallel to the normal $\vec{n}$ of the entrance surface of the specimen.
- $z$-component of $\vec{K}_i (k_z)$ very large compared to its $O_{xy}$-component ($\chi$) (i.e. 400 nm$^{-1}$ at 200 kV accelerating voltage).

Electron wave oscillates with very high frequency in $O_z$ direction.
Schrödinger equation

The Laplacian $\Delta = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}$ is written as: $\Delta \rho + \frac{\partial^2}{\partial z^2}$. As a result, $[\Delta + \ldots] e^{2\pi ik_z z} \Psi(\rho; z)$ is given by: $[\Delta \rho + \frac{\partial^2}{\partial z^2} + \ldots] e^{2\pi ik_z z} \Psi(\rho; z)$.

Performing the $z$-differentiation:

$$\frac{\partial^2}{\partial z^2} e^{2\pi ik_z z} \Psi(\rho; z) = e^{2\pi ik_z z} \left[ -4\pi^2 k_z^2 + 4\pi ik_z \frac{\partial}{\partial z} + \frac{\partial^2}{\partial z^2} \right] \Psi(\rho; z)$$

Inserting the last expression and dropping the term $e^{2\pi ik_z z}$:

$$[\Delta \rho + 4\pi^2 (K_i^2 - k_z^2 + V(\rho; z)) + 4\pi ik_z \frac{\partial}{\partial z} + \frac{\partial^2}{\partial z^2}] \Psi(\rho; z) = 0$$

Since $K_i^2 = k_z^2 + \chi^2$:

$$[\Delta \rho + 4\pi^2 \chi^2 + 4\pi^2 V(\rho; z) + 4\pi ik_z \frac{\partial}{\partial z} + \frac{\partial^2}{\partial z^2}] \Psi(\rho; z) = 0$$

Rearranging the last equation:

$$\frac{\partial \Psi(\rho; z)}{\partial z} = -\frac{1}{4\pi k_z} [\Delta \rho + 4\pi^2 \chi^2 + 4\pi^2 V(\rho; z) + \frac{\partial^2}{\partial z^2}] \Psi(\rho; z)$$
The term $|\frac{\partial^2 \Psi(\rho; z)}{\partial z^2}|$ being much smaller than $|k_z \frac{\partial \Psi(\rho; z)}{\partial z}|$ we drop it (this is equivalent to neglect backscattering).

Fundamental equation of elastic scattering of high energy mono-kinetic electrons with a potential within the approximation of small angle scattering:

$$i \frac{\partial \Psi(\rho; z)}{\partial z} = -\frac{1}{4\pi k_z} \left[ \triangle \rho + 4\pi^2 \chi^2 + 4\pi^2 V(\rho; z) + \frac{\partial^2}{\partial z^2} \right] \Psi(\rho; z)$$
The approximations of the fundamental equation are equivalent to assume that the scattering potential is small compared to the accelerating potential and that $k_z$ varies only slightly with $z$. It is in fact a quite good approximation, since the mean crystal potential is of the order of $10^{-20}\text{V}$.

Electron backscattering is neglected, the electron are moving forwards.

The fundamental equation is actually equivalent to a 2-dimensional Schrödinger equation ($\rho = \{x, y\}$) where $z$ plays the role of time. The system evolution is causal, from the past to the future.

Fundamental equation in Hamiltonian form:

$$i\frac{\partial}{\partial z} \Psi = H \psi$$

where:

$$H = -\frac{1}{4\pi k_z} [\Delta \rho + 4\pi^2 \chi^2 + 4\pi^2 V(\rho; z)] = H_0 + \frac{4\pi^2 V(\rho; z)}{4\pi k_z}$$

A fundamental postulate of quantum mechanics ([7, 10]) says that the evolution operator obeys the equation:

$$i\frac{\partial}{\partial z} U(z, 0) = H(\rho; z) U(z, 0)$$
Causal evolution operator

$U(z, 0)$: unitary operator (the norm of $|\Psi>$ is conserved), in general not directly integrable $\implies$ approximations.

$U(z, 0)$ can be directly integrated only when $H(\rho; z)$ and $\frac{\partial}{\partial z} H(\rho; z)$ commute. In that case the general solution is [7]:

$$U(z, 0) = e^{-i \int_0^z H(\tau) d\tau}$$

$H(\rho; z)$ and $\frac{\partial}{\partial z} H(\rho; z)$ commute when:

- $V(\rho; z)$ does not depend on $z$, i.e. $V(\rho; z) = V(\rho)$ (perfect crystal).
- $V(\rho; z)$ can be neglected (free space propagation).
- $H(\rho; z)$ is approximated by its potential term (phase object).

Three approximations will be considered:

- Multislice method.
- Bloch wave method.
- Howie-Whelan column approximation.
Diffraction and imaging
The transition probability from initial state $|\chi>\rightarrow q(z, 0)$ to final state $|q>\rightarrow q(z, 0)$. 

$$\omega_{\chi \rightarrow q(z, 0)} = |<q|\hat{U}(z, 0)|\chi>|^2$$

To calculate the intensity or transition probability $\omega_{\chi \rightarrow q(z, 0)}$ we must calculate $\hat{U}(z, 0)$.
Diffraction: transition probability

**Final state** \( |\Psi_z > \) of electron after interaction with crystal potential:

\[
|\Psi_z > = \hat{U}(z, 0) |\Psi_0 > = \hat{U}(z, 0) \hat{T} |\Psi_0 > = \sum_{q'} \hat{U}(z, 0) |q' > < q'|\Psi_0 >
\]

**Complex amplitude** of beam diffracted in direction \( |q > \) (dropping \( \sum_{q'} \) since each time \( |q' > < q'| \) appears it implies a summation):

\[
< q|\Psi_z > = < q|\hat{U}(z, 0) |q' > < q'|\Psi_0 >
\]

\[
< q|\Psi_z > = < q|\hat{U}(z, 0) |\chi >
\]

**Transition probability** from the initial state \( |\chi > \) to the final state \( |q > \), \( \omega_{\chi \rightarrow q}(z, 0) \), is the module square of \( < q|\hat{U}(z, 0) |\chi > \) (intensity of beam diffracted to \( |q > \)).

**Intensity diffracted to** \( |q > \):

\[
\omega_{\chi \rightarrow q}(z, 0) = | < q|\hat{U}(z, 0) |\chi > |^2
\]
Intensity of the wave function

Wave function intensity $\Psi(\rho; z)$ at exit face of crystal slab $\implies$ transition probability from initial state $|\chi\rangle$ to final state $|\rho\rangle$:

$$\omega_o \rightarrow \rho(z, 0) = | \langle \rho | \hat{U}(z, 0) | \chi \rangle |^2$$

With a discrete set of $|q\rangle$:

$$\omega_o \rightarrow \rho(z, 0) = | \sum_q < \rho | q \rangle < q | \hat{U}(z, 0) | \chi \rangle |^2$$

Where:

- $< q | \hat{U}(z, 0) | \chi \rangle$ Fourier transform of object wave function.
- $\sum_q < \rho | q \rangle < q | \hat{U}(z, 0) | \chi \rangle$ the Fourier synthesis of the image wave function.
Intensity in image plane

Intensity observed at point $\rho$ in the image plane is modified by the transfer function of the microscope $T(q', q)$ that couples states $|q'\rangle$ and $|q\rangle$ (matrix $T(q', q)$).

Image intensity is given by:

$$\omega_o \rightarrow \rho(z, 0) = \sum_q \sum_{q'} <\rho|q'\rangle <q'|T(q', q)|q\rangle <q|\hat{U}(z, 0)|\chi\rangle^2$$

Fourier components of the wave function in image plane of objective lens are modified by $< q'|T(q', q)|q\rangle$ (Abbe theory of image formation).
Abbe image formation: transfer function

The terms \( \sum_{q'} < \rho | q' > \) show that the wave function is obtained by inverse Fourier transform (Fourier synthesis).

In back focal plane of objective lens Fourier components of image wave function:

\[
< q' | T(q', q) | q > < q | \hat{U}(z, 0) | \chi >
\]

The microscope introduces a coupling between the diffracted beams when transferred by the objective lens.

When transfer is linear (for example in WPOA (weak phase object approximation) the transfer matrix \( < q' | T(q', q) | q > \) is diagonal:

\[
< q' | T(q', q) | q > = T(q) \delta(q' - q)
\]

In this particularly simple case the image intensity is:

\[
\omega_o \rightarrow \rho(z, 0) = | \sum_q < \rho | q > T(q) < q | \hat{U}(z, 0) | \chi > |^2
\]
Finally, let us remark that the symmetry properties of the diffraction patterns and high resolution images are intrinsically tight to the properties of the transition probabilities $\omega_{o} \rightarrow \rho(z,0)$ and $\omega_{\chi} \rightarrow q(z,0)$.

Evolution operator $\hat{U} \implies$ key to understand not only symmetries, but also contrast of bright and dark field images of defects.

This systematic approach due to D. Gratias and R. Portier ([2]) unifies all the different methods of image simulation in transmission electron microscopy.
Approximations

$H(z) = \frac{1}{2K} (\nabla^2 - \chi^2 + V(z)) = H^0 + \frac{V(z)}{2K}$

**z-independent potential: $\hat{V}$**

Zero-layer approximation
or
Projected-potential approximation

**Plane-wave expansion**
- Diffraction matrix
  - Darwin (1914)
  - Sturkey (1962)
  - Tournarie (1960, 1961, 1962)
  - Howie–Whelan (1961)

**Bloch-wave expansion**
- Secular equation
  - Bethe (1928)
  - Kambe (1967)
  - Howie–Whelan (1961)
- Fresnel propagator
  - Goodman–Moodie (1974)

**Multislice method**

**z-dependent potential**

**Perturbation expansion**
- Plane waves
- Bloch waves
- Sudden perturbation

**Physical optics**
- Cowley–Moodie
  - First order
  - Kinematical theory

**Interband transition**
- Buxton (1976, 1979)
  - First order
  - Single interband transition

**Phase object**
- First order
  - Weak phase object

**Figure:** From Gratias and Portier [2]
Dynamical theory: multislice method
Causal evolution equation

Hamiltonian $H = H(\rho; z)$ is $z$ dependent (through the potential term $V(\rho; z)$). When $V$ does not depend on $z$, a causal evolution operator $U(z, 0)$ is defined as:

$$\Psi(\rho; z) = U(z, 0) \Psi(\rho; 0) \text{ or } |\Psi_z > = \hat{U}(z, 0)|\Psi_0 >$$

The evolution from $z$ to $z$ (no evolution!) is the identity operator $\hat{I}$:

$$U(z, z) = \hat{I}$$

Assuming that the scattering potential $V(\rho; z)$ does not depend on $z$ is equivalent to make the hypothesis that the crystal can be replaced by a series of thin slices of constant $V'(\rho)$ given by:

$$V'(\rho) = \frac{1}{\tau} \int_{z}^{z+\tau} V(\rho; z')dz'$$

$V'(\rho)$ is the projected potential of the slice of thickness $\tau$. The causal evolution operator for the whole crystal is then written as:

$$U(z_n; 0) = U(z_n; z_{n-1}) U(z_{n-1}; z_{n-2}) ... U(z; 0)$$

where each thin slice is characterized by its own particular evolution operator.
The multislice method is heavily used when the unit cell of the crystal is large or contains defects. It can be used to simulate:

- **Amorphous** material (once a model is available!).
- CBED patterns and coherent nano-probe diffraction patterns.
- HAADF (high resolution STEM) images (frozen phonon).

It has severe limitations for the simulation of:

- LACBED patterns.
- SAED patterns and HRTEM images of large [uvw] zone axis indices.

In the multislice method the crystal is **cut** into thin slices and their potential projected on a plane. The wave function is **propagated** from slice to slice:

- At the projection plane the hamiltonian reduces to the **diffractor**.
- In between the planes it reduces to the **propagator**.

The **diffractor** takes into account the scattering events and involves a simple multiplication of the incident wave function by the phase object. The propagation is a convolution (best performed in Fourier space).
Multislice: slicing the crystal

**Figure:** Crystal is sliced and the potential of the slices is projected on a plane.
Figure: Wave function propagates through the stack of slices.
**Pt catalyst on amorphous carbon**

**Figure:** Pt catalyst on amorphous carbon.

**Figure:** HRTEM image simulation.
Pt octahedron: slice

**Figure:** Typical slice: unit cell with atoms at \((x, y, z)\) (\(0 \leq x, y, z < 1\)).

**Figure:** Propagator (real part, Fourier transform).
Phase object of a typical slice

**Figure:** Phase object (real part).

**Figure:** Phase object (imaginary part).
Dynamical theory: Bloch wave method
Electrons inside the crystal propagate as eigenstates of the interaction Hamiltonian.

**Figure:** Bloch wave 1 (maximum on the atomic column: "state 1s").

**Figure:** Bloch wave 2 (maximum between the atomic column).
The Bloch wave method is very versatile and is used to simulate HREM, SAED, CBED, LACBED images of small to medium unit cell crystals.

It provides a strong physical basis of how the electron propagates inside periodic crystals \(\rightarrow\) channeling.

It is a the basis of the quantitative CBED technique developed by Zuo and Spence.

It offers simple ways to simulate dynamical SAED, precession and hollow cone patterns.
The Bloch wave method provides explanation of electron channeling (here GaAs [110]).
**Figure:** Web site EM centre - Monash university (J. Etheridge).

**Figure:** Simulation using Bloch wave method.
The Howie-Whelan method is principally used to simulate bright-dark field, CBED or LACBED images of extended crystal defects like dislocations, stacking faults, precipitates.

It is the basis of the weak beam image simulation.
TEM and STEM image formation

1. Ray optics (remainder).
2. Perfect optical system.
3. Aberrations (spherical).
4. Properties of optical systems:
   - Linearity.
   - Space invariance.
5. Transfer function (coherent or partially coherent illumination - HRTEM).
6. Optical transfer function (incoherent illumination).
In the back focal plane of the lens T & D beams converge to points (secondary spherical sources).

Why is the objective lens so most important lens of the TEM?
$P_E$ entrance and $P_S$ exit pupils.

$O$ (object) and $I$ (image) are conjugate points.
Wavefront is deformed \( \rightarrow \) inclined incidents rays are not focused at same point.

Coefficient of spherical aberration \( C_s \) is close to the focal length \( f \) of the objective lens.
Wavefront aberrations to 6th order in cartesian coordinates

\[
\begin{align*}
\{ z, \pi (u^2 + v^2) \lambda \} (\text{defocus}) & \\
W(1, 1), 2\pi (u \cos(\phi(1, 1)) + v \sin(\phi(1, 1))) & \\
W(2, 2), \pi \lambda ((u - v)(u + v) \cos(2\phi(2, 2)) + 2uv \sin(2\phi(2, 2))) & \\
W(3, 1), \frac{2}{3} \pi (u^2 + v^2) \lambda^2 (u \cos(\phi(3, 1)) + v \sin(\phi(3, 1))) & \\
W(3, 3), \frac{2}{3} \pi \lambda^2 (u (u^2 - 3v^2) \cos(3\phi(3, 3)) - v (v^2 - 3u^2) \sin(3\phi(3, 3))) & \\
W(4, 0), \frac{1}{2} \pi (u^2 + v^2)^2 \lambda^3 (\text{spherical aberration}) & \\
W(4, 2), \frac{1}{2} \pi (u^2 + v^2) \lambda^3 ((u - v)(u + v) \cos(2\phi(4, 2)) + 2uv \sin(2\phi(4, 2))) & \\
W(4, 4), \frac{1}{2} \pi \lambda^3 ((u^4 - 6v^2u^2 + v^4) \cos(4\phi(4, 4)) + 4u(u - v)v(u + v) \sin(4\phi(4, 4))) & \\
W(5, 1), \frac{2}{5} \pi (u^2 + v^2)^2 \lambda^4 (u \cos(\phi(5, 1)) + v \sin(\phi(5, 1))) & \\
W(5, 3), \frac{2}{5} \pi (u^2 + v^2)^2 \lambda^4 (u (u^2 - 3v^2) \cos(3\phi(5, 3)) - v (v^2 - 3u^2) \sin(3\phi(5, 3))) & \\
W(5, 5), \frac{2}{5} \pi \lambda^4 (u (u^4 - 10v^2u^2 + 5v^4) \cos(5\phi(5, 5)) + v (5u^4 - 10v^2u^2 + v^4) \sin(5\phi(5, 5))) & \\
W(6, 0), \frac{1}{3} \pi (u^2 + v^2)^3 \lambda^5 & \\
W(6, 2), \frac{1}{3} \pi (u^2 + v^2)^2 \lambda^5 ((u - v)(u + v) \cos(2\phi(6, 2)) + 2uv \sin(2\phi(6, 2))) & \\
W(6, 4), \frac{1}{3} \pi \lambda^5 ((u^6 - 5v^2u^4 - 5v^4u^2 + v^6) \cos(4\phi(6, 4)) + 4uv (u^4 - v^4) \sin(4\phi(6, 4))) & \\
W(6, 6), \frac{1}{3} \pi \lambda^5 ((u^6 - 15v^2u^4 + 15v^4u^2 - v^6) \cos(6\phi(6, 6)) + 2uv (3u^4 - 10v^2u^2 + 3v^4) \sin(6\phi(6, 6))) & \\
\end{align*}
\]
When the optical system has these two important properties:

1. Linearity.
2. Space invariance.

it can be globally characterized by a transfer function $\tilde{T}(\vec{k})$. 
Linearity

\[ S\{a_1\Psi_1^o(\vec{x}) + a_2\Psi_2^o(\vec{x})\} = a_1S\{\Psi_1^o(\vec{x})\} + a_2S\{\Psi_2^o(\vec{x})\} \]
\[ S\{a_1\Psi_1^o(\vec{x}) + a_2\Psi_2^o(\vec{x})\} = a_1\Psi_1^i(\vec{x}) + a_2\Psi_2^i(\vec{x}) \]

Allows to:

- decompose the object wavefunction into points sources.
- consider only the transfer by the optical system of a point source (impulse).

\[ \Psi_o(\vec{x}) = \int_{-\infty}^{\infty} \Psi_o(\vec{u})\delta(\vec{x} - \vec{u})d\vec{u} \]
Impulse response

Image wavefunction $\Psi_i(\vec{x})$ is:

$$\Psi_i(\vec{x}) = S \left\{ \int_{-\infty}^{\infty} \Psi_o(\vec{u}) \delta(\vec{x} - \vec{u}) d\vec{u} \right\}$$

By linearity property ($\Psi_o(\vec{x}) \rightarrow$ coefficients real or complex):

$$\Psi_i(\vec{x}) = \int_{-\infty}^{\infty} \Psi_o(\vec{u}) S\{\delta(\vec{x} - \vec{u})\} d\vec{u}$$

Optical system *impulse response* $t(\vec{x}; \vec{u})$:

$$t(\vec{x}; \vec{u}) = S\{\delta(\vec{x} - \vec{u})\}$$

By linearity one has to consider only how the optical system transfer a spherical wave (point source).
Space invariance: point spread function

\[ t(\vec{x}; \vec{u}) = t(\vec{x} - \vec{u}) \]

Transfer is a convolution integral of \( \Psi_o(\vec{u}) \) and the point spread function \( t(\vec{x}) \):

\[ \Psi_i(\vec{x}) = \int_{-\infty}^{\infty} \Psi_o(\vec{u}) t(\vec{x} - \vec{u}) d\vec{u} = \Psi_o(\vec{x}) \otimes t(\vec{x}) \]

In Fourier space (or reciprocal space):

\[ \tilde{\Psi}_i(\vec{k}) = \tilde{\Psi}_o(\vec{k}) \tilde{T}(\vec{k}) \]

\[ \tilde{T}(\vec{k}) = e^{-2\pi i \left(\frac{C_s \lambda^3 k^4}{4} - \frac{\Delta f \lambda k^2}{2}\right)} \]

Electrical systems have the property of time invariance.
Abbe image formation: transfer function

\[ z \]

\[ f \]

\[ S_1 \]

\[ S_{-1} \]

Objet

Plan focal image

Diffusion & Interference

\[ \Psi_0(x) \]

\[ \tilde{\Psi}_0(u) \tilde{T}(u) \]

Remember \( \frac{\pi}{2} \) phase shift of the diffracted beams!
Image formation

- **Coherent illumination:**
  \[ \Phi_i(\vec{x}) = \Phi_o(\vec{x}) \otimes PSF(\vec{x}) \]

- **Partially coherent illumination:**
  \[ \tilde{I}(\vec{h}; z) = \int \tilde{T}_{cc}(\vec{h}'; + \vec{h}; \vec{h}; z) \phi(\vec{h}' + \vec{h})\phi^*(\vec{h}') d\vec{h}' \]

- **Incoherent illumination:**
  \[ I(\vec{x}) = |\Phi_o(\vec{x})|^2 \otimes [PSF(\vec{x})PSF^*(-\vec{x})] \]

where \( PSF(\vec{x}) = FT^{-1} \left[ \tilde{T}(\vec{h}) \right] \)
Detectors: CCD cameras

Figure: Comparison of the MTF of 2 CCD cameras. Note that the intensity of high spatial frequencies is severely attenuated.

Given a particular crystal, the contrast of the observed HRTEM micrographs depends on the TEM magnification. For quantitative work always record HRTEM micrographs at the highest possible magnification or introduce the detector MTF in the simulations.
MTF & Si (220) lattice planes

Figure: Si (220) spatial frequencies at low and high magnification (i.e. diffracted beams).
Effect of CCD MTF on simulated images: high magnification

Figure: Taking into account the MTF of the CCD camera (left: CCD MTF not included, right: CCD MTF included).
Effect of CCD MTF on simulated images: low magnification

Figure: Taking into account the MTF of the CCD camera (left: CCD MTF not included, right: CCD MTF included).
To conclude...

- **Learn:**
  2. Diffraction physics.
  3. Image formation.
  4. ...

- **Characterize** entirely your microscope (include the acquisition system).

- **Be critical** ⟷ always verify the results....
Notation for crystal space states: $\text{rep}\{\rho\}$

In crystal space $\rho = \{x, y\}$ represents the position (better notation would be $\vec{\rho}$ instead of $\rho$, but this would make notation cumbersome (context of equations clearly determines scalar or vectorial nature of objects).

Eigenvectors of position operator $\hat{\rho}$ are the $|\rho\rangle$. The $|\rho\rangle$ form a basis of representation $(\text{rep}\{\rho\})$ of $\hat{\rho}$:

$$<\rho'|\rho> = \delta(\rho - \rho')$$

$\text{basis}\{\rho\}$ is complete and has the closure relationship:

$$\int_{-\infty}^{\infty} d^2\rho \, |\rho\rangle <\rho| = \hat{I}$$

In this representation the wave function of the electron is:

$$\Psi(\rho) = <\rho|\Psi>$$

In the $\text{rep}\{\rho\}$ some operators are diagonal ($\hat{V}(\rho; z)$ is diagonal), some are not (for example $\Delta_{\rho}$).
Natural representation of momentum space given by plane waves $\text{rep}\{q\}$. The $\text{basis}\{q\}$ has the usual properties:

$$< q' | q > = \delta(q - q')$$

$$\int_{-\infty}^{\infty} d^2q \ |q > < q| = \hat{I}$$
Direct and momentum space: dual spaces

In their dual space $|\rho\rangle$ et $|q\rangle$ are written as:

$$
<\rho|q> = e^{i\rho\cdot q}
$$

$$
<q|\rho> = e^{-i\rho\cdot q}
$$

In the $\text{rep}\{q\}$ (plane waves), $<q|\Psi>$, is written using the closure relationship as:

$$
<q|\Psi> = \int_{-\infty}^{\infty} d^2\rho \ <q|\rho><\rho|\Psi>
$$

$$
<q|\Psi> = \int_{-\infty}^{\infty} d^2\rho \ e^{-i\rho\cdot q} \ \Psi(\rho)
$$

$\implies$ Fourier transform.

In the $\text{rep}\{\rho\}$:

$$
<\rho|\Psi> = \int_{-\infty}^{\infty} d^2\rho \ <\rho|q><q|\Psi> = \int_{-\infty}^{\infty} d^2\rho \ e^{i(\rho\cdot q)} \ \Psi(q)
$$

$\implies$ inverse Fourier transform!
Why $\text{rep}\{\rho\}$ and $\text{rep}\{q\}$

$\text{rep}\{\rho\}$ and $\text{rep}\{q\}$ are linked by Fourier transforms.

$\implies \text{rep}\{\rho\}$ used to calculate high resolution images.

$\implies \text{rep}\{q\}$ used to calculate diffraction patterns.

Initial state of incident electron $\Psi(\rho; 0)$ given:

in $\text{rep}\{\rho\}$ by $\Psi(\rho; 0) = <\rho|\Psi_0>$.

in $\text{rep}\{q\}$ by $|\chi>$

$$|\chi> = \int_{-\infty}^{\infty} d^2\rho ~ <q|\rho><\rho|\Psi_0> = \int_{-\infty}^{\infty} d^2\rho ~ e^{-i\rho\cdot q} \Psi(\rho, 0)$$

When convergent beam electron diffraction patterns are calculated, the incident conical illumination is decomposed in a set of plane waves $|\chi>$ and an independent dynamical calculation performed for each $|\chi>$.

Similarly, the final state at thickness $z$, after scattering by the potential during time $z$ is given by $\Psi(\rho; z) = <\rho|\Psi_z>$. 
Hermitic or self-adjoint operators can be decomposed on a basis formed by their eigenvectors (spectral decomposition). These operators have real eigenvalues and correspond to measurable physical properties. In our case, the eigenvalues of the hamiltonian operator $\hat{H}$ correspond to the total energy of the system, i.e. kinetic energy + potential energy. In the basis of its eigenvectors $|j>$ with corresponding eigenvalue $\gamma^{(j)}$, $\hat{H}$ is decomposed as:

$$\hat{H} = \sum_j \gamma^{(j)} |j><j| = \sum_j \gamma^{(j)} \hat{P}_j$$

where the $\hat{P}_j = |j><j|$ are projection operators ($\hat{P}_j^2 = \hat{P}_j$). Finally, using the spectral decomposition, any function of a self-adjoint operator (for example $\hat{H}$) can be written as:

$$f(\hat{H}) = \sum_j f(\gamma^{(j)}) |j><j|$$

(This follows by Taylor series development of $f()$).
In $\mathbb{R}^3$, $|1> = (1, 0, 0)$, $|2> = (0, 1, 0)$ and $|3> = (0, 0, 1)$.

$\hat{I} = \hat{P}_1 + \hat{P}_2 + \hat{P}_3$ where:

$\hat{P}_1 = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} \cdot (1 \ 0 \ 0) = \begin{pmatrix} 1 \ 0 \ 0 \\ 0 \ 0 \ 0 \\ 0 \ 0 \ 0 \end{pmatrix}$

$\hat{P}_2 = \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} \cdot (0 \ 1 \ 0) = \begin{pmatrix} 0 \ 0 \ 0 \\ 0 \ 1 \ 0 \\ 0 \ 0 \ 0 \end{pmatrix}$

$\hat{P}_3 = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} \cdot (0 \ 0 \ 1) = \begin{pmatrix} 0 \ 0 \ 0 \\ 0 \ 0 \ 0 \\ 0 \ 0 \ 1 \end{pmatrix}$

It is easy to verify that $\hat{P}_1^n = \hat{P}_1$, ...
Multislice: phase object or diffractor

The hamiltonian $\hat{H}$ reduced to the potential term is not diagonal on the \textit{basis} $\{q\}$, but is diagonal on the position vector \textit{basis} $\{\rho\}$.

The eigenvalue associated to position $\rho$ is $V(\rho, z)$:

$$\hat{H}^d |\rho > = \frac{1}{2k_z} \hat{V}(xy, z) |\rho > = \frac{1}{2k_z} \hat{V}(\rho, z) |\rho >$$

The corresponding evolution operator, diffractor, is:

$$\hat{U}^d (z, 0) = e^{\frac{-i}{2k_z} \int_0^z d\tau \hat{V}(\rho, \tau)} = \int d^2 \rho \ e^{\frac{-i}{2k_z} \int_0^z d\tau \hat{V}(\rho, \tau)} |\rho > < \rho |$$

The wavefunction $|\Psi(z) >$ at thickness $z$ is:

$$|\Psi(z) > = \hat{U}^d (z, 0) |\chi > = e^{\frac{-i}{2k_z} \int_0^z d\tau \hat{V}(\rho, \tau)} |\chi >$$

The wave function is the incident wave function multiplied by the phase factor $e^{\frac{-i}{2k_z} \int_0^z d\tau V(\rho, \tau)}$, only function of $\rho$ where $\int_0^z d\tau V(\rho, \tau)$ is the projected potential $V_p(\rho)$. 

Pierre Stadelmann CIME-EPFL CH-1015 Lausanne Switzerland Pierre.Stadelmann@epfl.ch Calculations of HRTEM and STEM images
When the projected potential is very weak the phase can be developed to the first order (weak phase object approximation):

$$|\Psi(z)\rangle = U^d(z,0)|\chi\rangle = \left[1 - \frac{i}{2k_z}V_p(\rho)\right]|\chi\rangle$$

The diffractor being diagonal in $\text{rep}\{\rho\}$ means that the transition probability from $|\rho_1\rangle$ to $|\rho_2\rangle$ is null except when $|\rho_1\rangle = |\rho_2\rangle$ (infinitely thin columns).
Multislice: free space propagation (Fresnel propagator)

Neglecting the potential term \( \Rightarrow \) free space propagation:

\[
\hat{H}^p = \frac{1}{2k_z} (-\Delta_{xy} - \chi^2)
\]

It is diagonal on the basis \( \{q\} \):

\[
\hat{H}^p |q> = \frac{1}{2k_z} (-\Delta_{xy} - \chi^2) |q> = \frac{q^2 - \chi^2}{2k_z} |q>
\]

or:

\[
\hat{H}^p = \int d^2q \frac{q^2 - \chi^2}{2k_z} |q><q|
\]

The eigenvalue is just \(-2\pi s_g\), \( s_g \) being the classical excitation error to the exact Bragg condition for the reciprocal lattice node \( G \) corresponding to \( |q> \).

The free space evolution operator \( \hat{U}^p(z, 0) \) is diagonal on this basis and is written:

\[
\hat{U}^p(z, 0) = e^{-i\hat{H}p z} = \int d^2q e^{-i \frac{q^2 - \chi^2}{2k_z} z} |q><q|
\]

Its action on the plane wave \( |q> \) is:

\[
\hat{U}^p(z, 0)|q> = e^{-i \frac{q^2 - \chi^2}{2k_z} z} |q>
\]

In Fourier space, Fresnel propagation is a simple product that changes the phase of the plane wave.
Multislice: Fresnel propagator in $\text{rep}\{\rho\}$

In $\text{rep}\{\rho\}$ the propagation between the point $\rho_1$ on the entrance surface of the crystal ($z = 0$) towards the point $\rho_2$ on the exit surface ($z$) is:

$$< \rho_2 | \hat{U}^p(z, 0) | \rho_1 > = \int d^2q \ e^{-i \frac{q^2 - \chi^2}{2kz}} < \rho_2 | q > < q | \rho_1 >$$

Since:

$$< \rho_2 | q > = \int d^2q \ \delta(\rho - \rho_2) e^{i q \cdot \rho}$$

and

$$< q | \rho_1 > = \int d^2q \ \delta(\rho - \rho_1) e^{-i q \cdot \rho}$$

The evolution operator $\hat{U}^p(z, 0)$ in the $\text{rep}\{\rho\}$ is:

$$< \rho_2 | \hat{U}^p(z, 0) | \rho_1 >= C \ \frac{ie^{ikz}}{2\pi z} e^{i k_z \frac{(\rho_2 - \rho_1)^2}{2z}}$$

where $C$ is a phase constant ($e^{i \chi z}$).

At thickness $z$ and at point $\rho$, the wavefunction is:

$$|\Psi(\rho, z) >= < \rho | \Psi(z) >= < \rho | \hat{U}^p(z, 0) | \Psi(0) >= < \rho | \int d^2q \ q > e^{-i \frac{q^2 - \chi^2}{2kz} z} |\Psi(0) >$$
Introducing the closure relation $\int d^2 \rho' |\rho' > < \rho'|$ before $\Psi(0)$:

$$|\Psi(\rho, z) > = \int d^2 \rho' \int d^2 q \, e^{-i \frac{q^2 - \chi^2}{2kz} z} < \rho|q > < q|\rho' > < \rho'|\Psi(0) >$$

$$|\Psi(\rho, z) > = \int d^2 \rho' \int d^2 q \, e^{-i \frac{q^2 - \chi^2}{2kz} z} e^{i(\rho - \rho')} |\Psi(\rho', 0) > = C \int d^2 \rho' e^{i k z \frac{(\rho - \rho')^2}{2z}} |\Psi(\rho', 0) >$$

The wave function at a small distance from the object is the convolution of the object transmission function by the Fresnel propagator.
Blochwave method: Hermitic operator

Since any hermitic operator $\hat{A}$ can be written on its complete basis as:

$$\hat{A} = \int \alpha_i |a_i><a_i|$$

where the $\alpha_i$ and $|a_i>$ are its eigenvalues (real numbers) and its eigenvectors.

The electron eigenfunctions in a periodic crystal are the Bloch waves $|j> (j, 1, 2, \ldots)$.

$$\hat{H}|j> = \sum_j \gamma_j |j><j|$$

($|j>$ anpassung parameter or transverse kinetic energy of electron in Bloch state $j$)

Evolution operator $\hat{U}(z, 0)$:

$$\hat{U}(z, 0) = \sum_j e^{-i\gamma_j z}|j><j|$$

Wave function $|\Psi(\rho, z)>$ at the exit surface of the specimen:

$$|\Psi(\rho, z)> = \hat{U}(z, 0)|\chi> = \left(\sum_j e^{-i\gamma_j z}|j><j|\right)|\chi>$$
Blochwave method: diffracted amplitude $\phi_q(z)$

$$\phi_q(\rho, z) = < q|\Psi(\rho, z) >= < q|\hat{U}(z, 0)|\chi > = \sum_j e^{-i\gamma_j z} < q|j >< j|\chi >$$

$< q|j > = c_j^i$ are the coefficients of Bloch wave $|j >$ in basis $\{q\}$

$< j|\chi > = c_j^{i*}$ are the excitation coefficient of Bloch wave $|j >$ in basis $\{q\}$

The excitation coefficients $c_j^{i*}$ give the decomposition of the incident wave function $|\chi >$ at the entrance surface of the crystal on the $|j >$.

**Wave function** $|\Psi(\rho, z) >$ at thickness $z$:

$$|\Psi(\rho, z) > = \sum_q \phi_q(\rho, z)|q > = \sum_j c_j^{i*} \sum_q c_q^i e^{-i\gamma_j z}|q >$$

**Bloch wave**:

$$|j > = \sum_q c_q^j |q >$$
Blochwave method: calculation of the Bloch waves

\[ \langle q | \hat{H} | j \rangle = \frac{1}{2k_z} \left( -\Delta \rho - \chi^2 + V \right) \langle q | j \rangle = \gamma_j \langle q | j \rangle \]

\[ \frac{q^2 - \chi^2}{2k_z} \langle q | j \rangle + \sum_{q'} \langle q | V | q' \rangle \langle q' | j \rangle = \gamma_j \langle q | j \rangle \]

or classically \((j = 1, 2, \ldots)\):

\[ \frac{q^2 - \chi^2}{2k_z} c_q + \frac{V_{q' - q} c_{q'}}{2k_z} = \gamma_j c_q \]

\(\rightarrow\) system of homogeneous linear equations
\(\rightarrow\) the unknown are the \(c_q^j\).

With \(M_{ij} = \frac{q^2 - \chi^2}{2k_z} \delta_{ij} + \frac{V_{q_j - q_i}}{2k_z}\) \(\rightarrow\) eigensystem:

\[ [M - \gamma I] \ C = 0 \]

where \(I\) is a diagonal matrix and \(C\) the column vector of coefficients \(c_q^j\).
After diagonalisation:

\[ M = S^{-1} [\gamma] S \]

[\gamma] is the diagonal matrix of the eigenvalues and S the matrix of the eigenvectors.

Consequently:

\[ \frac{d}{dz} \Phi = i \pi S^{-1} [\gamma] S \Phi \]
The wave function is expanded on basis \( \{ q \} \):

\[
|\Psi(\rho, z) > = \sum_q \phi_q(z) |q >
\]

The complex diffracted amplitude becomes:

\[
\phi_q(z) = < q | \Psi(\rho, z) > = < q | \hat{U}(z, 0) | \chi >
\]

Deriving with respect to \( z \) and using \( i \frac{d\hat{U}(z, z_0)}{dz} = \hat{H}(z) \hat{U}(z, z_0) \):

\[
\frac{d\phi_q(z)}{dz} = < q | \frac{d\hat{U}(z, z_0)}{dz} | \chi > = -i < q | \hat{H} \hat{U} | \chi >
\]

with the closure relation \( \sum_k |g > < g| \) (reciprocal vectors \( g \) associated with the \( |q > \)):

\[
\frac{d\phi_q(z)}{dz} = -i \sum_g < q | \hat{H} | g > < g | \hat{U}(z, 0) | \chi >
\]
Howie-Whelan: Howie-Whelan equations

Since \( a_g(z) = \langle g | \hat{U}(z, 0) | \chi \rangle \) and \( \hat{H} = \hat{H}^{(p)} + \hat{V} \):

\[
\frac{d\phi_q(z)}{dz} = -i \sum_g \left[ \langle q | \hat{H}^{(p)} | g \rangle + \langle q | \frac{\hat{V}}{2k_z} | g \rangle \right] \phi_k(z)
\]

This forms a system of coupled differential equations whose \( q \) line equation is:

\[
\frac{d\phi_q(z)}{dz} = \pi \left[ \frac{\chi^2 - q^2}{2\pi k_z} \phi_q(z) - \sum_g \frac{V_{g-q}}{2\pi k_z} \phi_g(z) \right]
\]

\( \frac{\chi^2 - q^2}{2\pi k_z} \) is twice the Bragg parameter and \( V_{g-q} \) is the Fourier coefficient of the potential \( \langle q | V | k \rangle \). Since diffraction only occurs when each \( q \) is very near a given \( g \), the Howie-Whelan equations are obtained:

\[
\frac{d\phi_g(z)}{dz} = \pi \left[ \frac{\chi^2 - (g + s_g)^2}{2\pi k_z} \phi_g(z) - \sum_h \frac{V_{g-h}}{2\pi k_z} \phi_h(z) \right] \tag{1}
\]
Howie-Whelan: Kinematical approximation

Writing $\Phi$ the column matrix of the diffracted amplitudes $\phi_g(z)$ and $M$ the diffraction matrix given by:

$$M_{ij} = \frac{\chi^2 - (g_i + sg_i)^2}{2k_z} \delta_{ij} + \frac{V_{qi-qj}}{2k_z}$$

the Howie-Whelan equations becomes:

$$\frac{d}{dz} \Phi = i \pi M \Phi$$

When the potential does not depend on $z$:

$$\Phi = e^{i \pi M} \Phi_0$$

where $\Phi_0$ is the column matrix of the incident conditions, usually $(1, 0, 0, ...)$.

In the kinematical approximation the amplitude of the transmitted beam stays equal to one and the only term of the summation of equation 1 comes from the transmitted beam:

$$\frac{d\phi_g(z)}{dz} = i \pi \left[ 2s_g \phi_g(z) - \frac{V_g}{2\pi k_z} \right]$$

(2)
Howie-Whelan: Kinematic solution

With $\phi_0(z) = 1$:

$$\phi_g(z) = i \frac{V_g}{2\pi k_z} \frac{\sin(\pi s_g z)}{\pi s_g} e^{i\pi s_g z}$$

Notice that the diffracted beams are out of phase by $\frac{\pi}{2}$, an important fact in (high resolution) imaging.
Very simple object model: weak phase object approximation (WPOA).

Crystal potential $V(\vec{r})$ (constant over $dz$ small enough), wavevector is $(e E + V(\vec{r}))$:

$$k = \sqrt{\frac{2 m e (E + V(\vec{r}))}{\hbar^2}}$$

Imagine a plane wave arriving on a crystal.
How does the wavefront deform?
Refraction index \( n \) of object \( \vec{n} \) \( \longrightarrow \) ratio object wavevector and vacuum wavevector

\(| V(\vec{r}) | << E \):

\[
n = \frac{\vec{k}_m}{\vec{k}_v} = \sqrt{\frac{E + V(\vec{r})}{E}} \approx 1 + \frac{V(\vec{r})}{2E}
\]

\( \implies \) phase change \( d\varphi \) as a function of object thickness \( dz \):

\[
d\varphi = (\vec{k}_m - \vec{k}_v) \cdot d\vec{r} = (n - 1)|\vec{k}_v|dz = \frac{\chi}{2E} V(\vec{r})dz
\]

with \( \chi = |k_v| \).

Question: value of \( n \) (compare to glass)?
Phase object transmittance

For object of thickness $\Delta z$, phase shift $\Delta \varphi$ written as:

$$\Delta \varphi = \frac{\chi}{2E} \int_{z}^{z+\Delta z} V(\vec{x}; z) \, dz = \frac{\chi}{2E} V_p(\vec{x}; z) \Delta z$$

Transmittance function of phase object over $\Delta z$ is:

$$\Psi_o(\vec{x}) = e^{2\pi i \Delta \varphi} = e^{2\pi i \frac{\chi}{2E} V_p(\vec{x}; z) \Delta z} = e^{i \sigma V_p(\vec{x}; z) \Delta z}$$

with $\sigma = \frac{\pi \chi}{E} = \frac{\pi}{\lambda E}$. 

Calculations of HRTEM and STEM images
\[ \Psi_o(\vec{x}) = e^{i \sigma V_p(\vec{x}; z)} \approx 1 + i \sigma V_p(\vec{x}; z) \Delta z \]

In the back focal plane of the objective lens (Fourier transform):

\[ \tilde{\Psi}_o(\vec{k}) = \delta(\vec{k}) + i \sigma \tilde{V}_p(\vec{k}; z) \Delta z \]
Abbe image formation model:

\[ \tilde{\Psi}_i(\vec{k}) = \tilde{\Psi}_o(\vec{k}) \tilde{T}(\vec{k}) = \tilde{\Psi}_o(\vec{k}) e^{-2\pi i \chi(\vec{k})} \]

where \( \chi(\vec{h}) \) is (defocus \( \Delta_f \) and spherical aberration \( C_s \)):

\[ \chi(\vec{k}) = \frac{C_s \lambda^3 \left( \vec{k} \cdot \vec{k} \right)^2}{4} - \frac{\Delta_f \lambda \left( \vec{k} \cdot \vec{k} \right)}{2} \]
\( \Psi_i(\vec{x}) \Psi_i^*(\vec{x}) \) image intensity (image plane)

In objective lens back focal plane:

\[
\tilde{\Psi}_i(\vec{k}) = \left[ \delta(\vec{k}) + i \sigma \tilde{V}_p(\vec{k}; z) \Delta z \right] [\cos 2\pi \chi(\vec{k}) - i \sin 2\pi \chi(\vec{k})]
\]

Choosing \( \sin 2\pi \chi(\vec{k}) = -1 \) et \( \cos 2\pi \chi(\vec{k}) = 0 \) for diffracted beams \( \vec{k} \), \( \tilde{\Psi}_i(\vec{k}) \) becomes:

\[
\tilde{\Psi}_i(\vec{k}) = \delta(\vec{k}) - \sigma \tilde{V}_p(\vec{k}; z) \Delta z
\]

Image intensity \((\Psi_i(\vec{x}) \Psi_i^*(\vec{x}))\) given by:

\[
I(\vec{x}) = (1 - \sigma V_p(\vec{x}; z))(1 - \sigma V_p(\vec{x}; z)) \approx 1 - 2\sigma V_p(\vec{x}; z) \Delta z
\]

Dark dots at the position of the atomic columns!
Remarks

1. Transfer function compensates the $\frac{\pi}{2}$ phase shift due to diffraction.
2. Direct interpretation of HRTEM micrographs possible.
3. Spots are darker when projected potential is important (heavy atoms).
4. Spots are darker when specimen thickness increases.
5. WPOA approximation only valid for very thin crystals ($\text{Au} \leq 1$ unit cell!).

$\sin 2\pi \chi(\vec{k}) = -1$ is selected by changing the specimen defocus $\Delta z$. 
Choosing $\sin 2\pi \chi(\mathbf{h}) = 0$ et $\cos 2\pi \chi(\mathbf{h}) = -1$, $(\Psi_i(\mathbf{x})\Psi_i^*(\mathbf{x}))$ is since:

$$\tilde{\Psi}_i(\mathbf{h}) = [\delta(\mathbf{h}) + i\sigma \tilde{V}_p(\mathbf{h}; z)\Delta z]$$

$$I(\mathbf{x}) = (1 - i\sigma V_p(\mathbf{x}; z)\Delta z)(1 + i\sigma V_p(\mathbf{x}; z)\Delta z)$$

$$= 1 + \sigma^2 V_p^2(\mathbf{x}; z)\Delta z^2$$

**White dots at the position of the atomic columns!**

Contrast no more proportional to projected potential. Very small $\Delta z$ change necessary to get contrast inverted micrographs.
In the image plane the image wave-function $\Psi_i(\vec{x})$ is the inverse Fourier transform of $\tilde{\Psi}_i(\vec{h})$ given by:

$$
\tilde{\Psi}_i(\vec{h}) = \tilde{\Psi}_o(\vec{h}) \tilde{T}(\vec{h})
$$

where $\tilde{\Psi}_o(\vec{h})$ is the object wave $\tilde{T}(\vec{h})$ is the transfer function of the microscope:

$$
\Psi_i(\vec{x}) = \int \tilde{\Psi}_o(\vec{h}) \tilde{T}(\vec{h}) e^{2\pi i \vec{h} \cdot \vec{x}} d\vec{h}
$$

In other words $\Psi_i(\vec{x})$ is the superposition (or interference) of waves transmitted by the optical system. The image intensity is then given by:

$$
l_i(\vec{x}) = \Psi_i(\vec{x}) \Psi_i^*(\vec{x})
$$

For a weak phase object the image intensity has a simple interpretation since it is proportional to the refraction index or the projected potential of the object.

What happen under incoherent illumination, i.e. scanning confocal image formation?
Incoherent illumination

When the illumination is incoherent (typically in scanning confocal microscopy) the image intensity is given by the temporal mean:

\[
I(\vec{x}) = \langle \Psi_i(\vec{x}; t) \Psi_i^*(\vec{x}; t) \rangle
\]

When the transfer function of the optical system is time invariant, each \( \Psi_i(\vec{x}; t) \) is given by:

\[
\Psi_i(\vec{x}; t) = \Psi_o(\vec{x}; t) \otimes T(\vec{x})
\]

Thus the temporal mean is:

\[
I(\vec{x}) = \langle [\Psi_o(\vec{x}; t) \otimes T(\vec{x})] [\Psi_o^*(\vec{x}; t) \otimes T^*(\vec{x})] \rangle
\]

Since \( T(\vec{x}) \) is time independent:

\[
I(\vec{x}) = [T(\vec{x}) T^*(\vec{x})] \otimes \langle \Psi_o(\vec{x}; t) \Psi_o^*(\vec{x}; t) \rangle
\]
Optical transfer function: OTF

Since $T(\vec{x})$ is time independent:

$$I(\vec{x}) = [T(\vec{x}) T^*(\vec{x})] \otimes \langle \Psi_o(\vec{x}; t) \Psi_o^*(\vec{x}; t) \rangle$$

When the spatial coherence is null, there is no phase correlation between object points:

$$\langle \Psi_o(\vec{x}; t) \Psi_o^*(\vec{x}; t) \rangle = |\Psi_o(\vec{x})|^2$$

$$I(\vec{x}) = |\Psi_o(\vec{x})|^2 \otimes [T(\vec{x}) T^*(\vec{x})]$$

which usually written as:

$$I(\vec{x}) = I_o(\vec{x}) \otimes OTF(\vec{x})$$

$$OTF(\vec{x}) = T(\vec{x}) T^*(\vec{x})$$

where the $OTF(\vec{x})$ is the point spread function under incoherent illumination.
Under incoherent, the optical system transfers the object intensity.

The OTF is the autocorrelation of the transfer function of the microscope.

The OTF is:

\[
\hat{\text{OTF}}(\vec{h}) = \hat{T}(\vec{h}) \otimes \hat{T}^*(\vec{h})
\]

The transfer function of a perfect diffraction limited optical system, i.e. the entrance pupil sets a limit \(u_{\text{max}}\) on the highest spatial frequency transferred by the system is \(\text{rect}(\vec{u})\) (in 1-D). Its impulse response is a \(\text{sinc}(\vec{x})\) function.

The OTF will be the convolution of the \(\text{rect}(\vec{u})\) by itself, i.e. a \(\Lambda(\vec{u})\) function, that extends to a maximum spatial frequency 2 \(u_{\text{max}}\).
Under partially spatial and/or temporal coherence the important quantity is the **mutual intensity** of 2 points of the object, the object acting as the light source for the optical system.

The Fourier transform of the image intensity is linked to the object wave-function by the transmission cross coefficients \( \tilde{T}_{cc}(\vec{h} + \vec{h}'; \vec{h}; z) \) that take into account the wavelength spread of the source and its angular size.

Noting \( \phi(\vec{h}) \) the beam diffracted in direction \( \vec{k}_0 + \vec{h} \), the Fourier transform of the image intensity is given by:

\[
\tilde{I}(\vec{h}; z) = \int \tilde{T}_{cc}(\vec{h}' + \vec{h}; \vec{h}; z) \phi(\vec{h}' + \vec{h})\phi^*(\vec{h}')d\vec{h}'
\]

This means that the image intensity contains non-linear interference terms (i.e. interference between diffracted beams).
Noting in Fourier space $P_E$ the entrance pupil of the optical system, the $T_{cc}$ of $h'$ and $h''$ are defined under the common area of the source and the pupil functions centered at $h'$ and $h''$. 

**Figure:** Integrating domains of the $T_{cc}$. 

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Calculations of HRTEM and STEM images
\[ \tilde{T}_{cc}(\vec{h}''; \vec{h}'; z) = \int \tilde{s}(\vec{h}) f(z') a(\vec{h}'' + \vec{h}) e^{2\pi i \chi(\vec{h}'' + \vec{h}; z' + z)} \times a(\vec{h}' + \vec{h}) e^{2\pi i \chi(\vec{h}' + \vec{h}; z')dz'} \]

where:

- \( \chi(\vec{h}; z) \) is the aberration function \( \frac{z\lambda \vec{h} \cdot \vec{h}}{2} + \frac{C_s \lambda^3 (\vec{h} \cdot \vec{h})^2}{4} \) when only defocusing and spherical aberration are introduced.

- \( \tilde{s}(\vec{h}) \) is the Fourier transform of the normalized intensity distribution of the source (lateral coherence).

- \( f(z) \) is the focal length distribution due to the spread of the wavelength (chromatic aberration).

- \( a(\vec{h}) \) is the pupil function of the optical system (the smallest of \( P_E \) or \( P_S \)).
A simple case

When the distributions are Dirac functions (coherent illumination), the transmission cross-coefficients reduce to:

\[ \tilde{T}_{cc}(\vec{h}''; \vec{h}'; z) = a(\vec{h}'') e^{2\pi i \chi(\vec{h}''; z)} a(\vec{h}') e^{2\pi i \chi(\vec{h}'; z)} \]

For the weak phase object (i.e. interference only possible between the transmitted beam and diffracted beams):

\[ \tilde{T}_{cc}(0; \vec{h}'; z) = a(\vec{h}') e^{2 \pi i \chi(\vec{h}'; z)} \]

which the transfer function of the coherent.
Weak phase object

The weak phase object approximation assumes that the object wave function can be approximated by:

\[ \tilde{\Psi}_o(\vec{h}) = \delta(\vec{h}) - i\sigma \tilde{V}_p(\vec{h}) = \delta(\vec{h}) + \varepsilon(\vec{h}) : \]

\[ \tilde{I}(\vec{h}; z) = \int \tilde{T}_c(\vec{h}' + \vec{h}; \vec{h}; z) \{ \delta(\vec{h}' + \vec{h}) + \varepsilon(\vec{h}' + \vec{h}) \} \times \{ \delta(\vec{h}') + \varepsilon^*(\vec{h}') \} d\vec{h}' \]

\( \tilde{I}(\vec{h}; z) \) decomposes into 3 contributions:

\[ \tilde{I}_0(\vec{h}; z) = \tilde{T}_c(\vec{h}; 0; z)\delta(\vec{h}) \]
\[ \tilde{I}_1(\vec{h}; z) = \tilde{T}_c(\vec{h}; 0; z)\varepsilon(\vec{h}) + \tilde{T}_c(0; -\vec{h}; z)\varepsilon^*(-\vec{h}) \]
\[ \tilde{I}_2(\vec{h}; z) = \int \tilde{T}_c(\vec{h}' + \vec{h}; \vec{h}; z)\varepsilon(\vec{h}' + \vec{h})\varepsilon^*(\vec{h}') d\vec{h}' \]
The 3 terms are:

- $\tilde{I}_0(\vec{h}; z) \implies$ image mean value.
- $\tilde{I}_1(\vec{h}; z) \implies$ contribution to the image intensity due to the interference of the transmitted beam with the diffracted beams.
- $\tilde{I}_2(\vec{h}; z) \implies$ contribution of the interference between the diffracted beams.

When the diffracted beams are very weak (biological objects, thin crystals, ...) one can neglect the contribution of $I_2$. 
Model: $Ti_2Nb_{10}O_{29}$
Questions: where are the atoms? Do we see the oxygen atoms?
Transfer function limited by gun and objective lens current instabilities (and incident beam convergence).
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