Transmission Electron Microscopy

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1931: (Conventional) Transmission Electron Microscope (CTEM)

1937: Scanning Transmission Electron Microscope (STEM)

1997: Aberration corrected optics
This lecture: atomic resolution

quantitative studies: measurements in atomic dimensions with picometer precision
Transmission Electron Microscopy
CTEM

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Part I
How is an atomic image formed in the transmission electron microscope?
The principles of imaging

\[ \psi(r) = \exp(2\pi ik_0 r) \]

\[ \rho(r) = \sum_i \rho_i(r) \delta(r - r_i) \]

unknown structure

exit-plane wave function

\[ \psi(r) = \int_g \psi(g) \exp\{2\pi i(k_0 + g)r\}dg \]

g - spatial frequency

\[ I(r) \propto |\psi|^2 \]
The principles of imaging

\[ \psi(r) = \exp(2\pi i k_0 r) \]

exit-plane wave function

\[ \psi(r) = \int_{g} \psi(g) \exp\{2\pi i (k_0 + g) r\} dg \]

\( g \) - spatial frequency

lens aberrations

aberration-induced phase shifts

\[ \psi(g) \exp\{-2\pi i \chi(g)\} \]

\[ \chi(g) = \frac{1}{4} C_s \lambda^3 g^4 + \frac{1}{2} Z \lambda g^2 + \ldots \]

aberration function

spherical aberration

defocus
The principles of imaging

\[ \psi(r) = \exp(2\pi i k_0 r) \]

\( \rho(r) = \sum_i \rho_i(r) * \delta(r - r_i) \)

unknown structure

exit-plane wave function

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\( g \) - spatial frequency

lens aberrations

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aberration function

spherical aberration
defocus
Spherical aberration

Object

Lens

Gaussian image plane

Point spread function

\[ R \propto \left| \frac{\partial \chi}{\partial g} \right|_{\text{max}} \]

\[ \chi(g) = \frac{1}{4} C_s \lambda^3 g^4 + \frac{1}{2} Z \lambda g^2 + \ldots \]

aberration disk
Spherical aberration in light microscopy

Object

Lens
crown glass (n=1.6)
flint glass (n= 1.9)

Gaussian image plane

R \approx 0

Ernst Abbe, 1875
Spherical aberration in electron microscopy

Gauss' law of magnetism

$\nabla B = 0$

always converging
1997
the world's first aberration-corrected electron microscope
As of today: about 800 aberration-corrected electron microscopes

All - CTEM and STEM - have our double-hexapole correction system (except for @ 25 Nion instruments)
Part II
How do the atoms produce contrast?
Amplitude contrast in atomic imaging

Electron diffraction channelling

plane wave

entry surface
thickness
exit surface

image
Amplitude contrast in atomic imaging

Electron diffraction channelling
Amplitude contrast in atomic imaging

Electron diffraction channelling

entry surface

thickenss

exit surface

high Z

low Z

image
increasing video time $t = \text{increasing depth in crystal } d$
Phase contrast in atomic imaging

High $E_{\text{pot}}$ / less $E_{\text{kin}}$

Low $E_{\text{pot}}$ / high $E_{\text{kin}}$

Electrons moving at the atoms are faster:
their phase is more advanced

Phase Contrast

Phase (differences) cannot be seen

Zernike’s technique allows to convert phase contrast into amplitude contrast
Zernike technique in the light microscope (1930)

\[ \psi(r) = \exp(2\pi i k_0 r) \]

Object \( \Rightarrow V(r) \) \( \Rightarrow \) Lens \( \Rightarrow \) Image

\[ \lambda/4 \]

Diffracted beams

Incident, transmitted beam

Im \( \pi/2 \)

Re

Dark contrast on a bright background
Zernike technique in the electron microscope typically by 5 orders smaller $\lambda$

Applications of phase plates mainly in biology

*In materials science:* defocus technique, i.e. the lens has two functions:

1. to image
2. and to provide contrast

R. Danev et al., PNAS 111, 15635 (2014)
Zernike technique in the electron microscope (1949)

\[ \psi(r) = \exp(2\pi ik_0 r) \]

Object

\[ V(r) \]

Lens

Image

\[ I(r) \propto |\psi|^2 \propto \sin 2\pi \chi(g) \]

exit-plane wave function

\[ \psi(r) = \int_g \psi(g) \exp\{2\pi i(k_0 + g)r\} dg \]

lens aberrations

aberration-induced phase shifts

\[ \psi(g) \exp\{-2\pi i\chi(g)\} \]

\[ \chi(g) = \frac{1}{4} C_s \lambda^3 g^4 + \frac{1}{2} Z \lambda g^2 + \ldots \]

spherical aberration

defocus

contrast transfer function

Otto Scherzer
Scherzer’s theory for phase contrast (1949)

\[ Z_S = -\left(\frac{4}{3}C_S\lambda\right)^{\frac{1}{2}} \]
\[ g_S = 2\left(3C_S\lambda^3\right)^{-\frac{1}{4}} \]
\[ d_S = g_S^{-1} \]

- Extend region of close to “1” to large \( g \)
- First “zero” at \( g_S \) as large as possible

The basis of “high resolution” for more than half a century

But there are issues:

Information is wasted
Severe delocalization

\[ \chi(g) = \frac{1}{4}C_S\lambda^3g^4 + \frac{1}{2}Z\lambda g^2 + \ldots \]
Delocalization

Object

Lens

Gaussian image plane

aberration disk

Point spread function

\[ R \propto \left| \frac{\partial \chi}{\partial g} \right|_{\text{max}} \]

\[ \chi(g) = \frac{1}{4} C_s \lambda^3 g^4 + \frac{1}{2} Z \lambda g^2 + \ldots \]

aberration function

inserting the Scherzer value for Z

\[ R = \left| \frac{\delta \chi}{\delta g} \right|_{\text{max}} = \left| C_s \lambda^3 g^3 + Z \lambda g \right|_{\text{max}} \approx 3d_s \]

delocalization 3 x the resolution
Jülich theory for phase contrast in the aberration corrected CTEM


\[ \chi(g) = \frac{1}{4} C_s \lambda^3 g^4 + \frac{1}{2} \frac{Z}{\lambda} g^2 + \cdots \]

spherical aberration \text{variable}  
defocus \text{variable}
Jülich theory for phase contrast (2002)


\[ Z_{opt} = -\frac{16}{9} (\lambda g_I^2)^{-1} \]
\[ C_{S,opt} = +\frac{64}{27} (\lambda^3 g_I^4)^{-1} \]
\[ R_{opt} = \frac{16}{27} g_I^{-1} \]

Optimization of contrast transfer exploiting variable \( C_S \) and variable \( Z \):

1) Information transferred up to \( g_I \)
2) No (!) Contrast delocalisation

\[ \chi(g) = \frac{1}{4} C_S \lambda^3 g_4^4 + \frac{1}{2} Z \lambda g^2 + \ldots \]

Contrast \( \propto \sin 2\pi \chi(g) \)

spherical aberration

variable

defocus

variable
Amplitude contrast and phase contrast

One problem is remaining

electron diffraction channelling

Amplitude image

bright contrast
Amplitude contrast and phase contrast

\[ Z_{opt} = -\frac{16}{9} (\lambda g_i^2)^{-1} \]

\[ C_{S, opt} = \frac{64}{27} (\lambda^3 g_i^4)^{-1} \]

electron diffraction channelling

weakening each other

Phase image

dark contrast

Amplitude image

bright contrast
The trick of NCSI (negative spherical aberration imaging)


\[ Z_{opt} = + \frac{16}{9} \left( \lambda g_i^2 \right)^{-1} \]

\[ C_{S, opt} = - \frac{64}{27} \left( \lambda^3 g_i^4 \right)^{-1} \]

- **electron diffraction channelling**

- **enhancing each other**

- **Phase image**
  - bright contrast

- **Amplitude image**
  - bright contrast
The trick of NCSI (negative spherical aberration imaging)
Part III
From the images to the unknown structure
Reconstruction of exit-plane wave function by TrueIMAGE™

(A. Thust, J. Barthel, 2011)

\[ \Psi_{j+1} = \Psi_j + \delta \Psi_j \]

\[ S^2 = \frac{1}{N} \sum_{n=1}^{N} \int |\delta I_n(g)|^2 \, dg \]

$$\rho(r) = \sum_i \rho_i(r) \cdot \delta(r - r_i)$$  \hspace{1cm} \text{object structure}$$

\[
\begin{array}{c}
- \frac{\hbar^2}{2m} \nabla^2 \\
\text{inversion of Schrödinger equation}
\end{array}
\]

$$\psi(r) = \int_g \psi(g) \exp\{2\pi i (k_0 + g)r\} dg$$  \hspace{1cm} \text{exit-plane wave function}$$

$$I(r) \propto |\psi|^2$$  \hspace{1cm} \text{intensity distribution}$$

First guess model

- calculate exit-plane wave function
- compare
- modify atom positions

Iteration
Traditional: *Focal series*

focal series of images:

$$\chi(g) = \frac{1}{4} C_s \lambda^3 g^4 + \frac{1}{2} Z \lambda g^2 + \ldots$$

task is finished, when the whole series of images are matched correctly
Traditional: **Focal series**

focal series of images:

\[ \chi(g) = \frac{1}{4} C_s \lambda^3 g^4 + \frac{1}{2} Z \lambda g^2 + \cdots \]

Interferometric imaging

**Disadvantages:**

- manipulates the contrast transfer function
- optical instabilities
- sample drift
- diminishes – in effect – the resolution