Central European Institute of Technology
BRNO | CZECH REPUBLIC

Electron Matter Interaction

Fall 2023

Ondrej L. Shanel, Ph.D.

with kind help of Andrea R. Konecna, Ph.D.

Why Electron Microscopy

- Electron benefits

  - Fundamental
    - Shorter Wavelength than light at the same energy
    - Interaction mechanisms with matter (signal types)

  - Technological
    - Creation
    - Manipulation
    - Detection

Electron description

Classical

Particle description ●

Quantum-mechanical

Wave function

Non/Relativistic

Controlled by fields (electric **E** / magnetic **B**)

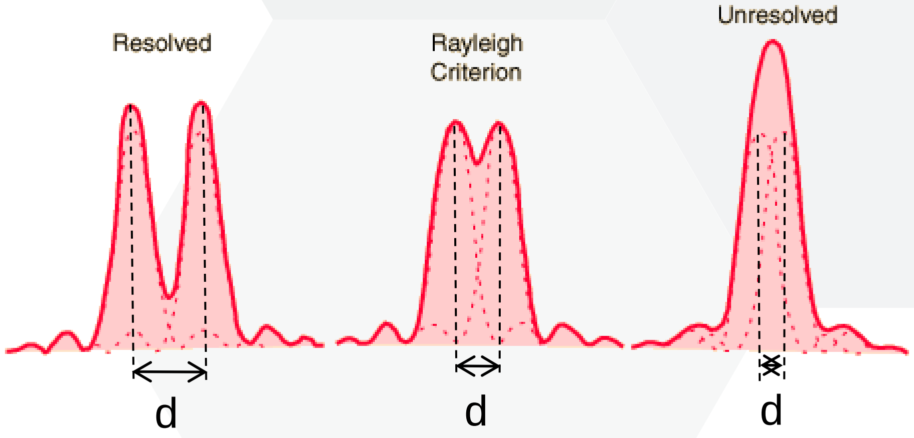
Electron properties

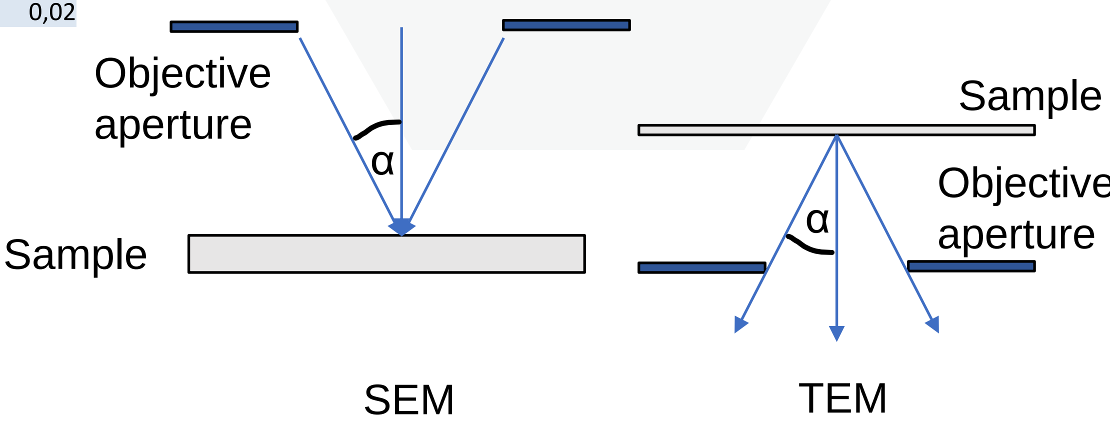
Energy of electron defines its main imaging properties

Rayleigh criterion

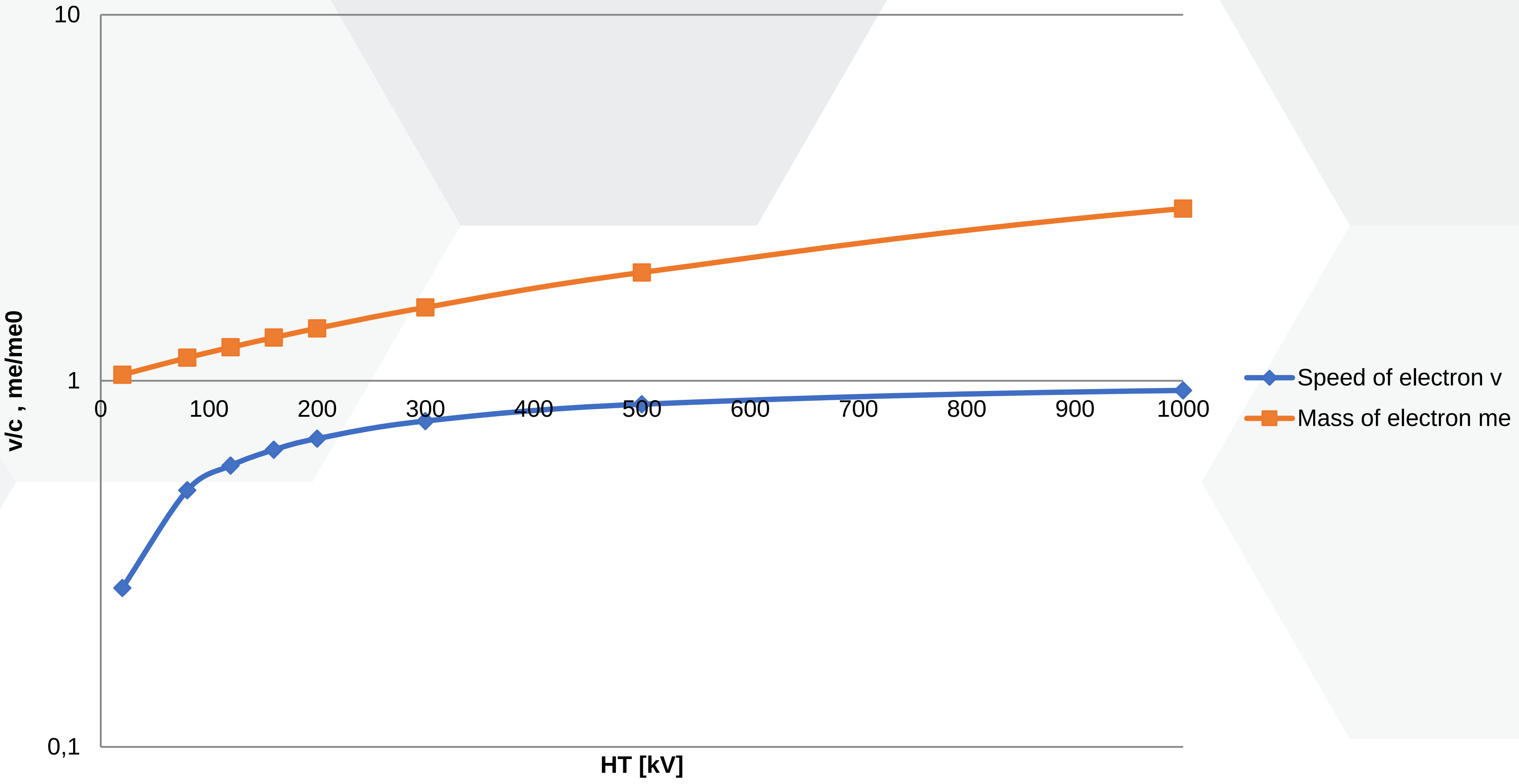
$$d= 1.22\ \lambda / n.\sin\alpha$$

Voltage accelerating electron [kV]	Speed of electron [v/c]	Relative mass of electron [m/m0]	Wave length [m]	Rayleigh criterion Alpha=14 mrad [nm]	Rayleigh criterion Alpha=100mrad [nm]
5	0,14	1,010	1,7E-11	1,51	0,21
10	0,19	1,020	1,2E-11	1,06	0,15
20	0,27	1,039	8,6E-12	0,75	0,11
30	0,33	1,059	7,0E-12	0,61	0,09
60	0,45	1,117	4,9E-12	0,42	0,06
80	0,50	1,156	4,2E-12	0,36	0,05
100	0,55	1,195	3,7E-12	0,32	0,05
120	0,59	1,234	3,4E-12	0,29	0,04
200	0,70	1,391	2,5E-12	0,22	0,03
300	0,78	1,586	2,0E-12	0,17	0,02





Electron properties Speed and Mass



Electron in Classical particle description

$$\mathbf{F} = \frac{\mathrm{d}\mathbf{p}}{\mathrm{d}t} = \frac{\mathrm{d}(m\mathbf{v})}{\mathrm{d}t} = -e(\mathbf{E} + \mathbf{v} \times \mathbf{B})$$

$$m = \gamma m_{\mathrm{e}}, \qquad \gamma = \frac{1}{\sqrt{1 - v^2/c^2}}$$

Relativistic mass

Lorentz contraction factor

E – Electrical intensity
B – magnetic flux
m_e – electron rest mass
c – speed of light

Wave function description

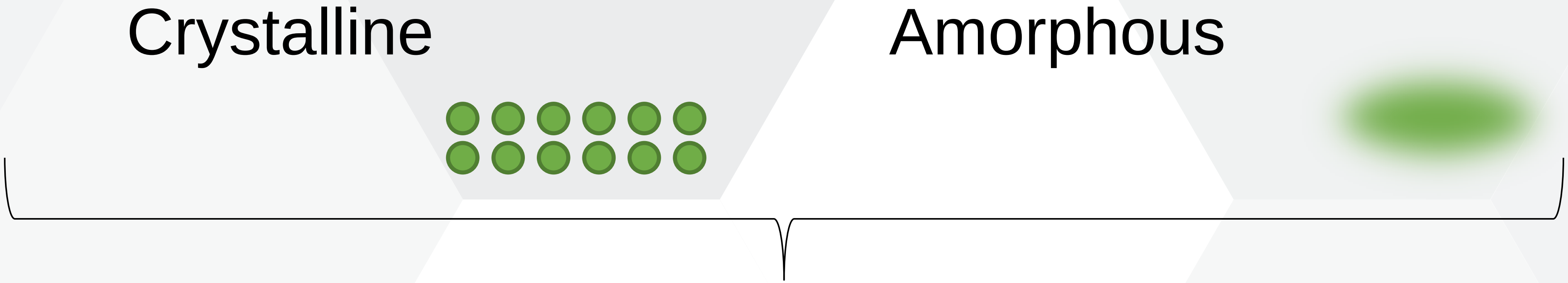
$$\frac{1}{2m_\mathrm{e}}(-\mathrm{i}\hbar\nabla + e\mathbf{A})^2\Psi - e\Phi^*\Psi = \frac{\mathrm{i}\hbar m}{m_\mathrm{e}}\frac{\partial\Psi}{\partial t}$$

$$\Phi^* = \Phi\left(1 + \frac{e}{2m_\mathrm{e}c^2}\Phi\right)$$

Relativistically corrected scalar potential

A – magnetic scalar vector
Ψ – wave function
Φ- electrical potential
m_e – electron rest mass
c – speed of light

Sample description

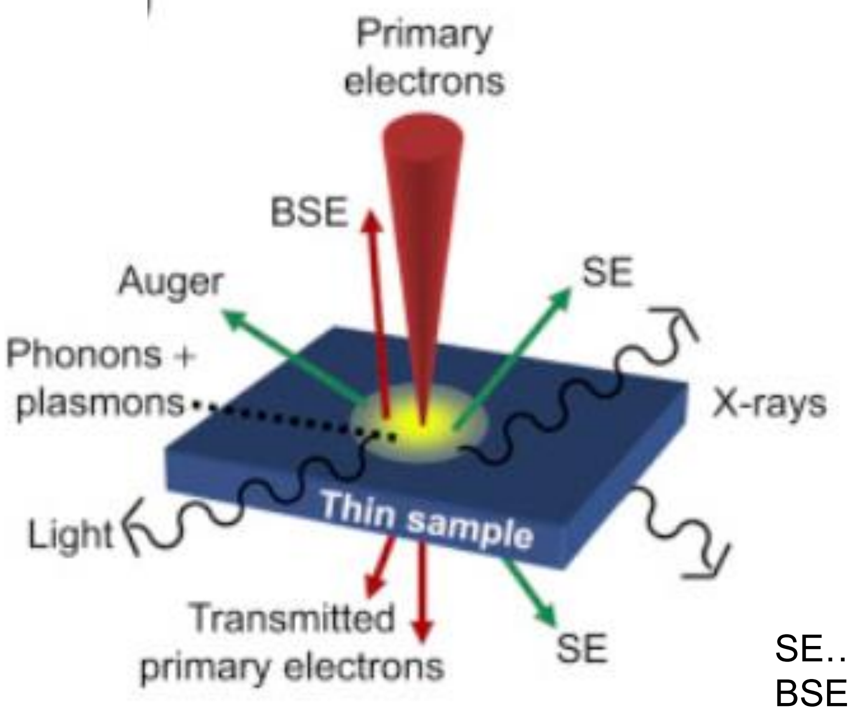


Described by a potential / scattering probability obtained

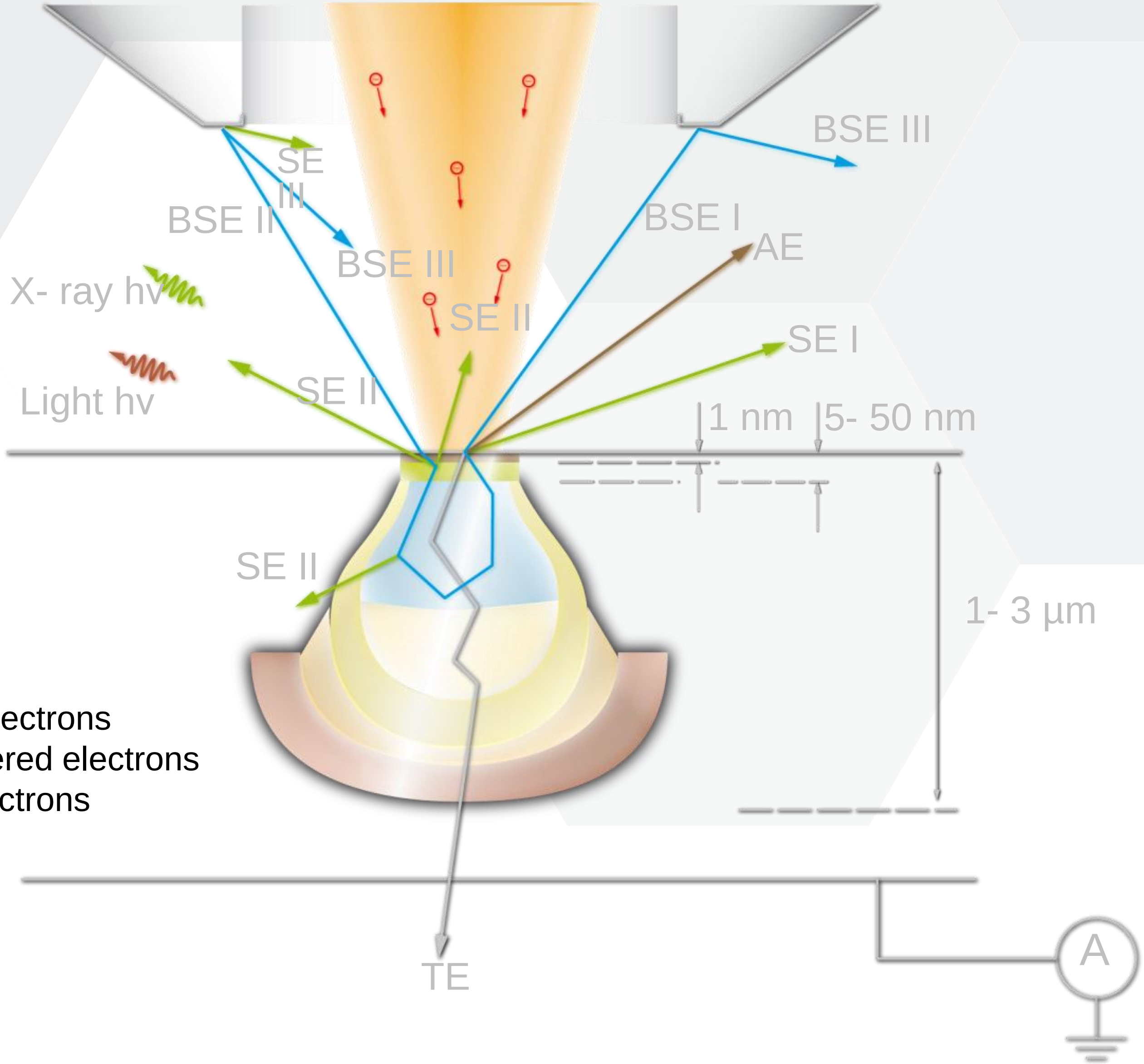
- From first principles
- Quasi-classically
- Empirically

Electron – Matter interaction types

Thin sample (S/TEM ≤ 200nm)

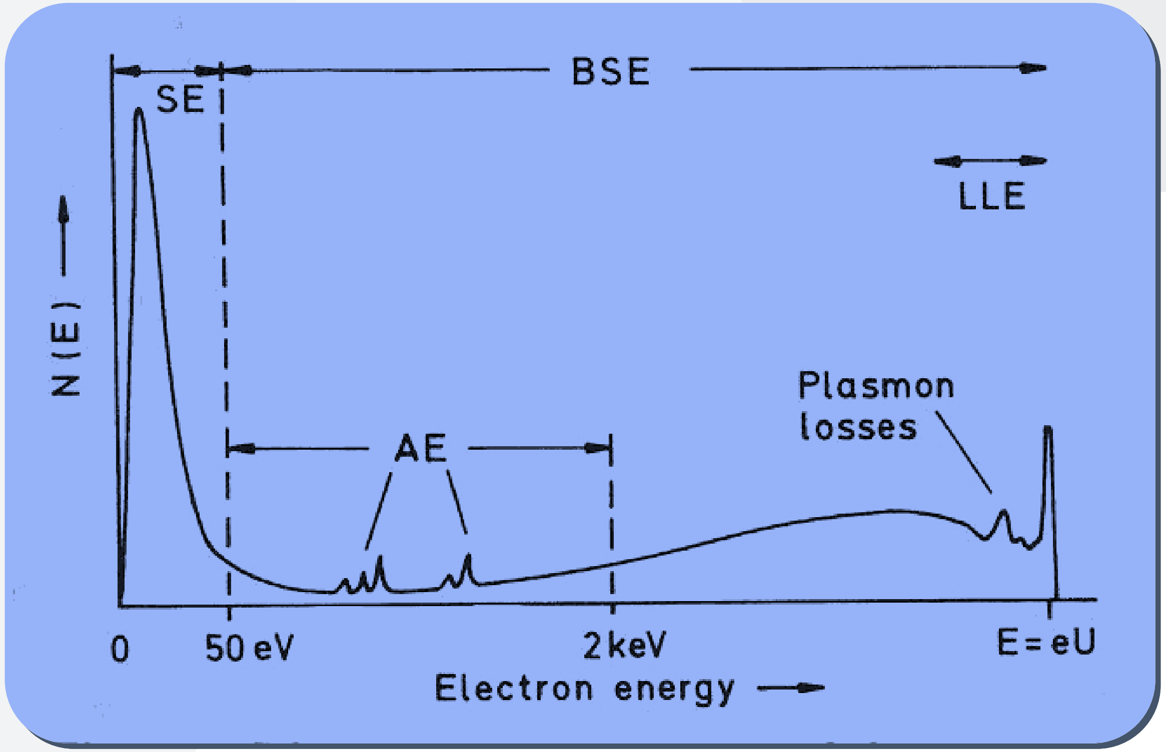


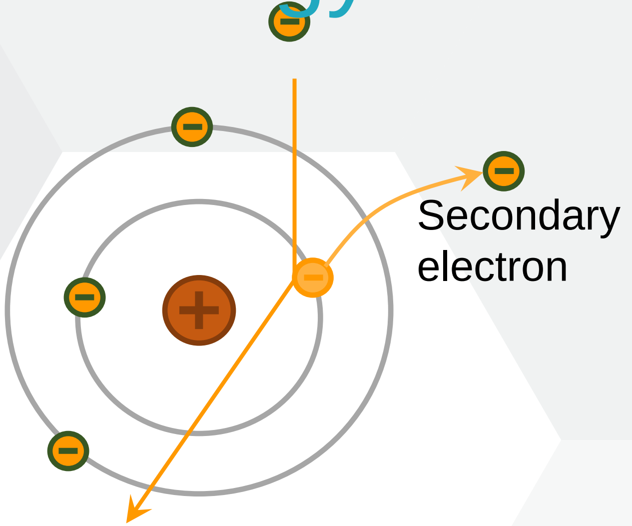
Thick sample (SEM)



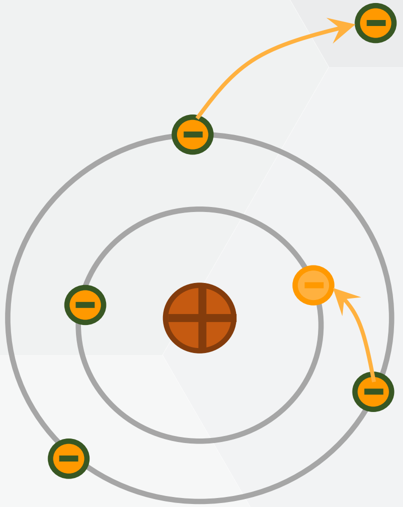
SE… secondary electrons
BSE… back-scattered electrons
Auger… Auger electrons

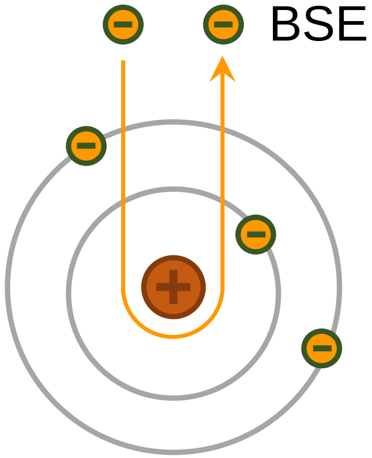
Electron – Matter interaction – Energy distribution

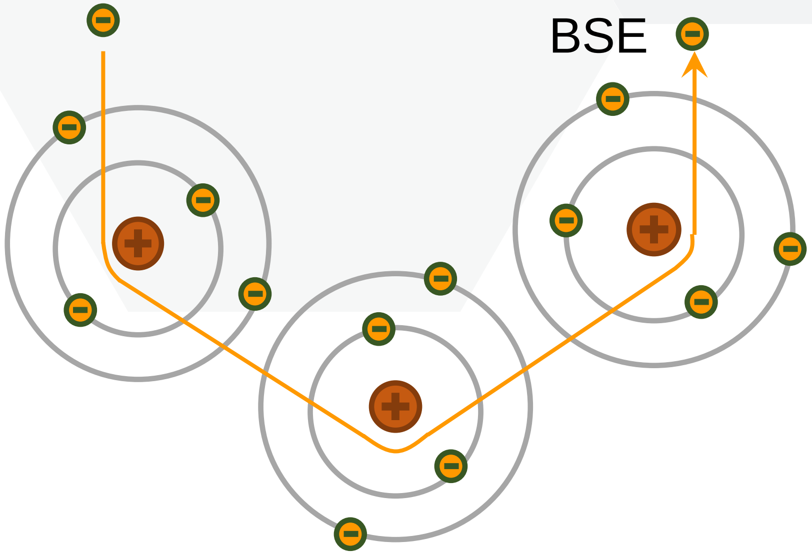



Secondary electron

Inelastically scattered incident electron

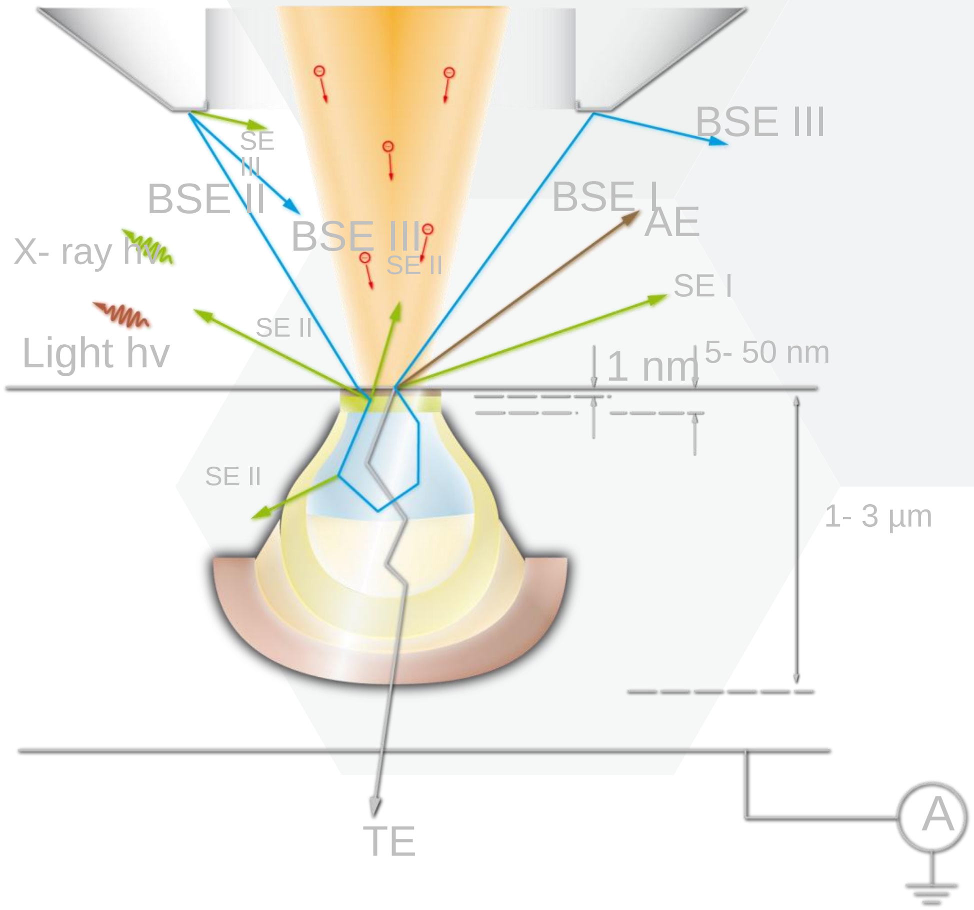

Auger electron


BSE


BSE

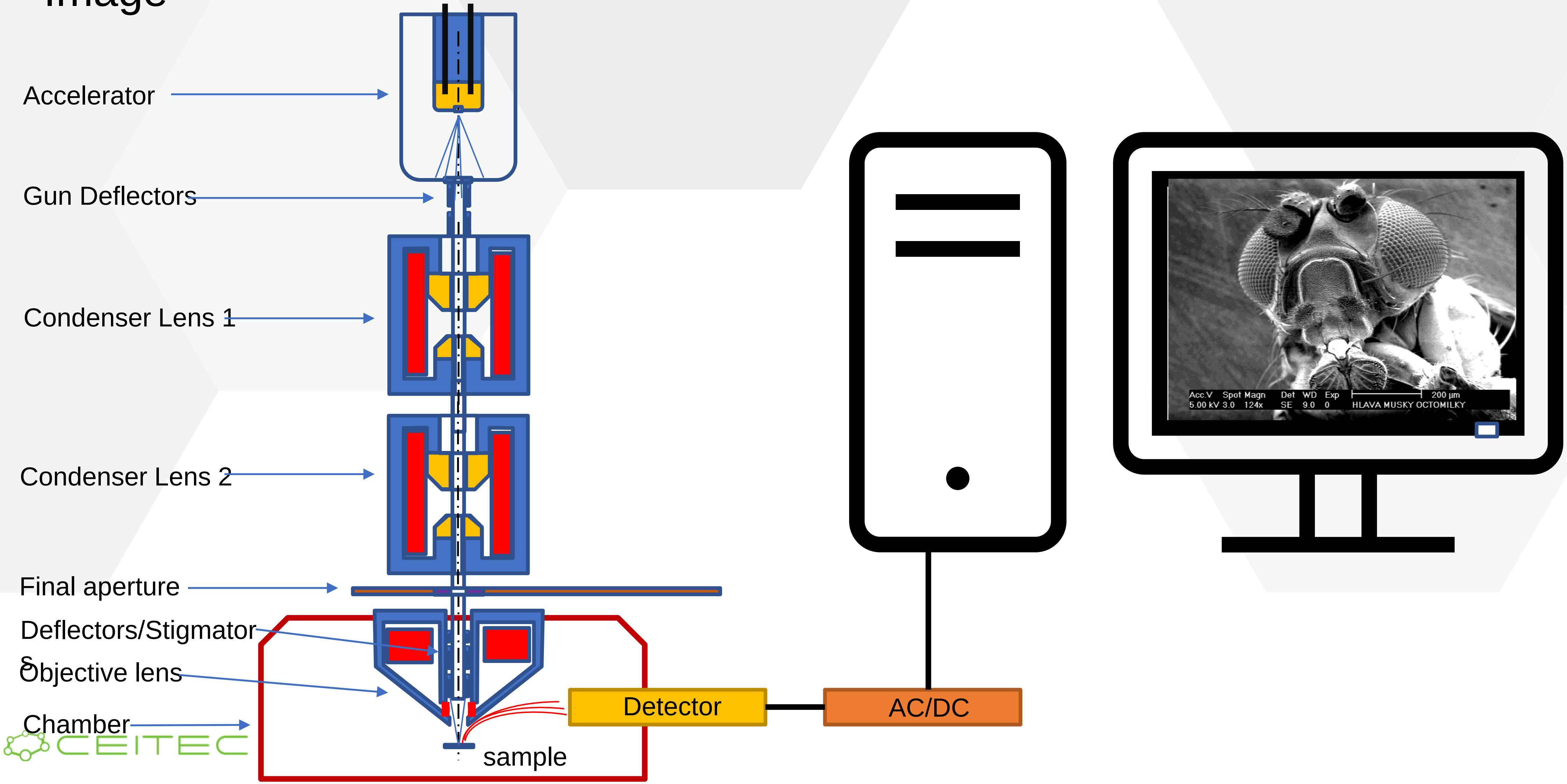
Scanning electron microscopy - SEM

- Electrons focused to small probe and scanning over the sample
- Electron energy: 1-30keV
- Resolution ~ 1nm
- Thick samples
- Signal depends on:
  - Sample morphology
  - Sample material
  - Crystal orientation



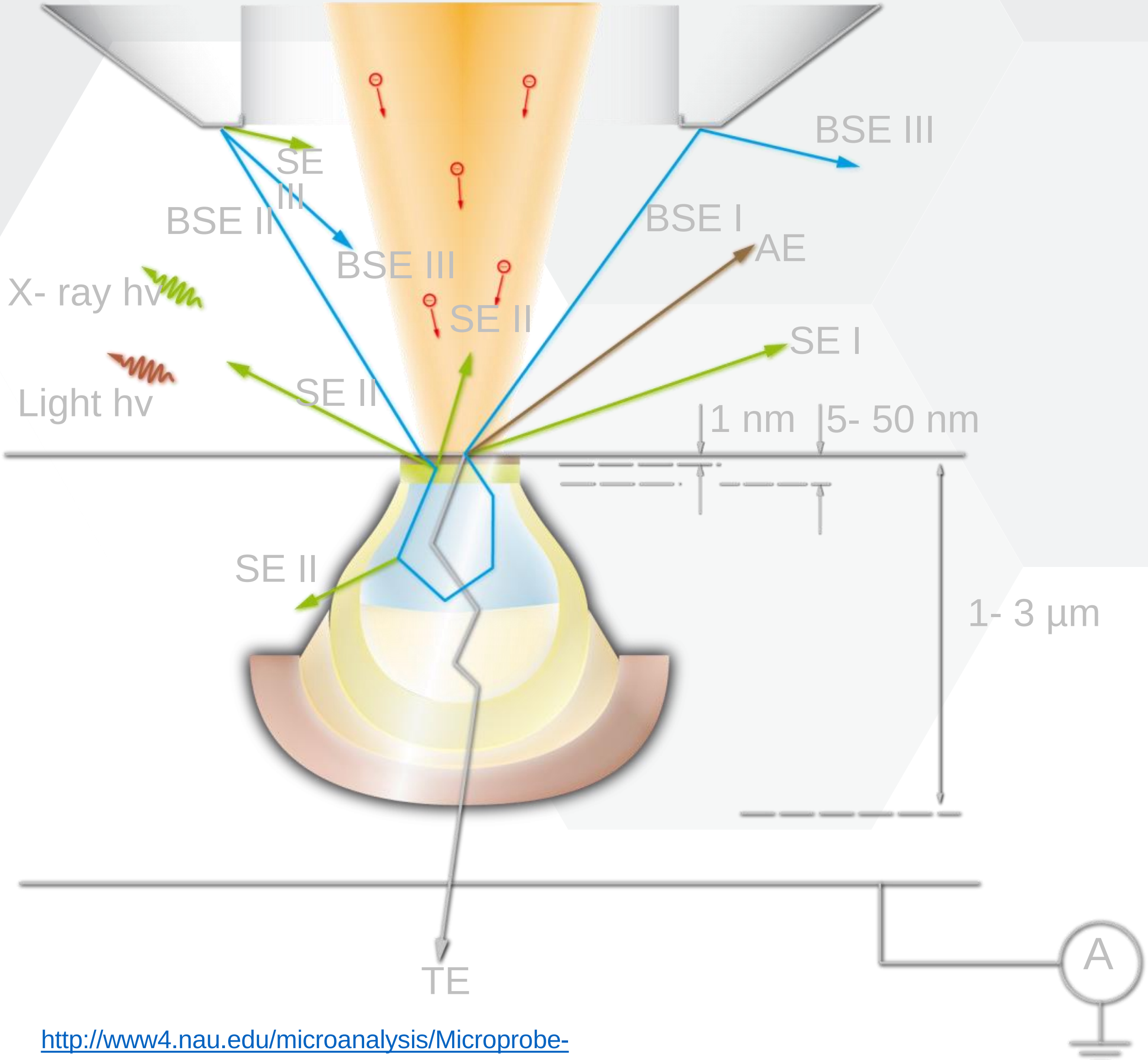
Scanning electron microscopy - Principle

- Using Focus Beam to Scan over the sample and process signal into Intensity map - Image



Scanning electron microscopy - Signals

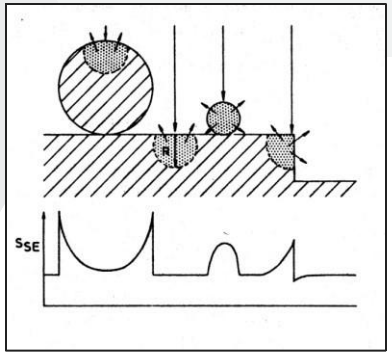
- Electron signals
  - Secondary electrons – (SE), E<50eV, small escape depth (~10nm) □ best resolution
  - Backscattered electrons – (BSE), 50eV<E≤E$_{\text{primary beam}}$, large interaction volume
  - Auger electrons, E>50eV, characteristic peaks, surface material composition information
  - Transmitted electrons (sample must be thin enough)
  - Absorbed electrons/current
- Photons
  - Cathodoluminescence
  - X-ray

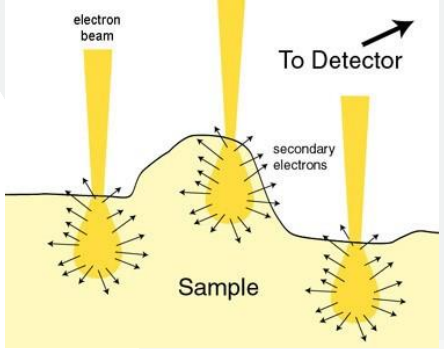


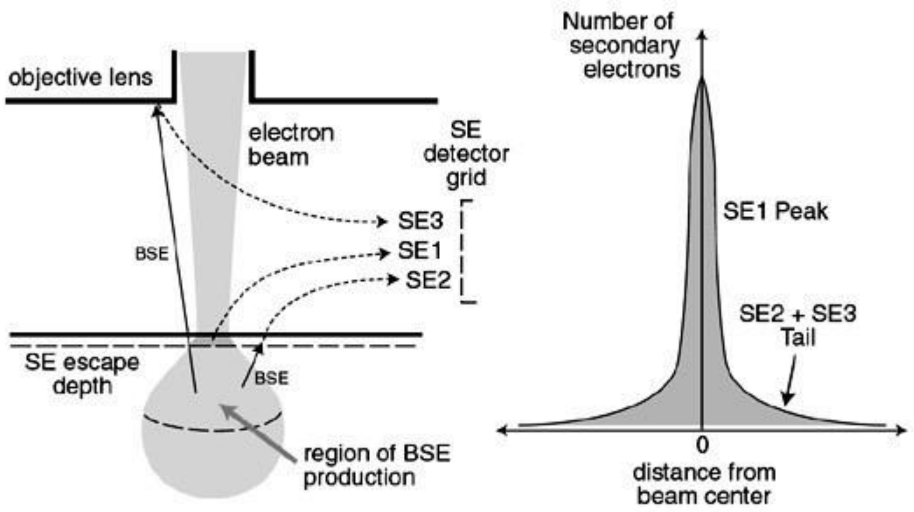
http://www4.nau.edu/microanalysis/Microprobe-SEM/Signals.html

Secondary electrons

- Electrons emitted by the sample under electron beam (inner shell ionization effects)
- Small escape depth high resolution
- Yield depends on local sample tilt Topography contrast
- Yield depends on local magnetic or electrostatic fields
- Signal is polluted by SE created by BSE in sample – SE2, or on some other surface in specimen chamber (usually final lens) – SE3 noise (information from different part of with different contrast)

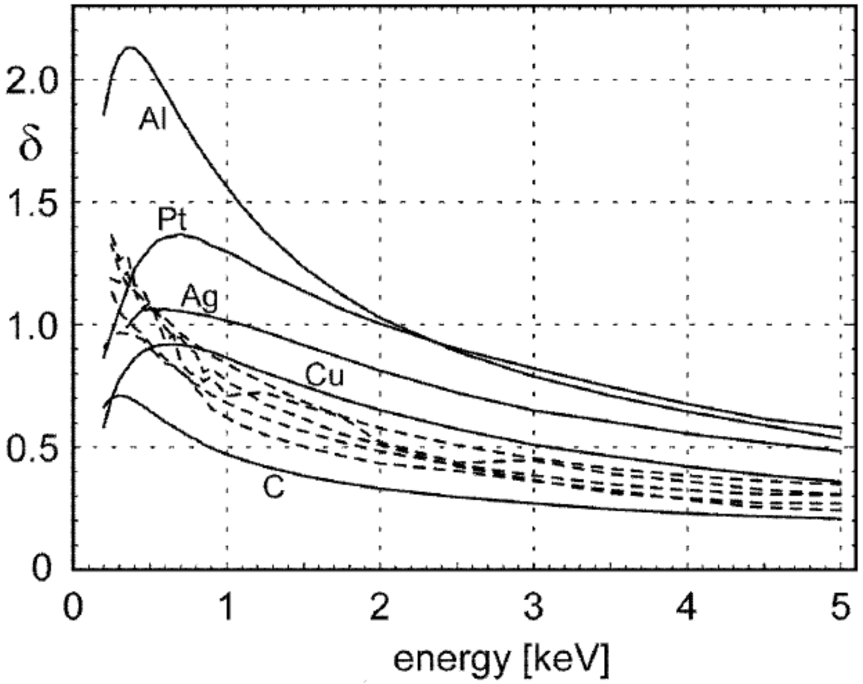






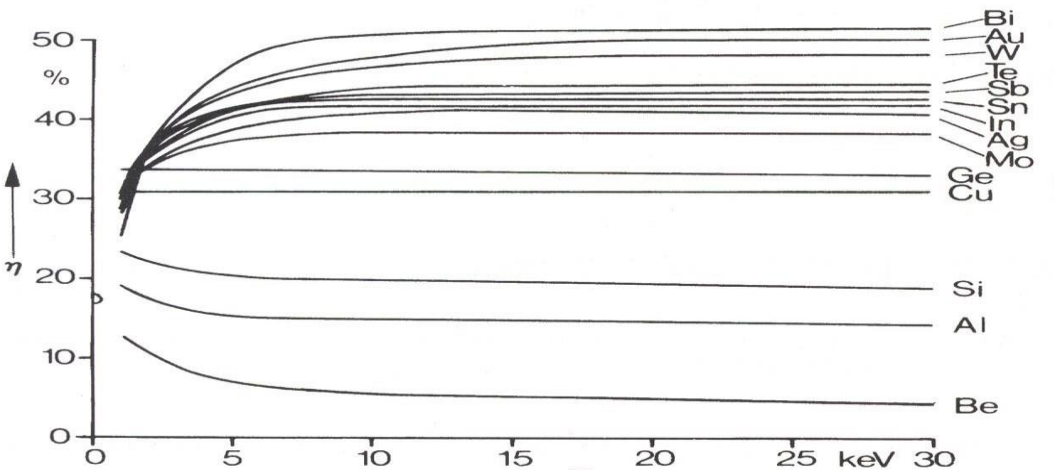
Secondary electrons

- Different yield for different materials □ material contrast
- Yield changes with primary beam energy □ for most materials there is equilibrium point where secondary emission balances primary beam current, i.e. no charging occurs even in case that sample is insulator.



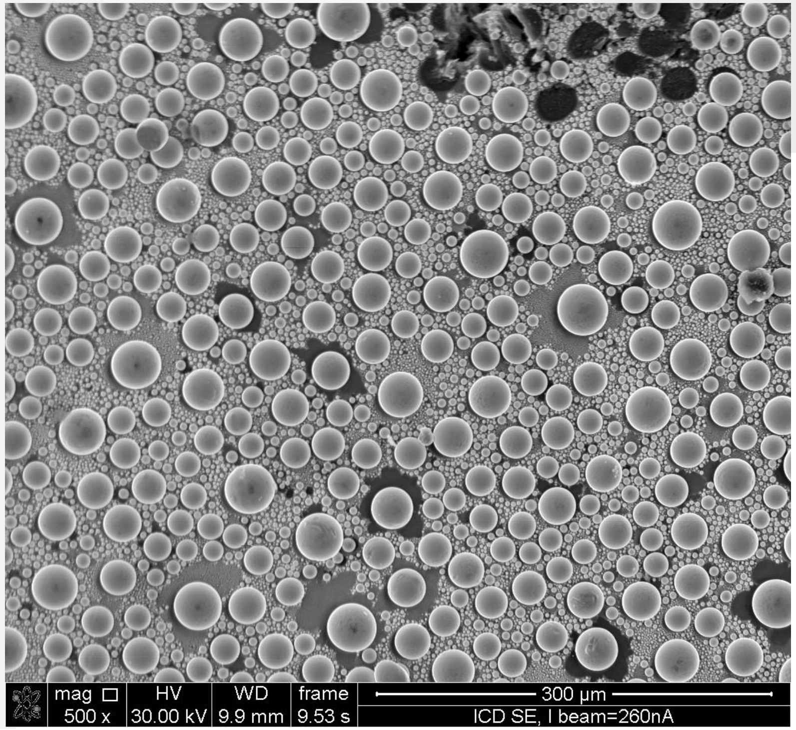
Backscattered electrons

- Primary beam electrons reflected by the sample (elastically or inelastically)
- Yield depends on atomic number of sample material □ low loss BSEs reflected close to beam axis – high take off angle
- Yield depends on local tilt of sample surface □ BSEs reflected far from beam axis – low take off angles
- Yield depends on crystal orientation □ channeling contrast & EBSD(P) = Electron Back Scattered Diffraction (Pattern)

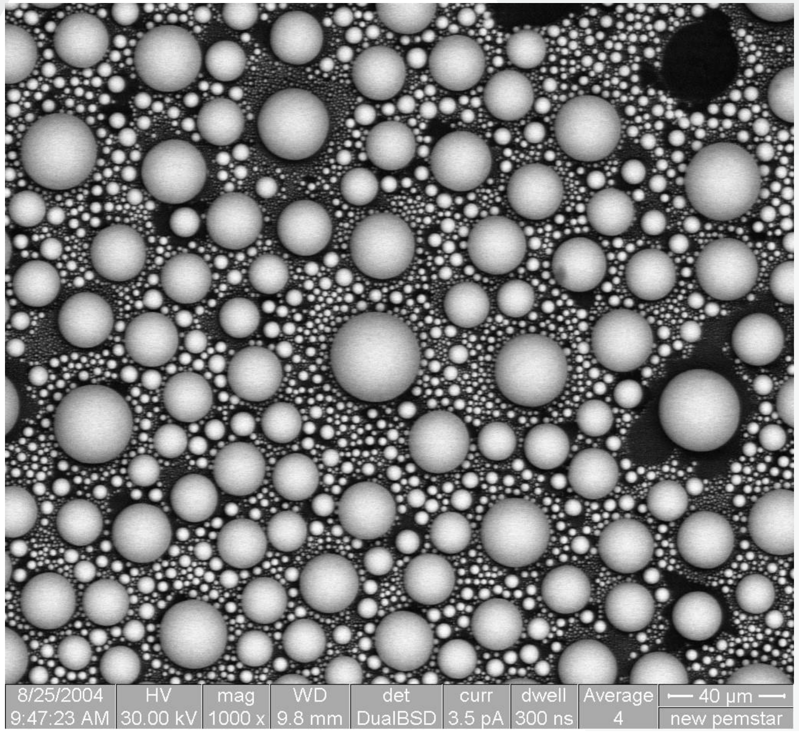


Examples of SE and BSE images

- SE image

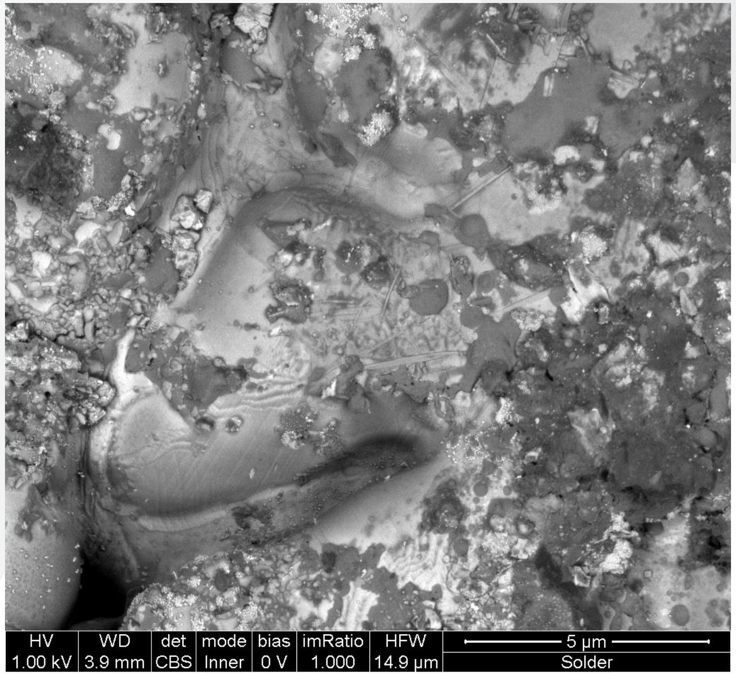


BSE image

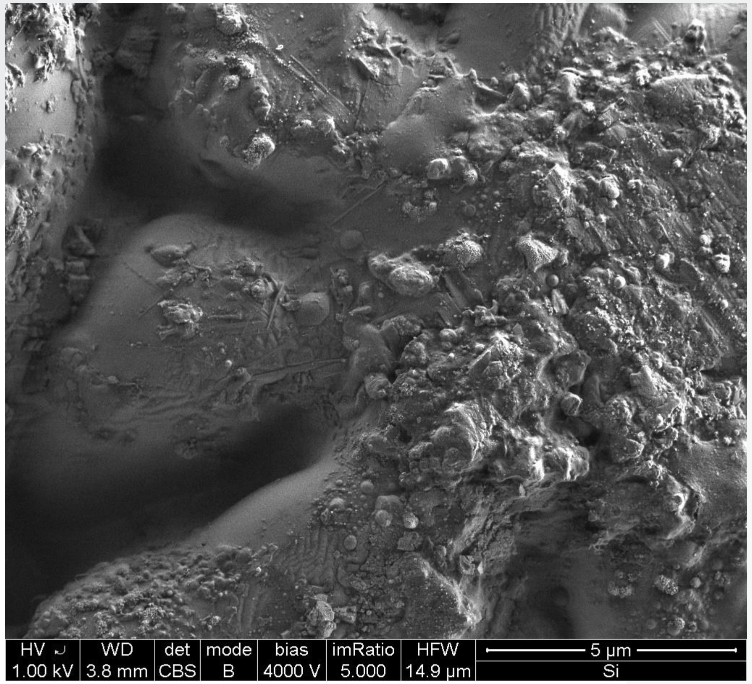


Material & topography contrast in BSE signal

Z- contrast

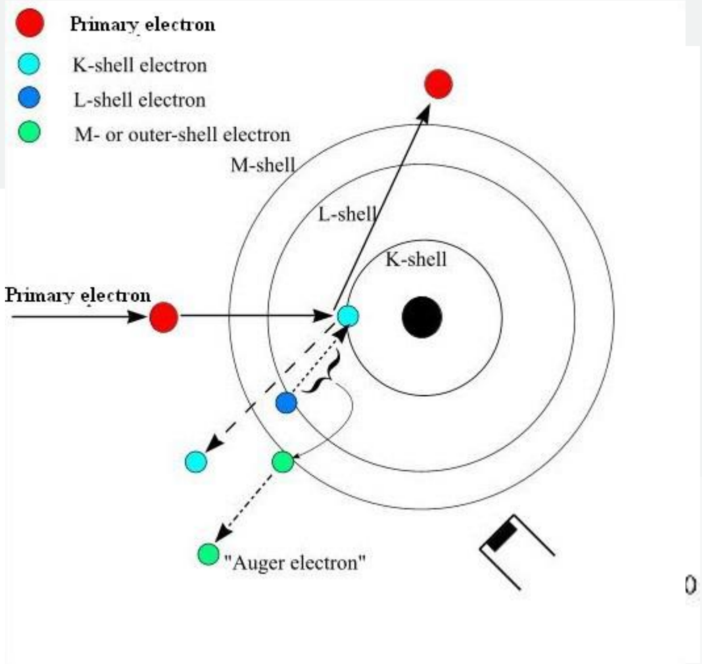


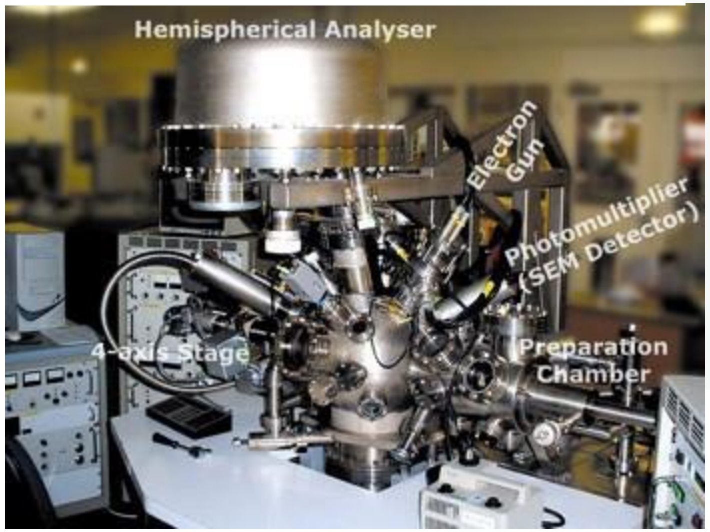
Topography



Auger electrons

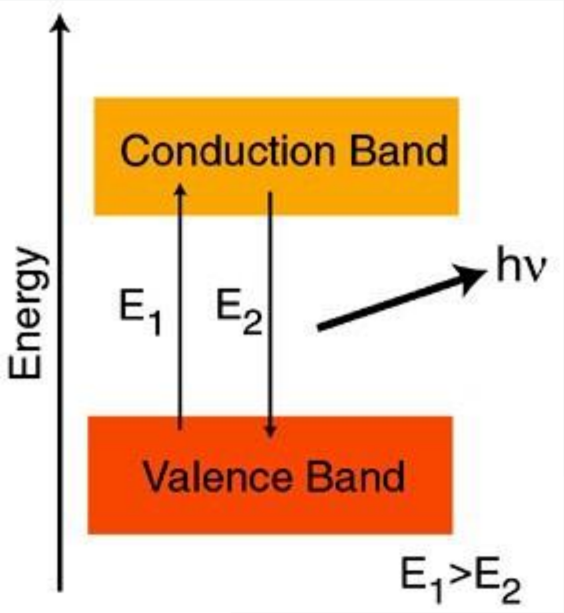
- Transition of electron in atom filling inner shell vacancy results in release of energy
- Energy may be transferred to another electron which is ejected from the atom
- Characteristic peaks for elements – analytical method AES- Auger Electron Spectroscopy
- Low energies (50eV-3keV)-> small escape depth = surface sensitive method
- Extreme surface sensitivity and weakness of signal require usually UHV setup

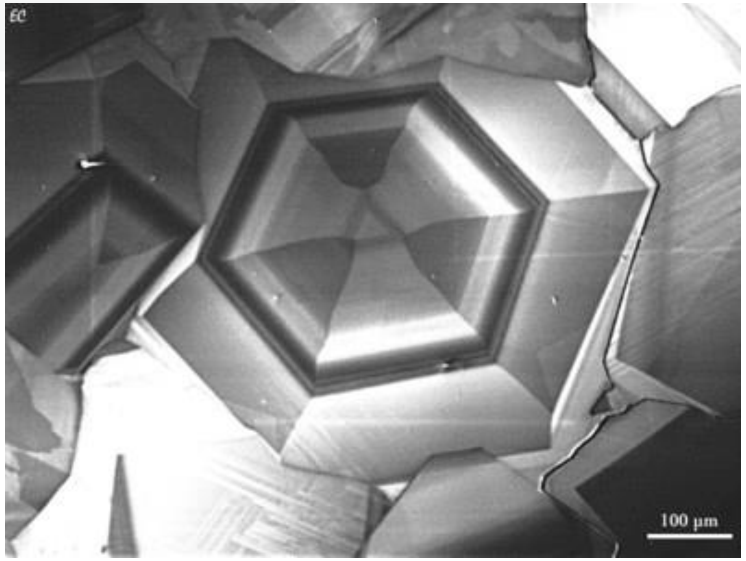


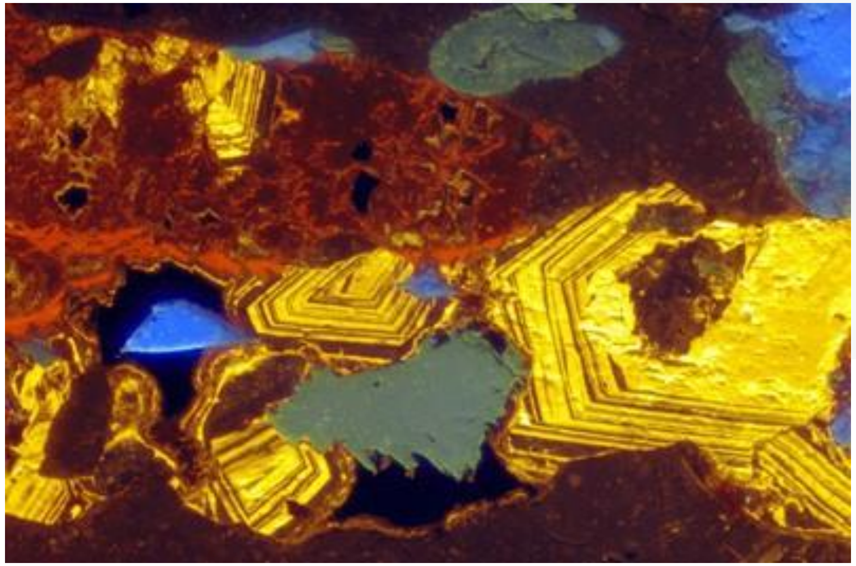


Cathodoluminescence

- UV to IR light (160nm-2000nm) emitted by the sample under electron irradiation
- Effect occurs only in certain materials (semiconductors, minerals, organic molecules)
- Direct detection of light emitted by sample, or more complex instruments with monochromator to obtain spectra of emitted light

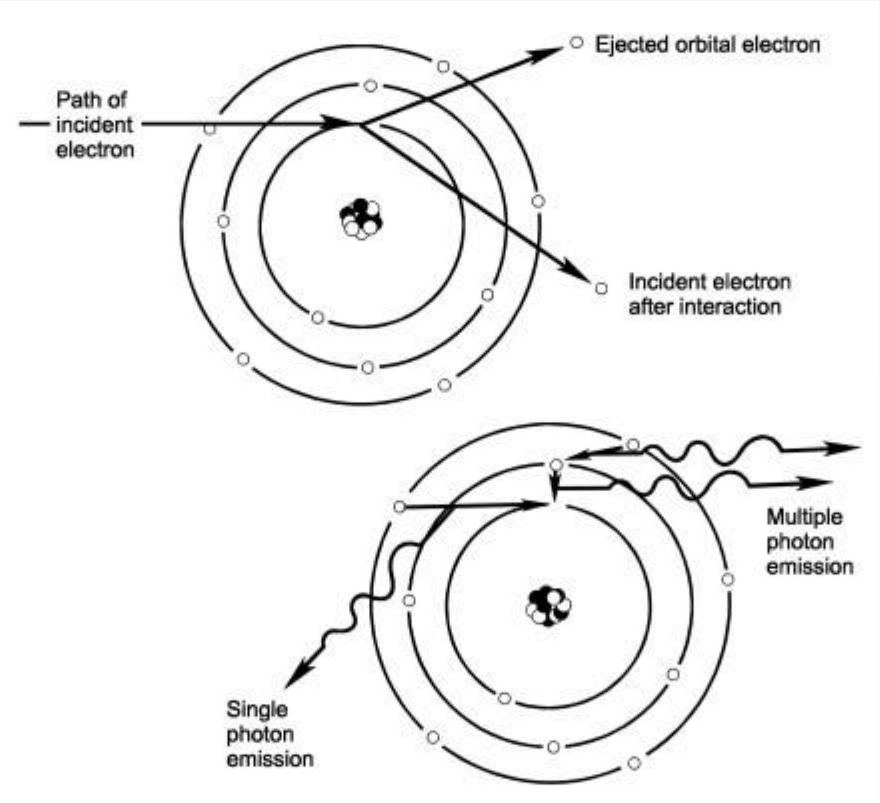


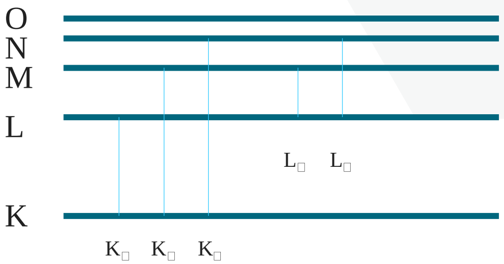




Characteristic X-Ray

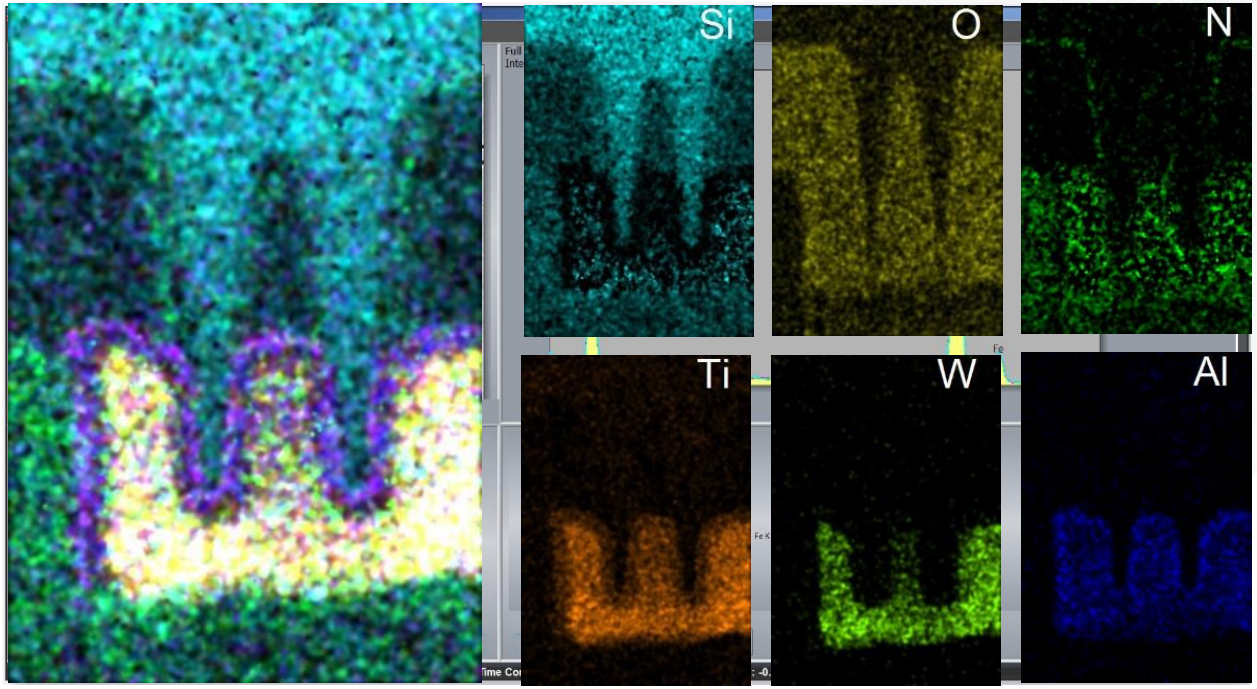
- Electron beam induced emission of X-ray has two components
  - Continuous (“brehmstrahlung”)
  - Characteristic X-ray – dependent on atomic structure of sample
- Peaks of characteristic X-ray corresponds to energy emitted by electron when changing energy levels in atom, thus they enable to determine atomic compound of sample (not chemical structure)





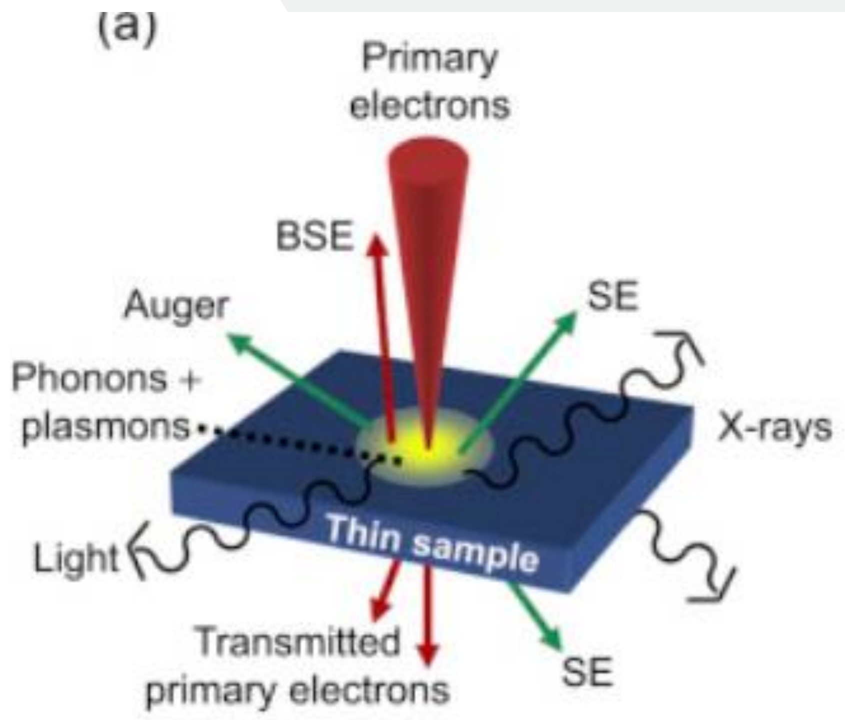
Characteristic X-ray

- EDS or WDS (also EDX, WDX)
  - Energy (Wave) Dispersive Spectroscopy (X-ray)
  - EDS – faster x WDS - more accurate (better energy resolution)
  - X-ray spectra
  - X-ray mapping



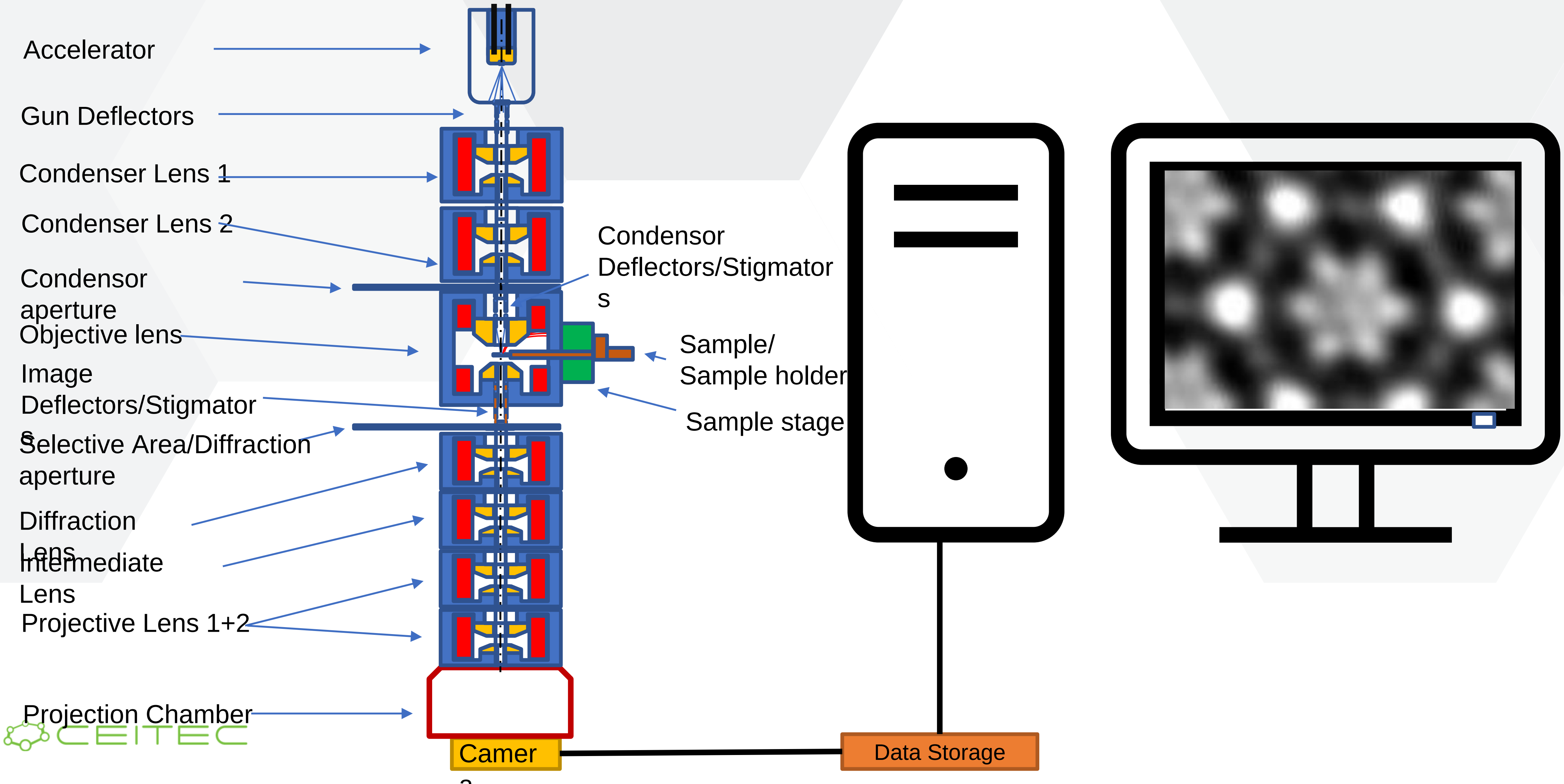
Transmision electron microscopy - TEM

- Electrons transmitted through sample without scattering or scattered to space below sample
- Only possible for samples with thickness smaller than interaction volume
- Electron energy: 30 - 300 keV
- Resolution ~ 0.05nm
- Signal depends on:
  - Sample thickness
  - Sample material
  - Crystal orientation
- Standard imaging - TEM
- Scanning transmission electron microscopy – STEM
- Electron energy loss spectroscopy - EELS



Transmision electron microscopy - TEM

- TEM mode – Image of an illuminated sample is magnified onto a camera
- STEM Mode - Focused Beam scanning over the sample → processed signal creates an image



Accelerator

Gun Deflectors

Condenser Lens 1

Condenser Lens 2

Condensor aperture

Objective lens

Image Deflectors/Stigmators

Selective Area/Diffraction aperture

Diffraction Lens

Intermediate Lens

Projective Lens 1+2

Projection Chamber

Condensor Deflectors/Stigmators

Sample/ Sample holder

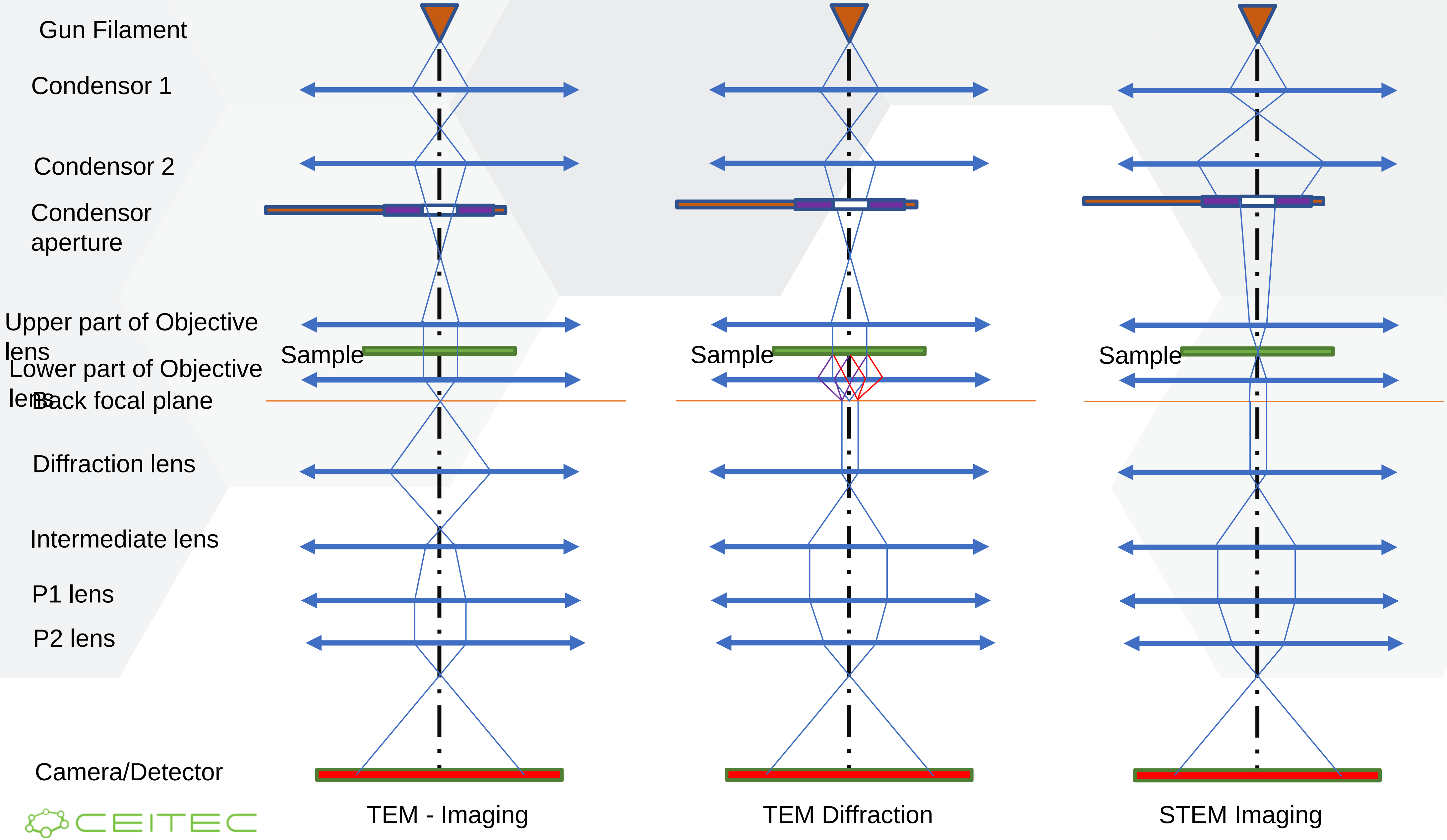
Sample stage

Camera

Data Storage

CEITEC

Transmision electron microscopy – Optical modes



Gun Filament

Condensor 1

Condensor 2

Condensor aperture

Upper part of Objective lens

Sample

Lower part of Objective lens

Back focal plane

Diffraction lens

Intermediate lens

P1 lens

P2 lens

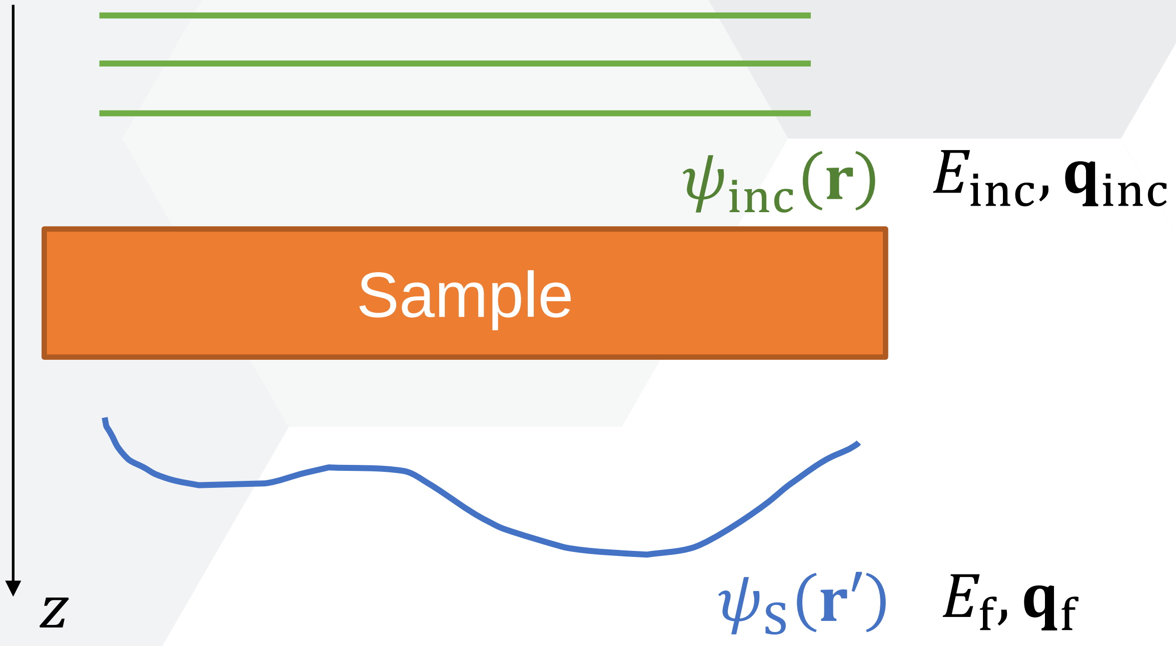
Camera/Detector

TEM - Imaging

TEM Diffraction

STEM Imaging

Transmitted primary electrons



Wavefronts representing incoming electron beam

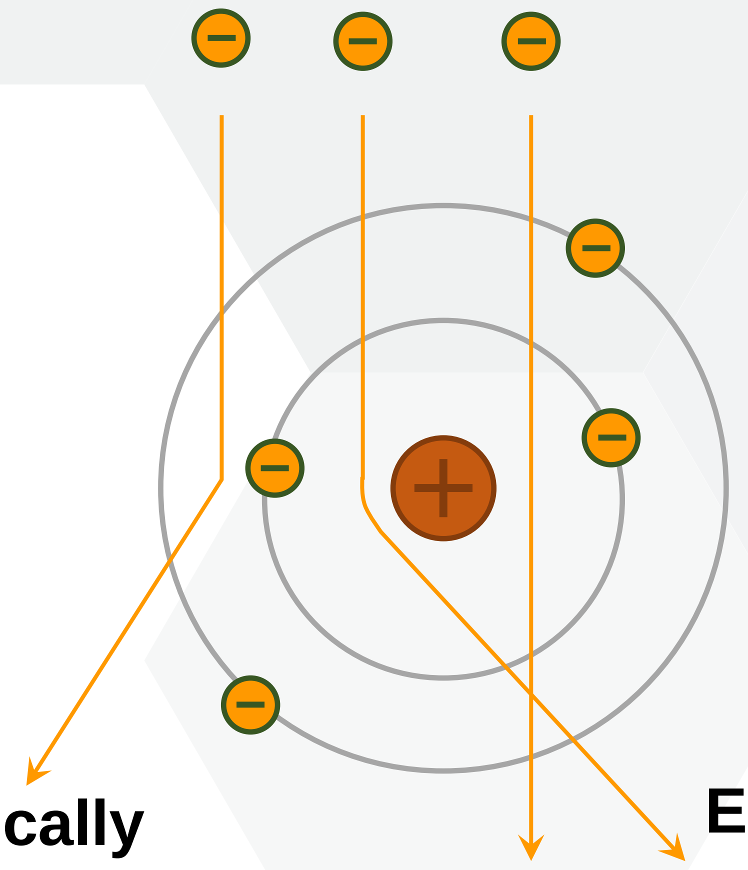
$\psi_{\text{inc}}(\mathbf{r})$ $E_{\text{inc}}, \mathbf{q}_{\text{inc}}$

Sample

z

$\psi_{S}(\mathbf{r}')$ $E_{\text{f}}, \mathbf{q}_{\text{f}}$

Resulting wavefront



Incident electrons

Specimen atom

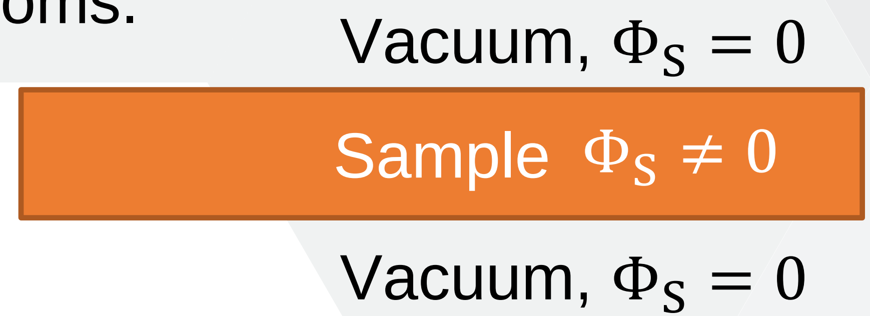
Inelastically scattered electron

Unscattered electron

Elastically scattered electron

Weak-phase object approximation

Suitable for description of thin samples with light atoms.



$$\psi(\mathbf{r}) = \exp(2\pi i z/\lambda)$$

Wave function inside the sample:

$$\psi_S(\mathbf{r}) \approx \exp\left(\frac{2\pi i z}{\lambda_S}\right) \approx \exp\left[\frac{2\pi i z}{\lambda}\left(1 + \frac{e\Phi_S(2m_e c^2 + 2e\Phi)}{2e\Phi(2m_e c^2 + e\Phi)}\right)\right] =$$

$$\exp\left(\frac{2\pi i z}{\lambda}\right)\exp\left[\Phi_S \frac{2\pi i z}{\lambda}\left(\frac{e(m_e c^2 + e\Phi)}{e\Phi(2m_e c^2 + e\Phi)}\right)\right] = \boxed{\exp\left(\frac{2\pi i z}{\lambda}\right)\exp(iz\sigma\Phi_S)}$$

$$\sigma = \frac{me\lambda}{2\pi\hbar^2}$$

Wave function after transmission through the sample:

$$\boxed{\psi_S(\mathbf{r}) \approx \exp\left(\frac{2\pi i z}{\lambda}\right)\exp(i\sigma v_z)}$$

$$v_z(\mathbf{R}) = \int \Phi_S(\mathbf{r})\,dz$$

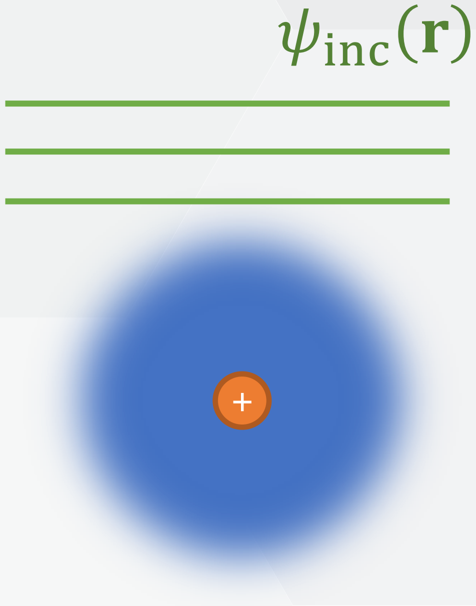
Elastic scattering on a signle atom

Let’s assume that prior to the interaction, the beam is described by a wave function:

$\psi_{\mathrm{inc}}(\mathbf{r})$, which fulfills $H\psi_{\mathrm{inc}} = E\psi_{\mathrm{inc}}$.

The wave function after scattering on an atom:

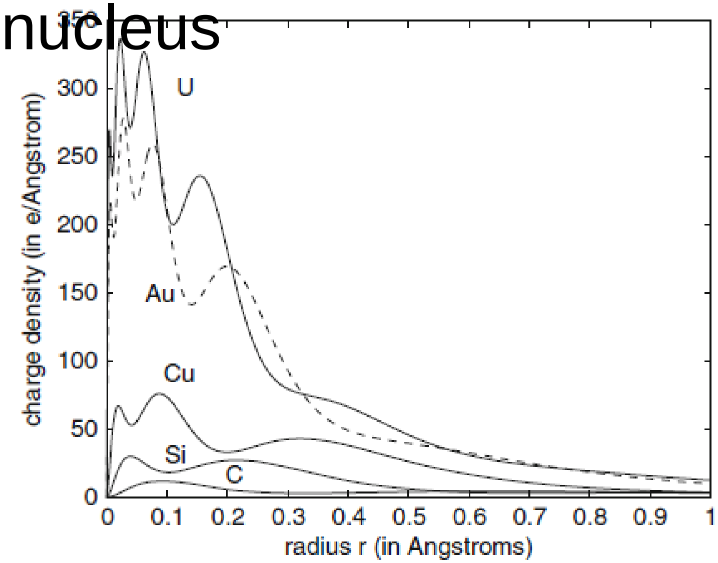
$\psi_S(\mathbf{r}) = \psi_{\mathrm{inc}}(\mathbf{r}) + f(\mathbf{r})$, ψ_S fulfills $\big(H + \Phi(\mathbf{r})\big)\psi_S(\mathbf{r}) = E\psi_S(\mathbf{r})$



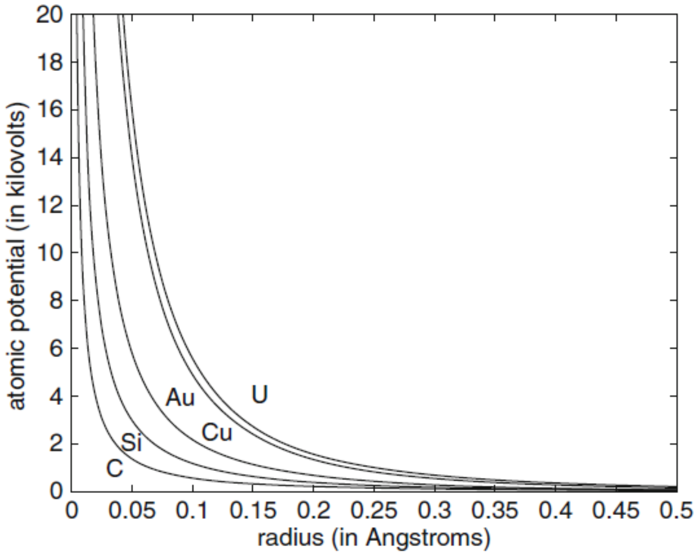
$\psi_{\mathrm{inc}}(\mathbf{r})$

$\psi_S(\mathbf{r}) =?$

Electron density $\rho(|\mathbf{r}|)$ as a function of distance from nucleus



Interaction potential $\Phi(|\mathbf{r}|)$



Ref.: Kirkland

Elastic scattering on a signle atom

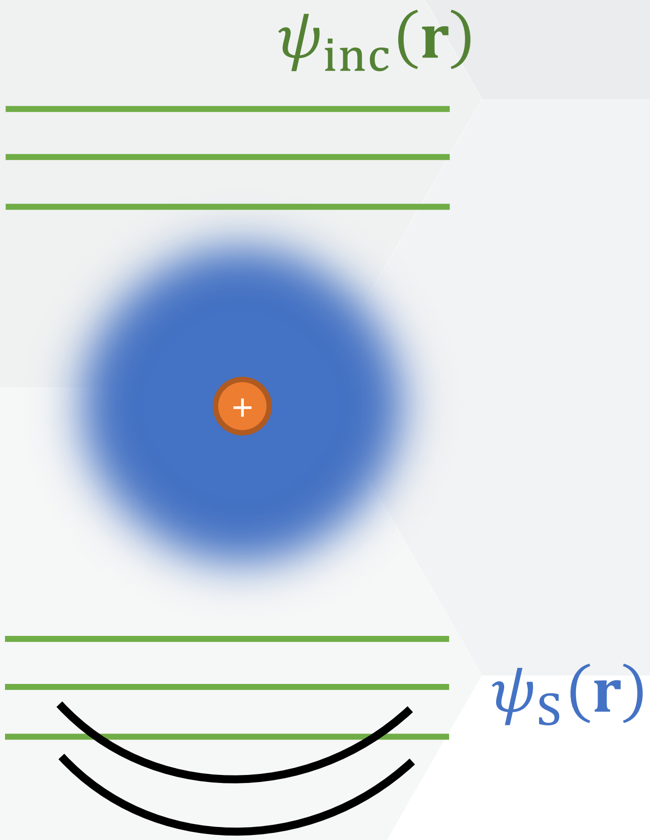
Final electron wave function after the interaction with an atom:

$$\boxed{\psi_\mathrm{S}(\mathbf{r}) = \psi_\mathrm{inc}(\mathbf{r}) + f_e(q)\,\frac{\exp(\mathrm{i}\,\mathbf{q}\cdot\mathbf{r})}{r}}$$

Scattering cross section:

$$\sigma = \frac{me\lambda}{2\pi\hbar^2}$$

$$f_e(q) = \frac{2\pi \mathrm{i}}{\lambda}\int_0^\infty J_0(qr)\left\{1 - \exp\left[\mathrm{i}\sigma\int \Phi(\mathbf{r})\mathrm{d}z\right]\right\} r\,\mathrm{d}r$$



For acquiring an image, we propagate $\psi_\mathrm{S}(\mathbf{r})$ through an electron-optical system:

$$I_\mathrm{detector} \propto \left|\mathrm{FT}^{-1}\{\psi_\mathrm{S}(\mathbf{Q})\ \mathrm{TF}(\mathbf{Q})\}\right|^2$$

Elastic scattering on a signle atom

Final electron wave function after the interaction with an atom:

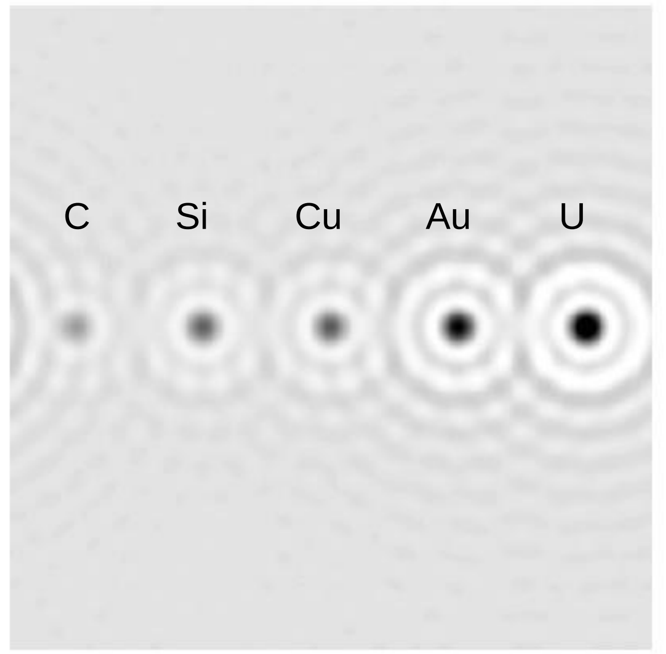
$$\boxed{\psi_S(\mathbf{r}) = \psi_{\mathrm{inc}}(\mathbf{r}) + f_e(q)\frac{\exp(\mathrm{i}\,\mathbf{q}\cdot\mathbf{r})}{r}}$$

Scattering cross section:

$$\sigma = \frac{me\lambda}{2\pi\hbar^2}$$

$$f_e(q) = \frac{2\pi\mathrm{i}}{\lambda}\int_0^\infty J_0(qr)\left\{1 - \exp\left[\mathrm{i}\sigma\int \Phi(\mathbf{r})\mathrm{d}z\right]\right\} r\,\mathrm{d}r$$

Calculation for $\psi_{\mathrm{inc}} \propto \exp(\mathrm{i}\,2\pi z/\lambda)$
200 keV electrons
(Kirkland; Advanced computing in EM)

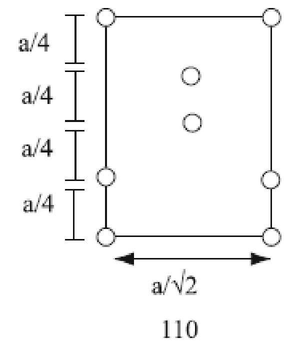


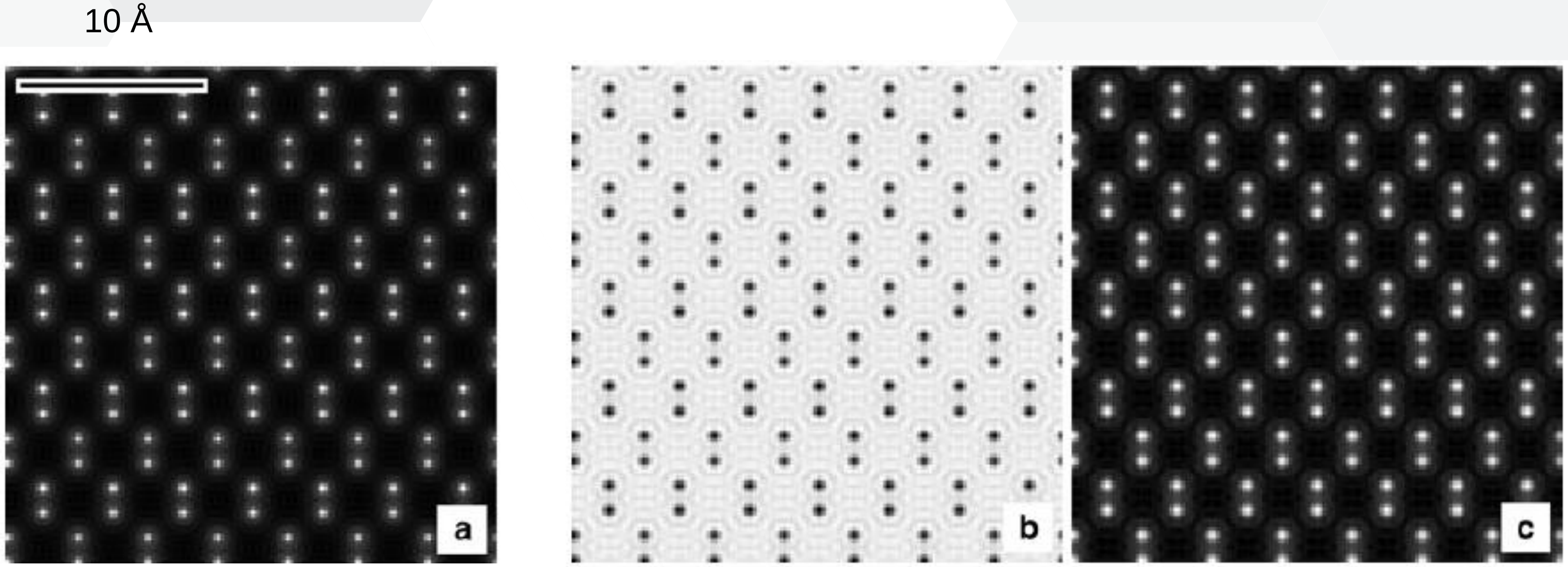
Weak-phase object approximation: Si lattice

The simplest approximation: superposition of potentials of independent atoms.

$$\Phi_S(\mathbf{r}) = \sum_{j=1}^{N} \Phi_j(\mathbf{r}) \qquad \psi_S(\mathbf{r}) \approx \exp(i\sigma \int \Phi_S(\mathbf{r}) dz) \exp(i\, 2\pi z/\lambda)$$

Example: Si lattice





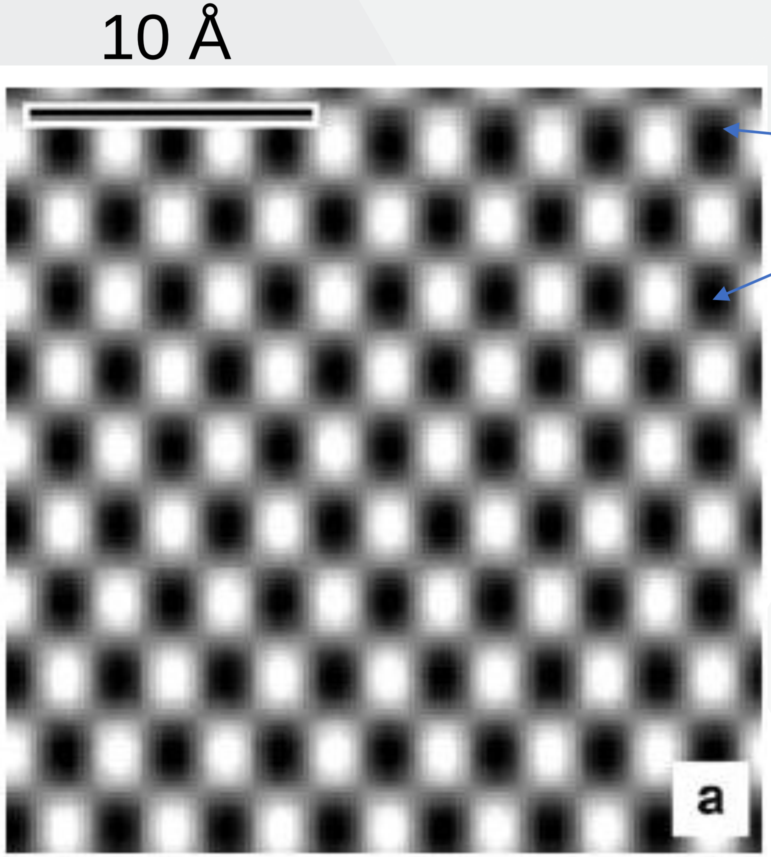
$\int \Phi_S(\mathbf{r}) dz$ $\quad$ $\mathrm{Re}\{\exp(i\sigma \int \Phi_S(\mathbf{r}) dz)\}$ $\quad$ $\mathrm{Im}\{\exp(i\sigma \int \Phi_S(\mathbf{r}) dz)\}$

Kirkland; Advanced computing in EM

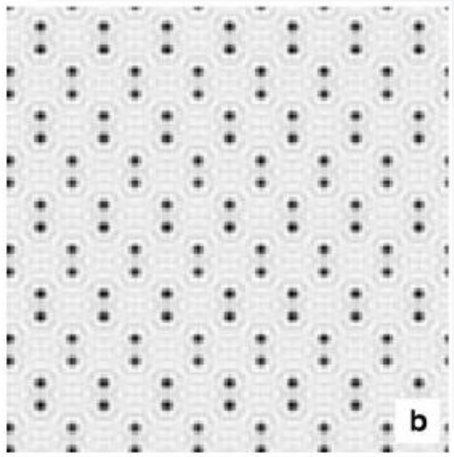
Weak-phase object approximation TEM: Si lattice

$$I_{\text{detector}} = \int \mathrm{d}^2\mathbf{R}_{\text{det}} \left|\psi_{\text{prop}}(\mathbf{R}_{\text{det}})\right|^2$$

Imaging in TEM (simulation):



Here approximately pairs of Si atoms



$$\mathrm{Re}\left\{\exp\left(\mathrm{i}\sigma \int V_S(\mathbf{r})\,\mathrm{d}z\right)\right\}$$

Kirkland; Advanced computing in EM

Thick sample

$$\left[-\frac{\hbar^2}{2m}\nabla^2 - e\,\Phi(\mathbf{r})\right]\psi_{\mathrm{tot}}(\mathbf{r}) = E\psi_{\mathrm{tot}}(\mathbf{r})$$

$\psi_{\mathrm{tot}}(\mathbf{r}) = \psi(\mathbf{r})\exp(\mathrm{i}\,2\pi z/\lambda)$ Slowly oscillating term * quickly oscillating term

$\left[\nabla_{\mathbf{R}}^2 + \frac{4\pi\mathrm{i}}{\lambda}\frac{\partial}{\partial z} + \frac{2\,m\,e\,\Phi(\mathbf{r})}{\hbar^2}\right]\psi(\mathbf{r}) \approx 0$ „Paraxial Schrödinger equation“

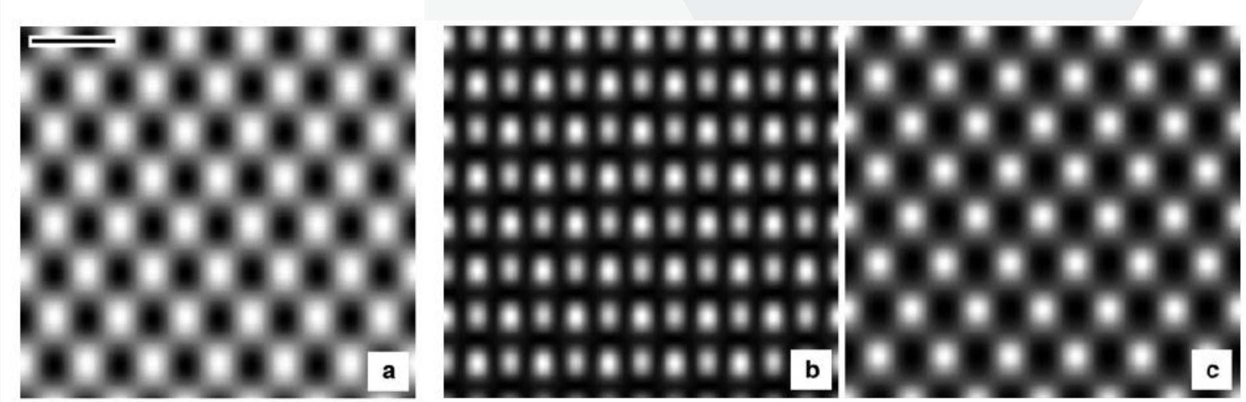
For a periodic crystal: $\Phi(\mathbf{r}) = \sum_{\mathbf{G}} \Phi_{\mathbf{G}} \exp(2\pi\mathrm{i}\,\mathbf{G}\cdot\mathbf{r})$

➡ $\psi(\mathbf{r}) = \sum_{\mathbf{G}} \psi_{\mathbf{G}}(z)\exp(2\pi\mathrm{i}\,\mathbf{G}\cdot\mathbf{r})$

Thick sample: GaAs

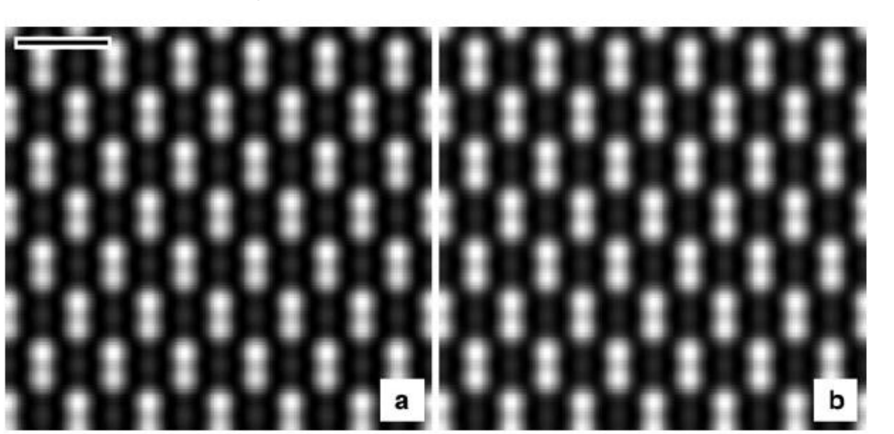
TEM (plane wave on a sample)

5 Å 2 layers 20 layers 100 layers



STEM (focused beam on a sample)

20 layers 100 layers



Contrast reversal!

Image Simulation SW

TEM: **JEMS**
https://www.jems-swiss.ch/

Inelastic mean-free path and thickness dependance

- Scattering is quite improbable process; subsequent scattering events can be considered as independent → Poisson statistics
- Probability that an electron experiences n scattering events after travelling distance z inside the sample:

$$p_n(z) = \frac{1}{n}\left(\frac{z}{\Lambda}\right)^n \mathrm{e}^{-z/\Lambda} \longleftarrow$$

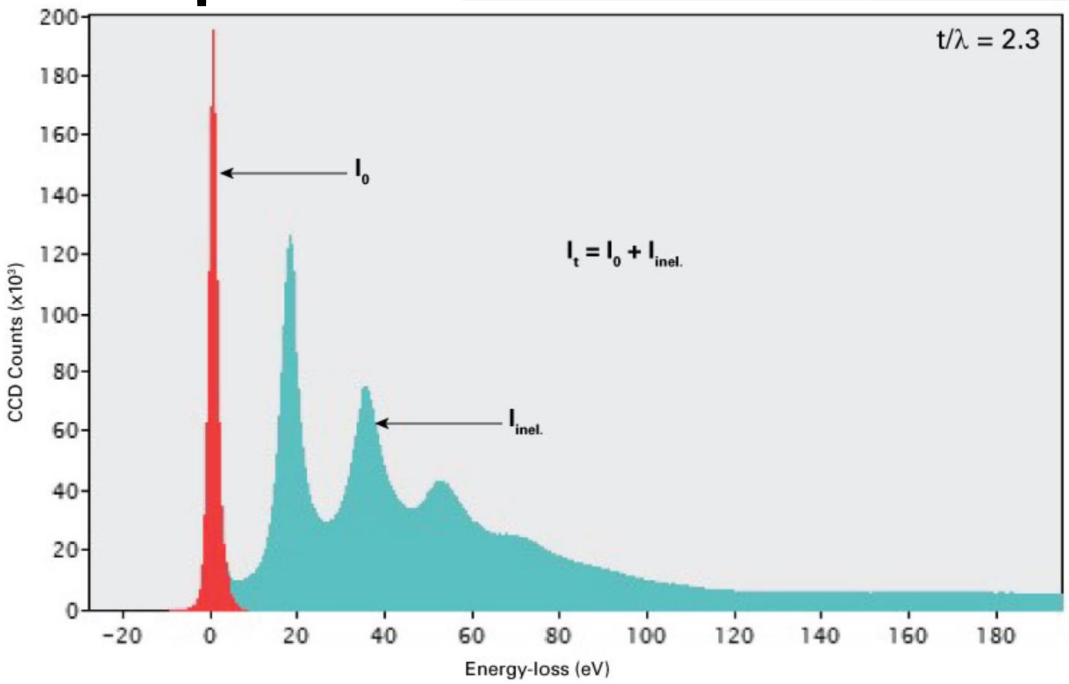
Inelastic mean free path

- Intensity of the EEL spectrum:

$$I_{\mathrm{tot}} = I_0 + I_{\mathrm{inel}}$$
$$= I'(p_0 + 1 - p_0)$$
$$= I_0 + I_0\mathrm{e}^{z/\Lambda}\left(1 - \mathrm{e}^{-z/\Lambda}\right)$$

$$\frac{I_{\mathrm{tot}}}{I_0} = \mathrm{e}^{z/\Lambda}$$

$$\boxed{\ln\frac{I_{\mathrm{tot}}}{I_0} = z/\Lambda}$$



eels.info

Inelastic mean-free path

$$\Lambda = 1/(n\,\sigma_{\text{inel}})$$

Number of atoms per unit volume

Inelastic scattering cross section

$$1/\Lambda = \frac{1}{\pi a_0 v^2}\left[A \ln\left(\frac{2v^2}{I}\right) - \frac{7C}{2v^2}\right]$$

Bohr radius

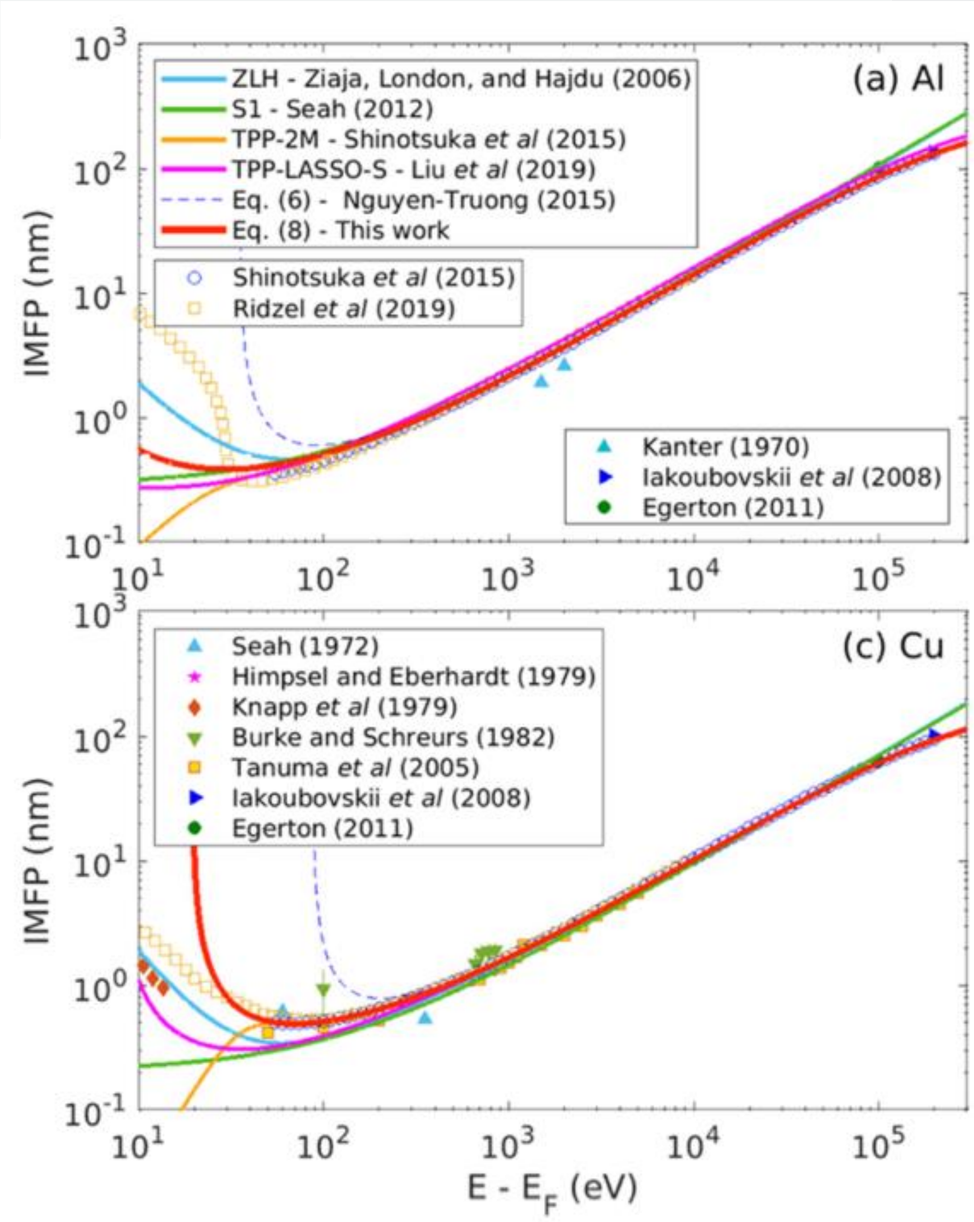
Electron velocity

Material-dependent constants:

$$A = \int_0^\infty \mathrm{Im}\left[\frac{-1}{\varepsilon(\omega)}\right] \mathrm{d}\omega$$

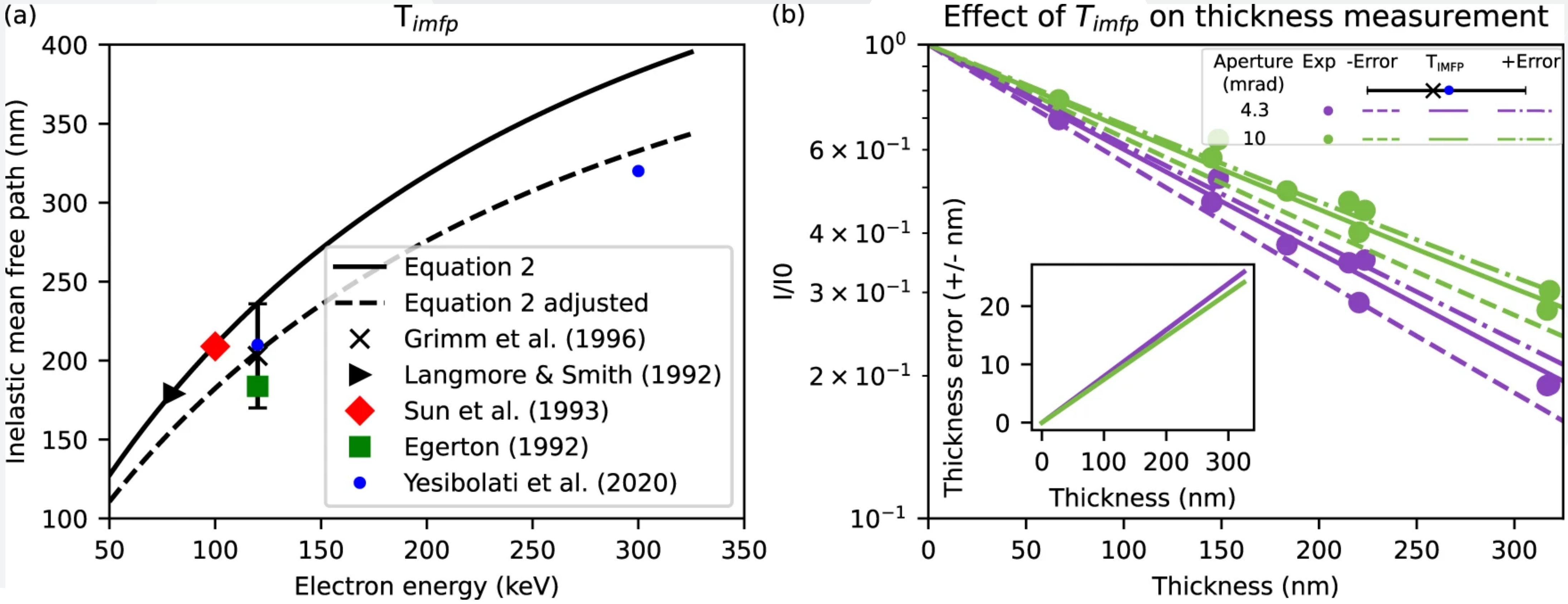
$$A \ln(I) = \int_0^\infty \mathrm{Im}\left[\frac{-1}{\varepsilon(\omega)}\right] \ln(\omega)\, \mathrm{d}\omega$$

$$C = \int_0^\infty \mathrm{Im}\left[\frac{-1}{\varepsilon(\omega)}\right] \omega\, \mathrm{d}\omega$$



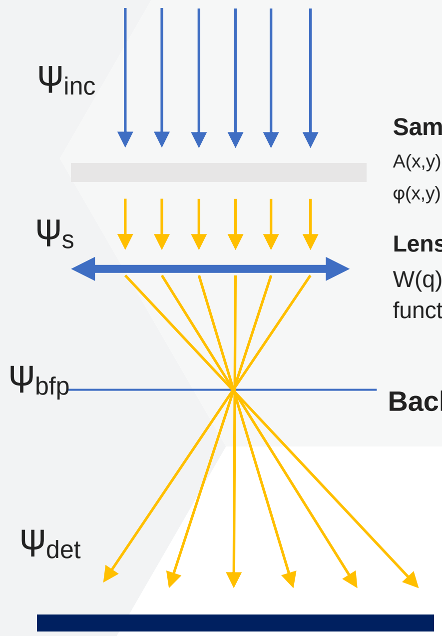
Le and Nguyen-Truong, J. Phys. Chem. C **2021** *125* (34), 18946

Inelastic mean-free path in ice



H. Bronw: MeasureIce: accessible on-the-fly measurement of ice thickness in cryo-electron microscopy

Transfer of Image through the optical system



Sample
A(x,y) – absorbtion
φ(x,y) – phase shift

Lens
W(q) – phase aberration function (lens properties)

Back focal plane (bfp)

Detector

DQE

I(R) - Intensity

Incoming Wave $\psi_{inc}(r)$

Sample Amplitude Influence $A(r)$

Sample Phase Influence $\varphi(r) = f_e(q)$

Exit Wave $\psi_s(r) = A(r)\psi_{inc}(r)e^{i\varphi(r)}$

when $A(r) \ll 1$ and $\varphi(r) \ll 1, \varepsilon(r) = lnA(r)$

And assumption $\psi_{inc}(r) = 1$ $(parallel\ illumination)$

Exit Wave $\psi_s(r) = \psi_{inc}(r)[1 + \varepsilon(r) + i\varphi(r)]$

$$\psi_{bfp}\,(q) = FT\psi_s(r)$$

$$\psi_{bfp}\,(q) = \delta(q) + E(q) + i\Phi(q)$$

Aberrations addition $W(q) = \frac{\pi}{2}(C_s q^4\lambda^3 + \Delta f q^2\,\lambda\,)$

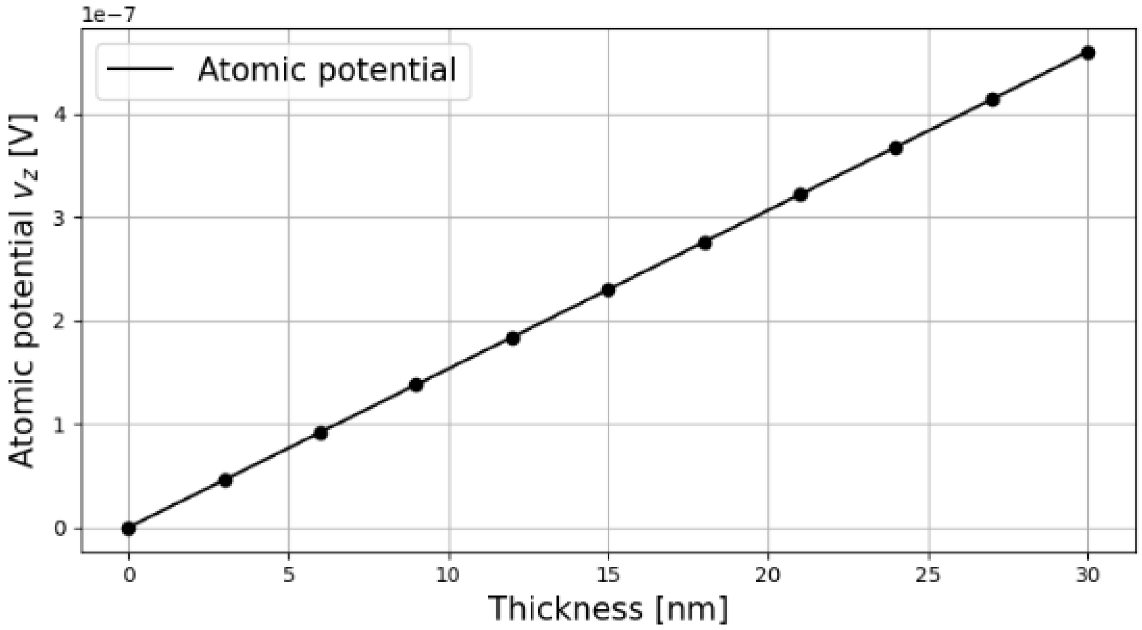
$$\psi_{bfp,ab}\,(q) = \delta(q) + E(q)e^{-iW(q)} + i\Phi(q)e^{-iW(q)}$$

Optical Intensity at Image Plane

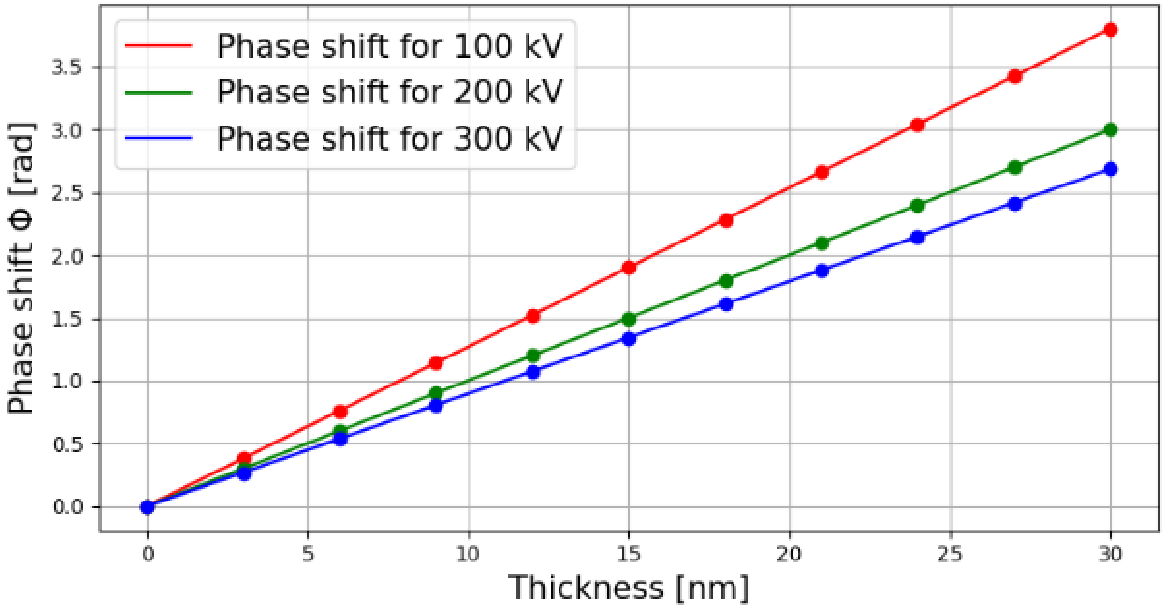
$$I(R) = |\psi_m(Rd_{et})|^2 = FT\psi_{bfp,ab}\overline{FT\psi_{bfp,ab}}$$

$$I(R) = |\psi_m(Rd_{et})|^2 = E_t * \{1 - 2\varphi(Q)\sin\big(W(Q)\big) + 2\varepsilon(Q)\cos(W(Q)\}$$

Phase shift – Carbon sample



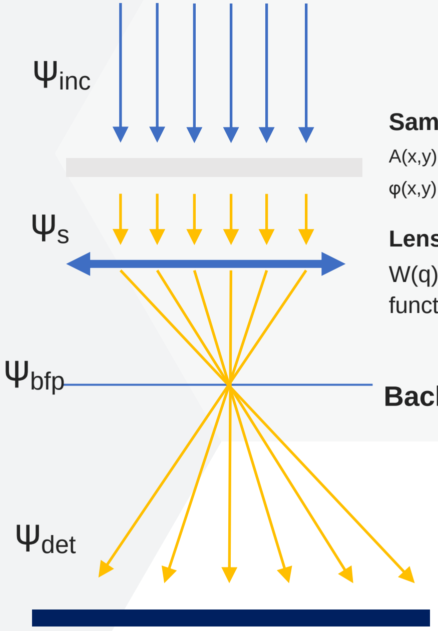
(a) Atomic potential of amorphous carbon.



(b) Phase shift of amorphous carbon for different accelerating voltages.

Michal Brzica bachelor thesis – derived from RICOLLEAU, C., et al. Random vs realistic amorphous carbon models for high resolution microscopy and electron diffraction. Journal of Applied Physics, 2013, 114.21: 213504. ISSN 0021-8979. Available from DOI: 10.1063/1.4831669.

Contrast Transfer Function



Sample
A(x,y) – absorbtion
φ(x,y) – phase shift

Lens
W(q) – phase aberration function (lens properties)

Back focal plane (bfp)

Detector
DQE
I(R) - Intensity

$$I(R) = |\psi_m(Rd_{et})|^2 = FT\psi_{bfp,ab}\overline{FT\psi_{bfp,ab}}$$

$$I(R) = |\psi_m(Rd_{et})|^2 = \{1 - 2\varphi(Q)\sin\big(W(Q)\big) + 2\varepsilon(Q)\cos(W(Q)\}$$

Contrast Transfer Function (CTF)

- Describing optical property of TEM

$$\mathrm{CTF}(\vec{q}\,')=E_\mathrm{t}(q')E_\mathrm{s}(\vec{q}\,')E_\mathrm{d}(\vec{q}\,')E_\mathrm{u}(\vec{q}\,') \cdot \mathrm{Intenzita}(\vec{q}\,') \in \langle -1; 1\rangle$$

where

$E_\mathrm{t}(q')$ - *temporal coherency*
$E_\mathrm{s}(\vec{q}\,')$ - *spatial coherency*
$E_\mathrm{d}(\vec{q}\,')$ - drift impact
$E_\mathrm{u}(\vec{q}\,')$ - *vibration dumping*

Observed Intensity on PC

CTF is not seen directly on our PC!

$$\mathrm{Intensity}_{\mathrm{ob}}(\vec{r'}) = \mathrm{I}_{\mathrm{rn}} + \mathrm{I}_{\mathrm{dc}} + \mathrm{CF} \cdot \mathrm{IFT}\left[\mathrm{FT}\left[P_{\mathrm{oiss}}\left(\Phi_{\mathrm{e}} \cdot \mathrm{IFT}^{-1}\left[\mathrm{CTF}_{\mathrm{optical}}(\vec{q'})\sqrt{\mathrm{DQE}(\vec{q'})}\right]\right)\right] \cdot \mathrm{NTF}(\vec{q'})\right]$$

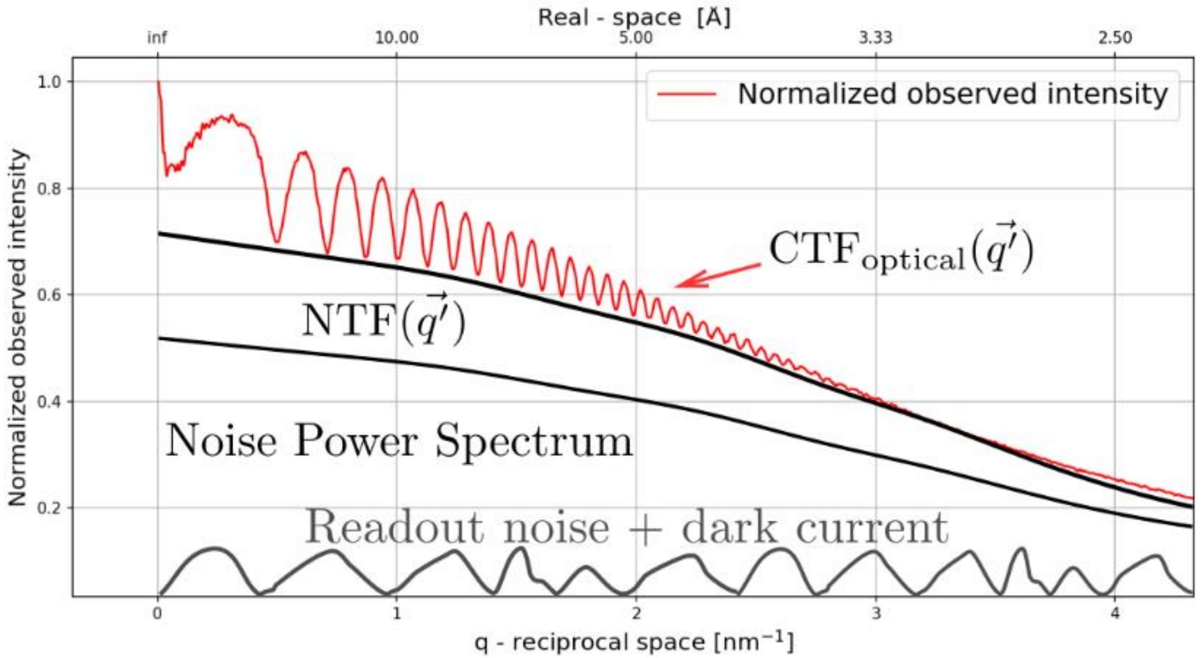


Figure 4.2: Scheme of the normalized observed intensity.

I_{rn} – Read-out noise
I_{dc} – dark current
CF – Conversion ration e/signal
Φ_{e} – Primary electron number
CTF – Contrast Transfer Function
DQE – Detector Quantum Efficiency
NTF – Noise Transfer Function

Michal Brzica bachelor thesis – derived VULOVIĆ, Miloš, et al. Image formation modeling in cryo-electron microscopy. Journal of structural biology, 2013, 183.1: 19-32. ISSN 1047-8477. Available from DOI: 10.1016/j.jsb.2013.05.008.

Conclusion

Electrons are powerful imaging particle

Undertading of imaging/interaction principles is the key for understanding of imaged data

Next – Design of Transmission Electron Microscopes