Scanning Techniques in Electron Microscopy

-Scanning Transmission Electron Microscopy (STEM)-

Berlin, Nov. 15th 2013

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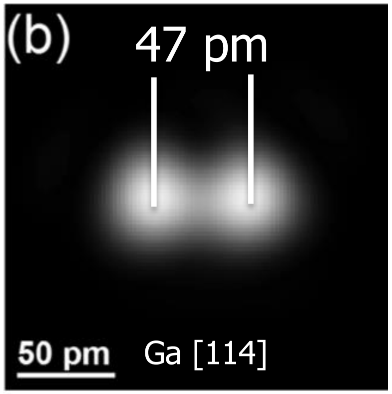
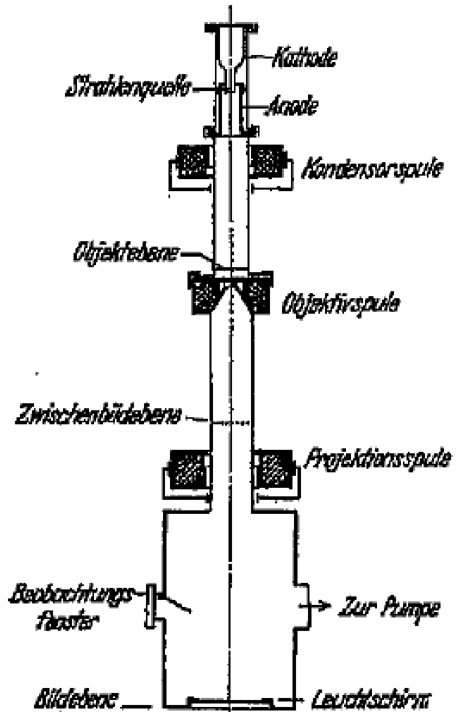
History

- 1897: Thompson – Discovery of the Electron
- 1925: de Broglie – Wave Nature of the Electron
- 1926: Bush – Magnetic/Electric Fields as Lenses
- 1931: Knoll and Ruska – 1st TEM built
- 1938: von Ardenne – 1st STEM built
- 1939: von Borries and Ruska – 1st Commercial TEM
  ~10nm resolution
- 1945: 1.0 nm resolution
- 1965: 0.2 nm resolution
- 1968: Crewe – 1st STEM with field emmission gun
  ~0.3 nm resolution
- 1999: < 0.1 nm resolution
- 2009: 0.05 nm resolution

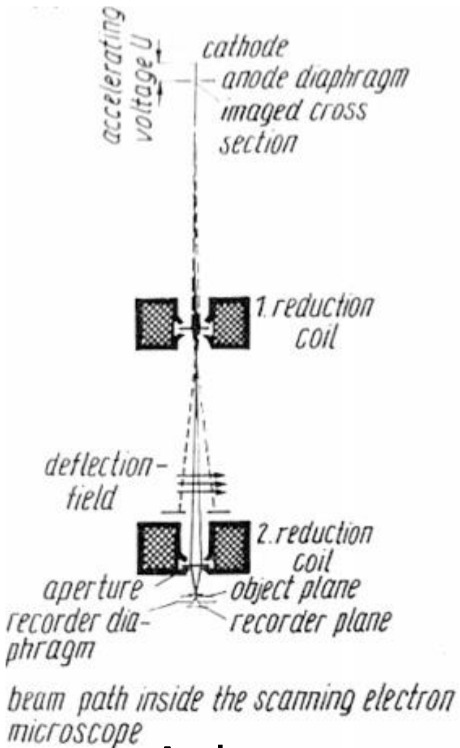


Erni et al. *Phys.Rev.Lett.* **2009**, 102, 096101.
Zaluzec Intorduction to Transmission/Sanning Transmission Electron Microscopy and Microanalysis

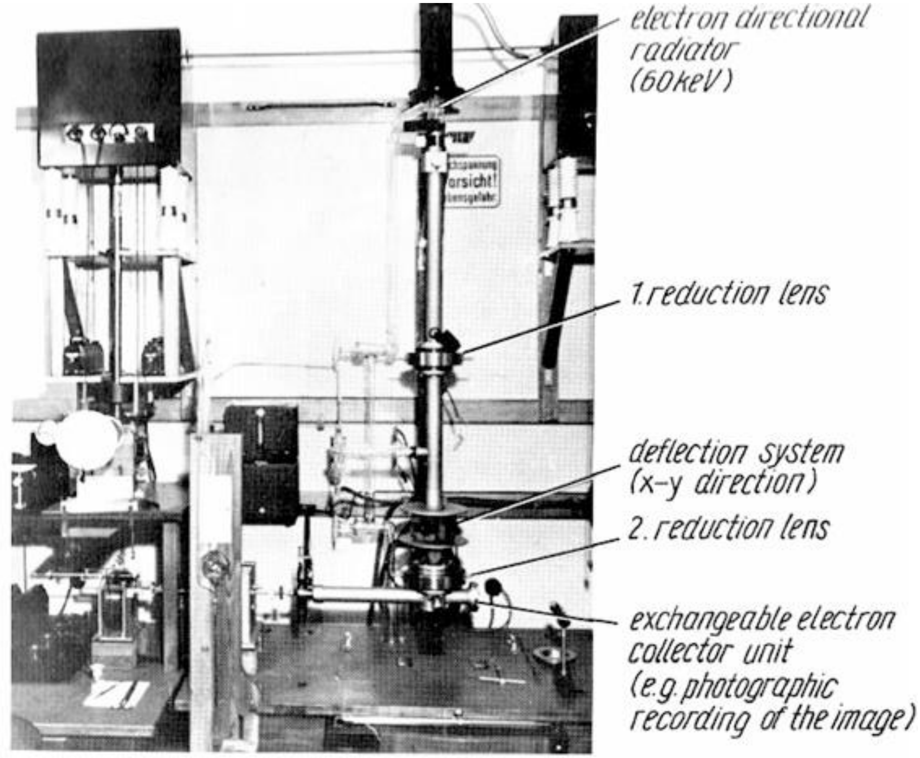
History

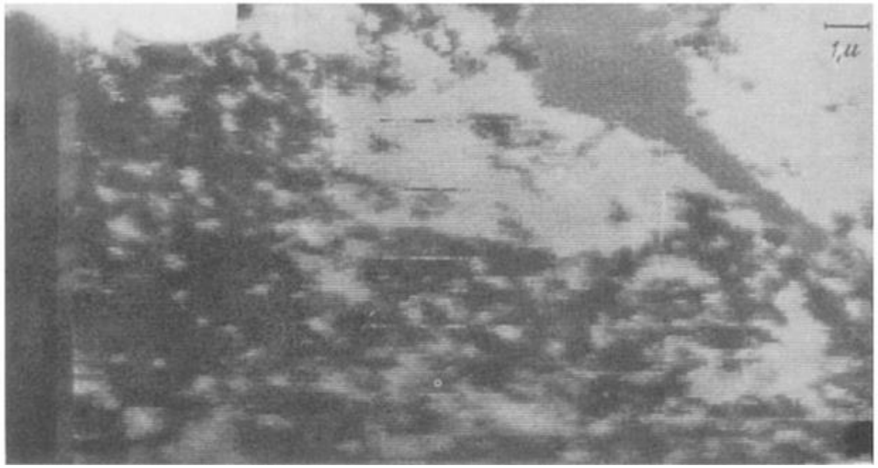


Ruska



von Ardenne





Knoll et al. *Z.Physik* **1932**, 76, 649-654.
v.Ardenne *Z.Tech.Physik* **1938**, 19, 4ß7-416.

Aim of the talk

- STEM is a very powerful and versatile instrument for atomic resolution imaging and nanoscale analysis

→What is STEM?

→What experiments can be done?

→What are the principles of operation?

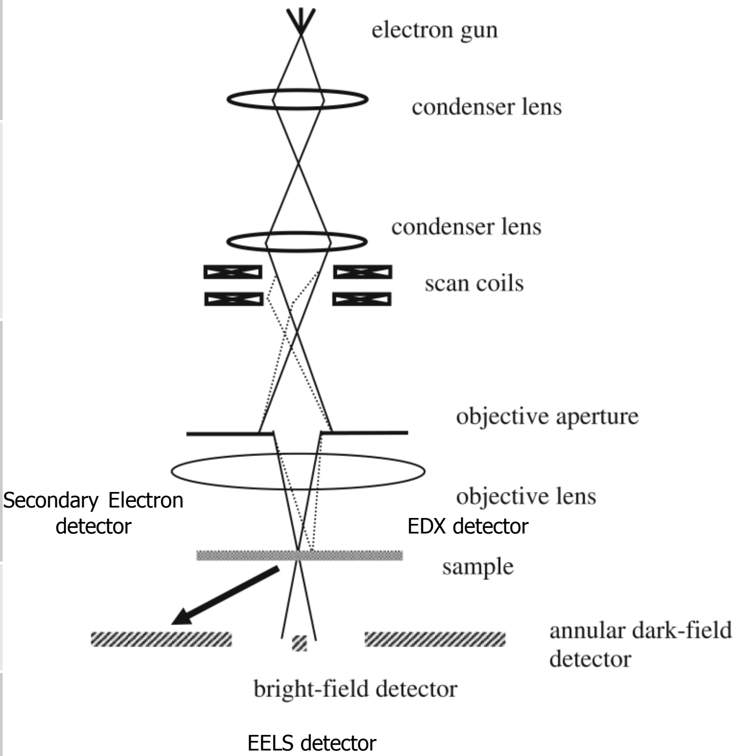
→What are limiting factors in performance?

Outline

- Principles of STEM
- STEM Probe
- Ronchigram
- Detectors
- Incoherent vs. Coherent Imaging
- Examples
- Literature

STEM vs. SEM

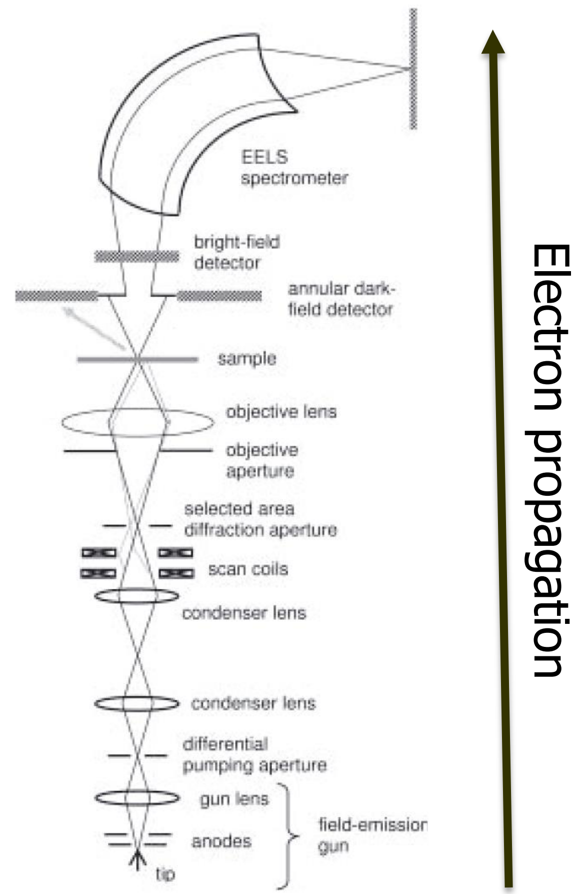
Similiarities	Differences
Electron gun →generates electron beam	SEM: bulk sample → Back scatterd/secondary electrons are detected
Lens system → Forms image of electron source at the specimen	STEM: electron transparent specimen → Detectors are placed after the sample
Electron spot (probe) can be scanned over the sample in a raster pattern → Exciting scanning deflection coils	
Scattered electrons are detected	
Image: Intensity plotted as a function of probe position	



Pennycook et al. Scanning Transmission Electron Microscopy **2012**

Principles of STEM

Historical: Dedicated STEM machines have electron gun at the bottom
(stability reason due to heavy UHV pumps)
→ electrons travel upwards



Pennycook et al. Scanning Transmission Electron Microscopy **2012**

Principles of STEM

Modern: Combined Conventional TEM (CTEM) and STEM instruments
CTEM coloumns and gun on top
important optical elements are identical

fei.com
jeol.com

Principles of STEM

- Confusing Literature

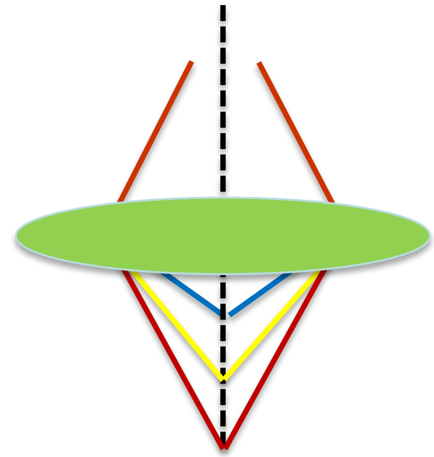
Probe forming lens and aperture:

Dedicated STEM: objective lens
Combined TEM/STEM: Condenser lens

Principle of STEM

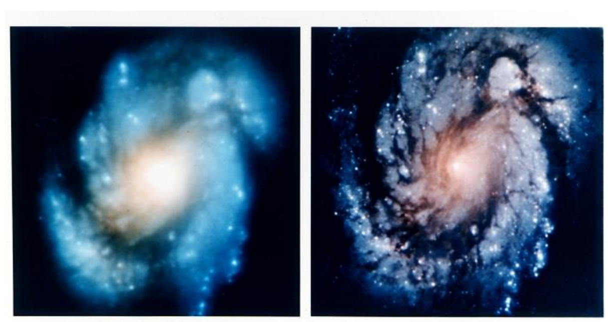
Lens aberration as resolution limiting factor

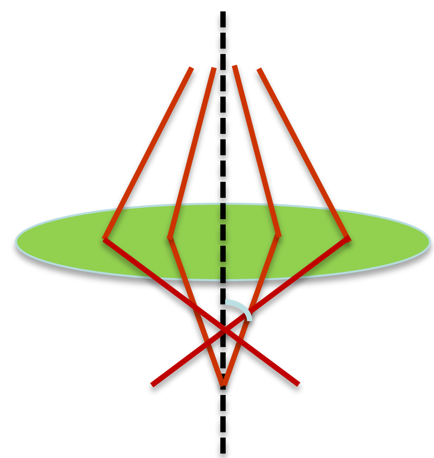
Chromatic Aberration



Spherical Aberration

Hubble telescope





Principles of STEM

Reciprocity of TEM and STEM

Reicprocity Theorem:

For elastical scattered electrons:
All electrons have same energy.

→ The propagation of electrons is
Time reversible

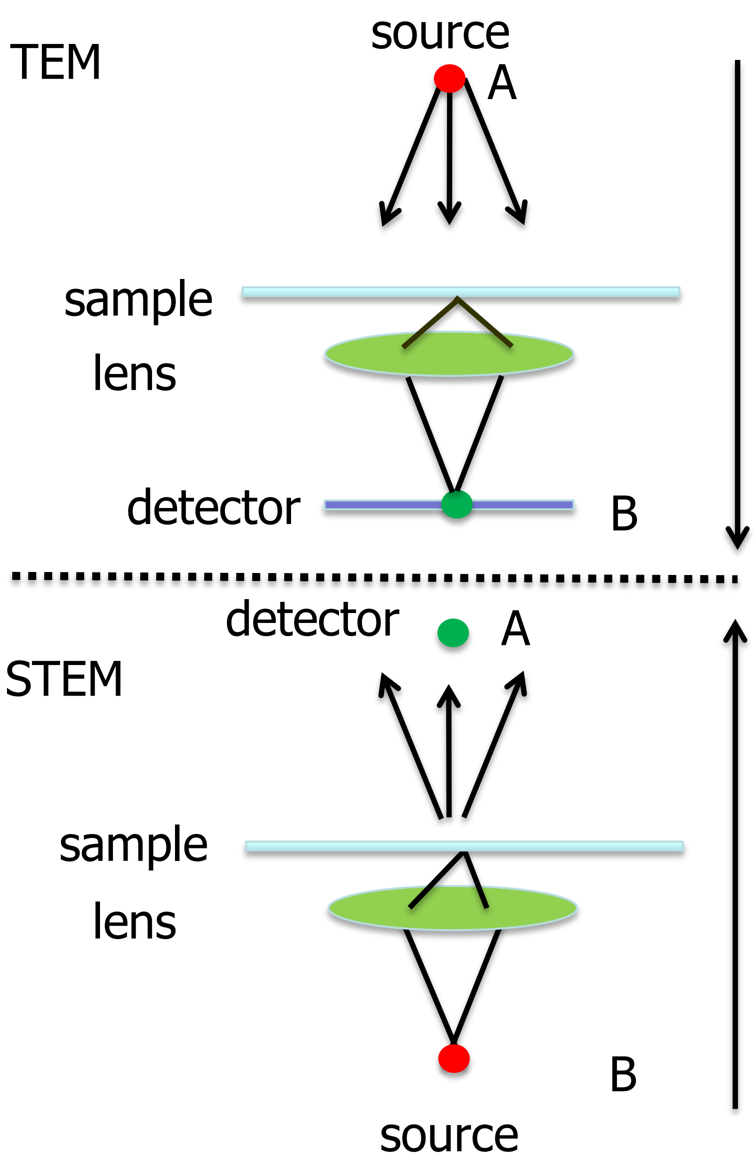
Electron intensities and ray paths in the Microscope remain the same if

the direction of rays is reversed and if

the source and detector are interchanged

→ Similar intensity

STEM imaging optics (before the sample)
Are the same than the imaging optics in
TEM (after the sample)



Zeitler, Thomson *Optik* **1970**, 31, 258-280 and 359-366
Cowley Appl.Phys.Lett. **1969**, 15, 58-59.

Scanning the sample

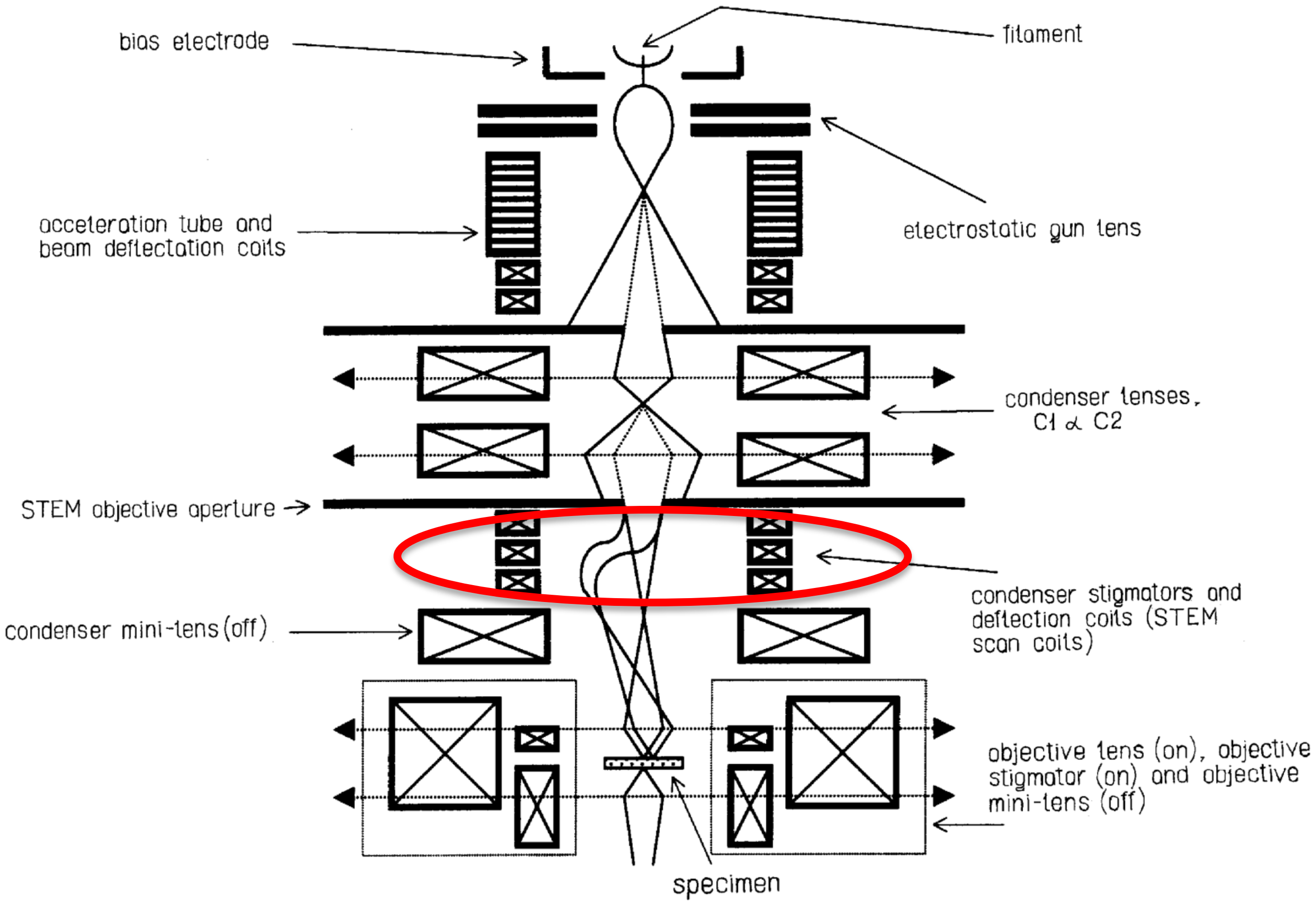


Fig. 2. Schematic of the probe forming optics in the JEOL 2010F STEM.

Browning et al. *Rev.Adv.Mat.Sci* **2000**, 1, 1-26.

Image Formation

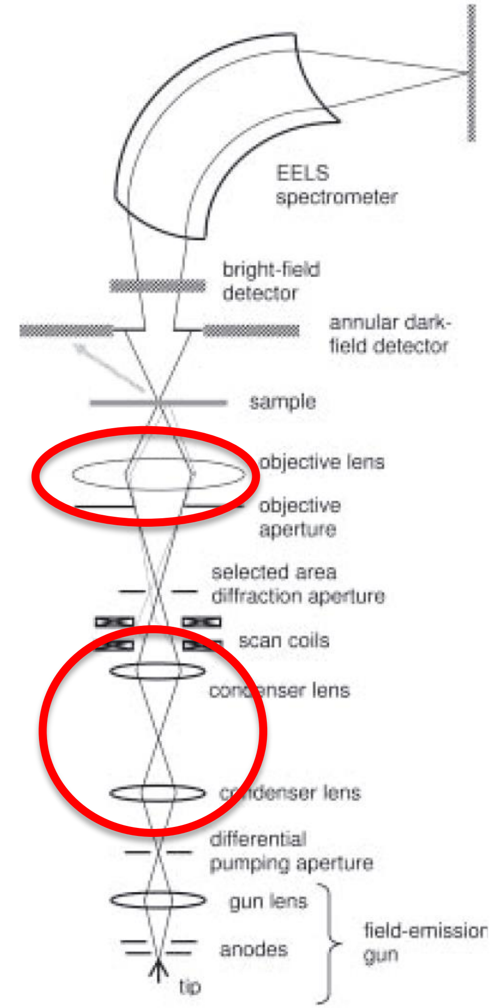
Thin sample (usually less than 50 nm)

→ Relatively small probe spreading

→ Resoultion dominated by the probe size

→ Important optics are the one that form the probe (dedicated STEM):
- objective lens: focuses the beam
- condenser lenses: demagnifies the electron source to form the probe

But: electron lenses suffer from inherent aberration: spherical and chromatic

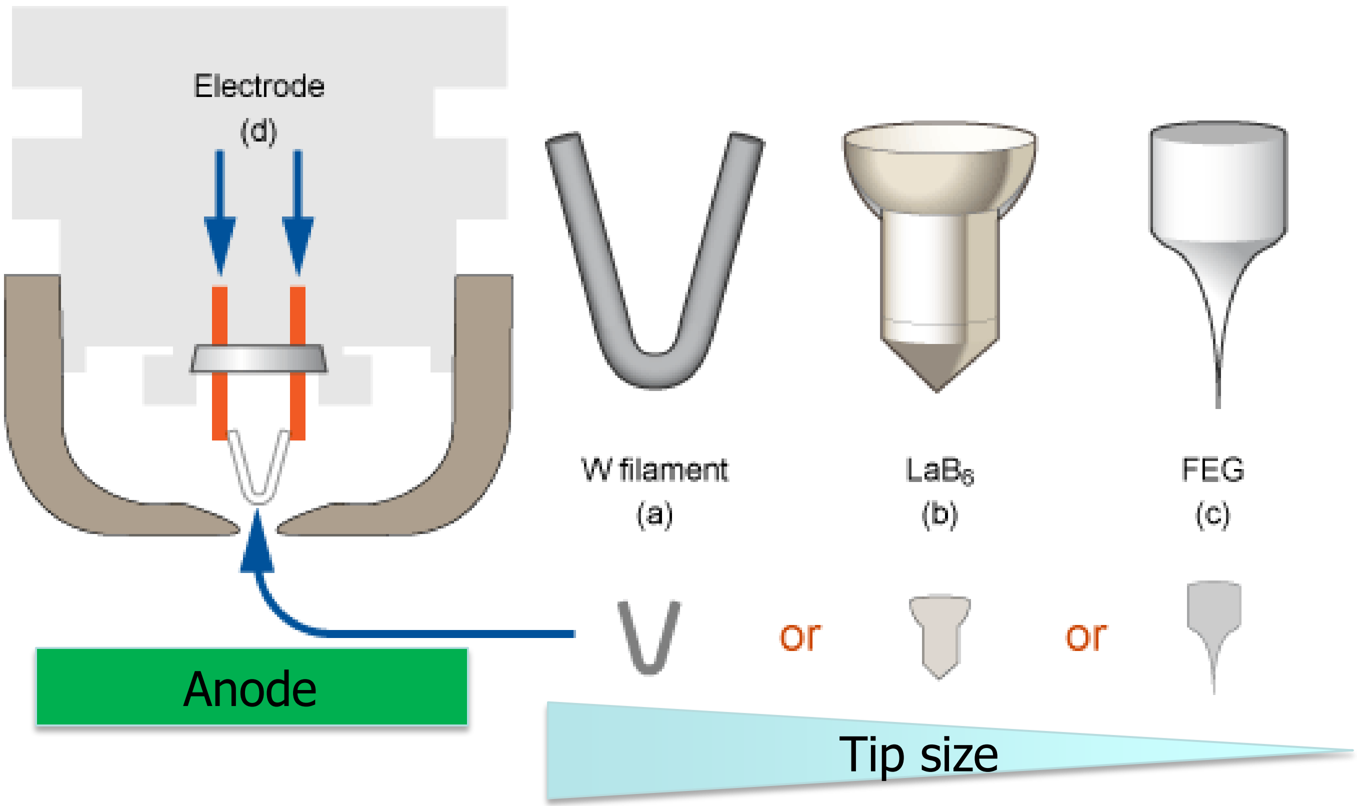


Probe size below the interatomic distances for atomic resolution images

Pennycook et al. Scanning Transmission Electron Microscopy **2012**

The electron source as resolution limiting factor

- Small and intense

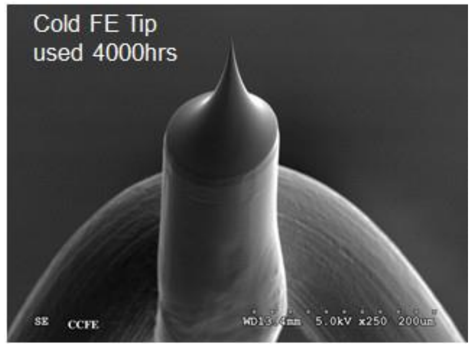


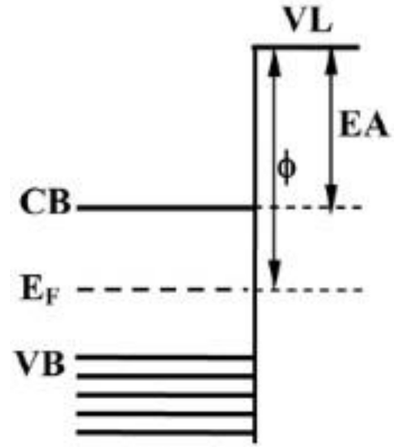
Principles of STEM

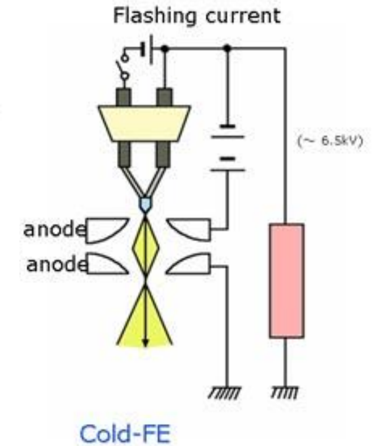
The electron source as resolution limiting factor

Cold FEG vs. Schottky FEG

Effective source size: 5 nm



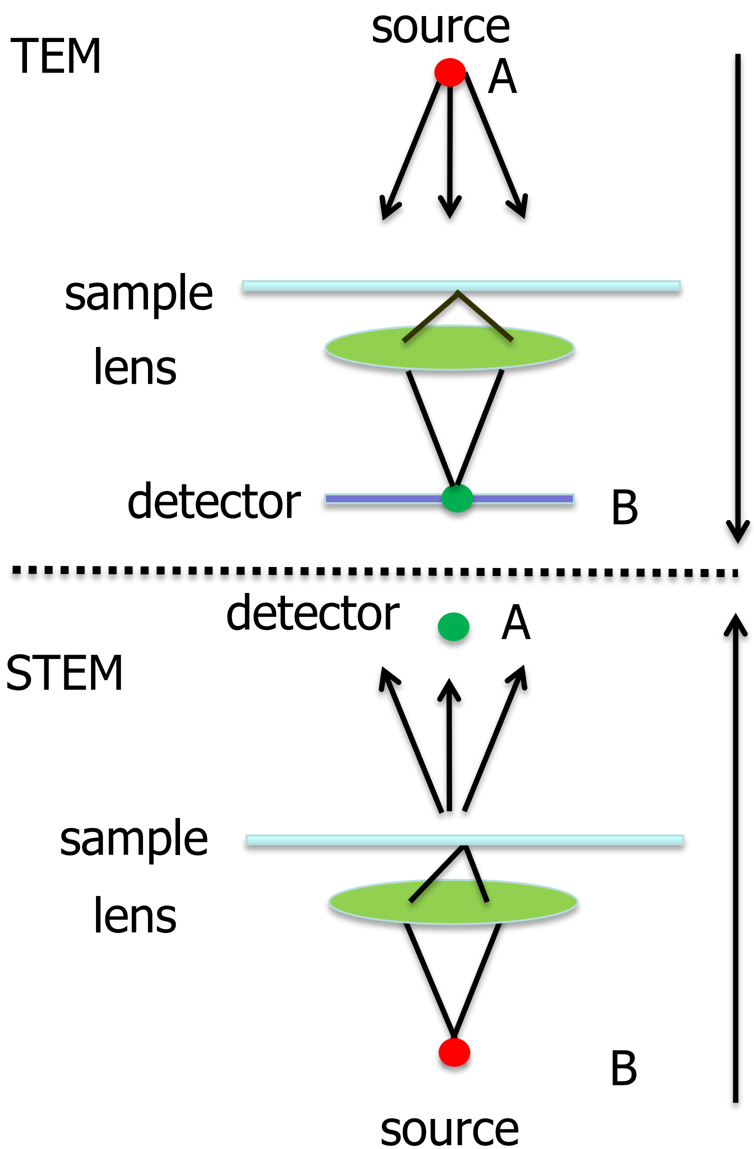




Source	Thermoionic	Thermoionic	FEG	Cold FEG
Material	W	LaB_6	**W(100) + ZrO**	**W(310)**
Work function [eV]	4.5	2.7	**2.7**	**4.5**
Tip radius [µm]	50-100	10-20	**0.5-1**	**<0.1**
Temperature [K]	2800	1900	**1800**	**300**
Normalized Brightness [$Acm^{-2}sr^{-1}$]	10^4	10^5	**10^7 / 10^8**	**2*10^7 / 10^9**
Energy spread at gun exit [eV]	1.5-2.5	1.3-2.5	**0.4-0.7 / 0.9**	**0.3-0.7 / 0.22**
Vacuum [Torr]			**10^{-9}**	**10^{-10}**

Dehm et al. *In-situ Electron Microscopy* 2012; spectral.se; spie.org

Reciprocity of TEM and STEM



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STEM Probe

What is the probe size?

Typical electron wavelength:

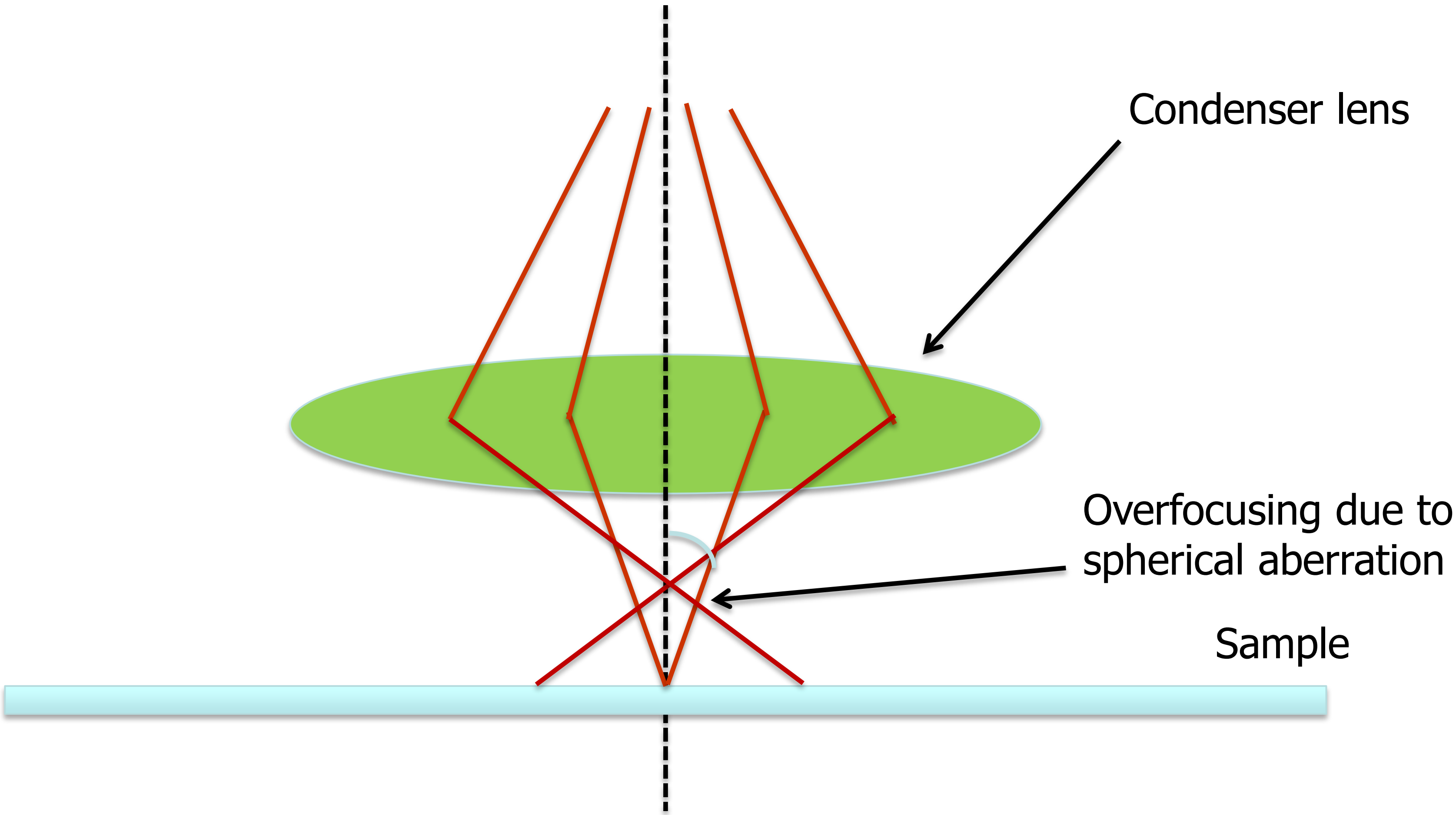
- 3.7 pm (for 100keV) and 1.9 pm (for 300keV)

Probe size should be close to these values!

Resolution is limited to about 0.2 nm

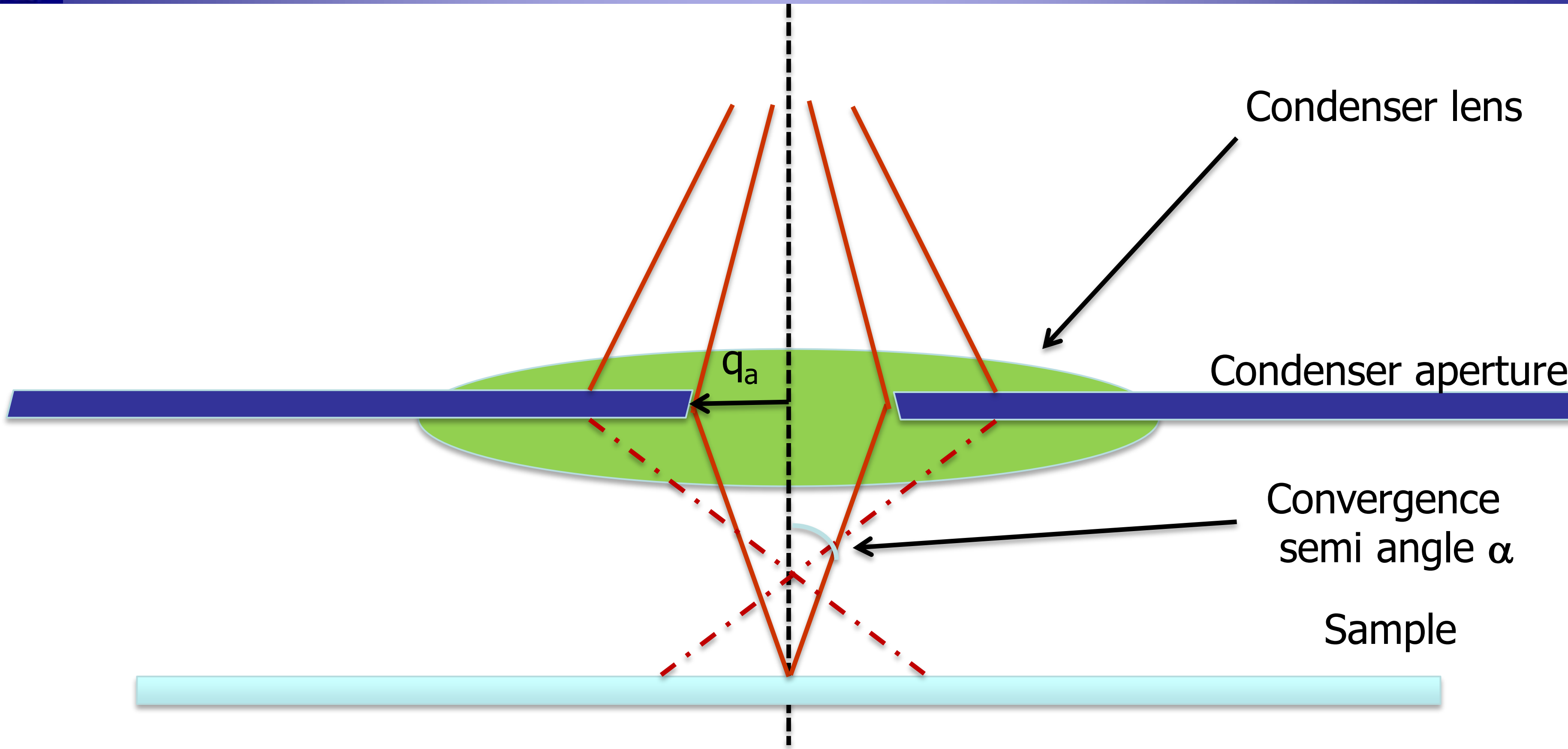
The most important aspect in STEM imaging is to focus a sub-nanometer sized probe at the sample

STEM Probe



Underfocusing the lens compensates the overfocusing effect of spherical aberration

STEM Probe

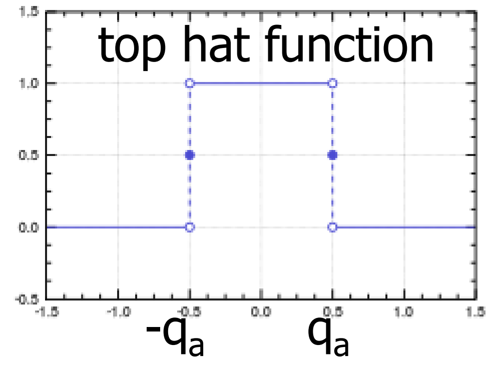


The probe is an image of the electron source. The probe size depends on the Same parameters as the resolution in a TEM image:

Aberration of the lens
Spatial and temporal coherence (energy spread of electron beam and source size)
Size of objective aperture

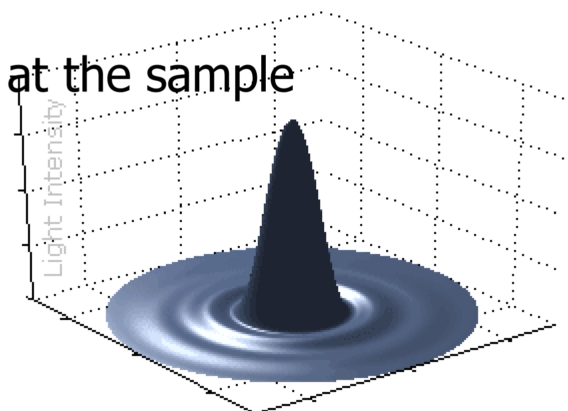
STEM Probe

For circular aperture
$$\psi_0(\boldsymbol{q}) = \begin{cases} 1 \text{ if } |\boldsymbol{q}| < |\boldsymbol{q}_\mathrm{a}| \\ 0 \text{ otherwise.} \end{cases}$$



Fermi function
$$\psi_0(\boldsymbol{q}) = \frac{1}{1 + \exp\left\{\frac{|\boldsymbol{q}|^2 - |\boldsymbol{q}_\mathrm{a}|^2}{\delta_\mathrm{a}^2}\right\}}$$

For a perfect lens (no aberration): FT → electron probe at the sample
Solely depending on the diffraction limit



3D Airy pattern

defocus

aberration funtion
$$\chi(\boldsymbol{q}) = \chi(q) = \frac{1}{2} q^2 \lambda^2 C_1 + \frac{1}{4} q^4 \lambda^4 C_3$$

Spherical aberration

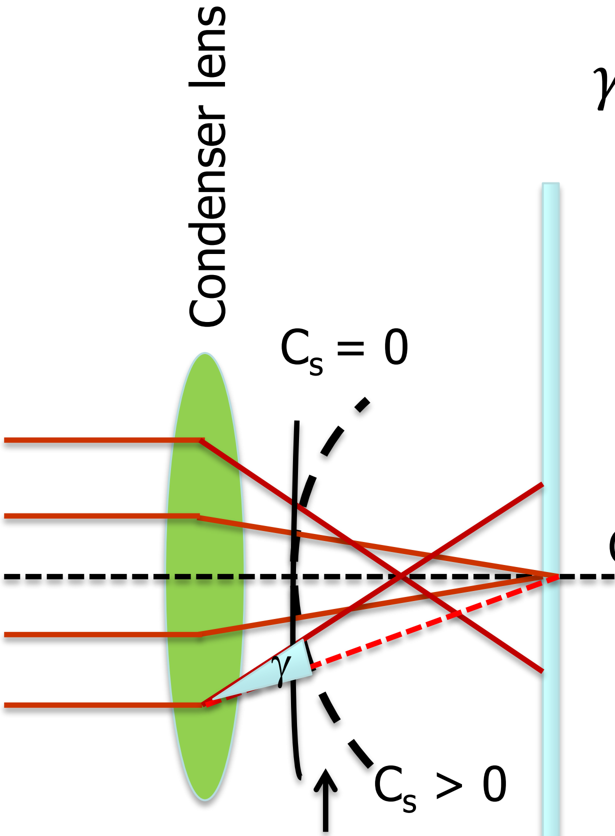
phase shift
$$\gamma(q) = 2\pi/\lambda \cdot \chi(q)$$

wave function of incident electron porbe $\psi_0 = A \cdot \exp\{\mathrm{i}B\}$

Erni - *Aberration-Corrected Imaging in Transmission Electron Microscopy* 2010.
cambridgeincolour.com

STEM Probe

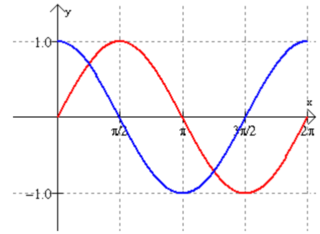
Optimization the probe



Phase difference
(wave front error)

Maximum tolerable phase shift

$$\gamma = \frac{\pi}{2} = \frac{\lambda}{4}$$

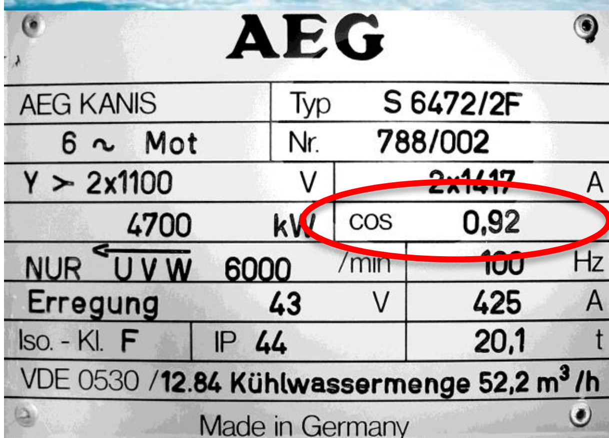


otherwise intensity loss
would be too high

Optimal conditions as derived
From Scherzer or Mory

$$f = -\frac{3}{4}\sqrt{C_1\lambda}$$

$$\alpha = 1.27(C_3\lambda)^{\frac{1}{4}}$$



Erni - *Aberration-Corrected Imaging in Transmission Electron Microscopy* 2010.

STEM Probe

Phase shift of the electron wave by the aperture (defocus and spherical aberration)

$$\psi_0(q) = \underbrace{\exp\left\{-\frac{2\pi \mathrm{i}}{\lambda}\chi(q)\right\}}_{\text{phase}} \underbrace{\frac{1}{1+\exp\left\{\frac{q^2-q_\mathrm{a}^2}{\delta_\mathrm{a}^2}\right\}}}_{\text{aperture function: amplitude}}$$

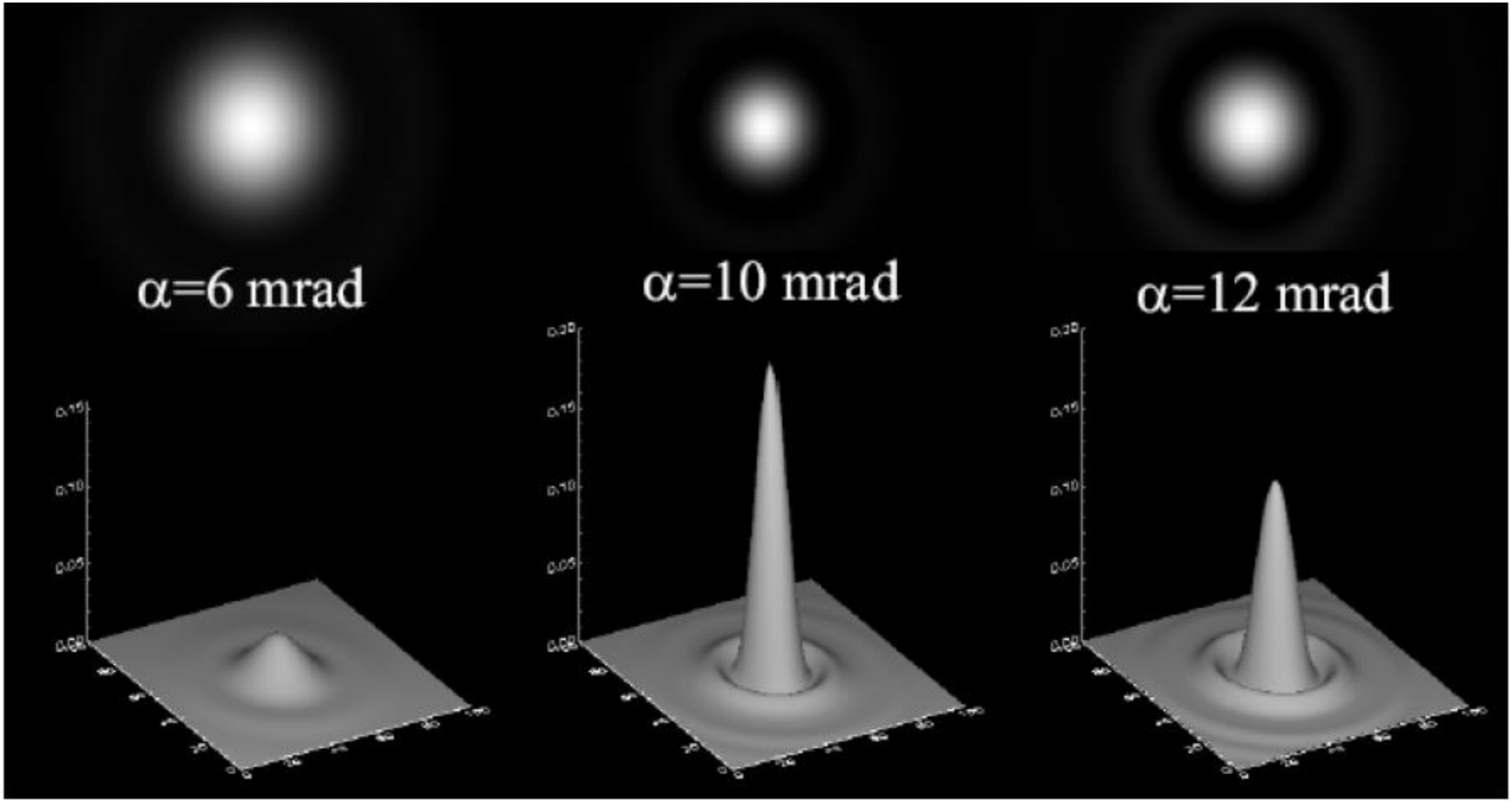
Coherent electron wave at the sample (electron probe)

$$\psi_0(\boldsymbol{r}) = \int\limits_{-\infty}^{\infty} \frac{\exp\left\{-\frac{2\pi \mathrm{i}}{\lambda}\chi(q)\right\}}{1+\exp\left\{\frac{|\boldsymbol{q}|^2-|\boldsymbol{q}_\mathrm{a}|^2}{\delta_\mathrm{a}^2}\right\}} \exp\{-2\pi \mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}\}\, d\boldsymbol{q}$$

Probe intensity distribution on sample $I_0(\boldsymbol{r}) = \psi_0(\boldsymbol{r})\overline{\psi}_0(\boldsymbol{r}) = |\psi_0(\boldsymbol{r})|^2$

Erni - *Aberration-Corrected Imaging in Transmission Electron Microscopy* 2010.

STEM Probe



STEM Probe

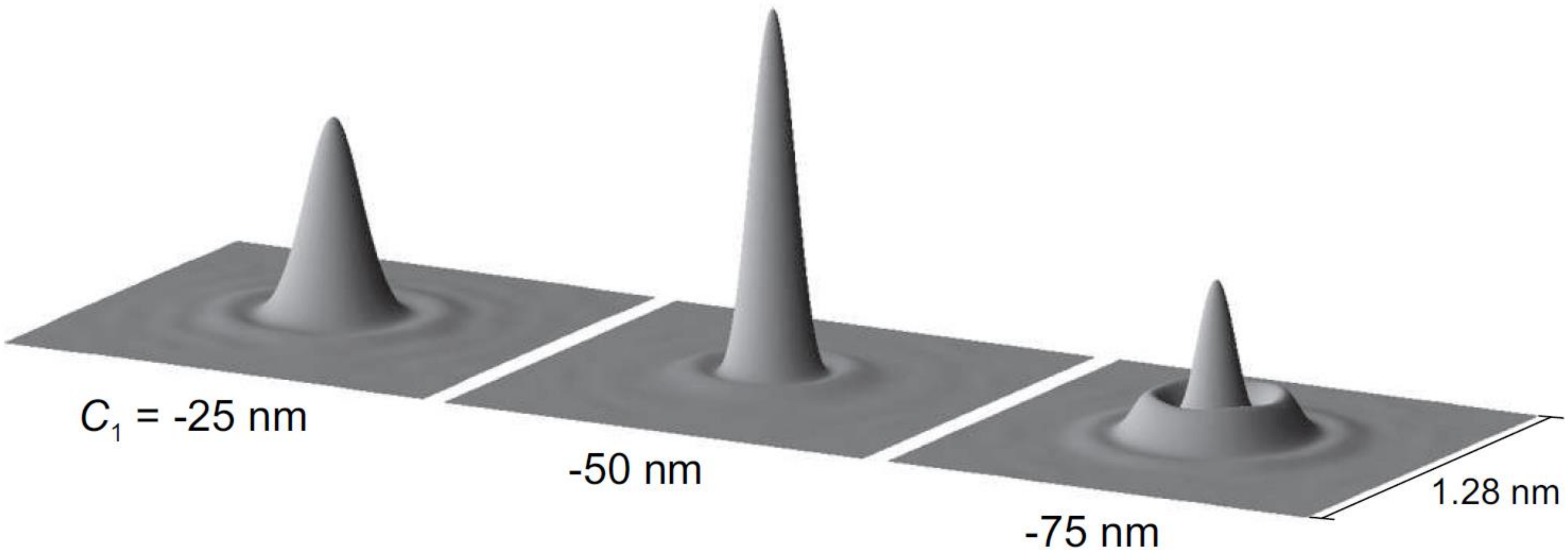


Fig. 3.8 Intensity profiles of electron probes calculated for 200 keV electrons ($\lambda = 2.5$ pm) and $C_3 = 1$ mm. The probes were calculated according to Eq. (3.17) for a defocus of -75 nm, for an optimized defocus of -50 nm (see Eq. (3.9)), and for -25 nm. The probe illumination semi-angle is in all three cases 10 mrad, corresponding to the optimum angle (see Eq. (3.10)).

How can we tune the electron probe experimentally?

Erni - *Aberration-Corrected Imaging in Transmission Electron Microscopy* 2010.

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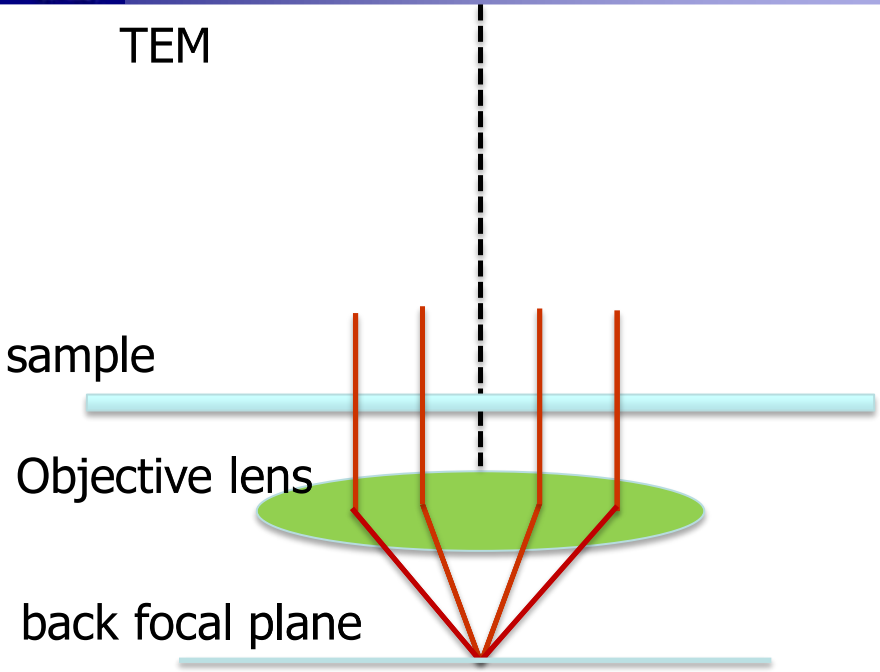
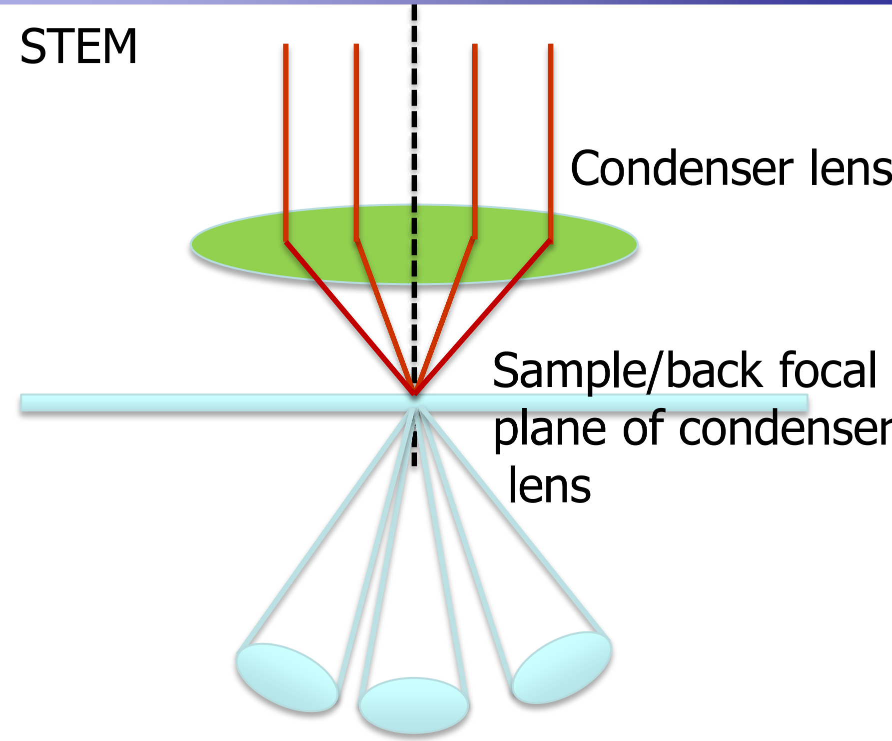
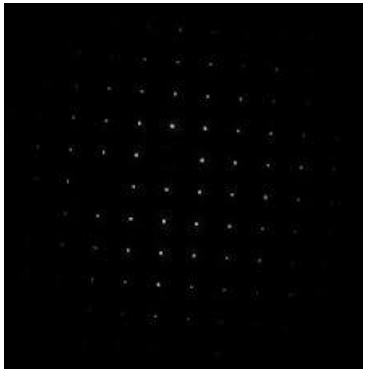
Ronchigram

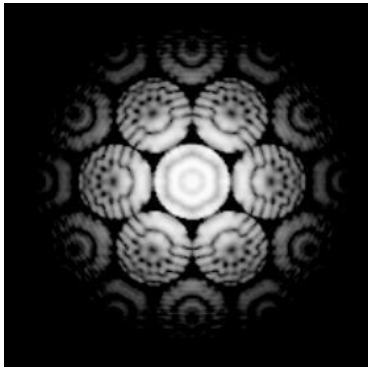






Electron Diffraction (ED)

The Ronchigram can emerge from the undiffracted disc of electrons at the center of the CBED pattern



Convergent Beam Electron Diffraction (CBED)

ammrf.org.au; hremresearch.com

Ronchigram

The shadow image (projection)

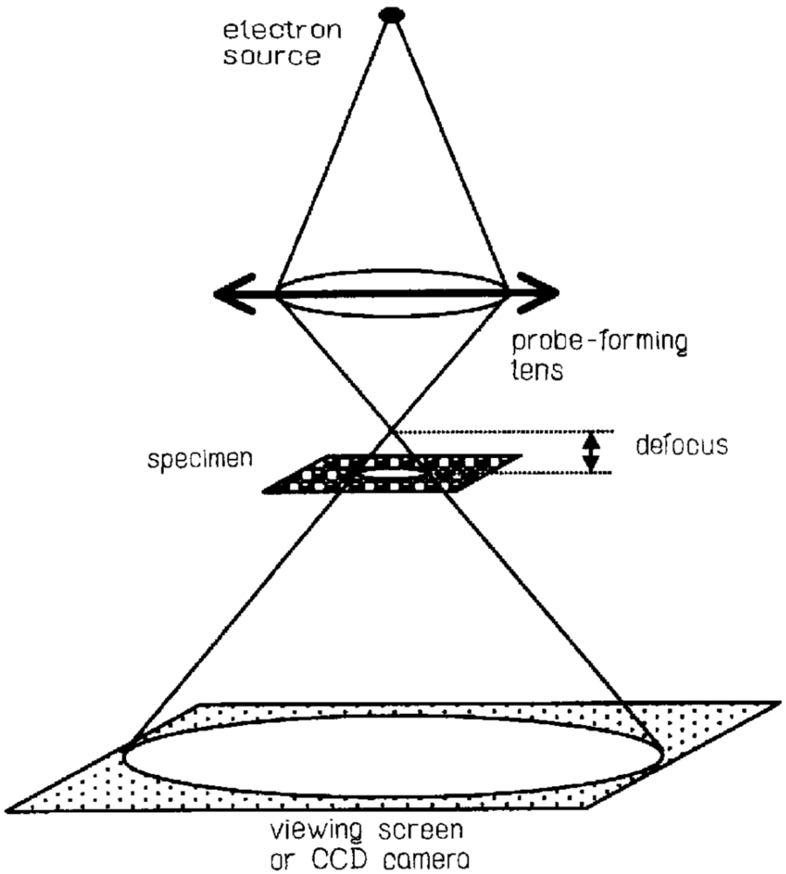


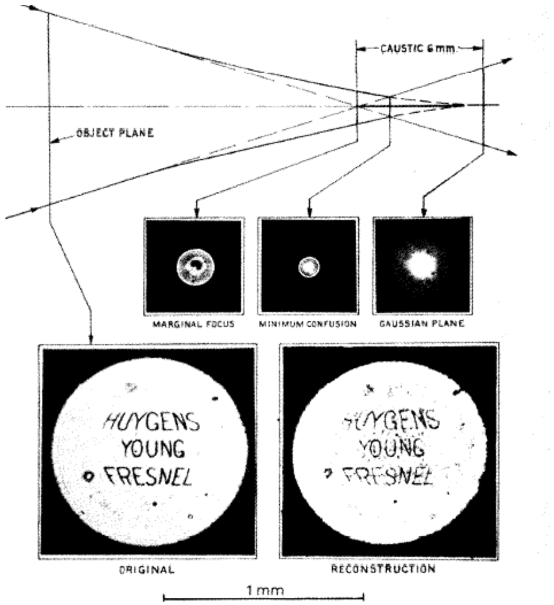
Fig. 3. Simplified ray diagram showing the formation of an electron Ronchigram.

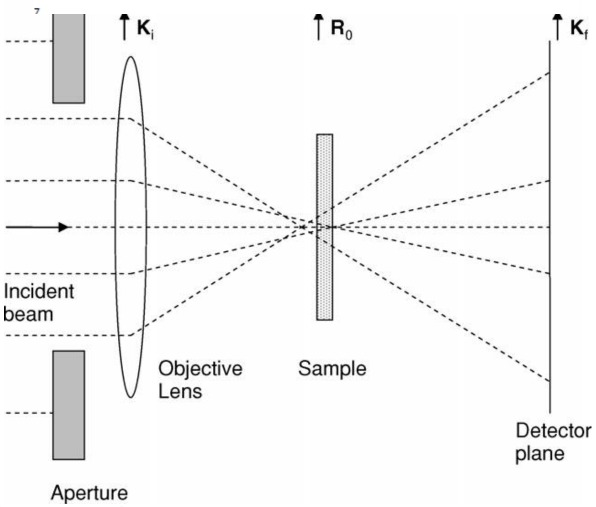
Discovered by Ronchi (1948)
During the investigation of the
Spherical aberration of optical
lenses

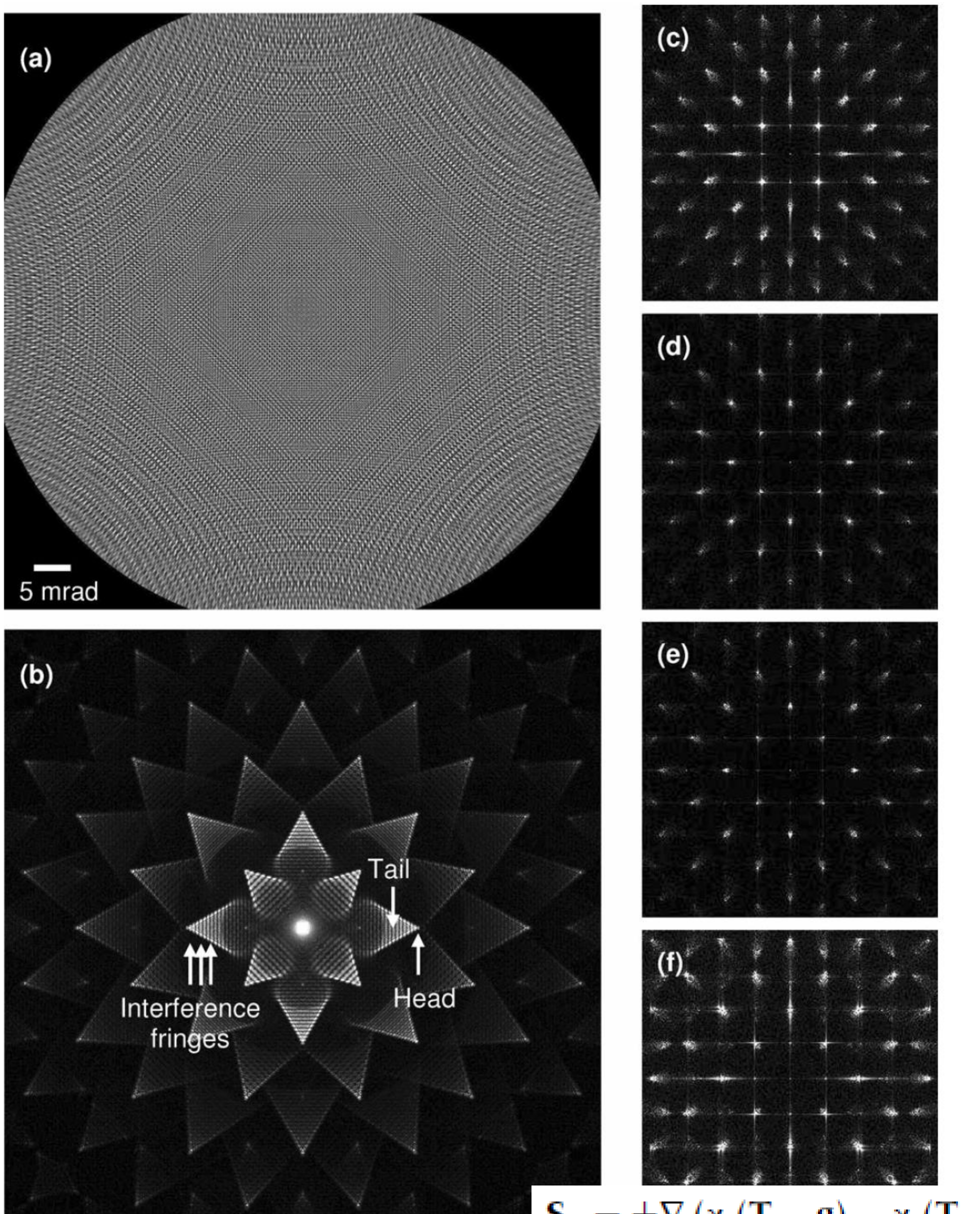
Browning et al. *Rev.Adv.Mat.Sci* **2000**, 1, 1-26.

Ronchigram

Inline hologram





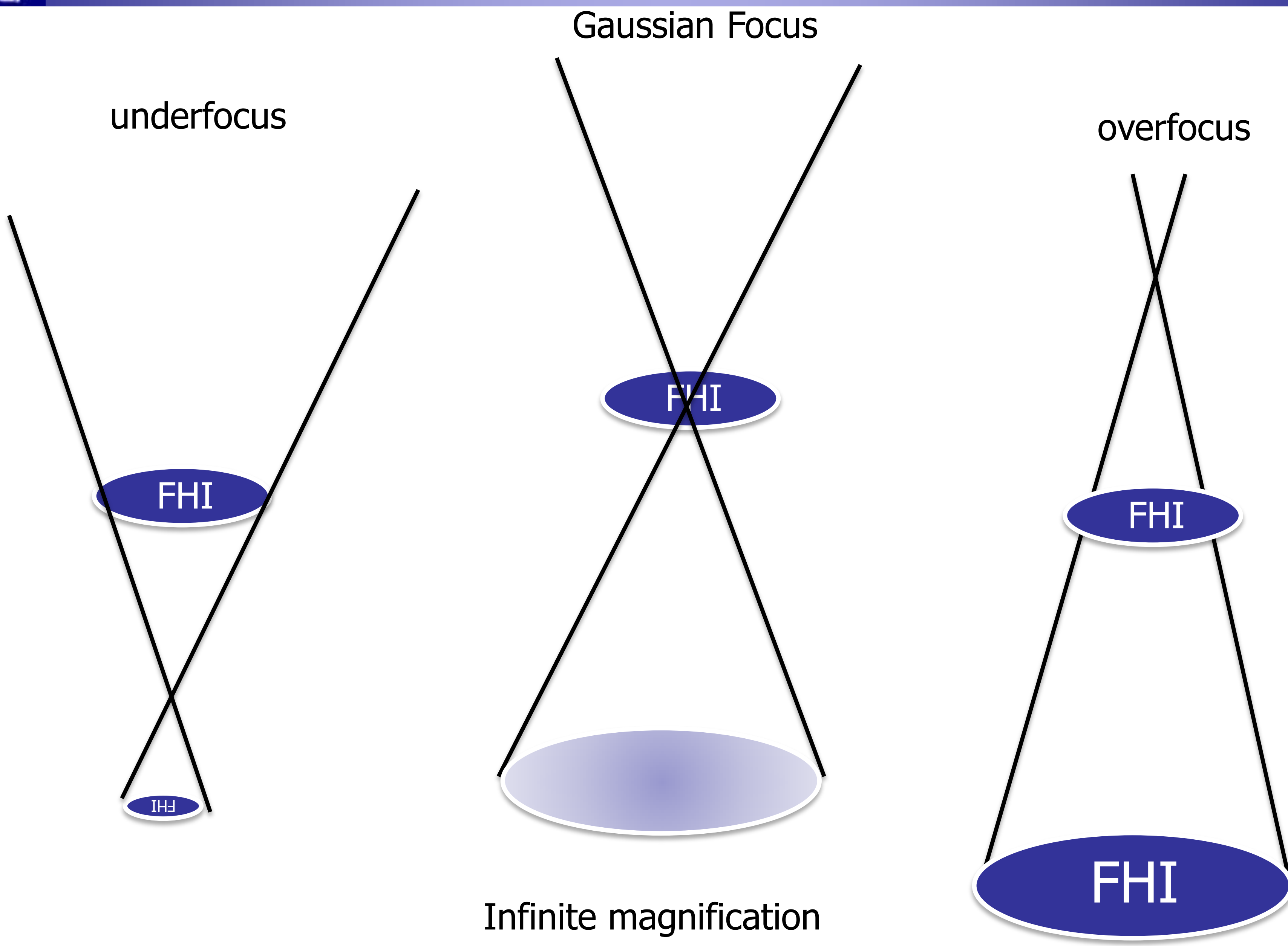


Gabor – Noble Lecture 1971

Lupini et al. *Journal of Electron Microscopy* **2008**, 57, 195–201.

$$S_T = \pm\nabla\left(\chi\left(T - g\right) - \chi\left(T - h\right)\right)$$

Ronchigram


Gaussian Focus
underfocus
overfocus
FHI
FHI
FHI
FHI
Infinite magnification

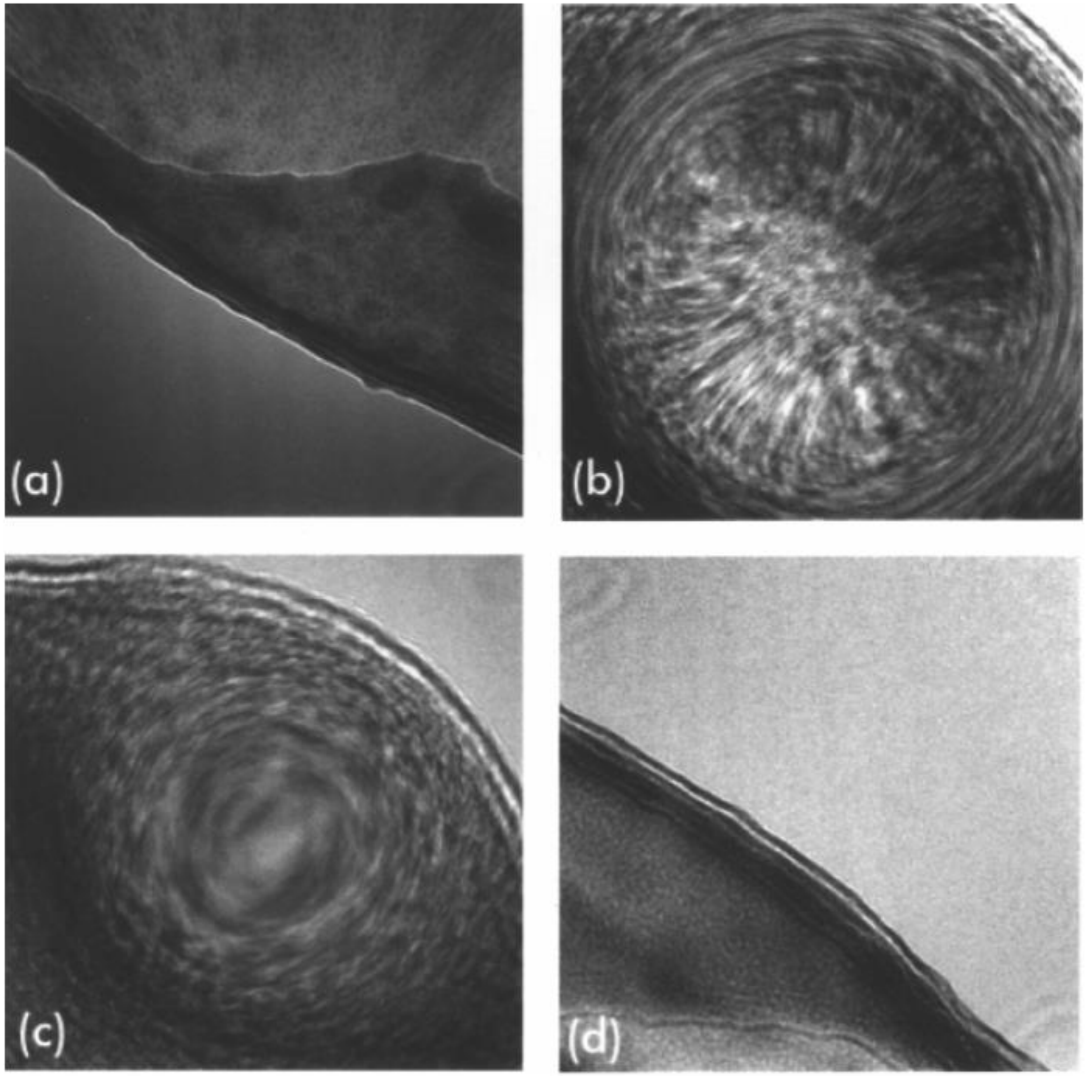
Ronchigram

underfocus

Underfocus
Close to Gaussian
focus

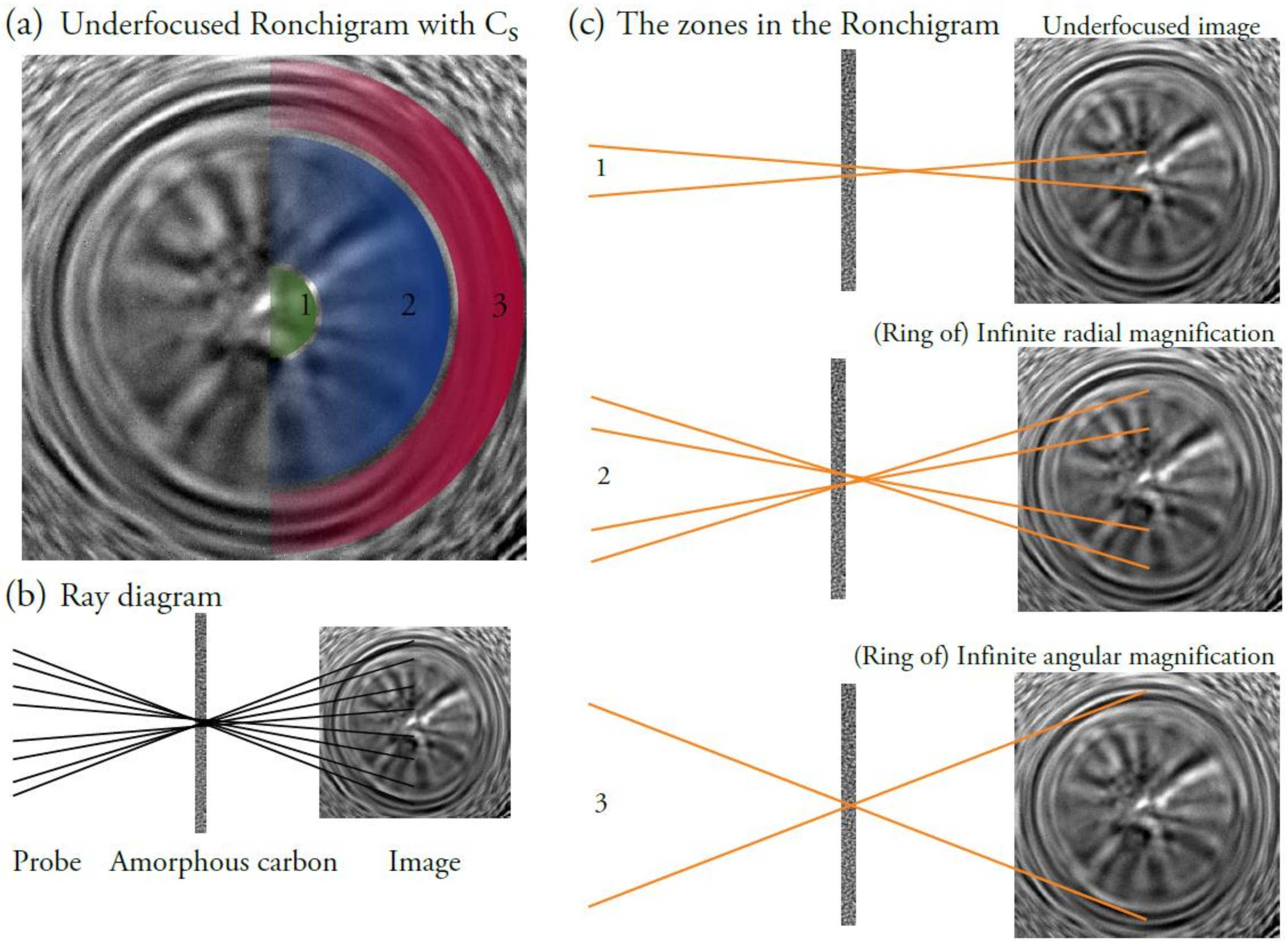
Gaussian focus
infinite
magnification

overfocus



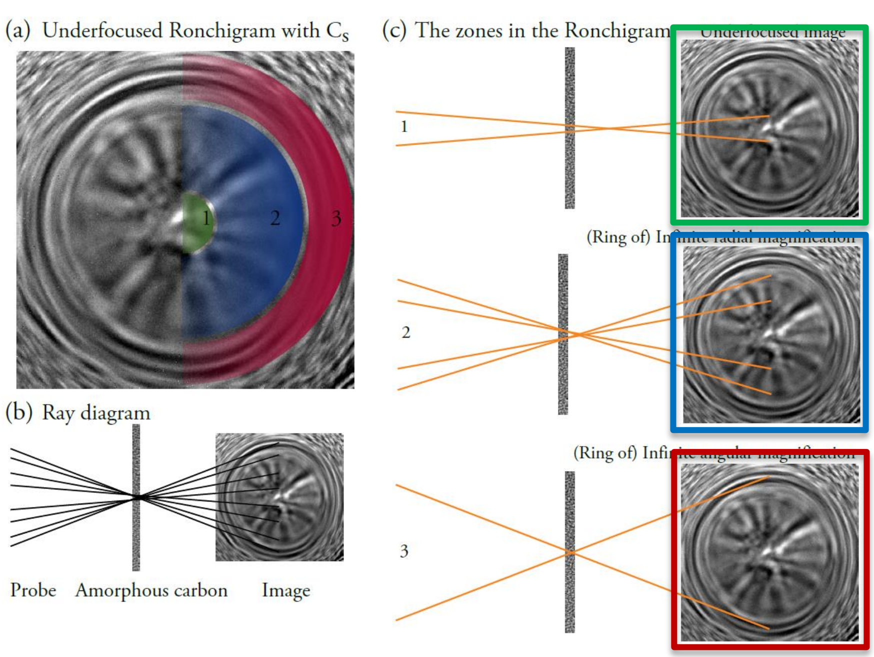
Browning et al. *Rev.Adv.Mat.Sci* **2000**, 1, 1-26.

Ronchigram

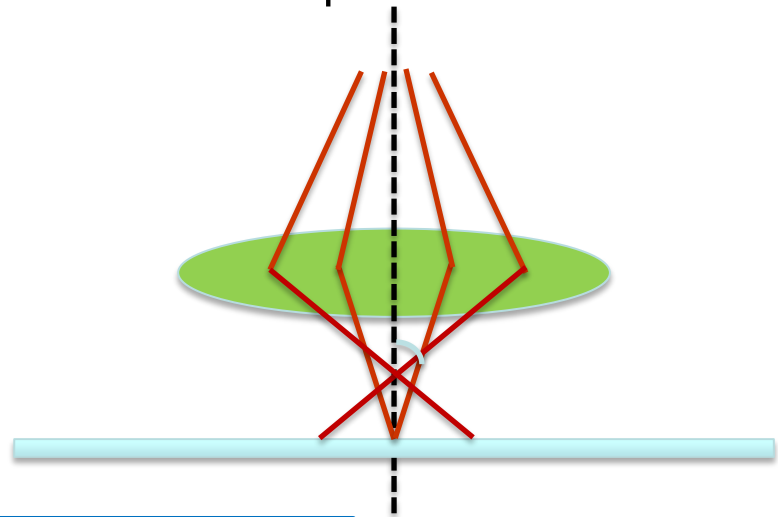


Ek – *A few concepts in TEM and STEM explained* 2011.

Ronchigram



spherical aberration
Underfocusing → partially compensation



High and equal angles
focused on the sample

→ Magnify single point on the Sample to the outer parts of the Ronchigram

→ Stretch into ring with infinite angular magnification

Medium angles
2 rays on the same site
Slightly different angles

Coincide on a ring
on the sample

→Points on this ring are stretched radially
→ infinite radial magnifcation

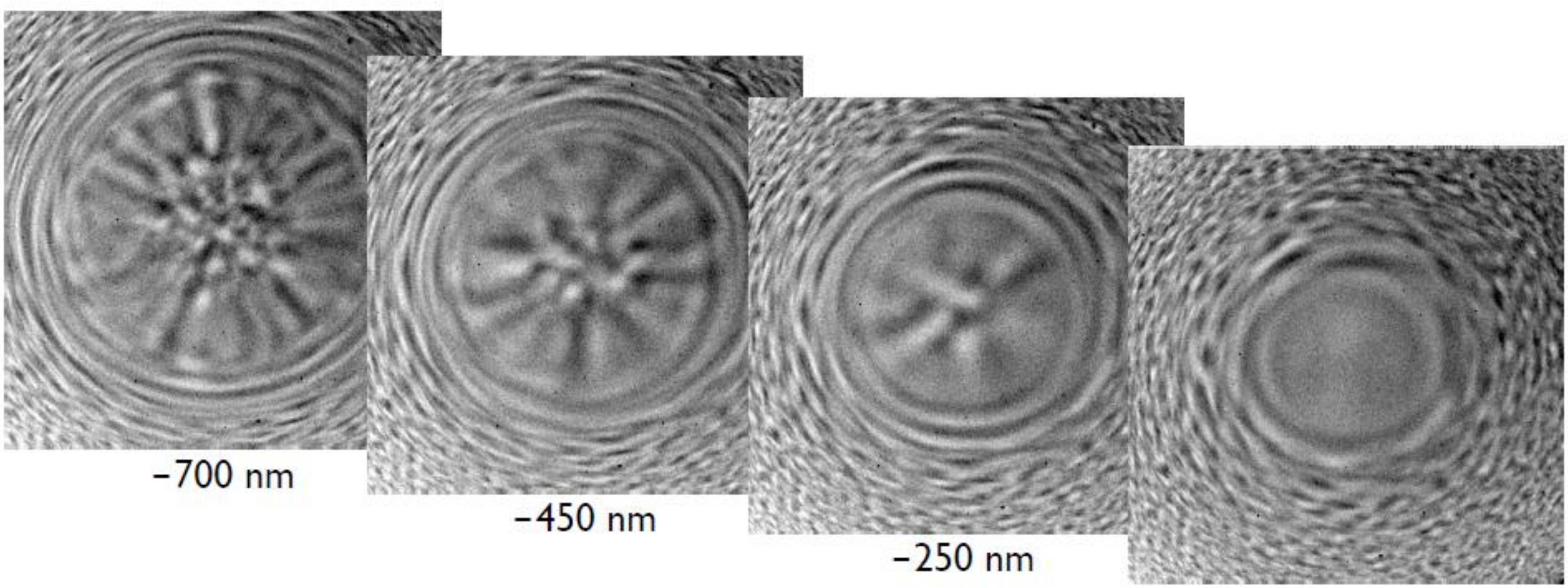
Low angle
Underfocused

→ Shadow image

Ronchigram

Reduce underfocus until infinite magnification rings are of minimum diameter
→ Scherzer like defocus

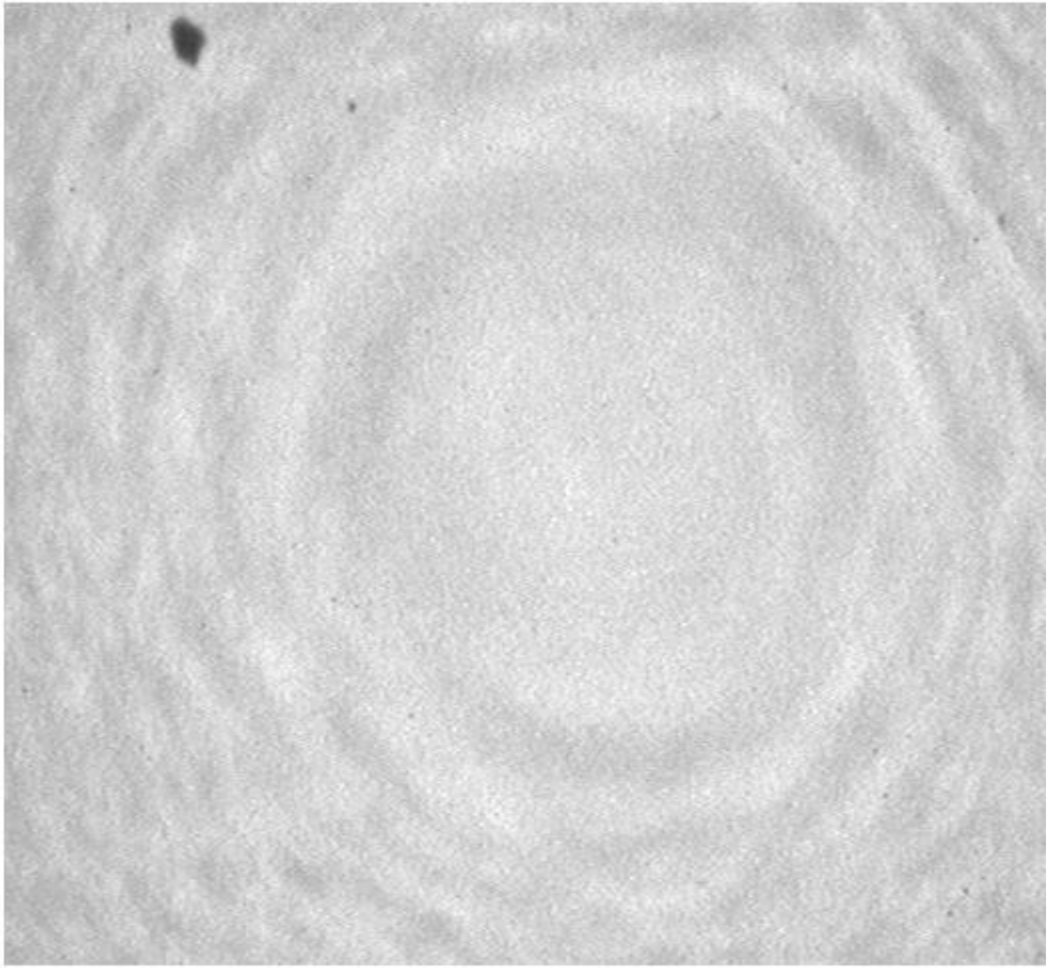
Fit condenser aperture to the sweet spot region of constant phase within this diameter



Alexander – *Looking through the fish-eye – the electron Ronchigram* 2012.

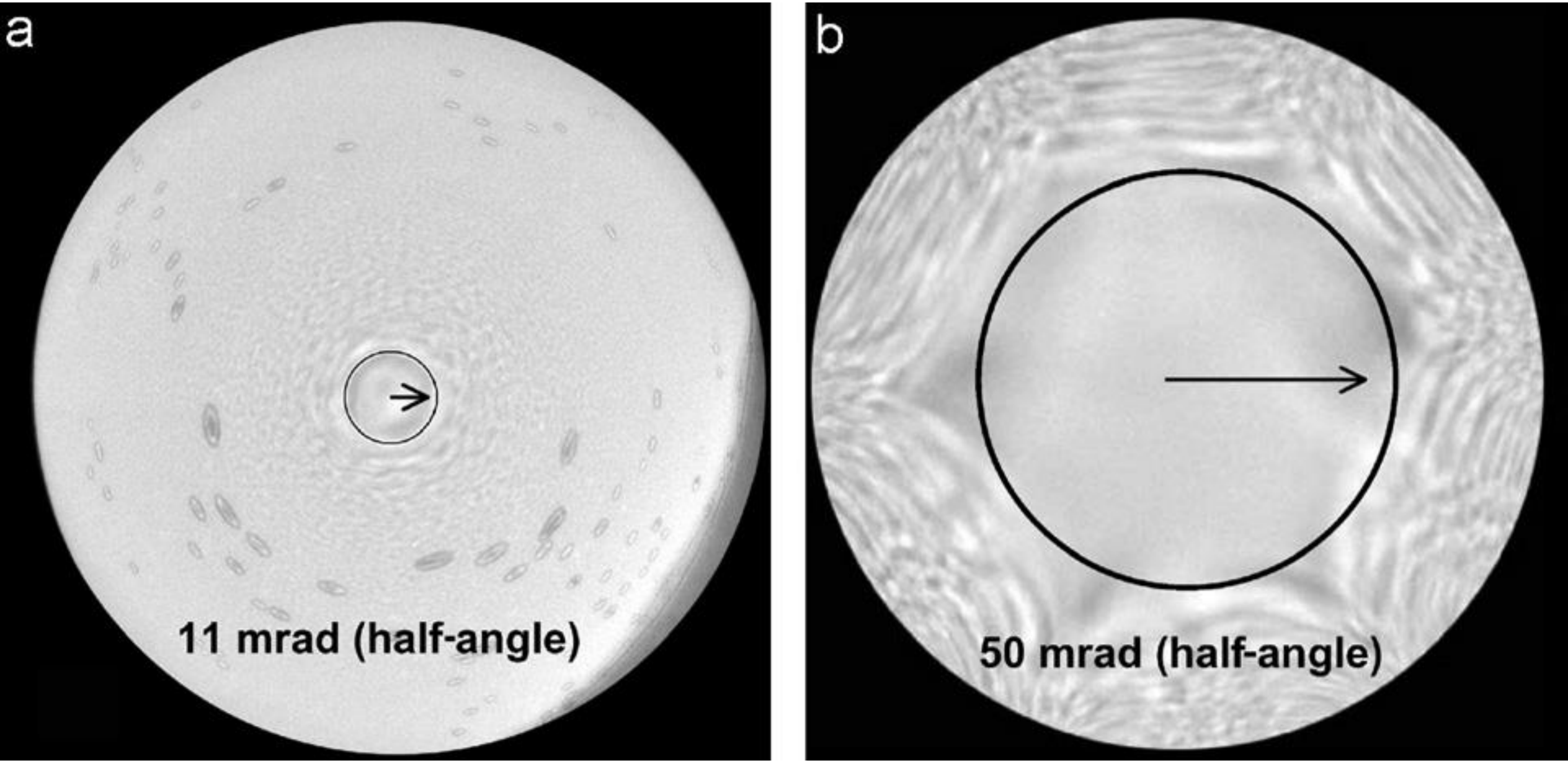
Ronchigram

Astigmatism



Alexander – *Looking through the fish-eye – the electron Ronchigram* 2012.

Ronchigram



uncorrected

Spherical abberation corrected

Sawada – *Ultramicroscopy* **2008**, 108, 1476-1475.

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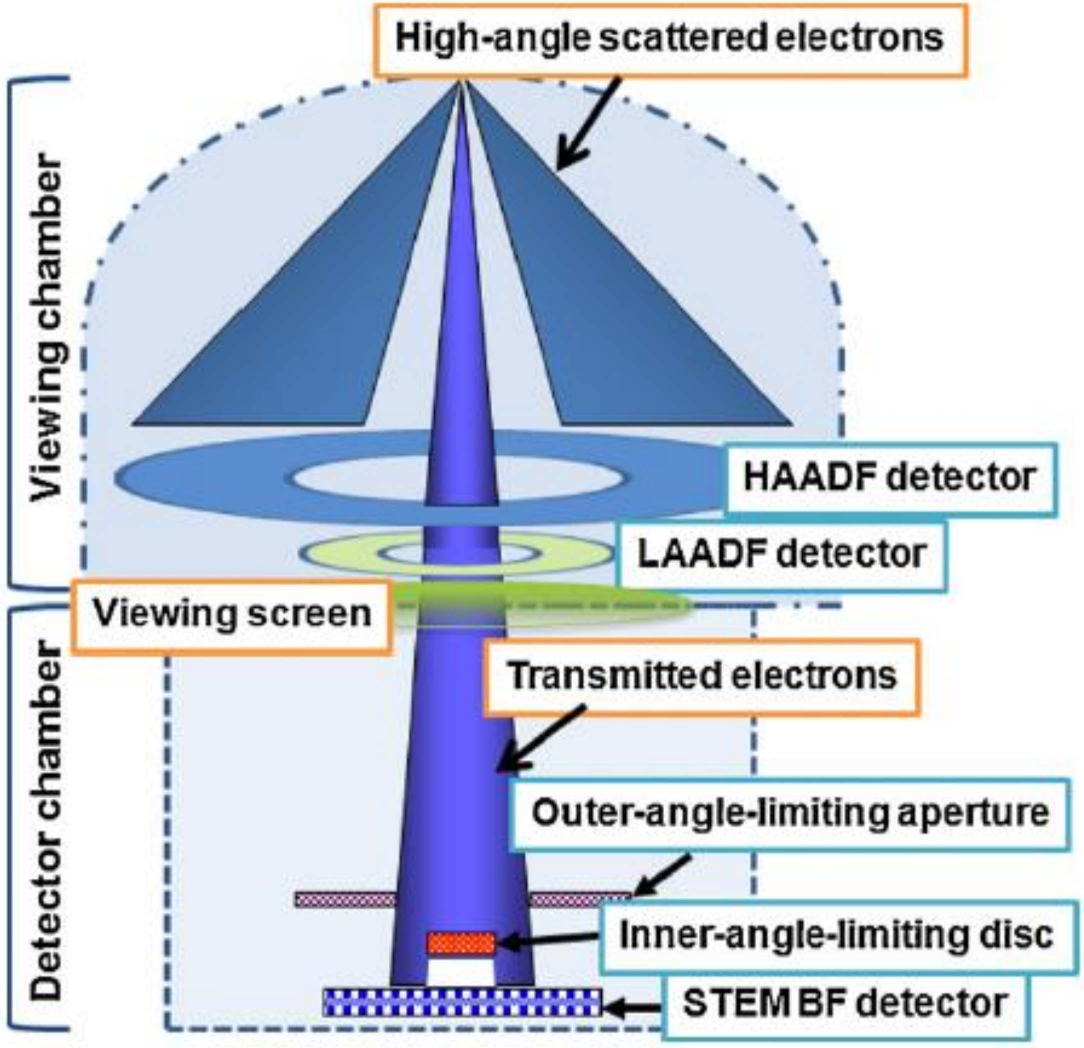
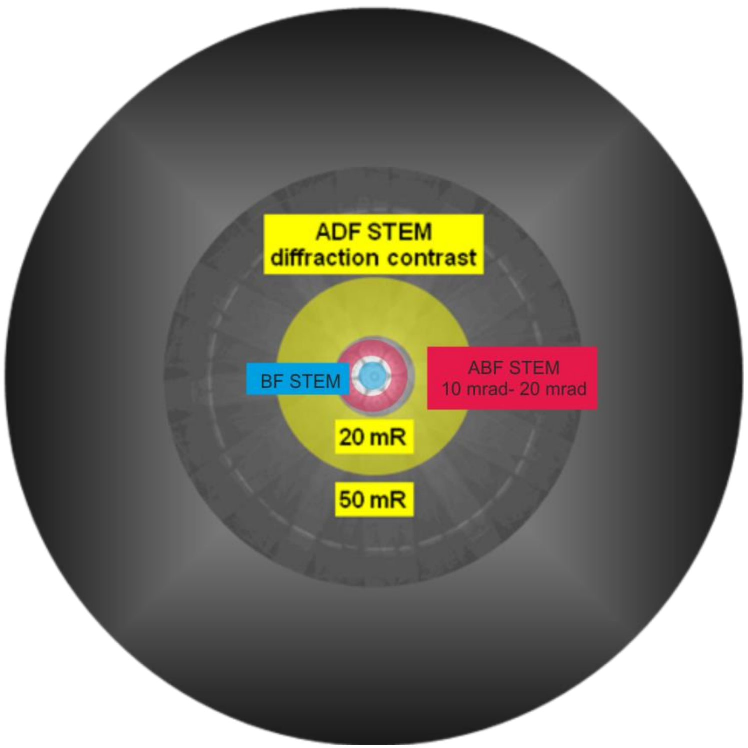
Detectors



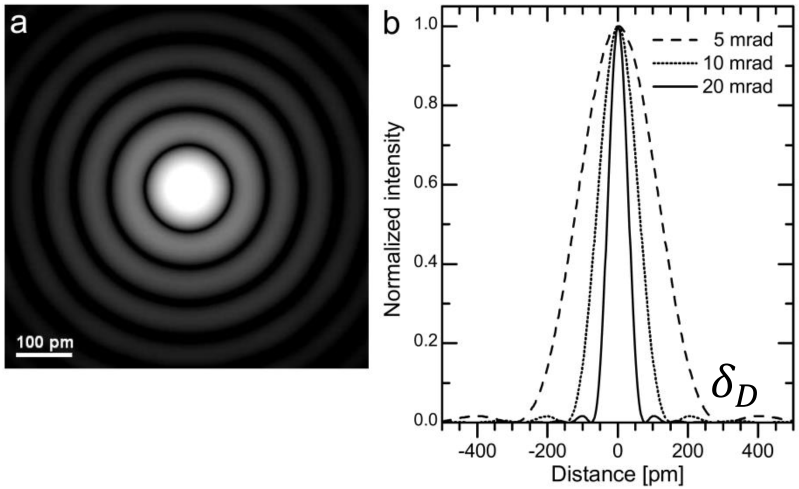
E. Okunishi et al. *Micron* **2012**, 43, 538–544.

Detectors

bright field detector

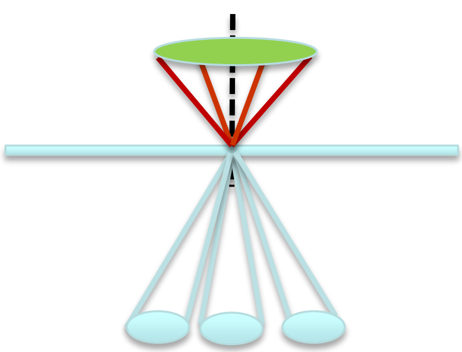


Airy disk

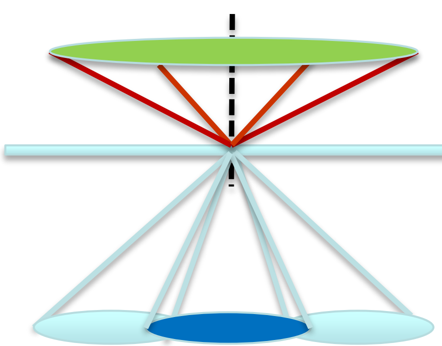


$$\delta_D = 0.61\frac{\lambda}{\alpha}$$

no lattice resolution



lattice resolution



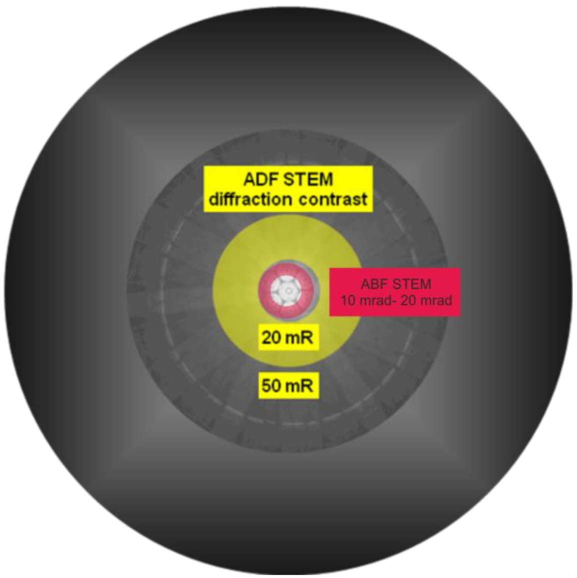
interference

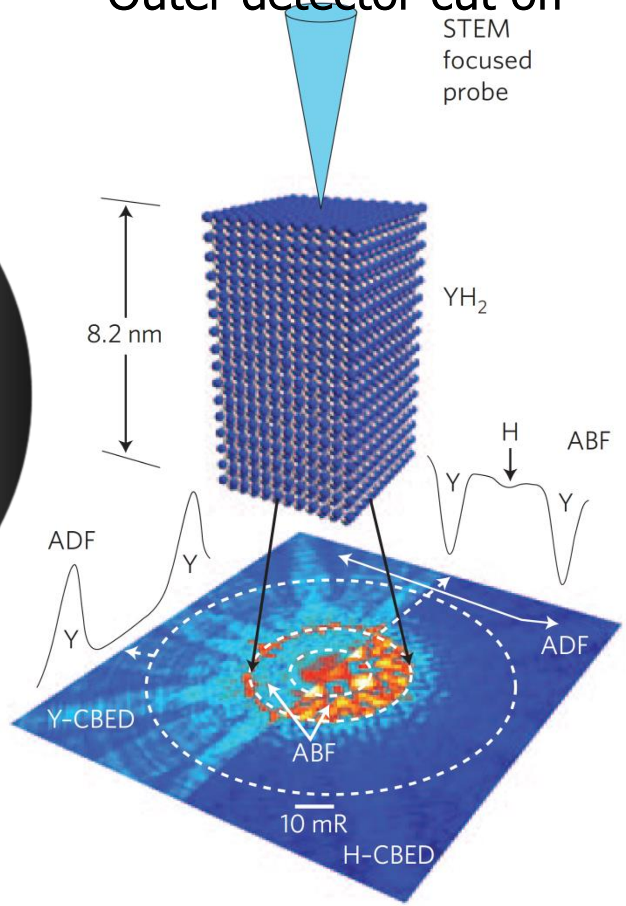
Erni - *Aberration-Corrected Imaging in Transmission Electron Microscopy* 2010.

Detector

Annular Bright Field (ABF)

Convergence angle =
Outer detector cut off





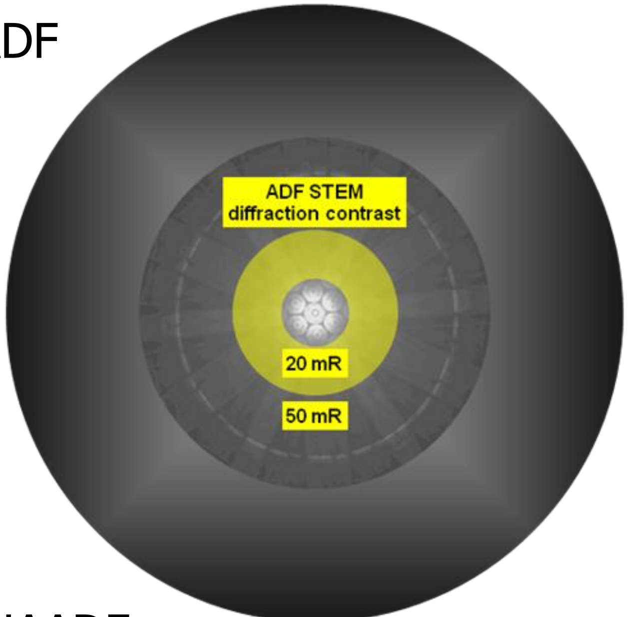
Small angle scattering occurs at the edges of the atoms where all atoms have similiar Charge densities.

gatan.com

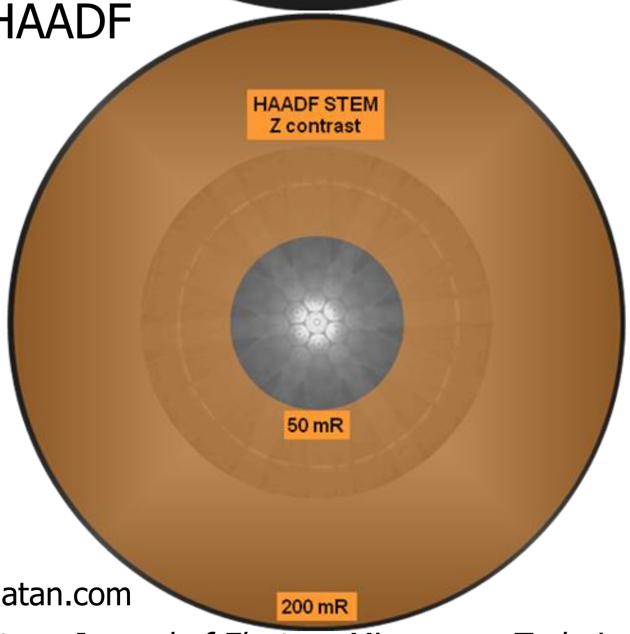
Batson *Nature Materials* **2011**, 10, 270-271.

Detectors

ADF

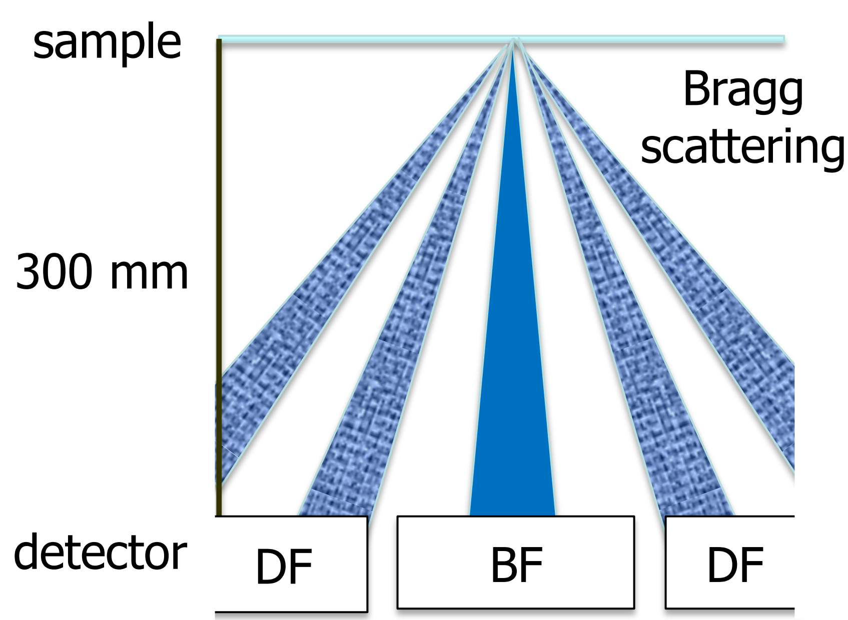


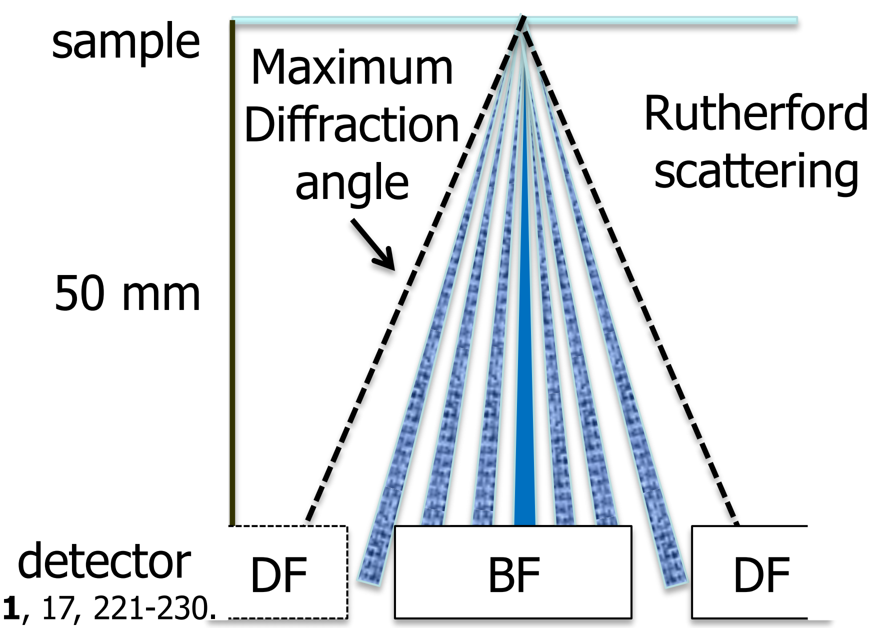
HAADF



gatan.com

Otten - *Journal of Electron Microscopy Technique* **1991**, 17, 221-230.





Combined STEM/ TEM

$$L\lambda = dD$$

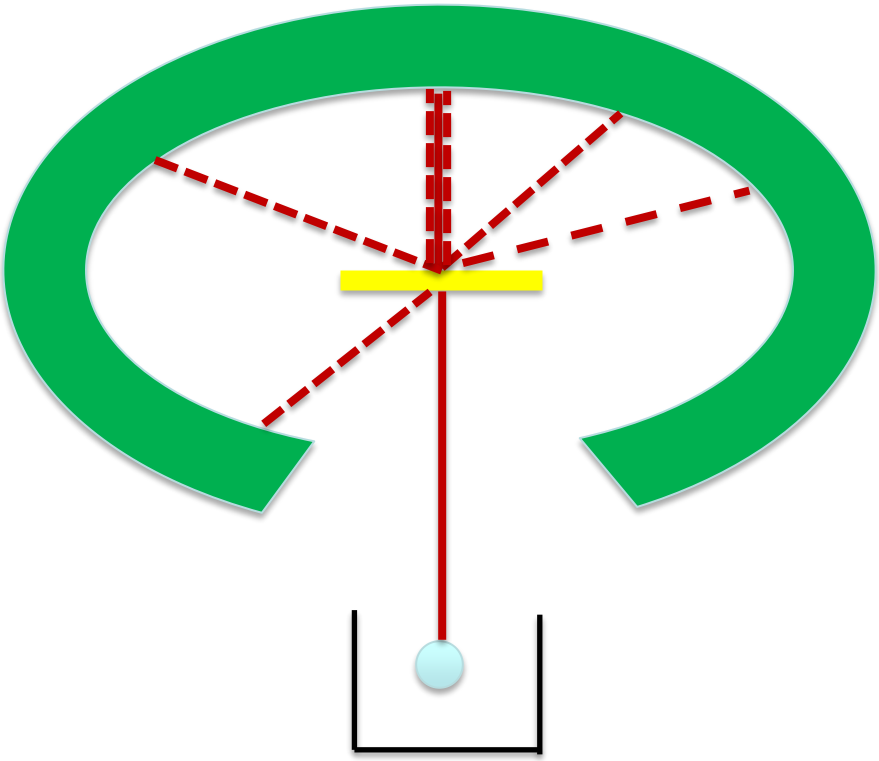
Detectors

High angle annular dark field (HAADF) incoherent elastical scattering

Rutherford scattering (elastic scattering)

Rutherford cross section

$$\left(\frac{d\sigma}{\alpha\Omega}\right)_{\theta} = \left(\frac{1}{4\pi\varepsilon_0}\right)^2 Z_2{}^2 \left(\frac{Z_1 e^2}{4E}\right)^2 \frac{1}{sin^4(\theta/_2)}$$



Detectors

Thermal Diffuse Scattering (elastic but incoherent scattering)

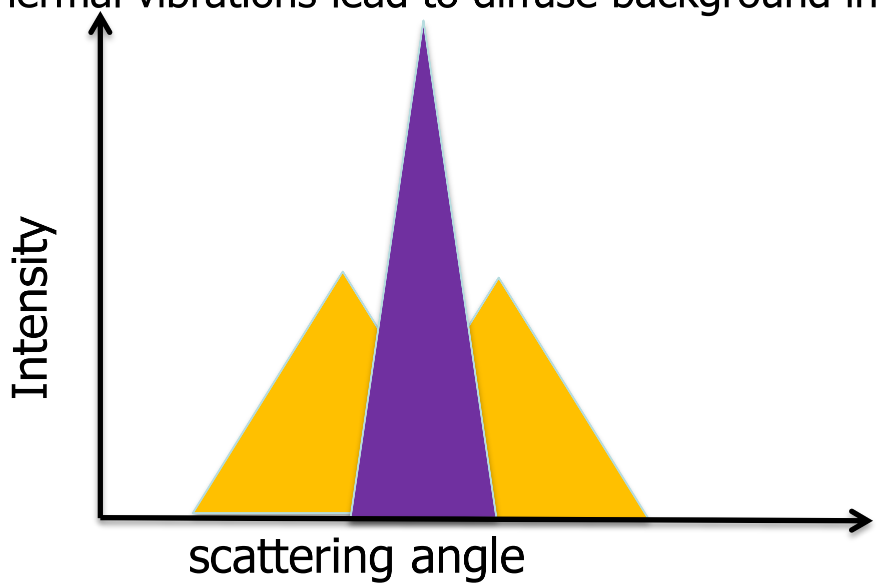
Atoms vibrate slightly

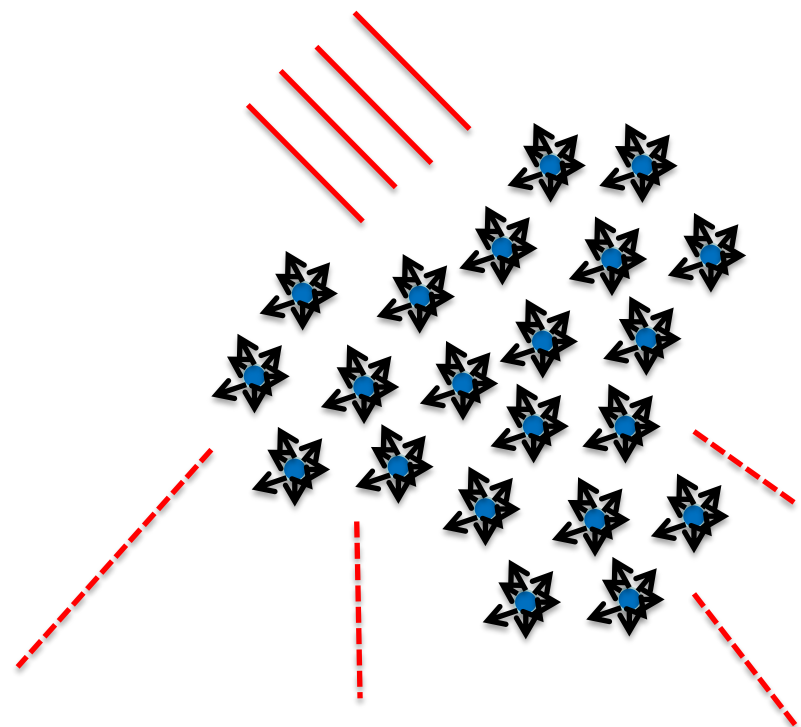
Einstein model: Every atom describes an **independent** oscillation in a harmonic potential.

Electrons are much faster (v~c) than the motion of vibrating atoms.

Each electron sees a snap shot of atoms randomly out of its equilibrium position

Thermal vibrations lead to diffuse background intensity

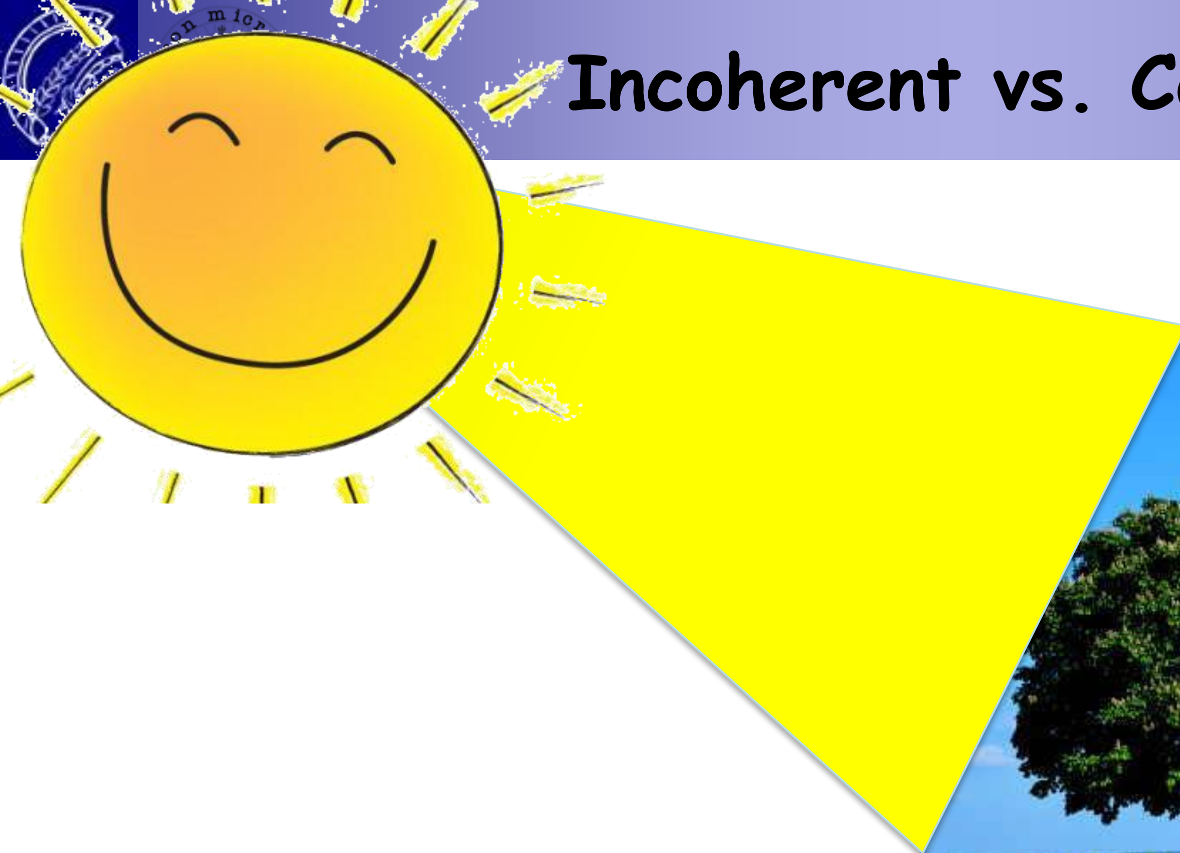




Outline

- Principles of STEM
- STEM Probe
- Ronchigram
- Detectors
- Incoherent vs. Coherent Imaging
- Examples
- Literature

Incoherent vs. Coherent Imaging

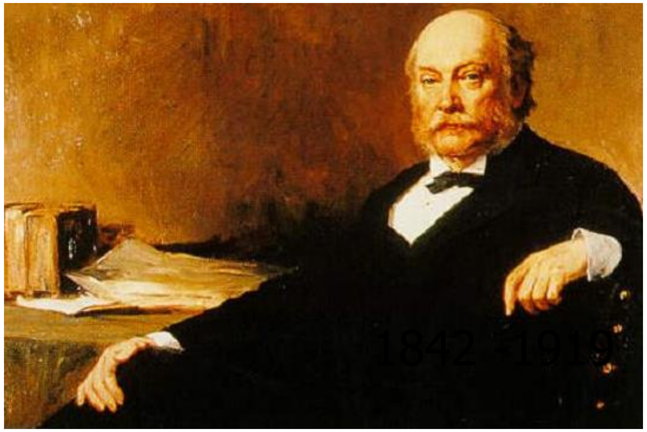


Incoherent imaging in nature

Coherence would lead to confusing interference effects!

Image simulation would be necassary!

Incoherent vs. Coherent Imaging

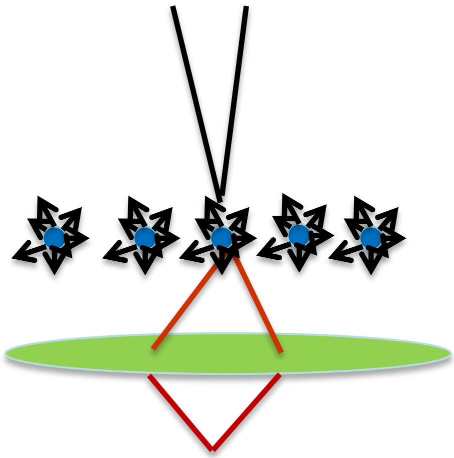


Lord Rayleigh
1842-1919

„The function of the condenser in microscopic practice is to cause the obeject to behave, at any rate in some degree, as if it were self-luminous, and thus to obviate the sharply-marked interference bands which arise when permanent and definite phase relationships are permitted to exist between the radiations which issue from various points of the object."

STEM

self-luminous object



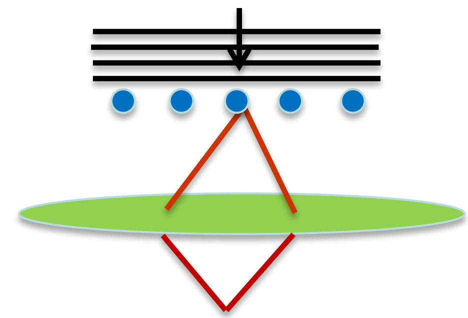
No phase relationship
(one atom column at one time)

No interference is observable

direct interpretation possible
(Z contrast)

TEM

plane wave



Permanent phase relationship between neighbours

Multi slit experiment

Interference occurs

No direct interpretation possible
(phase loss)

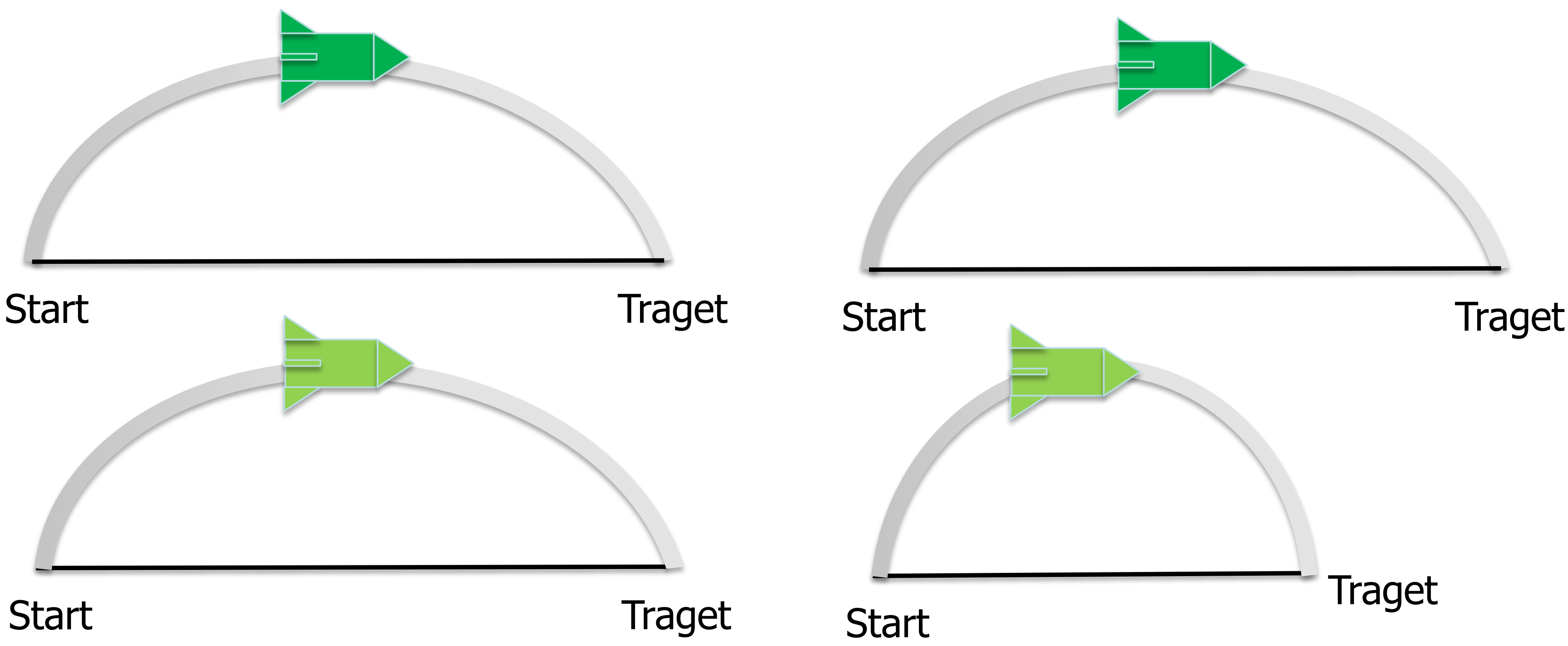
Incoherent Imaging gives significantly better resolution than coherent imaging

Nellsit et al. *Advances in Imaging and Electron Physics* 113, 147-203.

Incoherent vs. Coherent Imaging

same start and end point
same departure and arrival time
same velocity

Same start point, but
Different velocity and
Different end point



Coherent

Inherent

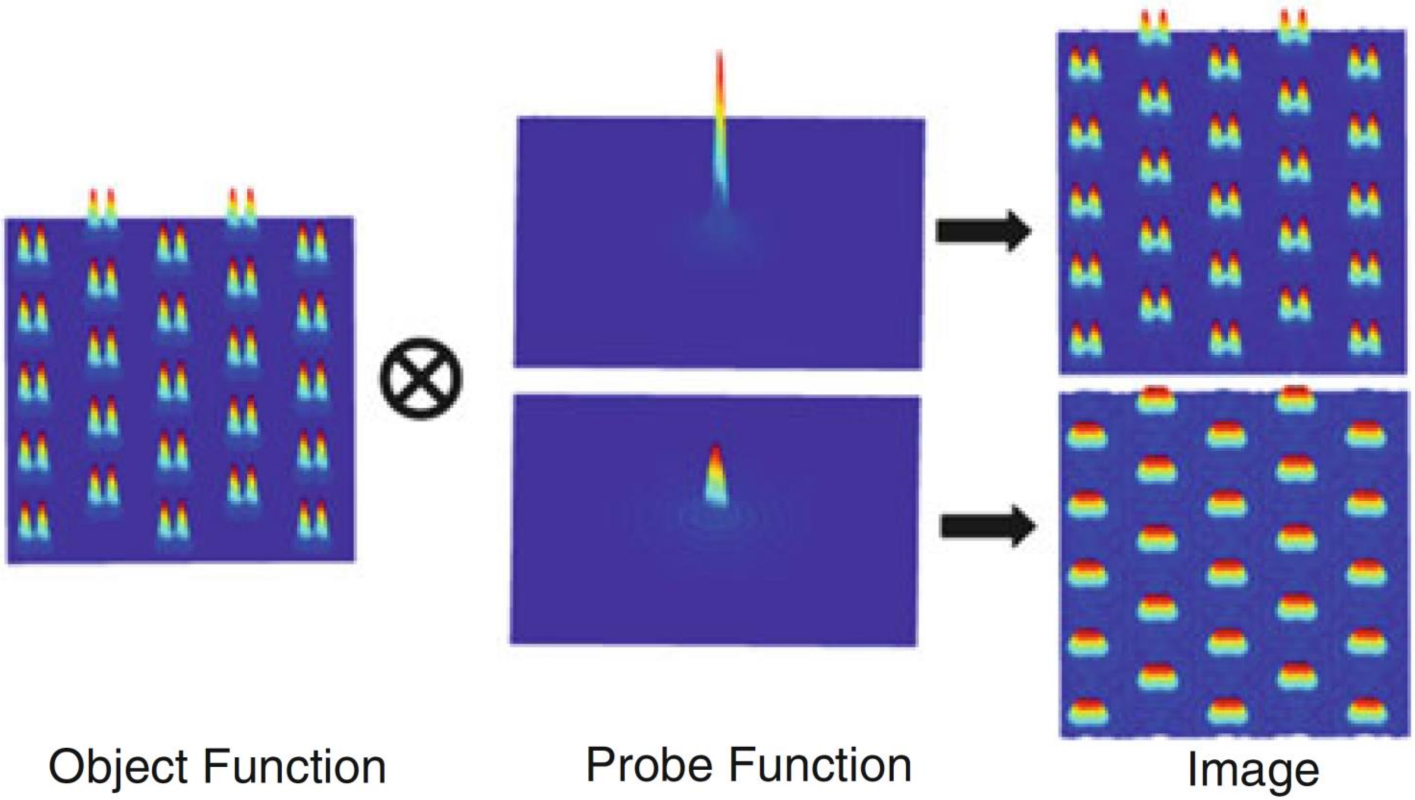
Shiojiri *J.Sci.* **2008**, 35, 495-520.

Incoherent vs. Coherent Imaging

Probe function (P(R)
Object function ($\psi(R)$)

STEM

$$I_{\text{incoh}}(\mathbf{R}) = |P(\mathbf{R})|^2 \otimes |\psi(\mathbf{R})|^2$$



$$I_{\text{coh}}(\mathbf{R}) = |P(\mathbf{R}) \otimes \psi(\mathbf{R})|^2$$

TEM

Vogt et al. – *Modelling Nanoscale Imaging in Electron Microscopy* 2012.

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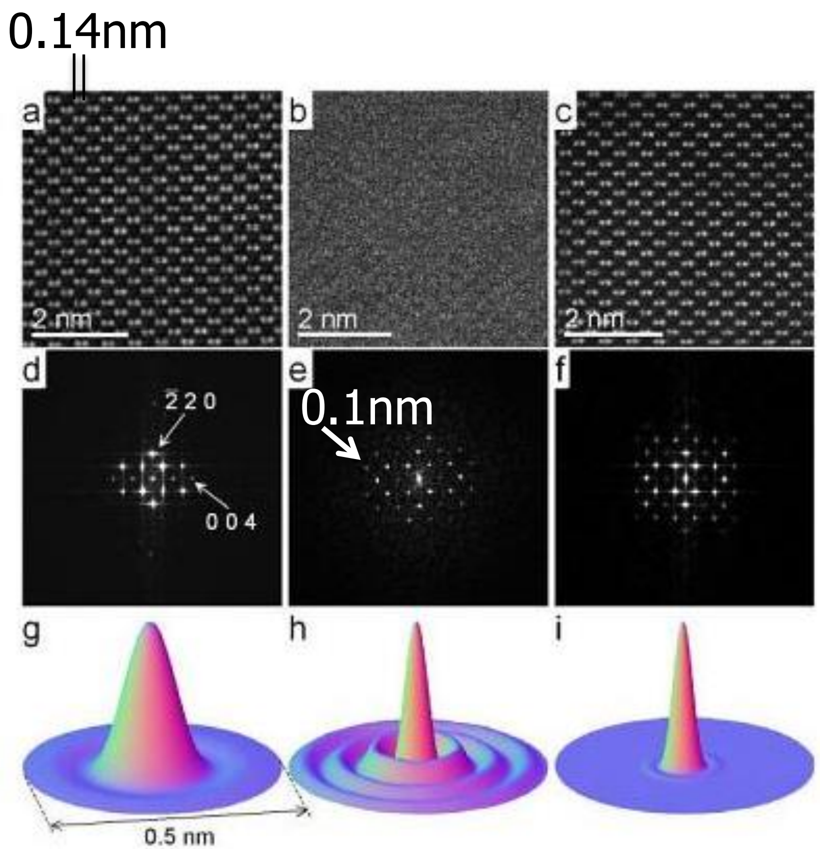
Example

C_s corrector

Si[110]
C_3=1.2 mm
300 kV



Scherzer condtion for incoherent imaging:
9 mrad
f=-200 nm
d=0.133 nm

15 mrad
f<-200nm
d=0.08 nm

close to super resolution mode

25 mrad
C_s corrected

Erni - *Aberration-Corrected Imaging in Transmission Electron Microscopy* 2010.

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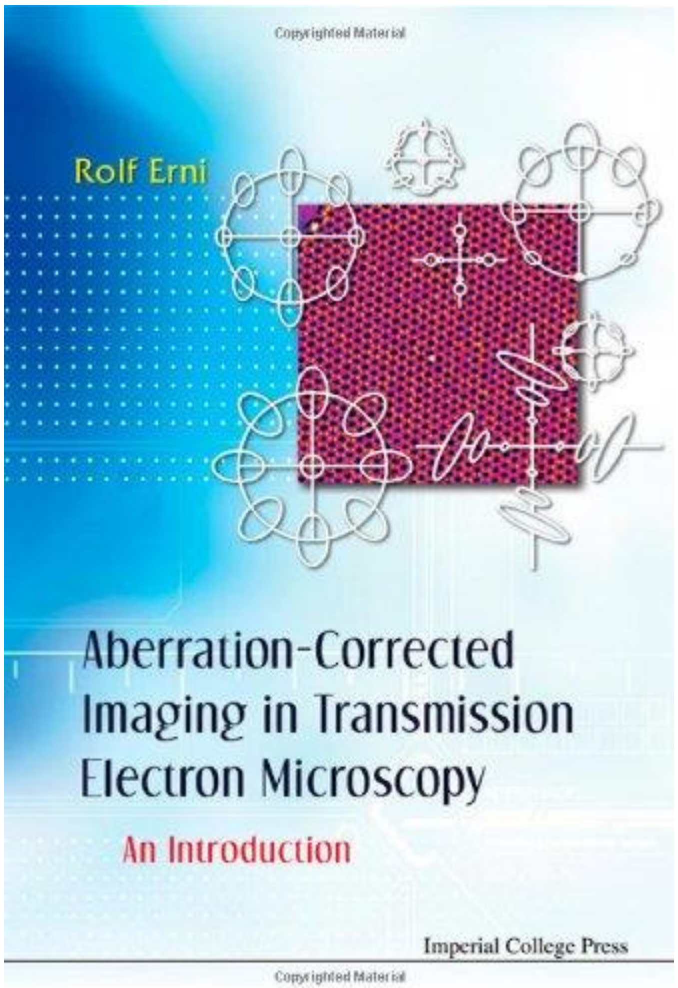
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An Introduction
Imperial College Press
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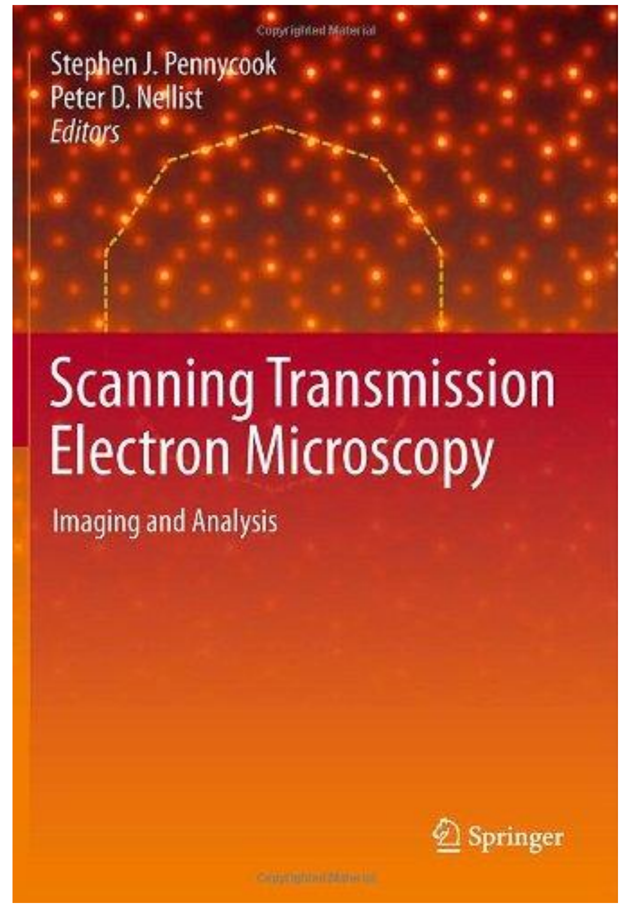
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