Transmission Electron Microscopy

Part VI: Scanning Transmission Electron Microscopy (STEM)

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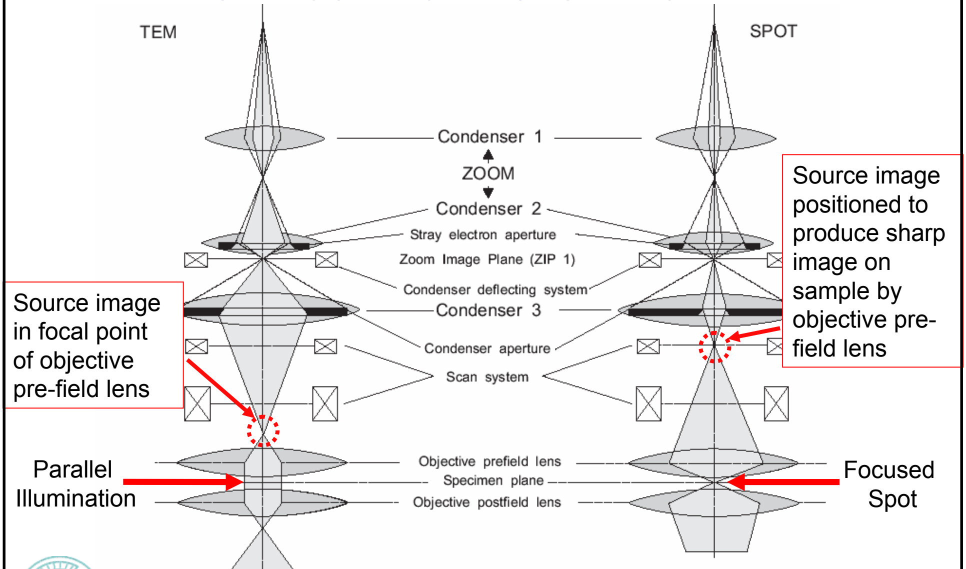
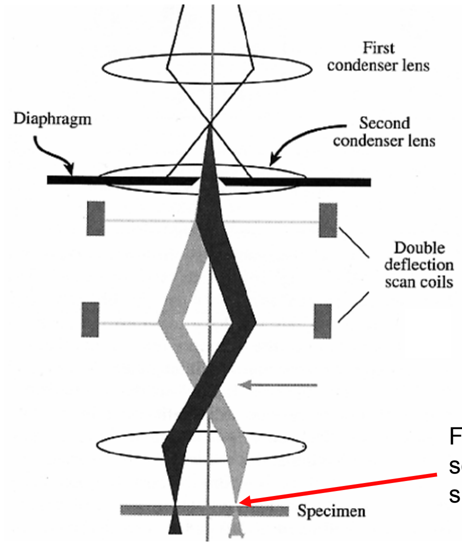
Websites: http://hrem.mpi-stuttgart.mpg.de/koch/Vorlesung/
http://hrem.mpi-stuttgart.mpg.de/koch/stem/

Reminder: Illumination in a TEM

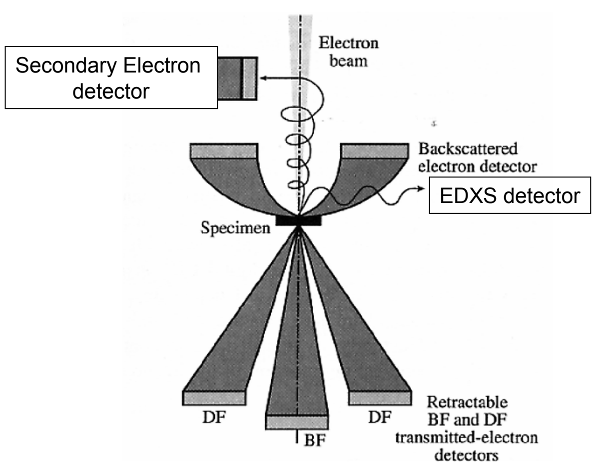


Example: 3-condensor system of a Zeiss Libra 200

STEM: Scanning a focused spot across the sample

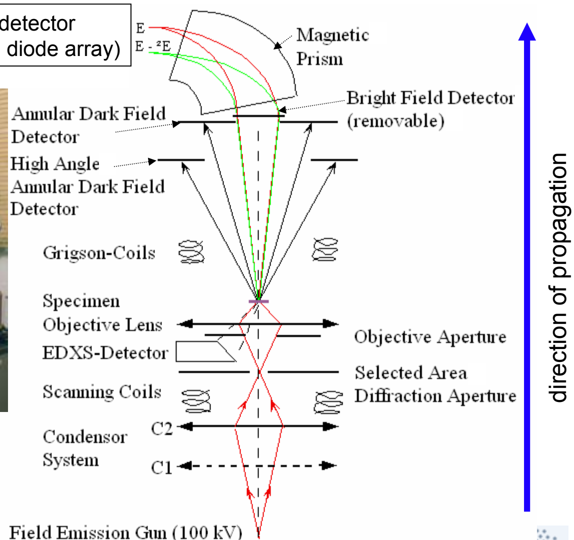


Detectors in a STEM



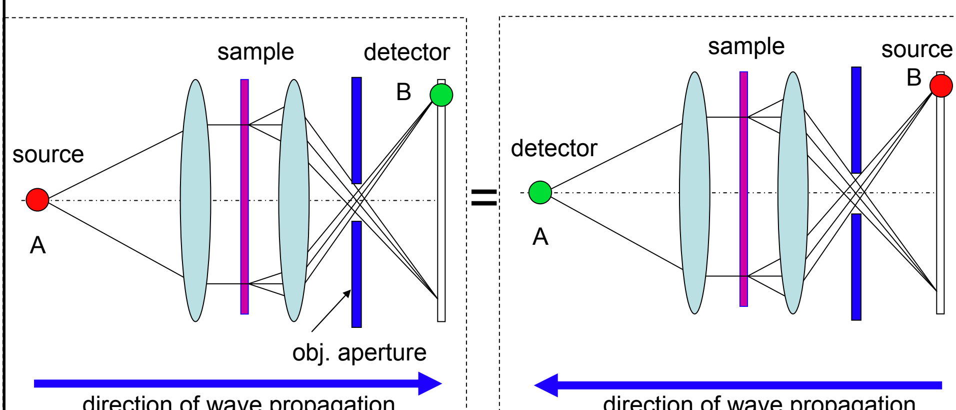
Normally, a STEM is equipped with several detectors which can collect their respective signals simultaneously. The BF detector may usually be replaced by an electron energy loss spectrometer (EELS).

Dedicated STEM: usually upside-down



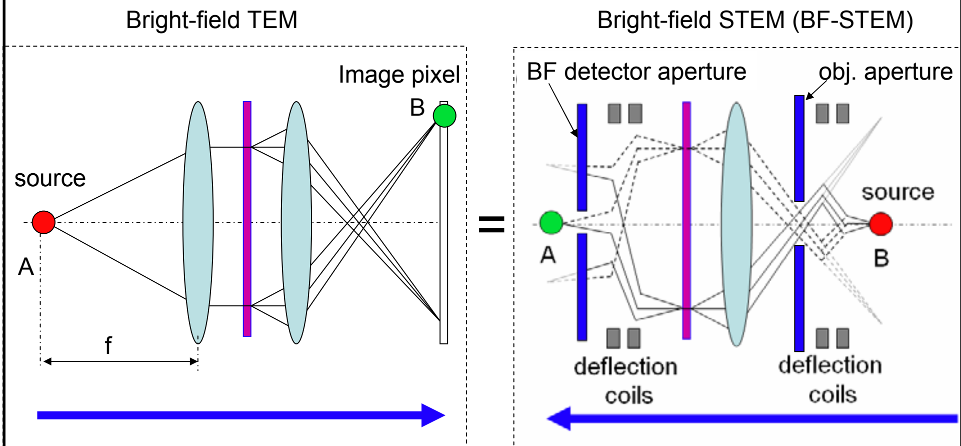
Heavy pumps needed for gun-UHV must be located at the bottom for stability reasons.

Reciprocity Theorem



Reciprocity Theorem (from geometric optics): “The amplitude of a wave at B due to an electron source at A is equal to the amplitude at A due to a source at B.”

Reciprocity Theorem: : Relating TEM and STEM

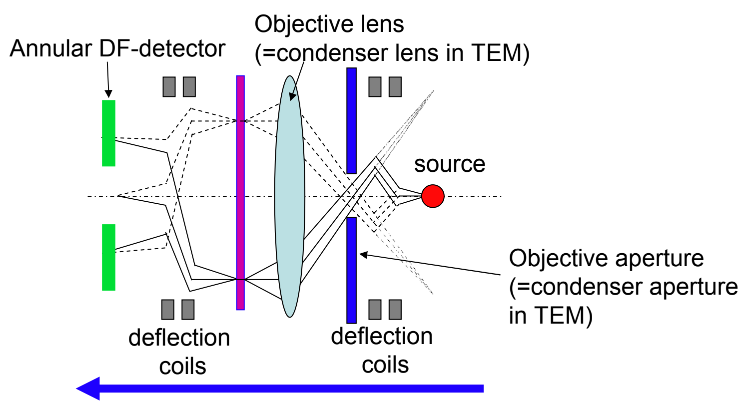


The scanning of the electron beam in a Scanning Transmission Electron Microscope (**STEM**) corresponds to the different pixel positions of a TEM image detector, if the range of scattering angles accepted by the bright-field detector equals the reciprocal-space source size.

(Note: the objective pre-field in the TEM performs a Fourier transform)

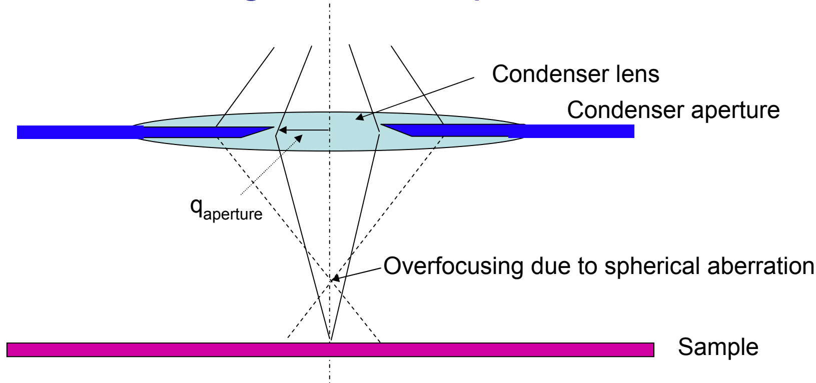
Annular Dark-Field STEM (ADF-STEM)

In Annular Dark-Field STEM (ADF-STEM) only those electrons which have scattered to high angles are collected.



Note: The analogue technique to ADF-STEM in the TEM is hollow-cone dark-field (HC-DF) TEM, in which the illumination angle is rotated about the optic axis during the exposure of the image.

Forming a focused probe



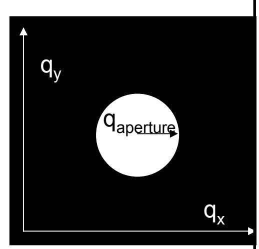
The probe is an image of the electron source. The probe size depends therefore on the same parameters as the resolution in a TEM image.

- Aberrations of the probe forming lens
- Spatial / temporal coherence (energy spread of electron beam / source size)
- Size of objective aperture

Condenser aperture = objective aperture in a dedicated STEM

Forming a focused probe

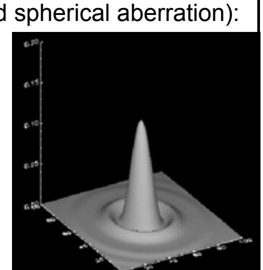
Electron wave in plane of condenser lens (reciprocal space):



$$\Psi(\vec{q}) = \begin{cases} 1, & |\vec{q}| \le q_{aperture} \\ 0, & |\vec{q}| > q_{aperture} \end{cases}$$

Phase shift by condenser lens (reciprocal space, defocus and spherical aberration):

$$\Psi_{aberrated}(\vec{q}) = \Psi(\vec{q}) \cdot \exp\left[-i\pi\left(\lambda\Delta f|\vec{q}|^2 + \tfrac{1}{2}\lambda^3 C_s|\vec{q}|^4\right)\right]$$



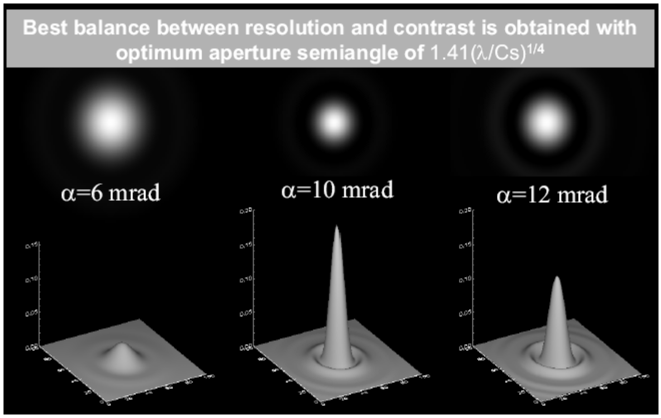
Probe current distribution on sample (real space)

$$I(\vec{r}) = |\Psi_{aberrated}(\vec{r})|^2 = FT^{-1}\left[\Psi_{aberrated}(\vec{q})\right]$$

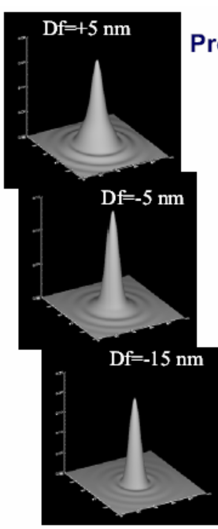
Choosing the optimum aperture size

Size of condenser aperture defines convergence semiangle

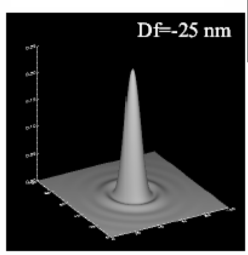
$$\alpha = \sin\left[\lambda q_{aperture}\right] \approx \lambda q_{aperture}$$

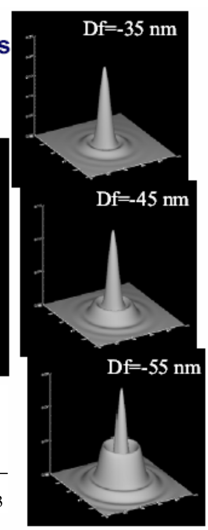


Choosing the proper defocus



Probe intensity profiles
(α=10 mrad)
200kV, Cs=0.48mm



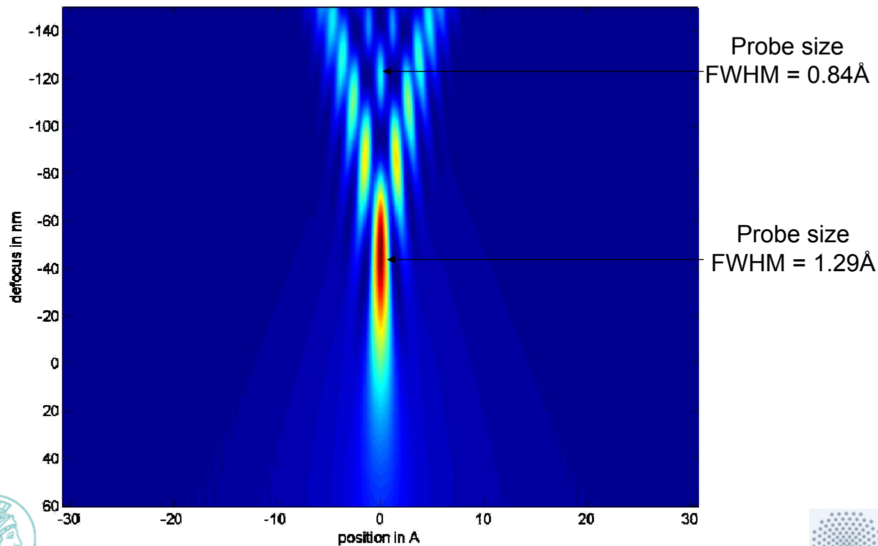


Df = deviation from Scherzer focus

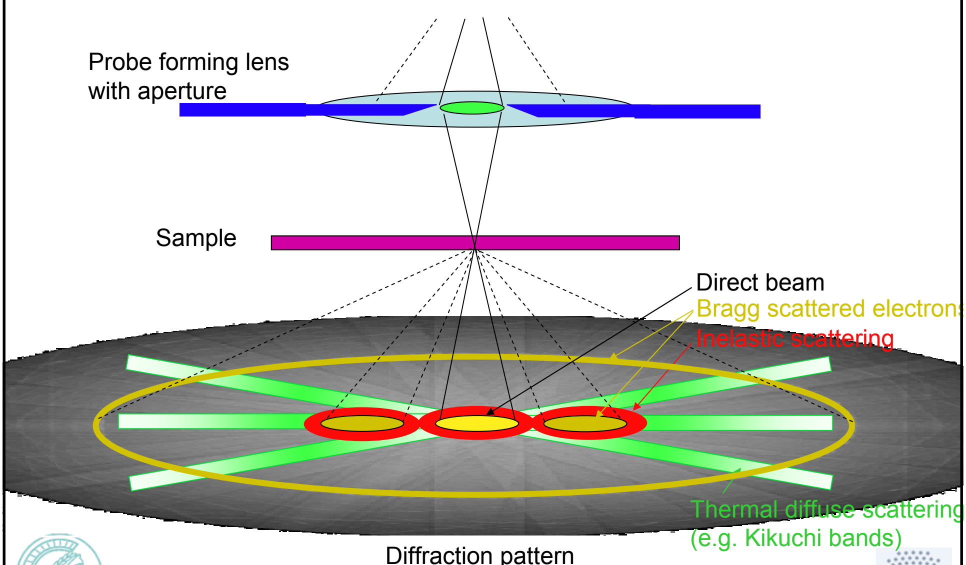
$$\Delta f_{Scherzer} = -1.2\sqrt{\lambda C_3}$$

Defocus map of a STEM probe

X-Z- cross section through a perfectly coherent probe with α=12mrad, E_0=200kV



Signals in a STEM

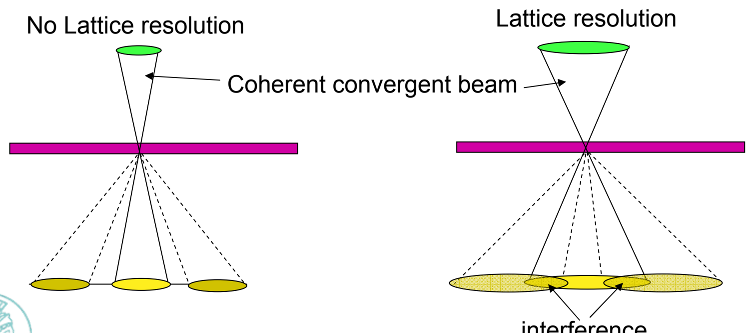


STEM Images

- Elastic BF-STEM images are equivalent to TEM bright-field images (reciprocity principle). They are mainly produced Bragg disks hitting the detector. They contain diffraction contrast and are therefore very sensitive to strain/diffraction conditions in the material.
- High-angle annular dark-field STEM (HAADF-STEM) images are mainly produced by thermal diffuse scattering (TDS), because at high scattering angles TDS has the highest scattering cross section.
- (medium angle) ADF-STEM images contain both, Bragg diffraction and TDS contributions.

Lattice resolution in BF-STEM

Being able to image the crystal lattice in STEM requires the probe to be small enough to sample details within a single unit cell of the crystalline material that it is probing. As the probe scans over a single unit cell, the diffraction intensities in each disc remain constant, only the phase of the diffracted beams change. BF lattice contrast is therefore only possible, if the condenser aperture is large enough to make Bragg discs overlap and the beam coherent enough to allow interference in the overlap region between neighboring beams.
An alternative argument is the reciprocity principle.



HAADF-STEM Image Simulation

Quantitative Image interpretation requires comparison of experimental data with simulations.
2 (out of several more) ways to simulate HAADF-STEM images are:

- Incoherent Imaging Model (Not quite correct, but helps to interpret most images): The Image is the convolution of object potential and probe intensity.

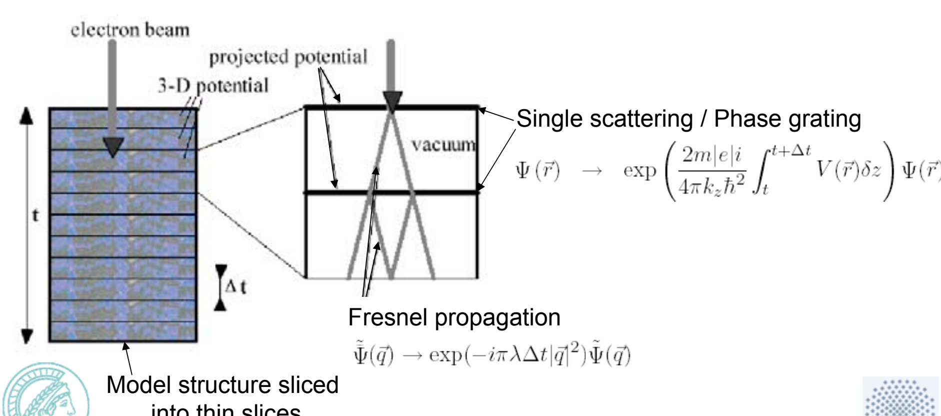
$$I_{image}(\vec{r}) = I_{probe}(\vec{r}) \otimes V_{proj}^2(\vec{r})$$

- Multiple Scattering Image Simulation (Quantitative agreement between simulation and experiment): For each beam position a complete multislice dynamical scattering simulation is performed, applying the frozen phonon approximation.

Note: The HAADF-STEM signal consists to a large part of thermal diffuse scattering (**TDS**). It is therefore essential for quantitative simulations to include TDS.

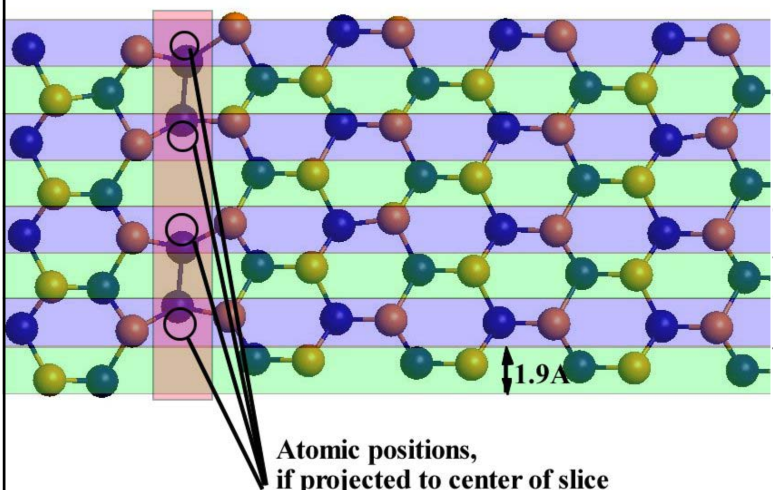
Dynamical Scattering Computation: Multislice algorithm

The multislice algorithm slices the 3-dimensional structure into thin slices, thin enough to make the phase grating approximation valid for each individual slice. It then iterates the following procedure:
1. Phase shift (and absorption) by the projected potential of the current slice.
2. Fresnel propagation through vacuum to the next slice.



Exact Computation requires 3D potential

•Frozen phonon approximation for TDS simulation includes z-position.
•Electron propagation is always normal to potential slices.
•Slices can be extremely thin.

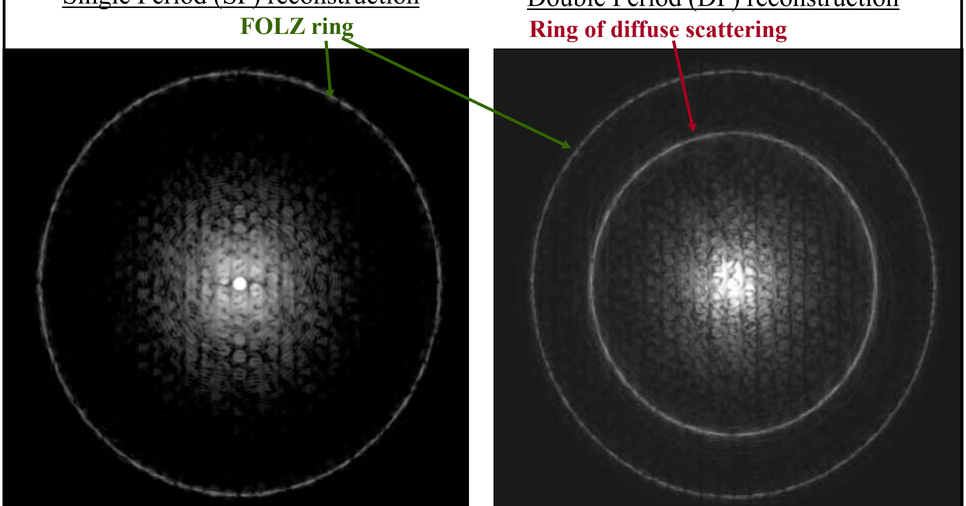


Model of a double period (DP) reconstructed partial dislocation core in silicon:
The dislocation core has a periodicity twice that of the lattice.

CBED patterns of 90° partial dislocation core

Single Period (SP) reconstruction

Double Period (DP) reconstruction

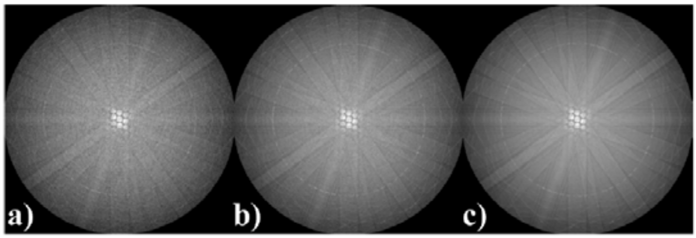


Simulation done with multislice, Debye-Waller factor: 0.44A^2, thickness: 460A (log-scale)

Computing the Thermal Diffuse Scattering

Within the **Einstein Model** for thermal vibration of atoms, every atom describes an **independent oscillation** in a harmonic (square well) potential. The electrons are much faster (v ≈ c) than the motion of vibrating atoms. Each electron therefore "sees" a **stationary "snap shot"** of the crystal with each atom randomly out of its equilibrium position.

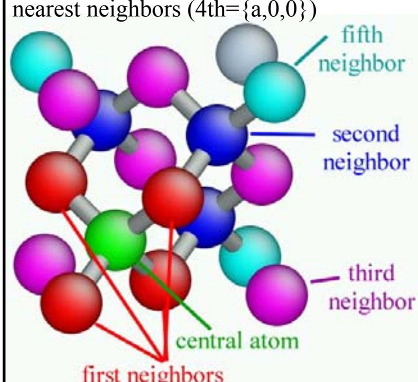
TDS may be simulated by **averaging the diffraction intensities** of several diffraction pattern of different such "snap shots".



Multislice CBED simulations of Si (110) including thermal diffuse scattering (TDS) according to the Einstein model. The sample thickness is 960Åthickness, beam energy 100kV, and the beam divergence angle α is 6 mrad. The pattern is plotted after a) 1, b) 4, and c) 20 iterations and becomes less and less noisy and finally converges.

Beyond the Einstein Model

Si unit cell indicating 1st through 5th nearest neighbors (4th={a,0,0})

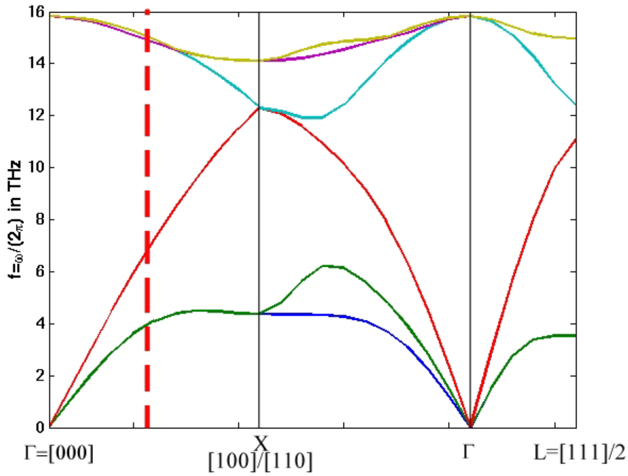


- Vibrational frequencies and modes are eigenvalues and eigenvectors of the **Dynamical Matrix** D [1,2].
- D is determined by the harmonic **force constants** (spring constants between atoms) for up to 5th order neighbors, which have been fitted to experimental phonon dispersion curves [3,4], obtained by neutron diffraction.
- Phonon modes are occupied according to **Bose-Einstein statistics**.

$$E_{mode} = \left\{ \frac{1}{\exp\left[\hbar\omega(\vec{q}, n_{Branch})/k_B T\right] - 1} + \frac{1}{2} \right\} \hbar\omega(\vec{q}, n_{Branch})$$

•[1] Born, Huang, "Dynamical Theory of Crystal Lattices", Oxford Univ. Press, London (1954)
•[2] Warren, "X-ray Diffraction", Dover, New York (1990)
•[3] Jiang et al, Solid State Comm. **86**, 731 (1993)
•[4] Hermann, J. Phys. Chem. Solids **8**, 405 (1959)

Vibrational modes and Eigenfrequencies

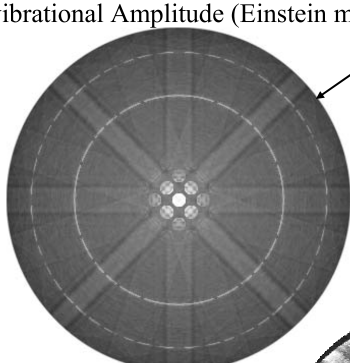


Si phonon dispersion curve fitted to exp. data

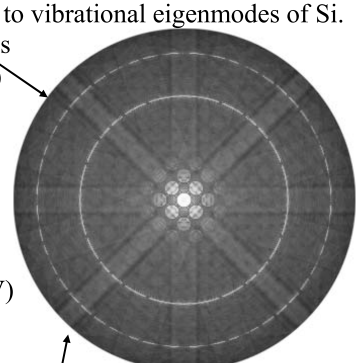
Vibrating Atoms => Thermal Diffuse Scattering (TDS)

Independent vibrations with same RMS vibrational Amplitude (Einstein model)

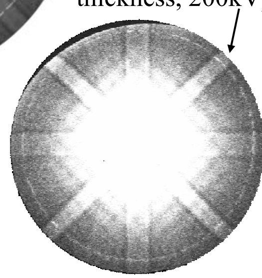
Atomic displacement according to vibrational eigenmodes of Si.



Simulated images (frozen phonon) log scale t=298Å 200kV

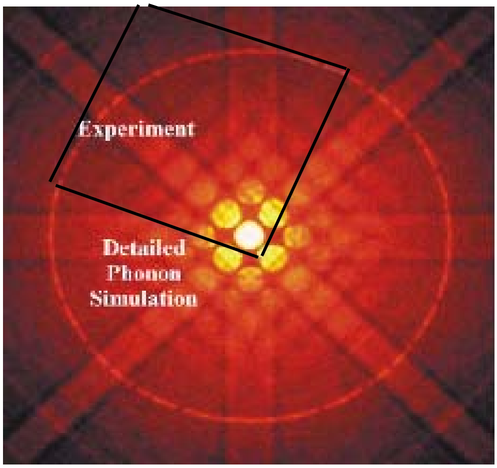


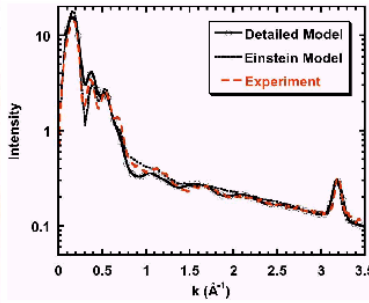
Experimental image (unknown thickness, 200kV)



Different thickness, therefore no quantitative agreement, but see Muller et al. Ultramicr. **86**, 371 (2001)

Frozen Phonon method seems quantitative





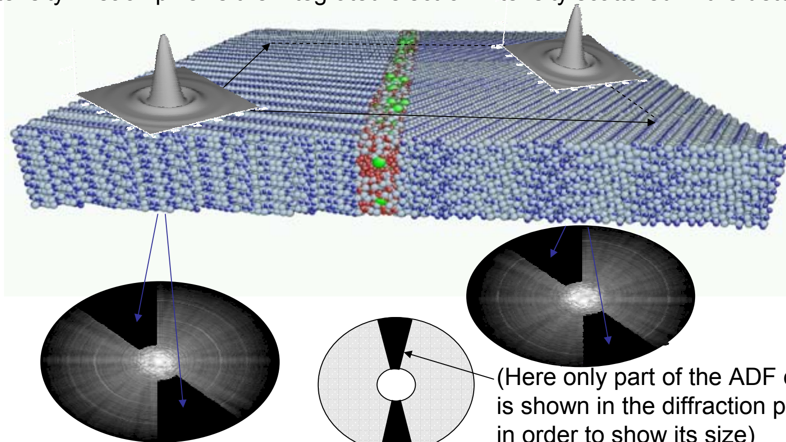
Calculations agree perfectly with experiment !

D. Muller, et al., Ultramicrocopy 86, 370 (2001)

The HAADF signal for a given beam position consists of the sum of the scattering intensity at a certain range of scattering angles.

STEM Simulation

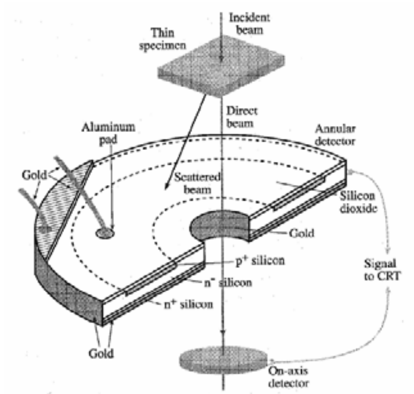
A diffraction pattern is computed for each pixel position using the Multislice algorithm.
The intensity in each pixel is the integrated electron intensity scattered in the detector area

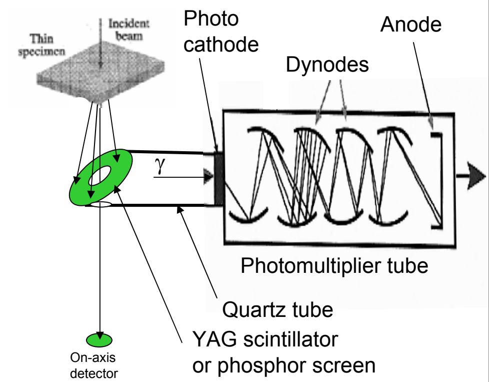


(Here only part of the ADF detector is shown in the diffraction patterns, in order to show its size)

The **effective source size** (partial spatial coherence) is included by convoluting the simulated STEM image by a Gaussian of the respective width.

ADF Detectors

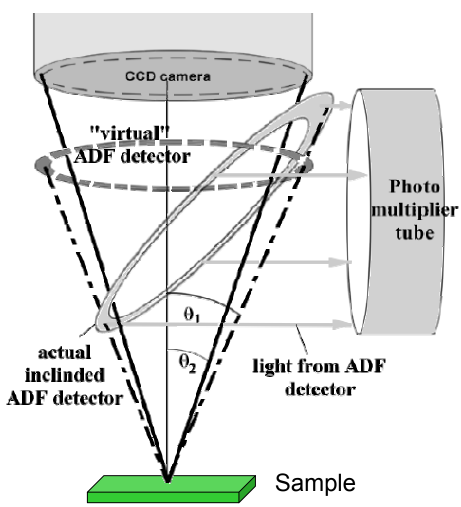




Silicon detector:
3,6 eV energy loss per electron-hole pair (exciton) ⇒ 100 keV-electron creates about $3 \cdot 10^4$ excitons.

Scintillator – Photomultiplier detector:
Photon creation by ionisation
Advantage: very fast response

Microdiffraction STEM: ADF + Diffraction Pattern



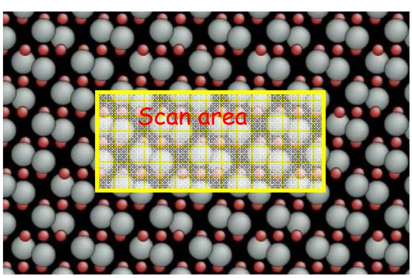
Perfect setup for Ptychography, a holographic method, which promises diffraction-limited resolution (i.e. <<1Å). …

STEM image signal

- STEM images are acquired **sequentially**, i.e. pixel by pixel. The precision of the image therefore depends on the reproducibility of the beam positioning.
- The emission current of some FEG sources (cold FEGs) may fluctuate. This leads to a **fluctuation** of counts between pixels.
- The signal recorded from the detector is usually amplified with an adjustable bias (threshold) and gain (amplification). This makes it often **impossible to quantify** the number of electrons per pixel in a given STEM image.
- Less than 10% of the incident electrons scatter to the HAADF detector (depends on detector geometry, of course). This makes HAADF images **very noisy**. However, the very high contrast of HAADF images compensates for some of the noise.

Example simulation: Al_2O_3

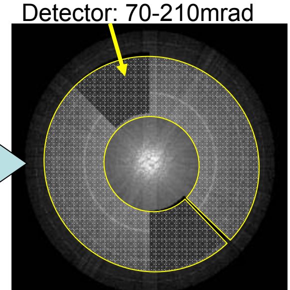
Al_2O_3 structure



Detector: 70-210mrad

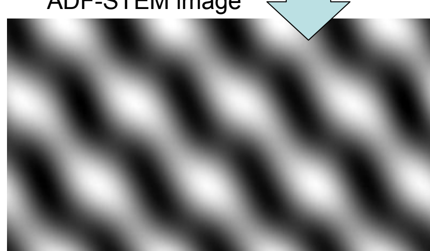
1 diffraction pattern

per beam position



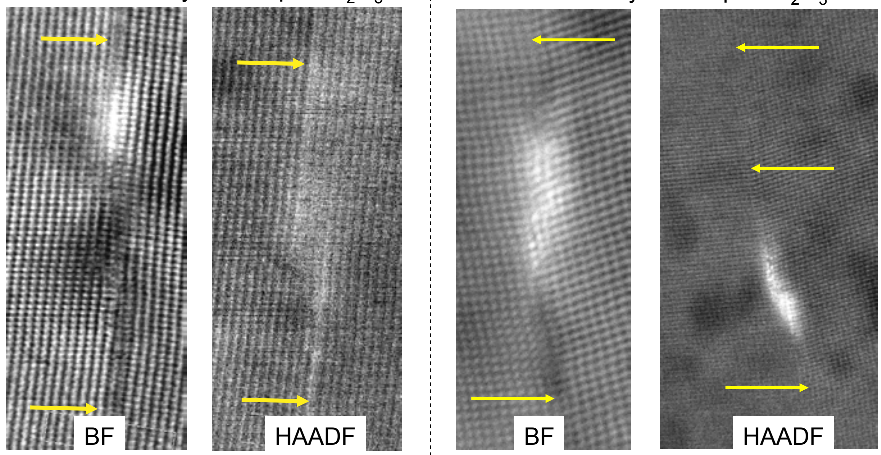
Since the probe tails extend over a few nm, the potential area must be larger than the image.

ADF-STEM image



HAADF-STEM better Z-contrast than BF-STEM

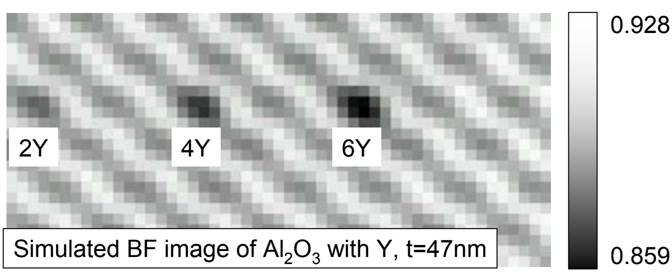
Twin Boundary in undoped Al_2O_3 | Twin Boundary in Y-doped Al_2O_3



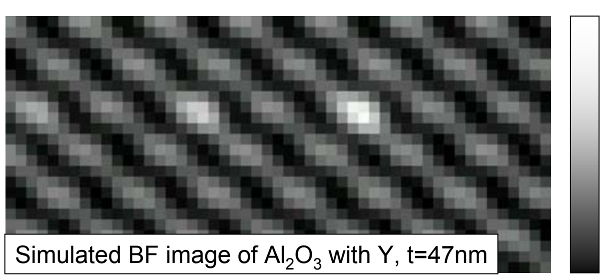
Images: D. Bouchet, C. Colliex, A. Bleloch

HAADF-STEM is much less sensitive to local diffraction conditions than BF-STEM. Because of its sensitivity mainly to the atomic number, it is also called Z-contrast STEM.

Simulation of Y in Al_2O_3



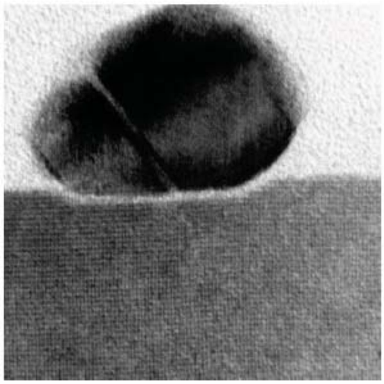
Simulated BF image of Al_2O_3 with Y, t=47nm



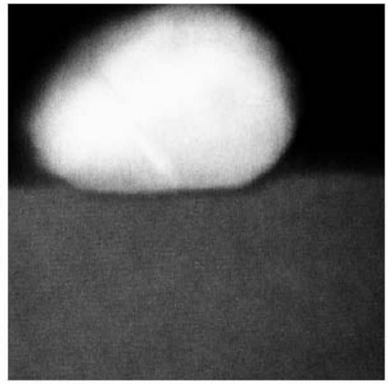
Simulated BF image of Al_2O_3 with Y, t=47nm

The fraction of electrons hitting the HAADF-detector is very small!
But contrast is very good.

Example Pd particle on $SrTiO_3$



BF-STEM image
(contains diffraction contrast)

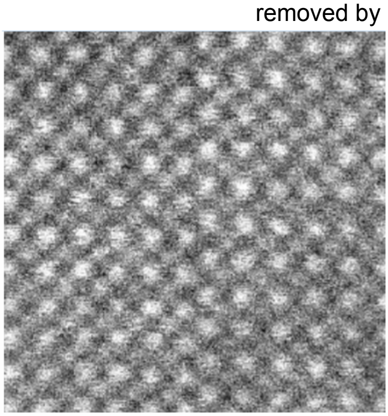


HAADF-STEM image
(chemical sensitivity because of Z-contrast)

Images: M. Ceh, S. Sturm, E. Tchernikova, Ljubljana

HAADF image: scan distortions

The sequential acquisition of STEM images causes artifacts which must be removed by image processing.



Raw image

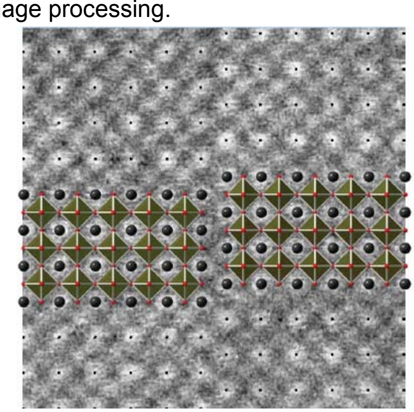
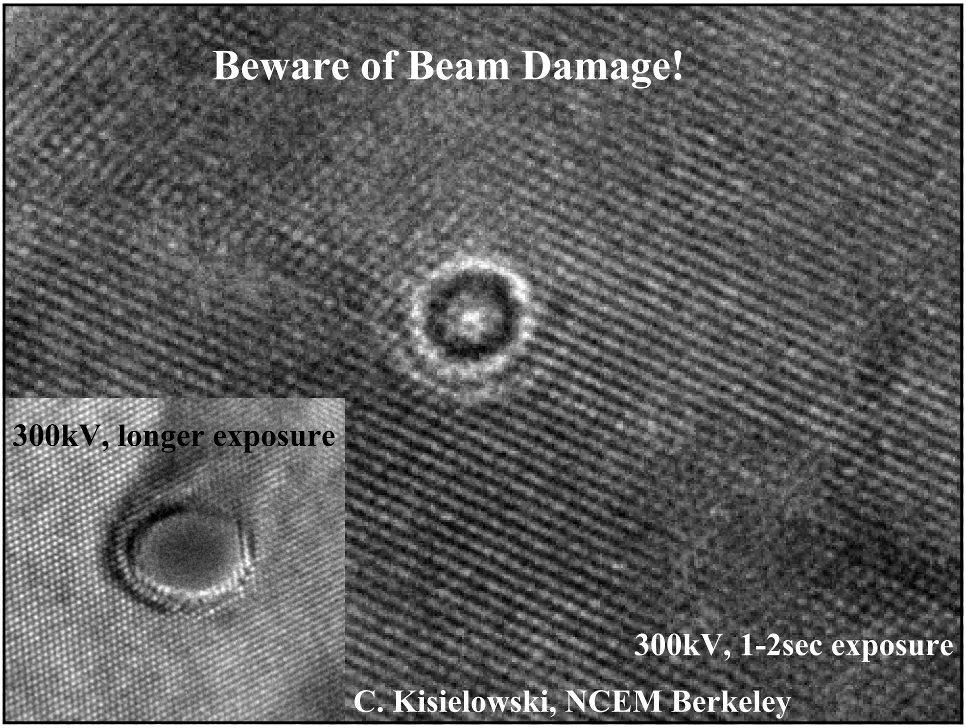
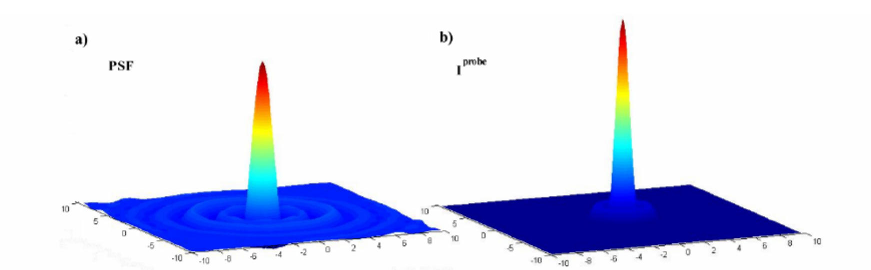


Image after warping (adjusting for scan distortions), including structural model.

Images: M. Ceh, S. Sturm, A. Recnik, Ljubljana



Resolution in STEM √2 better than TEM



2-dimensional impulse response function ($PSF(\vec{r})$) and STEM probe function $I^{probe}(\vec{r})$ for the same microscope parameters ($E = 200\text{kV}$, $C_s = 0.7\text{mm}$, $\alpha = 12.5\text{mrad}$, $df = -513\text{Å}$). The scale in x- and y-direction is in Å.