ÉCOLE POLYTECHNIQUE
FÉDÉRALE DE LAUSANNE

High Resolution Transmission Electron Microscopy (HRTEM)

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Doctoral Course MS-637

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Centre
Interdisciplinaire
de Microscopie
Électronique

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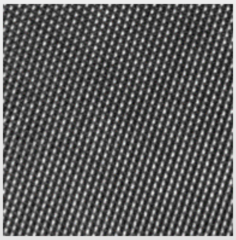
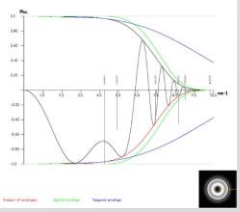
Summary



Contrast in TEM images results from the scattering of electrons in thin samples and changes in the phase of the electron waves. When 2 or more electrons beams scattered from the sample interfere and are transferred at high magnification to detector plane, there can form an interference pattern that we commonly refer as a high resolution TEM image. The phase shift of the electron waves can be used to map the atomic structure of the sample which appear as fringes (2 scattered beams) or white and dark spot patterns (>2 beams). The contrast mechanisms are difficult to interpret being very sensitive to many factors such as thickness, orientation and scattering of the sample, objective lens focus and aberrations, electron beam coherence and convergence angle on the sample. As such, we need simulations to interpret them properly.

HREM TEM simulations are needed to interpret the contrast mechanisms and complicated **Interference Patterns of HRTEM IMAGES.** Through careful experimentation, image acquisition and simulation of HRTEM images, we can reconstruct the projected potential of the specimen and determine its atomic structure. The common method for simulating HRTEM images is the multi-slice calculation in which the sample volume is sliced into sections and the associated phase shift due to scattering from the sample's crystal structure is sequentially calculated for each slice. The calculation can take into account the microscope performance and experimental conditions, such as aberrations, objective lens defocus and sample thickness. Because the HRTEM phase contrast is very sensitive to defocus and sample thickness and it is difficult to known these parameters experimentally with the required precision, we can simulate montages of HRTEM images that illustrate the variance in the pattern with defocus and thickness which can be compared to the experimental data.



Outline:

This section of the course gives a brief introduction to HRTEM imaging and multi-slice simulations which are used to interpret complicated patterns occurring HRTEM images

1) Introduction to Phase Contrast and HRTEM imaging
   A. What is a HRTEM image?
   B. HRTEM using Modern TEMs at CIME
   C. Complication of using real (imperfect) lenses
2) HRTEM simulations
   A. The problem
   B. Multi-slice calculations
   C. Weak Phase Object Approximation (WPOA)
   D. Transfer function
   E. Contrast Transfer Function (CTF)
3) Aberration Corrected HRTEM

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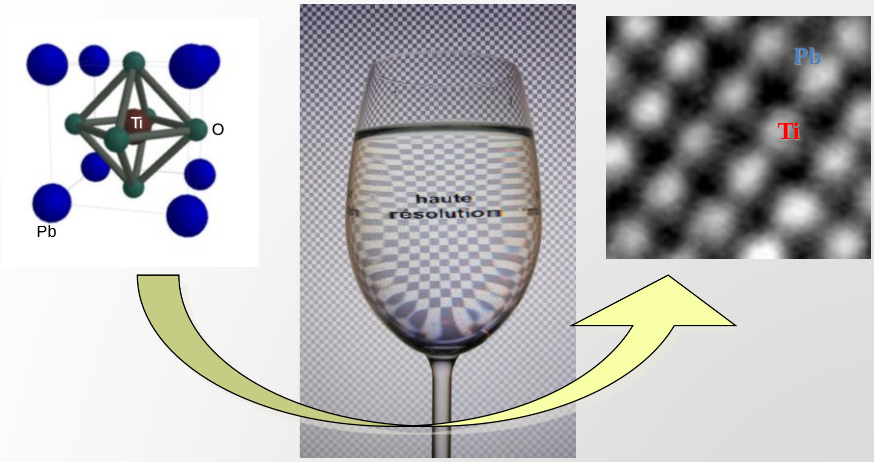
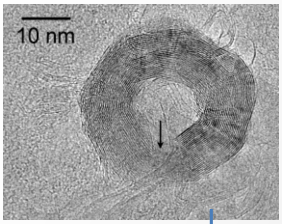
1) What does HRTEM imaging tell us about our samples?



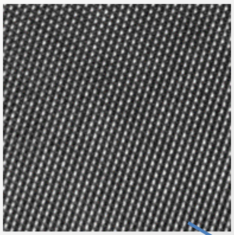
Information about the atomic structure of materials, interfaces, defects, etc…

1) Basic concept of Phase Contrast and HRTEM image formation

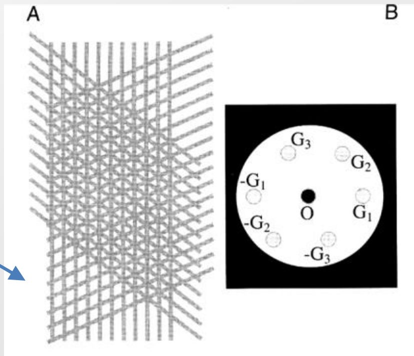
Phase Contrast or HRTEM image requires the more than one selected scattered beam, which interfere to produce a fringe, spot or more complex mosaic patterns

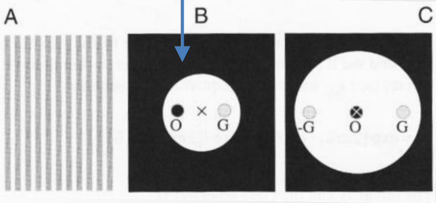


Graphite shell



Si [110]



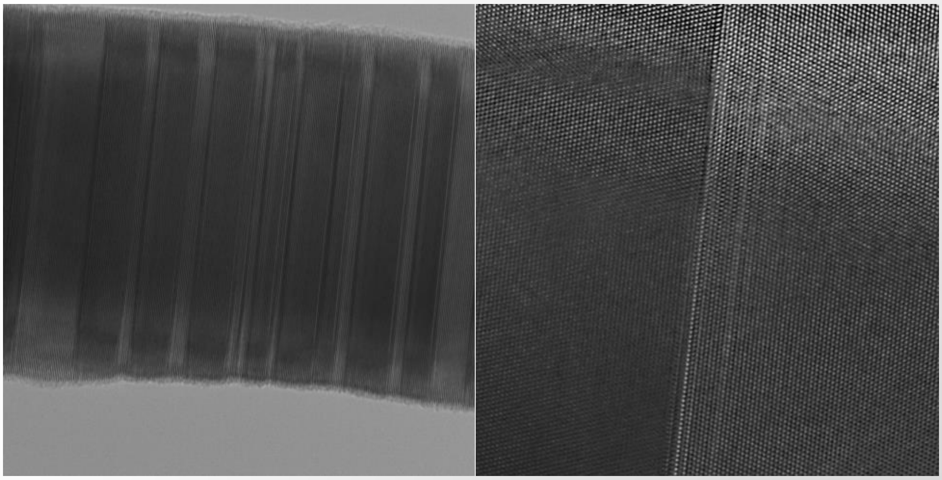


Lattice fringes are NOT direct images of the atomic structure, but carry information about the lattice spacing

1) Modern TEMS at CIME: Talos

Talos F200S at CIME-EPFL	
Electron Gun	S-FEG
Lens System	Constant Power
EDS System	2-SDD
Specimen loading	Remote with touch screen
STEM HAADF resolution	0.16 nm
TEM Information limit	0.12 nm
TEM point to point resolution	0.24 nm
Camera	CMOS (CETA)

1) HRTEM examples from the Talos: imaging nanowire defects



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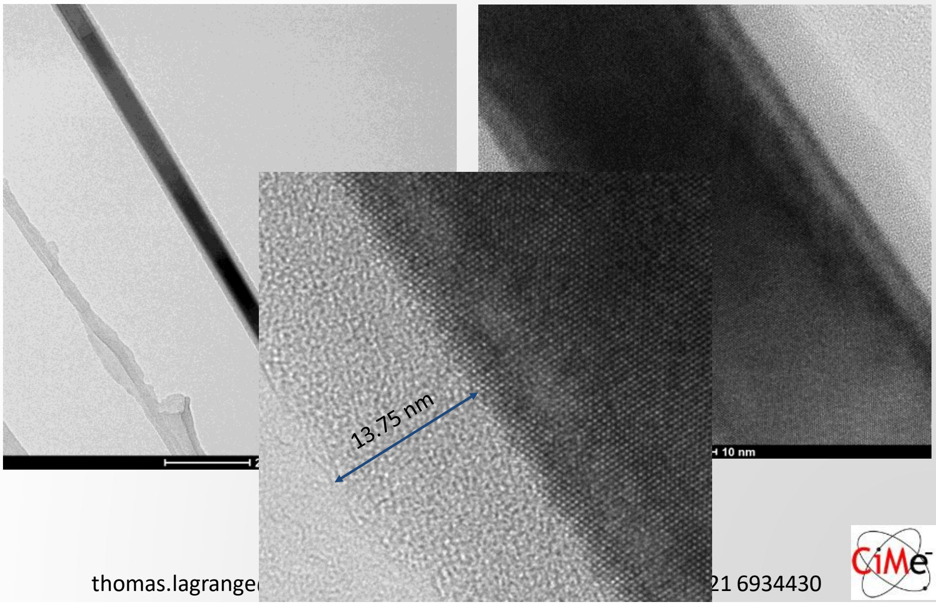
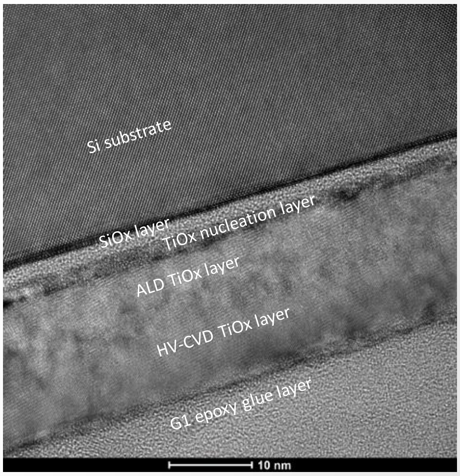
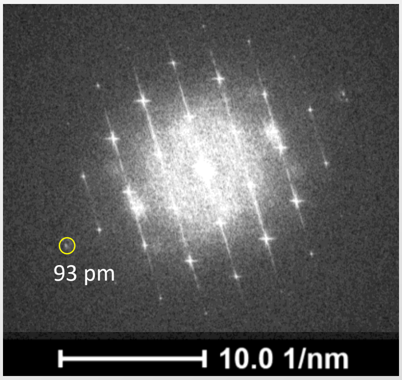
1) HRTEM examples from the Talos: imaging nanowire oxide coating thicknesses



1) HRTEM examples from the Talos: imaging layer thickness and defects in thin film coatings processed with different techniques





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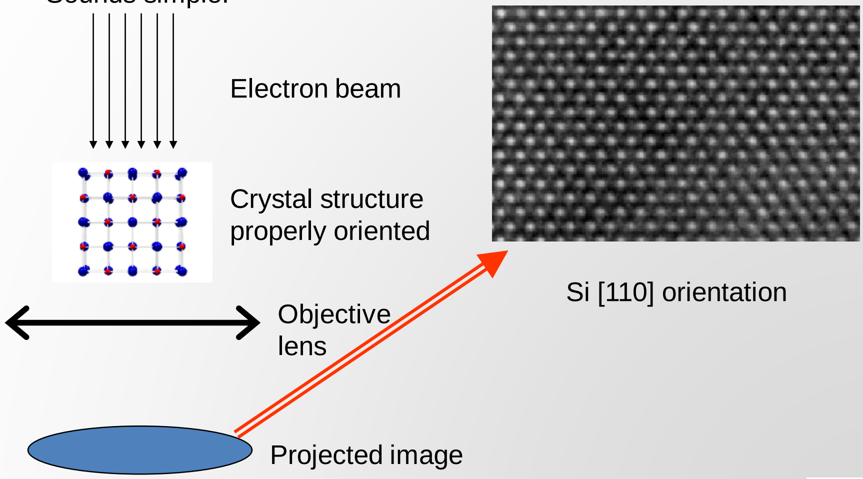
1) Problem: Phase Contrast images are not always direct projections of the atomic structure

Phase contrast for crystalline specimen

Sounds simple:


Electron beam

Crystal structure properly oriented

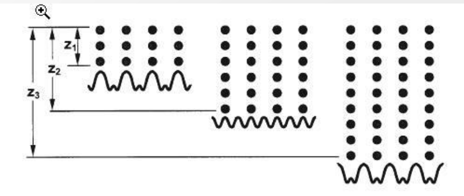
Objective lens

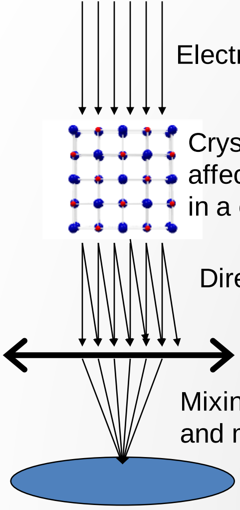
Projected image

Si [110] orientation

1) Problem: Phase Contrast images are not always direct projections of the atomic structure

Unfortunately things are much more complicated





Electron beam

Crystal thickness with a given orientation affects phase and amplitude of the beam in a complicated way

Direct and Diffracted beams (Phase shift!)

Objective lens (with aberrations affects phase shifts)

Mixing of information from the sample and microscope via INTERFERENCES!

Projected **amplitude** after **intereference**!

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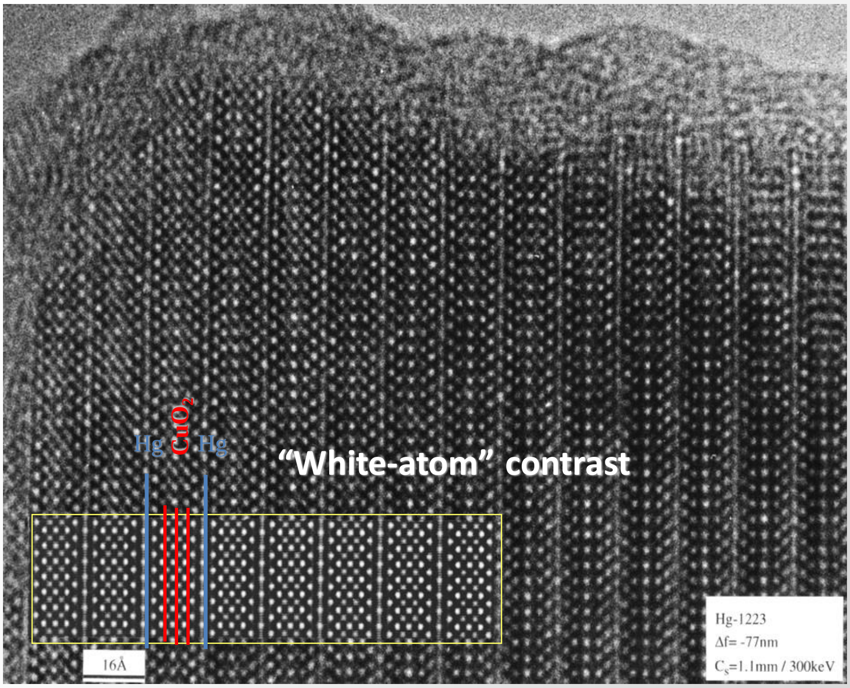
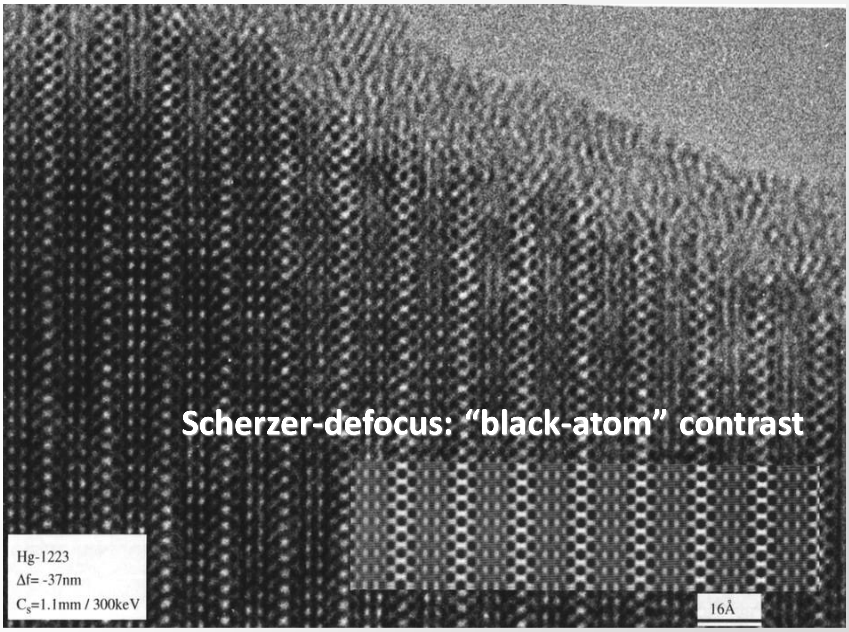


“White-atom” contrast



Scherzer-defocus: “black-atom” contrast

2) Why HRTEM perform image simulations?

HR(S)TEM ==⇒ to acquire knowledge on observed material (oriented in particular [uvw] directions):

- Specimen structure
- Chemical composition
- Functional properties
- …

But HR(S)TEM images depend of several adjustable microscope parameters, lens aberrations, sample thickness…for example,

Objective defocus strongly affects HRTEM images

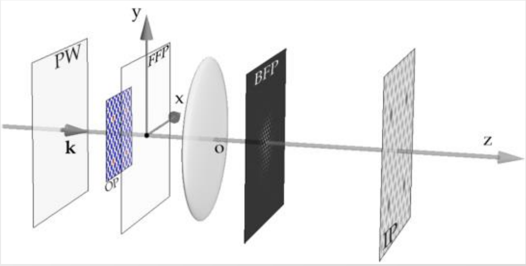
2) Formation of HRTEM images involves complex physical processes

Approximations and models of these physical processes are required in order to perform computer simulations.

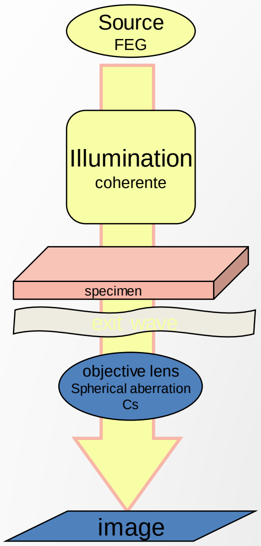
Models are based on electron scattering, diffraction, optics, ...

Needed: crystallography, optics, quantum mechanics, ... And appropriate computer programming – e.g.- JEMS

- **Source:** *coherent and monochromatic*
- **Illumination:** *parallel*
- **Sample:** thin, *nicely prepared (no amorphization), orientation (zone axis)*
- **objective lens:** low *aberrations, proper focus, high stability !*
- **projection** lens *(optical transfer to detectors and magnification)*
- **Detectors:** *good MTF*



2) HRTEM Image formation



Source
FEG

Illumination
coherente

specimen

exit wave

objective lens
Spherical aberration
Cs

image

- **Illumination:** *parallel beam*

$$\Psi(\vec{\mathbf{r}}) = \Psi_0 \exp^{2\pi i \vec{\mathbf{k}}\cdot\vec{\mathbf{r}}}$$

- **Sample:**

  weak phase object:
  weak phase object aproximation (WPOA)

- **Objective lens:**

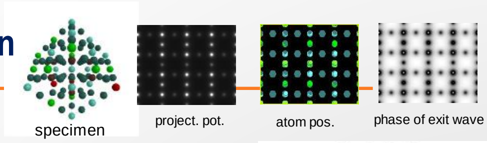
  Abbé's principle
  transfer function
  contrast transfer function (CTF)

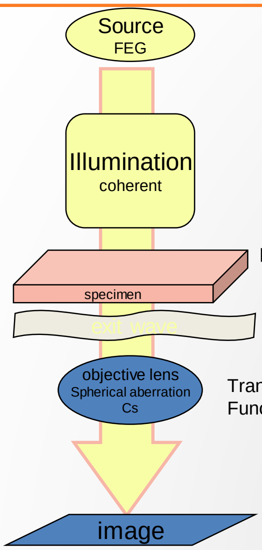
$$\Psi_i(\vec{\mathbf{x}}) = \Psi_o(\vec{\mathbf{x}}) \otimes T(\vec{\mathbf{x}})$$

- **Image contrast** (intensity)

$$\mathrm{I}_{\mathrm{i}}(\vec{\mathbf{x}}) = \Psi_i(\vec{\mathbf{x}})\Psi_i^*(\vec{\mathbf{x}})$$

2) HRTEM image formation

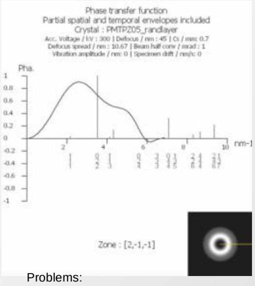


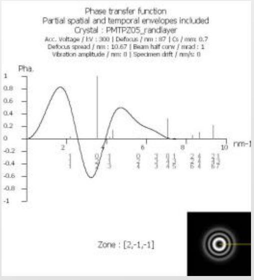


projected potential

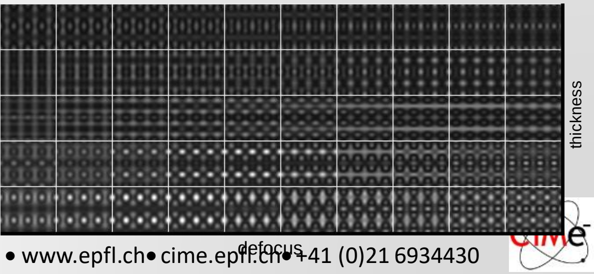
Transfer Function

image of “projected potential”

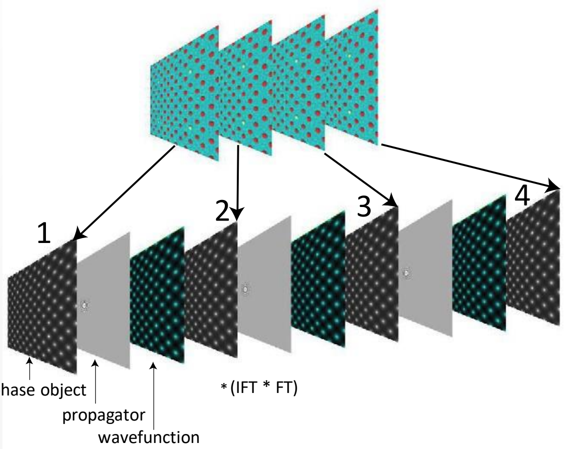




Problems:
defocusing for contrast: delocalization of information, information limit not used



2) Multi-slice calculation



The solid is sliced into thin sub-slices. The incident wave-function is transferred by the first slice (diffraction) and propagated to the next one. The propagation is done within the Fresnel approximation, the distance between the slices being 20 - 50 times the wavelength.

2) What do we need to know and characterize prior to performing simulations?

Prior to performing any calculation the following items (from the electron source to the detector) must characterized and modeled:

- The electron beam properties
- Convergence angle
- Source size
- Coherence (spatial and temporal)

Source and
illumination

- The specimen properties
- How is the incident electrons beam scattered by the specimen?

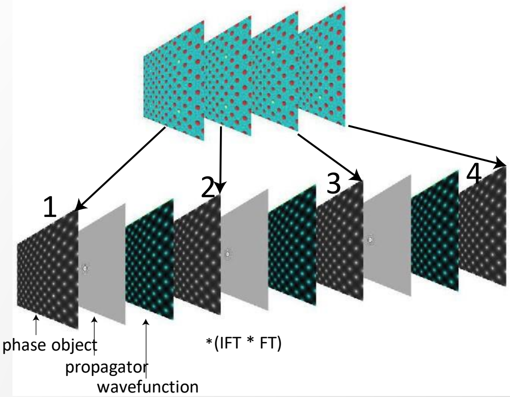
Sample Potential
Phase object
WPOA

- How does the microscope transfer the scattered electron beam?
- How do we measure the properties of the scattered electron beam (diffraction, image, hologram)?

Contrast Transfer
Function (CTF)

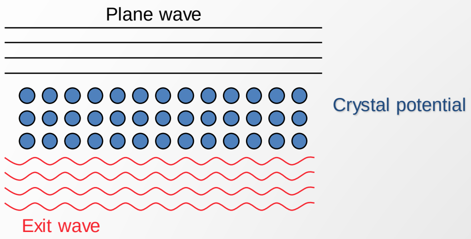
- What are the properties of the detection system? ← MTF of the Detector

2) Multi-slice calculation: FFT approach



- Develop a model of the perfect crystal (or supercell with defects or specific chemical ordering in the nanoparticles) ⇒ phase object
- Diffractor: transfer by a slice⇒multiplication by phase object function (POF(~ ρ))
- Propagator: propagation between slices⇒convolution by the Fresnel propagator (is nowadays performed by a FFT followed by a multiplication and an inverse FFT (FT−1, multiplication, FFT)).

2) Specimen shifts the phase and changes the amplitude of the electron wave



Wave vector in vacuum:

$$k = \sqrt{2me(E)/h^2}$$

Wave vector in a potential:

$$k = \sqrt{2me(E + V(\vec{r}))/h^2}$$

Phase shift $\Delta\alpha$ due to the crystal potential V_p:

$$\Delta\alpha = \frac{\sigma}{2\pi} V_p(\vec{x}, z)$$

$$\sigma = \pi/\lambda E$$

Exit wave function: $\Psi_o(\vec{\mathbf{x}}) = exp\lfloor -i\sigma V_p(\vec{\mathbf{x}}; z)\rfloor$

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2) Weak phase object approximation (WPOA)

Weak phase object approximation:

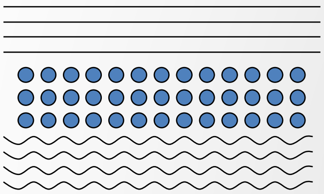
$$\Psi_o(\vec{\mathbf{x}}) = exp\lfloor -i\sigma V_p(\vec{\mathbf{x}}; z)\rfloor \cong 1 - i\sigma V_p(\vec{\mathbf{x}}; z)$$

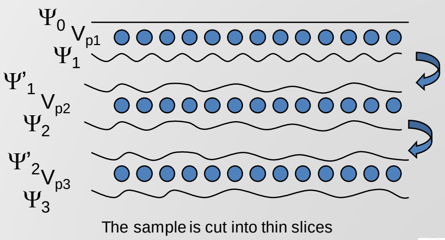
No absorbtion,effect of the object on the outgoing wave: only phase shift

The exit wave function contains the information about the structure of the sample

Multi-slice calculation:

Calculation of the exit wave function for complex structures:





The sample is cut into thin slices

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Weak Phase Approximation

In the Weak Phase Object Approximation under optimum transfer conditions the image intensity $I(\vec{x})$ is:

positive Cs (black atomic columns)

$$I(\vec{x}) \sim 1-2\sigma V_p(\vec{x})$$

negative Cs (white atomic columns)

$$I(\vec{x}) \sim \sigma V_p(\vec{x})$$

Where:

$V_p(\vec{x})$: projected potential

σ : electron matter interaction constant

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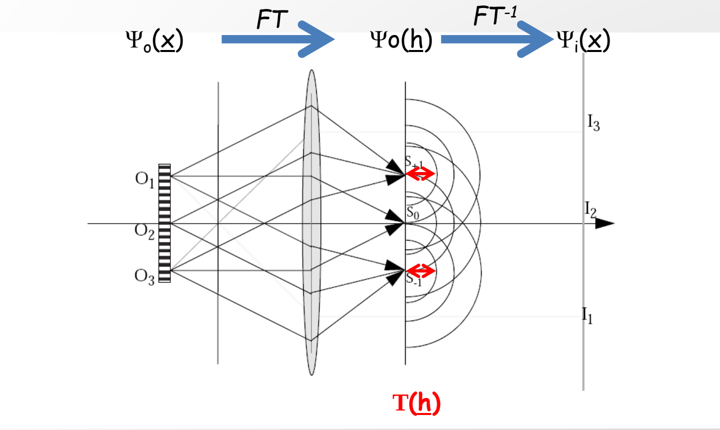
2) Abbé’s principle and Transfer Function



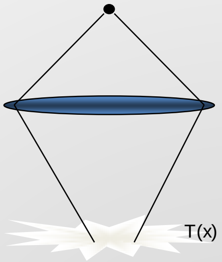
Objective lens is modelled as ***a thin lens*** that brings Fraunhofer diffraction pattern at finite distance (i.e. in its Back Focal Plane).

2) Transfer Function

- The optical system (lenses) can be described by a convolution with a transfer function T(x):

- Point spread function (PSF): describes how a point on the object side is transformed into the image.

$$\Psi_i(x) = \int_{-\infty}^{\infty} \Psi_o(\mathbf{u}) T(x\text{-}\mathbf{u}) d\mathbf{u} = \Psi_o(x) \otimes T(x)$$



- ***Transfer Function***:describes how an “object” wave-function is transformed into an “image”

$$\Psi_i(h) = \Psi_o(h) T(h)$$

- **The image INTENSITY** observed on a screen (or a camera)

$$I_i(x) = \Psi_i(x) \Psi_i^*(x)$$
$$I_i(x) = \Psi_i(h) \otimes \Psi_i^*(-h) = \int \Psi_i(h') \Psi_i^*(h - h') dh'$$
$$I_i(x) = |\Psi_o(h) T(h)| \otimes | \Psi_o^*(-h) T^*(-h)|$$

---

2) Transfer Function

$$T(\vec{\mathbf{h}}) = a(\vec{\mathbf{h}}) \exp\left[2\pi i \chi(\vec{\mathbf{h}})\right] E_s(\vec{\mathbf{h}}) E_t(\vec{\mathbf{h}})$$

- Phase factors:
  - Spherical Aberration
  - Defocus
- Amplitude factors:
  - (objective) apertures
  - spatial coherence envelope (non-parallel, convergent beam)
  - Temporal coherence envelope (non monochromatic beam, instabilities of the gun and lenses)

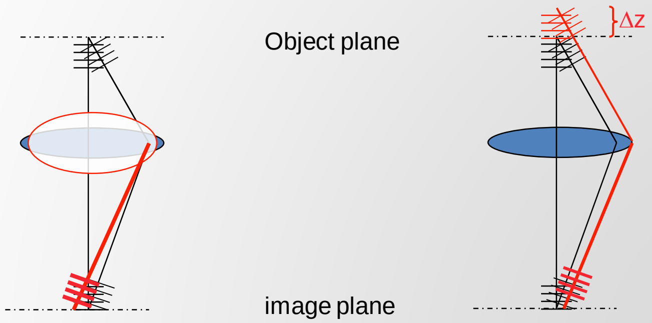
2) Transfer Function: phase factor

$$T(\vec{\mathbf{h}}) = \exp\left[2\pi i \chi(\vec{\mathbf{h}})\right]$$

$$\chi(\vec{\mathbf{h}}) = \underbrace{0.25 C_s \lambda^3 h^4}_{\text{Spherical aberration}} + \underbrace{0.5 \Delta z \lambda h^2}_{\text{defocus}}$$

Spherical aberration

defocus



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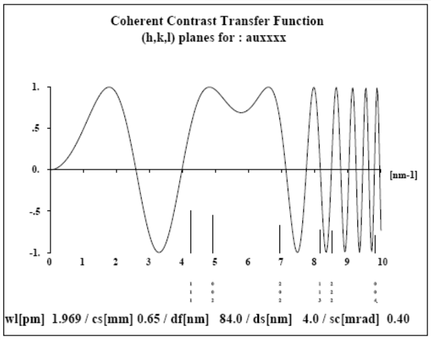
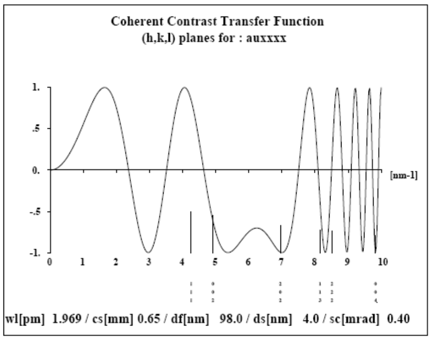
2) The contrast transfer function -CTF

CTF: contrast transfer function
(“useful part” = V_p)
Image contrast including
Defocus and Cs aberration

$$T(\vec{\mathbf{h}}) = \exp\left[2\pi i \chi(\vec{\mathbf{h}})\right]$$

$$\chi(\vec{\mathbf{h}}) = 0.25 C_s \lambda^3 h^4 + 0.5 \Delta z \lambda h^2$$



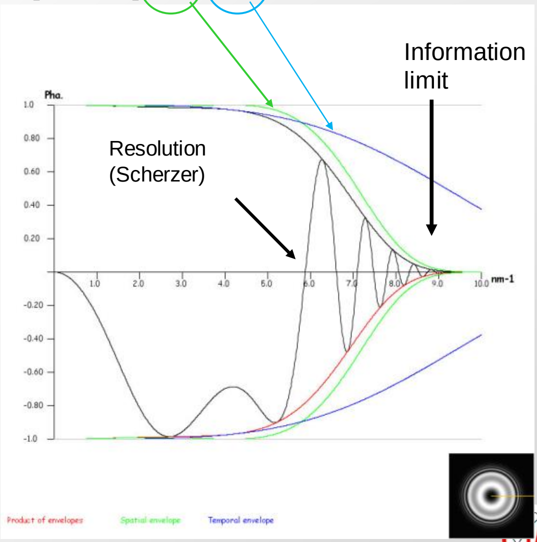


2) Spatial and temporal coherence

$$T(\vec{\mathbf{h}}) = a(\vec{\mathbf{h}})\exp\left[2\pi i\chi(\vec{\mathbf{h}})\right]\boxed{E_s(\vec{\mathbf{h}})}\boxed{E_t(\vec{\mathbf{h}})}$$

CM300UT FEG
Field emission
C_s: 0.7mm
Dz= 44nm

Resolution(point to point):1.7Å
Information limit : ~1.2Å



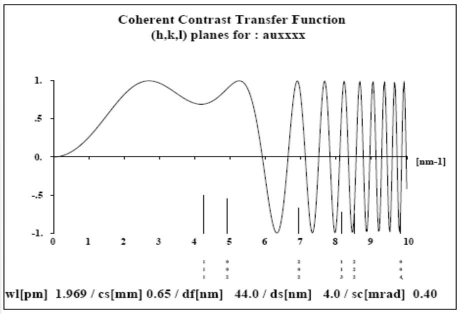
2) How is Scherzer Defocus defined?

With $Dz_{scherzer}$

$$\Delta z = -\sqrt{\tfrac{4}{3} C_s \lambda}$$

The CTF has a wide pass band

$$D_{scherzer} = 0.66\lambda^{3/4} C_s^{1/4}$$



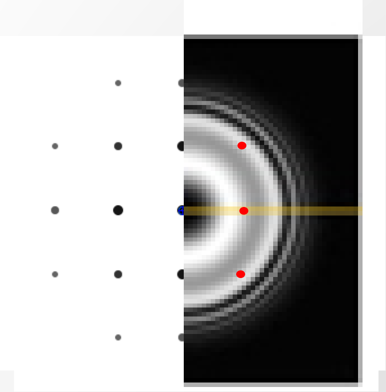
The first zero crossing of the CTF defines the point-to-point resolution of an electron microscope

The atom columns appear as dark areas on a bright background

$$I(x) = (1 - \sigma V_p(x))(1 + \sigma V_p(x)) = 1\text{-}2\sigma V_p(x) + (\sigma^2 V_p^2(x))$$

Dark atoms ***bright background***

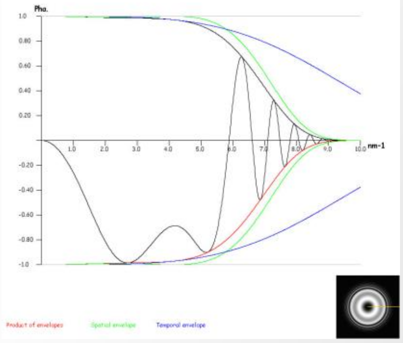
2) Example: Au

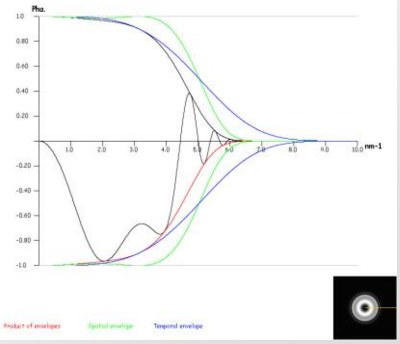


2) Some CTFs of good (old) microscopes

CM300UT FEG
Field emission
C_s: 0.7mm
Δz= 44nm

Resolution (point to point): 1.7Å
Information limit: ~1.2Å

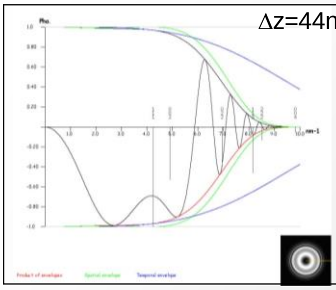




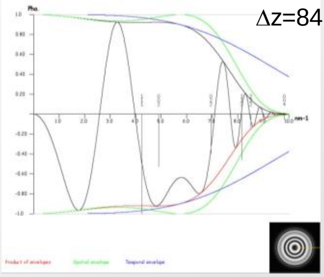
CM30ST LaB6
Thermal emitter
C_s: 2mm
Δz= 76nm

Resolution (point to point): 2.1Å
information limit: ~1.9Å

2) Pass bands as a function of defocus

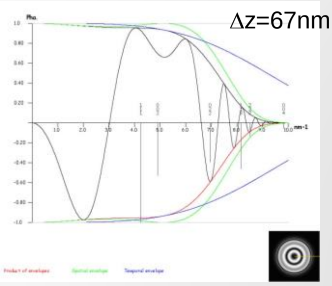


Δz=44nm

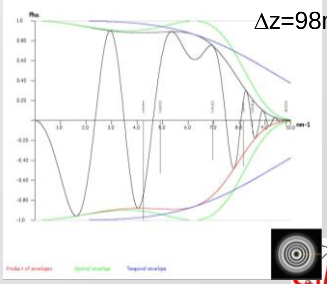


Δz=84nm

direct



Δz=67nm



Δz=98nm

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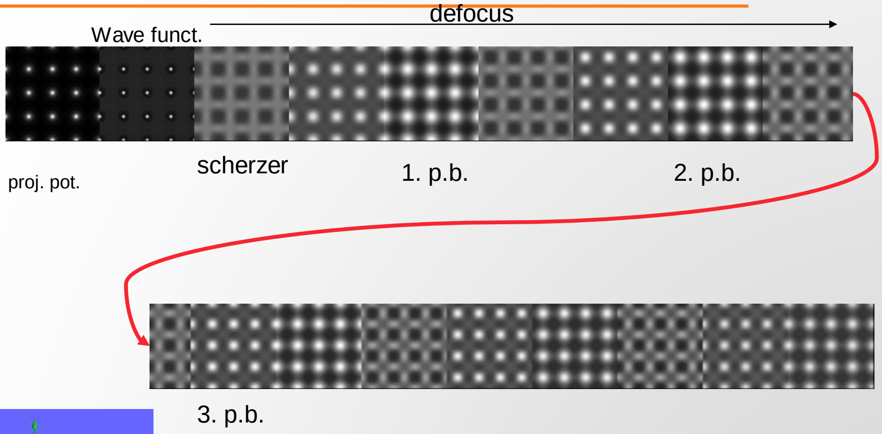
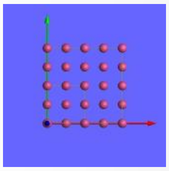
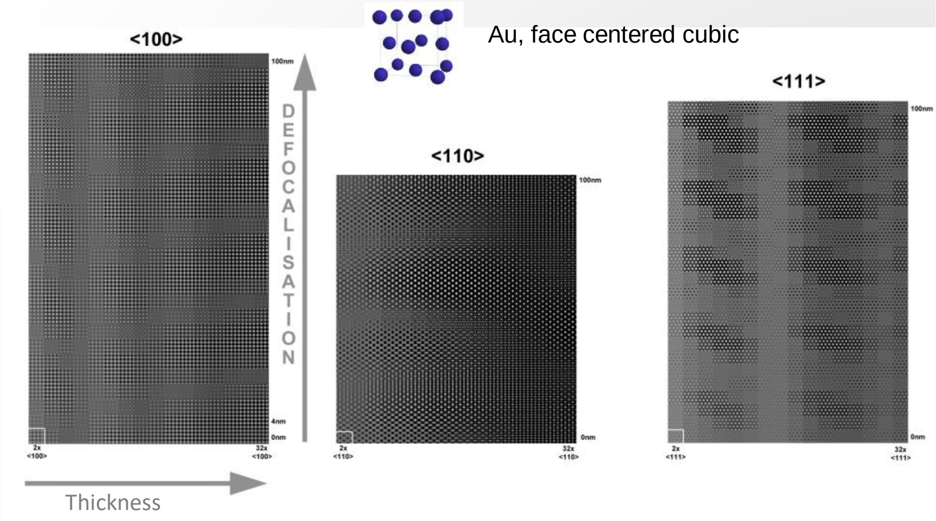
2) Variation in HRTEM Image Constrast with Defocus





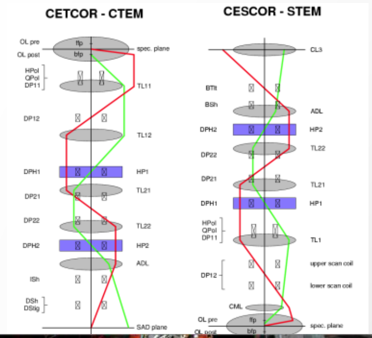
Au [100], thickness 20nm

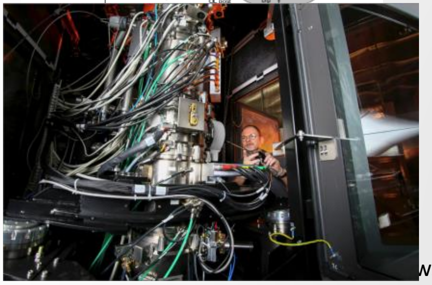
2) Variation of the contrast with: thickness, defocus, orientation

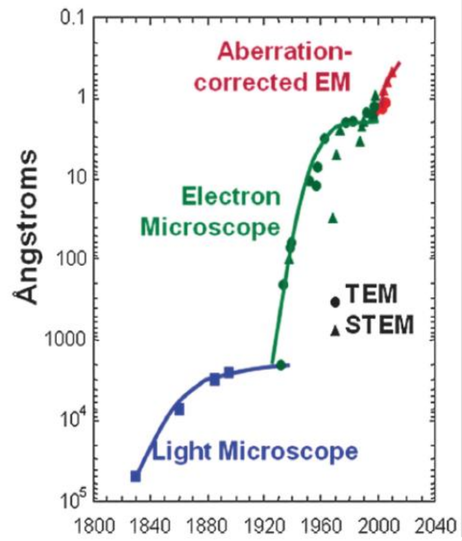


Au, face centered cubic

3) Aberration corrected (AC) HRTEM: Cs correctors

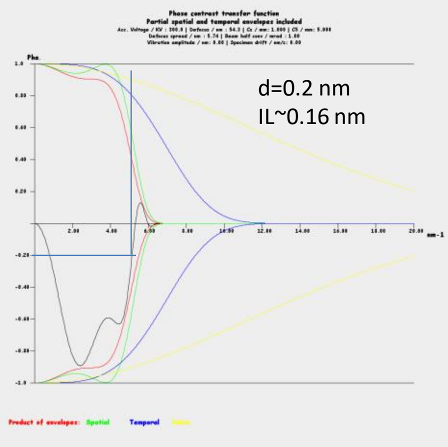






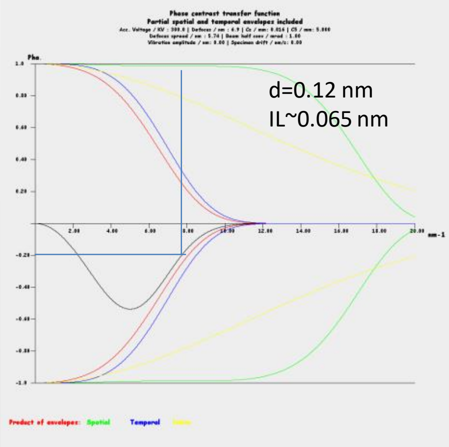
3) Cs correction greatly improves interpretable limit and information limit

Uncorrected with Cs=1mm



d=0.2 nm
IL~0.16 nm

Corrected with Cs=0.01mm



d=0.12 nm
IL~0.065 nm

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3) What are the advantages of Cs correction?

- Improved resolution
  - At lower voltage! (beam sensitive materials)
  - For large pole piece gap (in-situ experiments possible)
- Higher precision (better contrast)

  Lower dose possible

  Higher frame rate (video)
- Less delocalization

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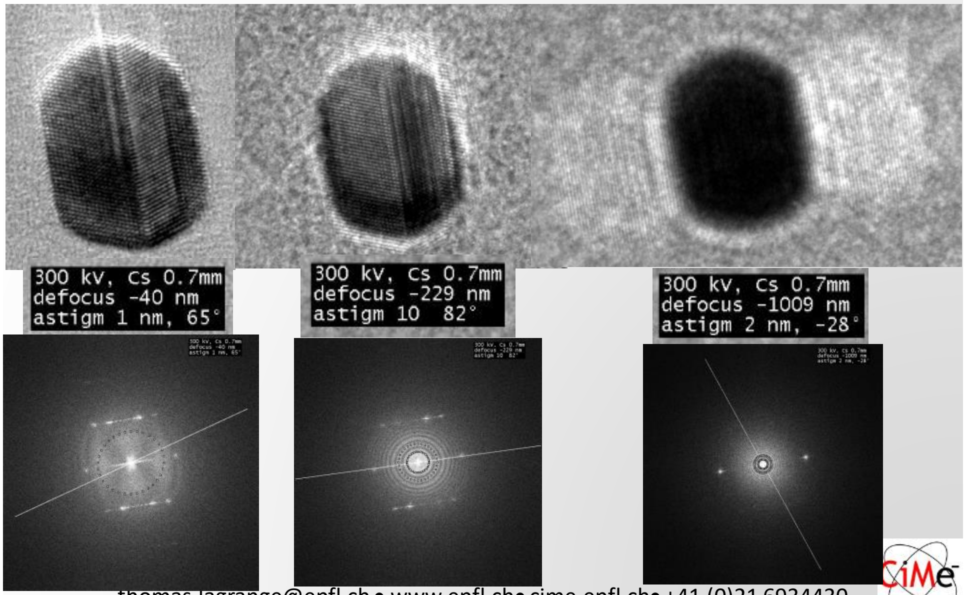
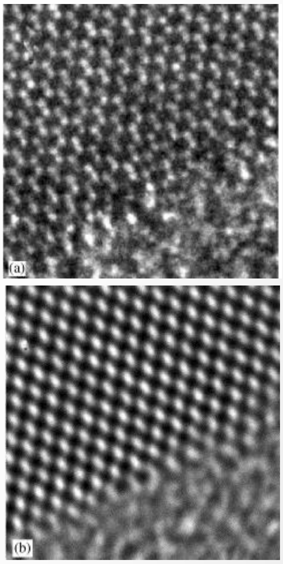
3) Delocatization occurs due to defocus of imaging which is necessary in non-corrected TEM to obtain highest interpretable spatial resolution

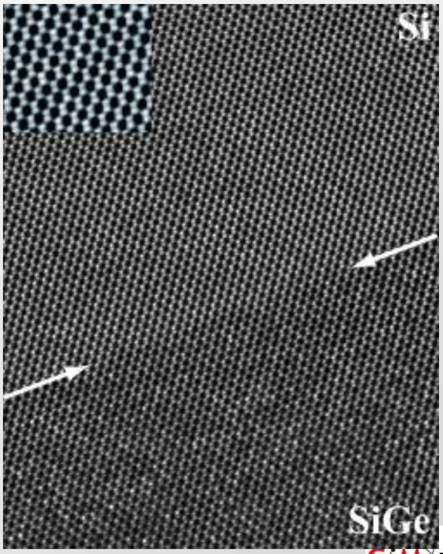


3) AC-HRTEM reduces delocalization at interfaces, i.e., provide clear images of sharp interfaces

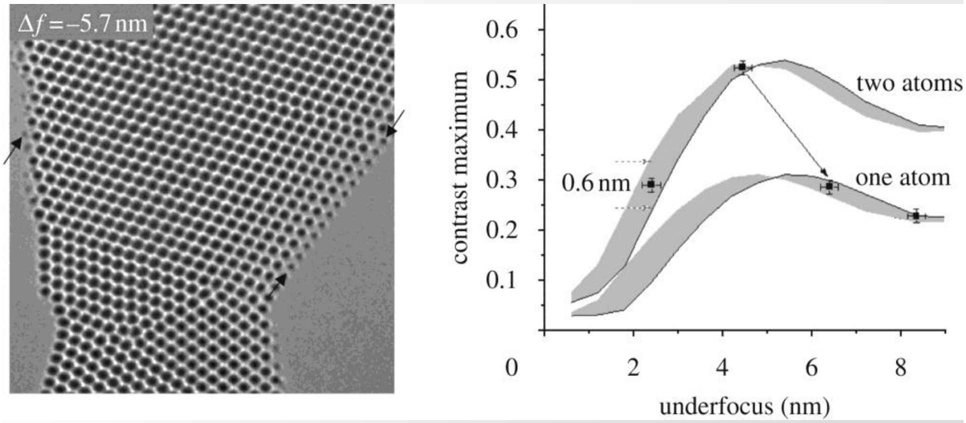


Cs corrector off

Cs corrector on

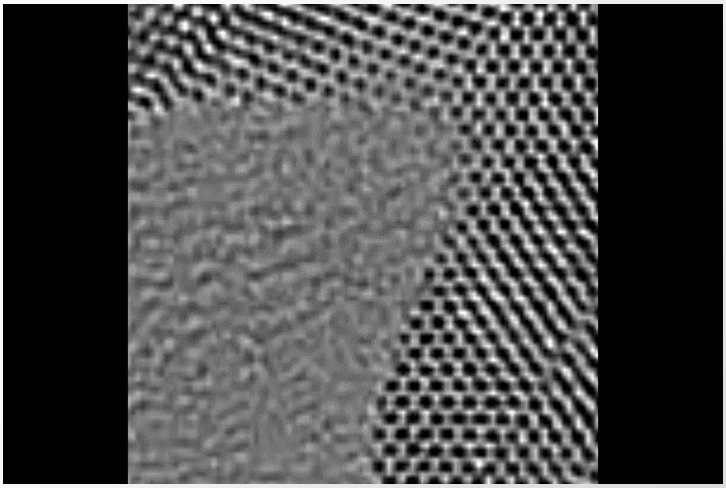


3) AC-HRTEM provides much higher contrast and can allow quantitative imaging



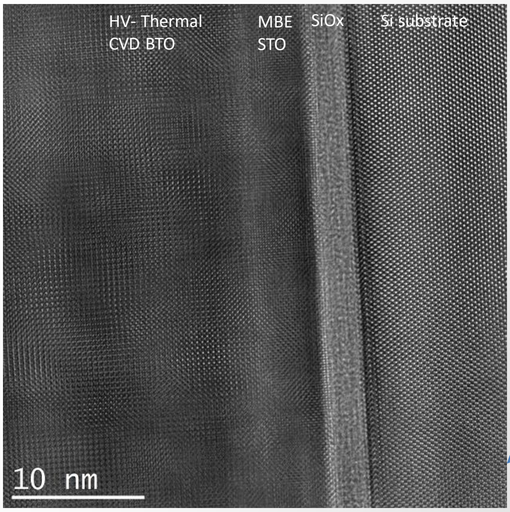
U. Dahmen et. al, “Background, status and future of the Transmission Electron Aberration-corrected Microscope project”, Phil. Trans of the Royal Society A, vol. 367 (2009)

3) Cs correction provides High current/signals allowing for high speed imaging:



We can observe in real-time atoms hopping around on the surface

3) Cs corrected HRTEM image for Titan Themis



Si dumbells from crossing of the {113} lattice fringes

Sub-angstrom spatial resolution

Films were prepared M. Reinke (EPFL-LPMAT, Prof. P. Hoffman)
TEM samples prepared by D. Laub
Images taken by T. LaGrange

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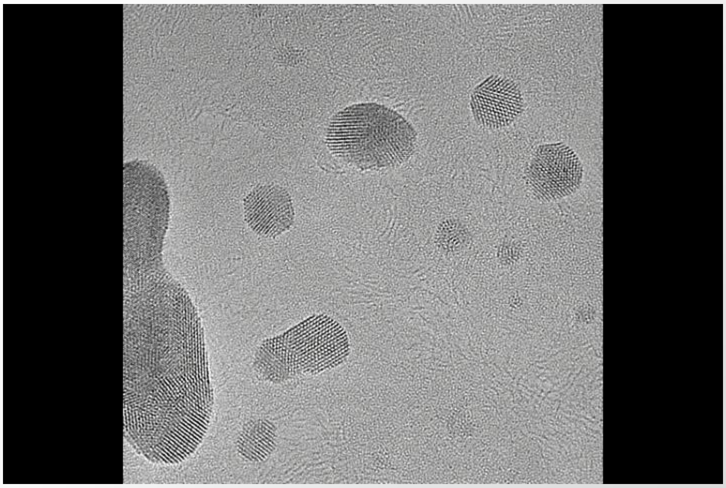
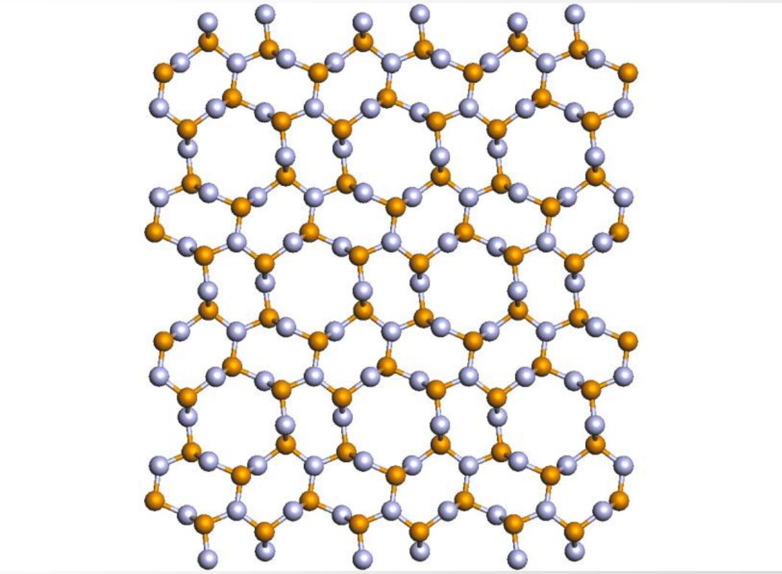
3) The Dance of the Nanoparticles particles



Beam induced Dynamics!

3) You still need simulations to interpret AC-HRTEM images Example: Crystalline Silicon Nitride on [001] Z.A.



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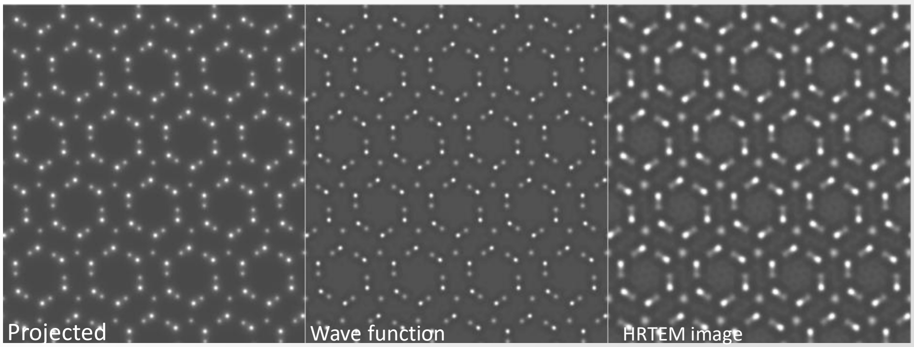
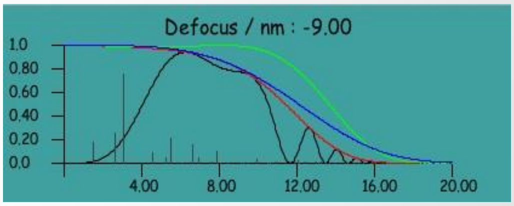
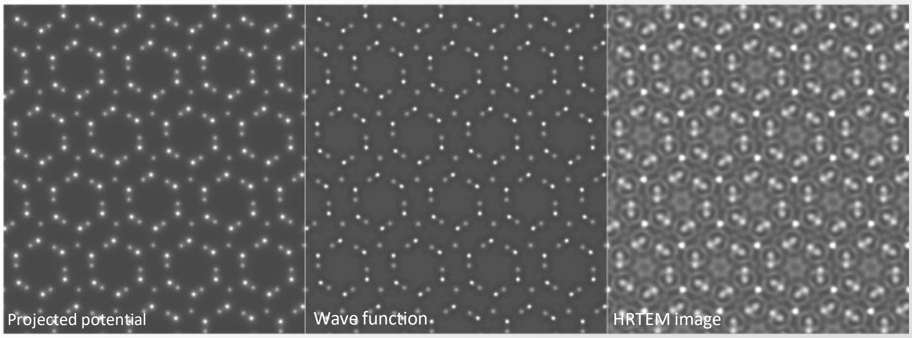
3) Silicon Nitride on [001] Z.A., 10nm thick, -9 nm defocus

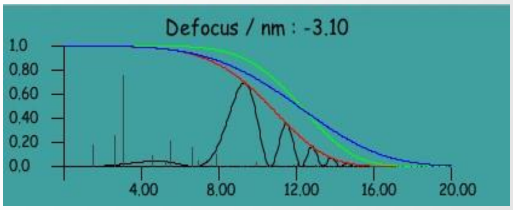




CTF for Titan 60-300 TEM with a image (Cs aberration) corrector operating at 300 KV and with Cs= -0.033 mm

3) Silicon Nitride on [001] Z.A., 10nm thick, -3 nm defocus





CTF for a Titan 60-300 TEM with a image (Cs aberration) corrector operating at 300 KV and with Cs= -0.033 mm

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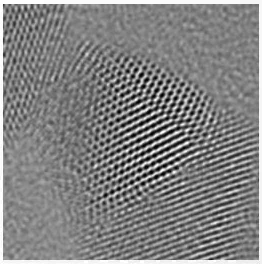
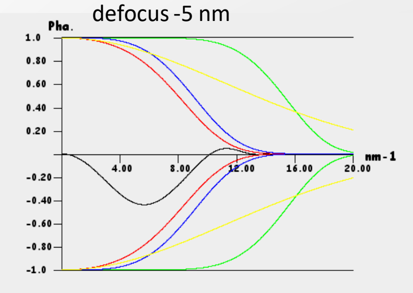
3) Simulations are still crucial for understanding aberration corrected HR-TEM images

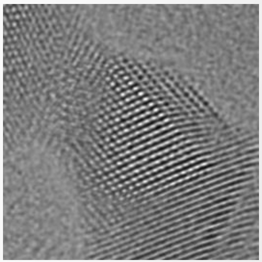
Black Atom Contrast



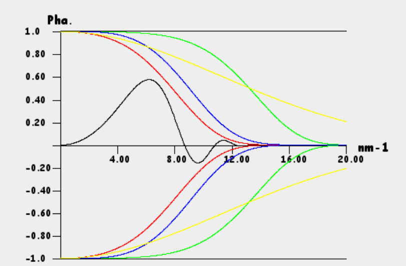
defocus -5 nm



White Atom Contrast



defocus +5 nm



Summary

1. HRTEM Images
   - ➢HRTEM can tell us about the atomic structure of our sample but remember the HRTEM image is an INTERFERNCE PATTERN and NOT A DIRECT IMAGE OF ATOMS
2. HRTEM Simulations
   - ➢HRTEM images are sensitive to many factors, e.g., crystal orientation, defocus, sample thickness and lens aberrations, and in order to properly interpret the images, we must do simulations
   - ➢Multi-slice technique is a common approach for simulating HRTEM images
   - ➢Simulations of the contrast transfer function can tell us which lattice spacings (spatial frequencies) we can properly interpret in our experiments at optimum focus (Scherzer Defocus)
   - ➢Making montages of how HRTEM image contrast changes with objective defocus and sample thickness is useful for interpreting HRTEM data
3. AC-HRTEM
   - ➢Gives improved spatial resolution, higher precision (better contrast) and less delocalization (better visualization of interfaces)
   - ➢Simulations are still needed for proper interpretation of AC-HRTEM data

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QUESTIONS?

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