Shaping the Future of Electron Microscopy: The TEAM Microscope

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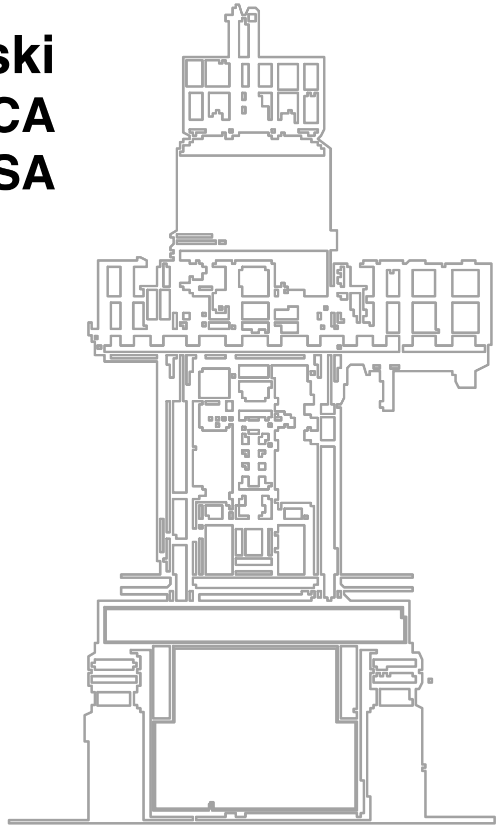


TEAM is a collaborative Project

Transmission Electron Aberration-corrected Microscope

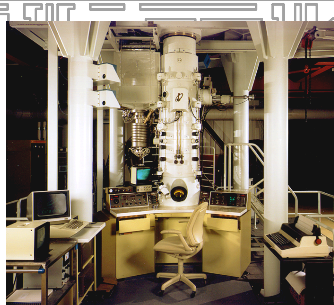
Lawrence Berkeley National Laboratory
Argonne National Laboratory
Brookhaven National Laboratory
Frederick-Seitz Materials Research Lab, UIUC
Oak Ridge National Laboratory

Industry: FEI Company, CEOS
NCEM: A.Schmid, A. Minor,
U. Dahmen (Head), P. Denes (Manager)

Supported by the US Department of Energy

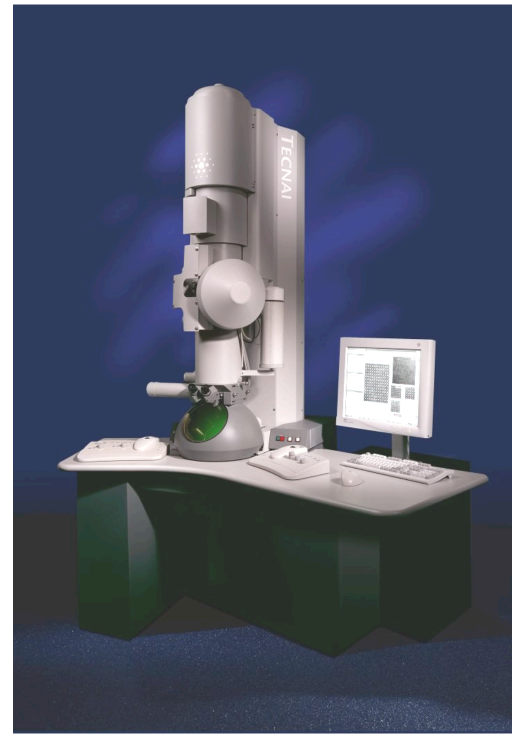
Selected Microscopes @ NCEM

Atomic Resolution Microscope (ARM)
Accessible to users since 1984
Point resolution: 0.16 nm, 400-800 kV



One Ångstrom Microscope (OAM)
Accessible to users since 1999
Spatial resolution: 0.08 nm, 300 kV

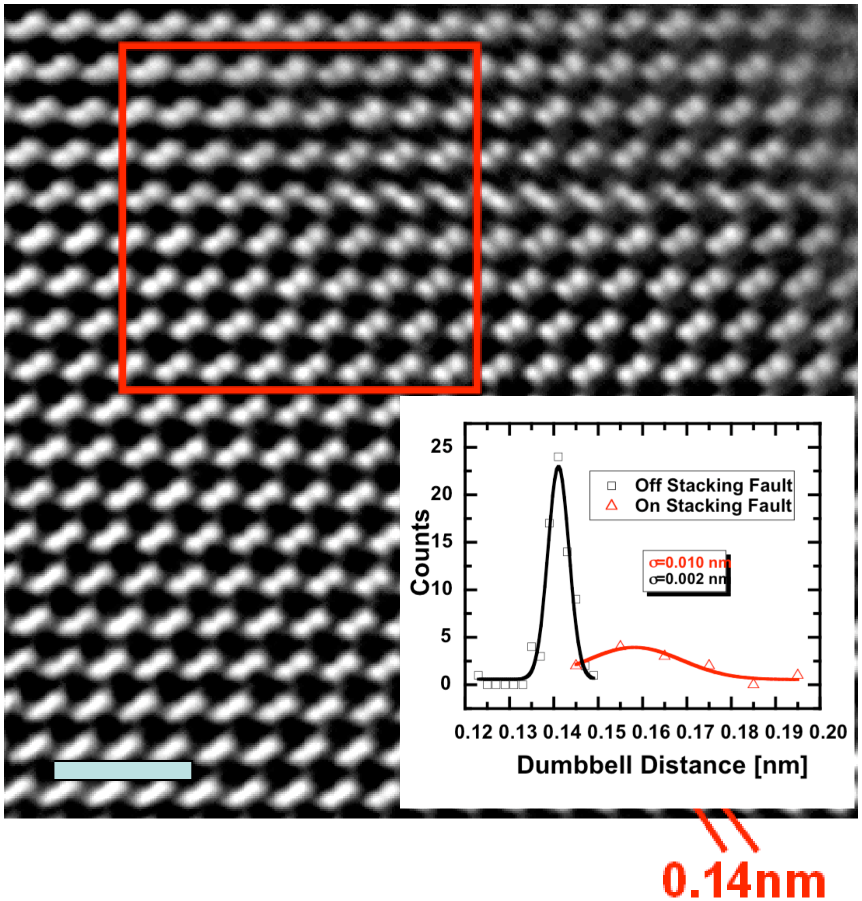
Highest TEM resolution today



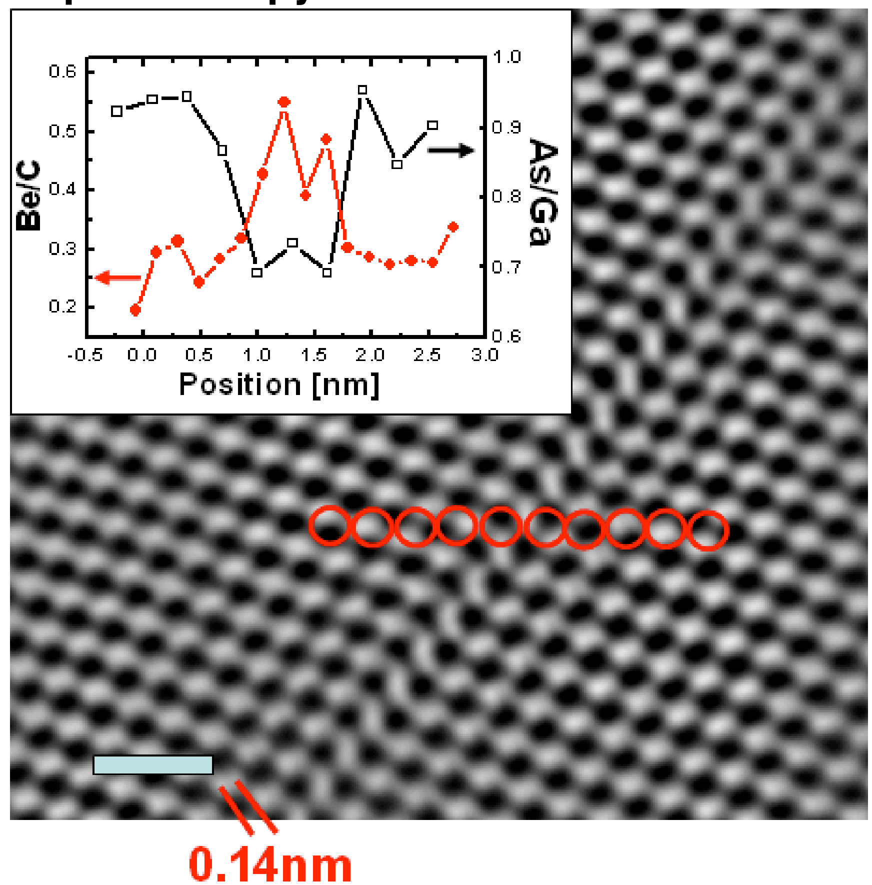
STEM / HRTEM : Tecnai G²
Accessible since 2003, 200 kV
Spatial resolution: 1.2 Ångstrom
Energy resolution: 150 meV

A STEM / TEM Study:
First full analysis of a single dislocation, GaAs:Be

Electron Exit Wave Reconstruction
Position determination



STEM (SuperSTEM, UK)
Spectroscopy

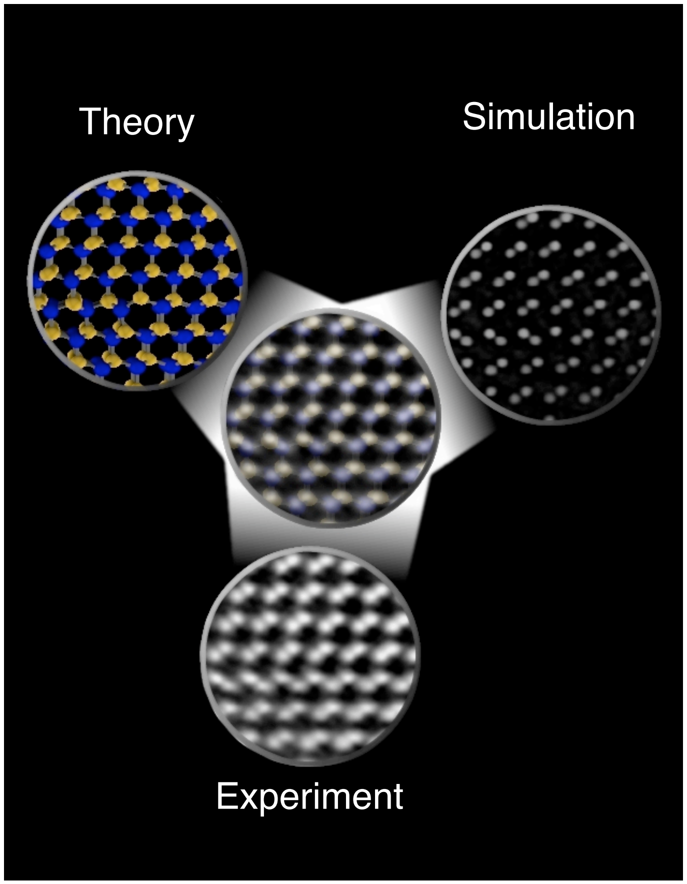


- Column positions can be determined to pm precision
- Compositional changes can be revealed on single atom columns

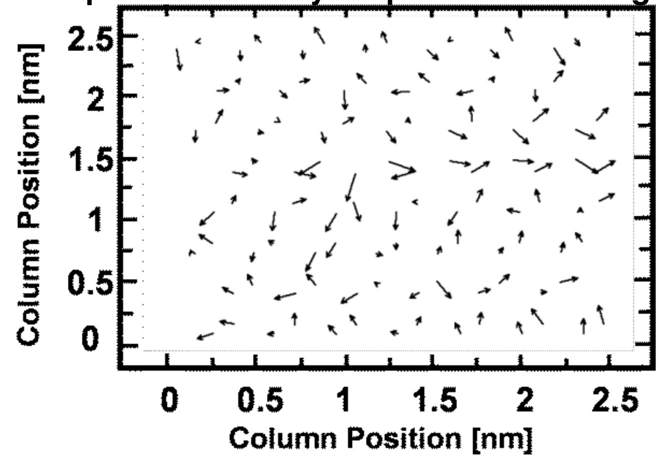
Combining Experiments & Simulations & Theory

Ga terminated 30^0 partial dislocation in GaAs: Be

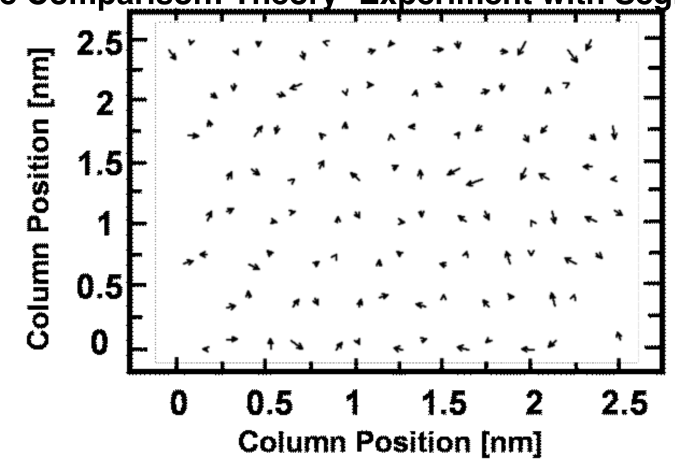
NCEM



Quantitative Comparison: Theory -Experiment w/o Segregation



Quantitative Comparison: Theory -Experiment with Segregation

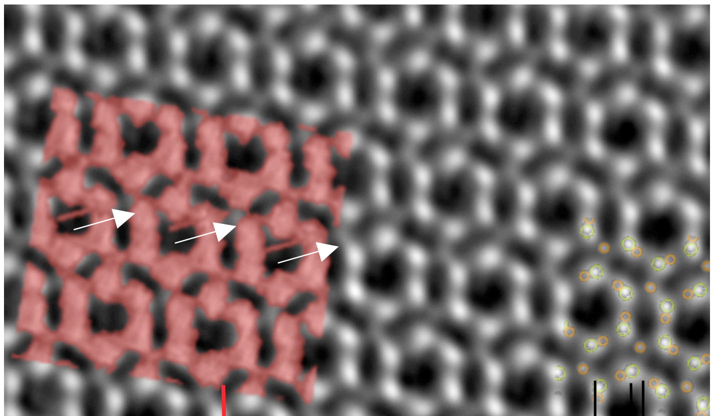


- **Theory and experiment can be merged on the same scale (~ 4000 atoms)**
- **Microscopy can guide theory**

X.Xu, S.P. Beckmann, P. Specht, E.R. Weber, D.C. Chrzan, R.P. Erni, I. Aslan, N. Brwoning, A. Bleloch, C. Kisielowski, **PRL 95, 2005, 145501**

Why Engage in New Developments

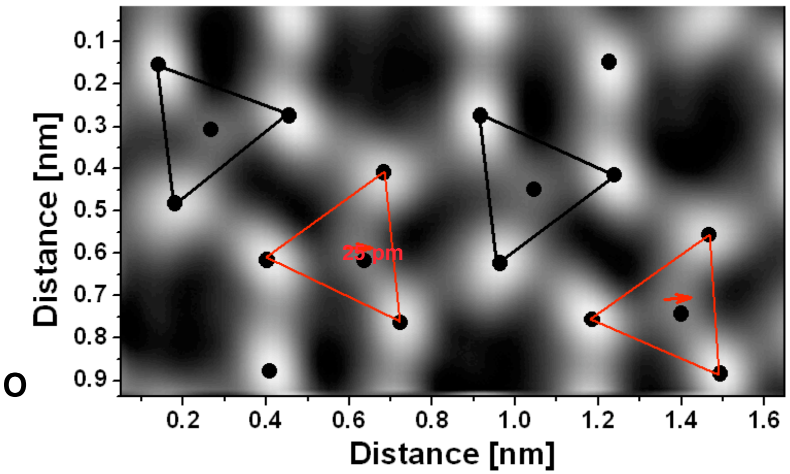
OAM reconstructed phase image (2005):



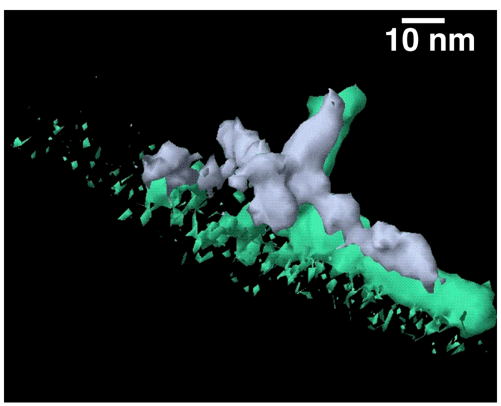
N$_2$ N$_1$ Si

ARM lattice image (1988):

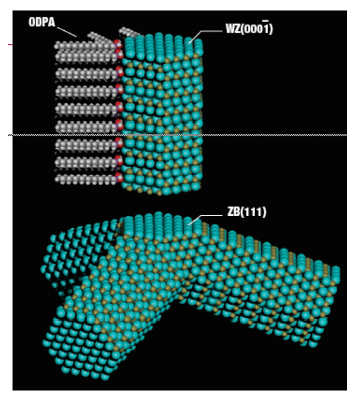
Si$_3$N$_4$: Al, O



- Projection of 3D object in 2D image
- Intensities are not interpreted

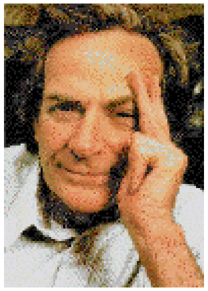


- 3D with atomic resolution



The Feynman Challenge

"It would be very easy to make an analysis of any complicated chemical substance; all one would have to do would be to look at it and see where the atoms are. The only trouble is that the electron microscope is one hundred times too poor … I put this out as a challenge: Is there no way to make the electron microscope more powerful?"

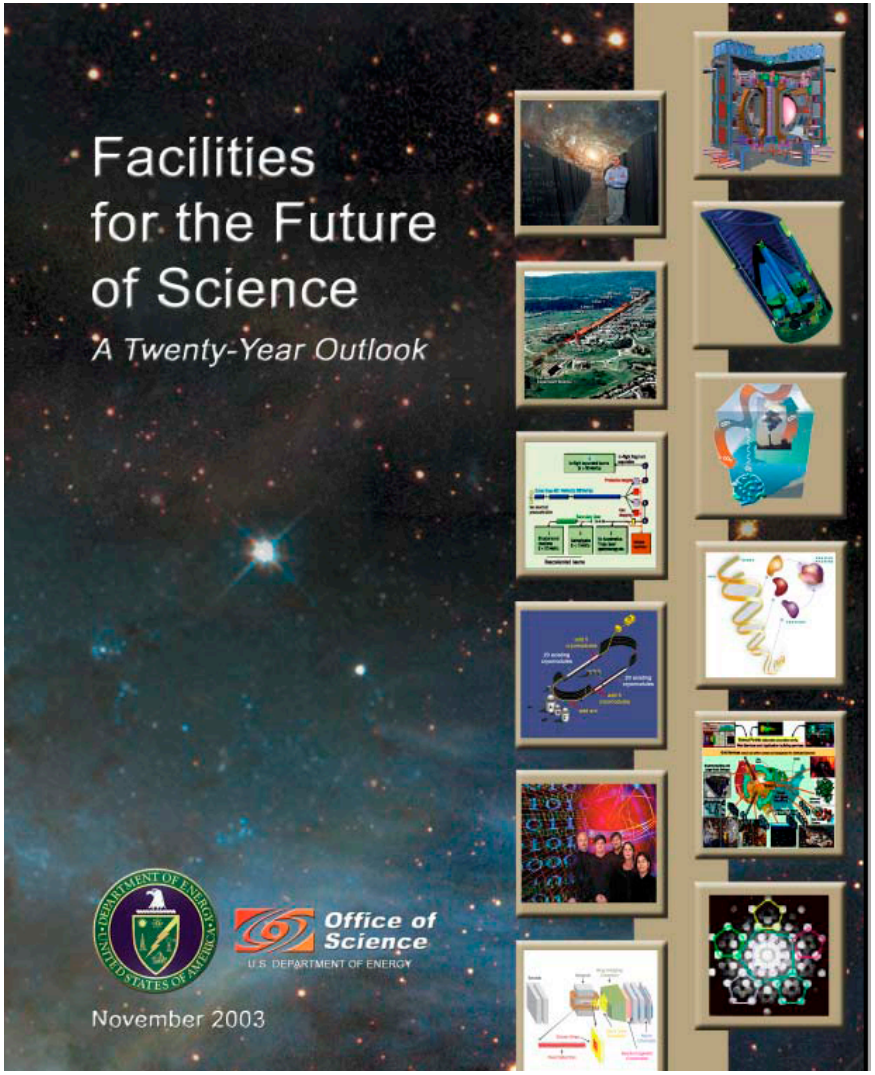


– Richard P. Feynman, 1959,
"There's Plenty of Room at the Bottom"

TEAM aims at meeting the Feynman challenge

There are numerous other advantages that are not mentioned

The DoE is Committed to Support TEAM



TEAM is a DoE Project

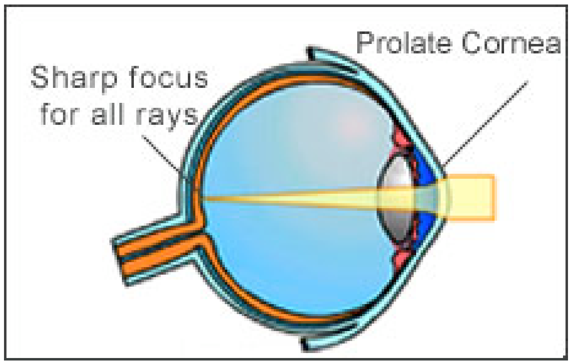
High visibility, near term goal

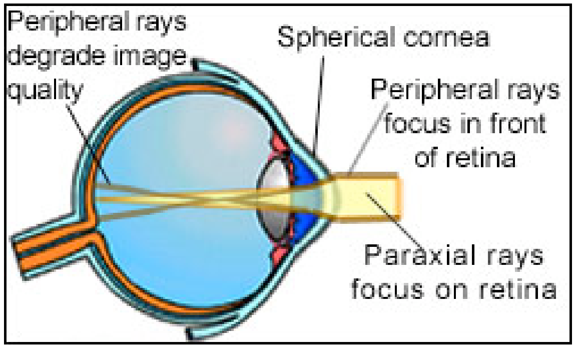
Platform: TITAN
Access: 2007 - 2009
Spatial resolution:
0.5 Ångstrom STEM/TEM
Energy resolution <100 meV
80 - 300 kV

Aberration corrected

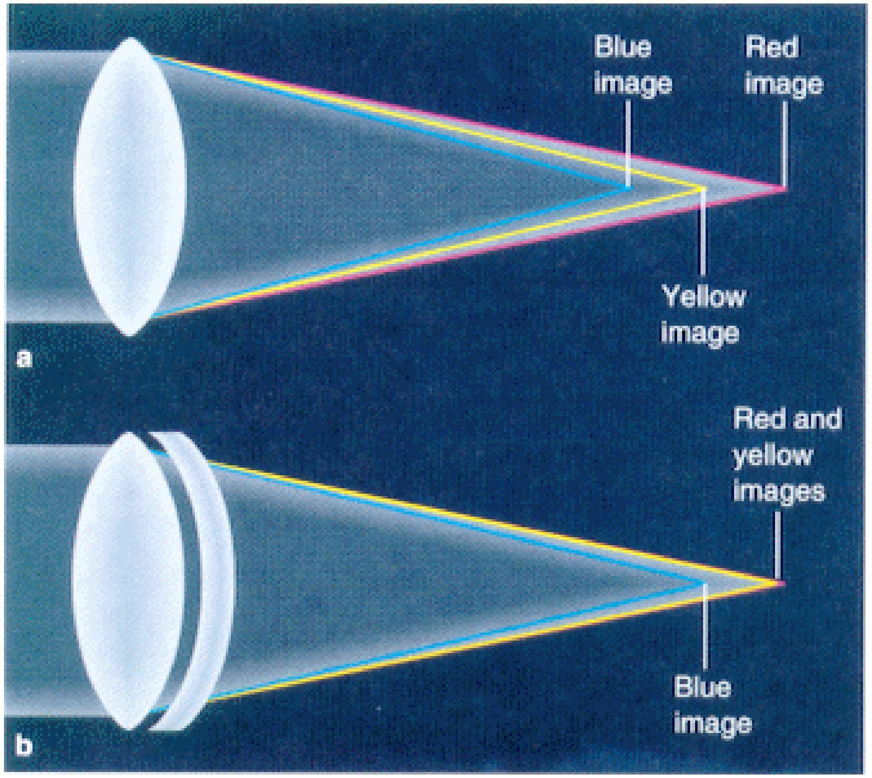
Well Known Aberrations

Spherical aberration



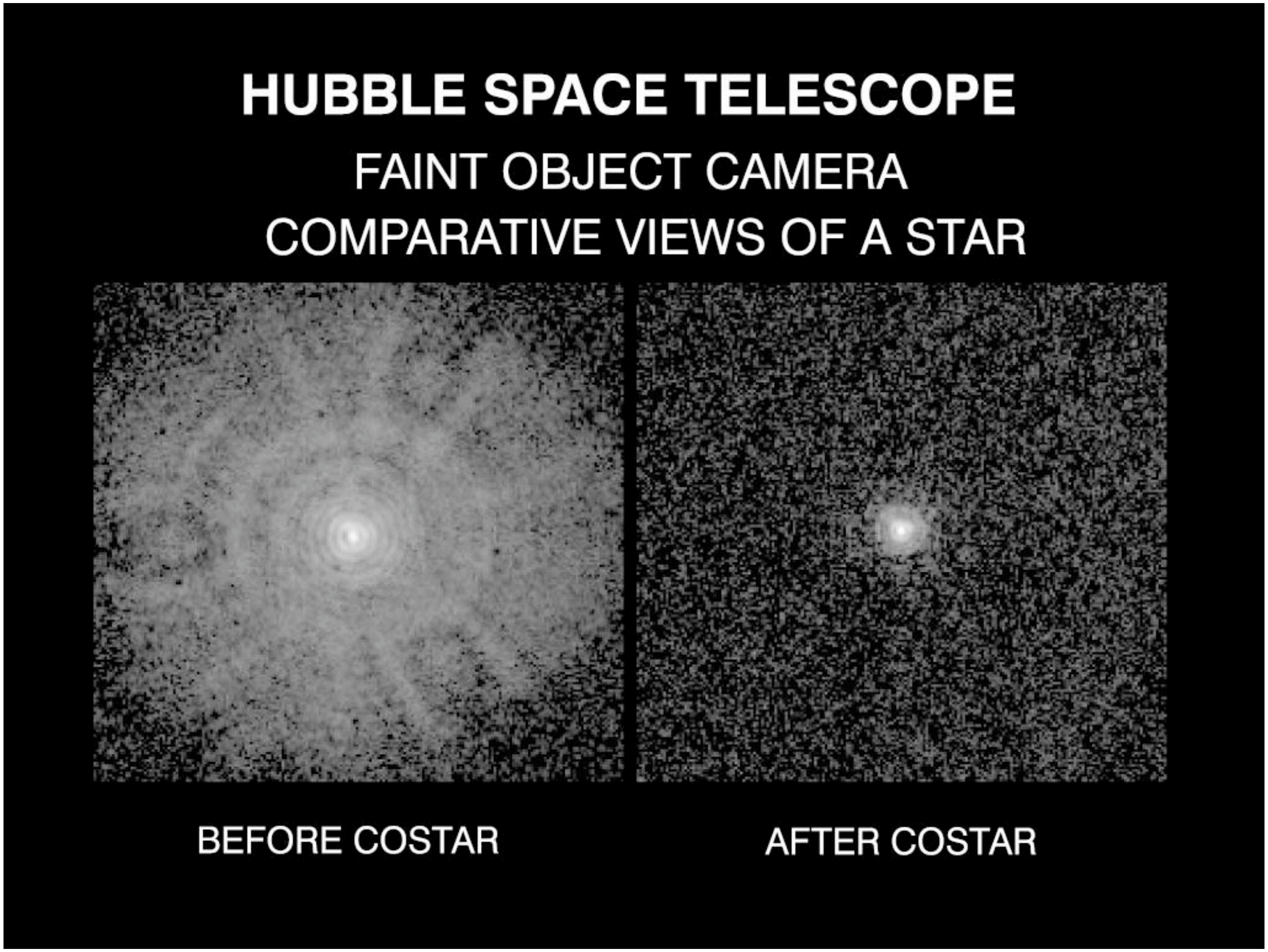


Chromatic aberration

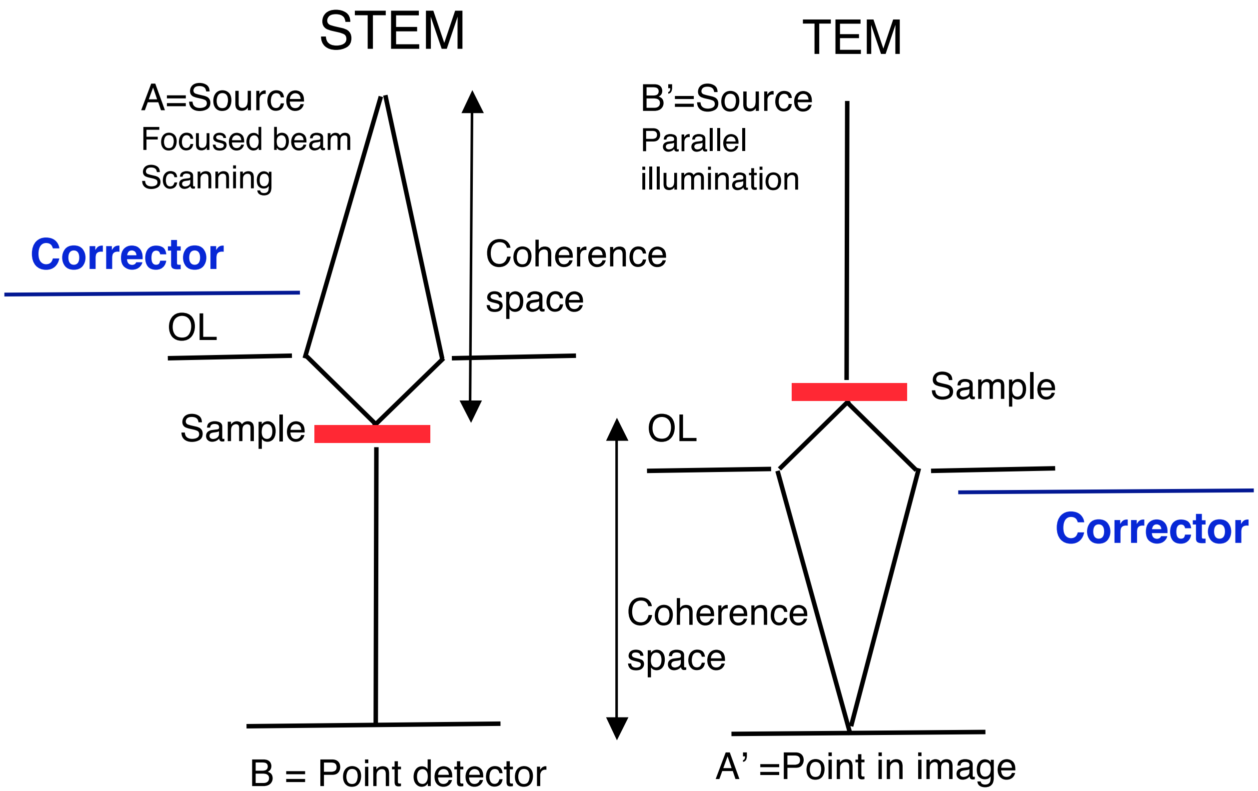


…and how they are corrected

A Famous Example



STEM & TEM



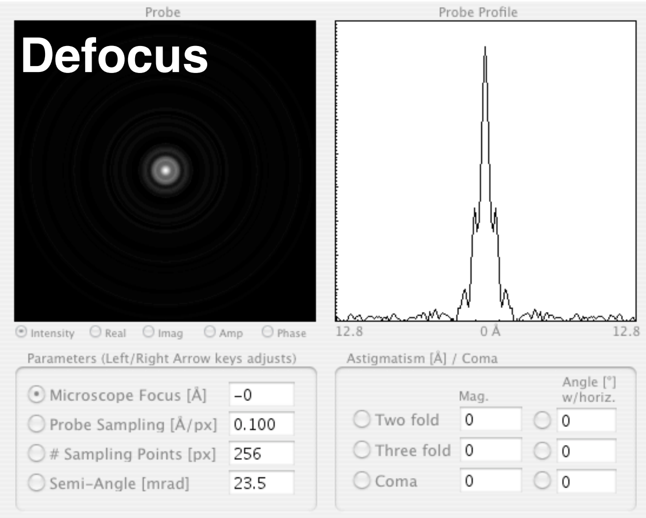
- **Principle of reciprocity:**

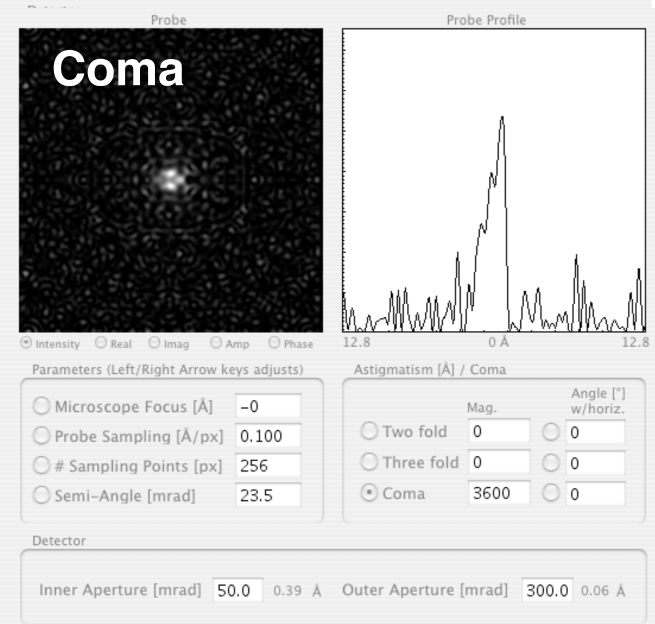
  Electron trajectories and elastic scattering processes have time reversal symmetry. J.M Cowley (APL 15, 1969,58)
- **TEM and STEM can perform identical tasks**

  Differences relate to S/N ratios & experimental set up
  Differences MATTER!

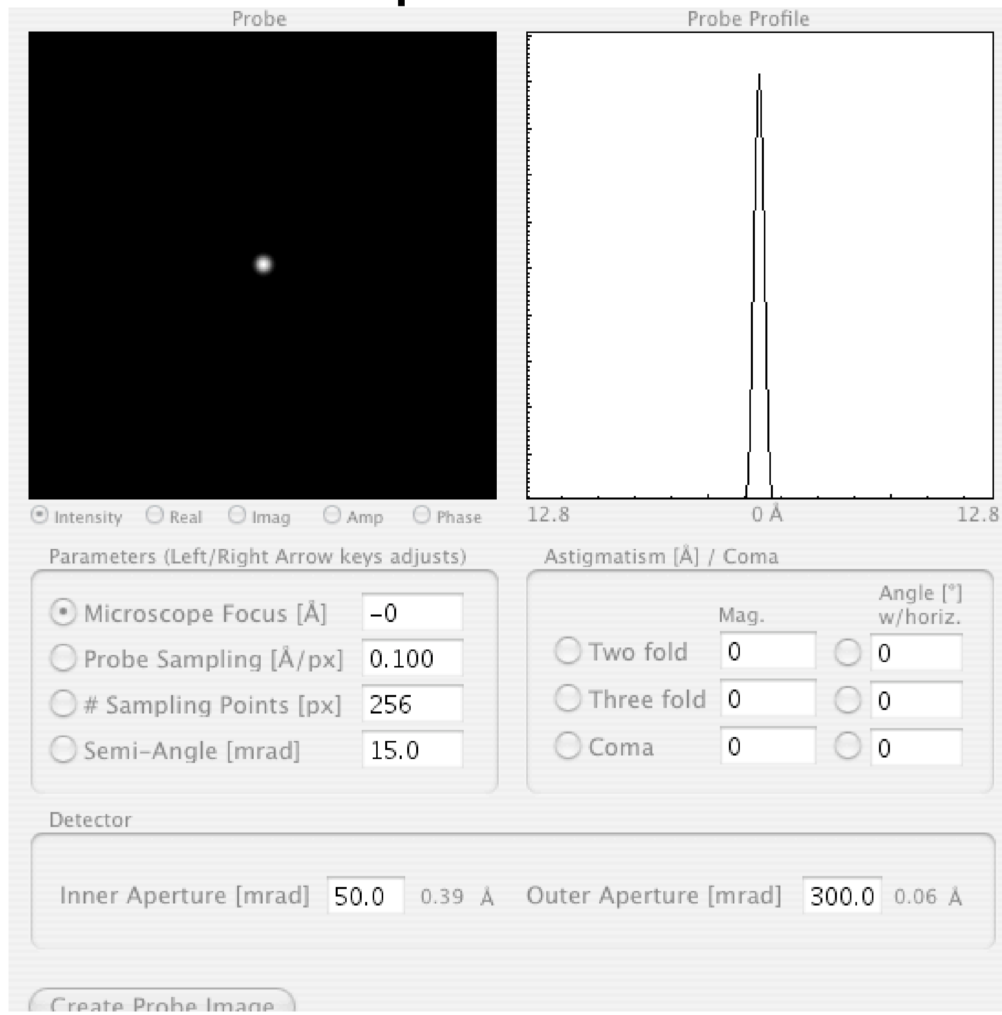
Lens Aberrations

STEM: Probe



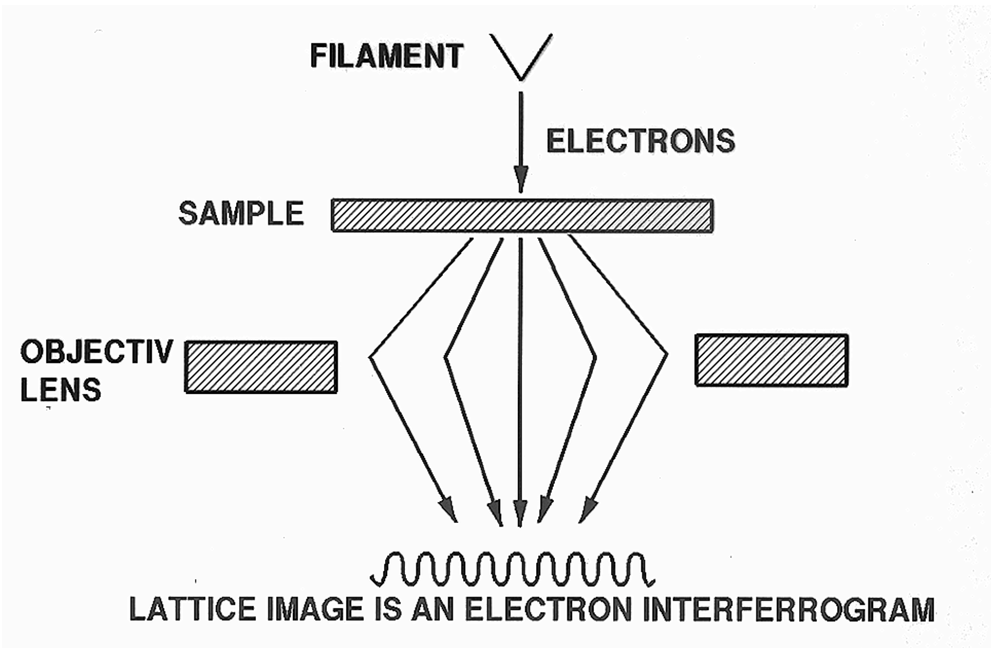


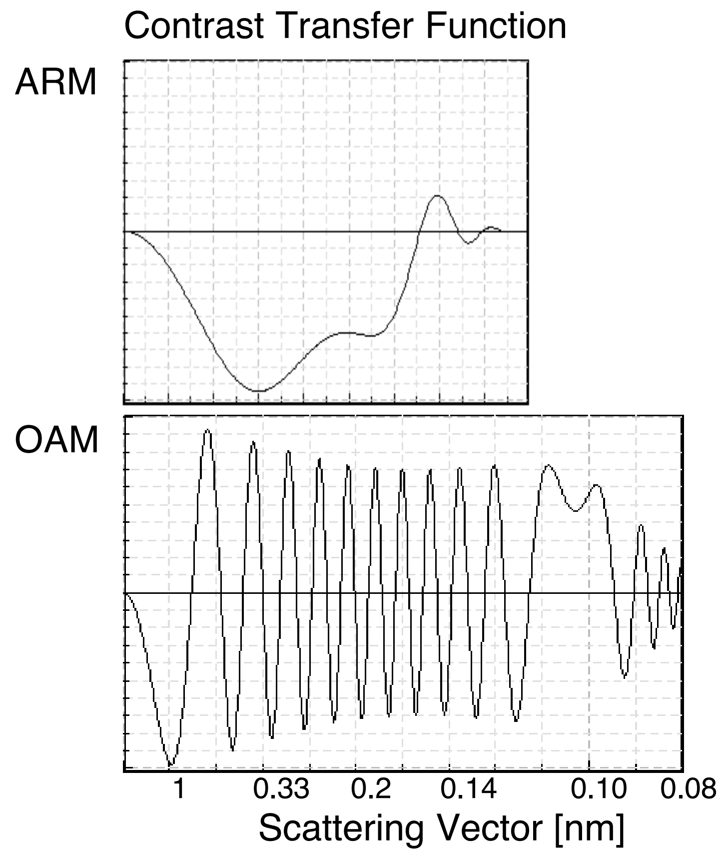
Cs corrected probe



In STEM aberration can only be corrected by hardware

Principle of HRTEM
Image formation

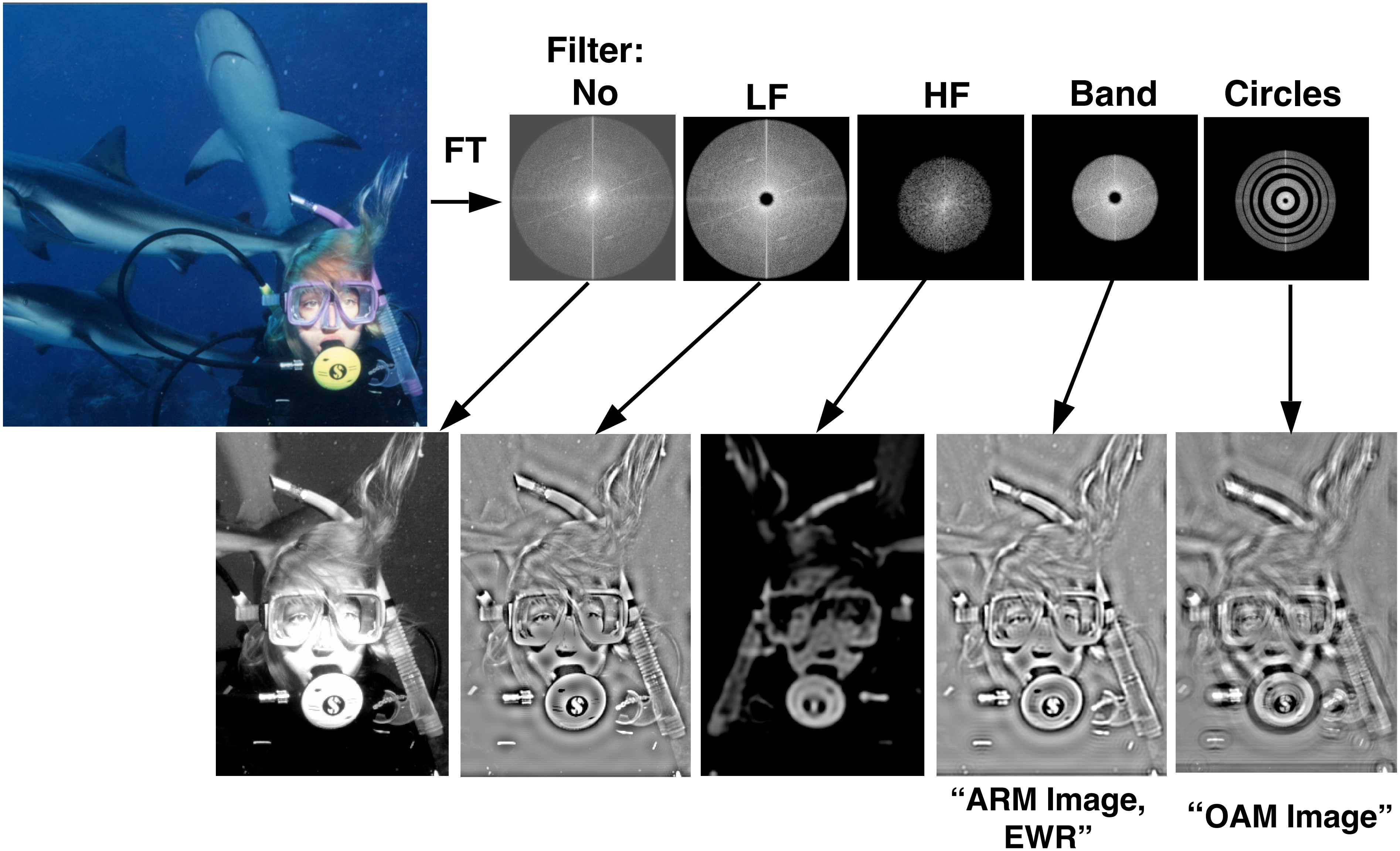




- We record Intensities: I ~ $\Psi\Psi^*$
  - Loss of phase information
  - CTF oscillations mix amplitudes and phases
  - CTF oscillations increase image delocalization
- We gain resolution - at a huge price….

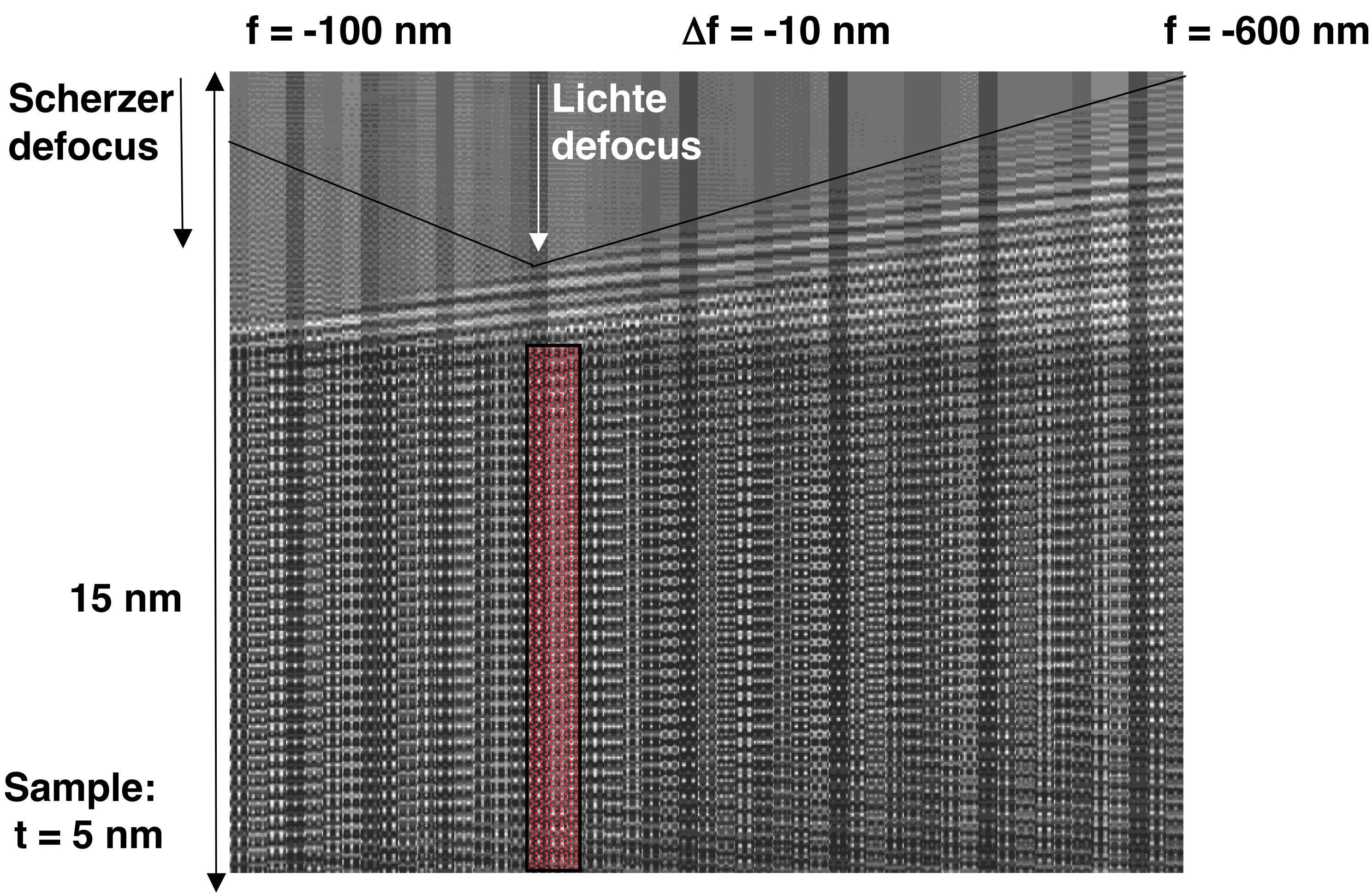
Principle of HRTEM

Image formation: delocalization



Principle of HRTEM

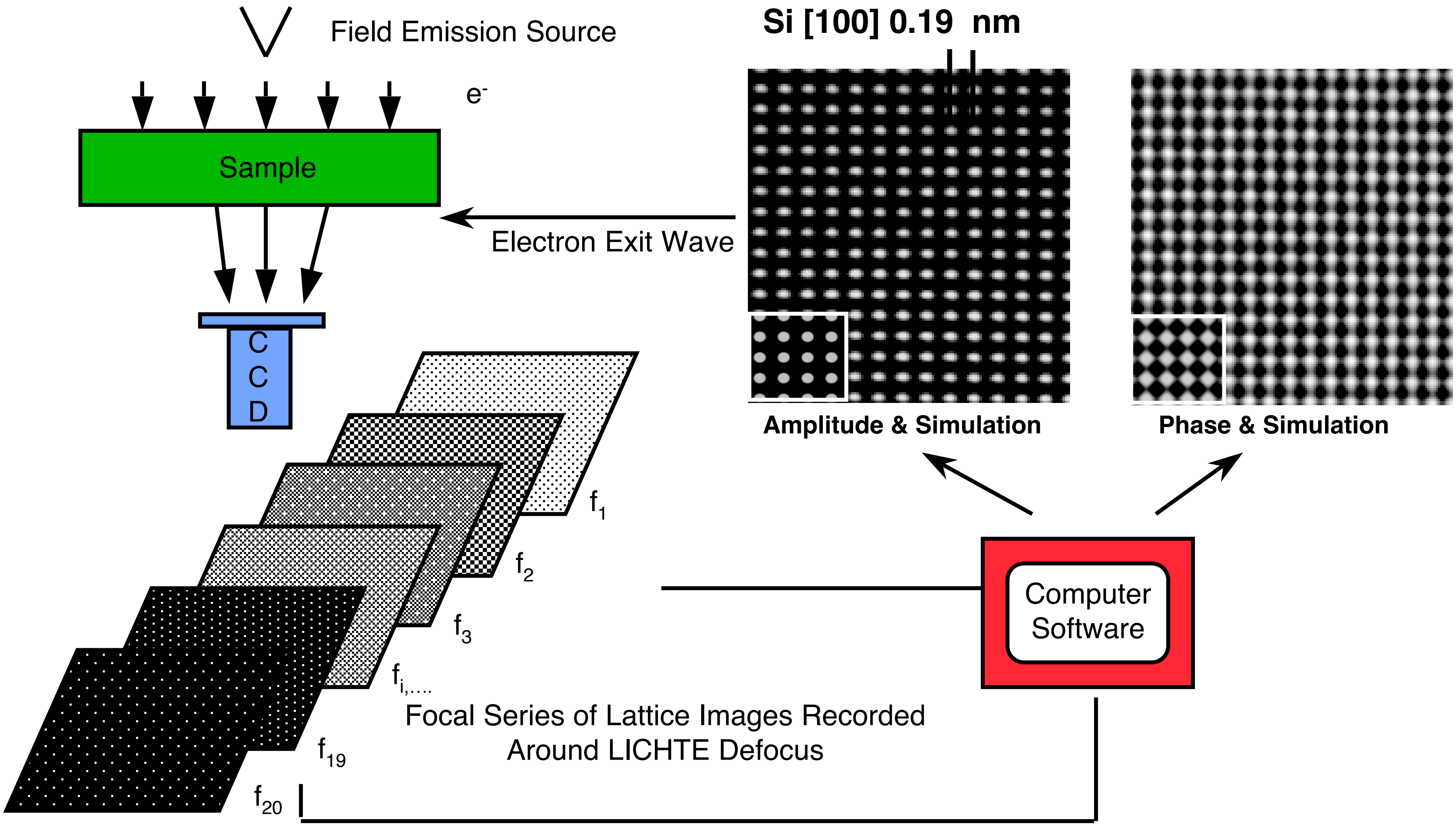
Image formation: delocalization



- We must fill the gaps in Fourier space (Focus series)
- We can solve the phase problem simultaneously

From Lattice Images to Electron Exit Waves

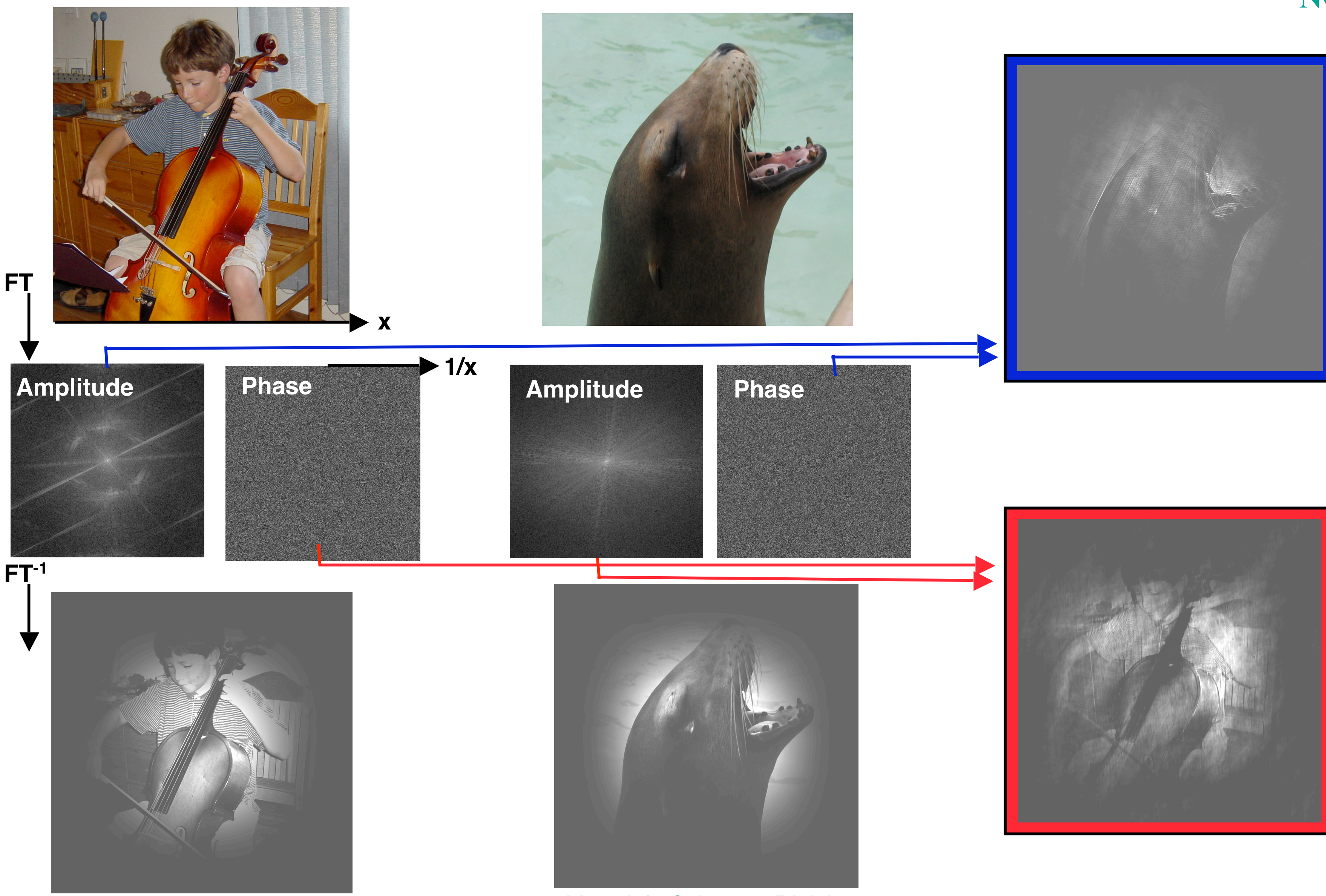
Solving the phase problem; removing aberrations & delocalization



W.M.J. Coene, A. Thust, Ultramicroscopy 64 (1996) 109 ,211
F.R. Chen, R. Kilaas et al. 2005

On the Importance of Phases

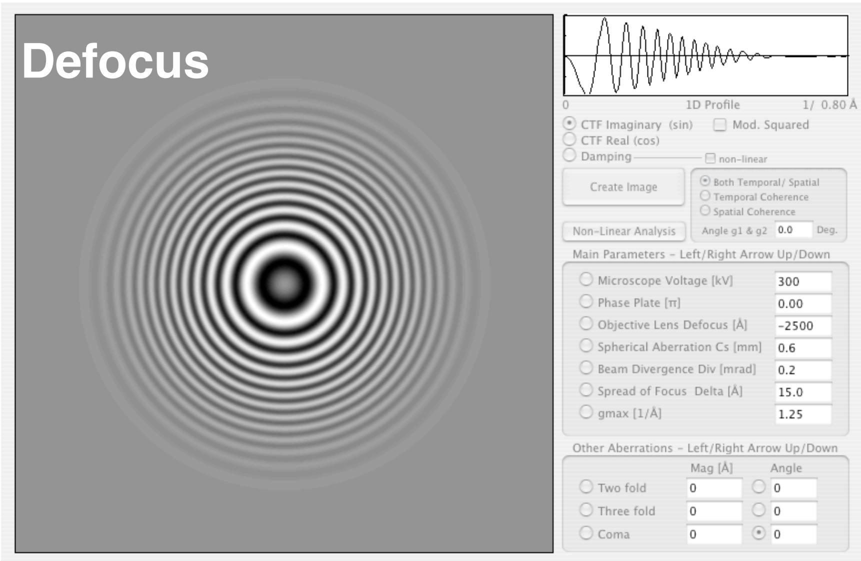
Phases carry most of the information in an image

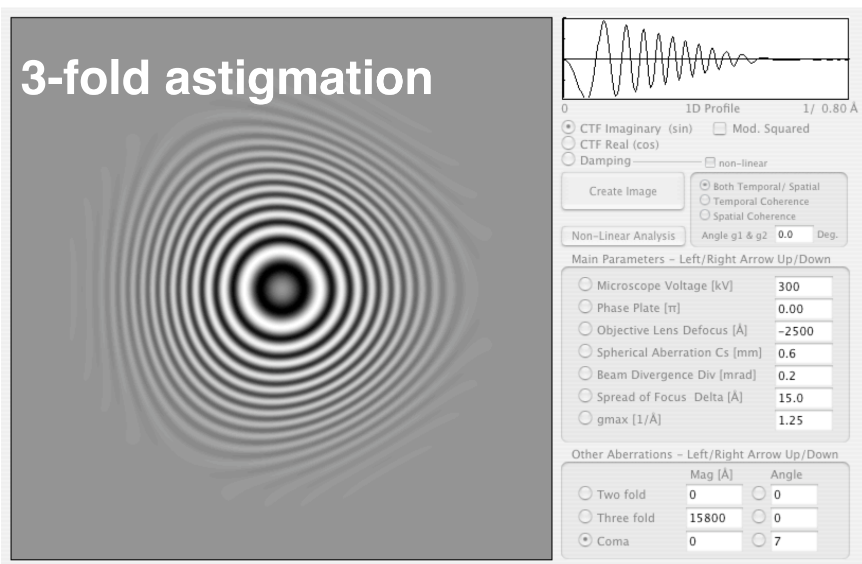


Lens Aberrations

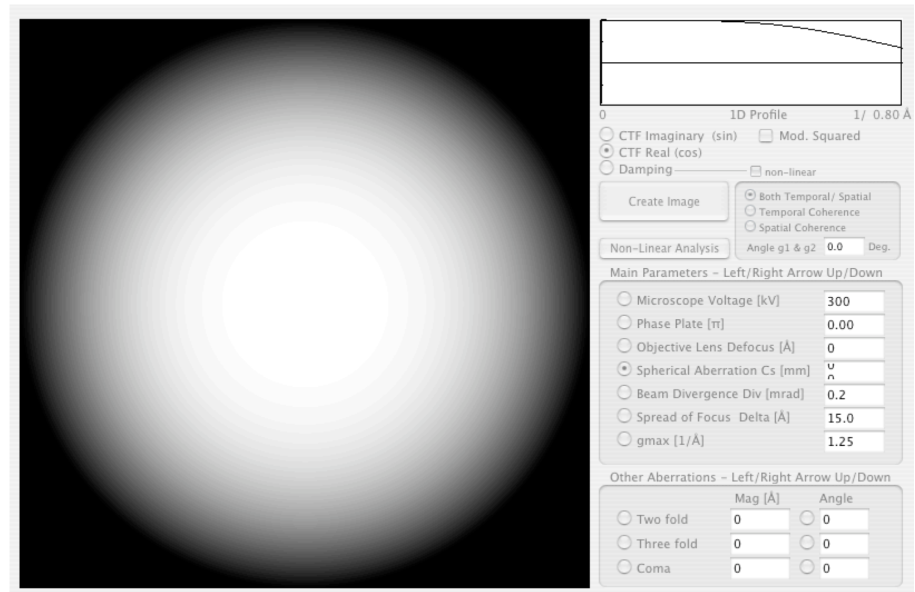
TEM: Point spread function

NCEM





Cs corrected amplitude imaging



In TEM aberration can be corrected by Software & hardware

Higher Order Lens Aberrations

$$
\begin{aligned}
E(\alpha,\overline{\alpha}) = \mathrm{Re}\{ & C_1\alpha\overline{\alpha} + \frac{1}{2}A_1\overline{\alpha}^2 \\
& + B_2\alpha^2\overline{\alpha} + \frac{1}{3}A_2\overline{\alpha}^3 \\
& + \frac{1}{2}C_3\alpha^2\overline{\alpha}^2 + S_3\alpha^3\overline{\alpha} + \frac{1}{4}A_3\overline{\alpha}^4 \\
& + B_4\alpha^3\overline{\alpha}^2 + D_4\alpha^4\overline{\alpha} + \frac{1}{5}A_4\overline{\alpha}^5 \\
& + \frac{1}{3}C_5\alpha^3\overline{\alpha}^3 + S_5\alpha^4\overline{\alpha}^2 + R_5\alpha^5\overline{\alpha} + \frac{1}{6}A_5\overline{\alpha}^6 \\
& + B_6\alpha^4\overline{\alpha}^3 + D_6\alpha^5\overline{\alpha}^2 + F_6\alpha^6\overline{\alpha} + \frac{1}{7}A_6\overline{\alpha}^7 \\
& + \frac{1}{4}C_7\alpha^4\overline{\alpha}^4 + S_7\alpha^5\overline{\alpha}^3 + R_7\alpha^6\overline{\alpha}^2 + G_7\alpha^7\overline{\alpha}^1 + \frac{1}{8}A_7\overline{\alpha}^8 \} .
\end{aligned}
$$

name	symbols						
defocus	C_1						
n-th order spherical aberration			C_3		C_5		C_7
n-th order axial coma		B_2		B_4		B_6	
n-th order star aberration			S_3		S_5		S_7
n-th order three-lobe aberration				D_4		D_6	
n-th order rosette aberration					R_5		R_7
n-th order pentacle aberration						F_6	
n-th order chaplet aberration							G_7
ν-fold astigmatism	A_1	A_2	A_3	A_4	A_5	A_6	A_7

M. Haider

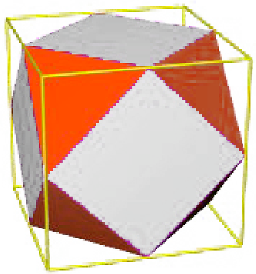
- **There are axial (listed) and off-axial aberrations**
- **Often only C_1 (Δf) and C_3 (C_s) are considered**

$$\chi(g) = 1/2\ \Delta f\ \lambda\ g^2 + 1/4\ C_s\ \lambda^3\ g^4$$

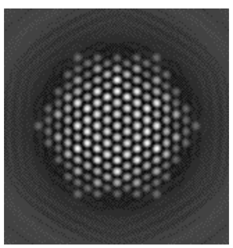
- **OÅM: A_1, A_2, A_3, B_2, (C_1 and C_3 by EWR)**
- **Higher order aberrations become important if the resolution increases (TEAM-Project --- 0.05 nm)**
- **It is challenging to correct all aberration by software only**

Cs Corrected Microscopy

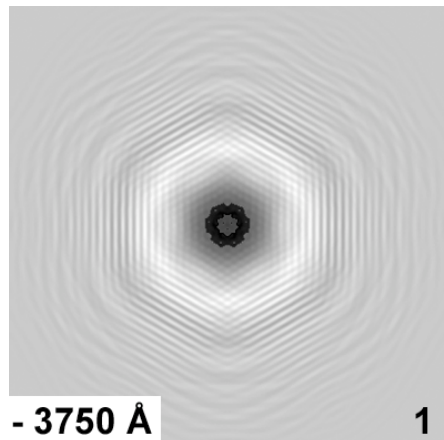
Advantages



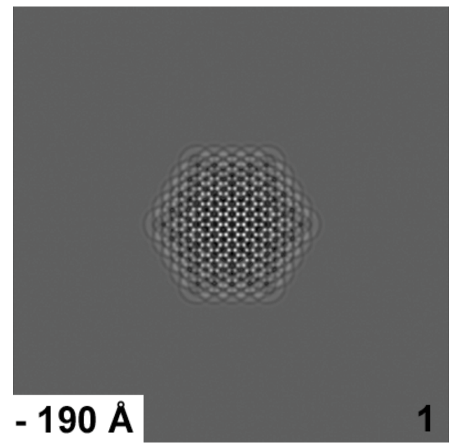
cuboctahedron



Exit wave



Focal series: CM300 (150 kV)

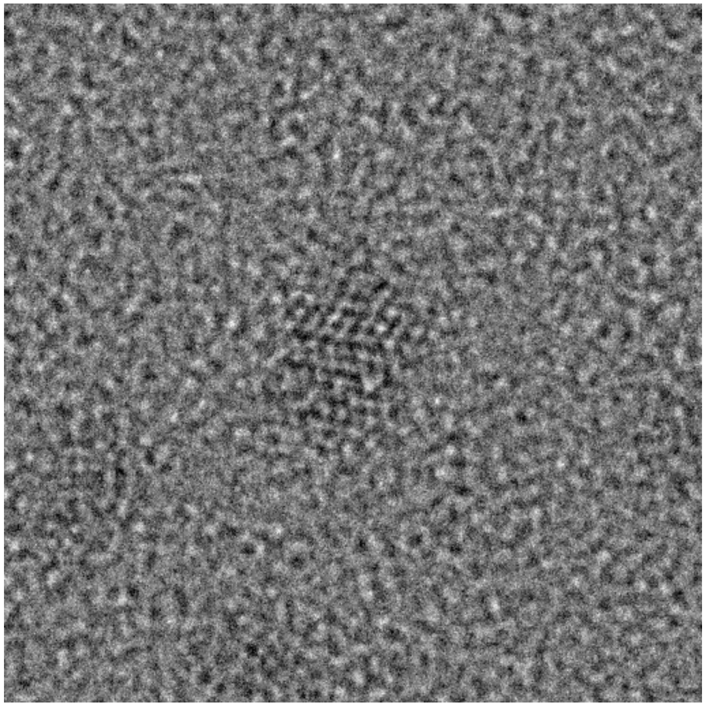


Cs corrected TEAM (150 kV)

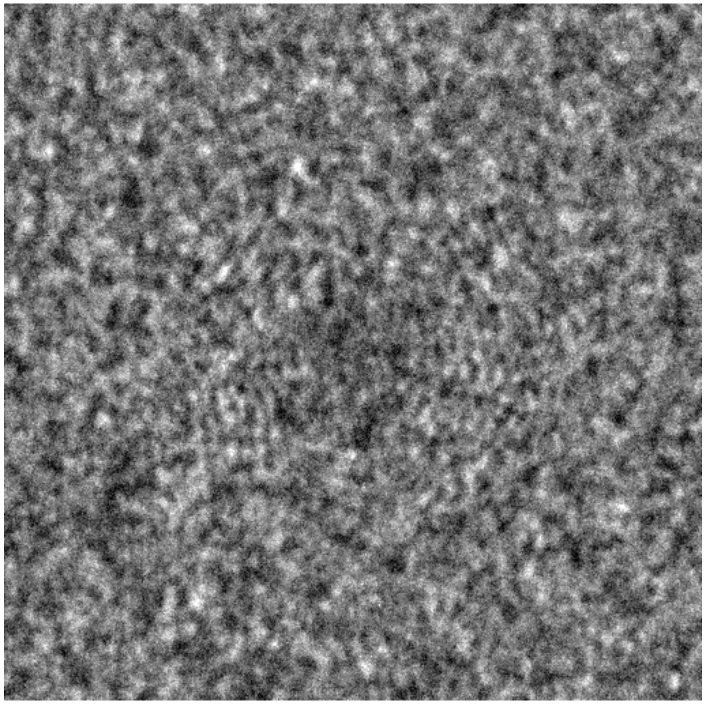
- **Image delocalization of small particles can wipe out lattice fringes**
- **Cs correction reduces the effect largely**
- **It will always be beneficial to do EWR in a Cs corrected microscope Access to waves**
- **What happens in the presence of noise?**

Software correction

Experimental limitations



FePt on carbon, Scherzer defocus



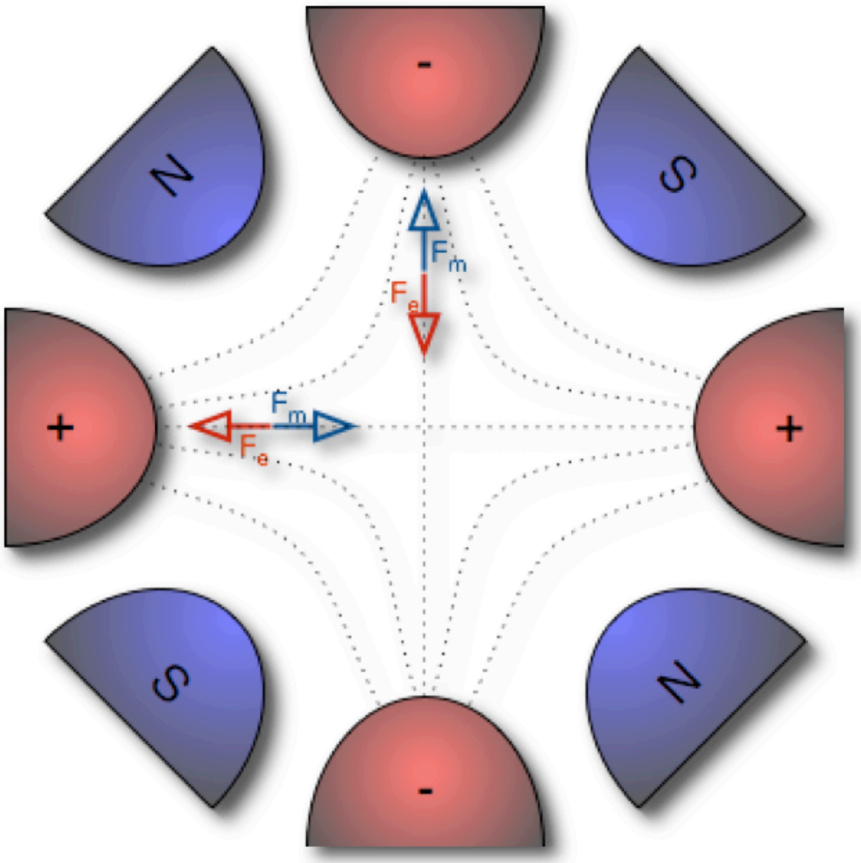
Δf~ Lichte defocus (-260 nm)

- **Experiment:**

  Image delocalization can be a significant limitation for the reconstruction of small particles on a carbon film

Hardware Correction: Wien Filter

Crossed electric + magnetic quadrupoles



Use magnetic and electric fields to deflect electrons in opposite directions.

$$for\ v_0 = E/B, \quad \mathbf{F} = 0$$

Electrons with v_0 will not be affected. All other "colors" will be distorted.

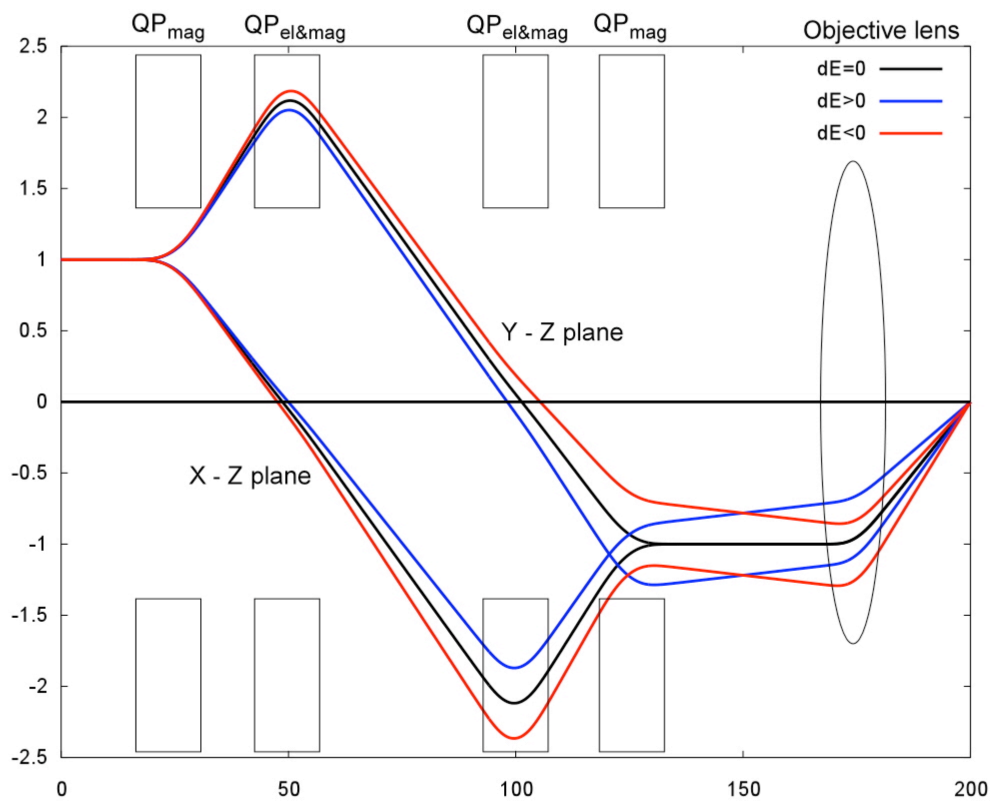
$$\mathbf{F} = q\{\mathbf{E} + (\mathbf{v} \times \mathbf{B})\}$$

Approach to Aberration Correction

Rose - CEOS

NCEM

- **Superaplanator - Cs/Cc corrector**
  - **2 Wien filters**
  - **3 octopole elements**
  - **Use of symmetry to cancel aberrations.**



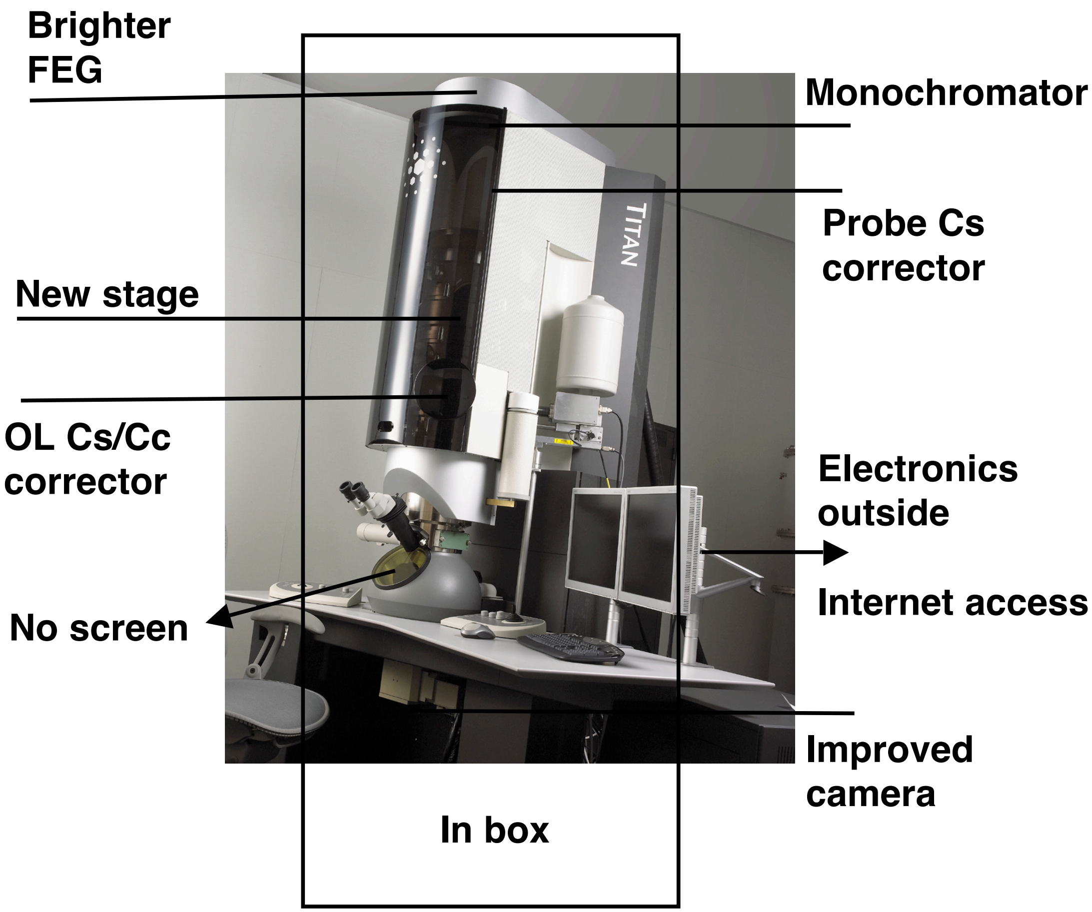
- **Cc/Cs correctors are tall**

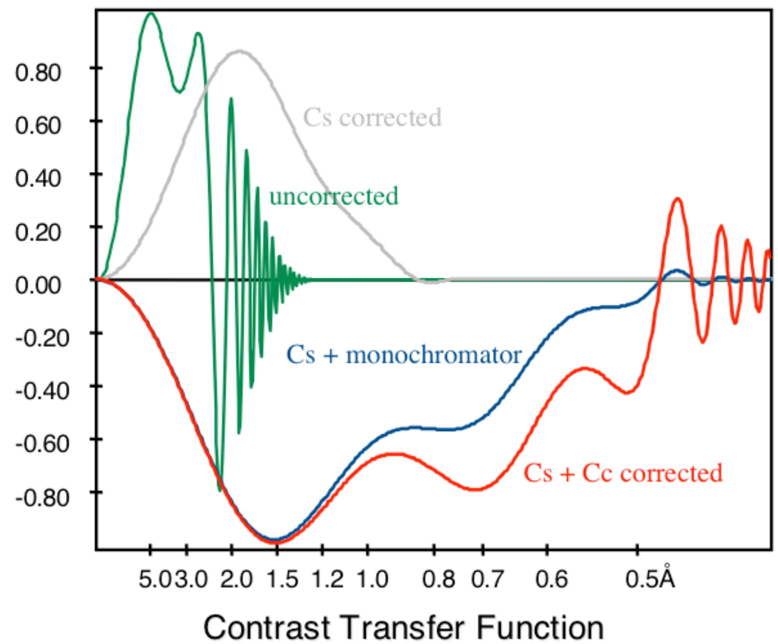
  0.8 - 1 m

Consequences for Design

Courtesy FEI Company

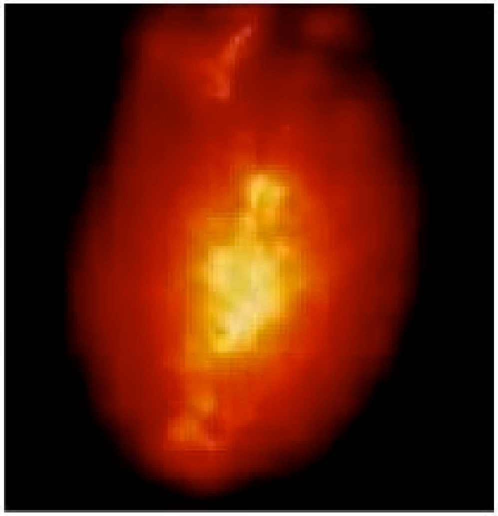
From Titan to TEAM

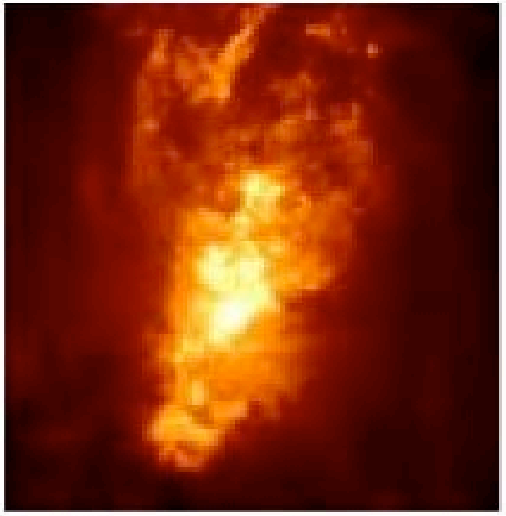




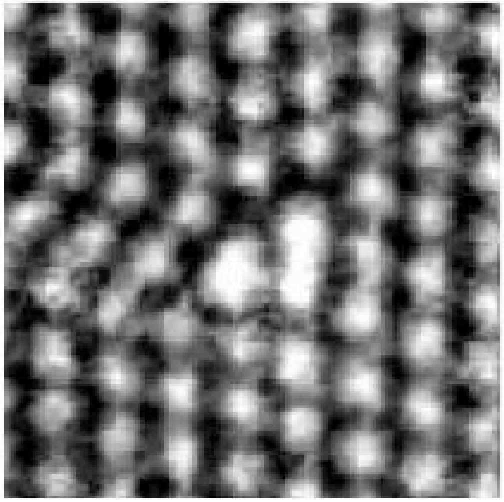
Numbers seem small but it is a long way from 0.8Å to o.5Å in STEM/TEM

TEAM Gets Us “Sharper” Images

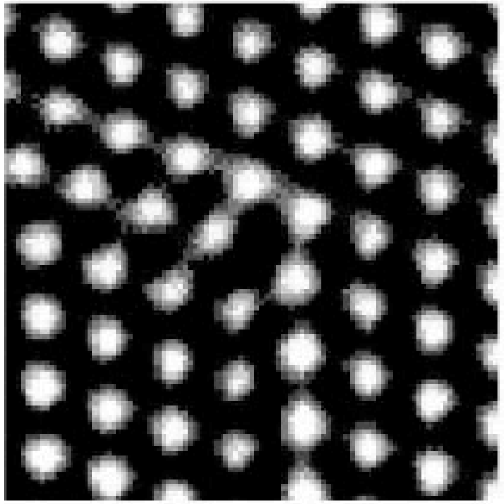




•Hubble telescope
before and after correction of
spherical aberration (1993)



Penisson, Berkeley ARM



Kisielowski, Juelich, 2002

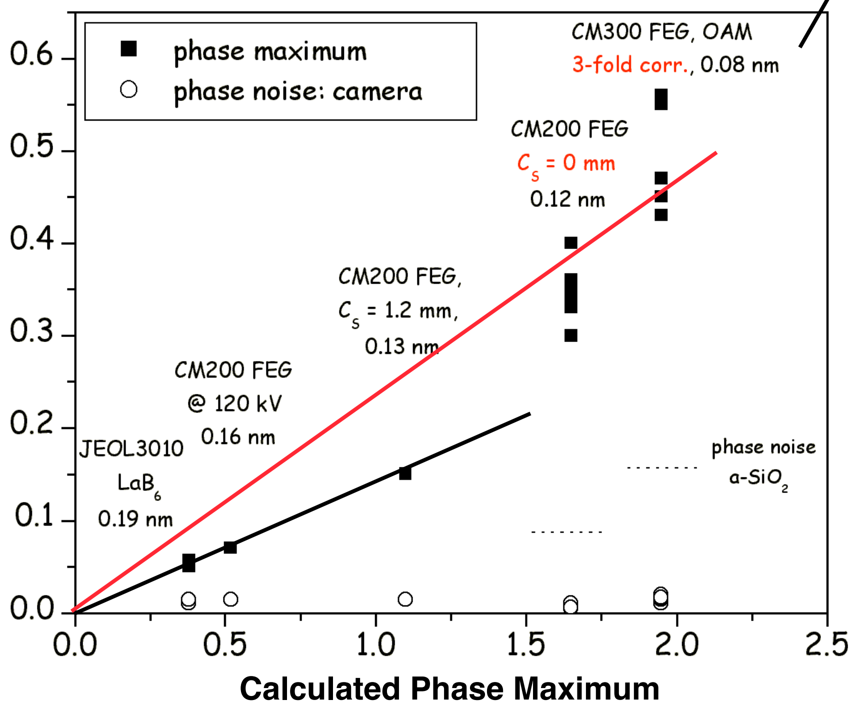
•Atomic resolution images of dislocation core in Au before and after correction of spherical Aberration

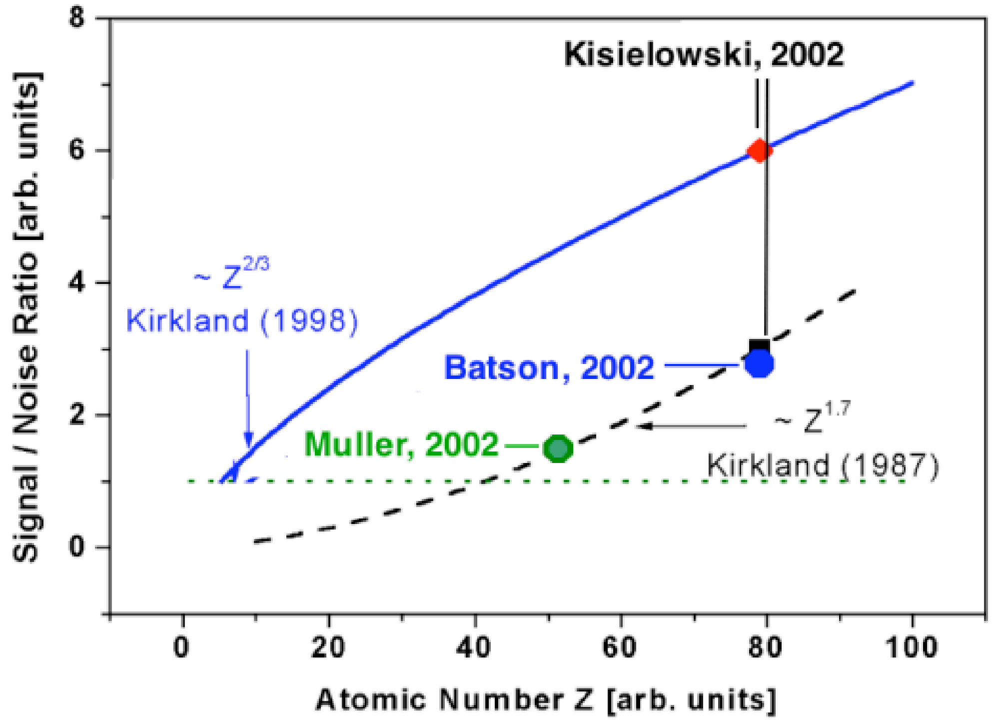
•How much is the improvement?

More Important than Resolution: Sensitivity

NCEM

TEAM Project



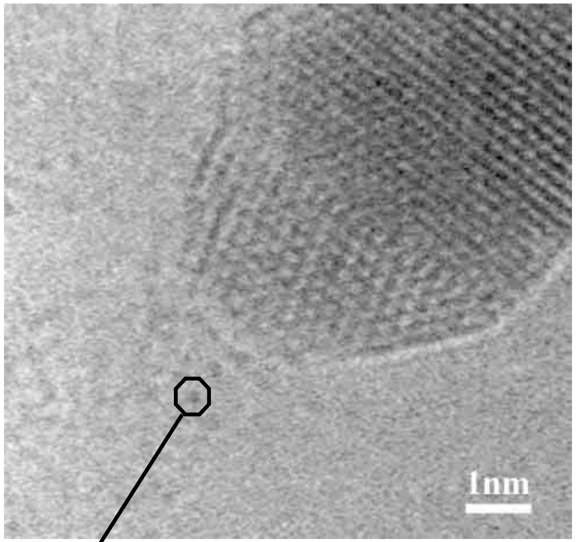


- Technological progress with EM has boosted sensitivity
  FEG, aberration correction,..
- Single atoms of most elements from the Periodic Table can be detected
- TEAM Project will provide the “ultimate” resolution / sensitivity

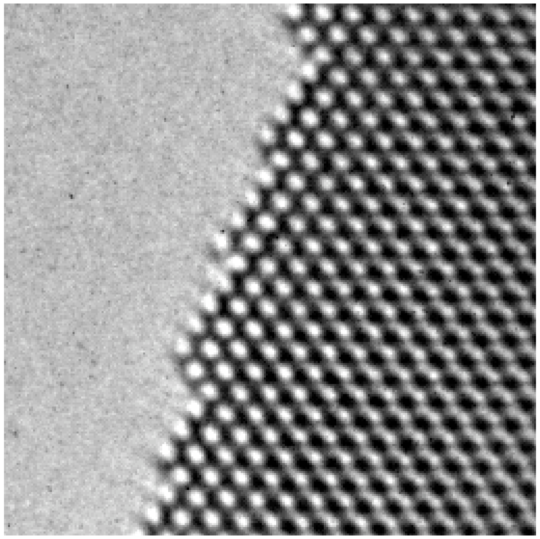
Single Atom Sensitivity:
Current abilities

NCEM

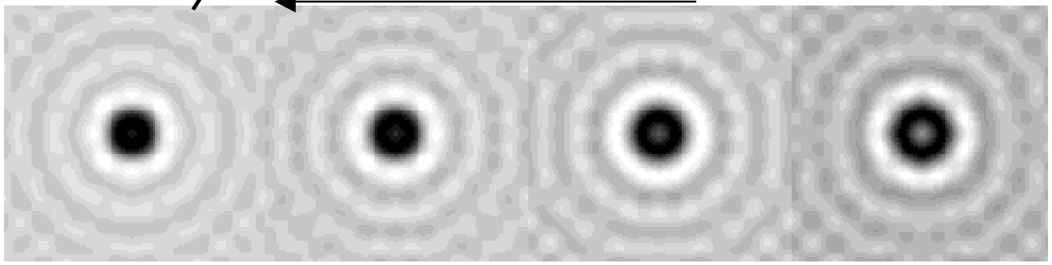
OAM: 30 frames/sec



Cs Correction: 1 frame/sec



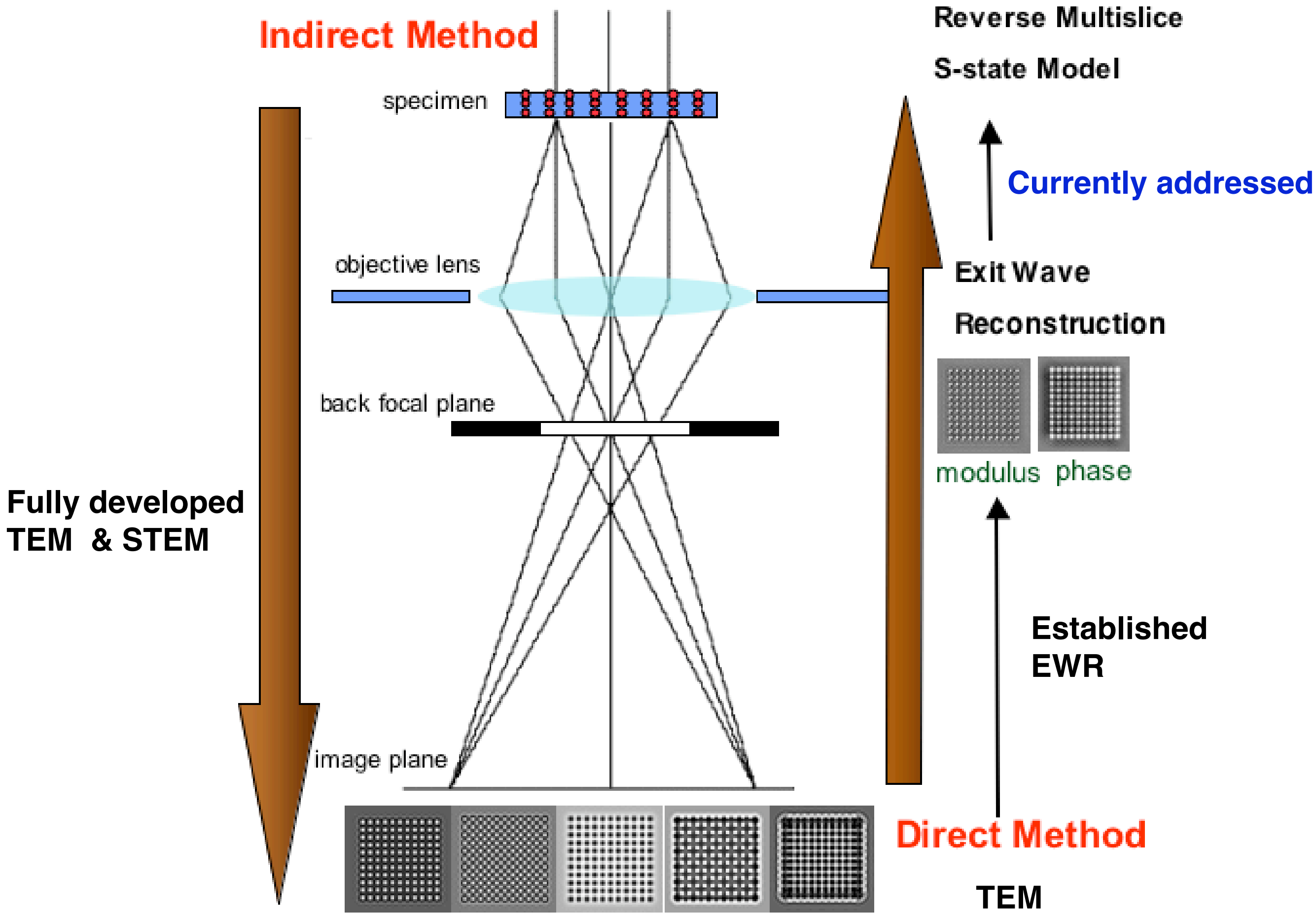
Δf (Simulation)



Intensity reflects atom position

Brighter gun & better cameras allow for better real time experiments with large S/N ratio

Experiments Must be Guided and Verified by Simulations



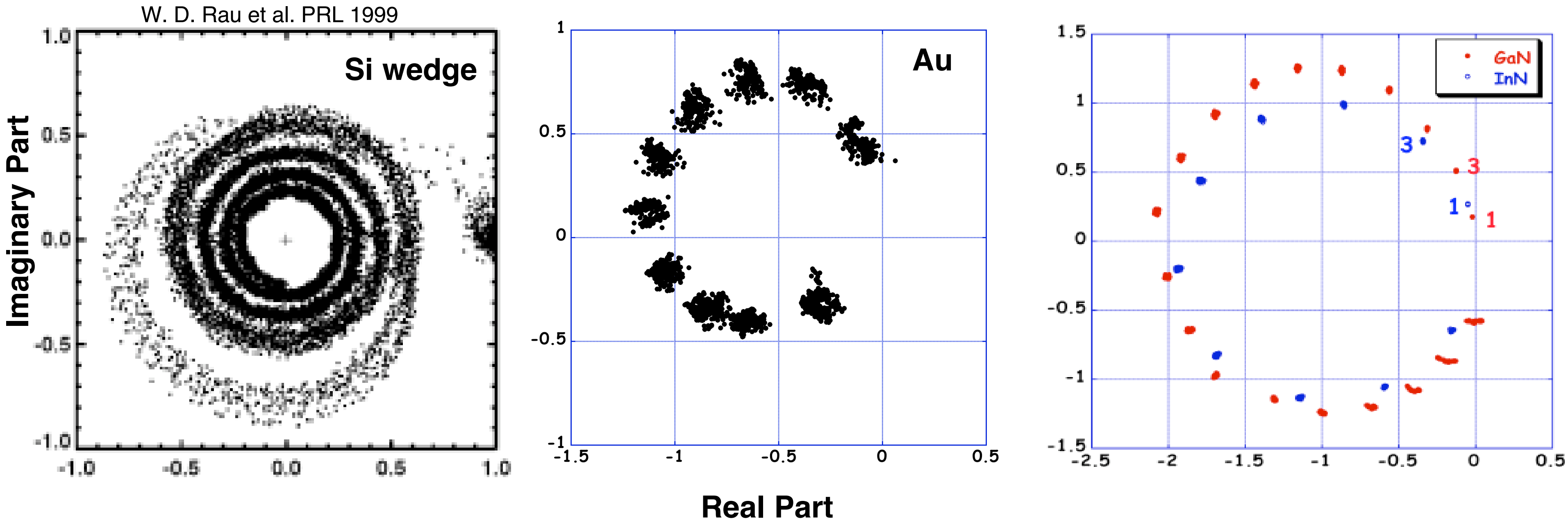
- Past: Utilization of indirect methods (lack of uniqueness)
- Today: Development of direct methods

  Progress with theory has enabled a reliable reconstruction of electron exit waves

  Structure reconstruction from exit ways becomes feasible

Interpretation of Waves

Amplitude - phase diagrams (Argand plot)

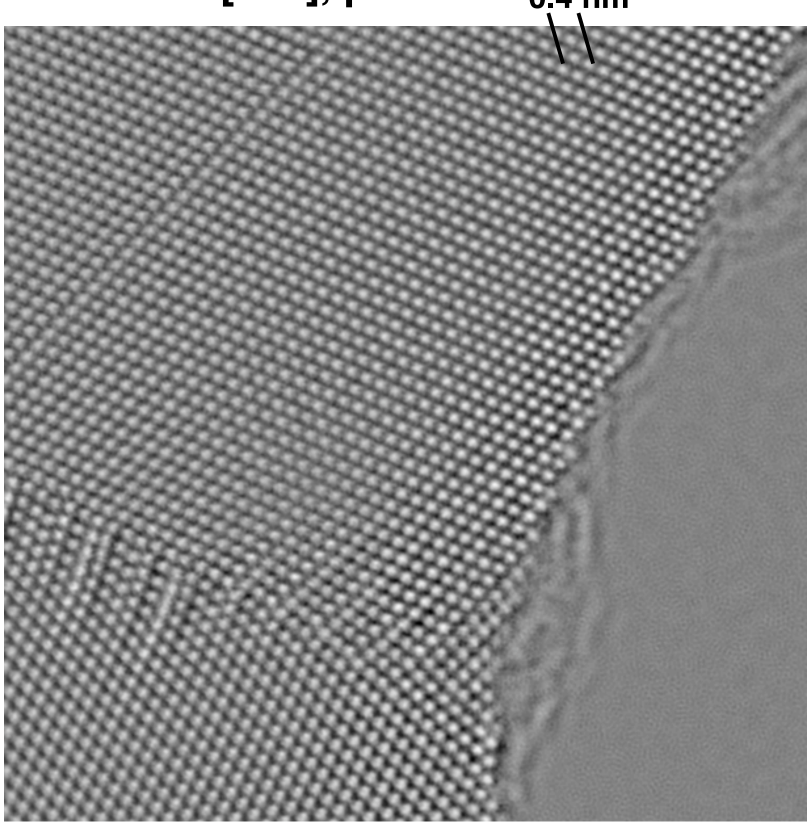


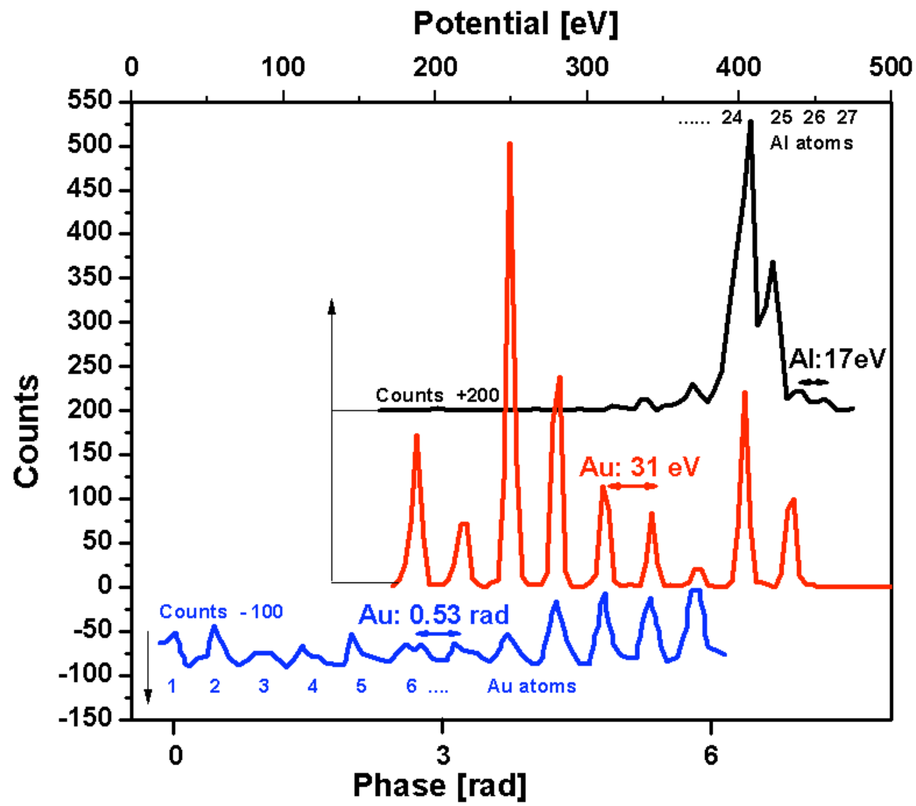
- A wave is entirely characterized by amplitude-phase diagrams
- In high performance microscopes Argand plots can be discrete
- Chemical composition can be revealed with single atom sensitivity
- Argand plots depict the full information - there is nothing more

Interpretation of Intensities

From exit waves to structure

Titan: Au [110], phase

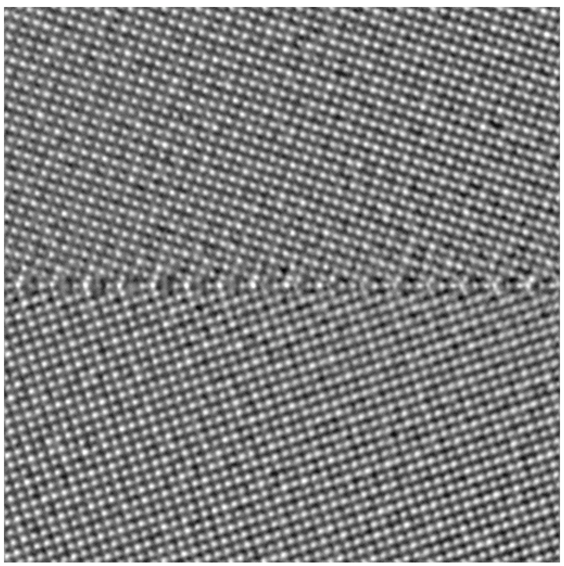




It is possible to count the number of atoms in columns by interpreting intensities

Intensity Interpretation
Current abilities; example Al:Cu

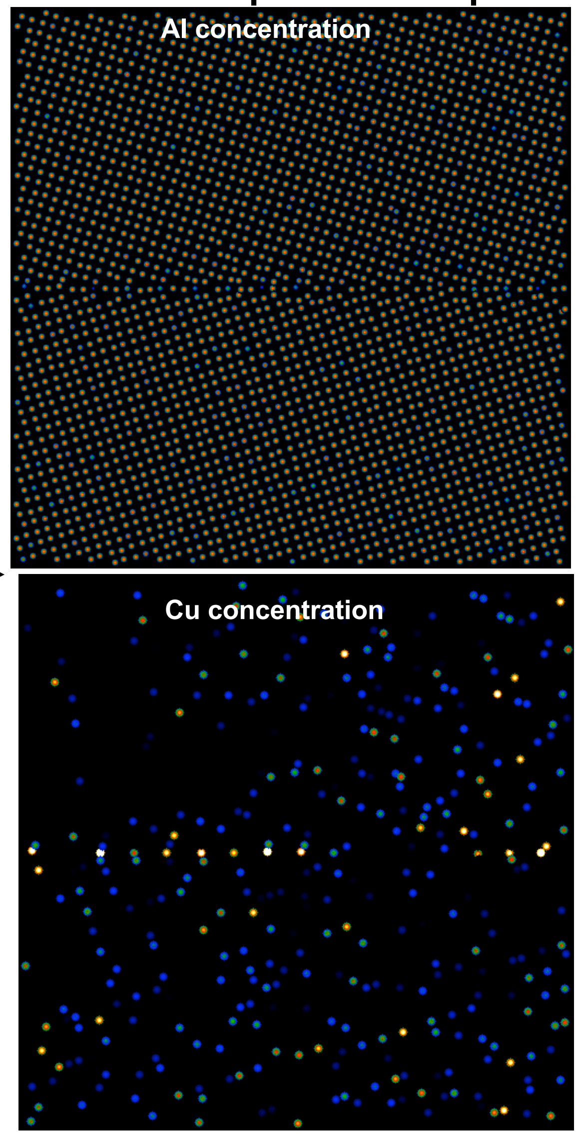
Exit wave (phase)



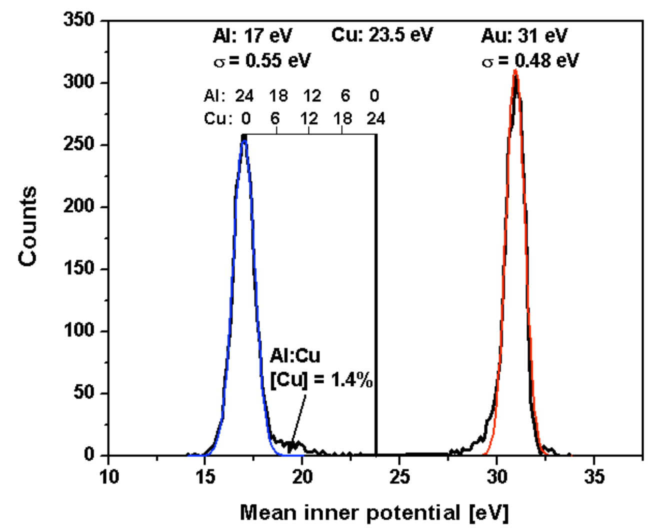
Al:Cu

F.R. Chen, C. Kisielowski, work in progress

Retrieved potential maps



Requirements:
- Recover waves
- Inversion of dynamic scattering

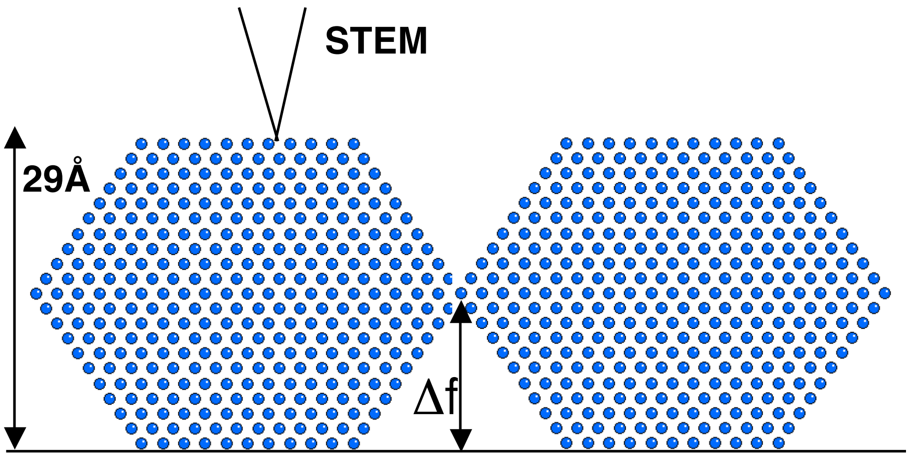


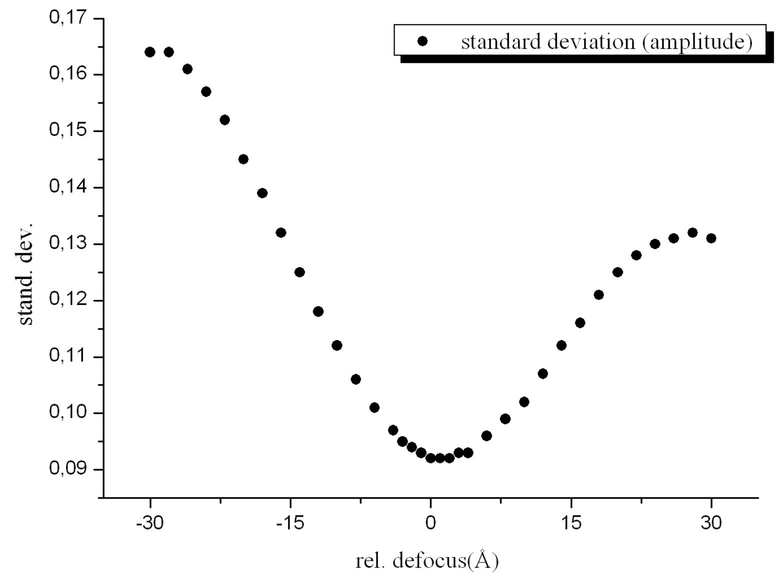
Limits
- Given by energy spread
- Recognizing a substitution of 1 Cu in column of 24 Al: σ = 0.27 eV

Limitations

Sample geometry, sample preparation & instrument stabilities

NCEM



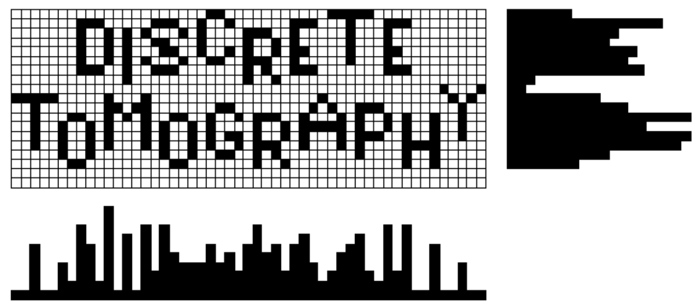


- The shape of nanoparticles can introduce local defocus changes
- Absolute amplitude/phase values noticeably over 2 Å
- Demands on stability are extremely high

Exploiting Sensitivity: Discrete Tomography

WORKSHOP ON

DISCRETE TOMOGRAPHY
AND ITS APPLICATIONS
June 13-15, 2005
New York City



Organizers

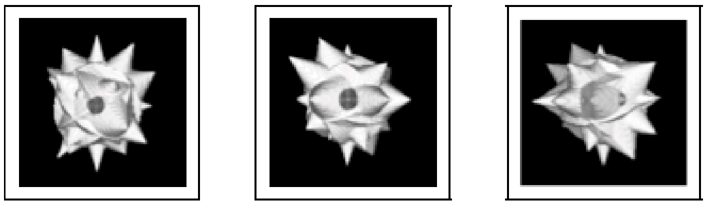
Gabor T. Herman   Attila Kuba

- •Workshops on DT are held regularly (~ 100 participants)
- •Discrete (binary) tomography is a general approach (biology, medicine, solids,…)
- •It is very suited to be applied to crystalline solids since atoms are discrete objects

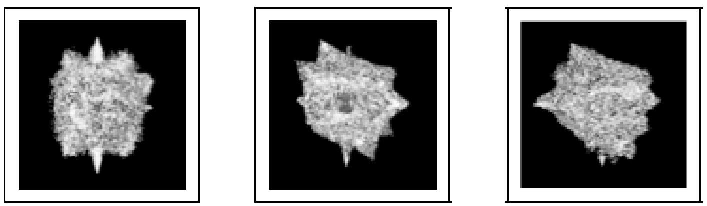
Discrete Tomography
Constraints

Binary Tomography

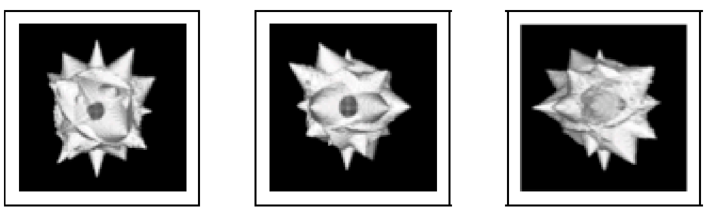
K.J. Batenburg / Electronic Notes in Discrete Mathematics 20 (2005) 247–261 259



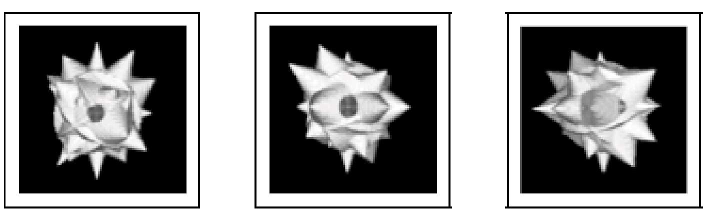
Original volume of size 128 × 128 × 128.



Reconstruction from two projections: (1, 0, 0) and (0, 1, 0).



Reconstruction from three projections: (1, 0, 0), (0, 1, 0) and (0, 0, 1).

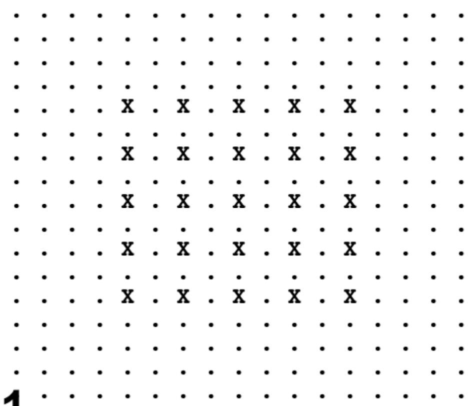


Reconstruction from four projections: (1, 0, 0), (0, 1, 0), (0, 0, 1) and (1, 1, 0).

- **Utilizing constraints reduces the number of projections**
- **Binary information is a powerful constraint**
- **Complex structures can be reconstructed**

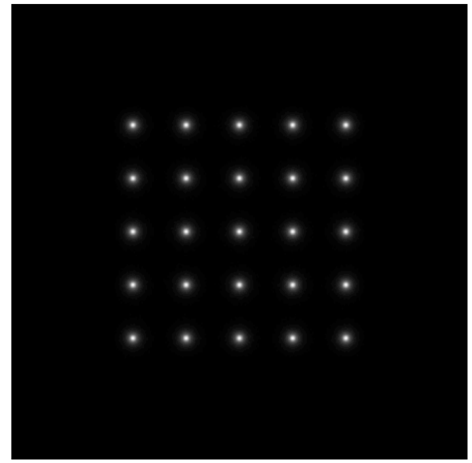
- **Atomic resolution (S)TEM may give binary information**
  - **Materials are made from single atoms**
  - **Constraint: atomicity**
- **Atomicity is NO limiting constraint for (S)TEM**
- **How can the number of atoms in columns be counted?**
  - **Utilize imaging along zone axis**
  - **(dynamic scattering)**

Discrete Tomography
Simulation



1

DT result



Projected potentials [100]

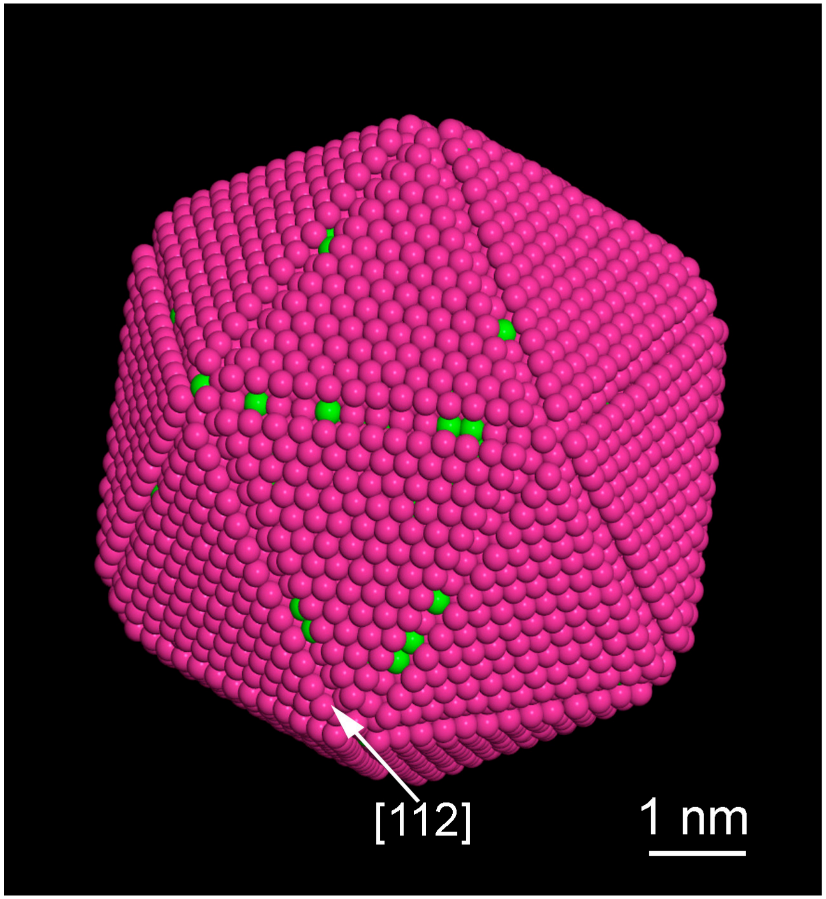
#projections	3				5				6			
orientations	[001], [110], [$\underline{1}$10]				[001], [110], [$\underline{1}$10], [011], [111]				[001], [110], [$\underline{1}$10], [011], [111], [1$\underline{1}$1]			
#count errors	10	20	40	80	10	20	40	80	10	20	40	80
% perfect	36	3	0	0	100	97	61	6	100	99	80	39
avg. #atom errors	1.28	4.51	11.92	27.13	0	0.03	0.57	3.73	0	0.01	0.23	1.18

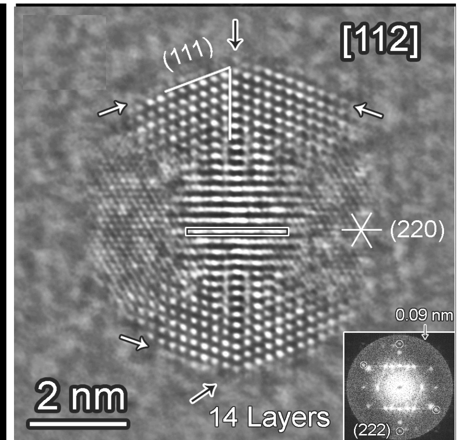
309 atoms

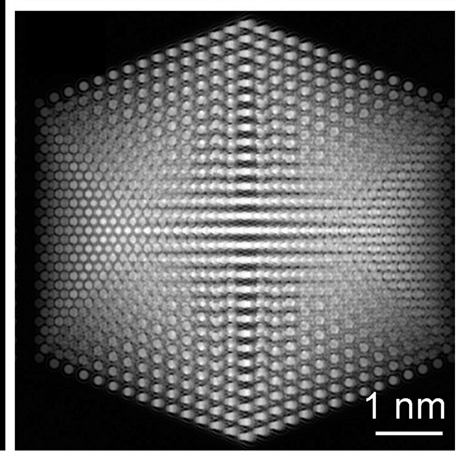
- **Reconstruction agrees with input structure: 3D can be done**
- **Number of projections determines error bars**

- **Experimentally, availability of sample holders is limiting**

A Glimpse into the Future:
Tomography with atomic resolution





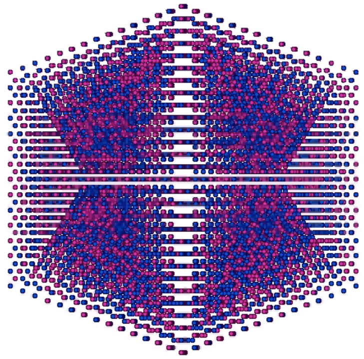


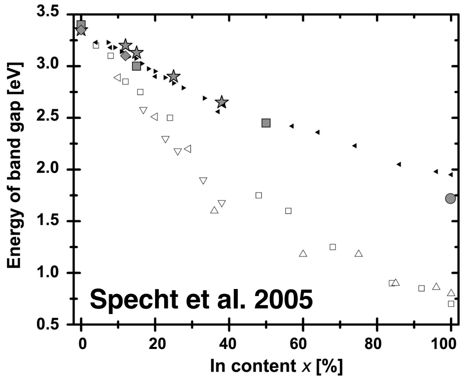
- **FePt icosahedra with core-shell structure (Nanomagnets with a catalytic shell)**
- **Strain relaxation by element redistribution and site-specific atom loss**

R. Wang et al. 2005, Nature Mat., 2005, in review

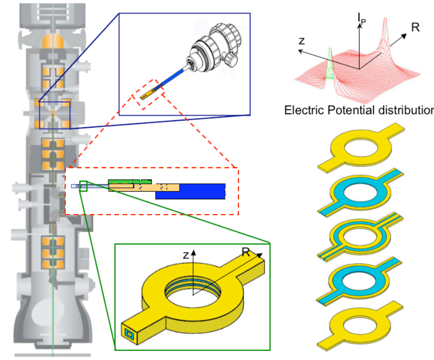
Conclusions

- **Aberration corrected EM enables novel science**
  - **TEAM will meet the Feynman Challenge**
  - **TEAM enables experiment design to meet scientific needs**
    - **Choose the best of STEM/TEM in one instrument**
    - **Do it in 3D**





- **Many Developments will largely benefit from TEAM Project**
  - **Monochromator**
  - **In-situ experiments (A. Minor)**
  - **Soft materials (phase plate)**
  - **Cameras (P.Denes)**
  - **Stages (A. Schmid)**
  - **…..**



W.K Hsieh et al. work in progress

- **There is plenty of room for innovation**