

# **Machine Learning in STEM 2023 Lecture 06**

## **Lecture 6: Introduction to Imaging**

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# Outline

## Introduction to Imaging

Image Formation Mechanisms

Imaging Techniques

HRTEM

## High Resolution Electron Microscopy

Scherzer Focus

The Damping Function

## Diffractogram

## Image Analysis

## Z-Contrast Imaging

Probe Formation

Scattering Process

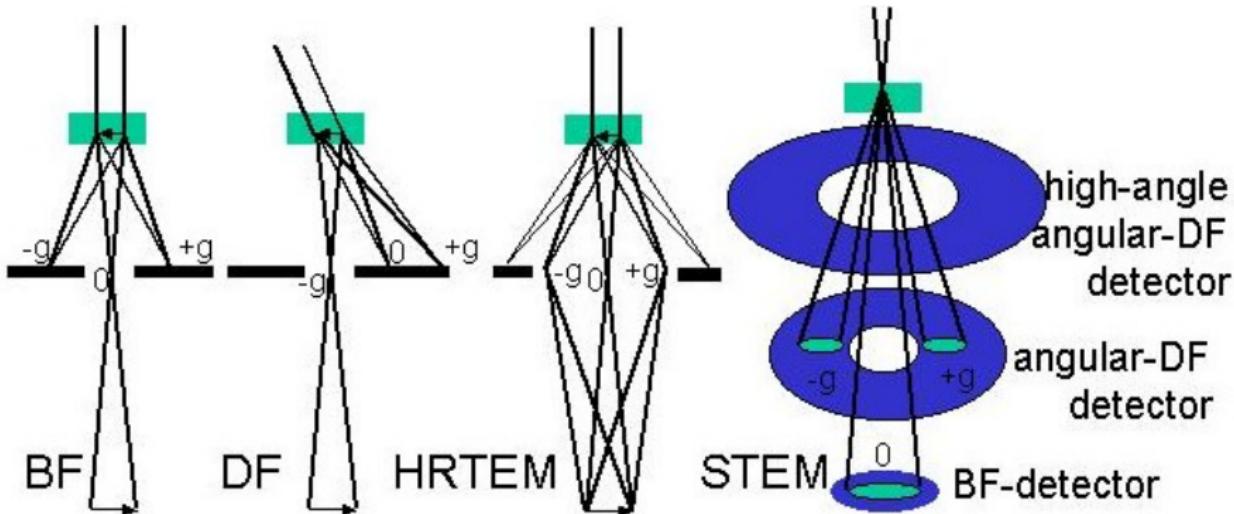
## Lens Aberrations

## Correction of Spherical Aberration

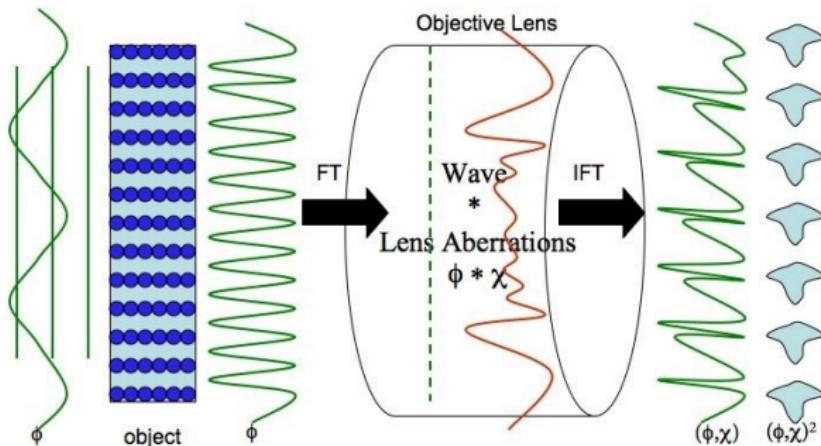
## High Resolution Electron Microscopy

- ▶ Diffraction/Strain Contrast
- ▶ Fresnel Contrast
- ▶ Phase Contrast
- ▶ Z-Contrast

# Imaging Techniques



## Fourier Optics of HRTEM

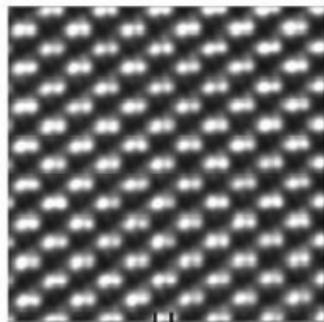


# Fourier Optic

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A plane waves hit the sample and get modified by the crystallographic structure. The object function consists of this modified plane waves directly after the sample. That is what we are after. The Fourier transformation of this object function is what we look at in selected area diffraction. Dynamic diffraction effects will play an important role here. Unfortunately, the image is now the square of the with each other interfering waves, which would not be quite as bad, but this is additionally mixed up by the microscope lens parameters. In the end, the phase information is lost and no calculation is possible to recapture the object function from the image.

Silicon



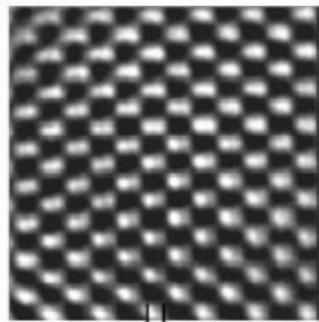
136 pm

Gallium Nitride



113 pm

Diamond



89 pm

# Resolution

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See Resolution Notebook for Rayleigh Criterion.

# Resolution in TEM

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A short summary of what we know so far

- ▶ The resolution in an electron microscope is not limited by the wavelength
- ▶ Resolution is dominated by aberrations of magnetic lenses
- ▶ In information science the appropriate description is a transfer function  $T$ .
  - ▶ How is the information transferred (distorted) by the system.
  - ▶ The transfer function is used in a convolution with the original information.

# TRANSFER FUNCTION

This transfer function  $T$  has several contributions:

$$T(u) = A(u)E(u)2\sin(\chi(u)) \quad (1)$$

- with:
- $T(u)$  Transfer function
  - $A(u)$  Aperture function
  - $E(u)$  Dampening function
  - $\chi(u)$  Aberration function

When  $T(u)$  is negative, positive phase contrast results, meaning that atoms would appear dark against a bright background. When  $T(u)$  is positive, negative phase contrast results, meaning that atoms would appear bright against a dark background. When  $T(u) = 0$ , there is no detail in the image for this value of  $u$ . (Note that we assume here that  $C_S > 0$ .)

The Scherzer focus is the defocus, which gives you an image that reflects (up to the point resolution) the sample (or object function) almost undisturbed for very thin sample locations.

The closest, we can get to the ideal curve of a contrast transfer function (nearly constant phase for all spatial frequencies) is when the aberration function  $\chi(u)$  is close to  $-120^\circ$  and  $\frac{d\chi}{du}$  is close to zero. If  $\chi(u)$  is close to  $120^\circ$  then  $\sin \chi(u)$  will be close to  $-1$ , and we want  $\sin(\chi)$  as large as possible.

# Spherical Aberration

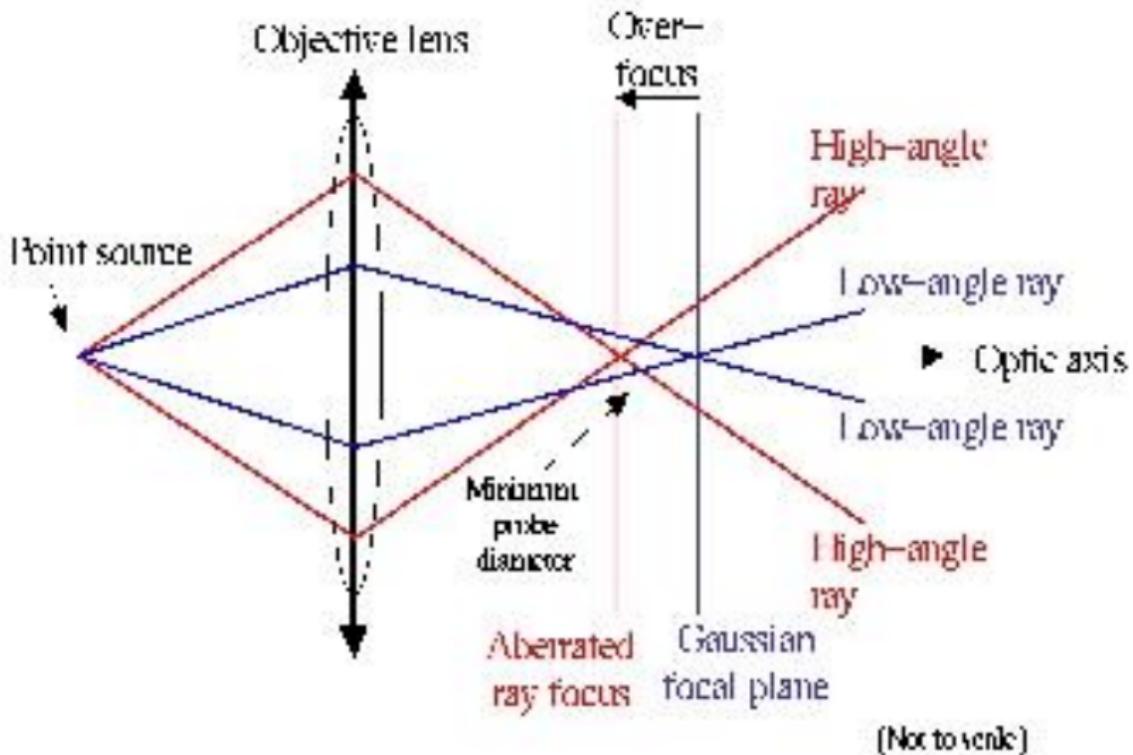


Figure: Simple electron trajectories in a lens with spherical aberration

# Spherical Aberration

A conical electron beam with angular convergence angle  $\alpha_0$  defined by the object aperture does not produce a sharp image point but the beam passes through a minimum  $d_{S,min}$ , in a plane of least confusion. In the Gaussian image plane (corresponds to the image plane if the plane of least confusion is the focal plane), the diameter is  $d'_{S,G} = 2C_S\alpha_0^3 M$ . The smallest diameter  $d_{S,min}$  can be estimated from  $d_{S,min} = 0.5C_S\alpha_0^3$ . All this originates from the wavefronts which are no longer perfectly spherical (see figure 1), that's where the name comes from.

The function  $\sin(\chi)$  is flat if  $\frac{d\chi}{du}$  is zero.

$$\frac{d\chi(u)}{du} = 2\pi\Delta f \lambda u + 2\pi C_s \lambda^3 u^3 \quad (2)$$

We set the left term to zero to get the flat portion and get:

$$0 = \Delta f + C_s \lambda^2 u^2 \quad (3)$$

When  $\chi$  is  $-120^\circ$  (or  $-2\pi/3$ ) we get:

$$-\frac{2}{3}\pi = \pi\Delta f \lambda u^2 + \frac{1}{2}\pi C_s \lambda^3 u^4 \quad (4)$$

Combining the two equations gives a certain defocus value the Scherzer focus:

$$\Delta f_{Sch} = - \left( \frac{4}{3} C_s \lambda \right)^{\frac{1}{2}} \quad (5)$$

At this value of the defocus we find that the first cross over of the contrast transfer function ( $\sin(\chi) = 0$  for  $\chi = \pi$ ) is:

$$u_{Sch} = 1.51 C_s^{-\frac{1}{4}} \lambda^{-\frac{3}{4}} \quad (6)$$

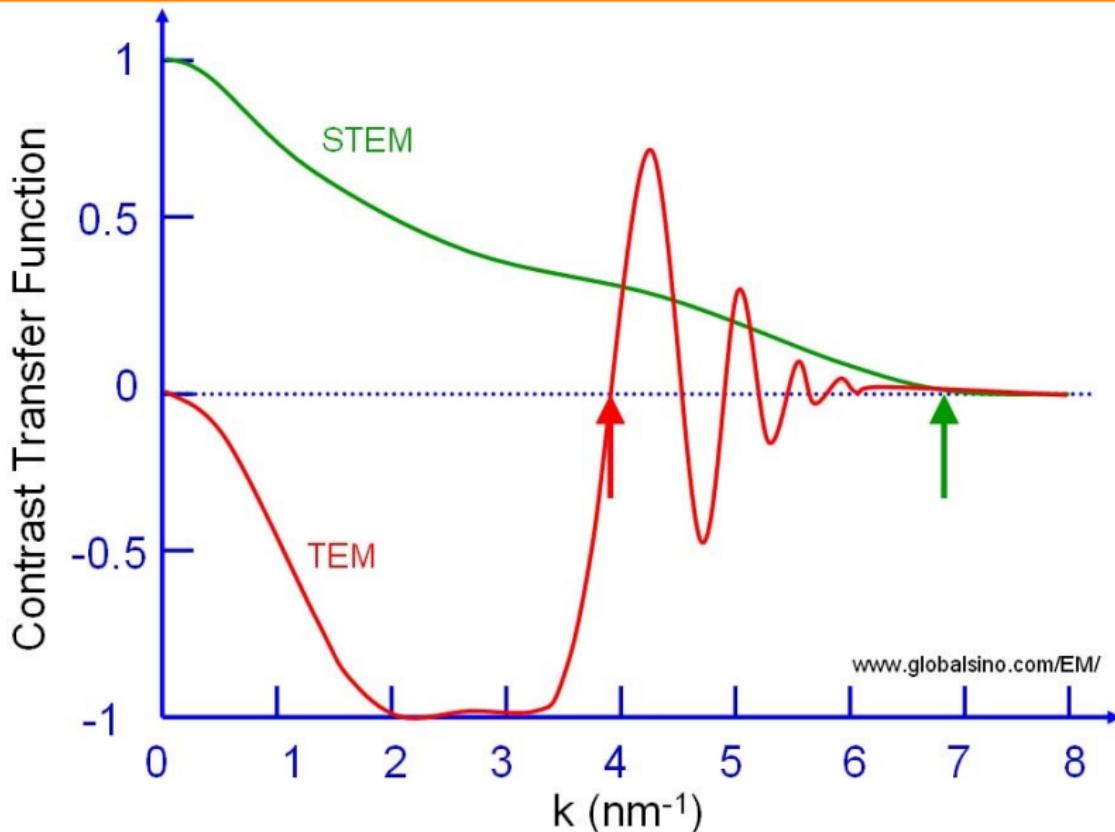
# Z-Contrast Imaging

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The point resolution is therefore defined as:

$$r_{Sch} = \frac{1}{1.51} C_s^{\frac{1}{4}} \lambda^{\frac{3}{4}} = 0.66 C_s^{\frac{1}{4}} \lambda^{\frac{3}{4}} \quad (7)$$

# Contrast Transfer Function



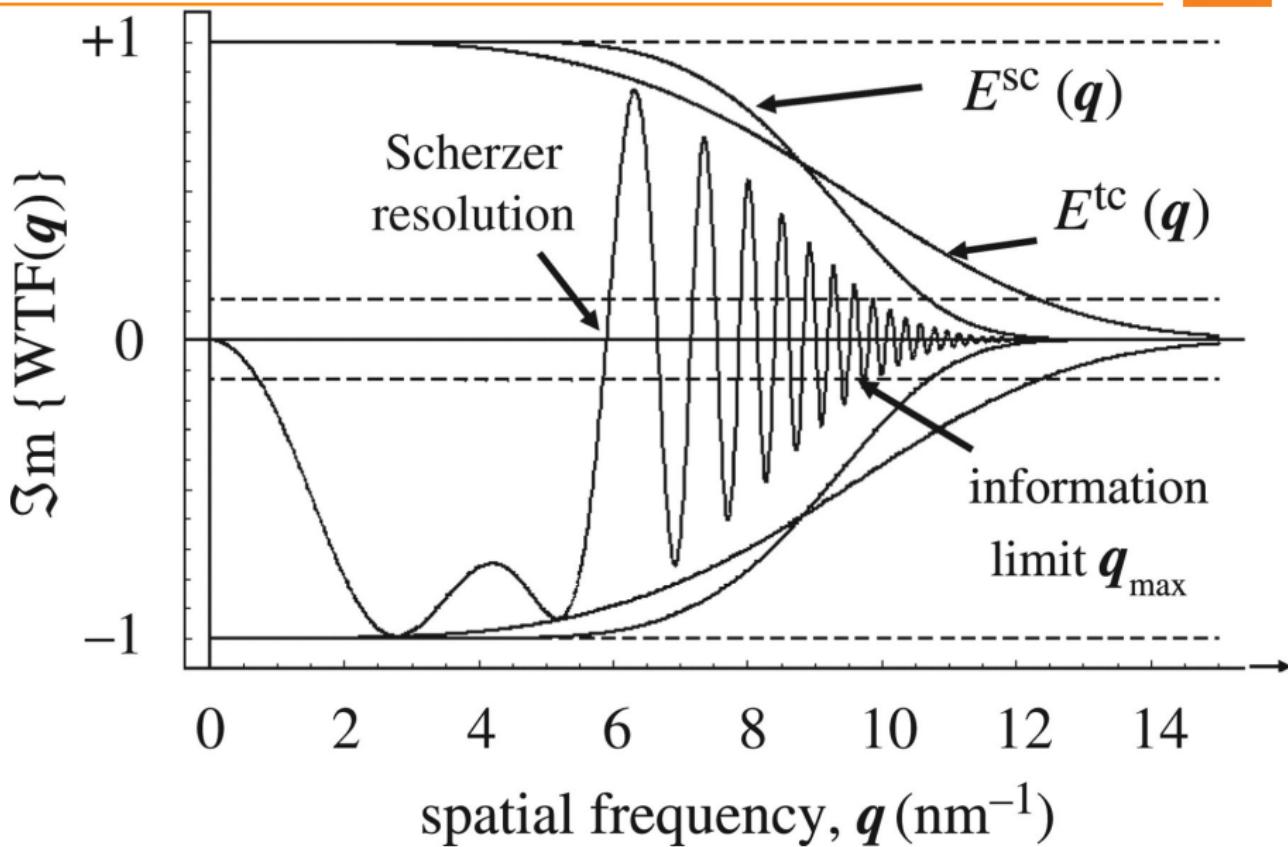
The plots of the  $\sin(\chi(\vec{u}))$  could extend as far out as you want, even beyond the resolution limit given by the Raleigh criterium. In practice, these plots extend to the so called information (retrieval) limit of the TEM, because of the envelope damping function. This damping function  $E(\vec{u})$  includes effects like the chromatic aberration and instabilities, that degrade the coherence of the electron beam. The effect of the damping function is a aperture function, which is independent of the defocus.

If you use a physical aperture to reduce noise in the picture, you should choose one of the size of the virtual aperture of this damping function. In other words, the biggest physical aperture should go out to the information limit.

The following terms contribute to the envelope function:

- ▶  $E_c(u)$  chromatic aberration
- ▶  $E_s(u)$  source dependence due to the small spread of angles from the probe
- ▶  $E_d(u)$  specimen drift
- ▶  $E_v(u)$  specimen vibration
- ▶  $E_D(u)$  detector

# Contrast Transfer Function



The envelope function  $E(u)$  is the product of all these:

$$E(u) = E_c(u)E_s(u)E_d(u)E_v(u)E_D(u) \quad (8)$$

The chromatic aberration is well known:

$$E_c(u) = \exp \left[ -\frac{1}{2}(\pi\lambda\delta)^2 u^4 \right] \quad (9)$$

The defocus spread  $\delta$  due to the chromatic aberration is defined as:

$$\delta = C_c \left[ 4 \left( \frac{\Delta I_{obj}}{I_{obj}} \right)^2 + \left( \frac{\Delta E}{V_{acc}} \right)^2 + \left( \frac{\Delta V_{acc}}{V_{acc}} \right)^2 \right]^{\frac{1}{4}} \quad (10)$$

The information limit given by the chromatic aberration is given by:

$$\rho_c = \left( \frac{2\pi\delta}{2} \right) \quad (11)$$

For FEG guns we also have to consider the source dependent of the defocus:

$$E_s(u) = \exp \left[ - \left( \frac{\pi \alpha}{\lambda} \right)^2 (\Delta f \lambda u + C_s \lambda^3 u^3)^2 \right] \quad (12)$$

This tells us, that the resolution limit can be degraded by choosing an too large of a semiangle  $\alpha$ .

The optimum defocus is then:

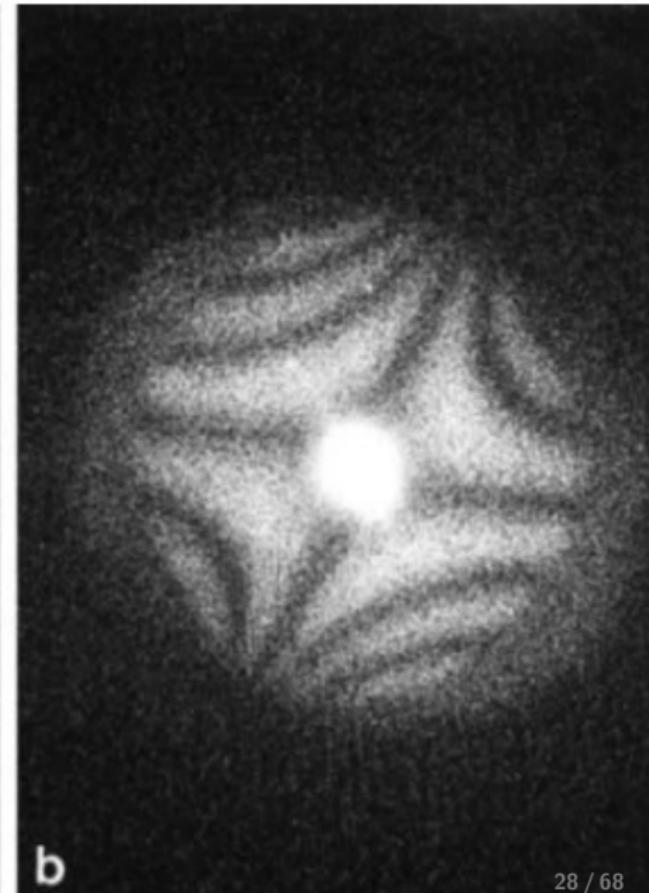
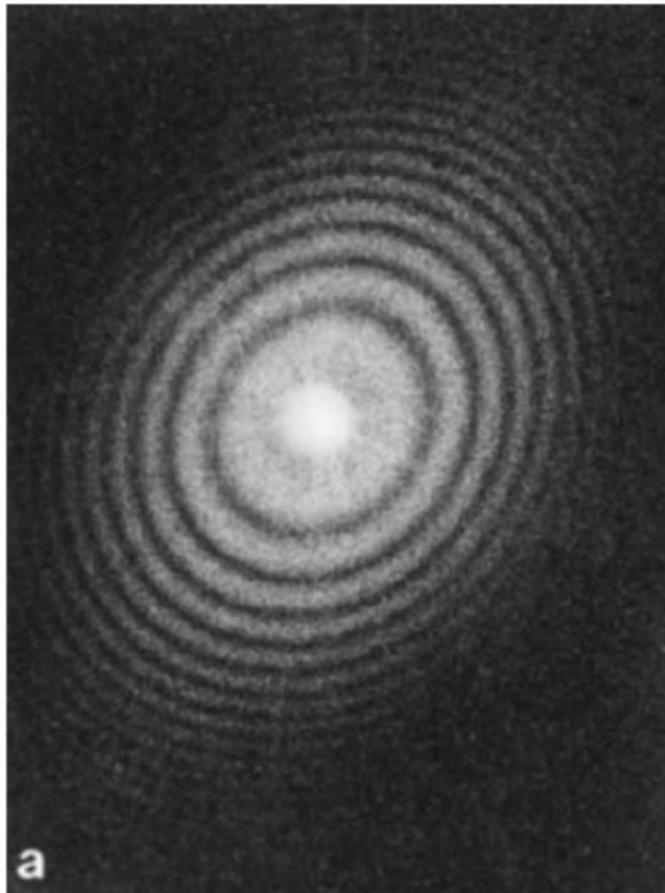
$$\Delta f_{opt} = -\frac{3}{4} C_s \lambda^2 u^2_{max} = -\frac{3}{4} \frac{C_s \lambda^2}{\rho_i^2}_{max} \quad (13)$$

# Diffractogram

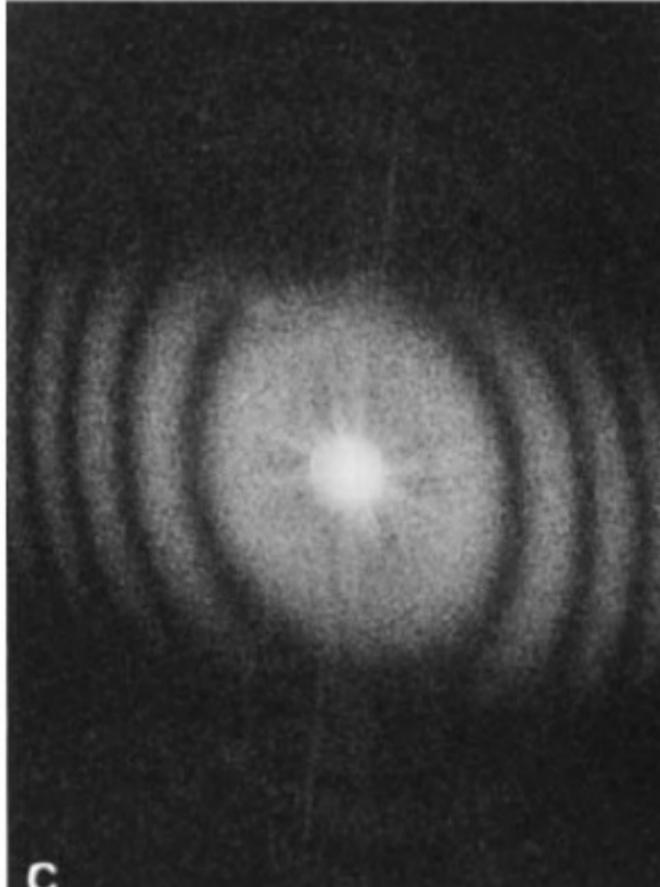
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We call a fourier transform of an atomic resolution image a diffractogram. It looks like a diffraction pattern, but it is really a diffraction pattern modified by the contrast transfer function.  
The diffractogram of amorphous materials allows us to characterize our imaging conditions.

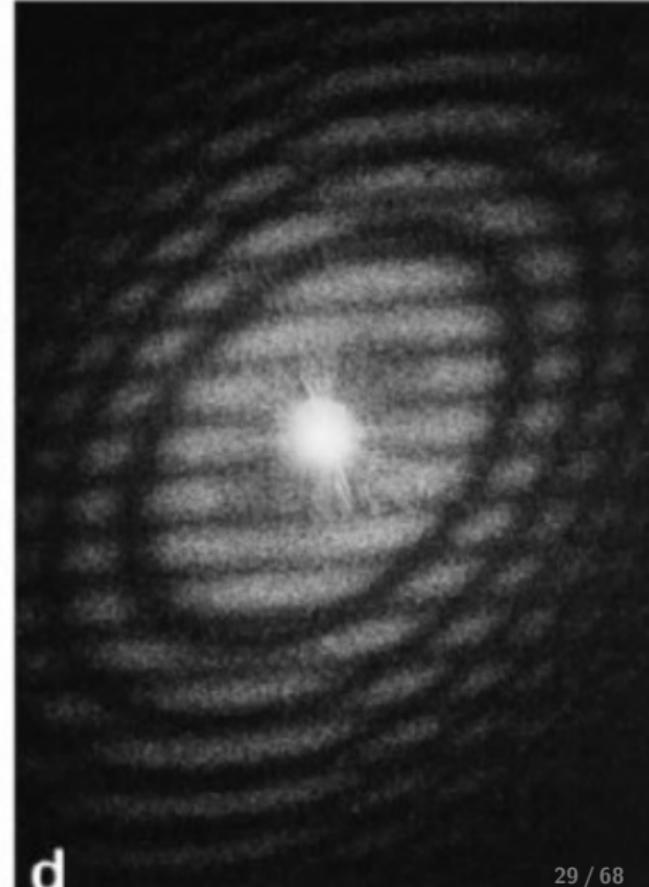
# Diffractogram



# Diffractogram



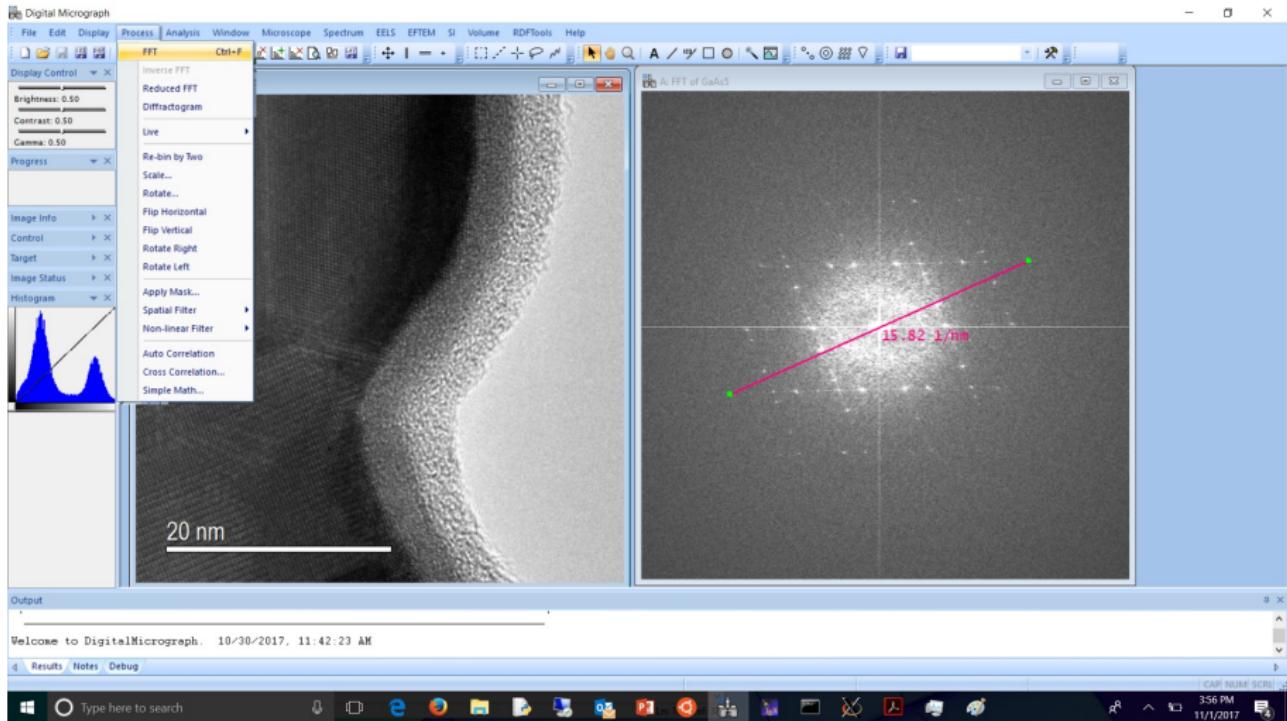
c



d

# Fourier Transform

Open the image ““GaAs.dm3”” with Digital Micrograph and do a Fourier Transform



What resolution was achieved?

# Simple Analysis of the Diffractogram

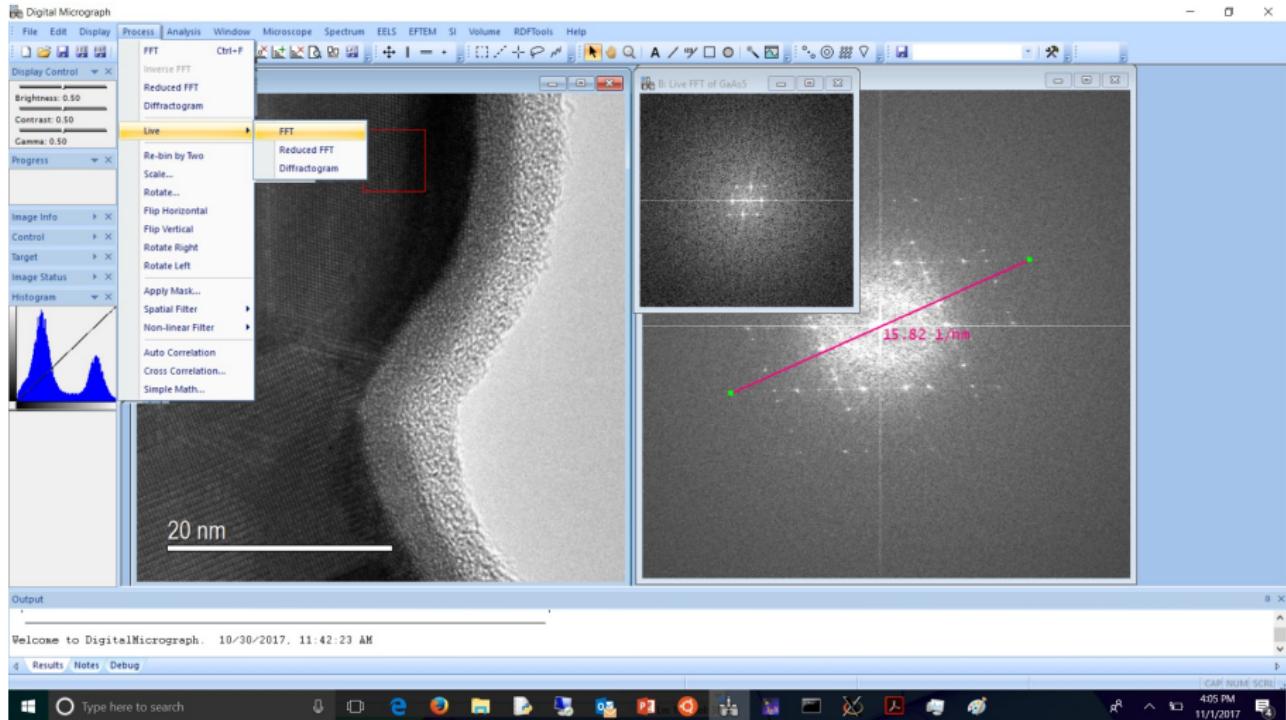
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1. What are the distances of the spots to the center?
2. What is the resolution?
3. Are there any aberrations?
4. What is the defocus?
5. Did anybody mess with this image before?

# Fourier Transform

Select a square area with a power of 2 (press down “Alt” button and select Live FFT.

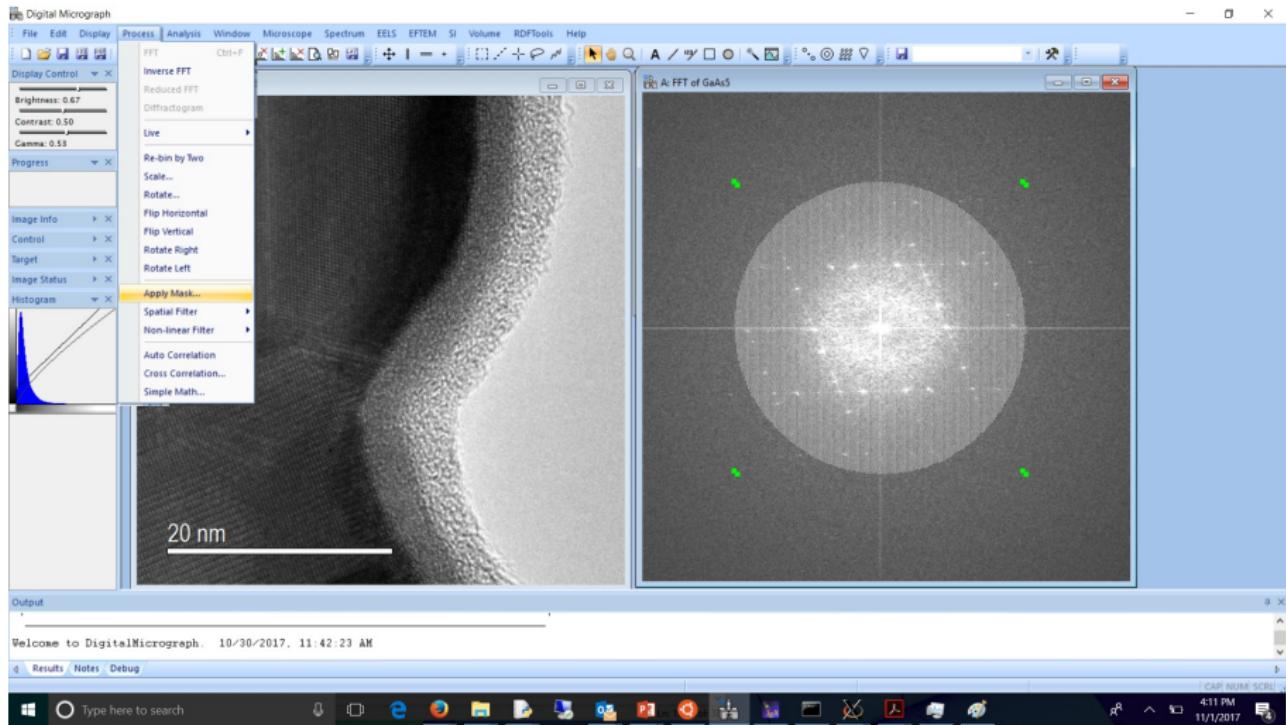


Where is what orientation?

# High Pass Fourier Filter

T

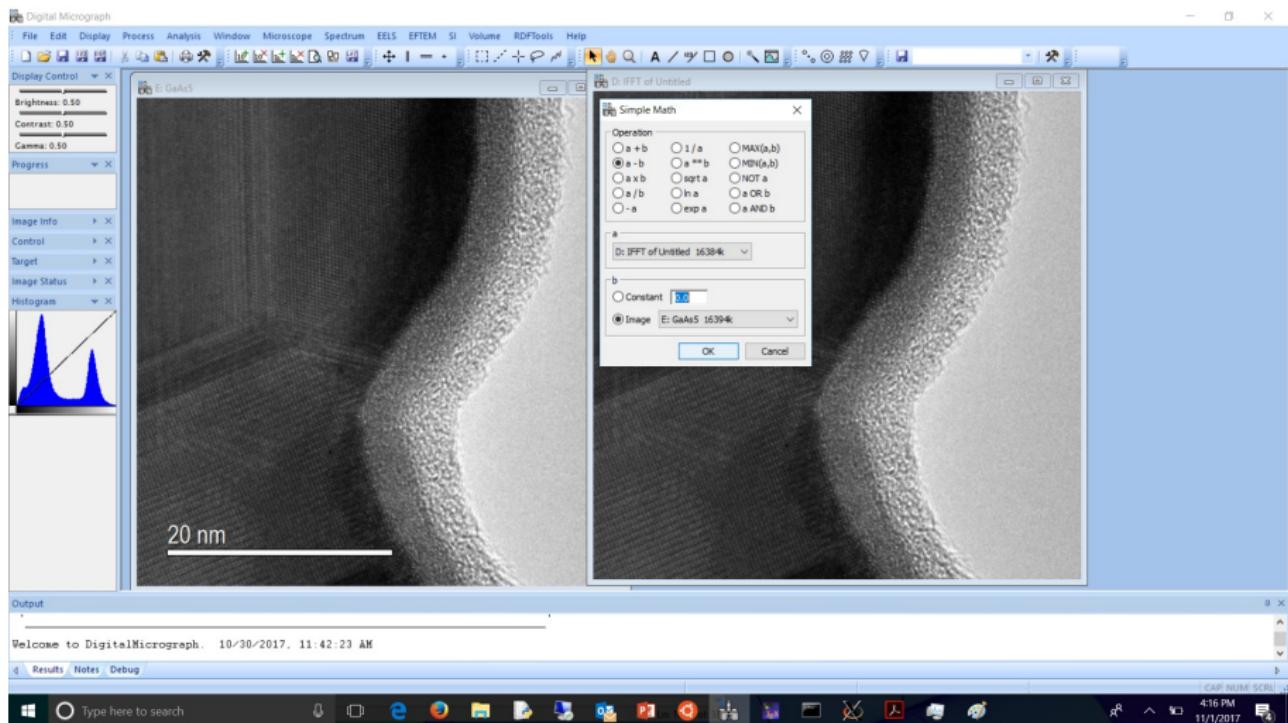
Select the “Disk Mask” in the Fourier transform; select Apply Mask and then do an inverse FFT.



What changes with size of the selected mask?

# High Pass Fourier Filter

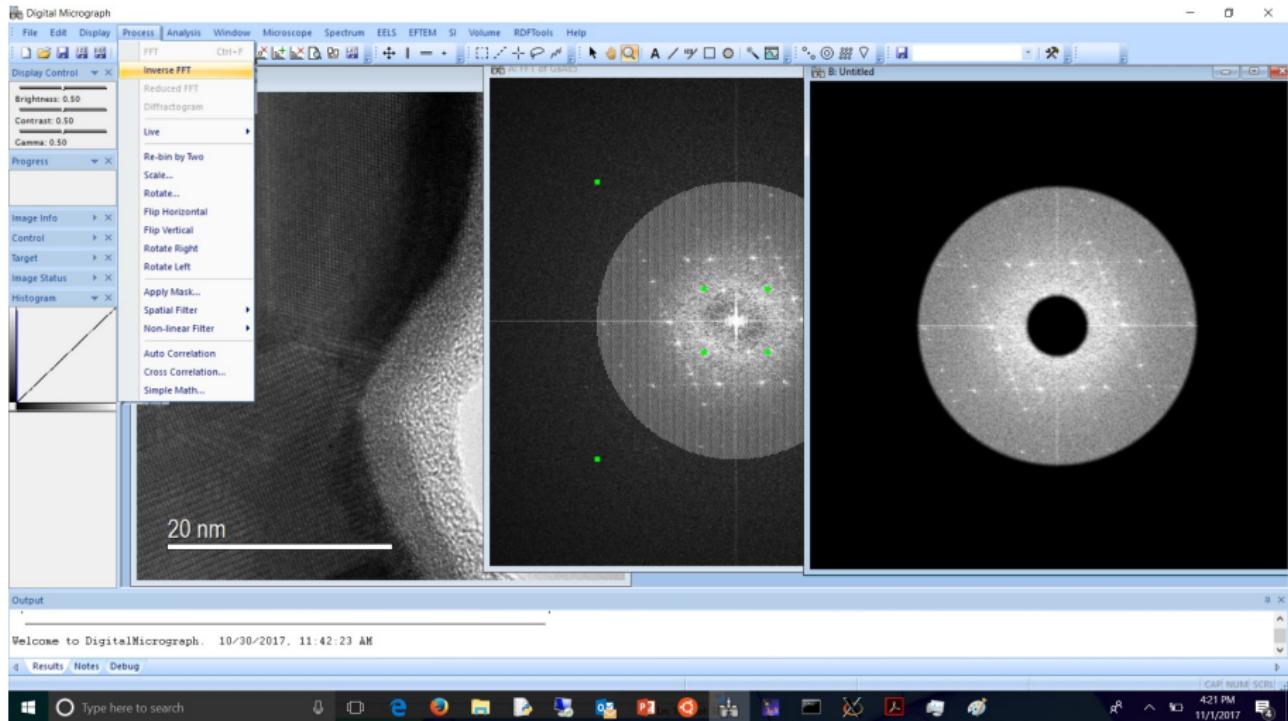
With the "Simple Math" menu we can subtract the unprocessed and processed image



What changes with size of the selected mask?

# Low Pass Fourier Filter

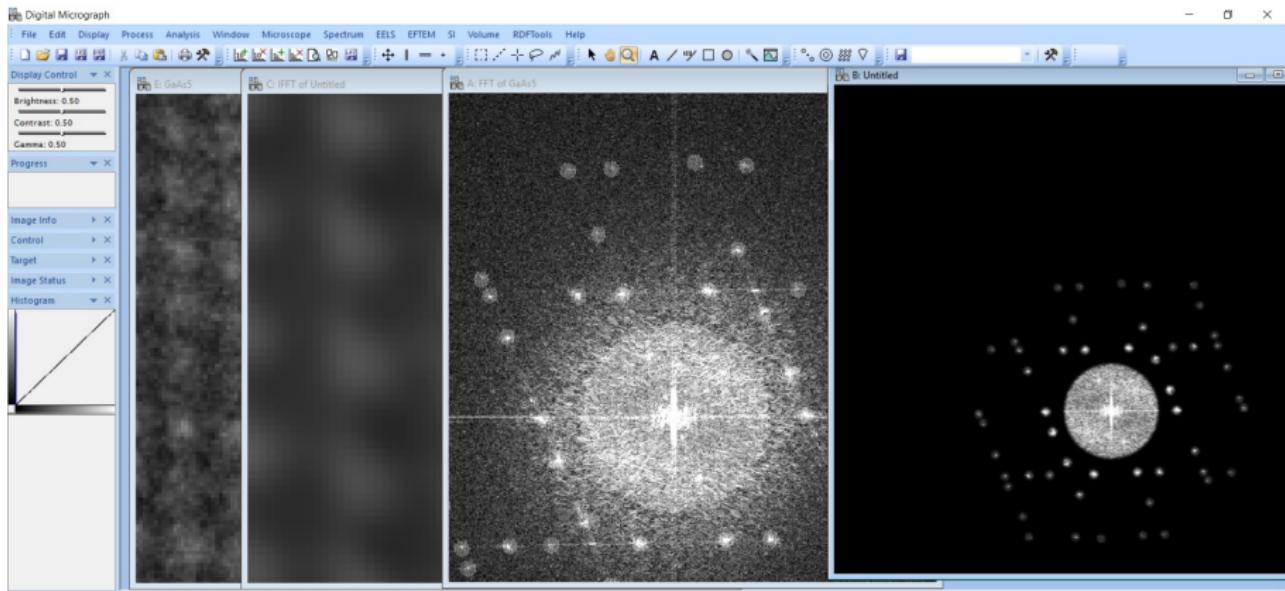
Select the “Low-High Pass Mask” in the Fourier transform; select Apply Mask and then do an inverse FFT.



What changes with size of the selected mask?

# Adaptive Fourier Filter

Select with the “disk” tool all diffraction spots in the Fourier transform; select Apply Mask and then do an inverse FFT.



Why do I select a large area close to the center?

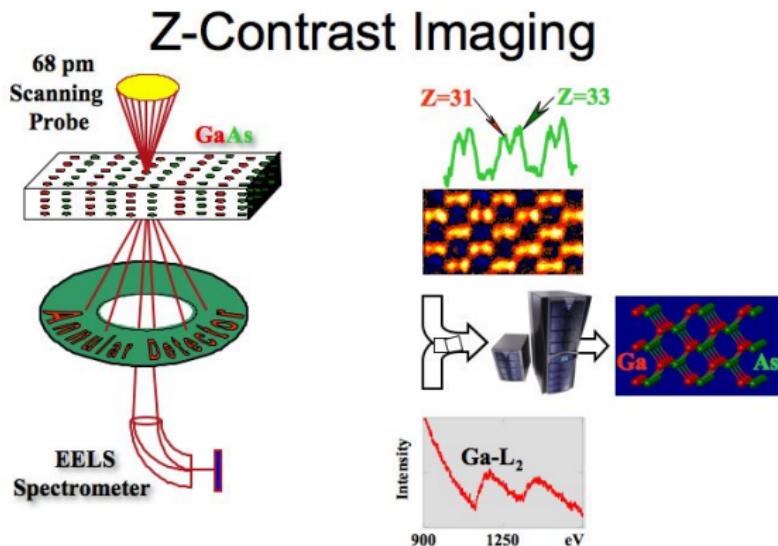
# Adaptive Fourier Filter

- ▶ zoom in a lot in the filtered image
  - ▶ how does the stacking fault look
  - ▶ how does the Moire look?
- ▶ change the size of the center area
- ▶ change the number of selected spots
- ▶ select random spots

**After each of the above processes**

How does the difference image change?

# Z-Contrast Imaging



# Z-Contrast Imaging

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We first form a small probe first, and the electrons can channel through the crystal, if the sample is in a small order zone axis. The electrons, which channel on the atomic columns, can be scattered to high angles by phonon excitation. This is an incoherent scattering event, that is why this imaging techniques is also called incoherent imaging. The electrons scattered to such high angles, that they are not longer fulfilling the Bragg condition, but Rutherford scattering condition with a probability (intensity) proportional to the square of the atomic number Z. We collect these electrons with a high angle annular dark field detector synchronized by the scanning of the beam. Please note that we only depend on channeling which means no dynamical oscillation of intensities but a higher phonon scattering probability with increasing thickness. The resolution is given by the probe diameter.

# Z-Contrast Imaging

The comparison is summed up in table

Technique	thickness dependence	Z dependence	detection
HRTEM	$I \sim \sin^2 \xi t$	$I \text{ not } \sim Z$	parallel
Z-contrast	$I \sim t$	$I \sim Z^2$	serial

Lets look into Z-contrast first.

# Z-Contrast Imaging

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What equipment do we need?

- ▶ A TEM
- ▶ A scanning unit
- ▶ A high angle annular dark field detector (HAADF)

The inner angle of the HAADF must be large compared to the angles of the Bragg reflections, for Silicon 20 mrad is enough while more than 100nm is advisable for materials with heavy elements.

# Z-Contrast Imaging

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The intensity in a Z-contrast image is given by

$$I(\vec{R}) = O(\vec{R}) * P(\vec{R})$$

A convolution (\*) between the object function ( $O(\vec{R})$ ) with the  $Z^2$  dependence and the probe profile ( $P(\vec{R})$ ).

# Z-Contrast Imaging

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Knowing the probe profile ( $P(\vec{R})$ ) will, therefore, enable us to recover the object function ( $O(\vec{R})$ ) by a simple deconvolution (division in Fourier Space) with the probe profile ( $P(\vec{R})$ ). The interpretation of Z-contrast images is, therefore, straight forward.

Now, we have to see how to get a small electron probe.

# Z-Contrast Imaging

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If we just reduce the condenser aperture we will get a smaller beam but also less and less intensity.

The smallest beam size you can reach with this method is about 1 nm.

# Z-Contrast Imaging

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The use of the coherency of your electron source can help you out here. If you have a completely coherent source, which in our case is given by a coherently filled aperture the beam will scatter at the rim and destructively interfere with the electron rays not going to the center. The result is a highly peaked function in the middle of the aperture, the Airy function. The result is a much smaller electron probe with much more intensity than with the other method. We use no or a large condenser aperture and a parallel illumination of the objective aperture. To achieve the plane illumination the condenser lenses have to be highly excited.

# Z-Contrast Imaging

The interesting thing now is that the larger the aperture the smaller your beam will be.

The limitation is given by the spherical aberrations  $C_S$ , which destroys the spatial coherence. The same is true for microscope instabilities. The overall limitation for the beam size  $d_p$  is given by

$$d_p = 0.5C_S\alpha^3$$

The optimum convergent angle  $\alpha$  for most microscopes is about 10 mrad.

# Z-Contrast Imaging

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How can we do incoherent imaging with a coherent microscope? We need the coherency for the probe formation and the channeling. In low order zone axis the electrons channel through the crystal. With a probe diameter in the atomic dimension focused on the top of the crystal the electrons are forced to channel wherever the probe is put. If the beam is located on an atomic column they channel down an atomic column. If located between atomic columns The electrons channel in these channels.

# Z-Contrast Imaging

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Using the reciprocity theorem you can look at it like there are plane waves hitting the samples from the detector side and you see the Bloch wave approach works.

# Z-Contrast Imaging

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The electrons channeling in channels have very little probability to scatter at phonons, but the electrons channeling on atomic columns have a very high probability to scatter at phonons to high angles. This scattering event is incoherent. If we collect with our ring detector only electrons scattered to high angles we get an incoherent image: the Z-contrast image. The high angle scattering process is described by the Rutherford scattering formalism and the intensity will be proportional to the square of the atomic number.

The resolution limit for incoherent imaging is according to Lord Rayleigh better than for coherent imaging by a factor  $\sqrt{2}$ .

# Lens Aberrations

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Here I give a summary of the results only. If you want to get deeper into lens errors, you have to study Fourier-optic.

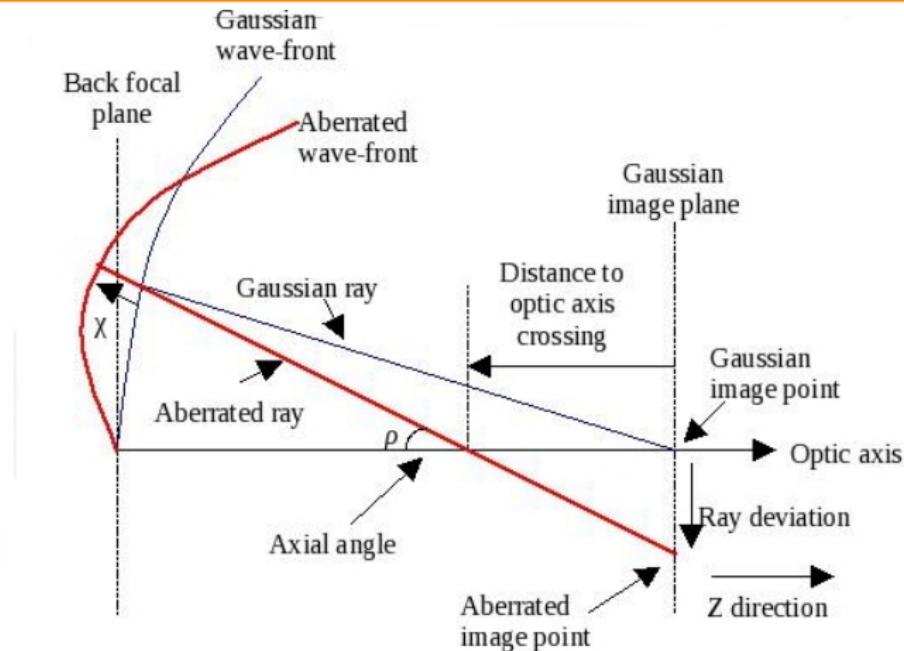
The aberrations of a transmission electron microscope are described by a scalar aberration function,  $\chi$ . This can be defined in a number of slightly different ways; the form used here is similar to that described by Krivanek et al (1999b) and was given in A. Lupini's PhD thesis.

# Lens Aberrations

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It is possible to consider the aberration function either in terms of the wave aberration or the ray deviation. Both views are related because the rays propagate normally to the wave-fronts. We define the aberration function as the distance between the aberrated wave-front and the un-aberrated (or "Gaussian") wave-front along the aberrated ray, as illustrated in the next figure. The wave aberration, which represents the phase change across the back focal plane of the objective lens, is thus given by  $2\pi\chi/\lambda$ . The ray deviation is defined as the vector from the Gaussian image point to the intersection of the aberrated ray and the image plane.

# Lens Aberrations



Effect of aberrations on rays in an aberrated system. The aberration function gives the distance from the un-aberrated Gaussian wave-front to the aberrated wave-surface as a function of angle to the optic axis.

# Lens Aberrations

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In a more general optical system, a ray is defined by its intersection with two planes, as described by Hawkes and Kasper (1994). However, in STEM, we are primarily concerned only with axial aberrations, where the Gaussian image points (those points which un-aberrated rays are imaged to) are always on-axis. In this case, it is possible to assume that the aberration function is a function only of the angle that a ray makes to the optic axis. This simplifies the following discussion and is sufficient for consideration of STEM instruments. Off-axial aberrations cannot be neglected in CTEM, but the additional terms make the aberration even more problematic.

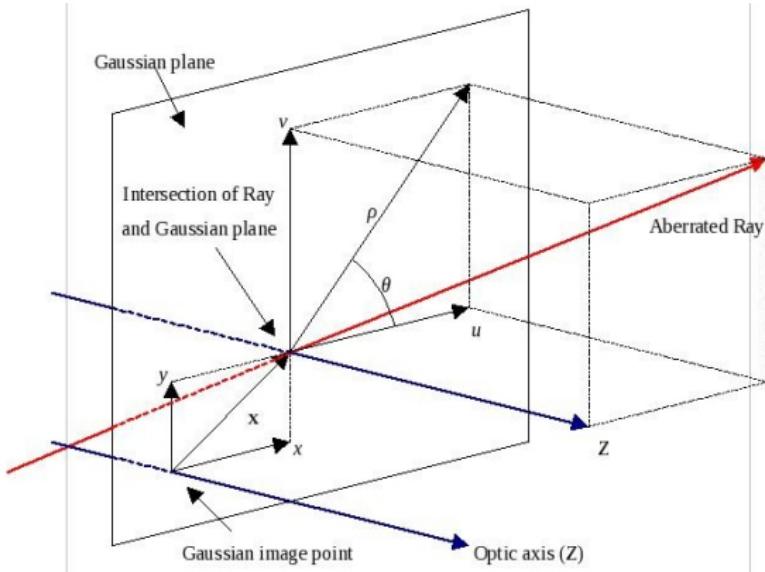
# Lens Aberrations

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The coordinates used to define the aberration function are shown in figures above and below.

The angles to the optic axis, the axial angle,  $\rho$ , and the azimuthal angle,  $\theta$ , are defined at the sample plane. These are extrapolated so that  $\rho$  and  $\theta$  also give the coordinates in the back focal plane. Strictly, this extrapolation gives a small error because these two definitions are not quite the same, however the error in this assumption is small for a large demagnification. In a magnetic lens, the trajectories will also be spiraling continuously. The optic axis is always defined as the Z-direction.

# Lens Aberrations



The coordinates used to define the aberration function.  $(\phi, \theta)$  and  $(u, v)$  are defined so that they give the slopes of a ray to the optic axis at the sample. It is assumed that demagnification is sufficient so these will also give the coordinates in the back focal plane, which implies that  $X$  is small.

The intersection of the ray with the image plane is given by the vector  $X = (x, y)$ . The optic axis defines the Z-direction.

# Lens Aberrations

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Using the notation of Krivanek et al (1999b), the aberration coefficients will be denoted by a capital C followed by up to two numbers and then a letter in subscript:  $C_{NSA}$ . The first number, N indicates the order of the ray aberration, which is referred to as “the order”. The second number, S indicates the symmetry of the aberration, and is the number of times that the function repeats on being rotated about the optic axis. The final letter indicates the orientation, which is specified as A or B type. These represent the cosine and sine terms respectively in the expansion below.

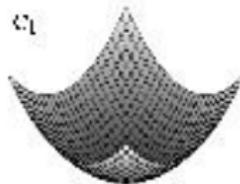
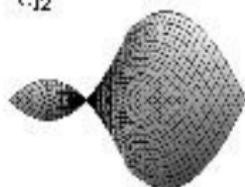
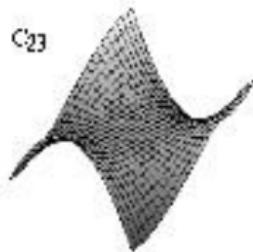
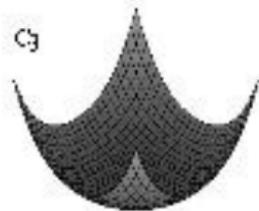
# Lens Aberrations

There is a whole variety of lens aberrations:

Notation	Order of ray aberr.	Order of wave aberr.	Name	Symmetry
$C_{01A}$	0	1	Probe shift in x direction	1-fold
$C_{01B}$	0	1	Probe shift in y direction	1-fold
$C_1$	1	2	Defocus	Rotational
$C_{12}$	1	2	Astigmatism	2-fold
$C_{21}$	2	3	Coma	1-fold
$C_{23}$	2	3	3-fold astigmatism	3-fold
$C_3$	3	4	Spherical aberration ( $C_S$ )	Rotational
$C_{32}$	3	4	2-fold astig of $C_S$	3-fold
$C_{34}$	3	4	4-fold astig of $C_S$	3-fold

**Table:** List of Aberrations

# Lens Aberrations

 $C_{01}$  $C_1$  $C_{21}$  $C_{21}$  $C_{23}$  $C_3$  $C_{34}$  $C_{32}$ 

# Lens Aberrations

Most of them we can organize in the aberration function  $\chi(\rho\phi)$ , where we express the different aberrations in the notation of Krivanek et al.

$$\frac{\rho^{N+1}}{N+1} [C_{NSA} \cos(S\phi) + C_{NSB} \sin(S\phi)] \quad (14)$$

We use spherical coordinates. In this expression  $\rho$  (or *theta* in the figure) is the (collateral or zenith) axial angle, and  $\phi$  the azimuthal angle. The axial angle is closely related with the distance from the center  $r$  in cylindriacal coordinates. Where  $N$  is the order of the aberration,  $S$  is the symmetry (2-fold, 3-fold,...),  $A$  and  $B$  is a direction.

# Lens Aberrations

With the above notation the aberration function is expressed as:

$$\begin{aligned}\chi(\rho, \phi) = & \{\rho [C_{01A} \cos(\phi) + C_{01B} \sin(\phi)] \\& + \frac{\rho^2}{2} [C_1 + C_{12A} \cos(2\phi) + C_{12B} \sin(2\phi)] + \\& + \frac{\rho^3}{3} [C_{21A} \cos(\phi) + C_{21B} \sin(\phi) + C_{23A} \cos(3\phi) + C_{23B} \sin(3\phi)] + \\& + \frac{\rho^4}{4} [C_3 + C_{32A} \cos(2\phi) + C_{32B} \sin(2\phi) + C_{34A} \cos(4\phi) + C_{34B} \sin(4\phi)] + \\& + \dots\}\end{aligned}$$

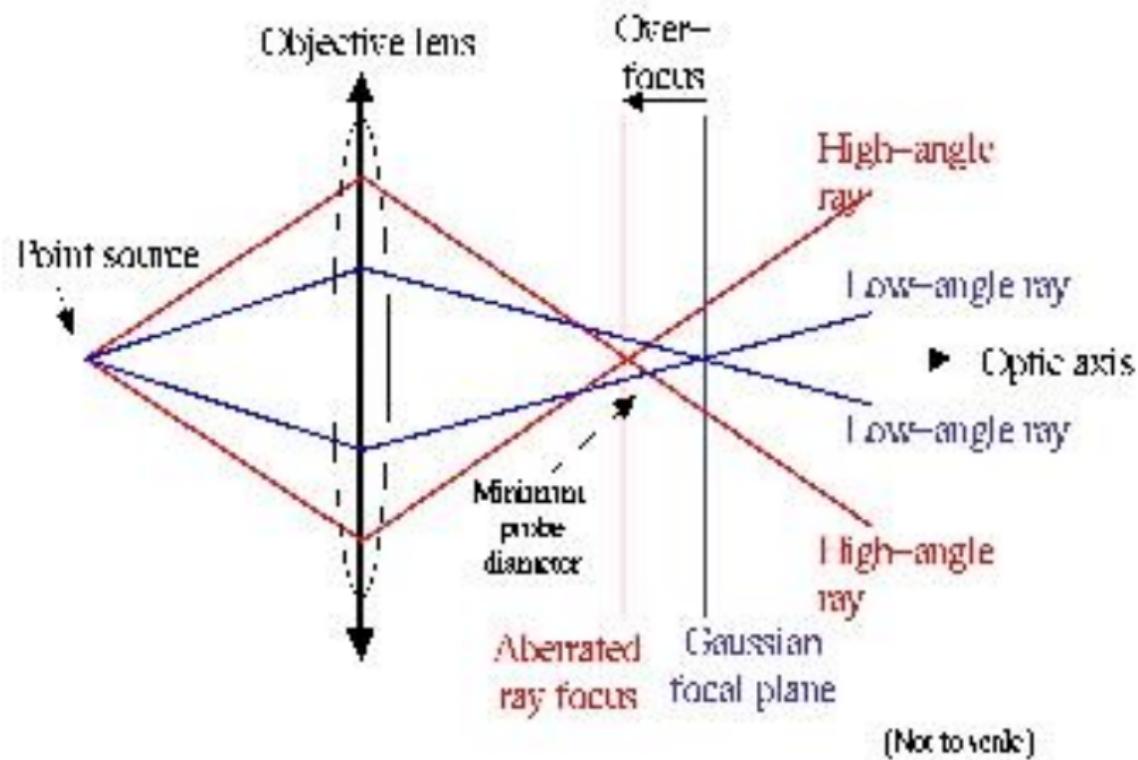
# Spherical Aberration

The focal length is reduced for the electron rays passing through the outer zones of the lens. Electrons crossing through the optic axis with an angle  $\theta$  or a scattered in the specimen under the angle  $\theta$  will intersect the Gaussian image plane at a distance

$$r'_S = C_S \theta^3 M \quad (16)$$

from the paraxial image point. The Gaussian image plane is the position of the image when very small apertures are used (paraxial rays).  $C_S$  is the spherical aberration coefficient and  $M$  is the magnification. The wave aberration term associated with spherical aberration is  $C_S \rho^4 / 4$ . Thus the ray deviation from the Gaussian image point is  $C_S \rho^3$ . Positive spherical aberration will cause higher angle rays to cross the axis before the Gaussian image plane.

# Spherical Aberration

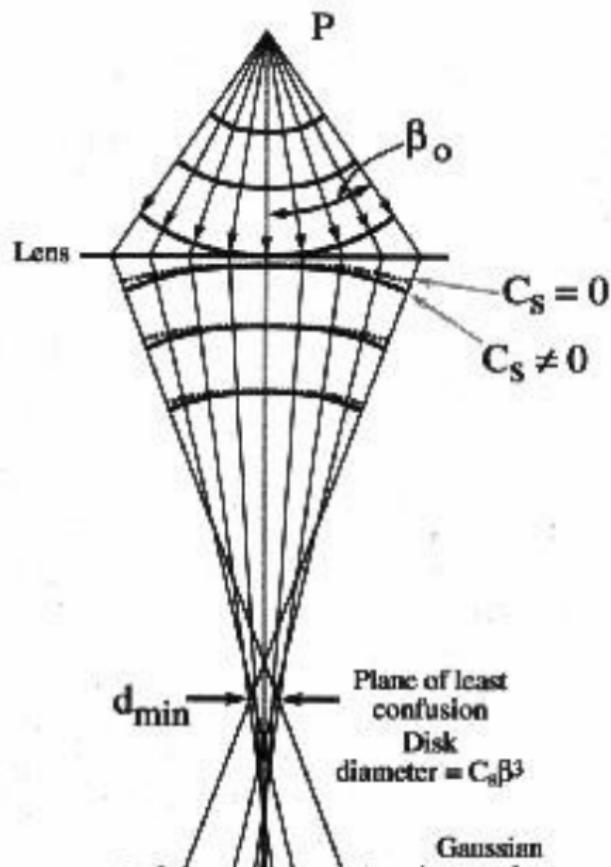


# Spherical Aberration

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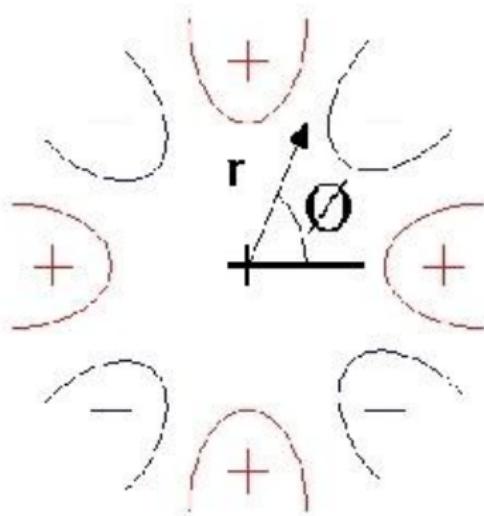
A conical electron beam with angular convergence angle  $\alpha_0$  defined by the object aperture does not produce a sharp image point but the beam passes through a minimum  $d_{S,min}$ , in a plane of least confusion. In the Gaussian image plane (corresponds to the image plane if the plane of least confusion is the focal plane), the diameter is  $d'_{S,G} = 2C_S\alpha_0^3 M$ . The smallest diameter  $d_{S,min}$  can be estimated from  $d_{S,min} = 0.5C_S\alpha_0^3$ . All this originates from the wavefronts which are not longer perfectly spherical, that's where the name comes from.

# Spherical Aberration



# Correction of Spherical Aberration

There are now two different designs of aberration correctors, which improve the resolution of a TEM. Both designs are based on multipole lenses. The figure shows the field of an octupole; its radial distribution goes as  $r^3$ . This kind of fields are necessary to correct for spherical aberration.

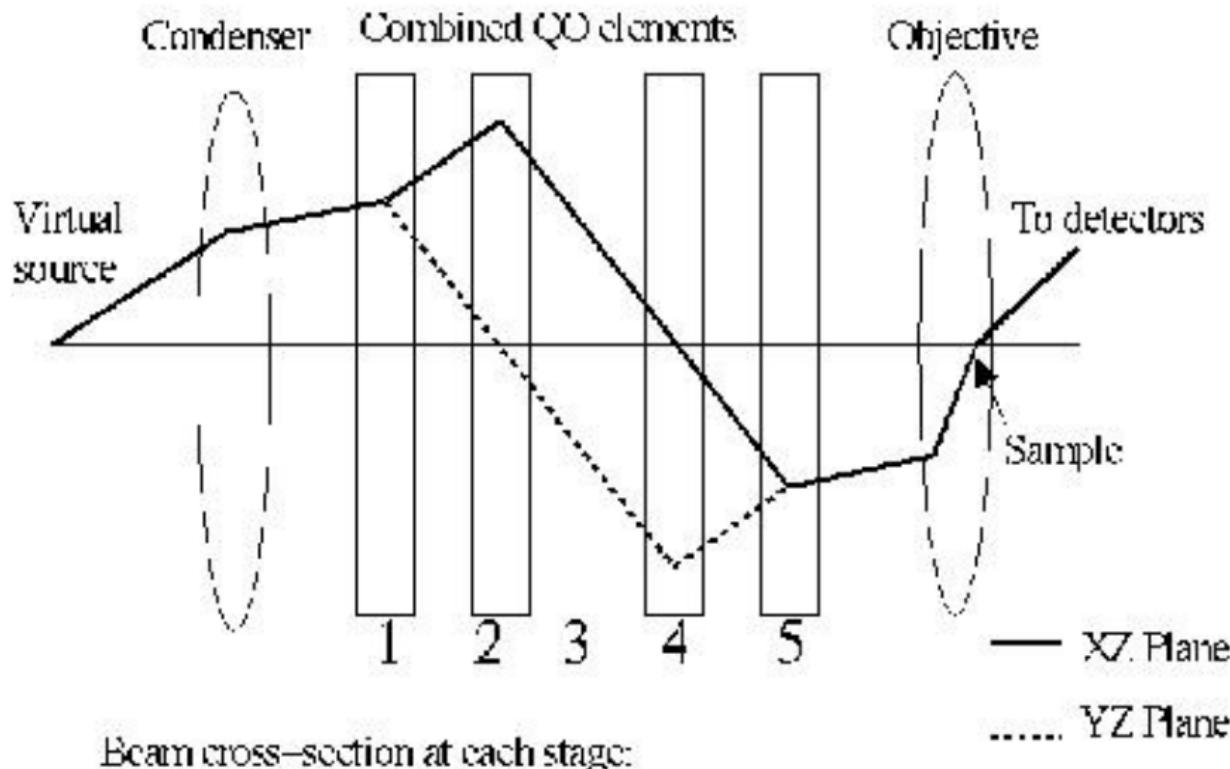


Ideal (infinite)  
octupole has field:

$$B_r = B_0 r^3 \cos(4\phi)$$

# Correction of Spherical Aberration

As for the correction of astigmatism, we use a whole set of lenses.



# Correction of Spherical Aberration

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That many electrical lenses (here we have about 84 degrees of freedom) need a sophisticated software for a computer based alignment. It can no longer be done manually. Only with new alignment algorithms and modern electronics was it possible to succeed.

So far we only talked about the advantages of Z-contrast imaging, but the biggest disadvantage we did not point out properly: The serial detection of the images results in wavy pictures.

The sample drift which is always present will prevent the unambiguous detection of the atom position in most cases. The parallel detection in HRTEM is far superior in the atomic site detection, if you know what you are looking at. Please, see this techniques not as competing, but as complimentary.

Of course we can fix that, watch lecture 6