TEM and STEM Image Simulation

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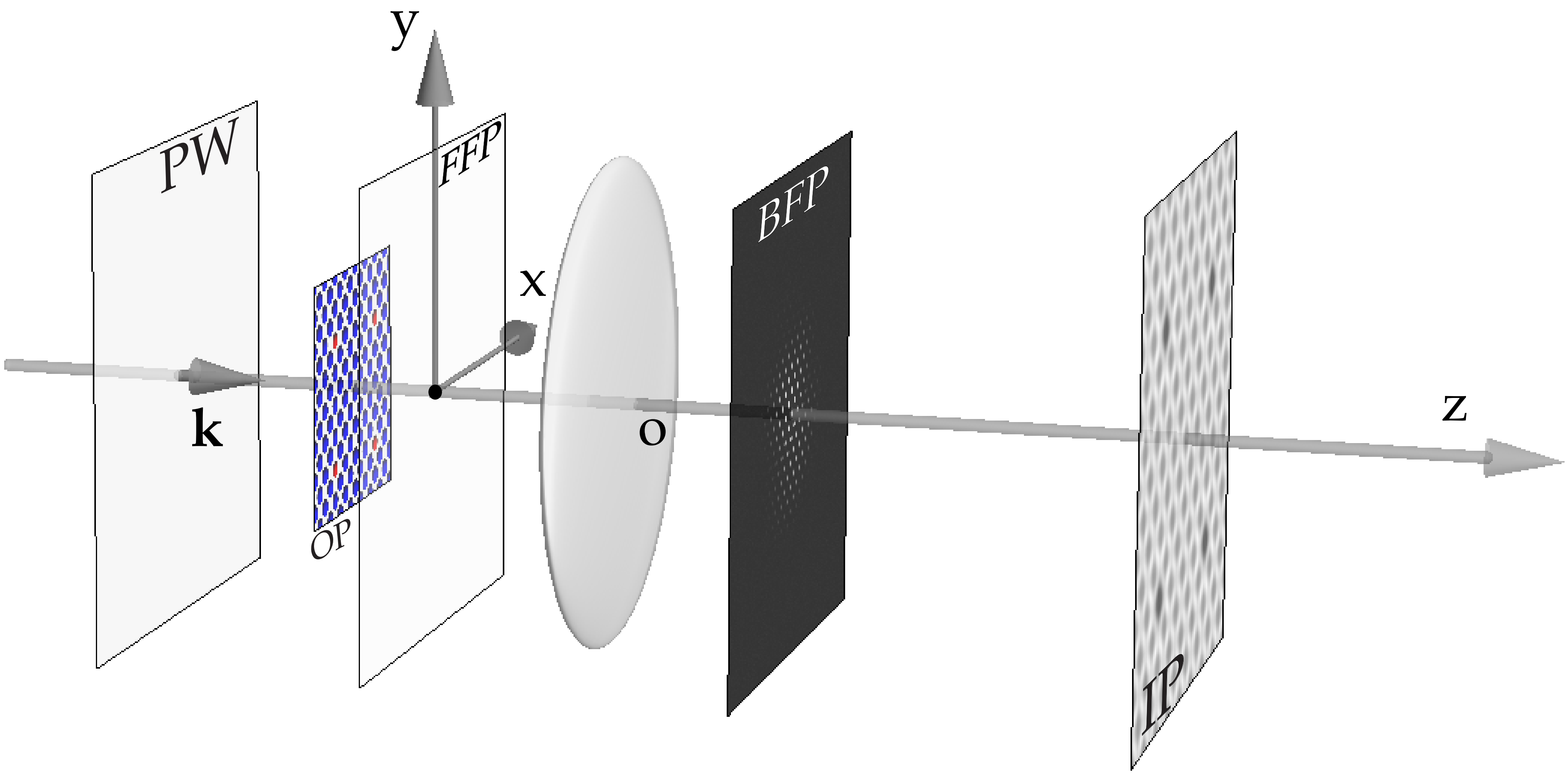
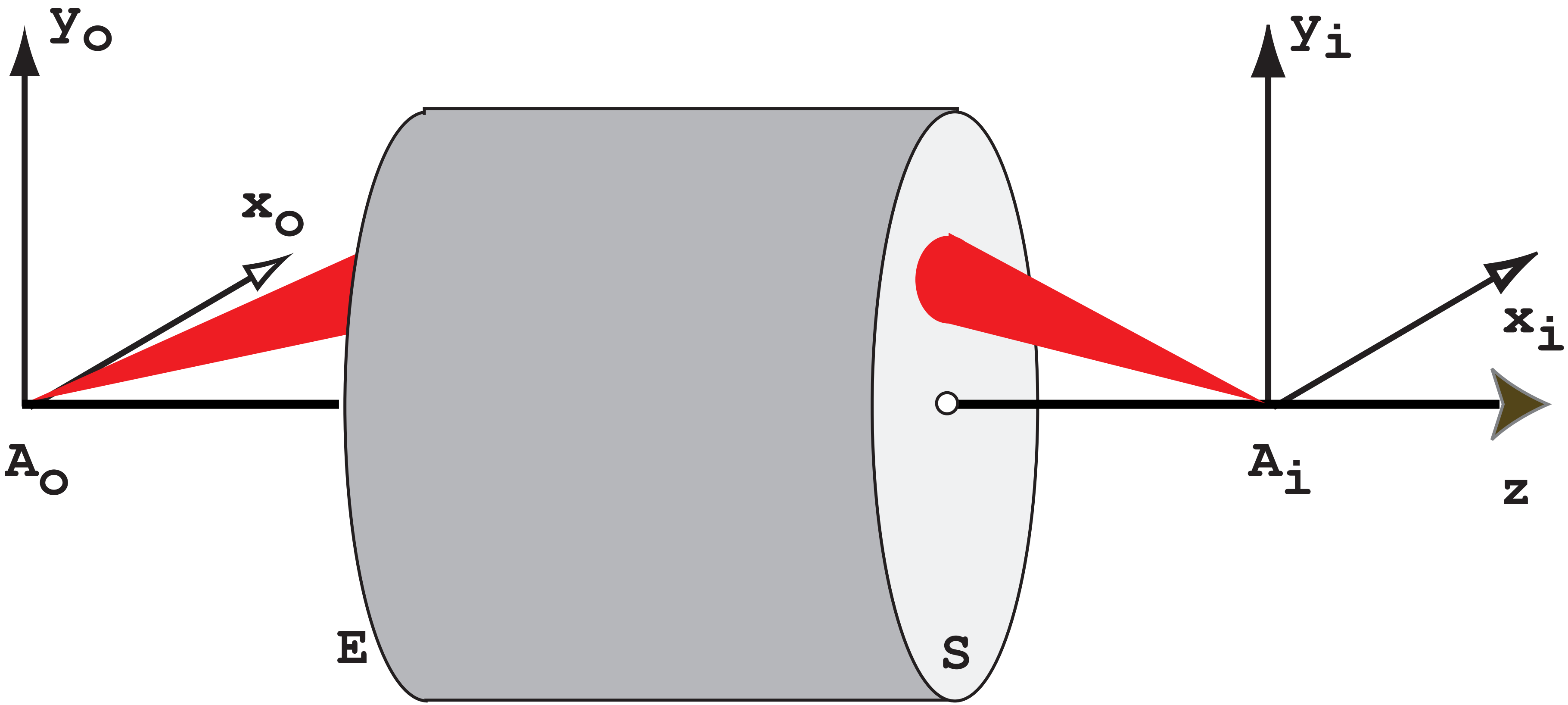
TEM: reminder



TEM modelling steps: incident wave (PW), crystal (OP), electron-matter interaction, Fraunhofer approximation, image formation (Abbe theory), ...

- **Optical system**.
- Aberrations.
- TEM transfer function & STEM: optical transfer function.
- Comparison HRTEM - HRSTEM.

Optical system



An optical system produces the image A_i of a point source object A_o. A_o and A_i are said to be conjugate. A_i is not a point since any optical system is diffraction limited. This limitation is introduced by the entrance and exit pupils of the optical system.

Transfer function

Optical system as a black box that gives an image wave-function $\Psi_i(\vec{x})$ of an object wave-function $\Psi_o(\vec{x})$:

$$\Psi_i(\vec{x}) = S\{\Psi_o(\vec{x})\}$$

- Linearity.
- Space invariance.

Linearity

$$\begin{aligned} S\{a_1\Psi_o^1(\vec{x}) + a_2\Psi_o^2(\vec{x})\} &= a_1 S\{\Psi_o^1(\vec{x})\} + a_2 S\{\Psi_o^2(\vec{x})\} \\ S\{a_1\Psi_o^1(\vec{x}) + a_2\Psi_o^2(\vec{x})\} &= a_1\Psi_i^1(\vec{x}) + a_2\Psi_i^2(\vec{x}) \end{aligned}$$

Linearity allows to decompose the object wave-function in a infinite sum of point sources:

$$\Psi_o(\vec{x}) = \int_{-\infty}^{\infty} \Psi_o(\vec{u})\delta(\vec{x}-\vec{u})\mathrm{d}\vec{u}$$

The image wave-function is then:

$$\Psi_i(\vec{x}) = S\left\{\int_{-\infty}^{\infty} \Psi_o(\vec{u})\delta(\vec{x}-\vec{u})\mathrm{d}\vec{u}\right\} = \int_{-\infty}^{\infty} \Psi_o(\vec{u}) S\{\delta(\vec{x}-\vec{u})\}\mathrm{d}\vec{u} = \int_{-\infty}^{\infty} \Psi_o(\vec{u}) T(\vec{x};\vec{u})\mathrm{d}\vec{u}$$

where $T(\vec{x};\vec{u})$ is ***Impulse response*** of the optical system.

Space invariance

Space invariance is realised when the image of a point source is independent of its position in the object plane, i.e. when the point source moves in the object plane its image moves similarly in the image plane without changing form and intensity.

$$T(\vec{x};\vec{u}) = T(\vec{x}-\vec{u})$$

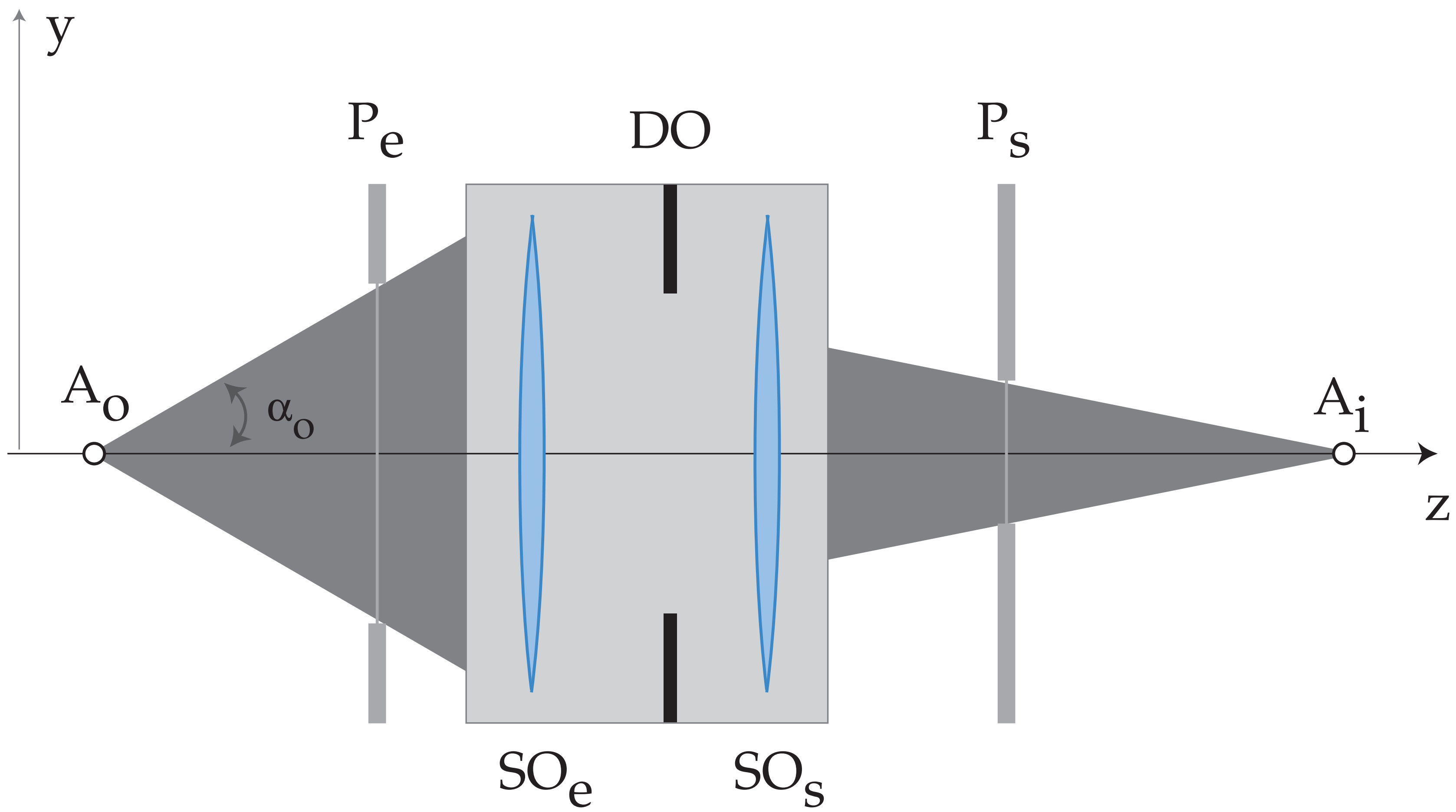
$$\Psi_i(\vec{x}) = \int_{-\infty}^{\infty} \Psi_o(\vec{u})\, T(\vec{x}-\vec{u})\mathrm{d}\vec{u} = \Psi_o(\vec{x}) \otimes T(\vec{x})$$

The convolution integral spreads the object information and degrades the performences of the optical system.
Taking its Fourier transform:

$$\widetilde{\Psi}_i(\vec{h}) = \widetilde{\Psi}_o(\vec{h})\, \widetilde{T}(\vec{h})$$

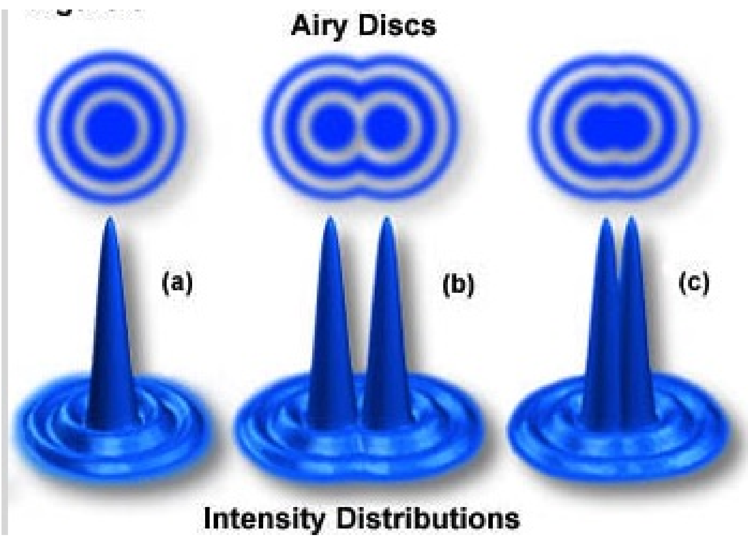
$\widetilde{T}(\vec{h}$ is the transfer function of the optical system.

Pupils

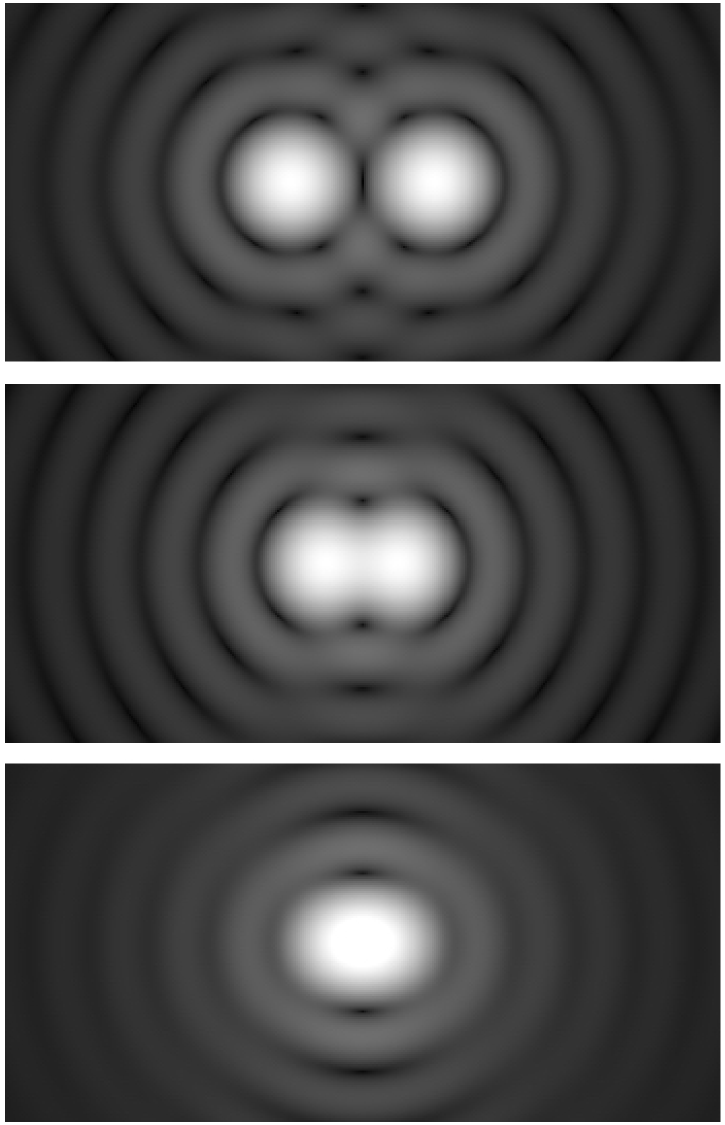


Any optical system can be characterised by an entrance pupil P_e and an exit pupil P_s. The pupils are the image of the opening aperture DO by the entrance and exit optical subsystems SO_e and SO_s. The portion of the object wave-function accepted by the optical system is limited by the P_e, while SO_s limits the extend of the image wave-function. For a perfect optical system, the image of a point source will be an Airy disk.

Airy disks and Rayleigh resolution criterion



Airy disks.



Airy disks near Rayleigh resolution criterion.

- Optical system.
- **Aberrations**.
- HRTEM transfer function & HRSTEM: optical transfer function.
- Comparison HRTEM - HRSTEM.

Some light rays emitted by object point A_o do not reach the image at point A_i.

Position of $A_i \longrightarrow$ intersection of the reference light ray (non deviated) and the image plane.

The image of a point source is a **spot** whose shape and intensity depend of the quality of the optical system.

Two types of aberrations:

1. Monochromatic.
2. Chromatic (λ dependent).

Monochromatic aberrations

In order to evaluate the monochromatic aberrations one must define a function characteristic of the optical system.

This function will depend on:

1. The selected reference planes.
2. The optical path followed by the light ray.

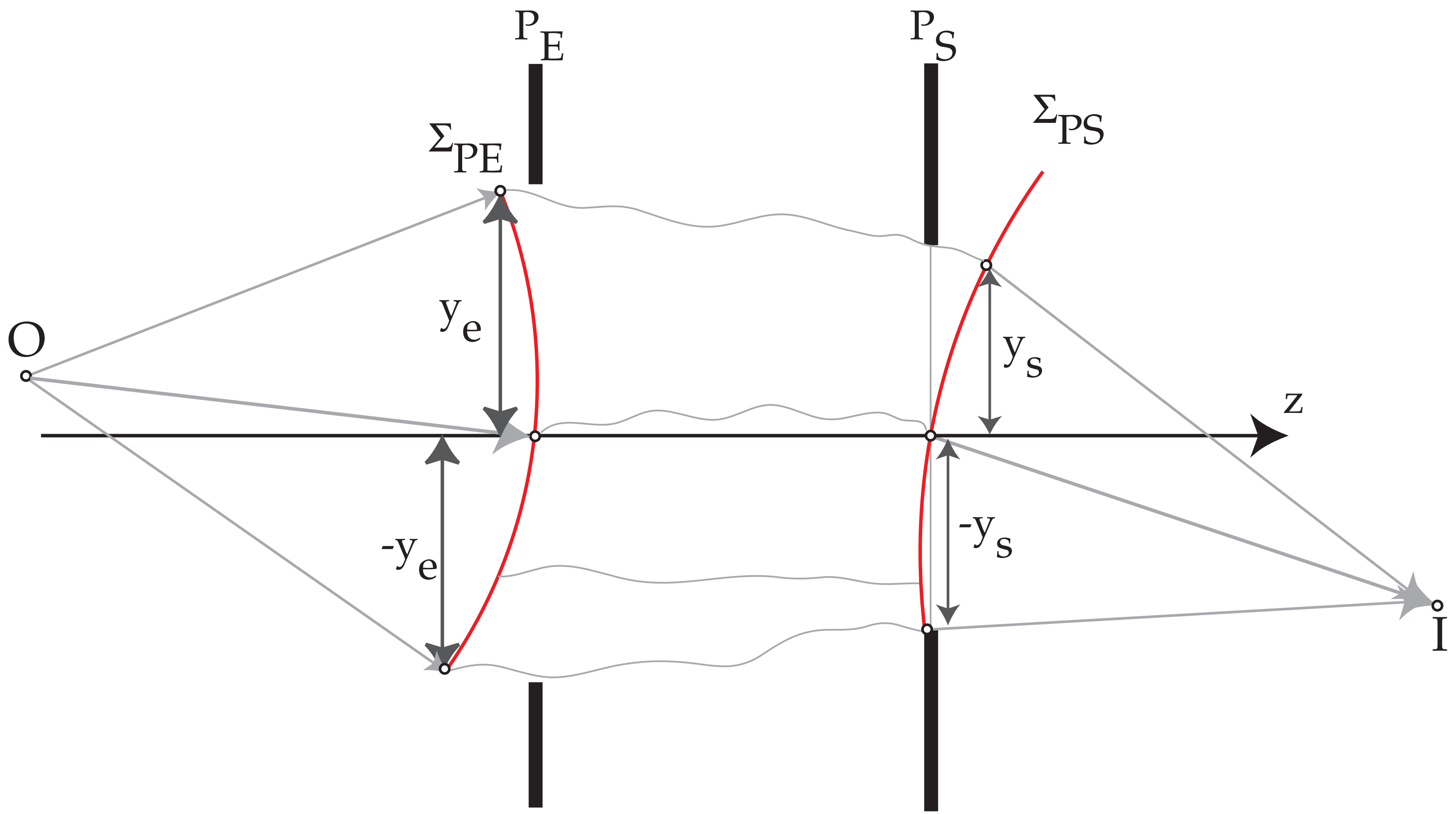
Optical path: length

The important feature is the optical path length (OPL).

$$OPL(P_1P_2) = \int_{P_1}^{P_2} n(\overrightarrow{r})\, ds$$

1. OPL is measured in meters ($n(\vec{r}) = \frac{c}{v(\vec{r})}$ has no unit).
2. OPL is proportional to the time spent by the light ray to travel from P_1 to P_2.
3. Surface of constant OPL $\longrightarrow$ wavefront (surface of constant travel time).
4. OPL is measured from the entrance pupil P_E to the exit pupil P_S.

Optical Path Length: OPL

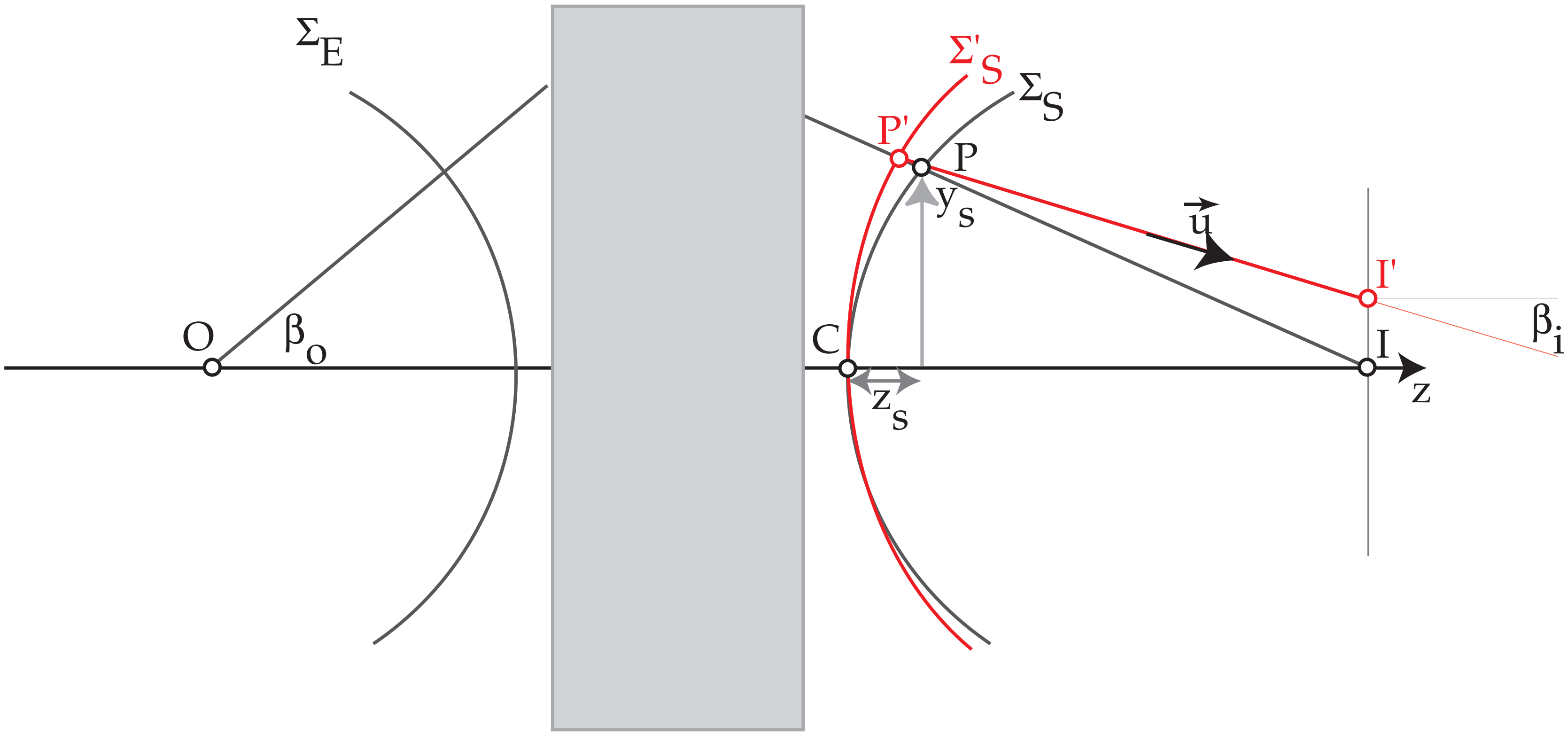


- Before P_E the reference wavefront Σ_{PE} is spherical (point source at O).
- After P_S the reference wavefront Σ_{PS} is spherical (converges towards I).

For a perfect optical system, both the entrance Σ_{PE} and exit Σ_{PS} wavefronts are spherical. The Optical Path Length form O to I is independent of the path.

Difference of OPL: OPD

The OPD measure the deviation of a wavefront from a perfect spherical wavefront (vacuum or homogenous medium).
At the exit pupil P_S, the spherical wavefront converging towards **I** defines the reference wavefront.



In the presence of aberrations the wavefront Σ_S' is no more spherical. The Optical Path Dinference (distance between the deformed Σ_S' and spherical waveform Σ_S) introduced the phase shift:

$$\delta\phi = e^{2\pi\imath\frac{OPD(x_S,y_S)}{\lambda}}$$

Transverse geometric aberrations: $\vec{\epsilon}$

The transverse geometric aberrations are proportional to $\frac{d}{d\theta}$ wavefront aberrations[1]:

$$\epsilon_x = -\frac{f}{n_i}\frac{\partial W}{\partial x_s}$$
$$\epsilon_y = -\frac{f}{n_i}\frac{\partial W}{\partial y_s}$$

f focal length.

The OPD's introduced by all the aberrations of the imaging system are collected in a function $\chi(\vec{u})$ and the phase shift is[2]:

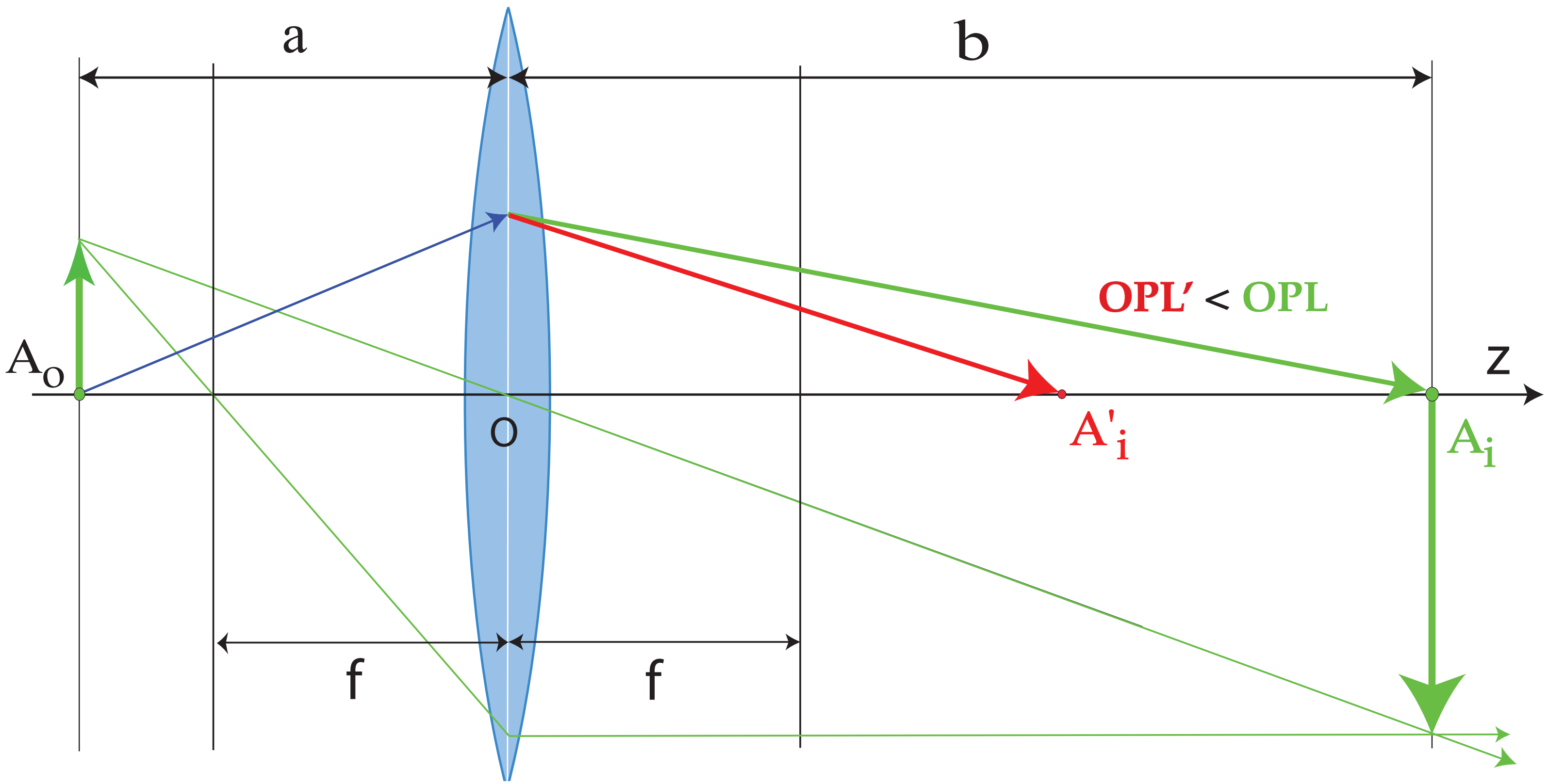
$$\widetilde{T}(\vec{u}) = e^{-\imath\chi(\vec{u})}$$

$\widetilde{T}(\vec{u})$ has been first employed by Abbe in his description of image formation (1866).

[1] $P(x_s, y_s)$ on the spherical reference wavefront can be characterised by the radial angle θ.

[2] The angle θ corresponds (through Bragg law) to a spatial frequency $\vec{u}$, i.e. a distance in the back focal plane.

OPD: spherical aberration

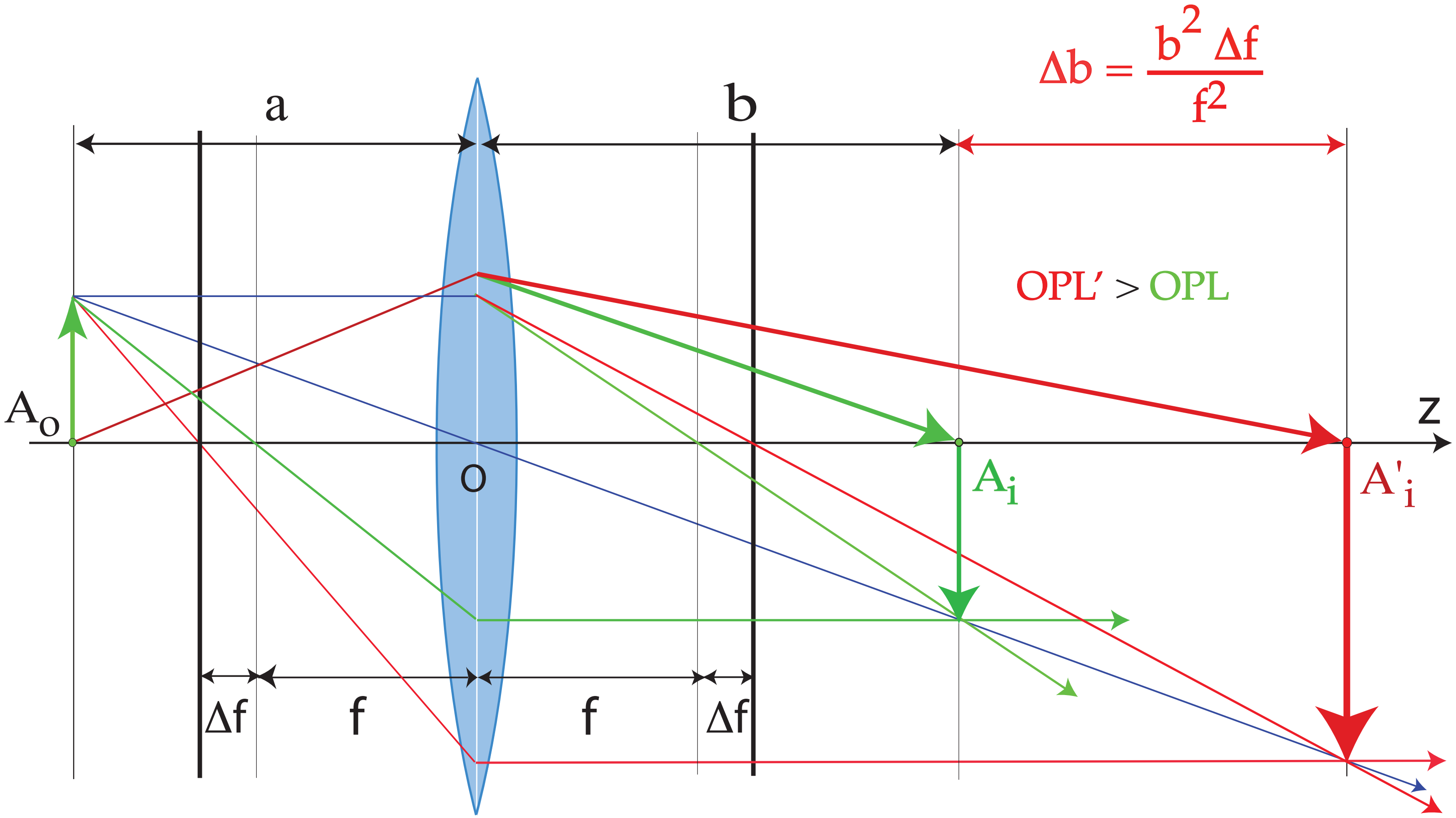


In presence of spherical aberration, the optical path length (OPL') form A_o to A_i' is smaller than OPL from A_o to A_i. The wavefront at A_i' is out-of-phase by[3]:

$$e^{-2\pi i \frac{C_s \lambda^3 (\vec{q}.\vec{q})^2}{4}}$$

[3]With our plane wave choice $e^{2\pi i \vec{q}.\vec{r}}$.

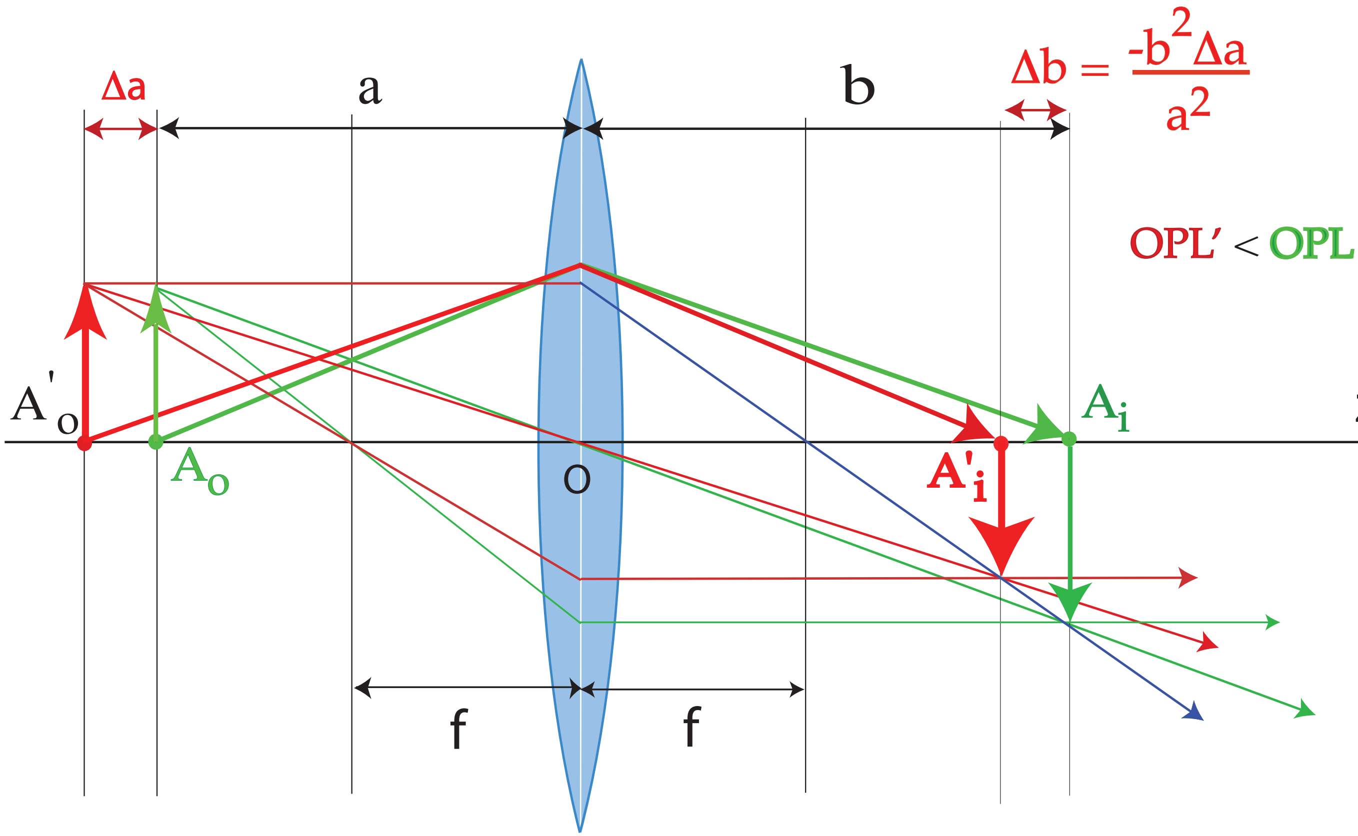
OPD: underfocus



Underfocus weakens the objective lens, i.e. increases **f**. As a consequence the OPL from A_o to A_i' is larger:

$$e^{2\pi i \frac{\Delta f \lambda (\vec{q} \cdot \vec{q})}{2}}$$

OPD: eccentricity



On the contrary keeping **f** constant and moving the object by Δa decreases the OPL.

Transfer function $\widetilde{T}(\vec{q})$

$$\widetilde{T}(\vec{q}) = e^{-\chi(\vec{q})} = \cos(\chi(\vec{q})) - i \underbrace{\sin(\chi(\vec{q}))}_{\text{CTF}}$$

$$\chi(\vec{q}) = \pi \left[-W_{20}\,\lambda\,(\vec{q}.\vec{q}) + W_{40}\frac{\lambda^3\,(\vec{q}.\vec{q})^2}{2} + \ldots \right]$$

Where:

- W_{20} : defocus (z)
- W_{40}: spherical aberration (C_s)

At present TEM and STEM aberration correctors only correct axial aberrations, i.e. aberrations that affect images of point sources located on the optical axis.

Wavefront aberrations to 6^{th} order (cartesian coordinates)

$\{z, \pi (u^2+v^2) \lambda\}$ *(defocus)*

$\{W(1,1), 2\pi(u\cos(\phi(1,1)) + v\sin(\phi(1,1)))\}$

$\{W(2,2), \pi\lambda((u-v)(u+v)\cos(2\phi(2,2)) + 2uv\sin(2\phi(2,2)))\}$

$\{W(3,1), \frac{2}{3}\pi (u^2+v^2) \lambda^2(u\cos(\phi(3,1)) + v\sin(\phi(3,1)))\}$

$\{W(3,3), \frac{2}{3}\pi\lambda^2 (u(u^2-3v^2)\cos(3\phi(3,3)) - v(v^2-3u^2)\sin(3\phi(3,3)))\}$

$\left\{W(4,0), \frac{1}{2}\pi (u^2+v^2)^2 \lambda^3\right\}$ (3^{rd} *order spherical aberration or* C_3)

$\{W(4,2), \frac{1}{2}\pi (u^2+v^2) \lambda^3((u-v)(u+v)\cos(2\phi(4,2)) + 2uv\sin(2\phi(4,2)))\}$

$\{W(4,4), \frac{1}{2}\pi\lambda^3 ((u^4-6v^2u^2+v^4)\cos(4\phi(4,4)) + 4u(u-v)v(u+v)\sin(4\phi(4,4)))\}$

$\left\{W(5,1), \frac{2}{5}\pi (u^2+v^2)^2 \lambda^4(u\cos(\phi(5,1)) + v\sin(\phi(5,1)))\right\}$

$\{W(5,3), \frac{2}{5}\pi (u^2+v^2) \lambda^4 (u(u^2-3v^2)\cos(3\phi(5,3)) - v(v^2-3u^2)\sin(3\phi(5,3)))\}$

$\{W(5,5), \frac{2}{5}\pi\lambda^4 (u(u^4-10v^2u^2+5v^4)\cos(5\phi(5,5)) + v(5u^4-10v^2u^2+v^4)\sin(5\phi(5,5)))\}$

$\left\{W(6,0), \frac{1}{3}\pi (u^2+v^2)^3 \lambda^5\right\}$ (5^{th} *order spherical aberration or* C_5)

$\left\{W(6,2), \frac{1}{3}\pi (u^2+v^2)^2 \lambda^5((u-v)(u+v)\cos(2\phi(6,2)) + 2uv\sin(2\phi(6,2)))\right\}$

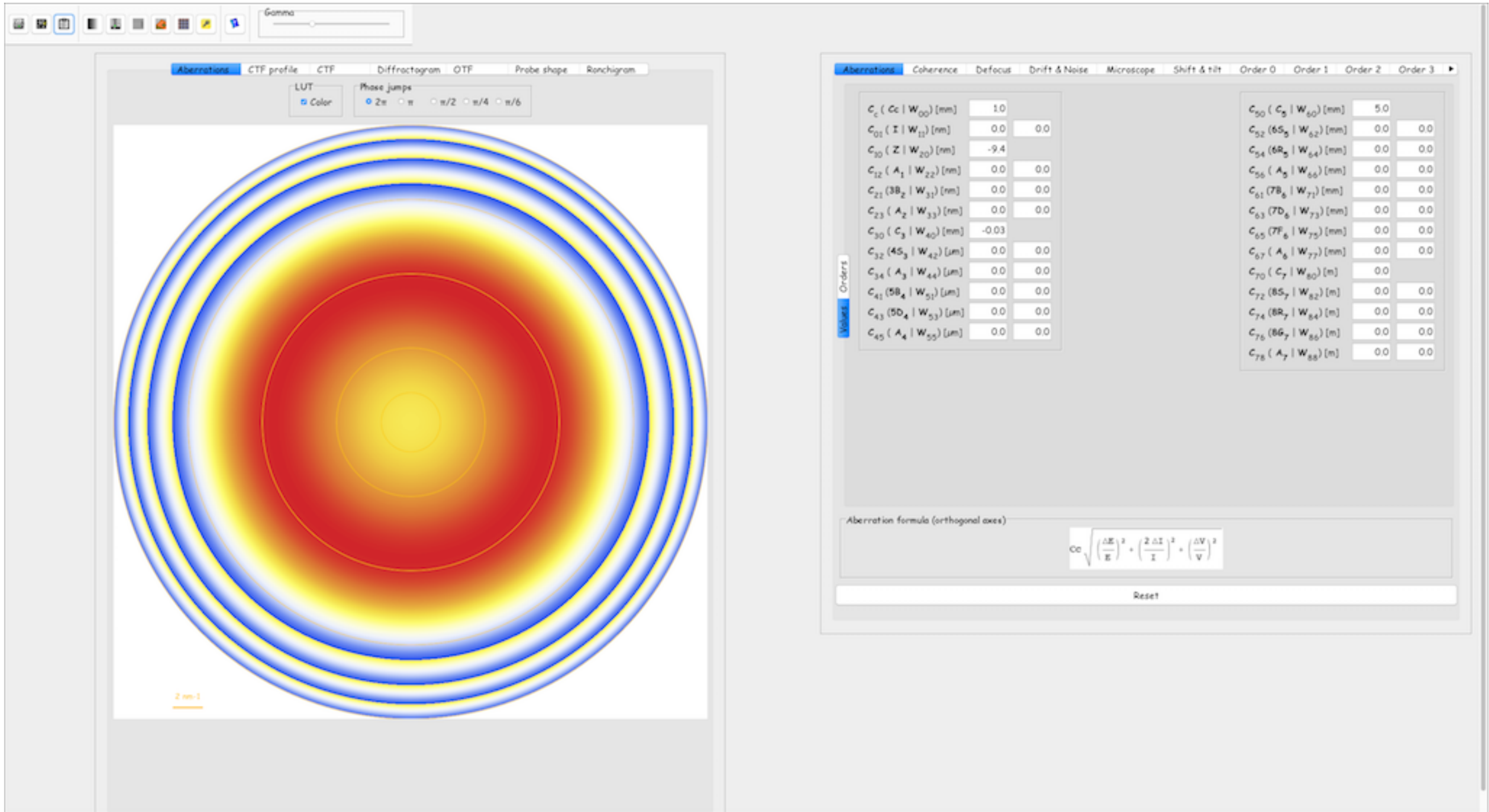
$\{W(6,4), \frac{1}{3}\pi\lambda^5 ((u^6-5v^2u^4-5v^4u^2+v^6)\cos(4\phi(6,4)) + 4uv(u^4-v^4)\sin(4\phi(6,4)))\}$

$\{W(6,6), \frac{1}{3}\pi\lambda^5 ((u^6-15v^2u^4+15v^4u^2-v^6)\cos(6\phi(6,6)) + 2uv(3u^4-10v^2u^2+3v^4)\sin(6\phi(6,6)))\}$

jems describes wavefront aberrations to order 8 [4].

[4] urlhttp://cimewww

Wave-front aberrations to order 8



Wave-front aberrations up to order 8 can be introduced in HRTEM image formation and HRSTEM probe formation.

- Optical system.
- Aberrations.
- **HRTEM transfer function & HRSTEM: optical transfer function**.
- Comparison HRTEM - HRSTEM.

HRTEM

coherent or partially coherent image formation process with coherent or partially coherent incident wave.

→ **TEM** ($\widetilde{T}(\vec{u})$: Transfer Function):

$$\begin{aligned}\widetilde{\Psi}_i(\vec{u}) &= \widetilde{\Psi}_o(\vec{u})\widetilde{T}(\vec{u})\\ \Psi_i(\vec{x}) &= \int \widetilde{\Psi}_o(\vec{u})\widetilde{T}(\vec{u})e^{2\pi\imath\vec{u}\cdot\vec{x}}\mathrm{d}\vec{u}\end{aligned}$$

HRSTEM

incoherent image formation process with coherent or partially coherent probe.

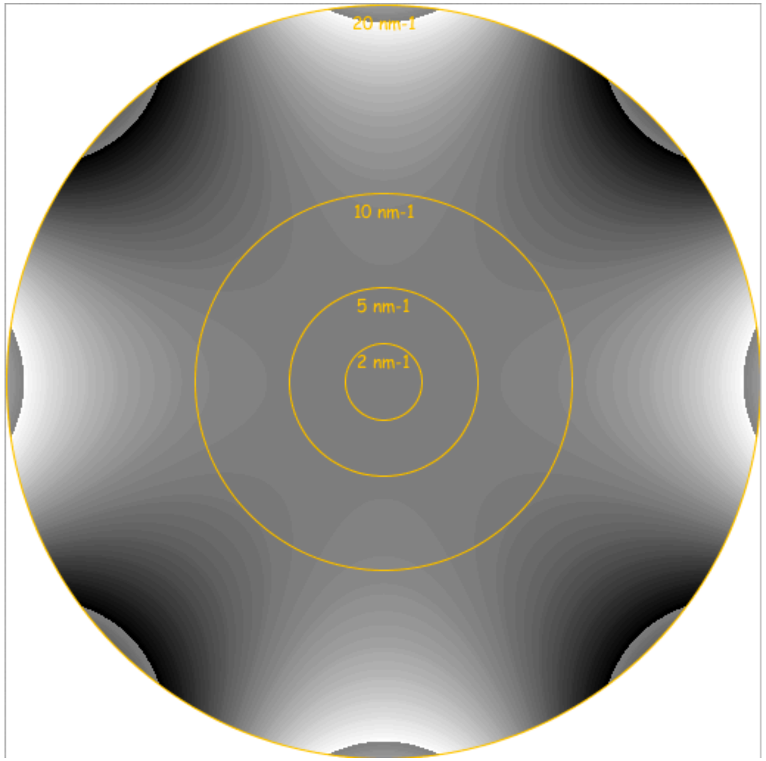
→ **STEM** ($\widetilde{OTF}(\vec{u}) = \widetilde{T}(\vec{u}) \otimes \widetilde{T}(-\vec{u})$: Optical Transfer Function):

$$\begin{aligned}
I(\vec{x}) &= \langle \Psi_i(\vec{x};t)\Psi_i^*(\vec{x};t)\rangle \\
\Psi_i(\vec{x};t) &= \Psi_o(\vec{x};t) \otimes T(\vec{x}) \\
I(\vec{x}) &= \langle [\Psi_o(\vec{x};t) \otimes T(\vec{x})][\Psi_o^*(\vec{x};t) \otimes T^*(\vec{x})]\rangle \quad (\otimes \text{ convolution.}) \\
I(\vec{x}) &= [T(\vec{x})T^*(\vec{x})] \otimes \langle \Psi_o(\vec{x};t)\Psi_o^*(\vec{x};t)\rangle \quad (T(\vec{x}) \text{ is time independent.}) \\
\langle \Psi_o(\vec{x};t)\Psi_o^*(\vec{x};t)\rangle &= |\Psi_o(\vec{x})|^2 \quad \text{(complete spatial incoherence)} \\
I(\vec{x}) &= |\Psi_o(\vec{x})|^2 \otimes [T(\vec{x})T^*(\vec{x})] \\
I(\vec{x}) &= I_o(\vec{x}) \otimes [T(\vec{x})T^*(\vec{x})] = I_o(\vec{x}) \otimes P(\vec{x})
\end{aligned}$$

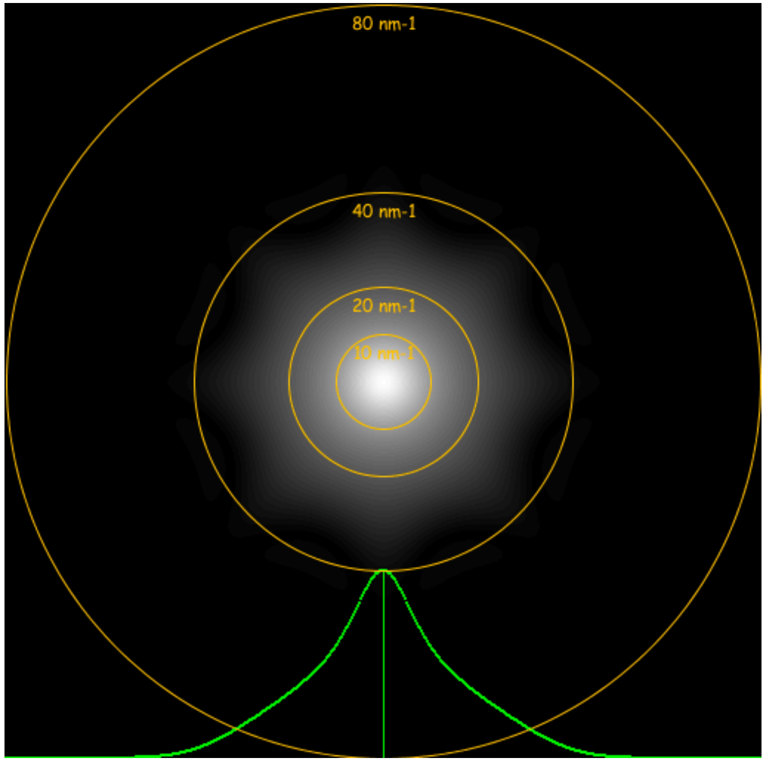
Probe function $P(\vec{x})$: source intensity distribution as measured at the sample plane.

HRTEM / HRSTEM problem: aberrations of optical system

Reaching 0.05 nm resolution sets very strong conditions on aberrations correction.



Aberration figure of $C_{34}(0.5\mu m)$, phase jump at $\frac{\pi}{4}$.

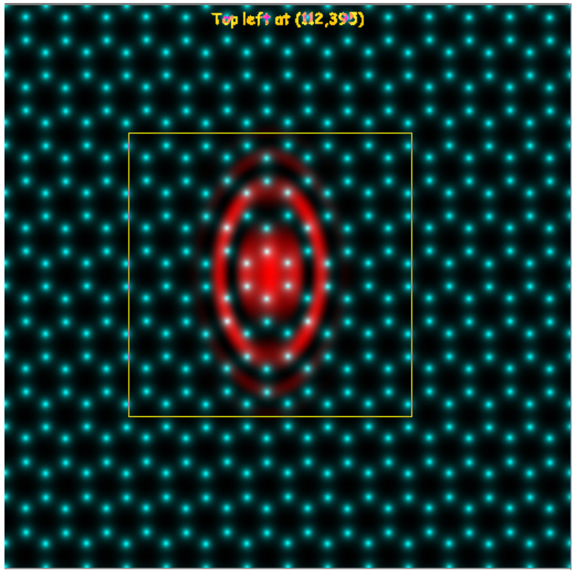


Optical Transfer Function.

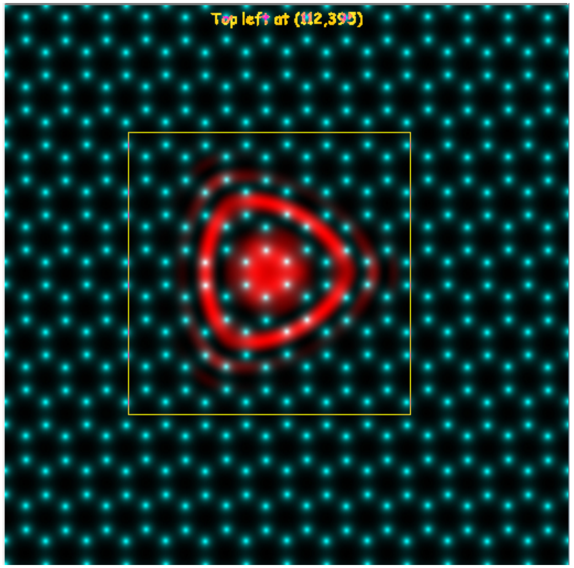
Note that Optical Transfer Function (HRSTEM) transfers higher spatial frequencies than Ccoherent Transfer Function (HRTEM).

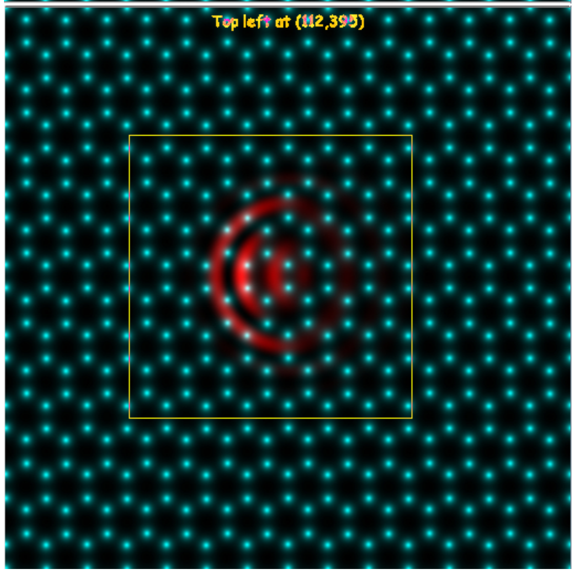
$P(\vec{x})$: source intensity distribution as measured at the sample plane

2 fold astigmatism.

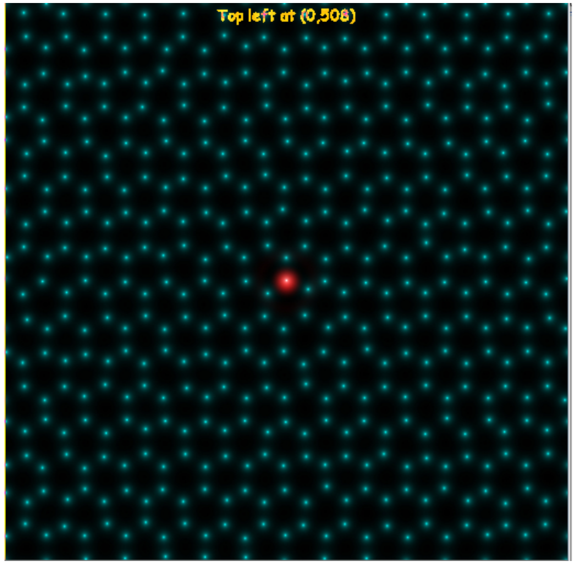


3 fold astigmatism.





Coma.



Corrected probe.

STEM imaging: calculating $I_0(\vec{x})$

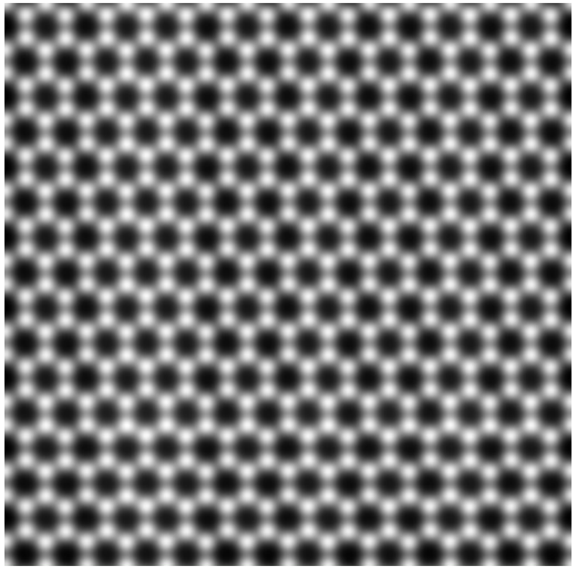
Numerous approximations are involved in calculating $I_0(\vec{x})$:

- Simple projected potential + convolution with probe intensity: no channeling effect (Weak Object Approximation).
- Multislice calculation: channeling + inelastic scattering (absorption or optical potential) + convolution with probe intensity.
- Frozen lattice (phonon) approximation: atoms of super-cell displaced out of equilibrium position, probe scanned on imaged area, intensity collected by annular detector. Allows to simulate HAADF (High Angle Annular Dark Field), BF (Bright Field), MAADF (Medium Angle Annular Dark Field), DPC (Differential Phase Contras), ...
- Pennycook, Nellist, Ishizuka, Shiojiri, Allen, Wang, Rosenauer, van Dyck, ...

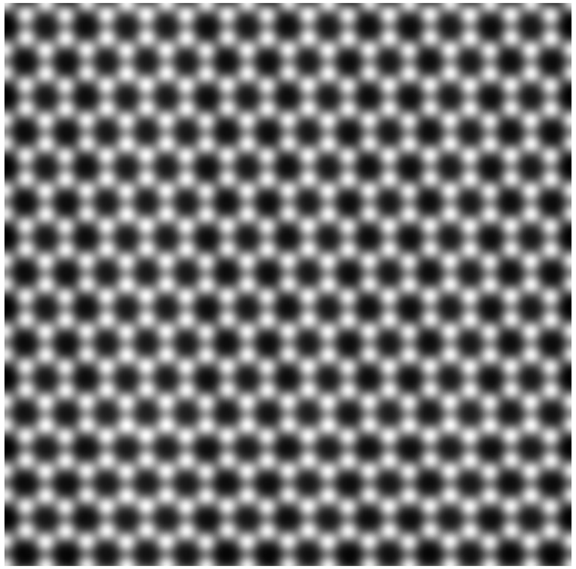
Except the first 2 methods, usually rather long simulation time (faster calculations using GPU).

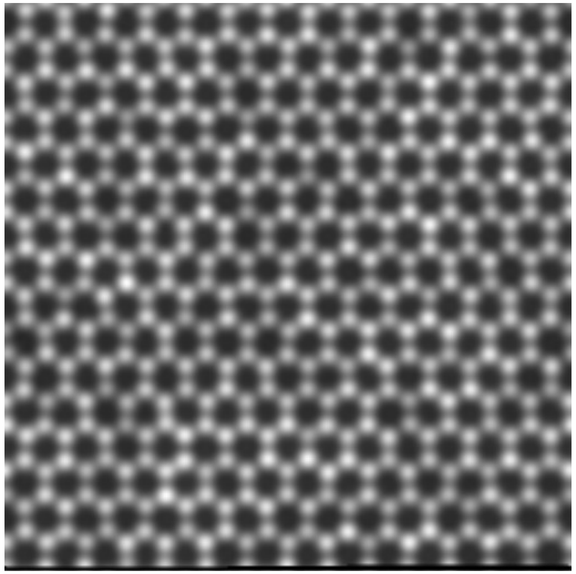
STEM imaging: graphene

Proj. pot. approx.

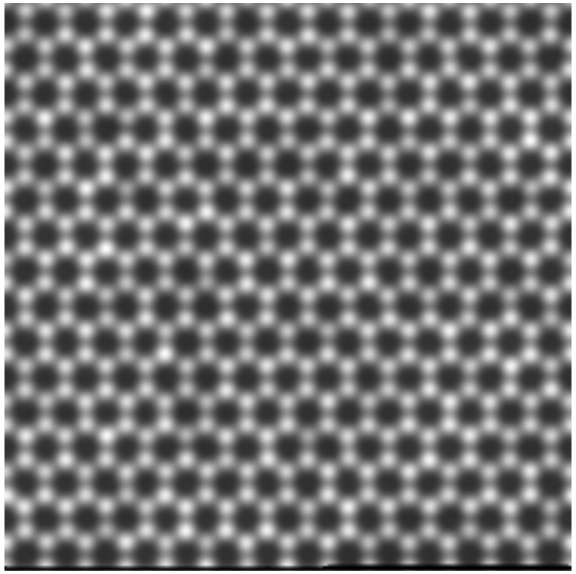


Channeling calculation.





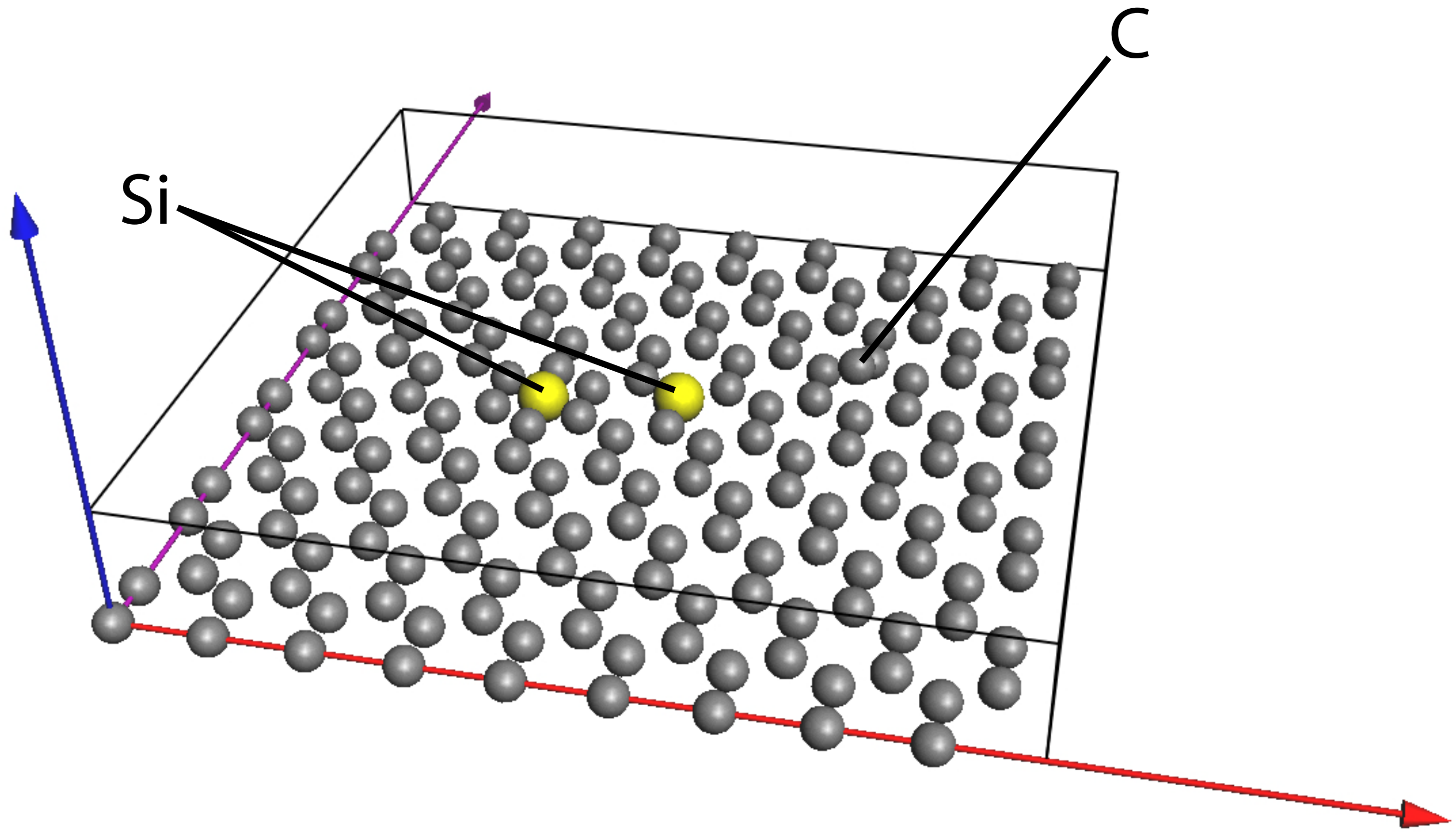
Frozen lattice 5 conf.



Frozen lattice 10 conf.

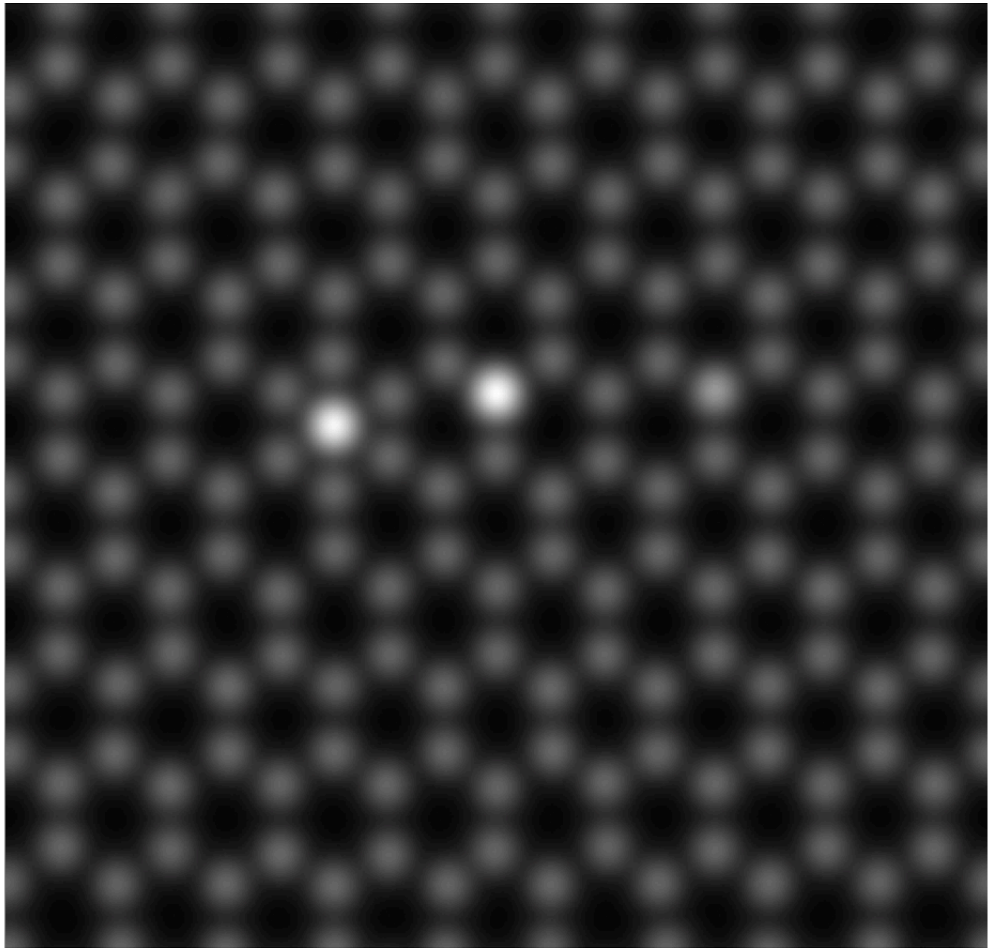
- Optical system.
- Aberrations.
- HRTEM transfer function & HRSTEM: optical transfer function.
- **Comparison HRTEM - HRSTEM**.

HRSTEM - HRTEM comparison: graphene with add atoms

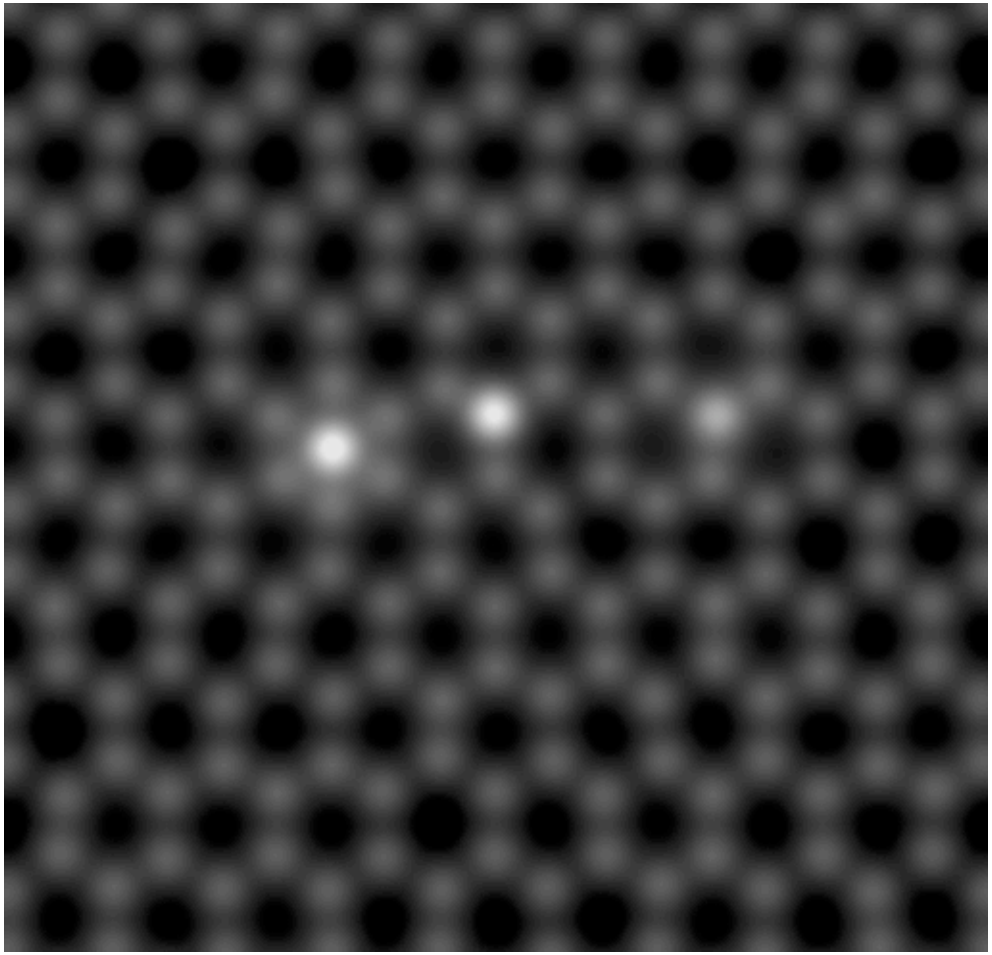


Graphene with Si in 6 C ring, Si substitutional and 2 C column.

Graphene: HAADF (100 kV, 70 -150 mrad)



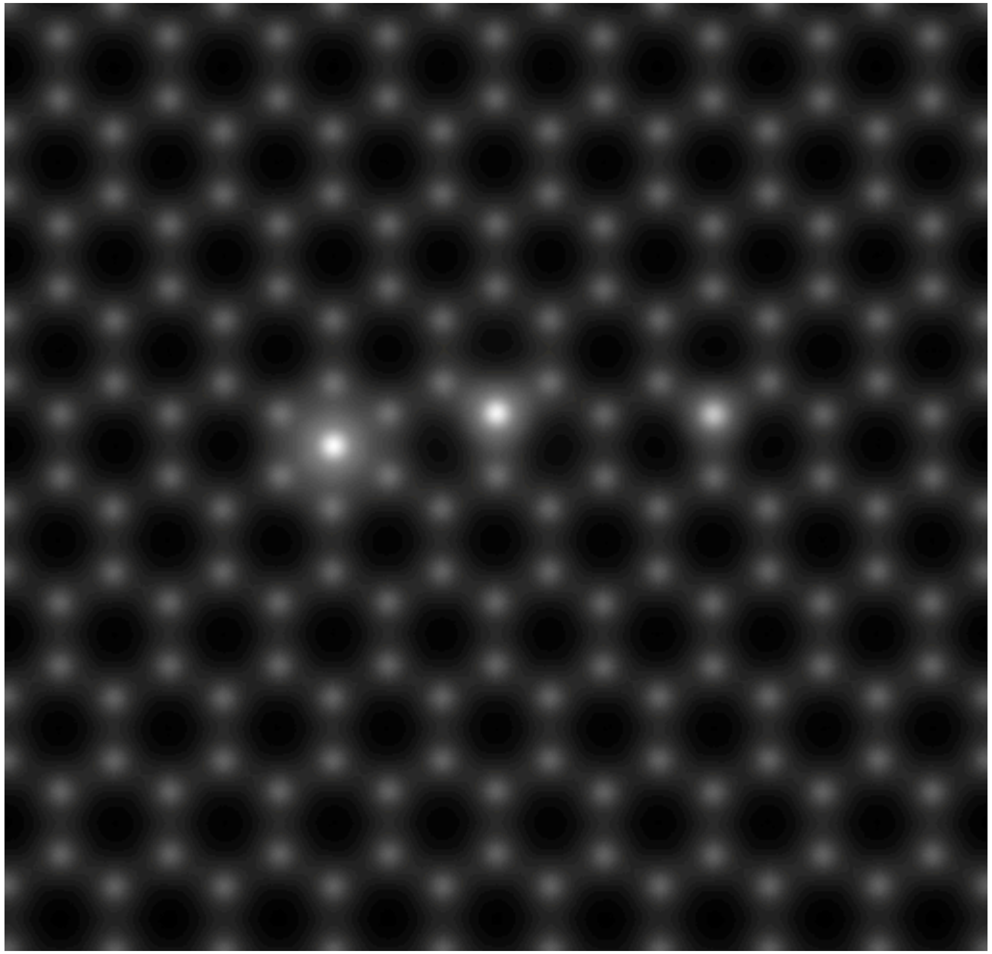
Frozen lattice (∼400 s).



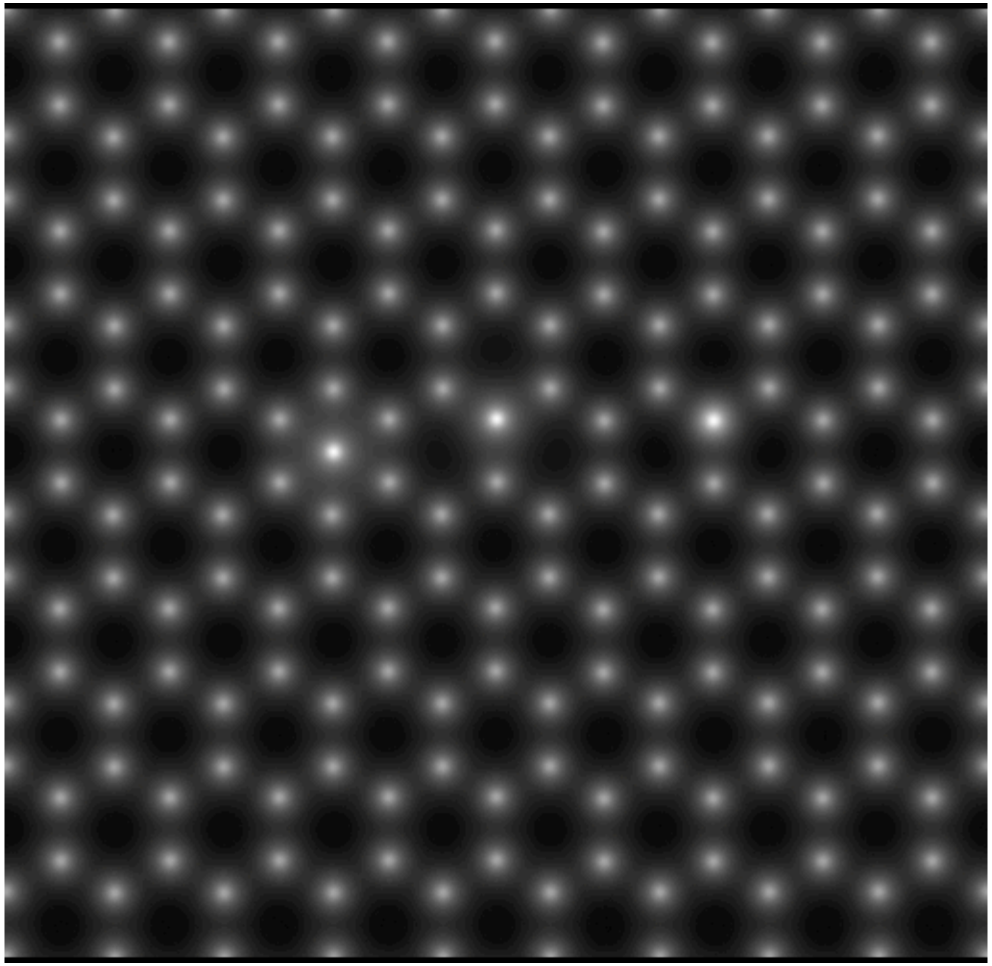
Channeling (∼2 s).

One Si shows more contrast than 2 C atoms ($i \sim Z^2$) : 14^2 compared to $\sim 2 \times 6^2$.

Graphene: HRTEM (100 kV, $C_s - 0.033mm$)



Weak phase object app., $C_c = 0.5mm$



Multislice, $C_s = -0.033mm$, $C_c = 0$, no thermal magnetic noise.

HRTEM does not display the strong contrast difference between one Si and two C as given by HAADF.

Thanks for your attention!