

Introduction to Image Simulation

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HRTEM image simulation

HRTEM or HRSTEM image simulation helps understand experimental images and provides valuable information about several crystal parameters pretty difficult to assess by any other technique (precise specimen orientation or thickness, atomic columns position, atomic species, etc). It is particularly useful when dealing with crystal defects (dislocations or interfaces). In this presentation the technique is briefly introduced.

Recommended books are:

- ▶ *J.M. Cowley, Diffraction Physics.*
- ▶ P.B. Hirsch, A. Howie, R.B. Nicholson, W.D. Pashley & M.I. Whelan, *Electron Microscopy of Thin Crystals.*
- ▶ J.-P. Morniroli, *Large-Angle Convergent-Beam Electron Diffraction.*
- ▶ S.J. Pennycook & P.D. Nellist, *Scanning Transmission Electron Microscopy.*
- ▶ *J.C.H. Spence, High Resolution Electron Microscopy.*
- ▶ Z.L. Wang, *Elastic and Inelastic Electron Scattering in Electron Diffraction and Imaging.*

Recommended papers:

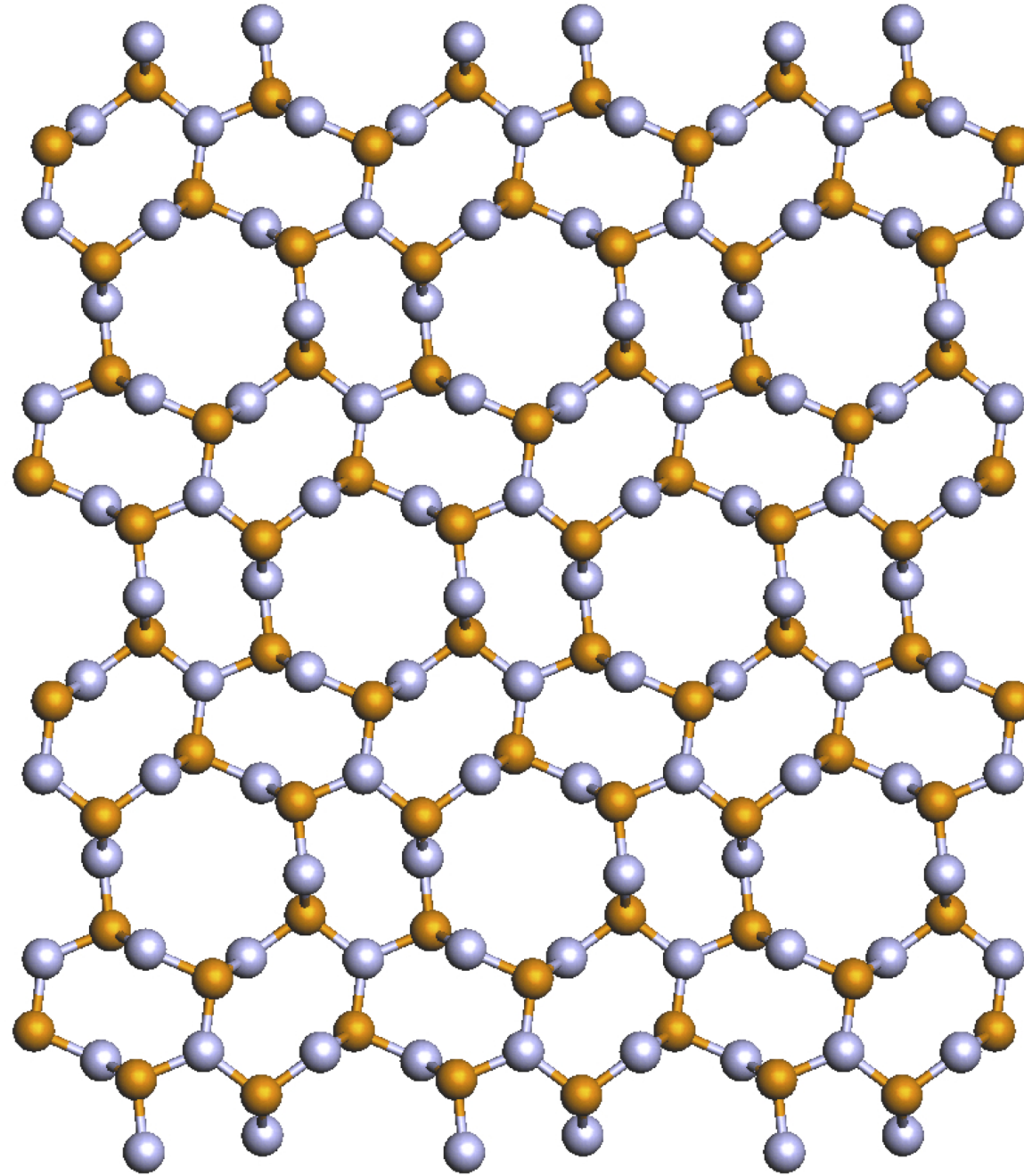
- ▶ J.M. Cowley & A.F. Moodie, *Acta Cryst.* **10**, (1957) 609-619.
- ▶ *D. Gratias & R. Portier, Acta Cryst.* **A39** (1983) 576-584.
- ▶ K. Ishizuka, *Ultramicroscopy* **5** (1980) 55-65, *Ultramicroscopy* **90** (2002) 71-83.
- ▶ D. Van Dyck, *Phys. Status Solidi* **72** (1975) 321-336.

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

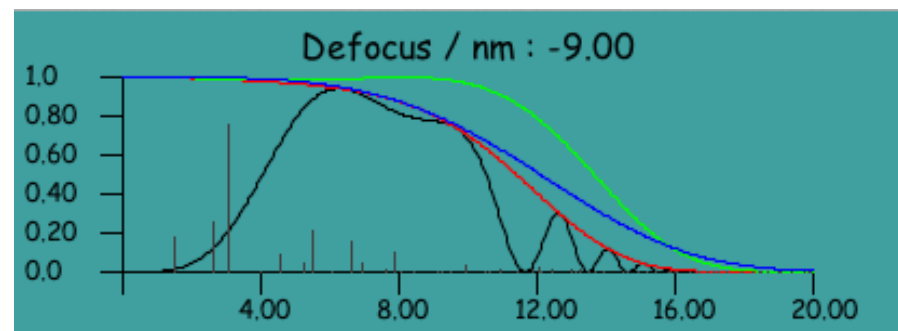
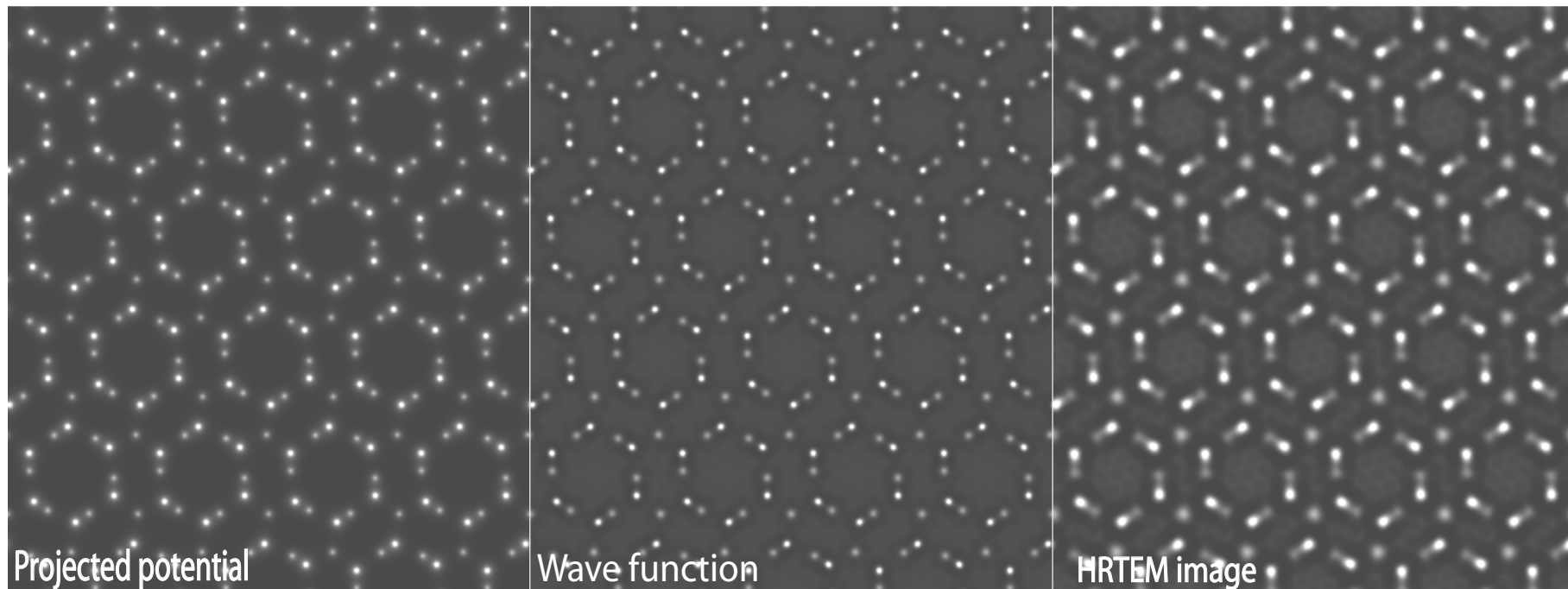
- ▶ Specimen structure..
- ▶ Chemical composition.
- ▶ Functional properties.
- ▶ ...

But HR(S)TEM images depend of several adjustable microscope parameters.

For example **object defocus** strongly affects HRTEM images.

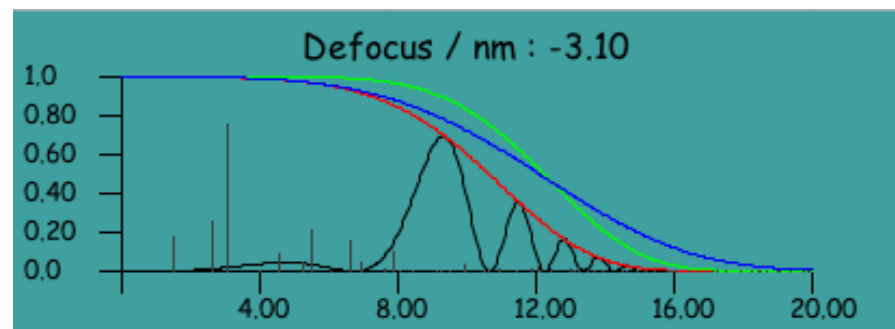
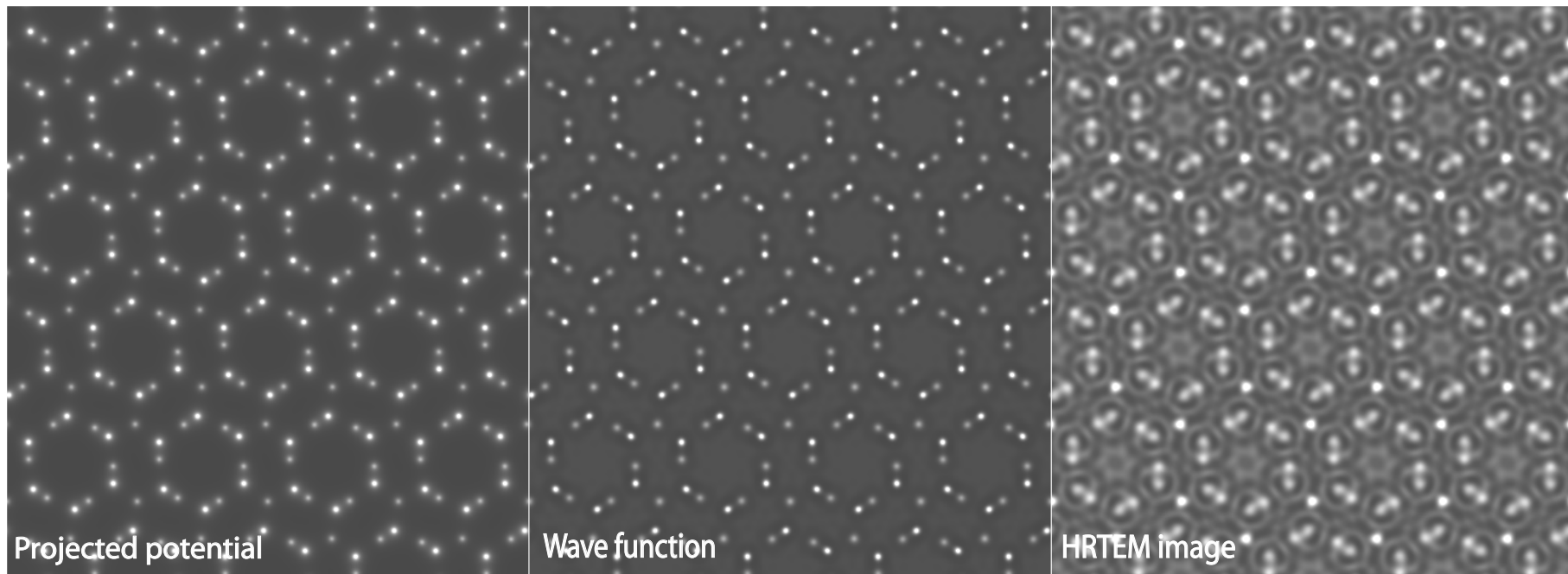


Images: Si_3N_4 P 63: [001], 10 nm thick, -9 nm defocus



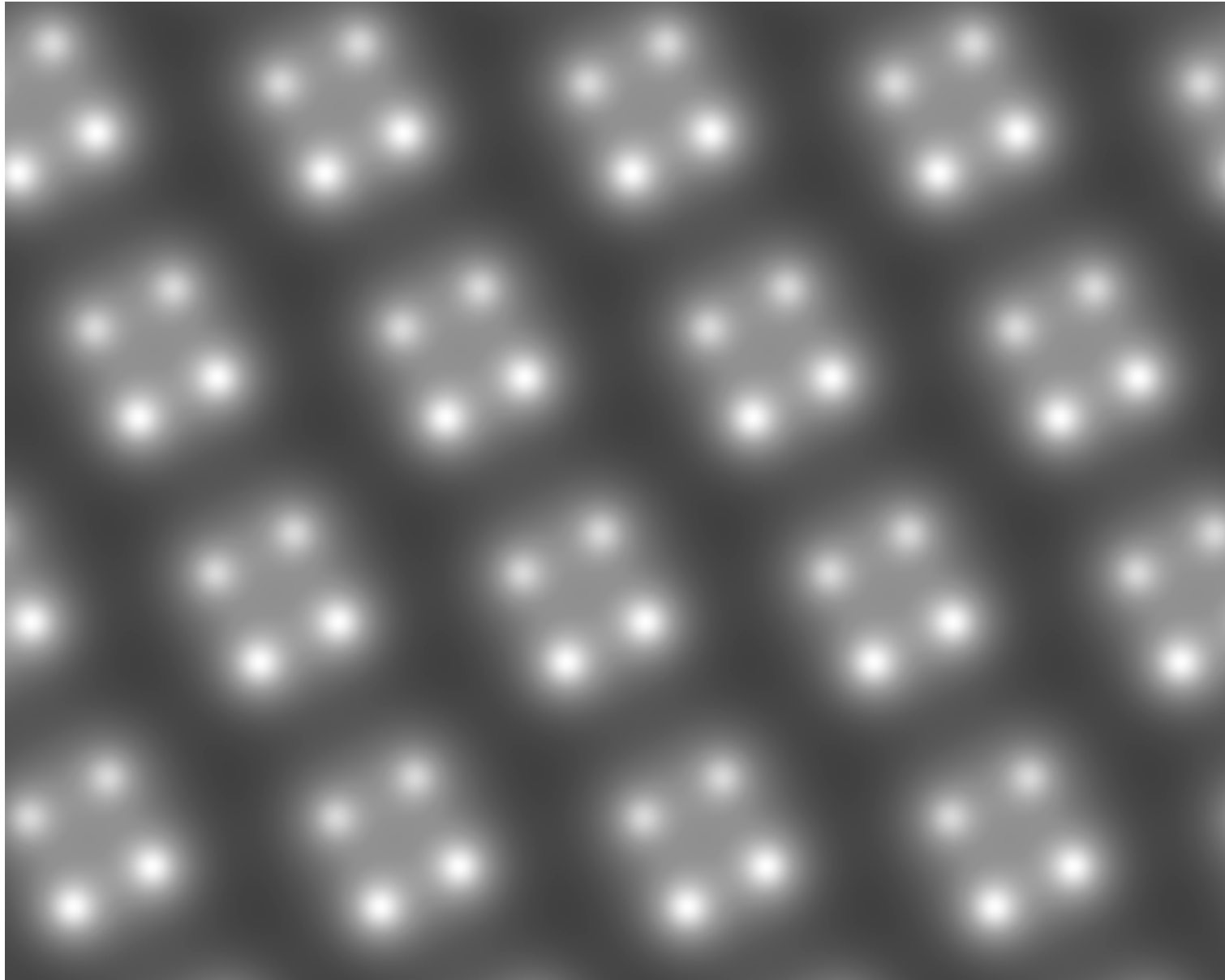
Imaging parameters: Titan 80-300 (300 kV), C_s : $-33 \mu\text{m}$

Si_3N_4 P 63: [001], 10 nm thick, -3 nm defocus



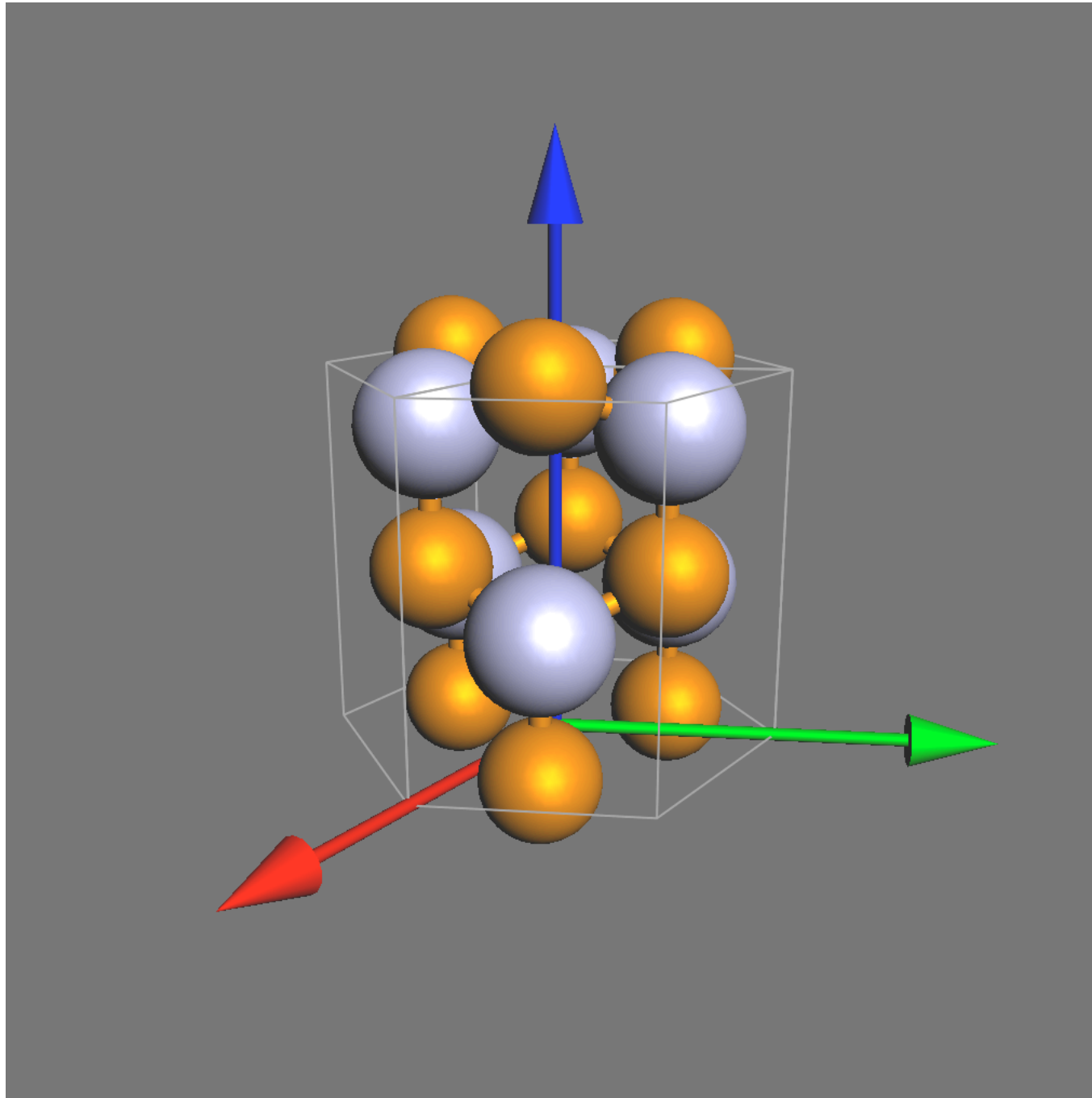
Imaging parameters: Titan 80-300 (300 kV), C_s : -33 μm

Projected potential: structure: ?



Projected potential of some structure. Is the crystal system cubic, tetragonal, orthorhombic (or other)?

Answer: BN hexagonal, $[uvw] = [1, 1, -2, 3]$ or $[1, 1, 1]$



- ▶ **Why?**
- ▶ **How?**
 - ▶ **Models (crystallography).**
 - ▶ **Approximations.**
 - ▶ **Methods.**
 - ▶ **Software.**

Books on crystallography that you may find useful:

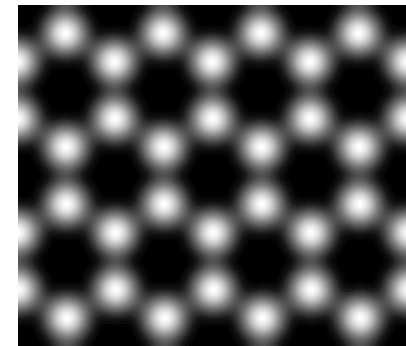
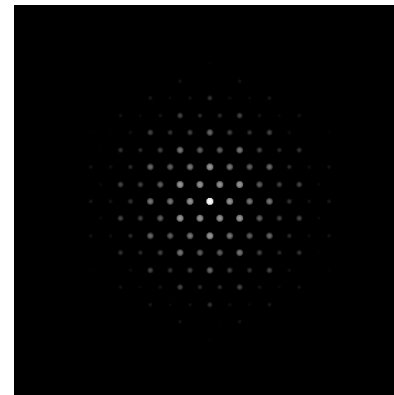
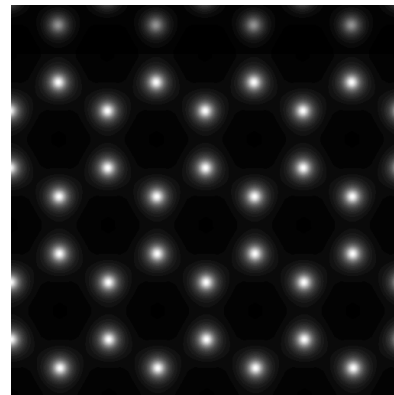
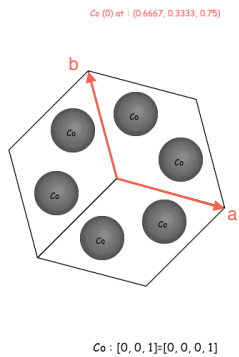
- ▶ D.E. Sands, *Introduction to Crystallography*, Dover Books on Chemistry.
- ▶ D.E. Sands, *Vectors and Tensors in Crystallography*, Dover Publications Inc.

We will consider 2 structures and show that image simulation helps understand HRTEM images. The first structure is very simple but hexagonal. In hexagonal crystals (hkl) plane normals are not parallel to [uvw] directions where (h=u, k=v & l=w) (except (001) // [001]).

- ▶ **Simple structure: Co**
- ▶ **More complicated structure: $MgZn_2$**

Simple structure: Co

When simple structures are imaged, image simulation may not be necessary at all. For example looking at Co (P63/mmc, magnetic!), model structure, projected potential, SAED, HRTEM are simple (and straight forward to interpret?):



Model structure
Cobalt, [001].

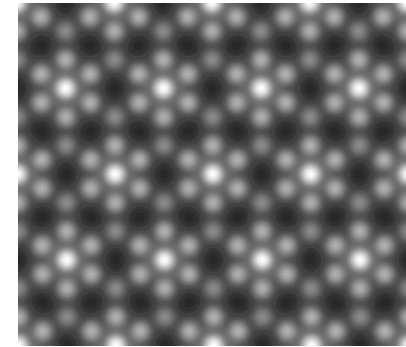
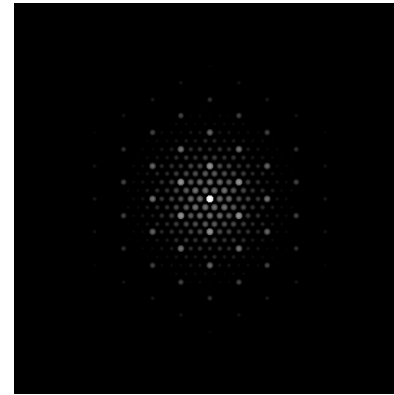
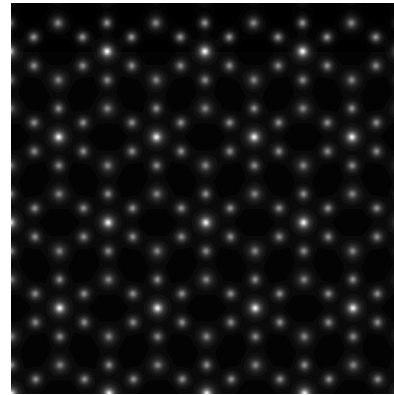
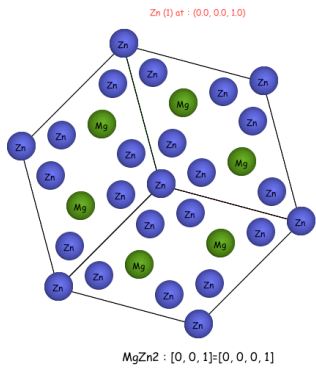
Projected potential
Co [001]

Selected Area
Electron Diffraction
(SAED) Co [001].

HRTEM image Co
[001], Titan negative
Cs.

More complicated structure: $MgZn_2$

When moderately complicated structures are imaged, image simulation may still not be necessary. For example looking at $MgZn_2$ (P63/mmc), model structure, projected potential, SAED, HRTEM are simple (SAED straight forward to interpret?):



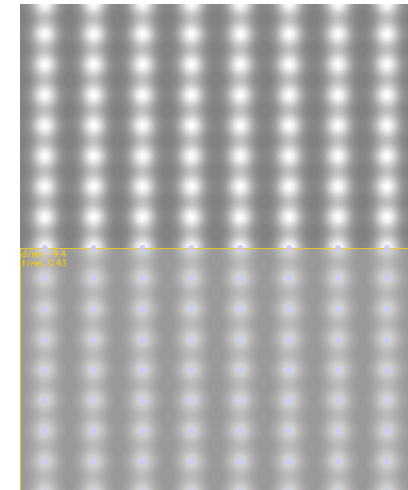
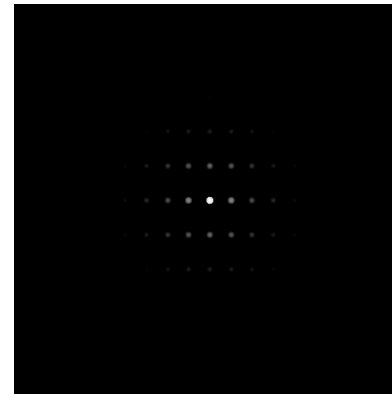
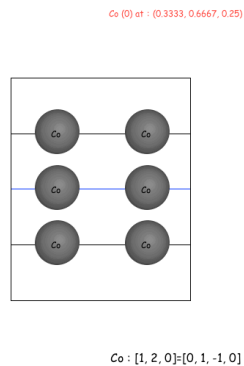
Model structure
 $MgZn_2$, [001].

Projected potential
 $MgZn_2$ [001]

Selected Area
Electron Diffraction
(SAED) $MgZn_2$
[001].

HRTEM image
 $MgZn_2$ [001], Titan
negative Cs.

Co observed in $[120]$ projection (Weber notation $[10-10]$)



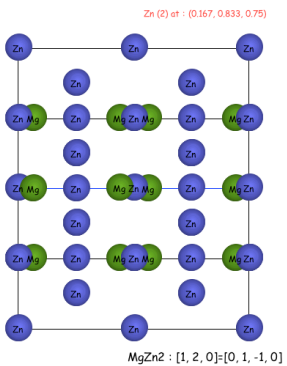
Model structure
Cobalt, $[120]$.

Projected potential
Co $[120]$

Selected Area
Electron Diffraction
(SAED) Co $[120]$.

HRTEM image Co
 $[120]$, Titan negative
Cs.

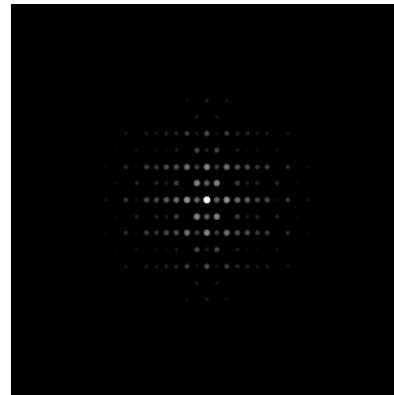
$MgZn_2$ observed in $[120]$ projection (Weber notation $[10-10]$)



Model structure
 $MgZn_2$, $[120]$.



Projected potential
 $MgZn_2$ $[120]$



Selected Area
Electron Diffraction
(SAED) $MgZn_2$
 $[120]$.

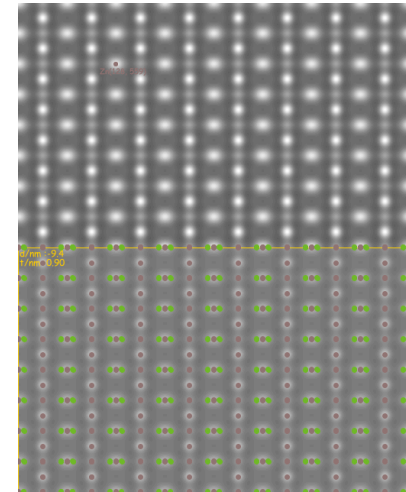


Figure: HRTEM image
 $MgZn_2$ $[120]$, Titan
negative Cs.

How to do HRTEM or HAADF image simulation?

We need models, (good) approximations and methods.

1. Models

1.1 Crystal

- ▶ Lattice parameters.
- ▶ Symmetries (space-group, regular point system).
- ▶ Atoms position.
- ▶ Orientation, ([uvw] zone axis indices, (hkl) Laue circle center indices with $u h + v k + w l = 0$).
- ▶ Shape (thickness, defect, ...).

1.2 Microscope

- ▶ Source coherence (i.e. size, energy spread).
- ▶ Accelerating voltage.
- ▶ Objective lens properties (Cs: spherical aberration coefficient, Cc: chromatic aberration coefficient, ...).

1.3 Detector: Modulation Transfer Function (MTF).

2. Approximations

2.1 Elastic scattering (under small angle scattering approximation, i.e. acc. voltage ≥ 50 kV):

- ▶ Kinematical: single scattering event.
- ▶ Dynamical: multiple scattering events.

2.2 Inelastic scattering:

- ▶ Single inelastic scattering.
- ▶ Multiple inelastic scattering.
- ▶ Frozen lattice or frozen phonon.

2.3 Abbe imaging theory (transmission cross-coefficients or transfer function + envelopes).

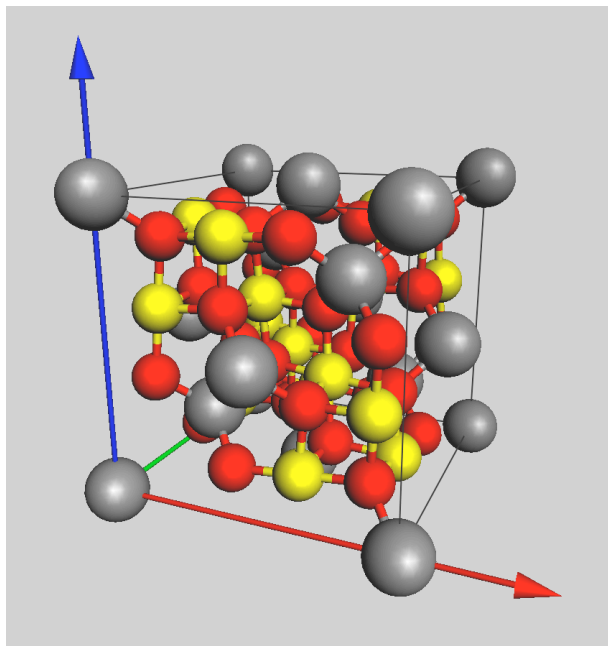
3. Methods

3.1 Bloch wave (solid state physics).

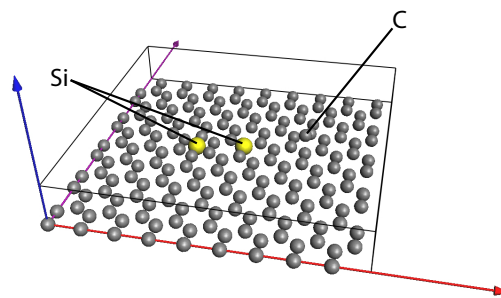
3.2 Multislice (physical optics).

3.3 Howie-Whelan (column approximation).

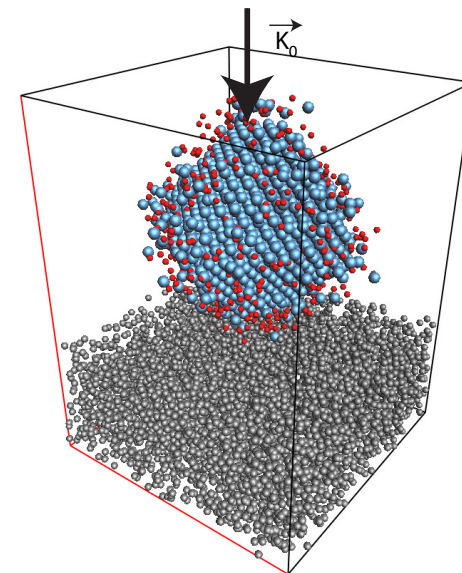
Models are **not** necessarily crystalline.



CoCr₂O₄ (cubic, $Fd\bar{3}m$, 3 atoms).



Graphene sheet with add atoms (448 atoms).

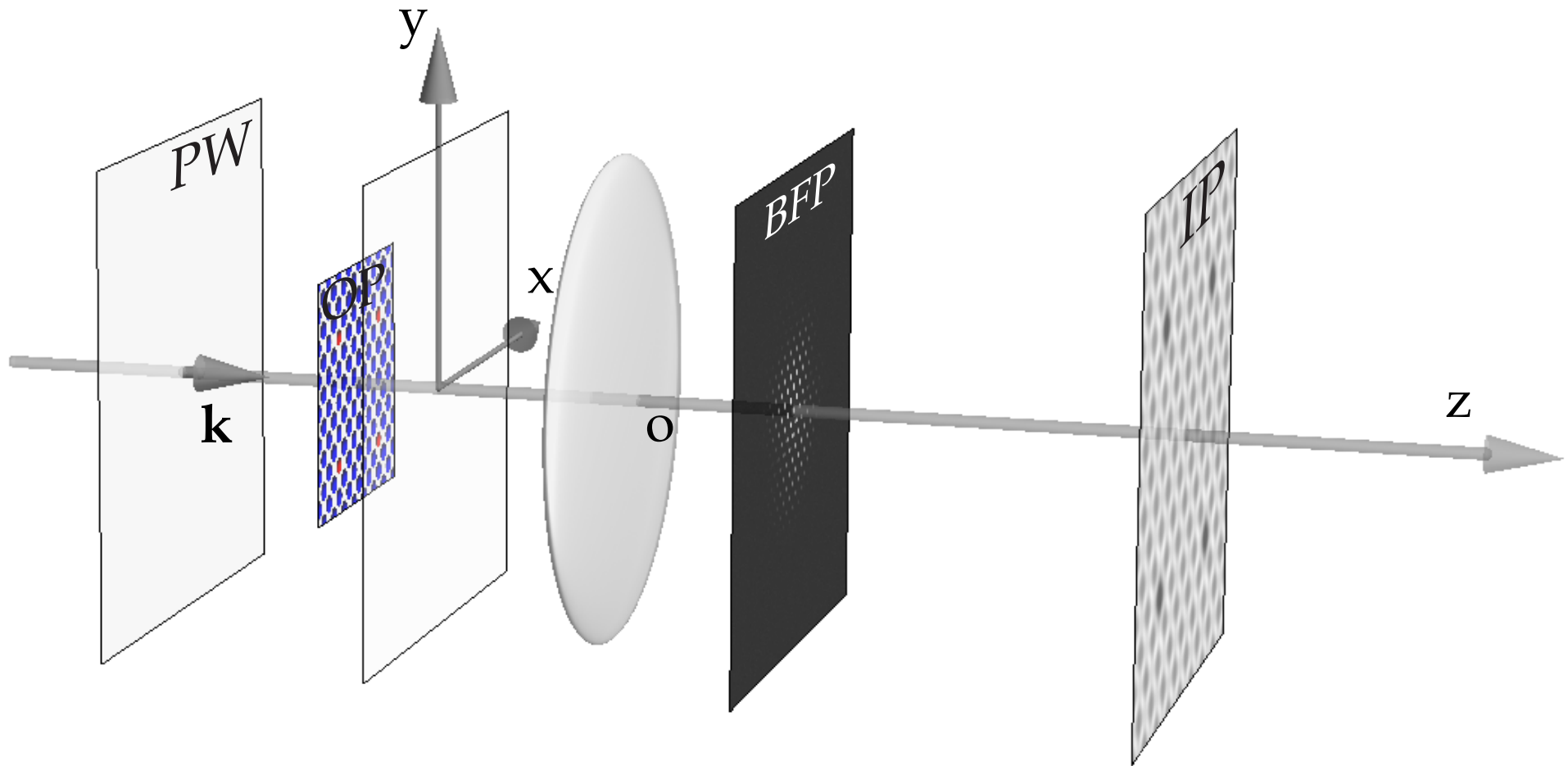


A

Pt catalyst (Pt cube octahedron on amorphous carbon film, 10'000 atoms).

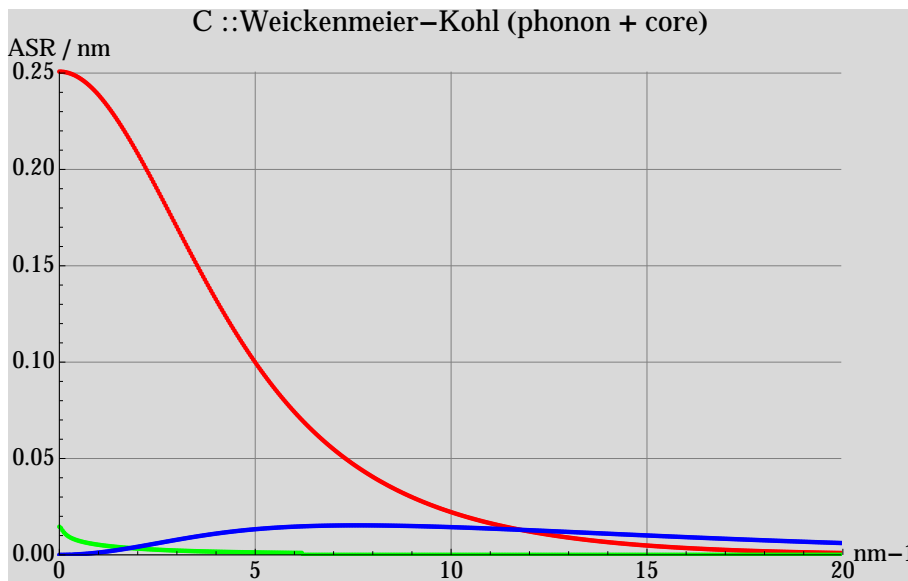
A model is a box of parameters $(a, b, c, \alpha, \beta, \gamma)$ with atoms at (x, y, z) such that $0.0 \leq (x, y, z) < 1.0$. The symmetries (space-group) helps defining the structure, the extinctions, etc. Wrong models do not provide reliable HRTEM or HRSTEM simulated images.

Model: the electron microscope (paraxial optics or gaussian optics)

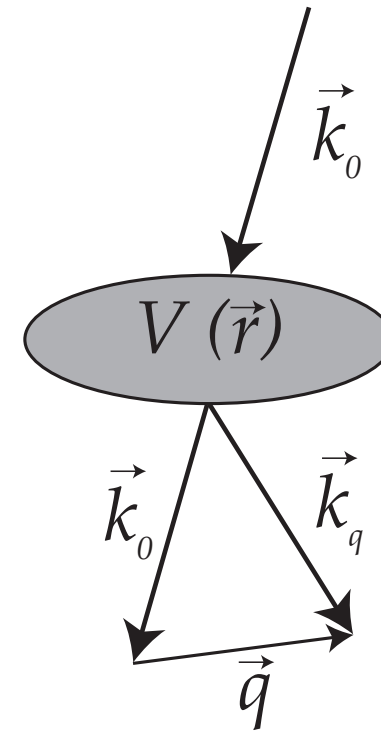


PW: incident Plane Wave, OP: Object Plane, BFP: Back Focal Plane, IP: Image Plane.
Only objective lens is modelled and axial aberrations considered, since it is the first imaging lens and its lateral magnification G_l is very large (HRTEM). Lateral magnification corresponds to angular compression $G_a = \frac{1}{G_l}$ ($G_l G_a = 1$).

Approximation: elastic scattering



Atomic Scattering Amplitude (carbon),
red: elastic, green: core absorption, blue:
TDS (Thermal Diffuse Scattering).



Electrons interact with
the crystal potential
 $V(\vec{r})$.

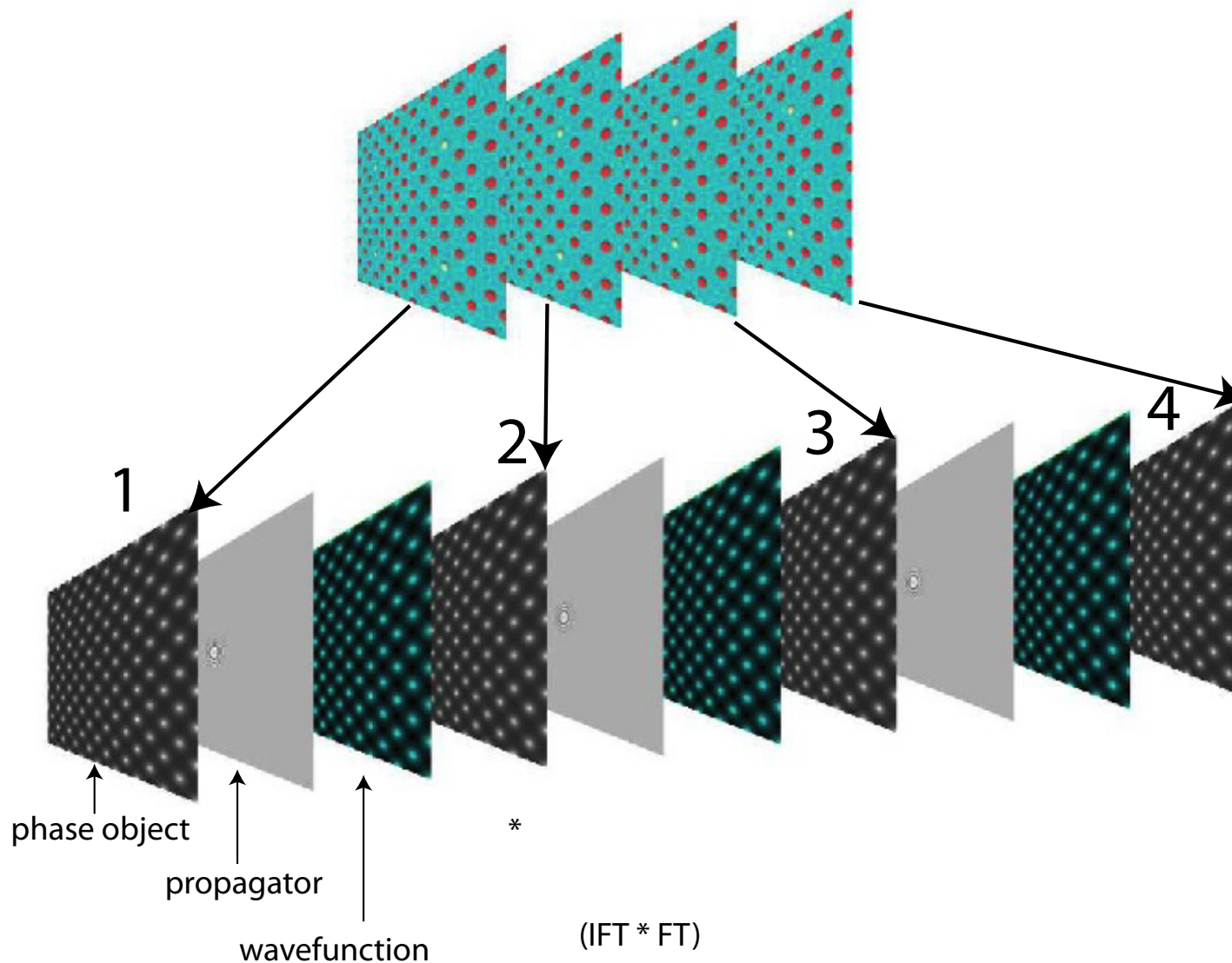
Scattering:

$$\vec{k}_q = \vec{k}_0 + \vec{q}$$

Elastic scattering:

$$||\vec{k}_q|| = ||\vec{k}_0||$$

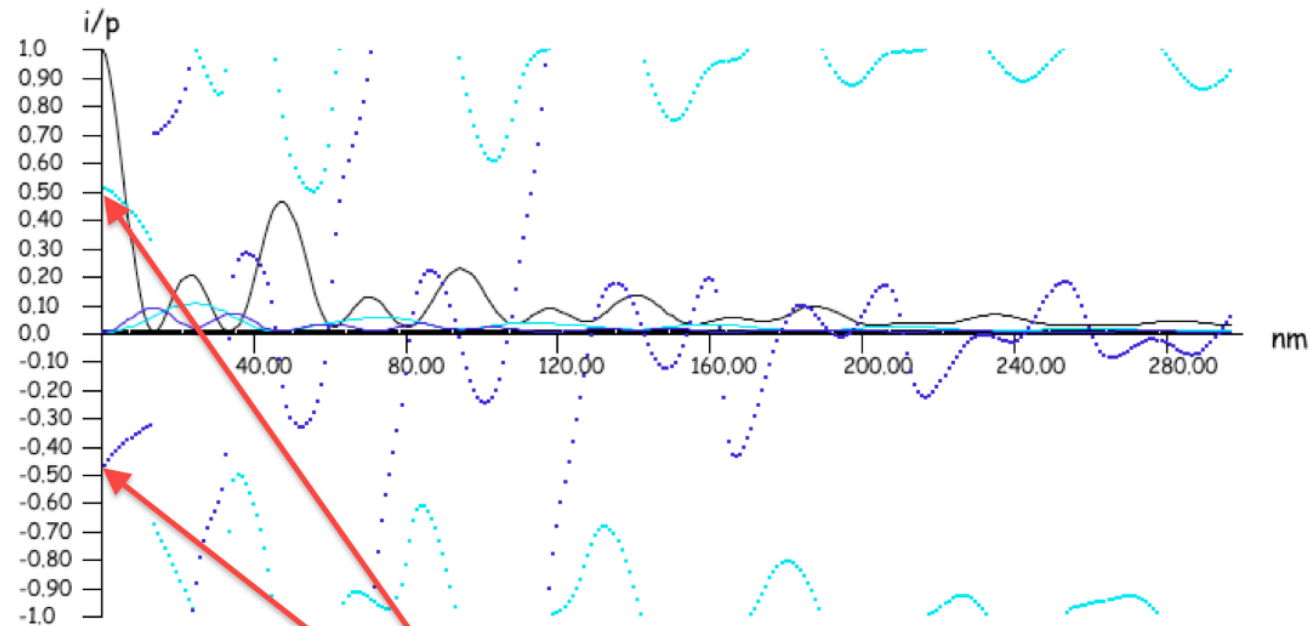
Dynamical scattering calculations: multislice method



A very popular (and powerful) method for performing dynamical calculation has been proposed by J. M. Cowley and A.F. Moodie. It makes use of several principles of Optical Physics.

Dynamical scattering: CoCr_2O_4 [001]

$\text{CoCr}_2\text{O}_4::(4, 0, 0)::[0, 0, 1]::(0.000, 0.000, 0.000)$

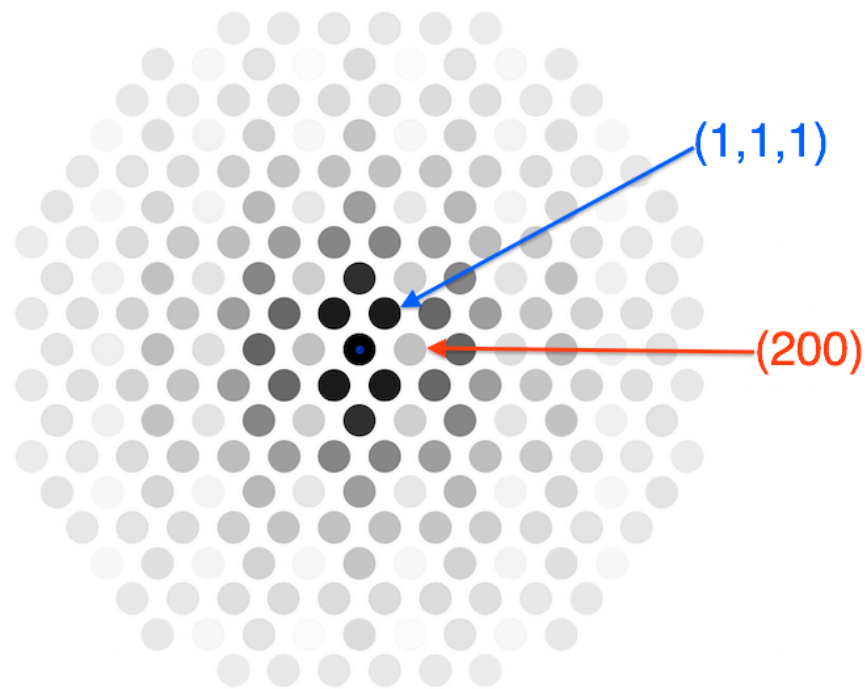


$\pi / 2$ phase shift!

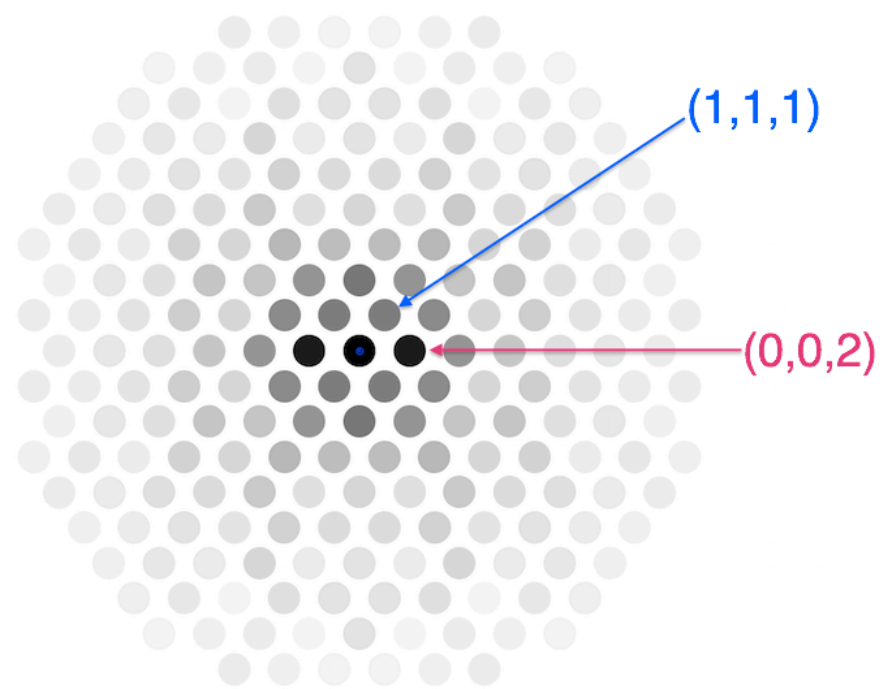
0 2 4
0 2 0
0 0 0

Dynamical scattering, i.e. electrons undergo multiple scattering events during their interaction with $V(\vec{r})$, makes the amplitude & phase of the transmitted (i.e. un-scattered) beam and the reflections (scattered beams) vary widely as a function of specimen thickness.

Dynamical scattering: Si [110]

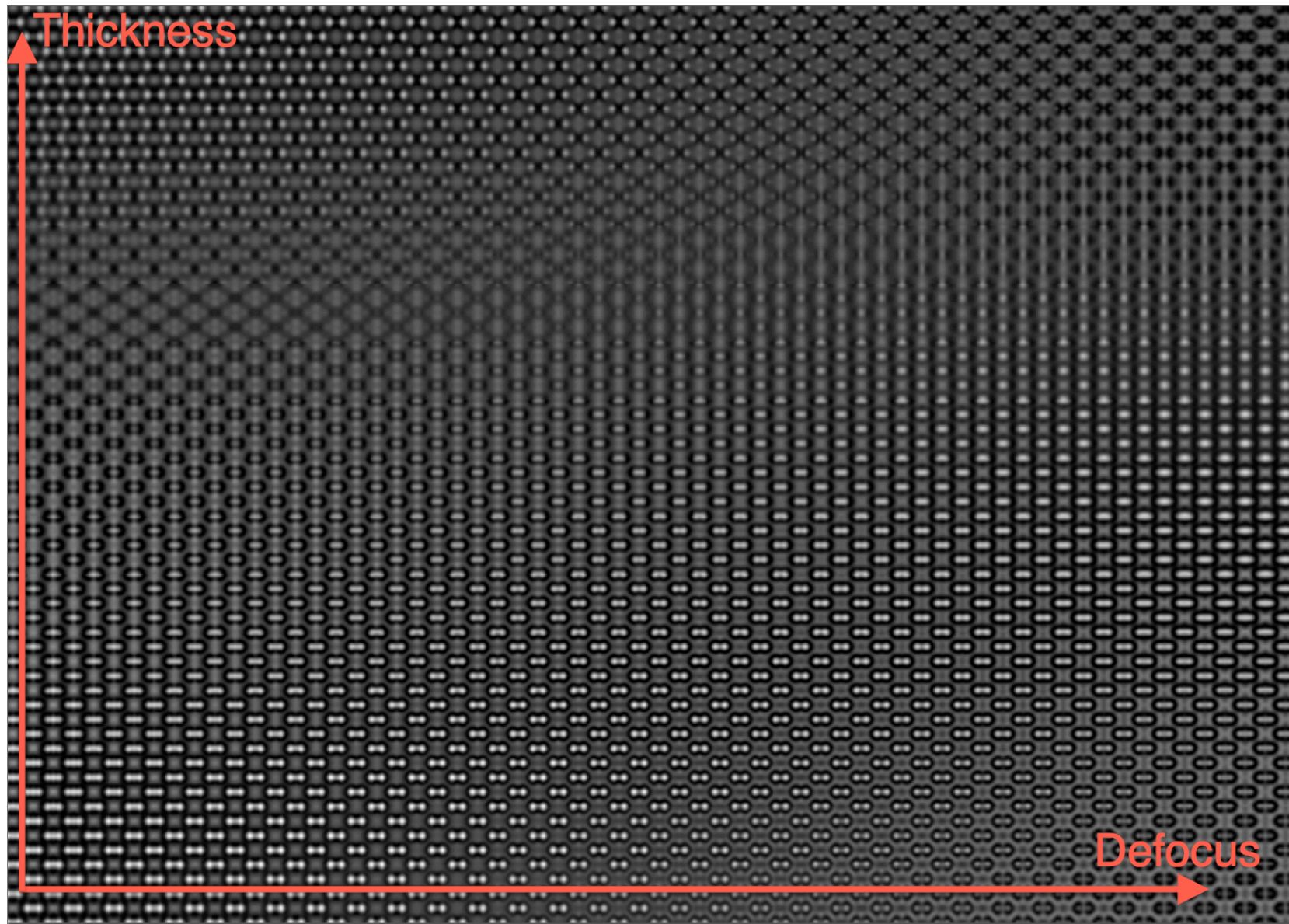


2 unit cell thick Si [110].



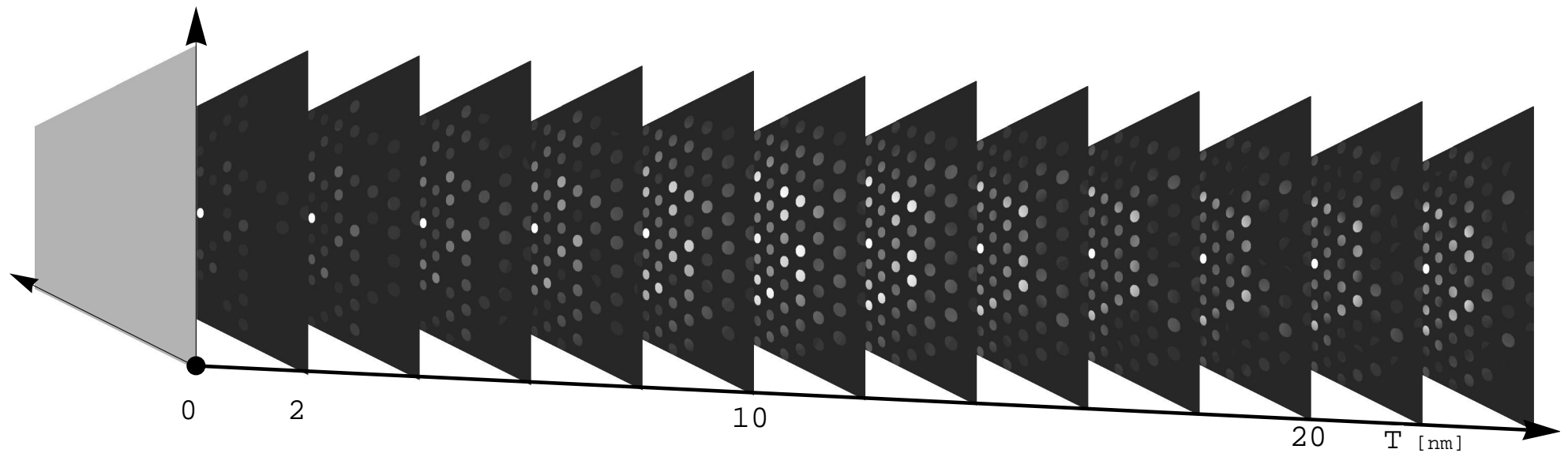
50 unit cells thick Si [110].

Very often for particular crystal thicknesses forbidden reflections are more intense than regular reflections. Example Si [110] where $I_{(002)} > I_{(111)}$.



Note that similar images appear at particular thickness or defocus (Fourier images).

Dynamical scattering: thickness series



The extinction distance is related to the Pendellösung period.

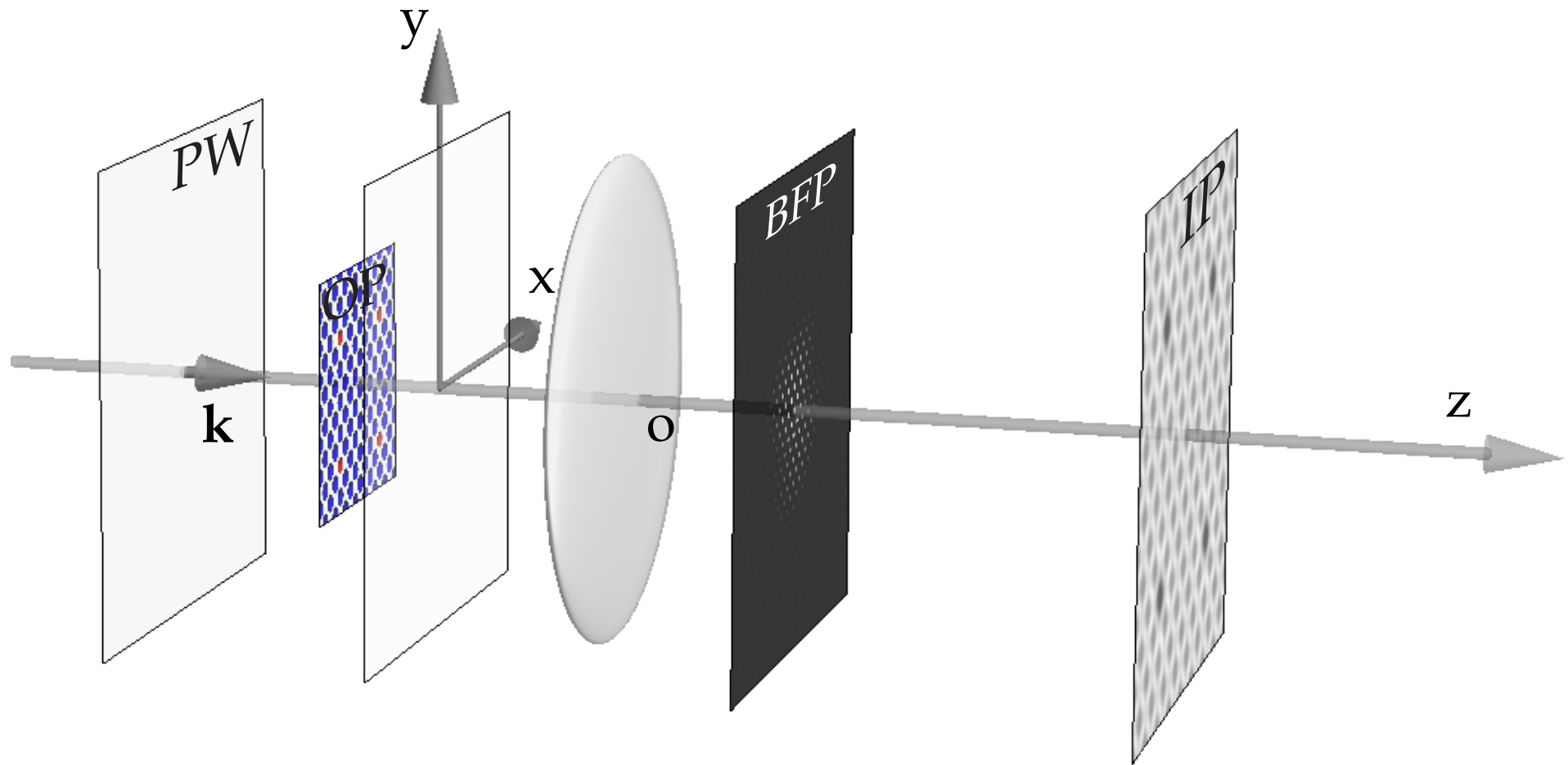
1. Bad

- ▶ Phase relationship between transmitted and diffracted beams depends on thickness → HRTEM lattice fringes or contrast not always on atomic column position.
- ▶ Amplitude of transmitted and diffracted beams depends on thickness → Some HRTEM lattice fringes may be missing or supplementary ones present.

2. Good

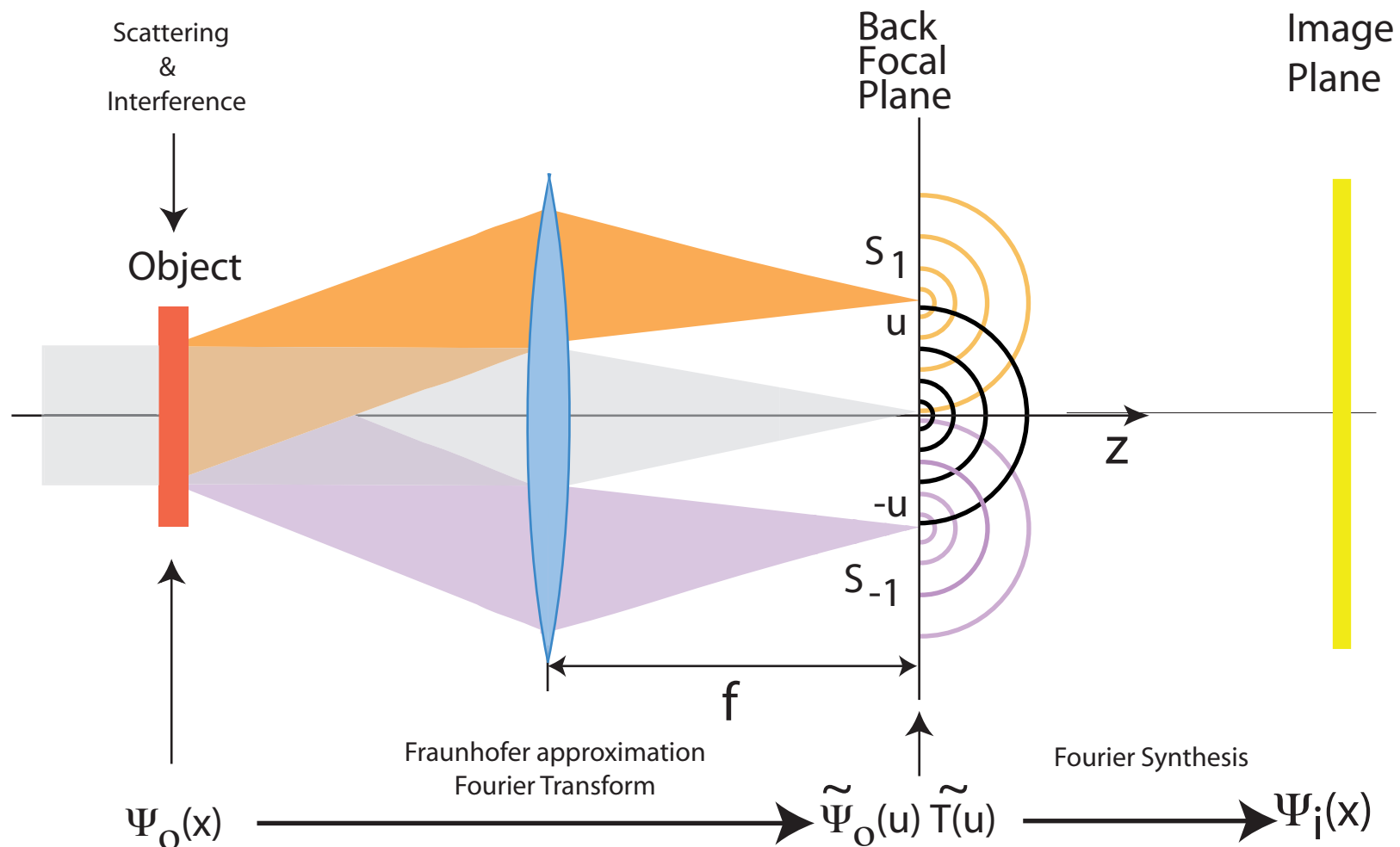
- ▶ Specimen thickness can be determined precisely (~ 1 nm or better).
- ▶ Specimen polarity can be determined.
- ▶ Specimen deformation can be determined precisely.

Model: electron microscope



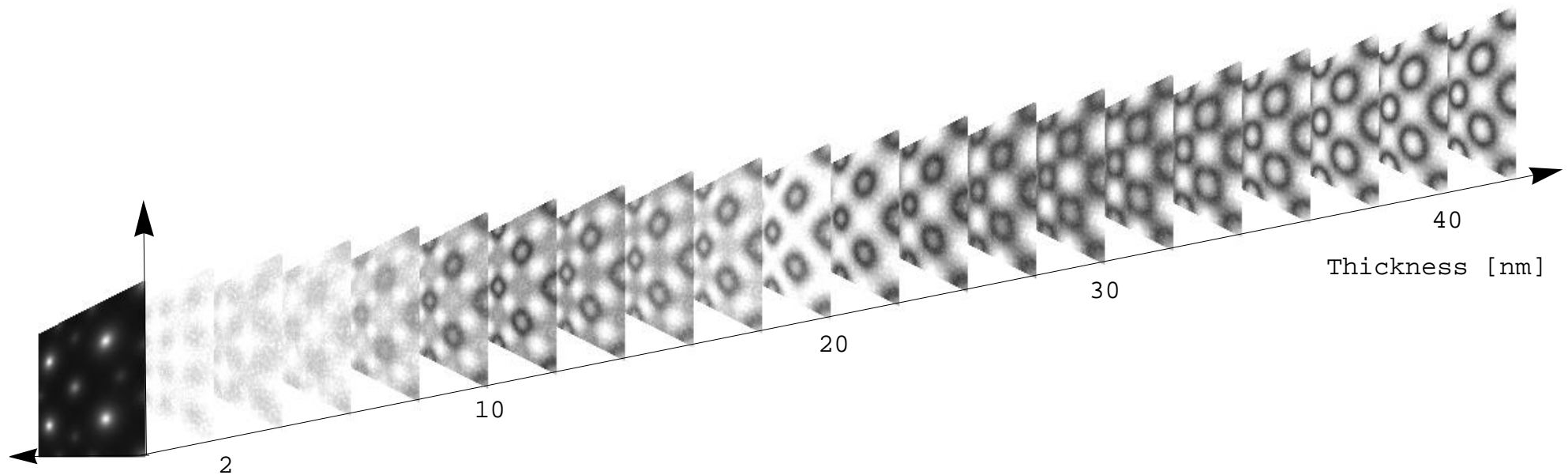
The electron microscope transfer is modelled by a transfer function $\tilde{T}(\vec{u})$ that acts only in its Back Focal Plane (BFP). The BFP is the plane where the Fourier transform of the object wave function $\tilde{\Psi}_o(\vec{u})$ is formed. In the Image Plane, the image intensity is $\Psi_i(\vec{x}) \Psi_i(\vec{x})^*$ i.e. the modulus square of the image wave function $\Psi_i(\vec{x})$.

Model: Abbe image formation



The objective lens changes the phase relationship between the transmitted and diffracted beams. Moreover not all diffracted beams are transmitted, due to its small acceptance angle. High spatial frequencies (i.e. beams diffracted at large angles) are damped due to partial spatial and temporal coherence, electronic noise, mechanical vibrations or drift.

Thickness series



For a very small crystal thickness, a positive C_s objective lens and optimal defocus the atomic columns position appear as black dots in HRTEM (Weak Phase Object Approximation).

Increasing the crystal thickness makes the HRTEM images less straight forward to interpret. In high symmetry zone axis orientation, the electrons channel along the atomic columns and images present contrasts not correlated to them.

Two cases:

→ **TEM** ($\tilde{T}(\vec{u})$: **T**ransfer **F**unction):

$$\tilde{\Psi}_i(\vec{u}) = \tilde{\Psi}_o(\vec{u}) \tilde{T}(\vec{u})$$

$$\Psi_i(\vec{x}) = \int \tilde{\Psi}_o(\vec{u}) \tilde{T}(\vec{u}) e^{2\pi i \vec{u} \cdot \vec{x}} d\vec{u}$$

→ **STEM** ($\widetilde{OTF}(\vec{u}) = \tilde{T}(\vec{u}) \otimes \tilde{T}(-\vec{u})$: **O**ptical **T**ransfer **F**unction):

$$I(\vec{x}) = \langle \Psi_i(\vec{x}; t) \Psi_i^*(\vec{x}; t) \rangle$$

$$I(\vec{x}) = I_o(\vec{x}) \otimes \widetilde{OTF}(\vec{x}) \quad (\otimes \text{ convolution})$$

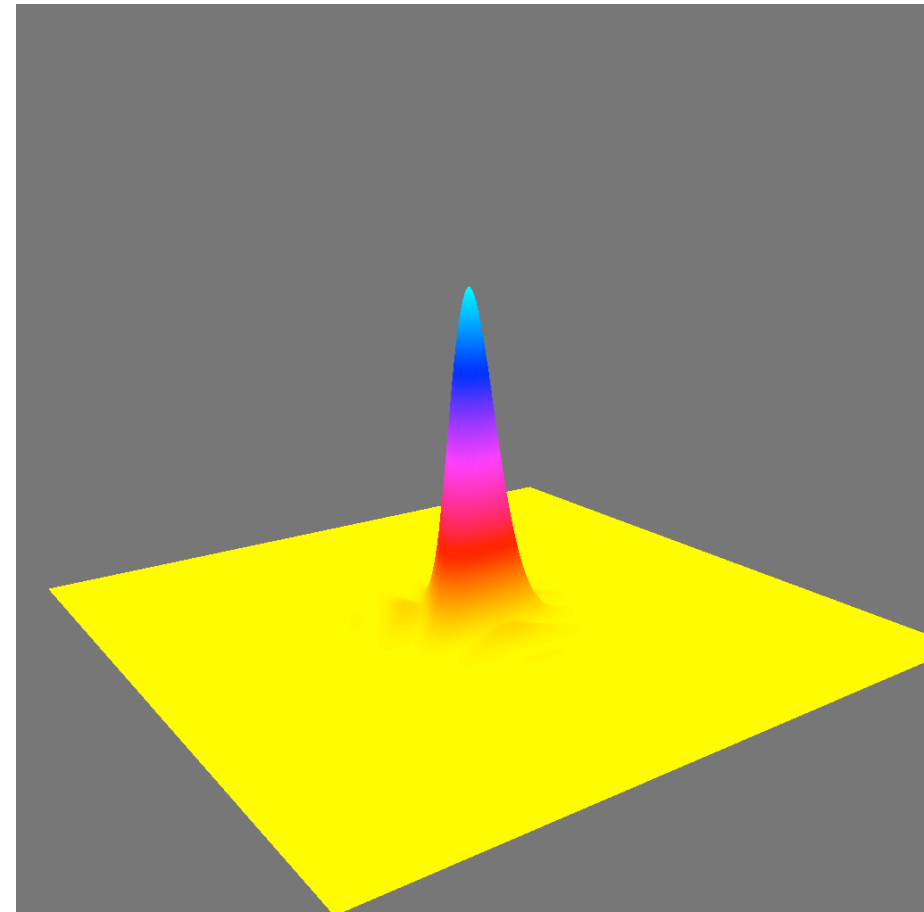
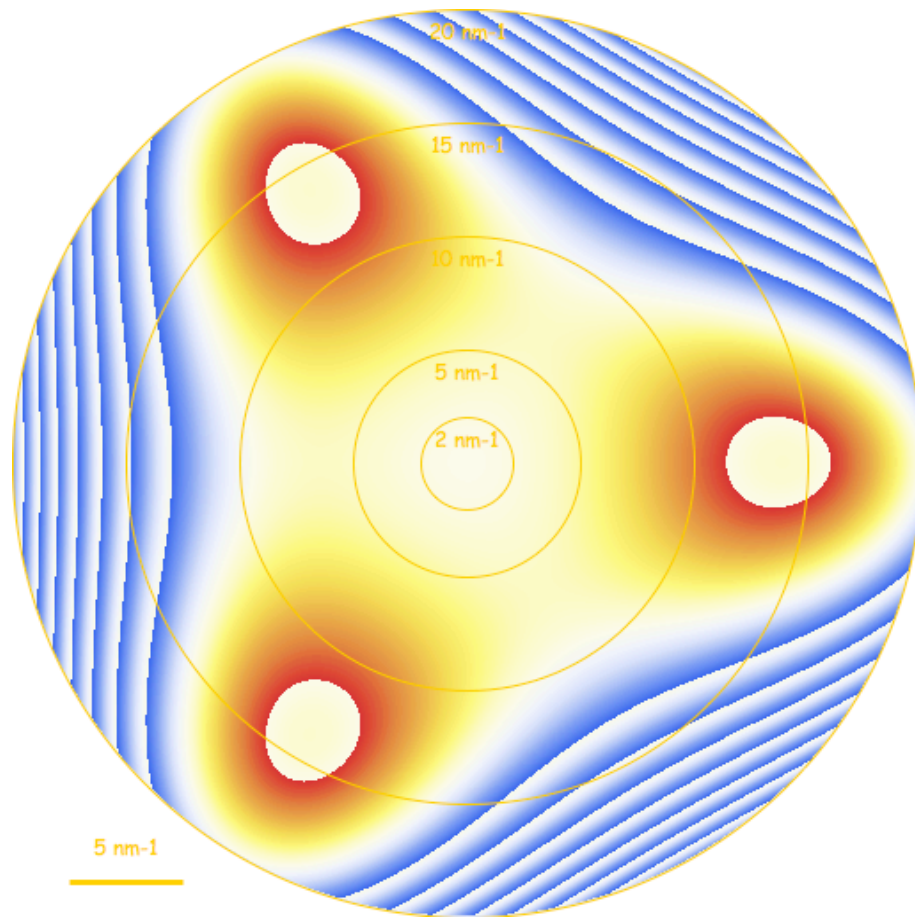
STEM: how to calculate $I_o(\vec{x})$?

JEMS: STEM probe formation and aberrations

The screenshot shows the JEMS software interface for configuring a STEM probe. The left panel displays a diffraction pattern with a central three-lobed probe and a scale bar of 0.1 nm. The right panel shows a list of aberrations with sliders for amplitude and angle. The C_{43} aberration is highlighted in red, and its value is set to 931.0 micrometers.

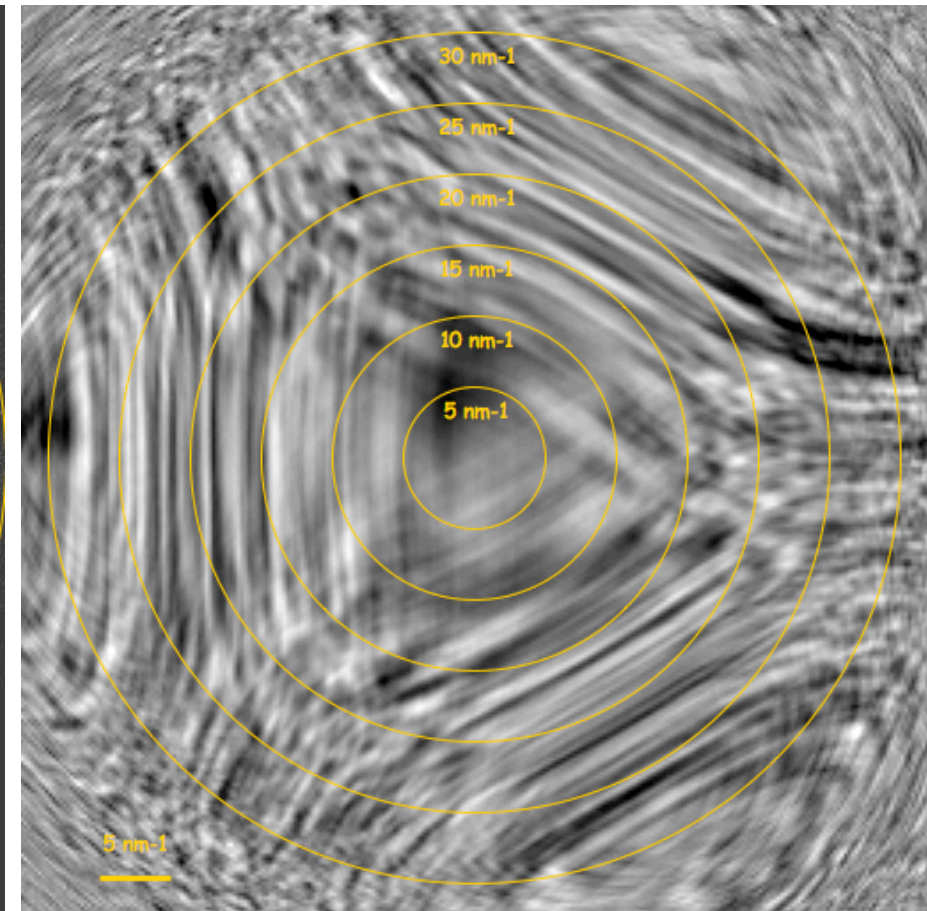
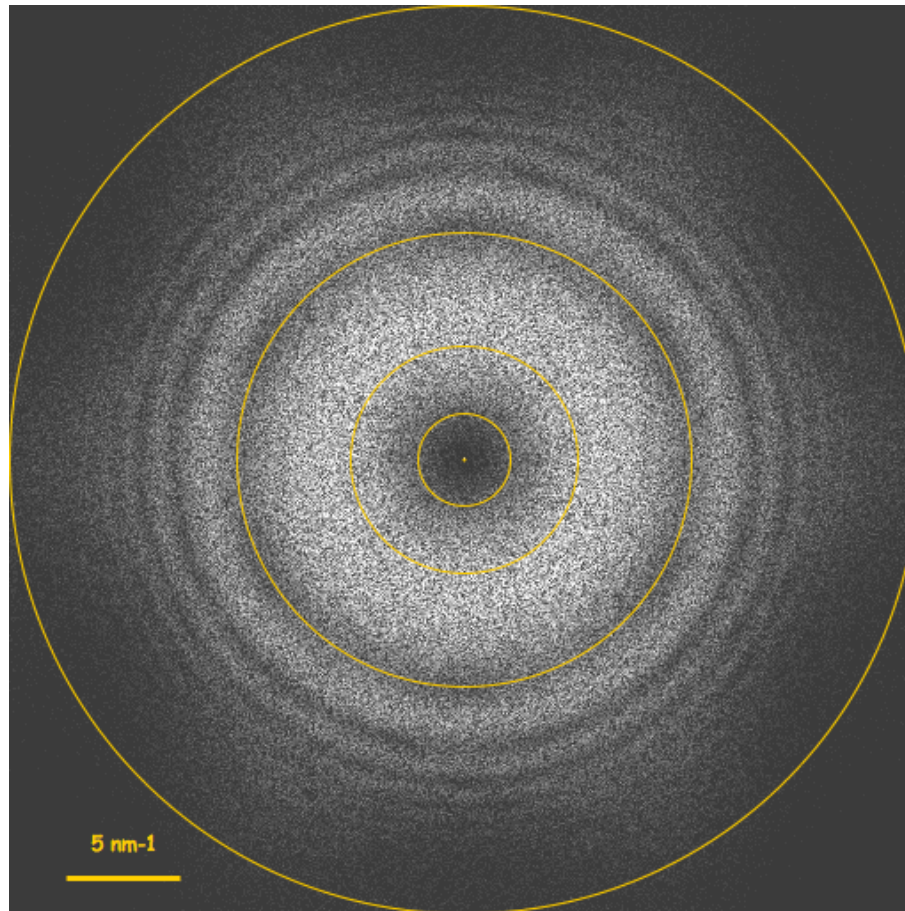
Aberration	Amplitude	Angle
C_c (C_c W_{00}) [mm]	0.0	
C_{01} (I W_{11}) [nm]	0.0	0.0
C_{10} (Z W_{20}) [nm]	-5.8	
C_{12} (A_1 W_{22}) [nm]	0.0	0.0
C_{21} ($3B_2$ W_{31}) [nm]	0.0	0.0
C_{23} (A_2 W_{33}) [nm]	0.0	0.0
C_{30} (C_3 W_{40}) [mm]	-0.03	
C_{32} ($4S_3$ W_{42}) [μ m]	0.0	0.0
C_{34} (A_3 W_{44}) [μ m]	0.0	0.0
C_{41} ($5B_4$ W_{51}) [μ m]	0.0	0.0
C_{43} ($5D_4$ W_{53}) [μ m]	931.0	49.7
C_{45} (A_4 W_{55}) [μ m]	0.0	0.0
C_{50} (C_5 W_{60}) [mm]	5.0	
C_{52} ($6S_5$ W_{62}) [mm]	0.0	0.0
C_{54} ($6R_5$ W_{64}) [mm]	0.0	0.0
C_{56} (A_5 W_{66}) [mm]	0.0	0.0
C_{61} ($7B_6$ W_{71}) [mm]	0.0	0.0
C_{63} ($7D_6$ W_{73}) [mm]	0.0	0.0
C_{65} ($7F_6$ W_{75}) [mm]	0.0	0.0
C_{67} (A_6 W_{77}) [mm]	0.0	0.0
C_{70} (C_7 W_{80}) [m]	0.0	
C_{72} ($8S_7$ W_{82}) [m]	0.0	0.0
C_{74} ($8R_7$ W_{84}) [m]	0.0	0.0
C_{76} ($8G_7$ W_{86}) [m]	0.0	0.0
C_{78} (A_7 W_{88}) [m]	0.0	0.0

STEM probe with C_{43} geometrical aberration (Krivanek) or W_{53} wavefront aberration or 4th order three-lobe (Haider).



Wavefront and STEM probe with C_{23} geometrical aberration (Krivanek), W_{33} wavefront aberration or A_2 three-fold astigmatism (Haider).

Aberrations can be measured



Diffractogram (TEM) and Ronchigram (STEM) (C_{23} geometrical aberration (Krivanek), W_{33} wavefront aberration or A_2 three-fold astigmatism (Haider)).

Please have jems installed on your PC or Mac before the labs (from the website below). I'll also provide jems on USB keys in case you can't download it.

<http://www.jems-saas.ch/>

http://www.jems-saas.ch/jemsv4_4831u2016.htm

3 versions are available for download:

1. MacOS-X 10.7 to 10.12 (Sierra)
2. Windows 7 to 10.
3. Linux ubuntu 15.04 or later.