Basics of High Resolution Transmission Electron Microscopy

MAX-PLANCK-GESELLSCHAFT

AC FHI

Outline

1. Image contrast in TEM
   (a) Mass-thickness contrast (Absorption contrast)
   (b) Diffraction contrast
   (c) Phase contrast

2. Abbe interpretation of imaging

3. Defocus and Aberrations ¾ transfer function

4. Weak phase object approximation (WPOA) and contrast transfer function (CTF)

5. Interpretation of image contrast and determination of structure

6. Image simulation

8. Reference

Image contrast in TEM

I. Mass-thickness contrast

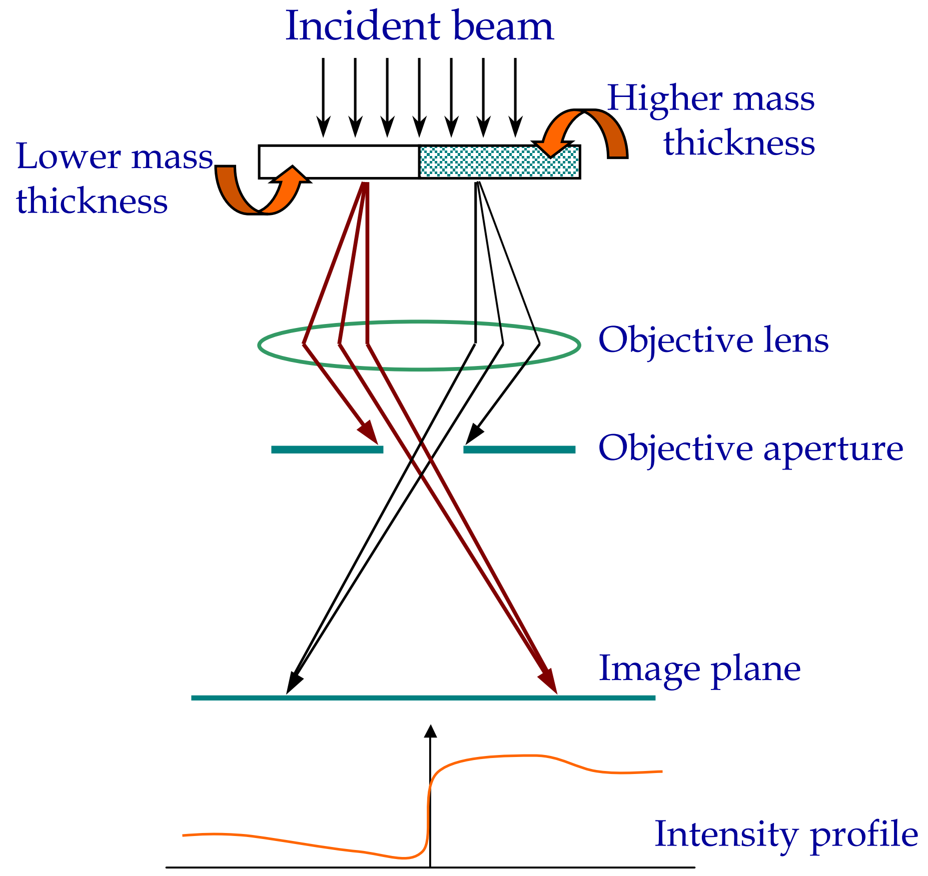
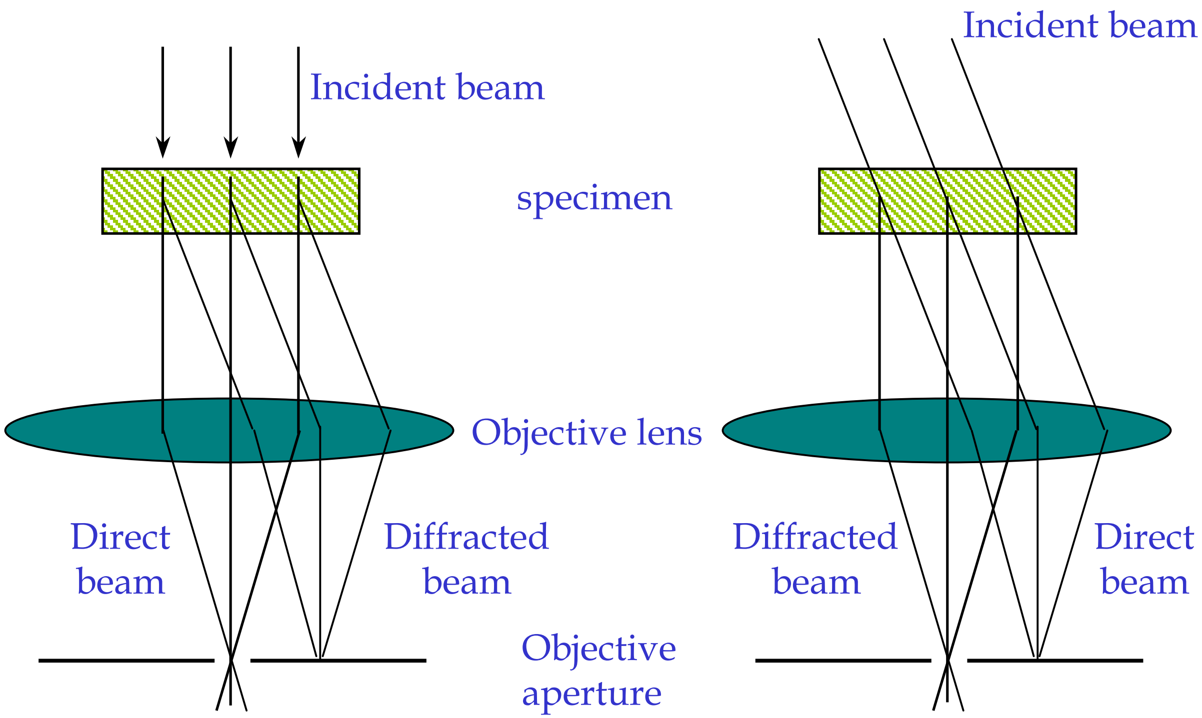


Image contrast in TEM

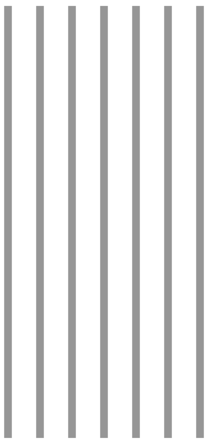
II. Diffraction contrast

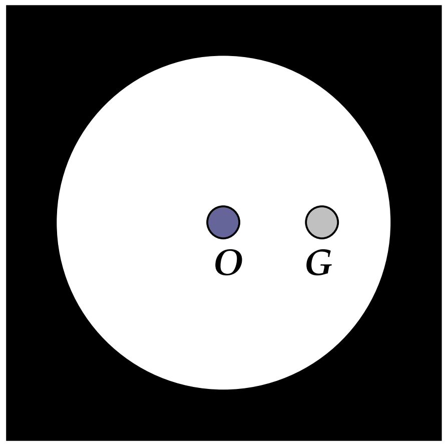


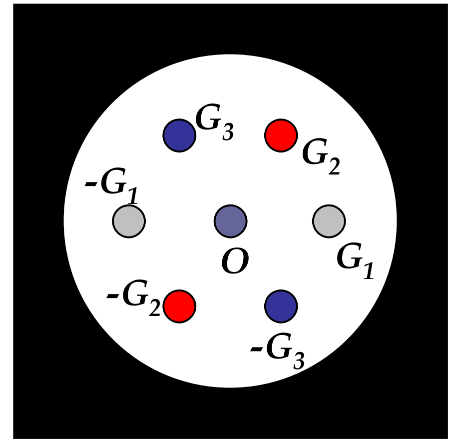
Bright field (BF) **Dark field (DF)**

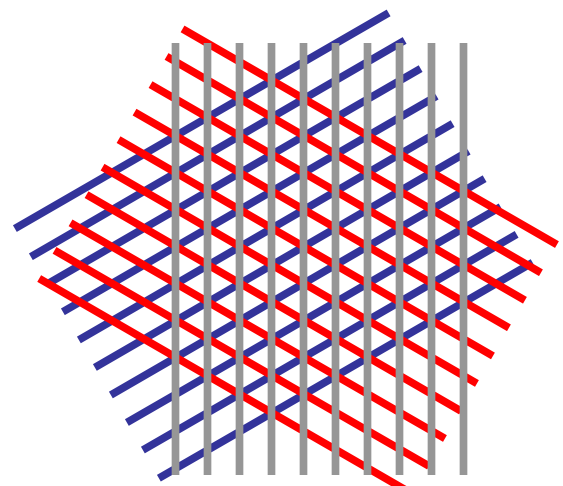
Image contrast in TEM

III. Phase contrast









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Fourier Transform (FT), Inverse FT (IFT) and Convolution

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FT and IFT

$$F(u) = \mathscr{F}\{f(x)\} = \int_{-\infty}^{+\infty} f(x)\exp(2\pi ixu)dx$$

$$f(x) = \mathscr{F}^{-1}\{F(u)\} = \int_{-\infty}^{+\infty} F(u)\exp(-2\pi ixu)du$$

Function of space	FT $\rightleftharpoons$ IFT	Distribution of spatial frequency
Function of time		Distribution of frequency

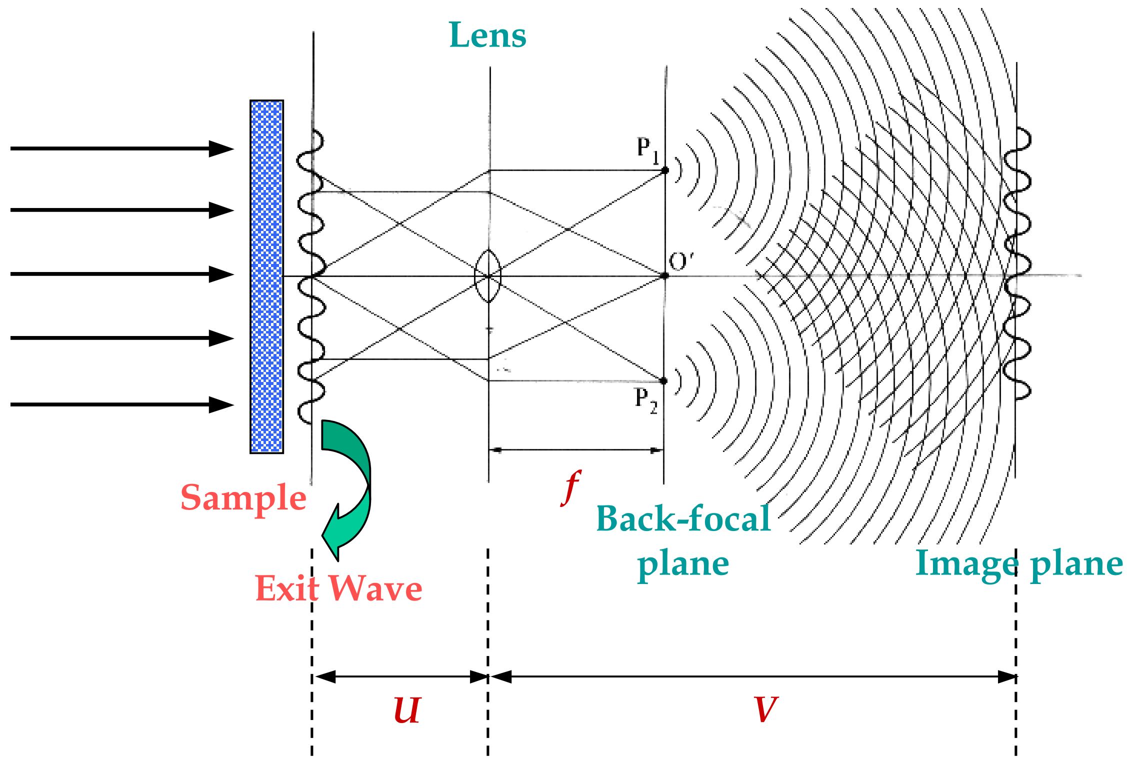
Convolution

$$f(x) * g(x) = \int_{-\infty}^{+\infty} f(X)g(x-X)dX$$

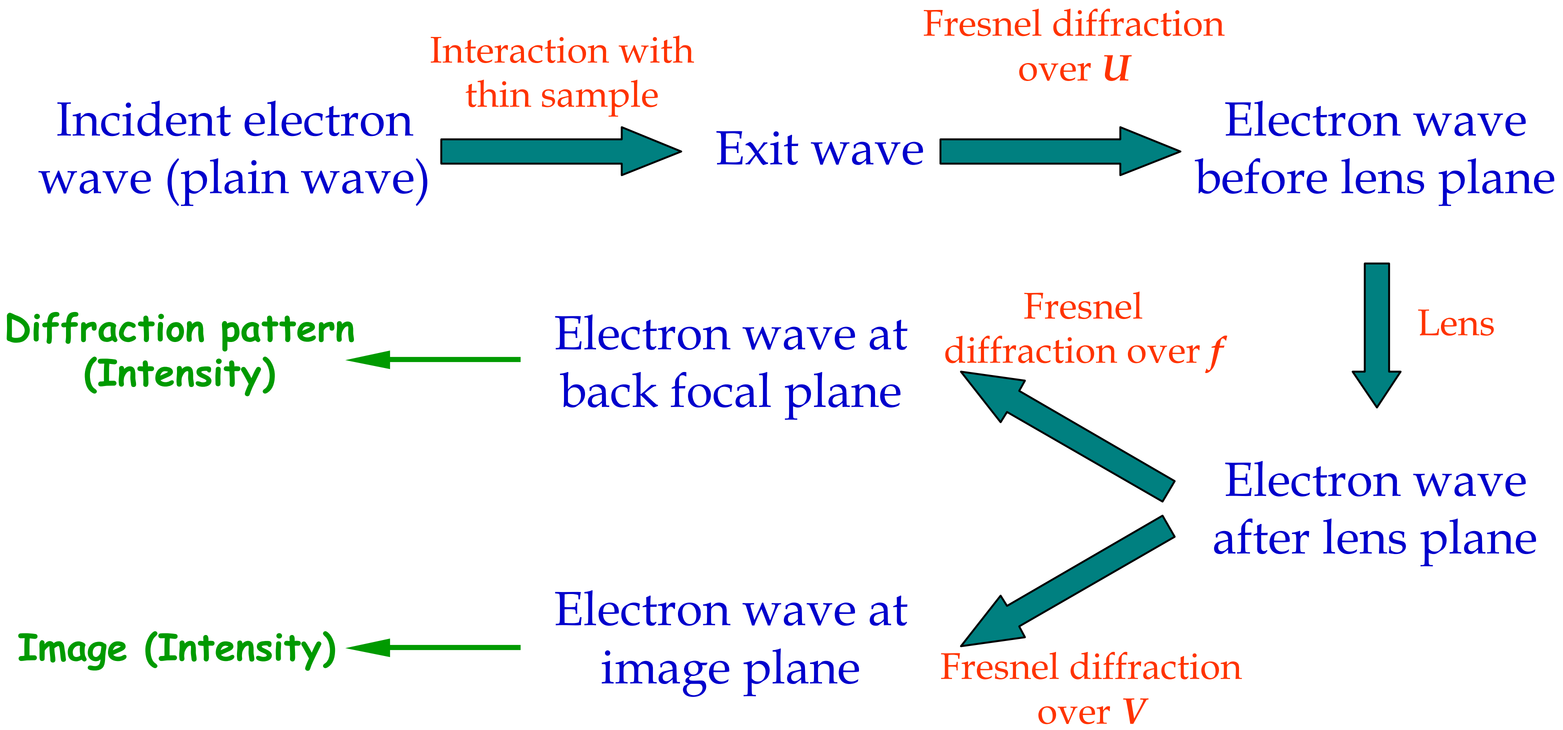
Important Property

$$\mathscr{F}\{f(x) * g(x)\} = \mathscr{F}\{f(x)\} \cdot \mathscr{F}\{g(x)\} = F(u) \cdot G(u)$$

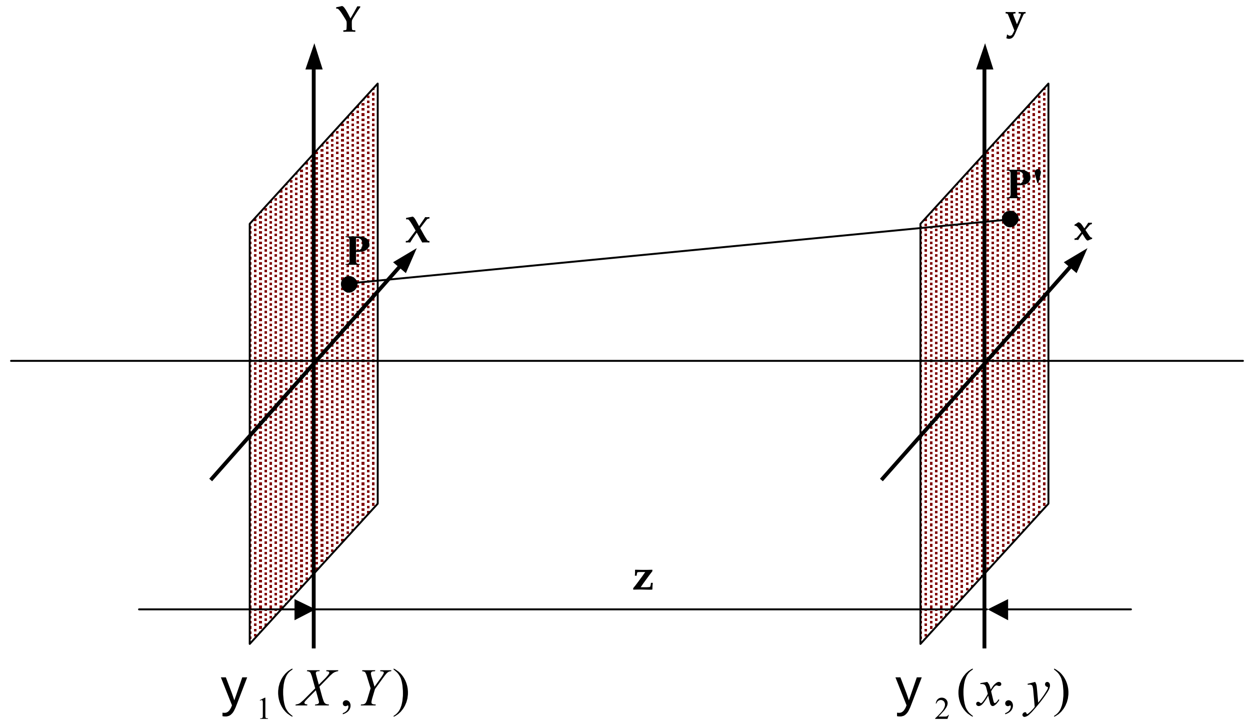
Abbe Interpretation of imaging



Abbe Interpretation of imaging



Fresnel Diffraction



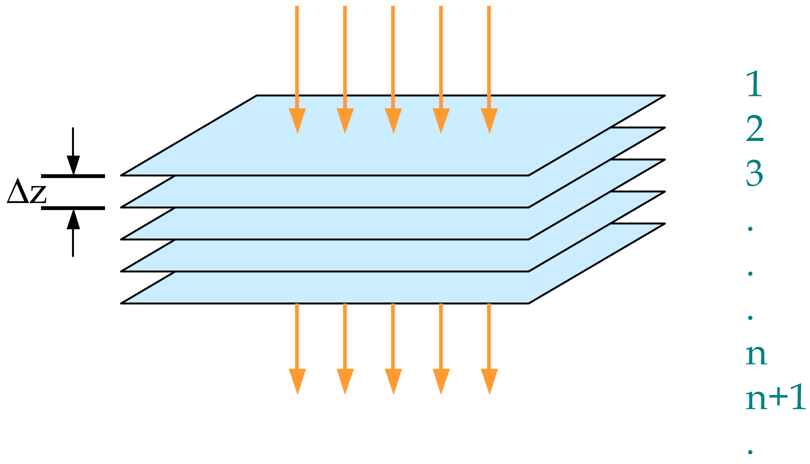
$$\psi_2(x,y) = \frac{i}{\lambda z}\iint \psi_1(X,Y)\exp\left(-\frac{2\pi i[(x-X)^2+(y-Y)^2+z^2]^{1/2}}{\lambda}\right)dXdY$$

$$\approx \frac{i\exp(-2\pi i z/\lambda)}{\lambda z}\iint \psi_1(X,Y)\exp\{-\frac{i\pi}{\lambda z}[(x-X)^2+(y-Y)^2]\}dXdY$$

$$= A\psi_1(x,y) * \exp\{-\frac{i\pi}{\lambda z}[x^2+y^2]\}$$

Propagation Factor $p_z(x,y)$

Propagation of electron wave in crystal



Multislice Method

- Transmission through each slice
- Fresnel diffraction between slices

$y_n(x,y)$: Electron wave function at the exit plane of crystal

Electron wave function in image plane (ideal lens system)

According to Abbe theory

Trasmission function of lens

$$L(x,y) = \exp[\frac{\pi i(x^2 + y^2)}{f\lambda}]$$

$$\psi_{back\_focal}(x,y) = \{[\psi_{exit}(x,y) * \underline{p_U(x,y)}]L(x,y)\} * \underline{p_f(x,y)}$$

$$\psi_{image}(x,y) = \{[\psi_{exit}(x,y) * p_U(x,y)]L(x,y)\} * \underline{p_V(x,y)}$$

Fresnel propagation factor over distance of *U*, *f*, and *V*, seperately

!

$$\psi_{back\_focal}(u,v) \sim A\exp(i\varphi_1(x,y))\mathscr{F}\{\psi_{exit}\}$$

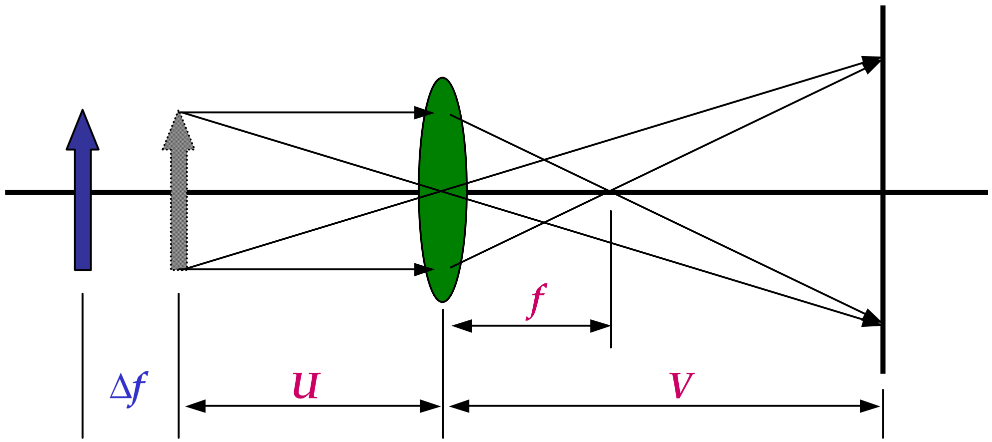
$$\psi_{image}(u,v) \sim B\exp(i\varphi_2(x,y))\psi_{exit}(-\frac{U}{V}(x,y))$$

when $\frac{1}{U} + \frac{1}{V} = \frac{1}{f}$

$$(u,v) = \frac{1}{\lambda f}(x,y)\Big|_{back\_focal}$$

Defocus and aberrations

I. Defocus



Propagation factor $p_{\Delta f}(x,y)$

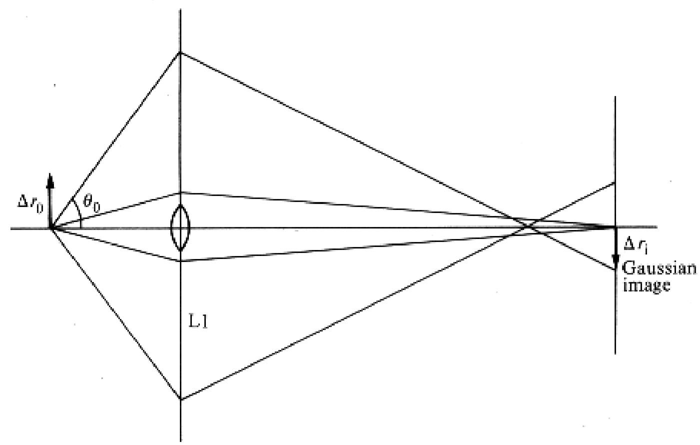
$$\psi_{object}(x,y) = \psi_{exit}(x,y) * \exp\left(\frac{-i\pi(x^2+y^2)}{\lambda \Delta f}\right)$$

$$\psi_{back\_focal}(u,v) = \mathcal{F}\{\psi_{exit}(x,y)\}\mathcal{F}\left\{\exp\left(\frac{-i\pi(x^2+y^2)}{\lambda \Delta f}\right)\right\}$$

$$\mathcal{F}\left\{\exp\left(\frac{-i\pi(x^2+y^2)}{\lambda \Delta f}\right)\right\} = \exp\{i\pi\lambda\Delta f(u^2+v^2)\} = \exp\{i\pi\lambda\Delta f\mathbf{H}^2\} = \exp(i\chi_1)$$

Defocus and aberrations

II Spherical aberration



$$\Delta r_0 = C_s \theta_0^{\,3}$$

C_s : Spherical aberration coefficient

Phase shift in back focal plane due to spherical aberration

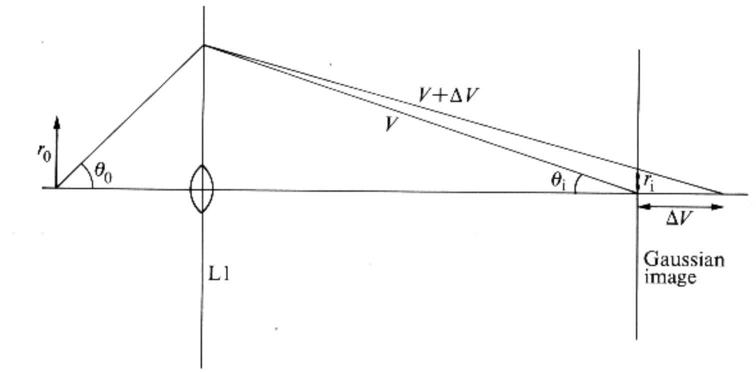
$$\chi_2 = \frac{1}{2}\pi C_s \lambda^3 (u^2+v^2)^2 = \frac{1}{2}\pi C_s \lambda^3 \mathbf{H}^4$$

Factor $\exp(i\chi_2)$

Defocus and aberrations

III Chromatic aberration

Faster electrons are brought to a focus beyond the Gaussian image plane.



- **Fluctuations of the acceleration voltage**
- **Fluctuation of lens current**

Spread of focal length

$$\Delta f = C_c\left(\frac{\Delta V_0}{V_0} - \frac{2\Delta I}{I}\right)$$

D: Standard deviation of Gaussian distribution due to the chromatic aberration

Envelope in back focal plane $\exp(-c_3)$

$$c_3 = \frac{1}{2} p^2 l^2 D^2 \mathbf{H}^2$$

Defocus and aberrations

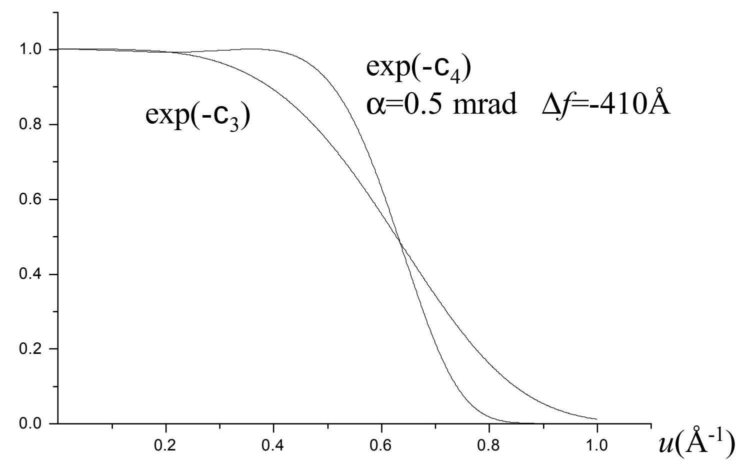
IV Beam divergence

Paralell incident beam (ideal condition)
Divergence angle a~ 0.5 mrad (real condition)

Envelope in back focal plane

$$\exp(-c_4)$$

$$c_4 = p^2 a^2 \mathbf{H}^2 (C_s l^2 \mathbf{H}^2 + \Delta f)^2$$



Transfer function

$$W(\mathbf{H}) = \exp(i\chi_I)\exp(-\chi_{II})$$

Complex transfer function

$$\chi_I = \chi_1 + \chi_2 = \frac{1}{2}\pi C_s \lambda^3 \mathbf{H}^4 + \pi\lambda\Delta f \mathbf{H}^2$$

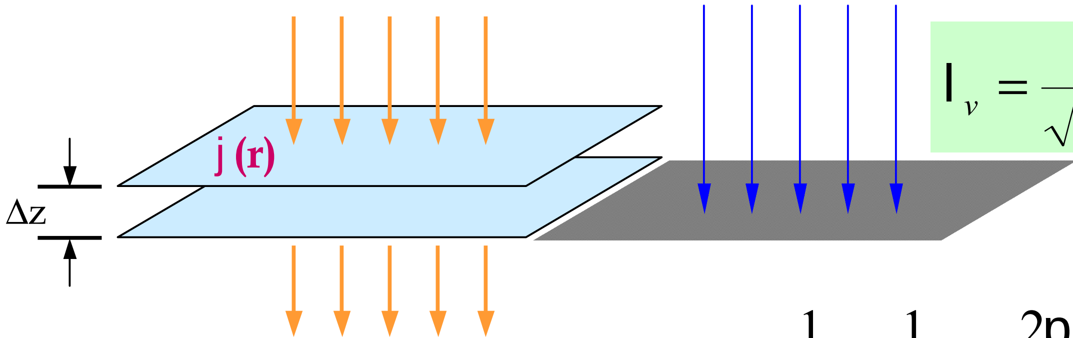
$$\chi_{II} = \chi_3 + \chi_4 = \frac{1}{2}\pi^2\lambda^2 D^2 \mathbf{H}^2 + \pi^2\alpha^2\mathbf{H}^2(C_s\lambda^2\mathbf{H}^2 + \Delta f)^2$$

Electron wave function and intensity in the image plane

$$\psi_{image} = \mathscr{F}^{-1}\{\mathscr{F}[\psi_{exit}]\cdot W(\mathbf{H})\}$$

$$I = \psi_{image}\cdot\psi^*_{image}$$

Weak phase object approximation (WPOA)



$$l_v = \frac{h}{\sqrt{2meU}}$$

$$l_c = \frac{h}{\sqrt{2me(U + j(\mathbf{r}))}}$$

$$\Delta\Phi = 2p\Delta z\left(\frac{1}{l_c} - \frac{1}{l_v}\right) = \frac{2p\Delta z}{h}\left(\sqrt{2me(U + j(\mathbf{r}))} - \sqrt{2meU}\right)$$

$$\approx \frac{2p\Delta z}{h}\sqrt{2meU}\,\frac{j(\mathbf{r})}{2U} = \frac{p}{lU}\,j(\mathbf{r})\Delta z = s\,j(\mathbf{r})\Delta z$$

Transmission function $q(\mathbf{r}) = \exp\{-is\,j(\mathbf{r})\Delta z - m(\mathbf{r})\Delta z\}$

For small Dz, m(r) ® 0 $q(\mathbf{r}) \approx \exp\{-is\,j(\mathbf{r})\Delta z\}$ **Phase object**

$$s\,j(\mathbf{r})\Delta z = \frac{p}{lU}\,j(\mathbf{r})\Delta z << 1$$ **WPOA**

$$q(\mathbf{r}) \approx \exp\{-is\,j(\mathbf{r})\Delta z\} \approx 1 - is\,j(\mathbf{r})\Delta z = 1 - is\,j(x, y)$$

Image intensity for WPOA

$$\psi_{image} = \mathscr{F}^{-1}\{\mathscr{F}[\psi_{exit}]\cdot W(\mathbf{H})\}$$
$$I = \psi_{image}\cdot\psi^{*}_{image}$$

with

$$\psi_{exit} = 1 - i\sigma\varphi(x,y)$$
$$W(\mathbf{H}) = \exp(i\chi_I)\exp(-\chi_{II})$$

$$\begin{aligned}\psi_{image} &= \mathscr{F}^{-1}\{\mathscr{F}[1-i\sigma\varphi(x,y)]\cdot W(\mathbf{H})\}\\ &= \mathscr{F}^{-1}\{\delta(\mathbf{H})\cdot W(\mathbf{H}) - i\sigma F(\mathbf{H})\cdot W(\mathbf{H})\}\\ &= 1 - i\sigma\varphi(x,y) * \mathscr{F}^{-1}\{[\cos\chi_I + i\sin\chi_I]\exp(-\chi_{II})\}\end{aligned}$$

$$\begin{aligned}I(\mathbf{r}) &= [1 - i\sigma\varphi(\mathbf{r}) * \mathscr{F}^{-1}\{(\cos\chi_I + i\sin\chi_I)\exp(-\chi_{II})\}]\\ &\quad\cdot[1 + i\sigma\varphi(\mathbf{r}) * \mathscr{F}^{-1}\{(\cos\chi_I - i\sin\chi_I)\exp(-\chi_{II})\}]\\ &\approx 1 + 2\sigma\varphi(\mathbf{r}) * \mathscr{F}^{-1}\{\sin\chi_I\exp(-\chi_{II})\}\end{aligned}$$

($[\sigma\varphi(\mathbf{r})]^2$ is negligible)

$\sin\chi_I\exp(-\chi_{II})$ **Contrast transfer function (CTF)**

Contrast transfer function (CTF) and Scherzer focus

$$\sin\chi_I \exp(-\chi_{II}) = \sin\left(\frac{1}{2}\pi C_s \lambda^3 u^4 + \pi\lambda\Delta f u^2\right)\exp(-\chi_{II})$$

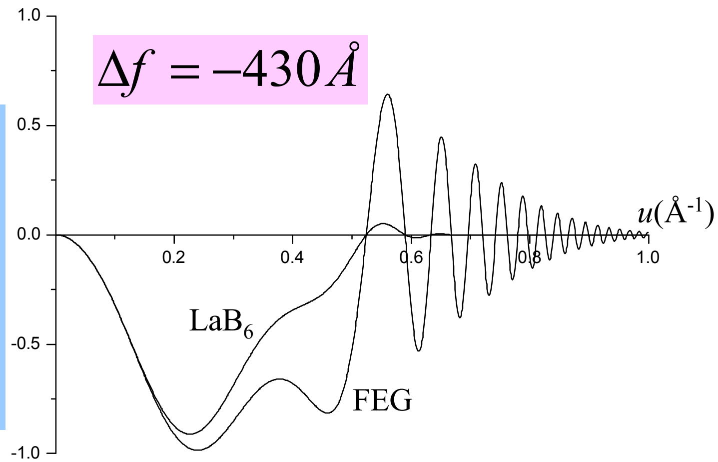
Parameters:

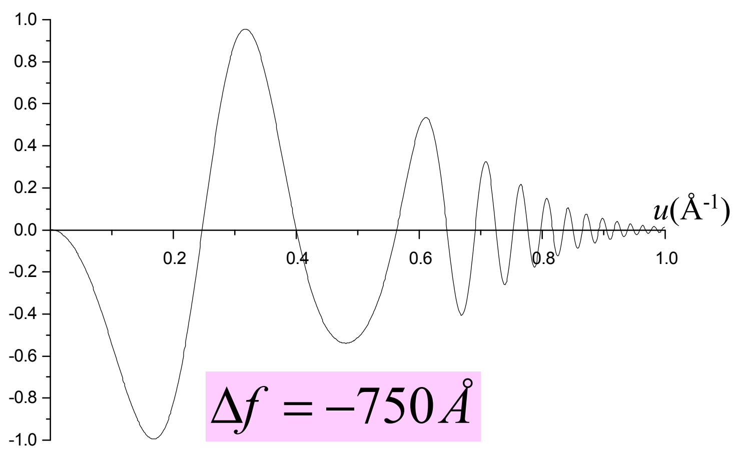
$U = 200kV$

$C_s = 0.5mm$

$D_{FEG} = 38\mathring{A}$

$D_{LaB_6} = 100\mathring{A}$





Scherzer focus

$$\begin{cases} \dfrac{d\chi_I}{du} = 2\pi C_s \lambda^3 u^3 + 2\pi\Delta f \lambda u = 0 \\ \dfrac{1}{2}\pi C_s \lambda^3 u^4 + \pi\Delta f \lambda u^2 = -\dfrac{2}{3}\pi \end{cases}$$

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

Point resolution

$$\frac{1}{2}\pi C_s \lambda^3 u^4 + \pi\Delta f_{Sch}\lambda u^2\Big|_{u\neq 0} = 0$$

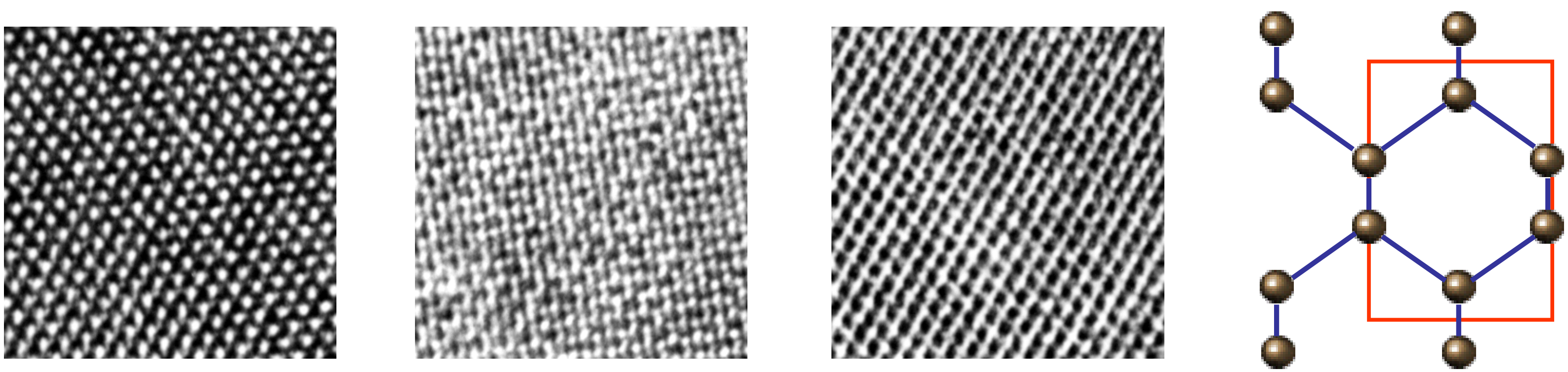
$$\Rightarrow u_0 = 1.51 C_s^{-1/4}\lambda^{-3/4},\quad r_0 = 0.66 C_s^{1/4}\lambda^{3/4}$$

Interpretation of image

$$\Delta f = \Delta f_{Sch}, \sin \chi_I \approx -1\,(0 < u < u_0)$$

$$I(\mathbf{r}) \approx 1 - 2\sigma\varphi(\mathbf{r})$$ **Structure image**

! **Only for thin crystal (WPOA) and the focus value close to Scherzer focus, the contrast of HREM image can be interprated as crystal structure up to point resolution.**
In general, the black or white dots in HREM image DO NOT correspond to atoms or atom groups.



Si [110] image by JEM-2010 FEG electron microscope with different defocus values

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Structure determination

- Go back from image(s) to structure

Through-focus images → **Exit wave** → **Structure**

Deconvolution method → **Projected potential j (x,y)**

Thin crystal and defocus value can be obtained

- Image simulation and matching

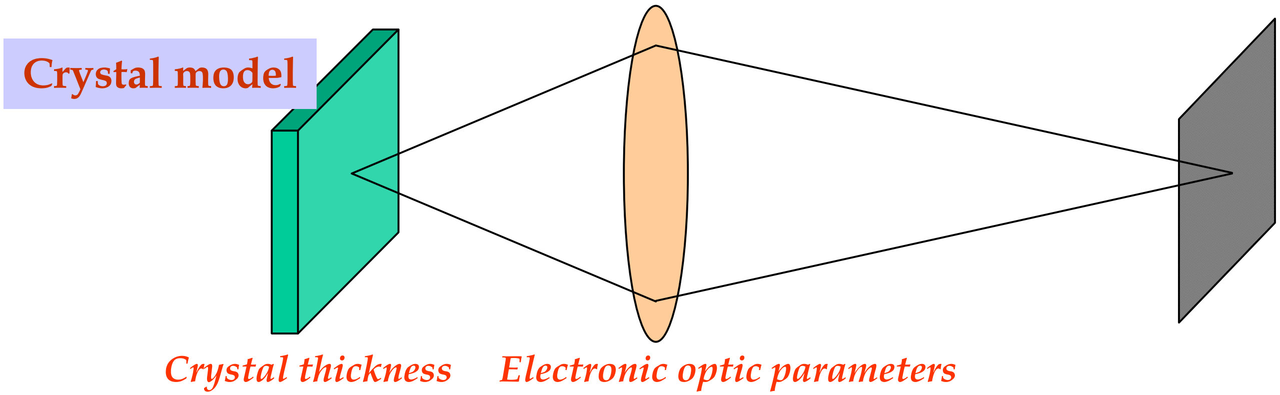
Initial model ← Experimental image

Initial model → Image simulation

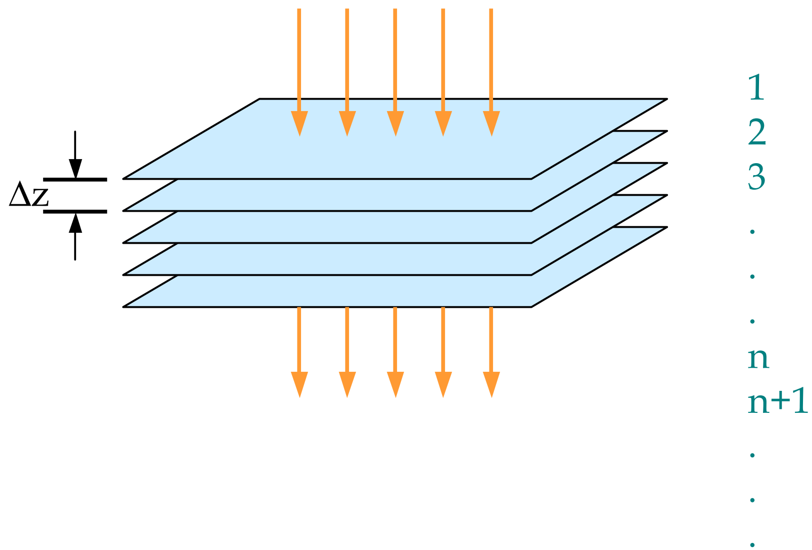
Experimental image + Image simulation → Modified model

Modified model → Image simulation

Image simulation



How to calculate exit wave? ¾ ¾Multislice method



Multislice method

$\psi_n(\mathbf{r})$: **Electron wave function after nth slice**

Then $\psi_{n+1}(\mathbf{r}) = [\psi_n(\mathbf{r}) * p(\mathbf{r})]q_{n+1}(\mathbf{r})$

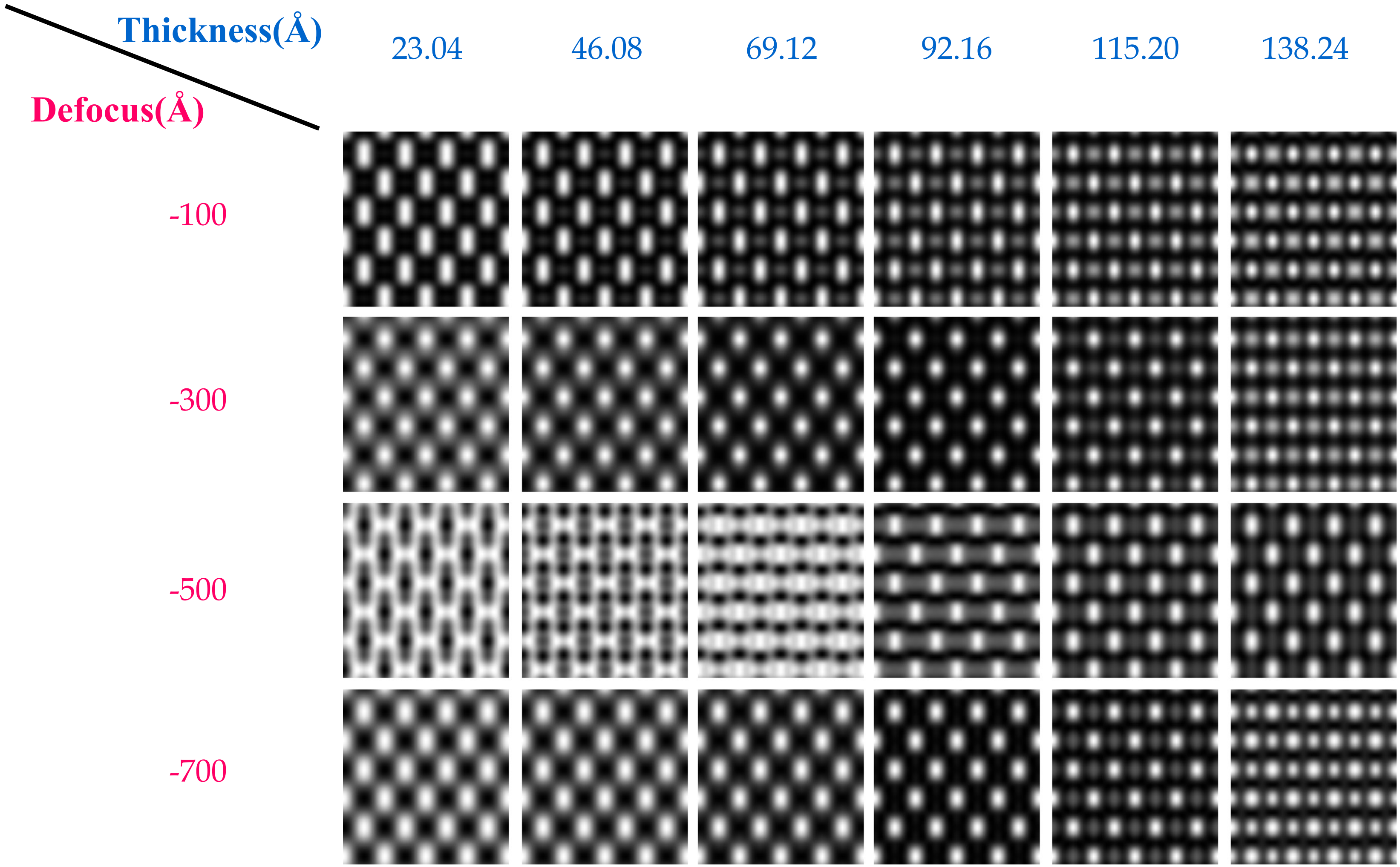
Electron wave function after N slices can be written as:

$$\psi_N(\mathbf{r}) = q_N(\mathbf{r})\underset{N}{[}\cdots\underset{3}{[}q_2(\mathbf{r})\underset{2}{[}q_1(\mathbf{r})\underset{1}{[}q_0(\mathbf{r}) * p_0(\mathbf{r})\underset{1}{]} * p_1(\mathbf{r})\underset{2}{]} * p_3(\mathbf{r})\underset{3}{]} * \cdots\underset{N}{]} * p_N(\mathbf{r})$$

When Δz being the crystal period along incident direction

$$\psi_N(\mathbf{r}) = q(\mathbf{r})\underset{N}{[}\cdots\underset{3}{[}q(\mathbf{r})\underset{2}{[}q(\mathbf{r})\underset{1}{[}q(\mathbf{r}) * p(\mathbf{r})\underset{1}{]} * p(\mathbf{r})\underset{2}{]} * p(\mathbf{r})\underset{3}{]} * \cdots\underset{N}{]} * p(\mathbf{r})$$

Si [110] simulated images



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