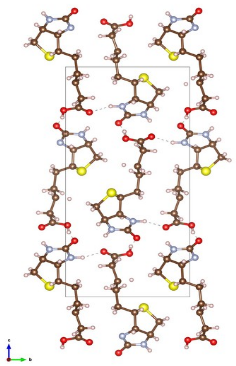
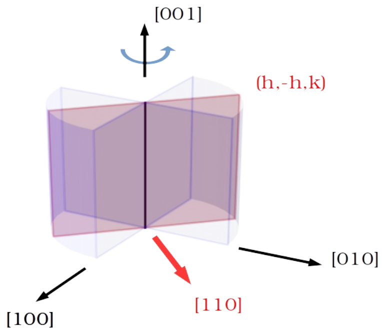
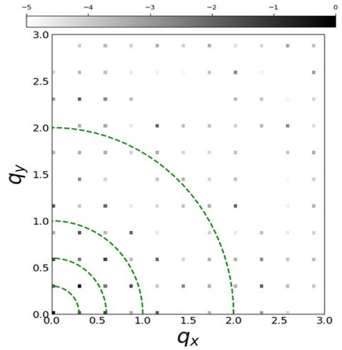
Simulation of Dynamical Scattering Effect in Electron Diffraction Patterns

Tarik Drevon, David Waterman, Eugene Krissinel







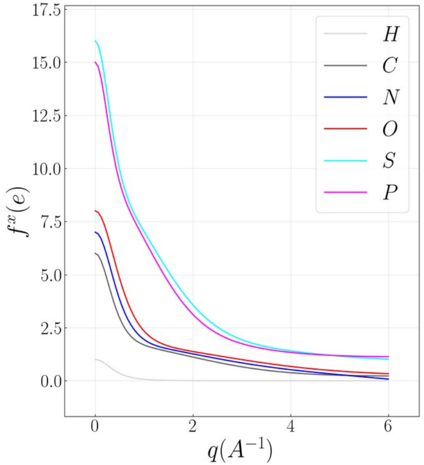
UK Research and Innovation

X-ray (MX) vs electron diffraction (ED) for biotin

MX 12keV ≅ 1Å

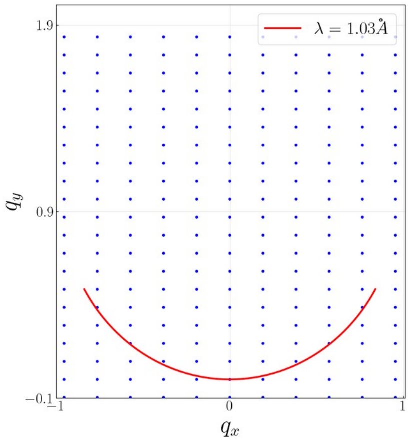
Form factor

- Electronic density
- Thomson scattering
- Small cross section
- Kinematic scattering

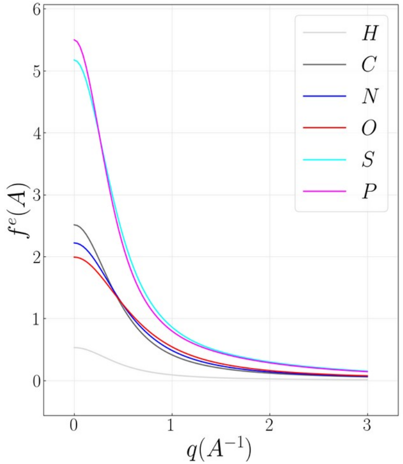


Ewald sphere curvature

- Large curvature
- High order Laue zones
- Thick sample t=20μm
- Narrow rocking curve



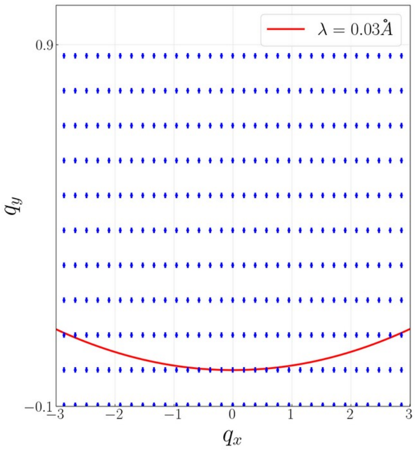
ED 200keV ≅ 0.025Å



Form factor

- Electrostatic potential
- Coulomb scattering
- Large cross section
- Dynamical scattering
- Mott-Bethe formula

$$f^e(q) = \frac{1}{2\pi^2 a_0}\frac{Z - f^x(q)}{q^2}$$

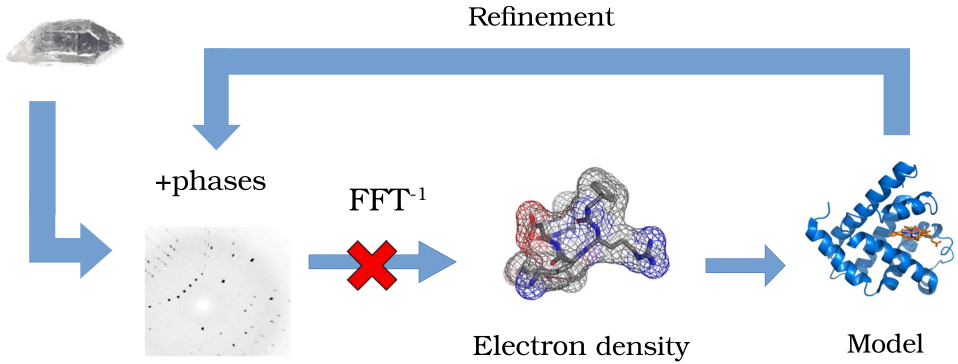


Ewald Sphere curvature

- Flat Ewald sphere
- First order Laue zone
- Thin sample t=0.2μm
- Wide rocking curve
- Thickness dependent rocking curve shape

Numerical simulation tools of ED patterns

Kinematic approximation



Schroedinger's fast electron wave equation

$$\left\{ \frac{\hbar^2}{2m_0}\nabla^2 + V(\mathbf{r}) \right\}\Psi(\mathbf{r}) = E\Psi(\mathbf{r})$$

$$\partial_z^2 \ll 2ik_0\partial_z$$

$$\frac{\partial\Psi(x,y,z)}{\partial_z} = \left\{ \frac{i\lambda}{4\pi}\nabla_{xy}^2 + i\sigma V(x,y,z) \right\}\Psi(x,y,z)$$

Kinematic theory of scattering

Kinematic solution

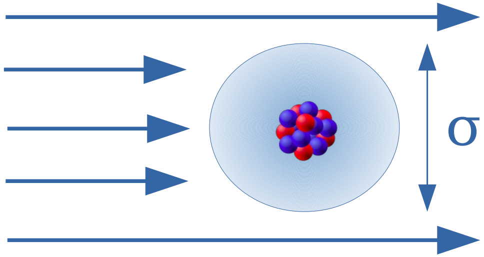
$$\Psi(\mathbf{r}) = e^{ikz} + f(\theta)\frac{e^{i\mathbf{k}\cdot\mathbf{r}}}{|\mathbf{r}|}$$

Born approximation

$$f(\theta) = -\frac{2me}{h^2}\int d^3 r e^{i\mathbf{q}\cdot\mathbf{r}} V(r) \ , \quad \frac{d\sigma}{d\Omega} = |f(\theta)|^2$$

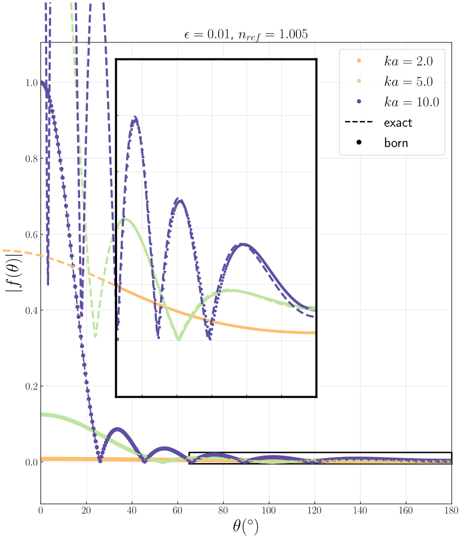
$$P_{scattered} = \sigma \phi_{incident}$$

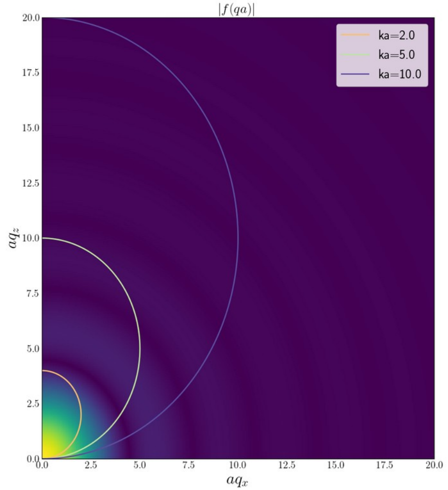
$$q = 2k_0 \sin\frac{\theta}{2}$$



Application to scattering by a sphere

$$f(q) = \frac{V_0}{E}\frac{a^3 k_0^2}{q^2 a^2}\left(-\cos qa + \frac{\sin qa}{qa}\right)$$



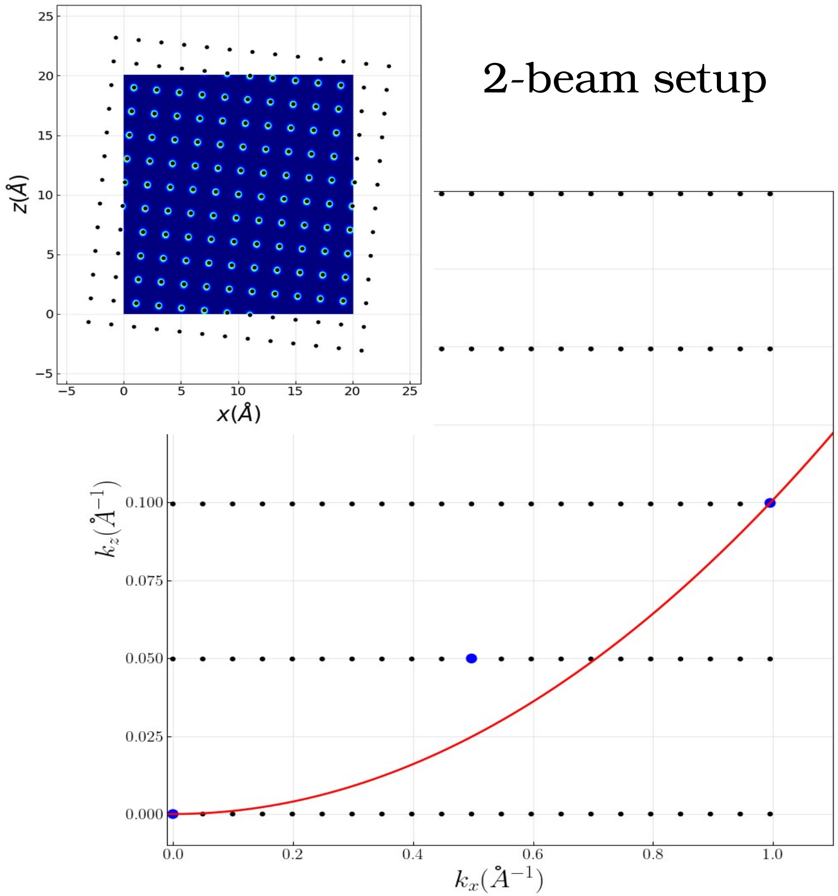


Numerical simulation tools of ED patterns

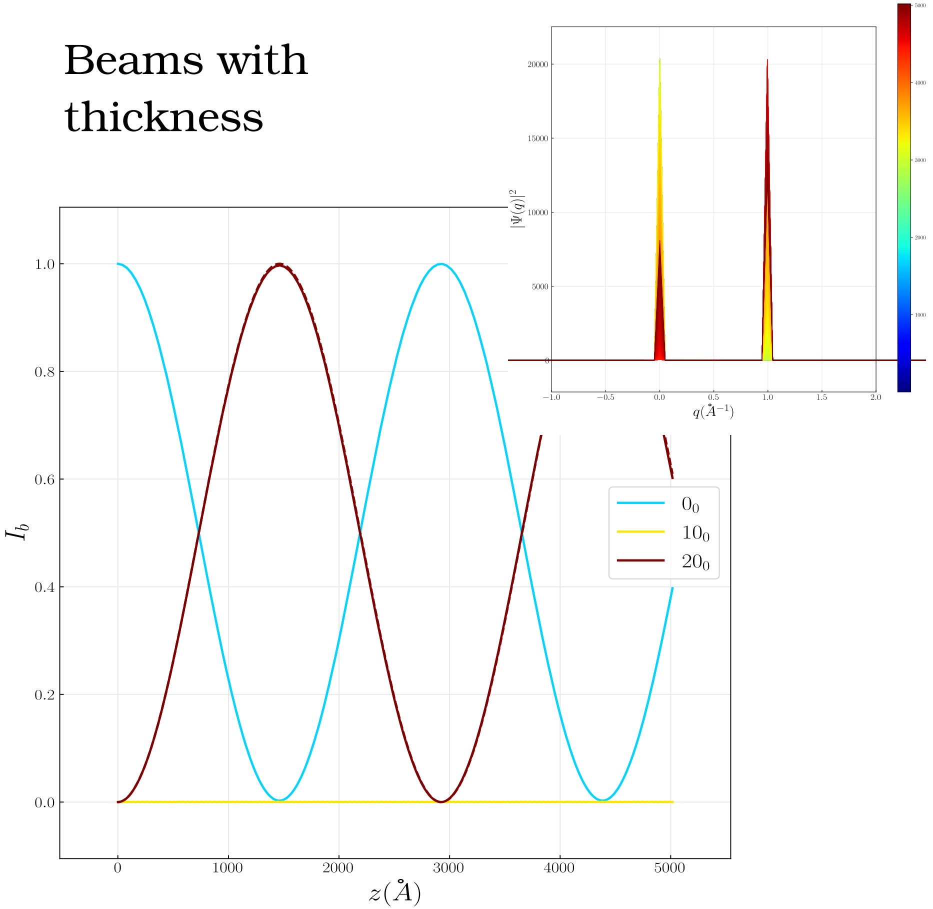
Method	Exact	Speed	Mem (beam per atom)	Periodic structure	Grid based	Parallelization type	Package
Multislice (MS) (physical optics based approach)	no	$N_zN_b\log N_b$	100	**yes**	**yes**	FFTw one slice after another	**TEMSIM (pyMS)** PRISM,…
Near bragg (real space path differences)	no	$N_zN_b^2N_p$	1	**no**	**no**	per pixel	NearBragg (James Holton)

Multislice 2-beam diffraction case

2-beam setup



Beams with thickness

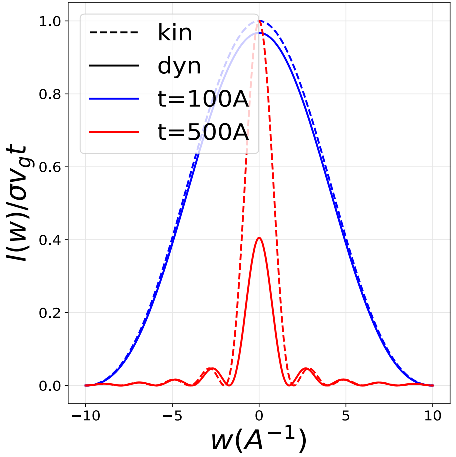


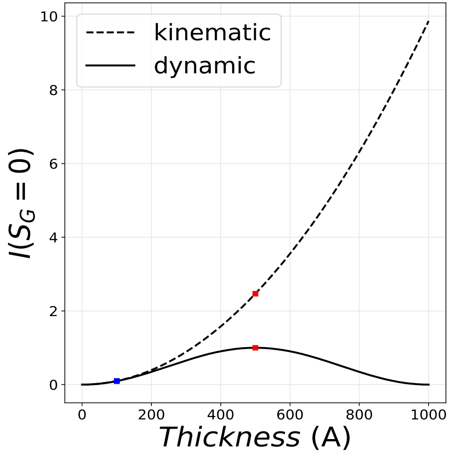
2-beam Rocking Curves

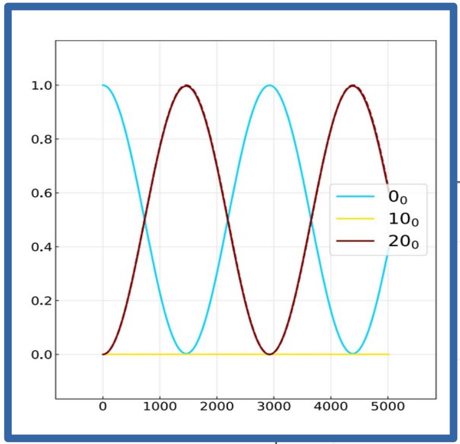
2-beam rocking curve

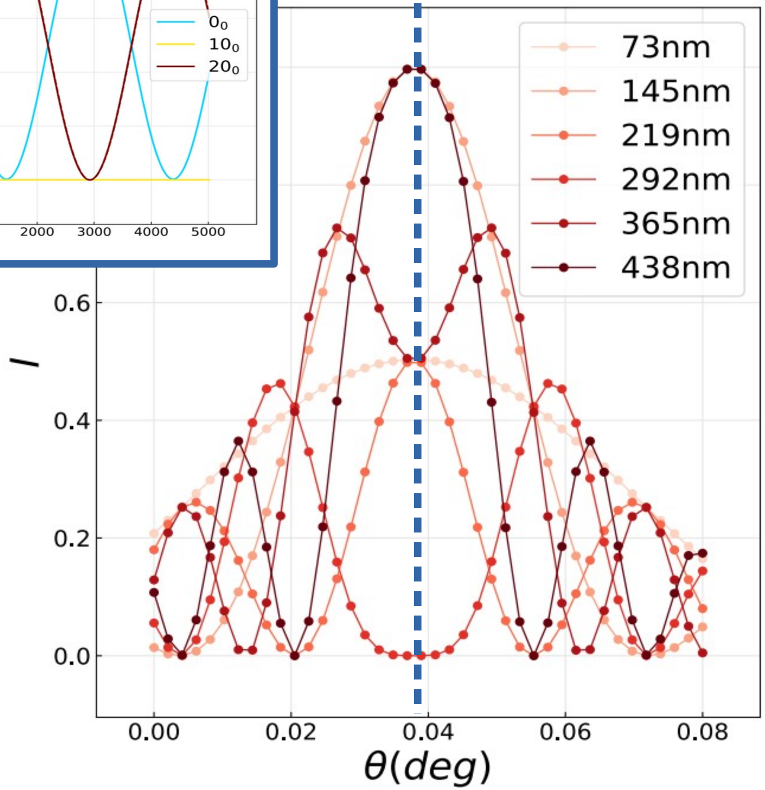
$$I_{dyn-2}(w_g; t, \xi_g) = \left(\frac{\pi t}{\xi_g}\right)^2 sinc^2\left(\frac{t}{\xi_g}\sqrt{1+w_g^2}\right)$$

$$\boxed{I(0; t, \xi_g)} = \left(\frac{\pi t}{\xi_g}\right)^2 sinc^2\left(\frac{t}{\xi_g}\right) \underset{t \ll \xi_g}{\rightarrow} \left(\frac{\pi t}{\xi_g}\right)^2$$

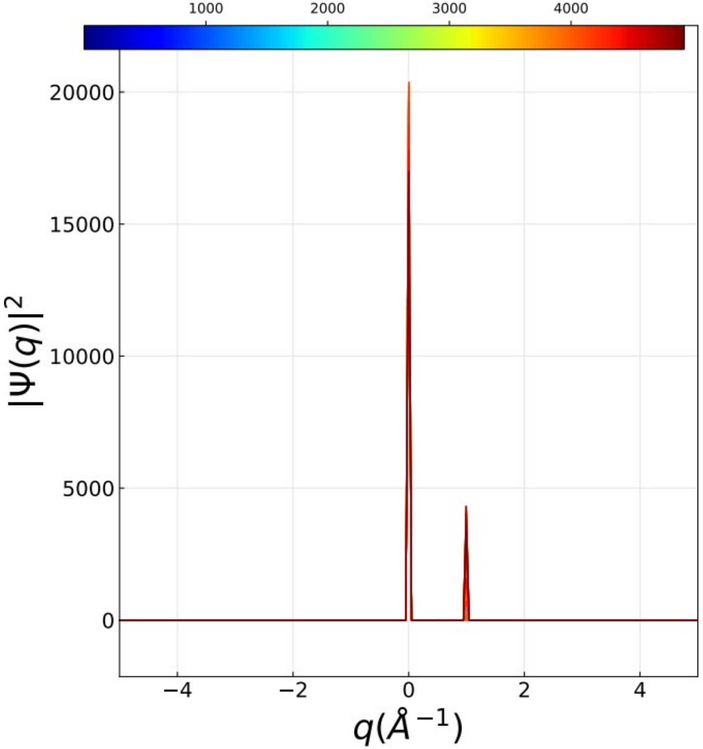




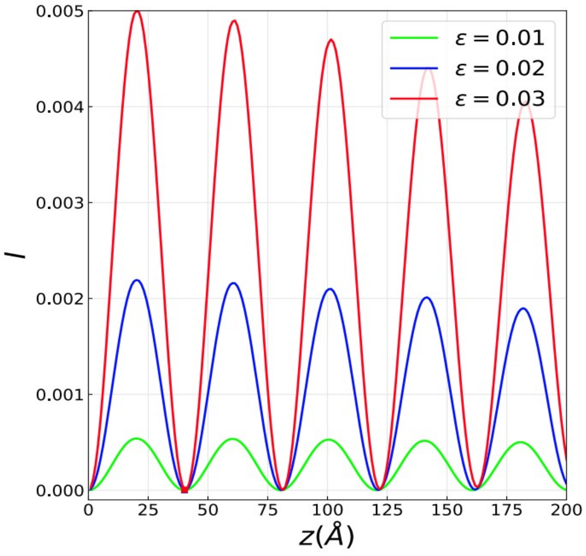




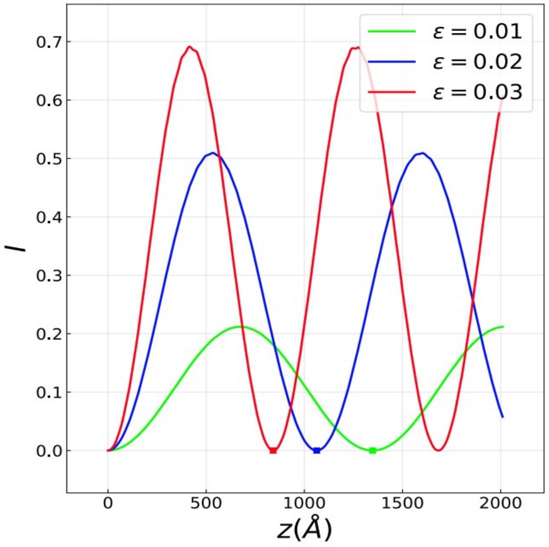
Dynamical diffraction extinction distance



Kinematic Ewald sphere curvature effect weakly diffracted beam



Potential dependent extinction distance for strongly diffracting beam

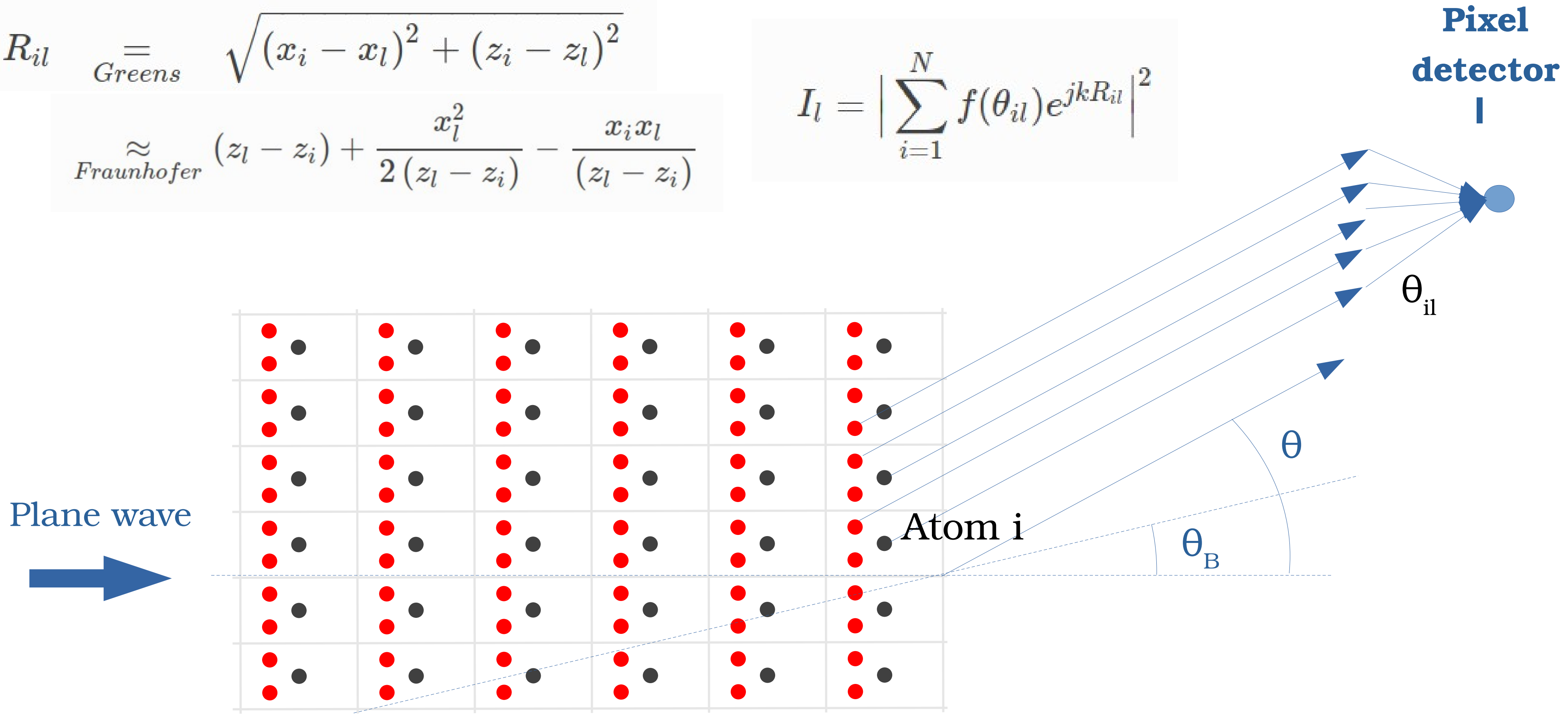


Near Bragg

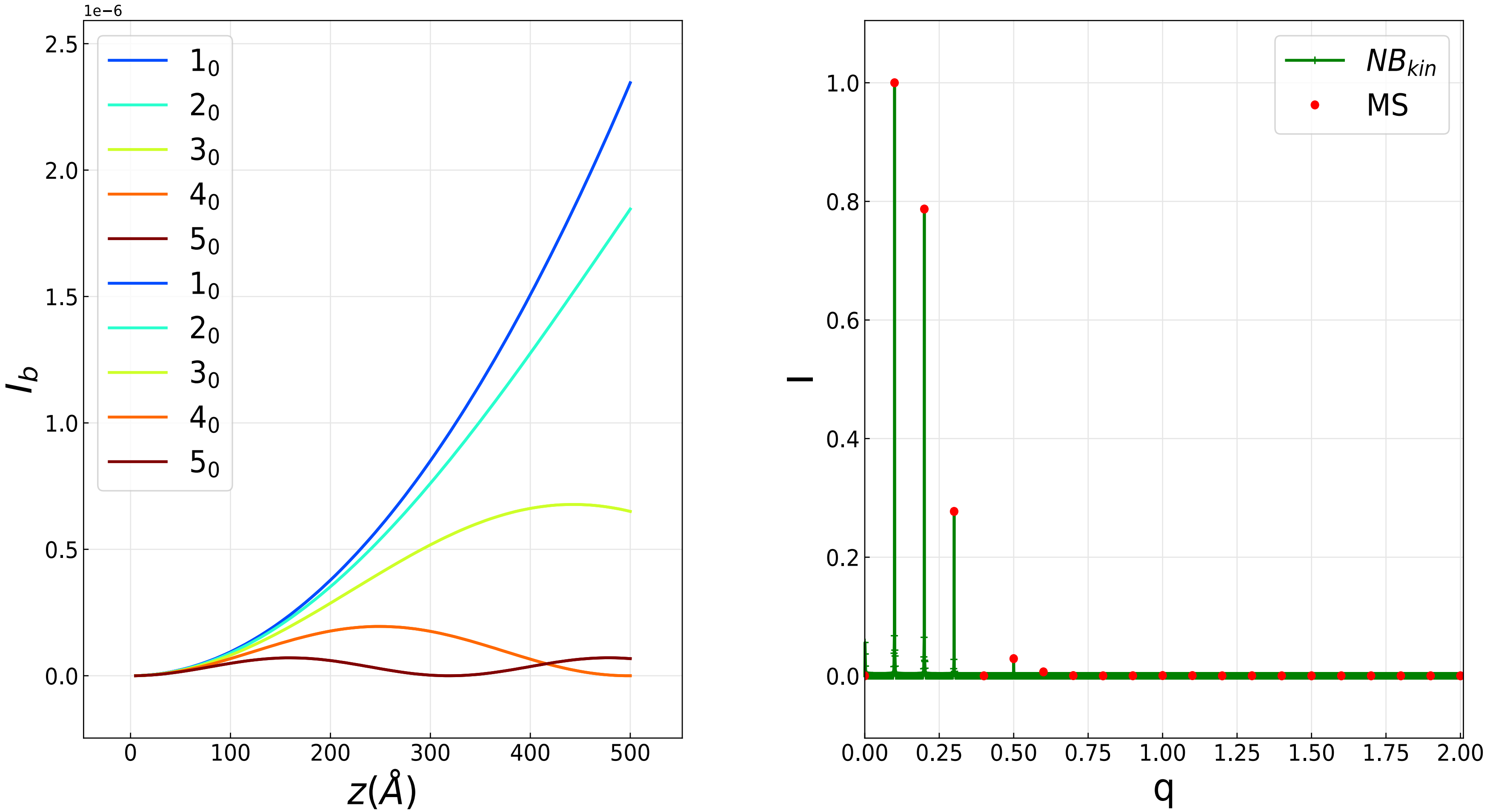
$$R_{il} \underset{Greens}{=} \sqrt{(x_i - x_l)^2 + (z_i - z_l)^2}$$

$$\underset{Fraunhofer}{\approx} (z_l - z_i) + \frac{x_l^2}{2(z_l - z_i)} - \frac{x_i x_l}{(z_l - z_i)}$$

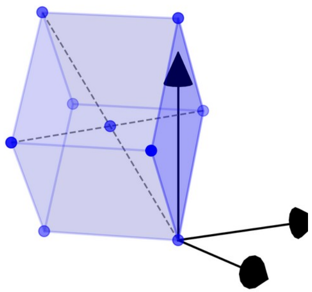
$$I_l = \left| \sum_{i=1}^{N} f(\theta_{il}) e^{jkR_{il}} \right|^2$$



Weak potential kinematic approximation



Diffraction patterns of biotin

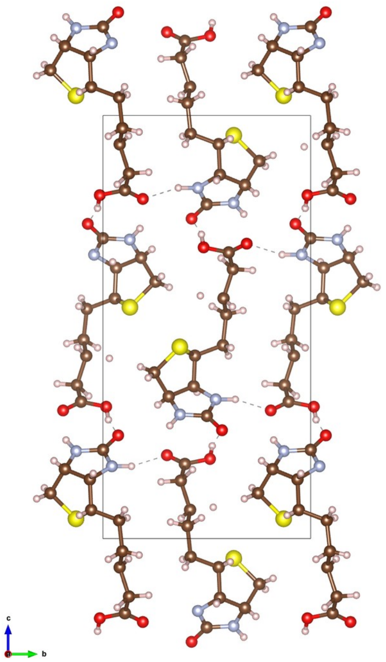


$C_{10}H_{16}N_2O_3S$

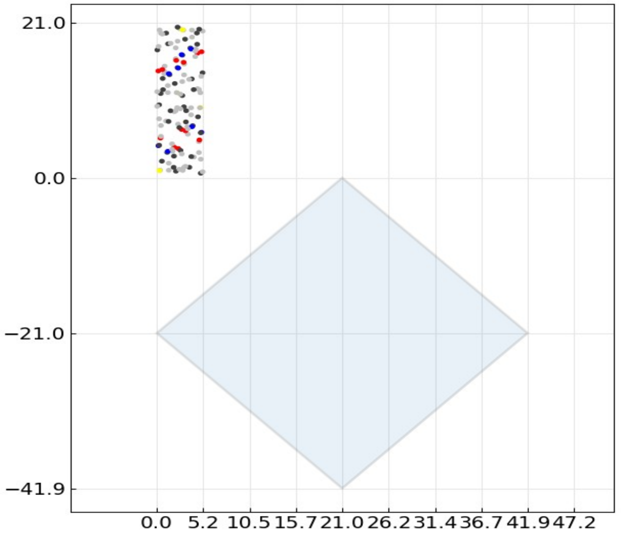
Structure : $P2_12_12_1$

alpha=90, beta=90, gamma=90

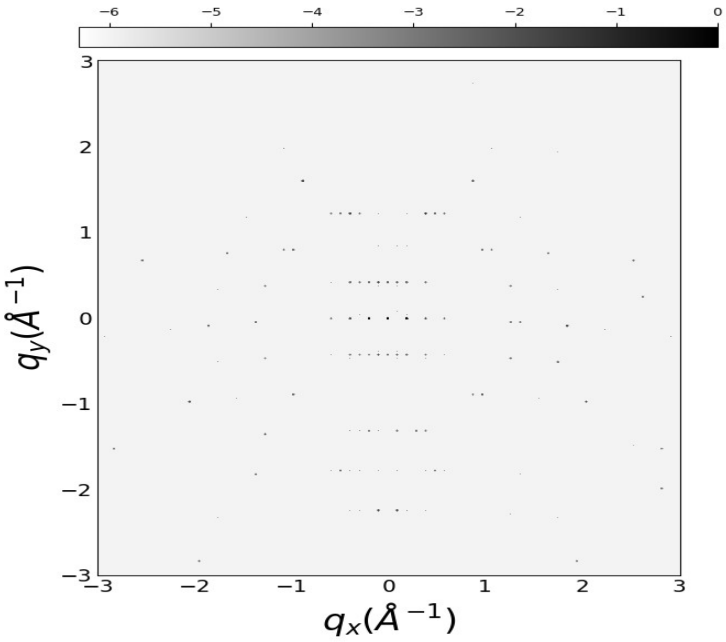
a=5.24A, b=10.35A, c=21.04A



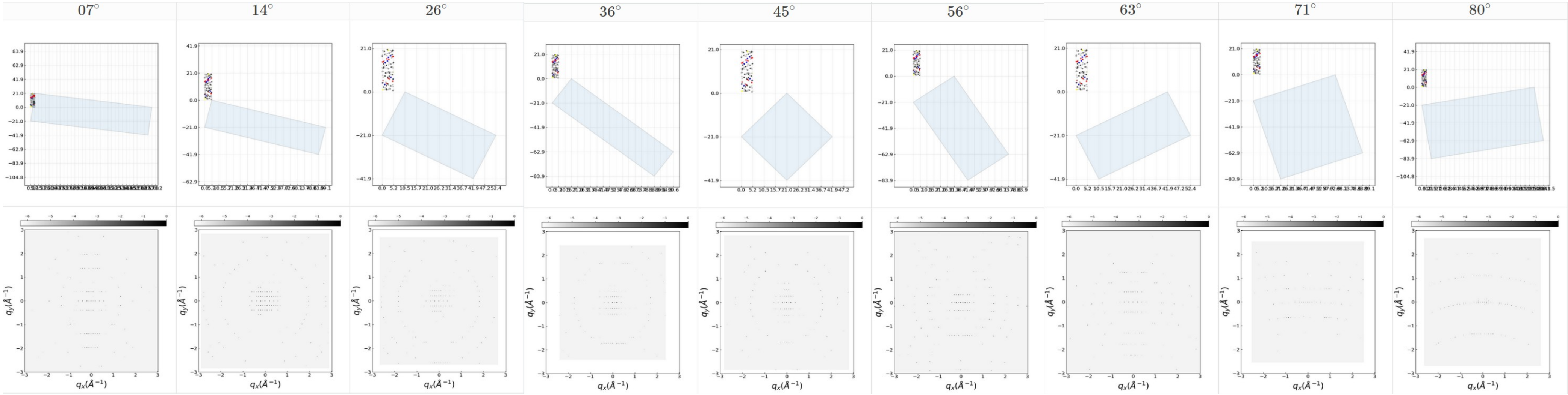
[401]=45deg



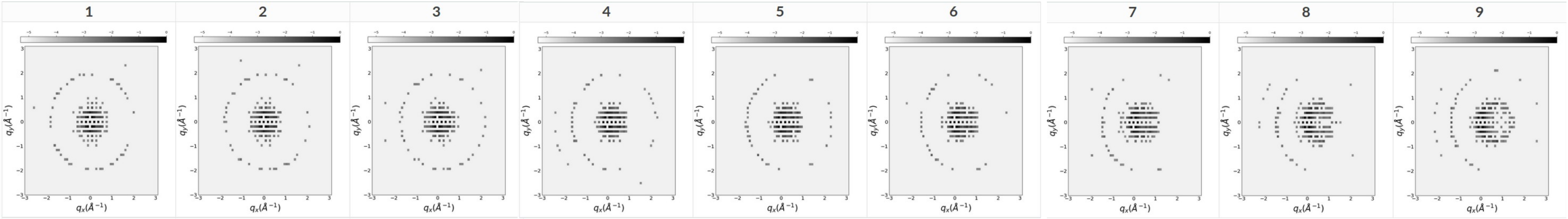
Diffraction pattern



Full rotation simulation

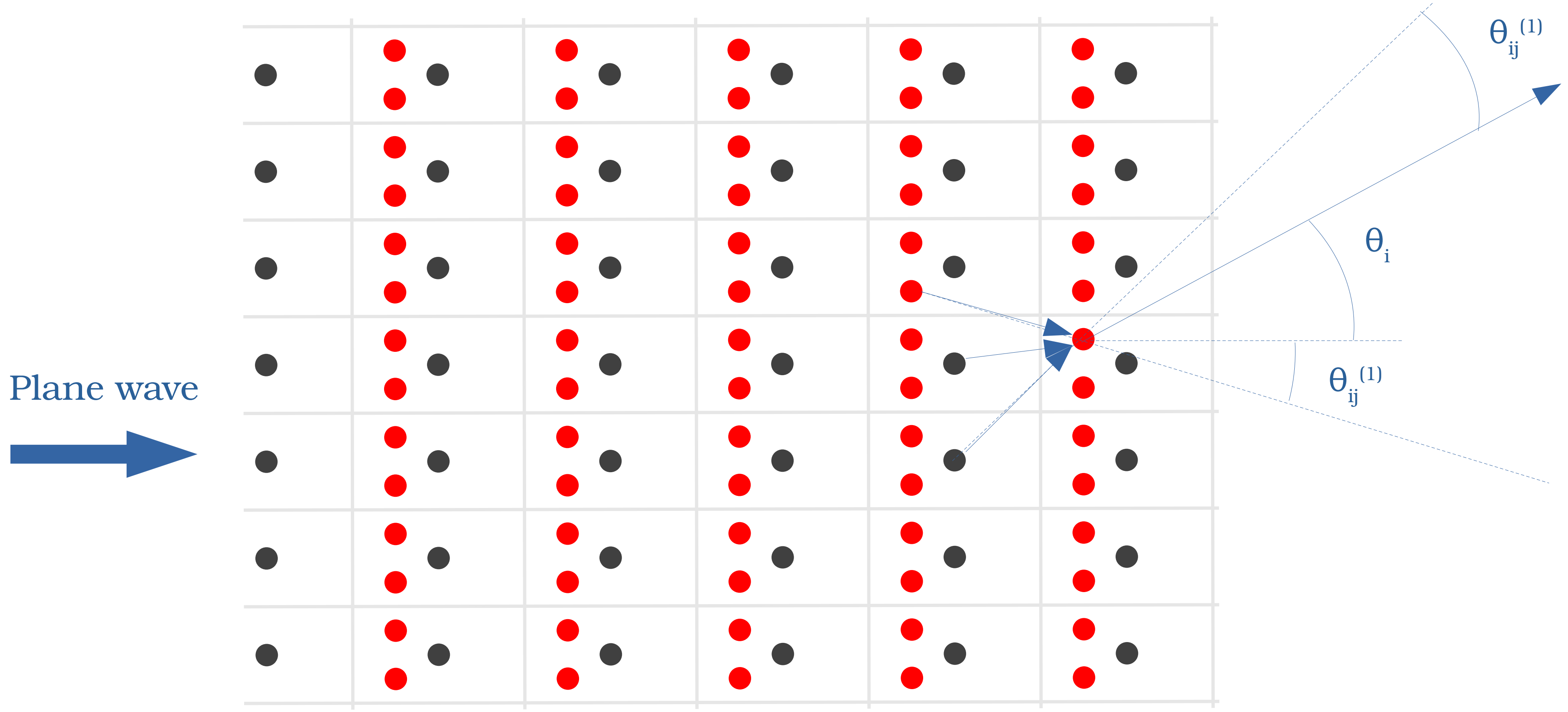


Simulations with small tilts [-5,+5] deg[1]



[1] J.H. Chen, D. Van Dyck, and M. Op De Beeck, “Multislice Method for Large Beam Tilt with Application to HOLZ Effects in Triclinic and Monoclinic Crystals,” Acta Crystallogr. Sect. A Found. Crystallogr., vol. 53, no. 5, pp. 576–589, 1997

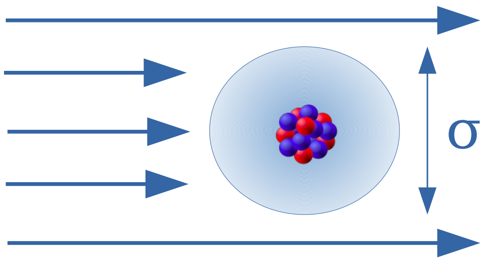
Extension to multiple scattering



Multiple Scattering in Electron Diffraction

Atomic interation cross section

$$P_{scattered} = \sigma\phi_{incident}$$



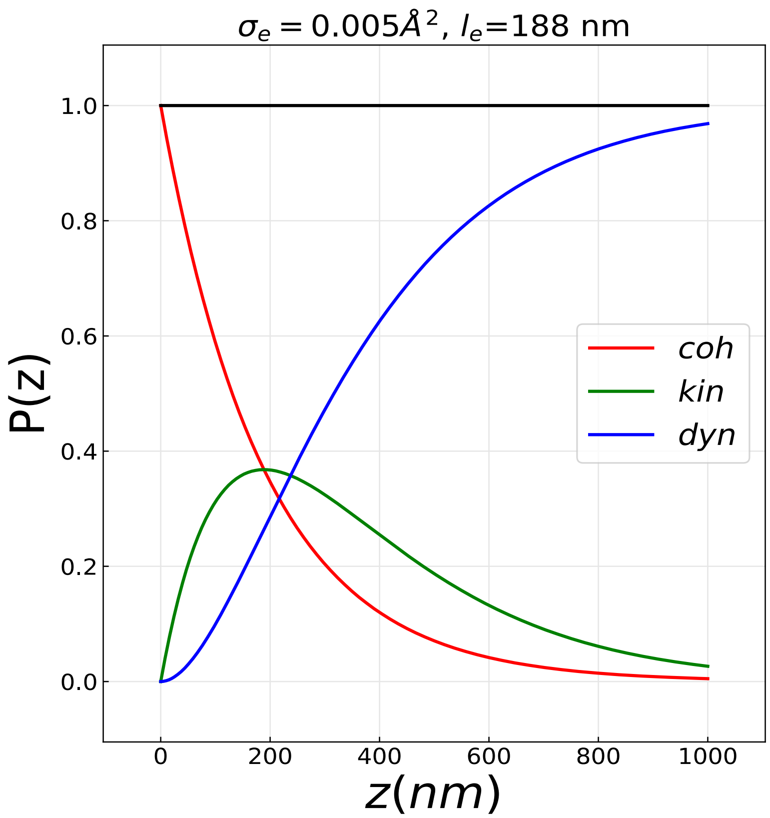
X-rays (Thomson scattering)

$$\sigma_{th} = \frac{8\pi}{3} r_e^2 = 66 fm^2$$

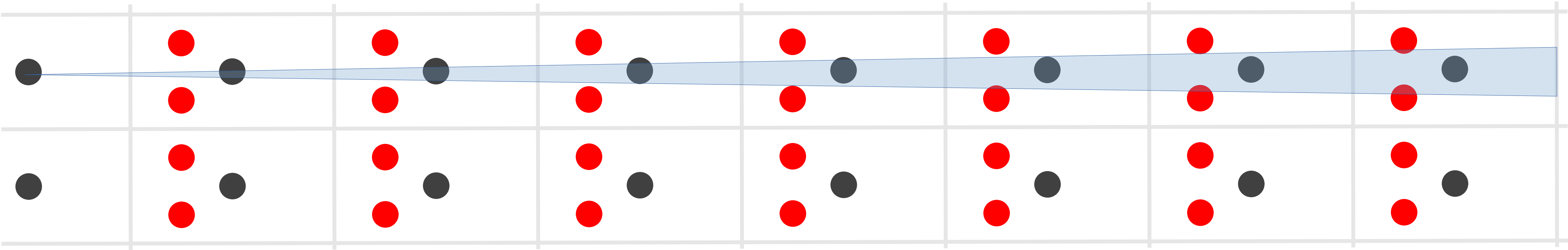
Electrons (Coulomb scattering)

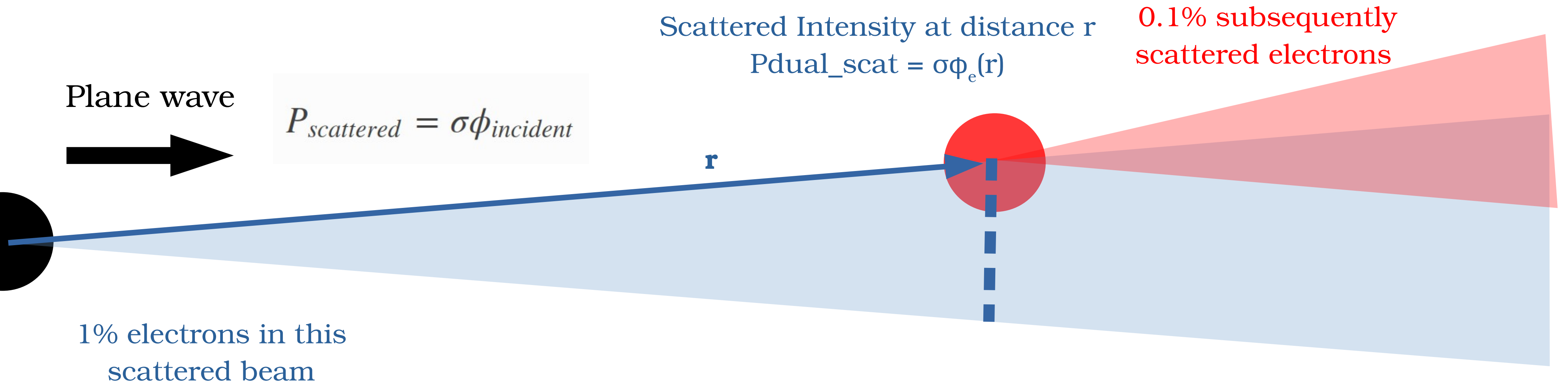
$$\sigma_{th} \approx 1.87 \times 10^6 Z^{4/3} (c/v)^2 = 5 \times 10^7 fm^2$$

Mean free path, l_e = 200nm

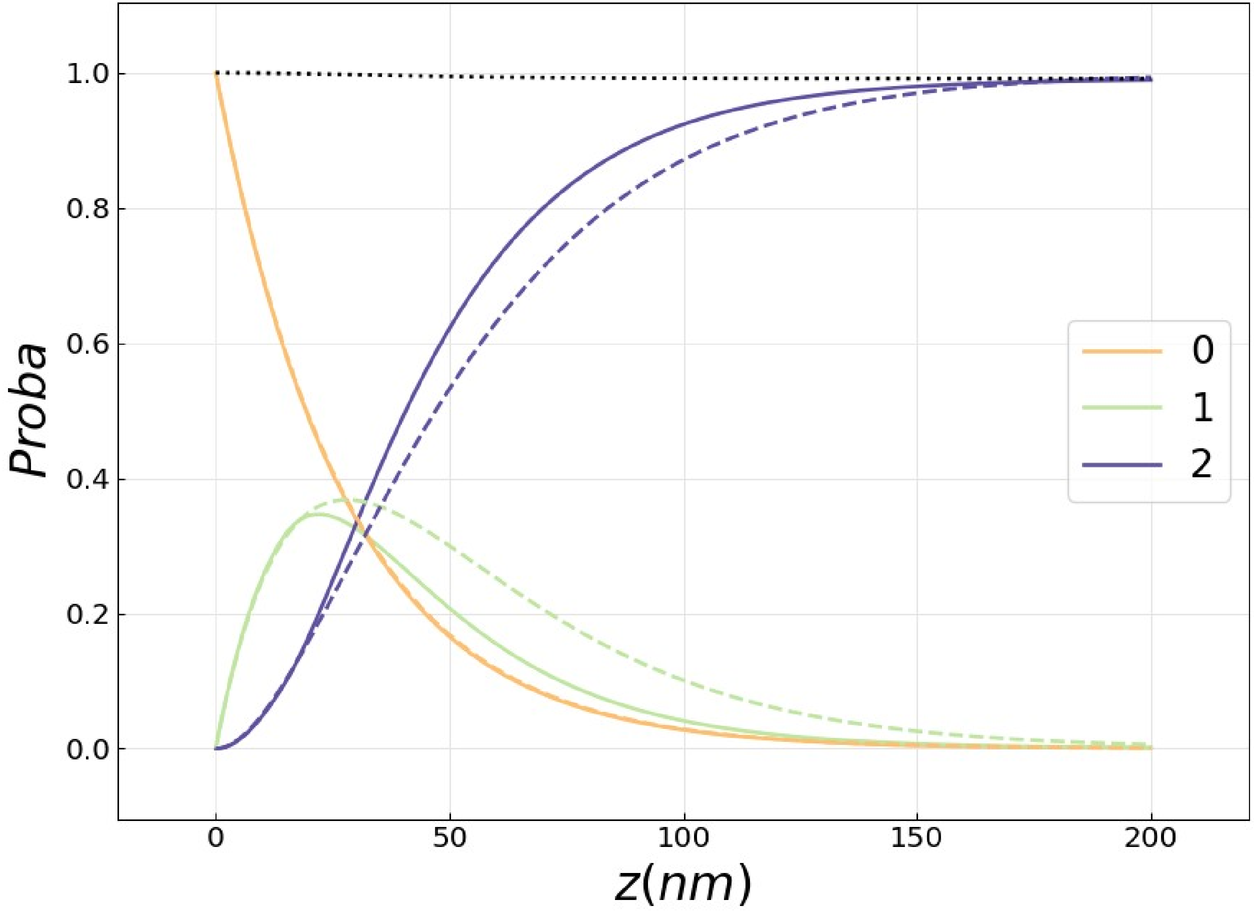


Extension to multiple scattering





Extension to multiple scattering

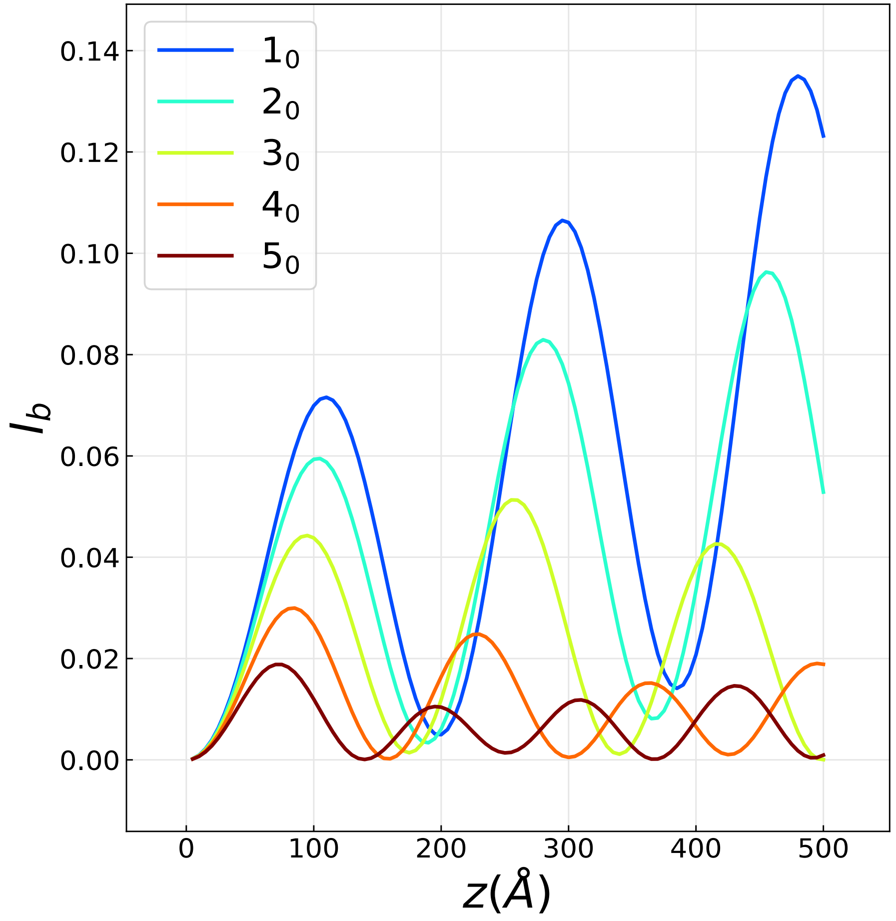


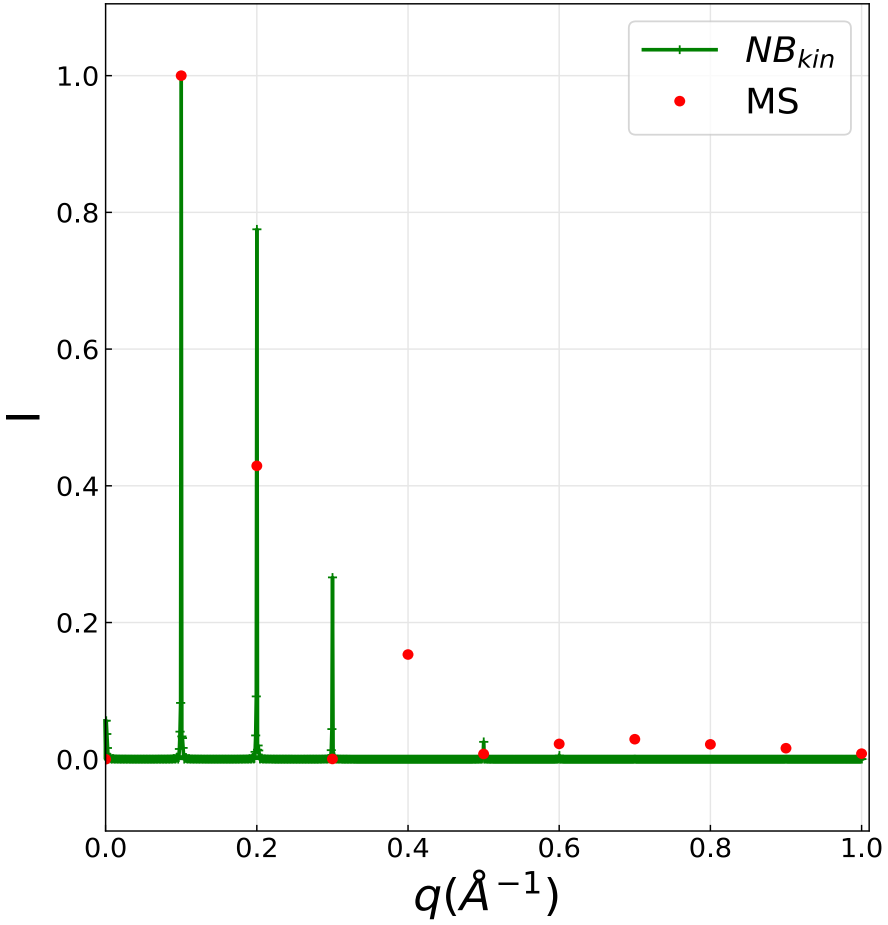
Dashed : near bragg

solid : Theory

0 no scattering
1 single scattering
2 multiple scattering

Near Bragg vs multislice





Current challenges

- adapt nearBragg approach to multiple scattering events

- Compare simulation to experimental dataset

- Simulate defects (modelling and computational), Thermal diffuse scattering, inelastic scattering

- model partially coherent beam as produced by LaB_6 guns