# Simulation of high energy electron diffraction patterns of organic crystals

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## Contribution

We provide a comparison of the 2 most popular methods used for simulating electron diffraction(ED), namely the multislice(MS) and blochwave(BW).
Dynamical diffraction is described by both methods while advantages and limitations of both approaches are highlighted and discussed.
As proof of concept, the multislice method is subsequently used to reproduce a continuous rotation experiment based on $\alpha$-glycine.

## Motivations for ED simulations

Macromolecular structures can be solved from ED patterns using standard X-ray diffraction (XD) techniques. ED is attractive as it:

- Enables **solving structures from nanocrystals** due to the *strong electron-atom interaction*.
- Produces **high resolution maps** due to the very *small electronic wavelength $0.025A@200keV$*.
- Resolves **atomic nucleus positions** through the *electrostatic potential map*.

However, **Rfactors obtained through ED remain larger than those of X-ray** due to a lack of theoretical understanding of *dynamical diffraction*.

## Theoretical background
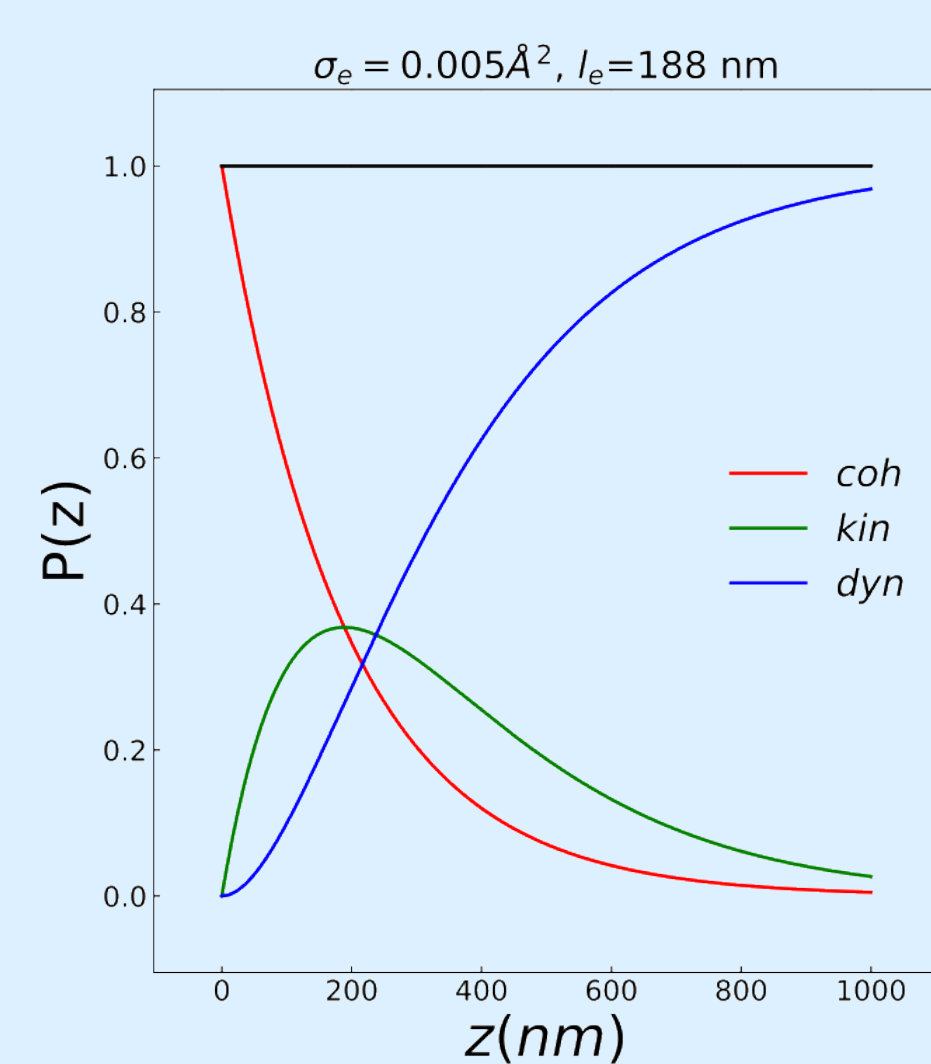
### Standard XD pipeline
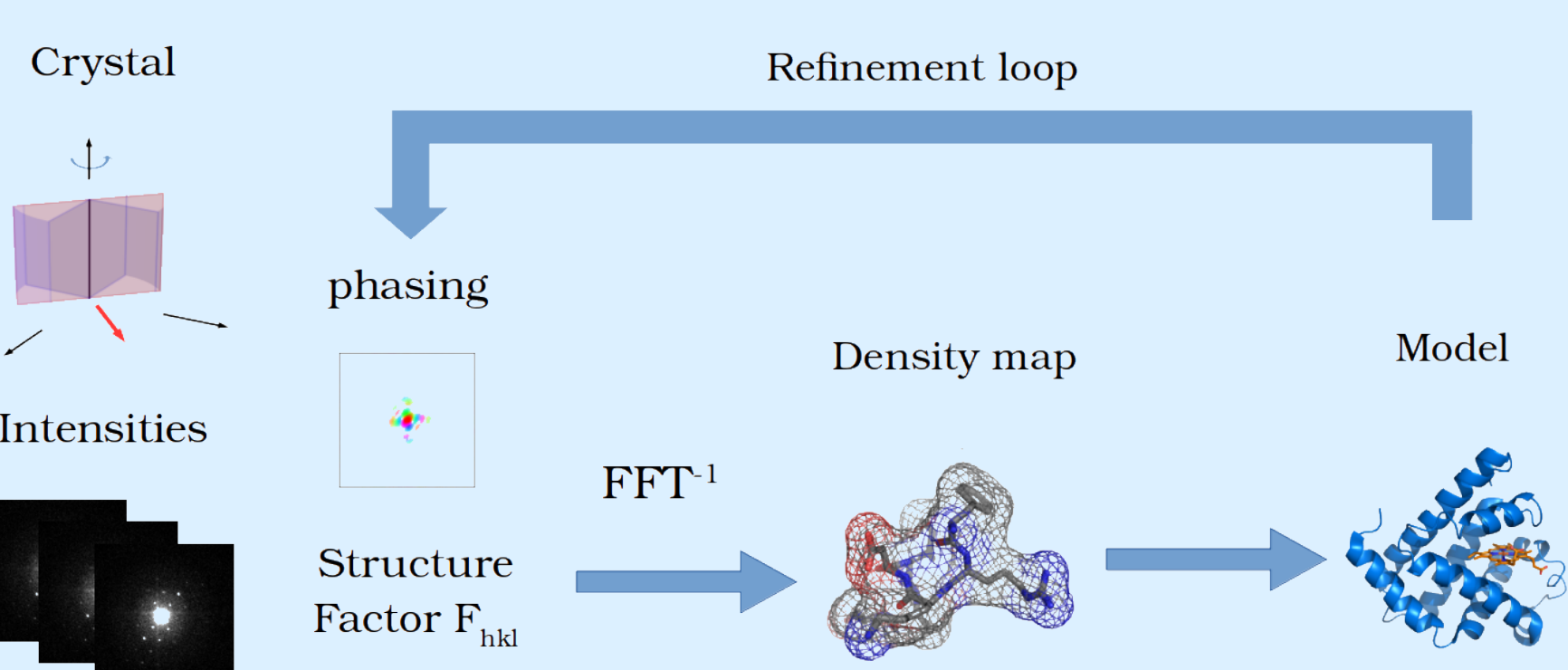


XD is based on the *kinematic theory of diffraction* due to the weak interaction between X-rays and the electron density. Kinematic theory can not rigorously be applied to ED.

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### Schrödinger fast electron wave equation

In ED, the structure factor is related to the electrostatic potential through :

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

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### Particle oriented picture



200keV electrons have typical elastic mean free path of 200nm in organic atoms. This results in an appreciable probability of multiple scattering in nanocrystals.

## Multislice(MS) method  [1]

The sample is **sliced in real space** into regular slices $\Delta z$ and the solution is propagated from slice to slice through:

$$\Psi(z + \Delta z) = \mathcal{F}^{-1}\left\{ p(k_x, k_y)\mathcal{F}\left( e^{i\sigma\nu_{\Delta_z}(z)}\Psi(z) \right) \right\}$$

where the Fresnel propagator $p(k_x, k_y) = e^{-i\pi\lambda\Delta z(k_x^2+k_y^2)}$, $\nu_{\Delta_z} = \int_z^{z+\Delta z} V(x,y,z')dz'$ is the projected potential.
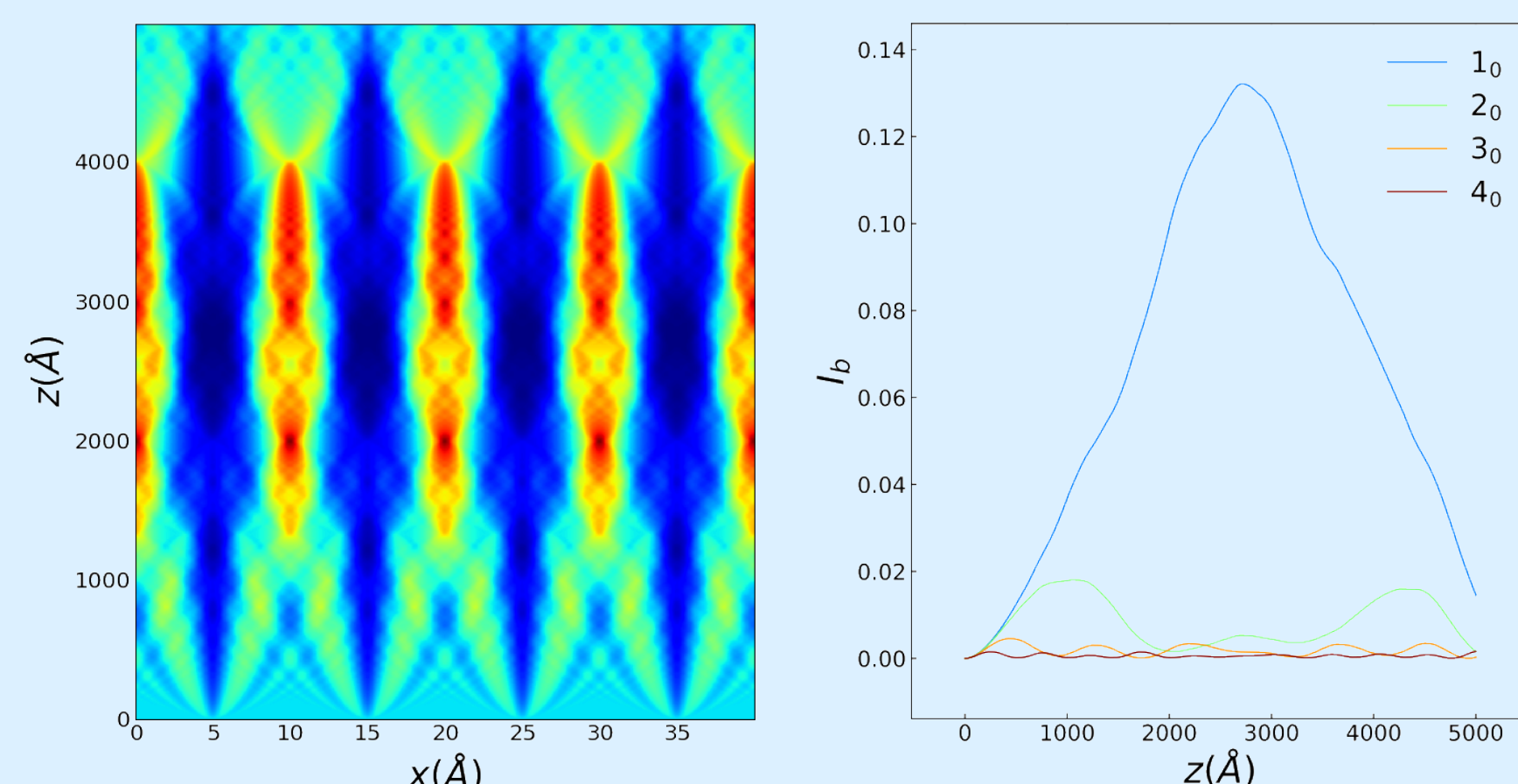


a) Typical $\Psi(x,z)$      b) Strong beams intensities

- The Discrete Fourier Transform $\mathcal{F}$ provides **fast and efficient $N\log N$ time complexity**.
- **Padding and large transverse super cells** necessary for non zone-axis orientations.
- Possiblity to model **solvent,inelastic scattering, disorder, partial coherency....**
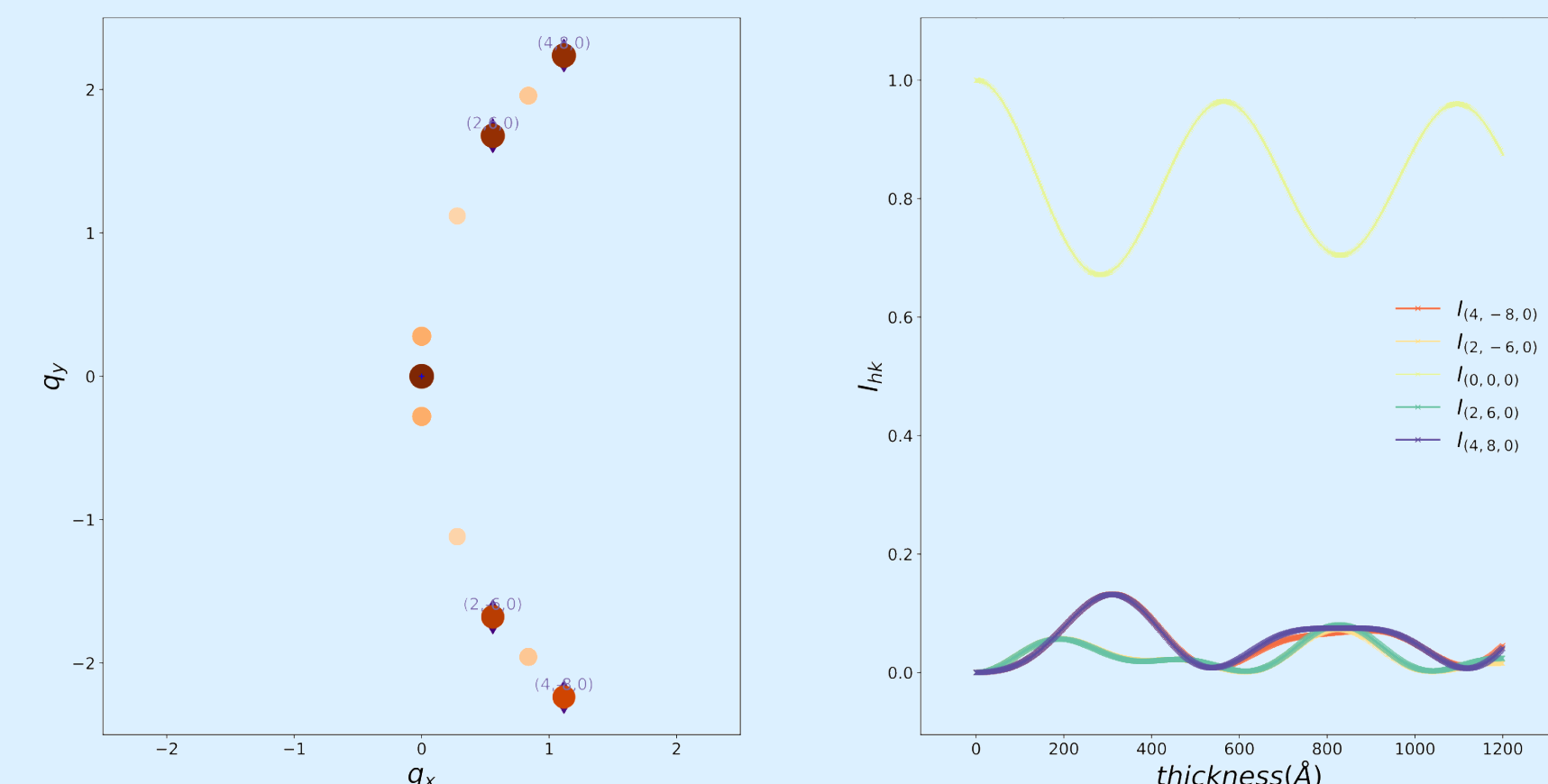
Padded simulation for a IRELOH :
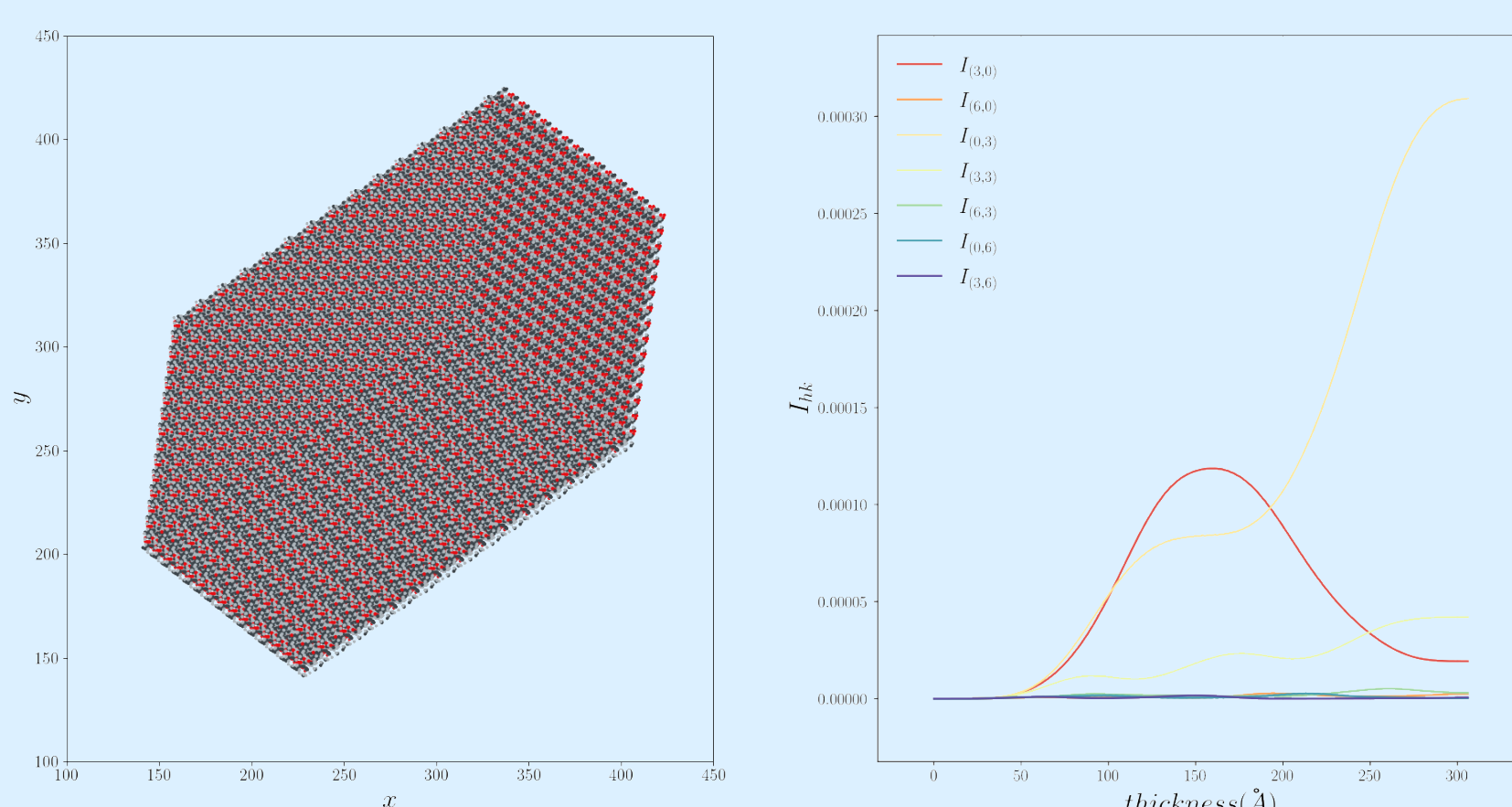


a) real space setup      b) strong beams intensities

## Blochwave(BW) approach [2]

The wave function is **solved in reciprocal space** by finding the eigen values $\gamma_j$ and eigen vectors $C_{j,\mathbf{G}}$:

$$S_{\mathbf{G}}C_{j,\mathbf{G}} + \sum_{\mathbf{G'}}\frac{U_{\mathbf{G}-\mathbf{G'}}}{2k_0}C_{j,\mathbf{G'}} = \gamma_j C_{j,\mathbf{G}}$$

The diffracted intensities of the strong beams $\mathbf{G}$ (small excitation error $S_{\mathbf{G}}$) are computed for any thickness sample $H$ with $I_{\mathbf{G}} = |\mathbf{C}e^{2i\pi\gamma_j H}\mathbf{C}^{-1}|^2$.



a) $S_{\mathbf{G}}$ and potential $U_{\mathbf{G}}$      b) Strong beams intensities $I_{\mathbf{G}}$

- Diagonalization **time complexity scales as $N^3$** but **only strongest beams need be included**.
- **Random orientations of any lattice** can be simulated.
- Inelastic scattering can be modelled but **not solvent scattering, disorder and defects**.
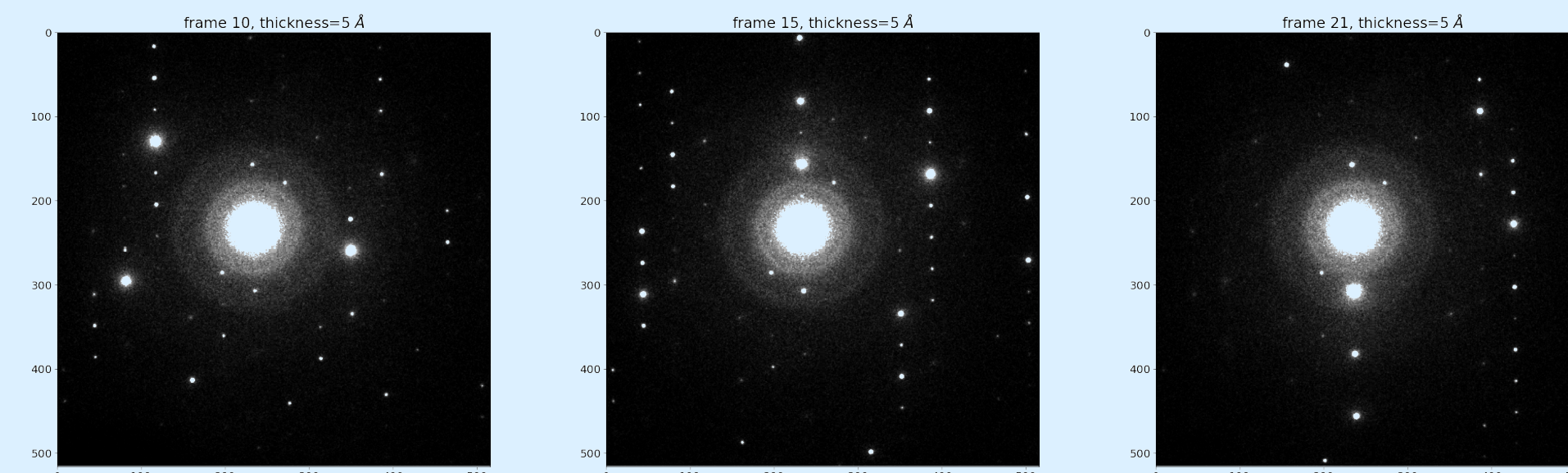
Rocking curves for diamond in different configurations :
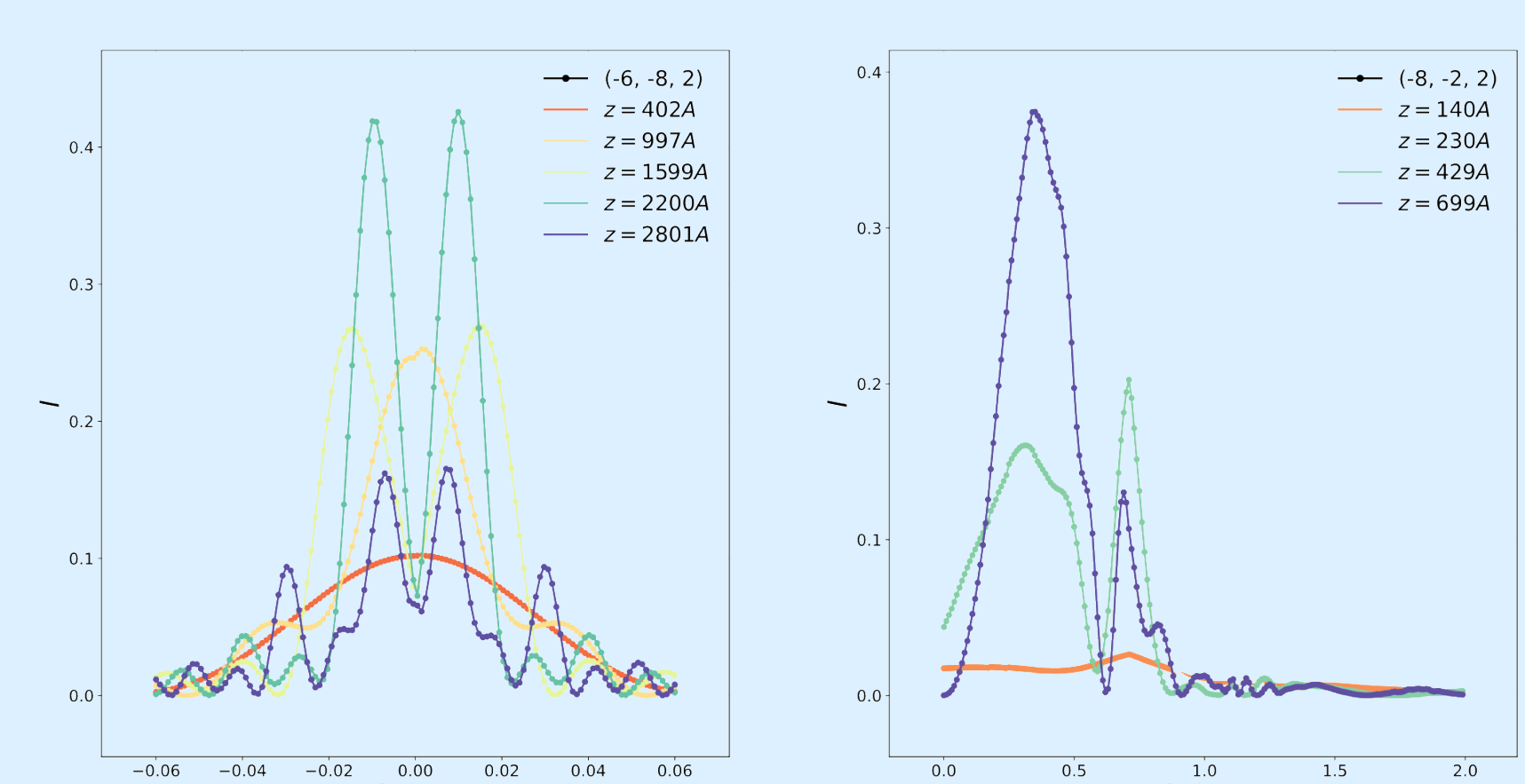


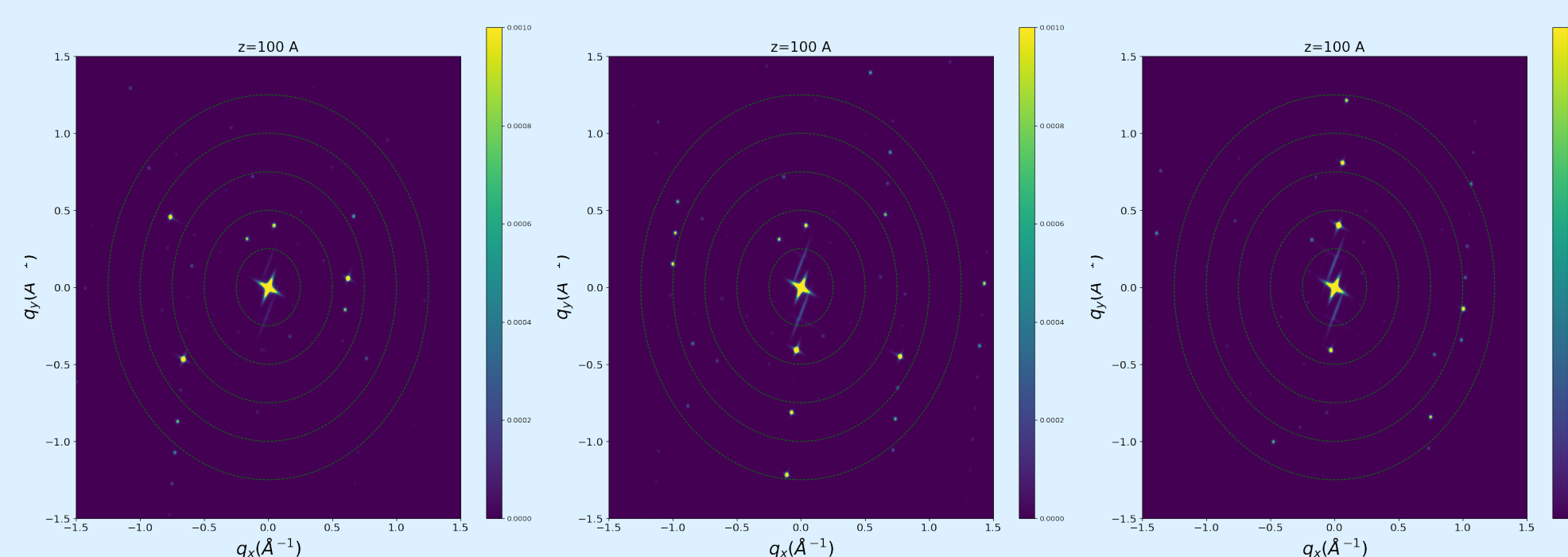a) 3 beam setup      b) arbitrary orientation

## Application to $\alpha$-glycine

Experimental continuous rotation electron diffraction dataset of $\alpha$-glycine



a) frame 10      b) frame 15      c) frame 21

Simulated frames using multislice. Padding was used to simulate the experimental orientations as retrieved by data processing software PETS2(http://pets.fzu.cz/)



a) frame 10      b) frame 15      c) frame 21

The main reflections are correctly predicted, although the patterns need to be rotated in the plane of the figure to get an exact match. The reflection broadening is related to finite size effects and window function of the padded domain.

Excitation errors $(-\log_{10}(S_{\mathbf{G}}))$ of main beams over frames [1-30] and simulated rocking curve of main reflection $(0,0,2)$:
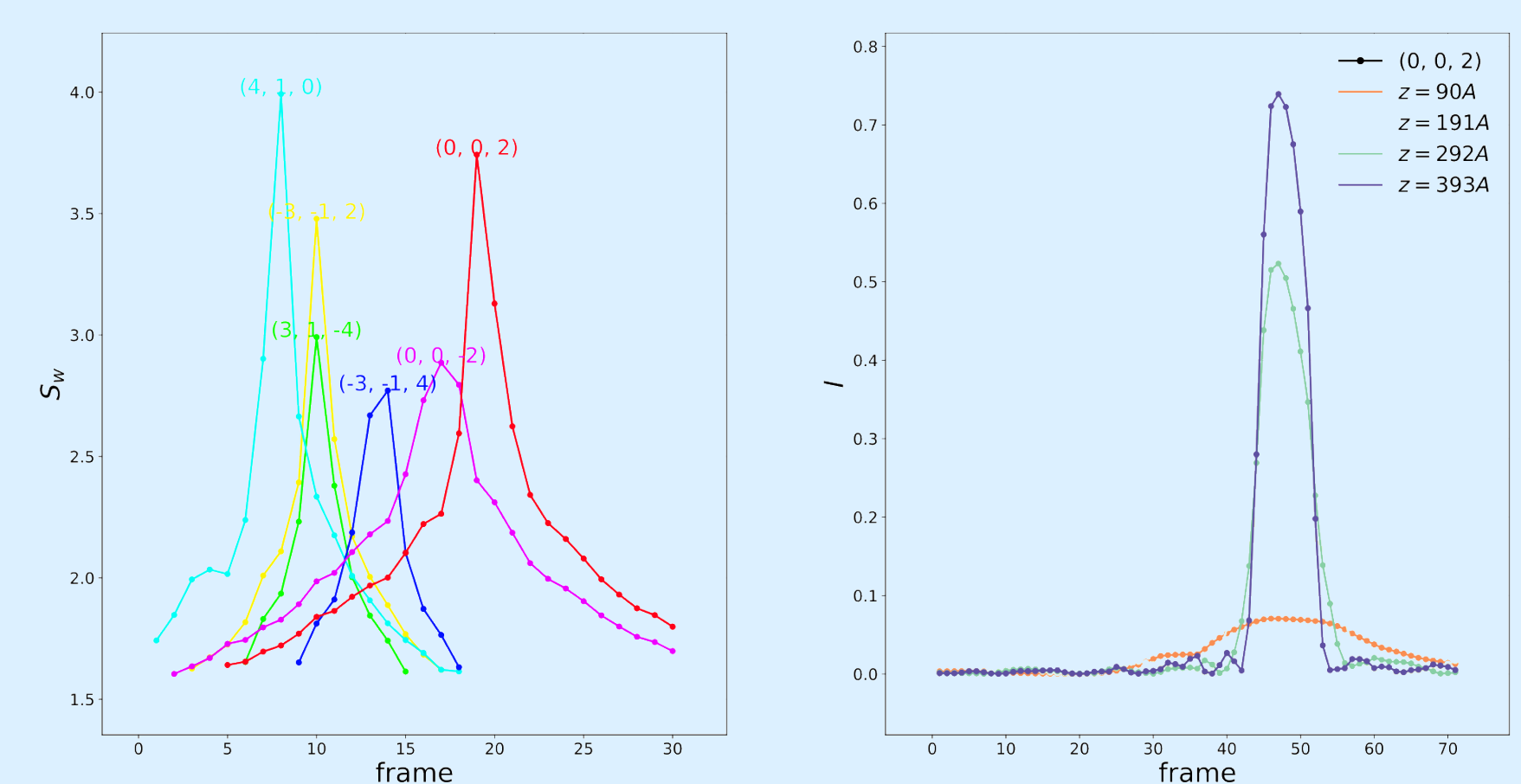


a) Excitation error      b) Rocking curve for (0,0,2)

Experimental intensities :
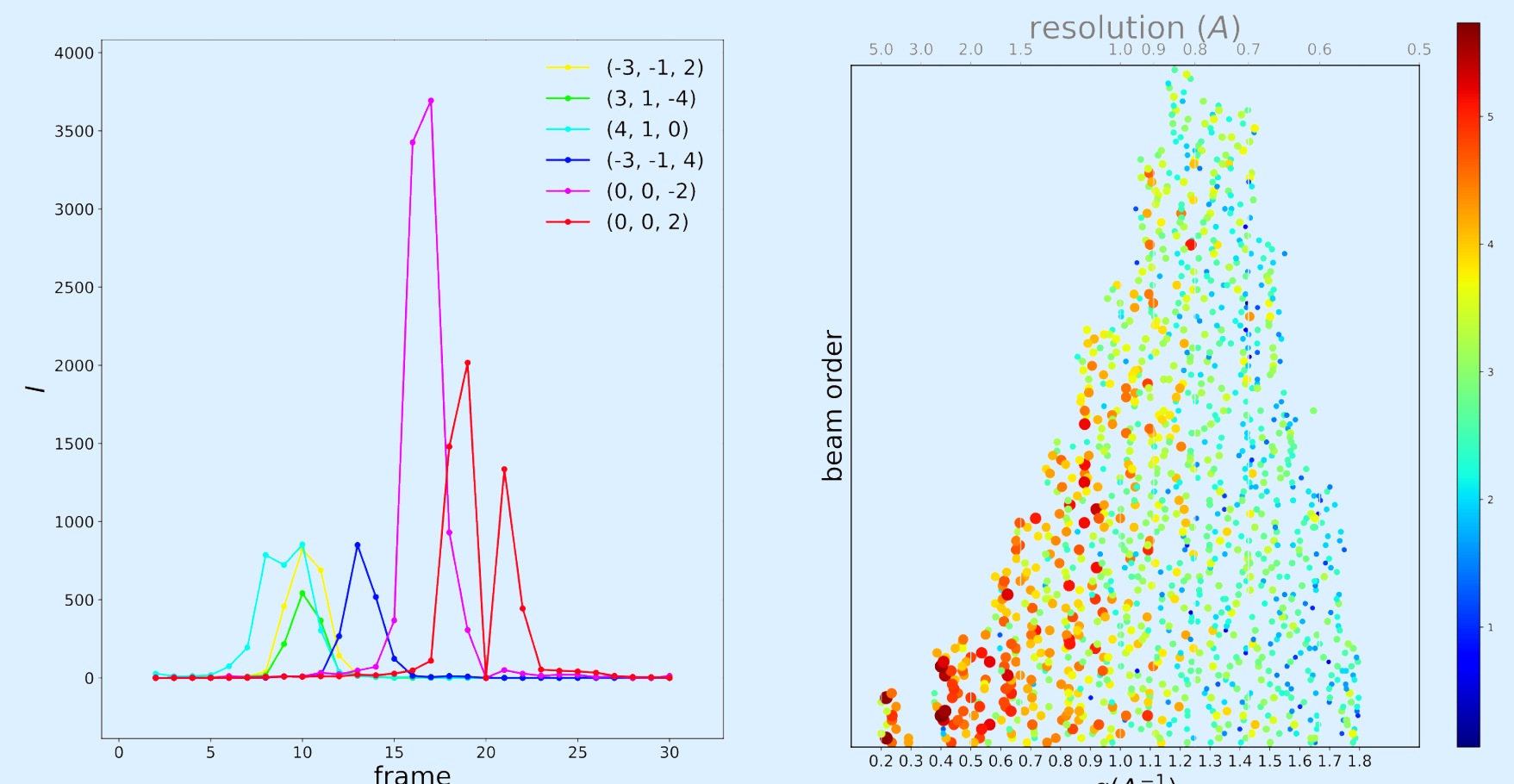


a) Experimental intensities      b) Distribution with resolution

## References

[1] E. J. Kirkland. *Advanced Computing in Electron Microscopy*. Springer, third edit edition, 2019.

[2] J. M. Zuo and A. L. Weickenmeier. *Ultramicroscopy*, 57(4):375–383, 1995.

## Acknowledgements