X-Ray Imaging of Correlated Disorder in Crystals

Victoria Kovalchuk[1], Jacob Ruff[2], and Jooseop Lee[2]

Department of Physics, University of Virginia[1] and Department of Physics, Cornell University[2]

Motivation

X-Ray crystallography has advanced a lot in the last century, but so has the number and variety of materials that need their crystal structure analyzed. A new technique has been developed by researchers at ETH Zurich, called 3D Δ-PDF (pair distribution function), which specializes in analysing crystals with correlated disorder[2]. The goal of this project was to write Python code that implements this new method and to study it's performance on models with the intent of applying this method to the high resolution X-ray diffraction data collected at Cornell's Synchrotron Source CHESS.

Background

X-ray crystallography is the analysis of X-ray scattering data to determine the atomic or molecular structure of a crystal.

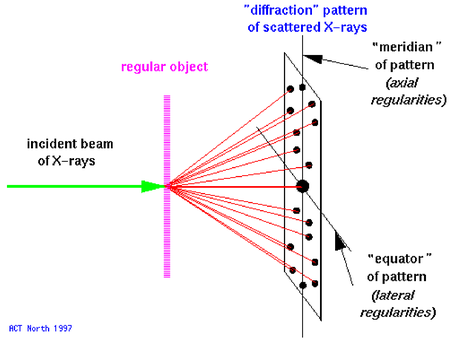

Figure: A simple visualization of how X-ray diffraction works[1].

The data that is collected with X-ray diffraction is in the reciprocal (momentum) space and the intensity $I(q)$ can be modeled as the absolute value squared of the Fourier Transform of the real space, where $\rho(x)$ is the electron density of a simulated atom.

$$I(q) = |\sum_{j-1}^{N} \rho(x_j) e^{2\pi i q x_j}|^2 \qquad (1)$$

To obtain the Δ-PDF, we take the Fourier transform of the diffuse scattering, which is the part of the diffraction data that represents deviations from the average crystal structure.

1D Δ-PDF

First we implemented the method in 1D because it is a lot more intuitive.

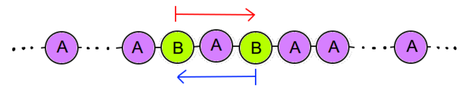

Figure: Example of 1D chain with defects where atoms B have ten times the scattering power of atoms A.

Unlike in Fig. 2, the atoms were actually treated as point particles with different scattering power. Then, Eq. 1 was applied to produce Fig. 3, which is a simulation of the 1D X-Ray diffraction data. The sharp peaks at integer values are called Bragg peaks and represent the average crystal structure.

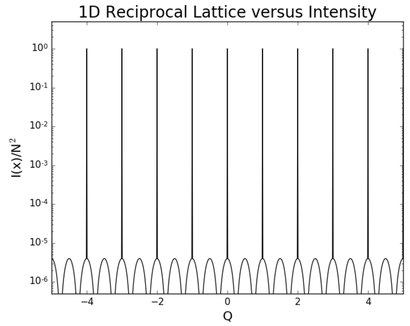

Figure: Reciprocal space of the 1D chain.

In order to calculate the Δ-PDF, we had to separate the Bragg peaks and the diffuse scattering in the simulated diffraction data.

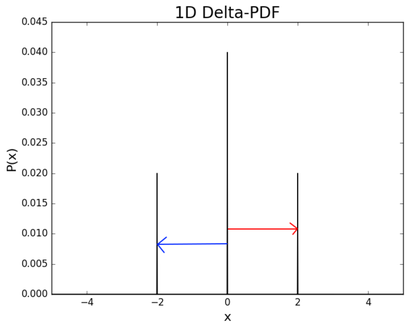

Figure: The 1D Δ-PDF of the 1D chain.

This model was simple enough that we knew what the Δ-PDF should look like, and the results matched our predictions.

2D Δ-PDF

Next, we simulated a simple 2D model, where the Δ-PDF should emphasize the blue atoms.

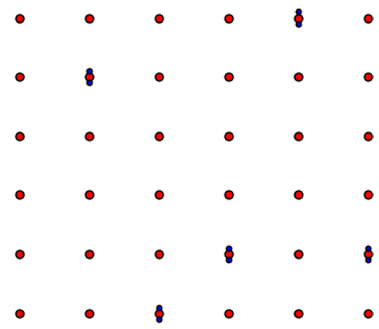

Figure: The real space of a 2D lattice where 10% of the atoms have a defect that attaches a blue atom above and below it.

These simulations did produce the predicted result, but only when the reciprocal space that was simulated was very large.

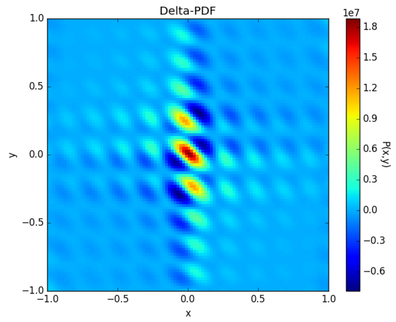

Figure: The 2D Δ-PDF of the 2D lattice when the reciprocal space was large (50 by 50).

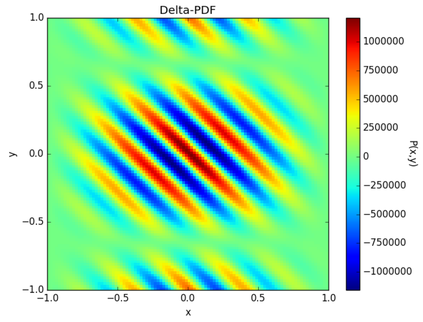

Figure: The 2D Δ-PDF of the 2D lattice when the reciprocal space is small (3x3).

Conclusions and Next Steps

The Δ-PDF method seems capable of providing useful information about systems that contain correlated disorder. However, current implementation would require extremely large reciprocal space lattices that are not within the scope of current X-ray diffraction techniques. Some next steps in this project include:

- Implement the Fast Fourier Transform technique
- Include more multiprocessing to speed up simulations
- Use properties of Fourier Transform to make the code more efficent.
- Test different models with Δ-PDF, and expand to 3D models

References

[1] North, A.L., X-Ray Crystallography, `http://life.nthu.edu.tw/∼labcjw/BioPhyChem/EM/BBS_xray.htm` (August 8, 2017).

[2] Weber, Thomas and Simonov, Arkadiy. The three-dimensional pair distribution function analysis of disordered single crystals: Basic concepts. *Zeitschrift fur Kristallographie* **227**, 238-247 (2012).

Acknowledgements

- Thank you to my mentors Jacob Ruff and Jooseop Lee and to my fellow REU peers for making this research really enjoyable.
- Thanks to the NSF and Cornell University (particularly the CLASSE REU program) for providing this amazing opportunity.