Diffraction Imaging with Coherent X-rays

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The Phase Problem: A Coherence Effect

The phase problem is due to the fact that there are no ways to tell where each photon is scattered from.

No phase problem for X-ray inelastic scattering.

Phase ambiguities: $\rho(r)$, $\rho(r + r_0)e^{i\theta_0}$, $\rho^*(-r + r_0)e^{i\theta_0}$
Regular Sampling: Sampling at the Bragg-peak Frequency

\[ |F(k)| = \left| \sum_{r=0}^{N-1} \rho(r) e^{-2\pi i k \cdot r / N} \right| \quad (i) \]

\[ k = 0, 1, 2, \ldots N - 1 \]

\[ \rho(r): \text{real} \Rightarrow |F(k)| = |F^*(-k)| \quad \rho(r): \text{complex} \]

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Independent equations: intensity points have no crystallographic symmetry.

Oversampling: Sampling at a Spacing Finer than the Bragg-peak Frequency

\[ |F(k)| = \left| \sum_{r=0}^{N-1} \rho(r) e^{-2\pi i k \cdot r / (2N)} \right| \quad \text{(ii)} \]

\[ k = 0, 1, 2, \ldots 2N - 1 \]

\( \rho(r): \text{real} \Rightarrow |F(k)| = |F^*(-k)| \quad \rho(r): \text{complex} \]

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The Oversampling Method

\[ g(\mathbf{r}) = \begin{cases} \rho(\mathbf{r}) & 0 \leq \mathbf{r} \leq N - 1 \\ 0 & N \leq \mathbf{r} \leq 2N - 1 \end{cases} \]  

(iii)

\[ |F(\mathbf{k})| = \left| \sum_{\mathbf{r}=0}^{2N-1} g(\mathbf{r}) e^{-2\pi i \mathbf{k} \cdot \mathbf{r}/(2N)} \right| \]

(iv)

\[ \mathbf{k} = 0, 1, 2, \ldots 2N - 1 \]

\[ \sigma = \frac{\text{electron density region} + \text{no-density region}}{\text{electron density region}} \]  

(v)

\[ \sigma > 2: \text{the phase information exists inside the diffraction intensity!} \]
Multiple Solutions

1D Case:
\( (\leq 2^N \text{ multiple solutions}) \)
\[
F(k_j) = \sum_{j=0}^{N-1} \rho(j) e^{-2\pi i k_j j/N}
\]
\[
a_j \equiv \rho(j), \quad z \equiv e^{-2\pi i k_j/N}
\]
\[
F(z) = \sum_{j=0}^{N-1} a_j z^j = \prod_{j=0}^{N-1} (z - z_j)
\]
\[
F^*(z) = F(1/z) = \prod_{j=0}^{N-1} (z - 1/z_j)
\]
\[
I(z) \propto F(z) F^*(z) = \prod_{j=0}^{N-1} (z - z_j)(z - 1/z_j)
\]

2D & 3D Case:
(Multiple solutions are rare)
Mathematically, 2D and 3D polynomials usually can not be factorized.

The Physical Interpretation of the Oversampling Method

Better coherence ⇒ More correlated intensity points ⇒ Phase information
Oversampling and Coherence

Oversampling vs. spatial coherence: \[ \Delta \theta \leq \frac{\lambda}{2Oa} \quad a: \text{sample size} \]

Oversampling vs. temporal coherence: \[ \frac{\lambda}{\Delta \lambda} \geq \frac{Oa}{d} \quad d: \text{desired resolution} \]

\[ O = \begin{cases} \sqrt[2]{\sigma} & \text{for a 2D sample} \\ \sqrt[3]{\sigma} & \text{for a 3D sample} \end{cases} \]

An Iterative Algorithm

(I) \[ F_j'(k) = |F_{\exp}(k)| \times e^{\phi_{j-1}(k)} \]

(II) \[ \phi_j'(0,0,0) = 0 \]

(III) \[ \rho_j'(r) = \text{FFT}^{-1}(F_j'(k)) \]

(IV) \[ \rho_j(r) = \begin{cases} \rho_j'(r) & \text{if } r \in S \text{ and } \rho_j'(r) \geq 0 \\ \rho_{j-1}(r) - \beta \times \rho_j'(r) & \text{if } r \notin S \text{ or } \rho_j'(r) < 0 \end{cases} \]

(V) \[ F_j(k) = \text{FFT}(\rho_j(r)) \]

(VI) Adopt \( \phi_j(k) \) from \( F_j(k) \)

Experimental Demonstration of Coherent Imaging

(a) A SEM image

(b) An oversampled diffraction pattern (in a logarithmic scale) from (a).

(c) An image reconstructed from (b).


(d) The convergence of the reconstruction.
Phase Retrieval as a Function of the Oversampling Ratio

\[
\sigma = 5 \text{ (180 x 180 pixels)} \quad \sigma = 4 \text{ (160 x 160 pixels)}
\]

\[
\sigma = 2.6 \text{ (130 x 130 pixels)} \quad \sigma = 1.9 \text{ (110 x 110 pixels)}
\]
Imaging a Nanostructure at 7 nm Resolution

(a) A SEM image of a non-crystalline sample made of Au

(b) A coherent diffraction pattern from (a) (the resolution at the edge is 7 nm)

(c) An image reconstructed from (b)
Imaging Buried Nanostructures

(a) A SEM image of a double-layered sample made of Ni (~2.7 x 2.5 x 1 μm³)

(b) A coherent diffraction pattern from (a) (the resolution at the edge is 8 nm)

(c) An image reconstructed from (b)
3D Imaging of Nanostructures

The reconstructed top pattern

The reconstructed bottom pattern

An iso-surface rendering of the reconstructed 3D structure
Determining the Absolute Electron Density of Disordered Materials at Sub-10 nm Resolution

(a) A coherent diffraction pattern from a porous silica particle

(b) The reconstructed absolute electron density

(c) The absolute electron density distribution within a 100 x 100 nm$^2$ area

Imaging *E. Coli* Bacteria

(a) Light and fluorescence microscopy images of *E. Coli* labeled with manganese oxide

(b) A Coherent X-ray diffraction pattern from *E. Coli*

(c) An image reconstructed from (b).

Coherent X-ray Diffraction of Bone Samples

(a) -74°

(b) -76°

(c) -77°

(d) -78°
Summary

- Proposed a theoretical explanation to the oversampling method.
- Carried out the first (to our knowledge) definitive experimental demonstration of coherent imaging.
- The results potentially open a door to near atomic resolution 3D X-ray diffraction microscopy.
- The methodology can in principle be extended to electrons.
- Future – the potential of imaging large biomolecules using X-FELs.
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