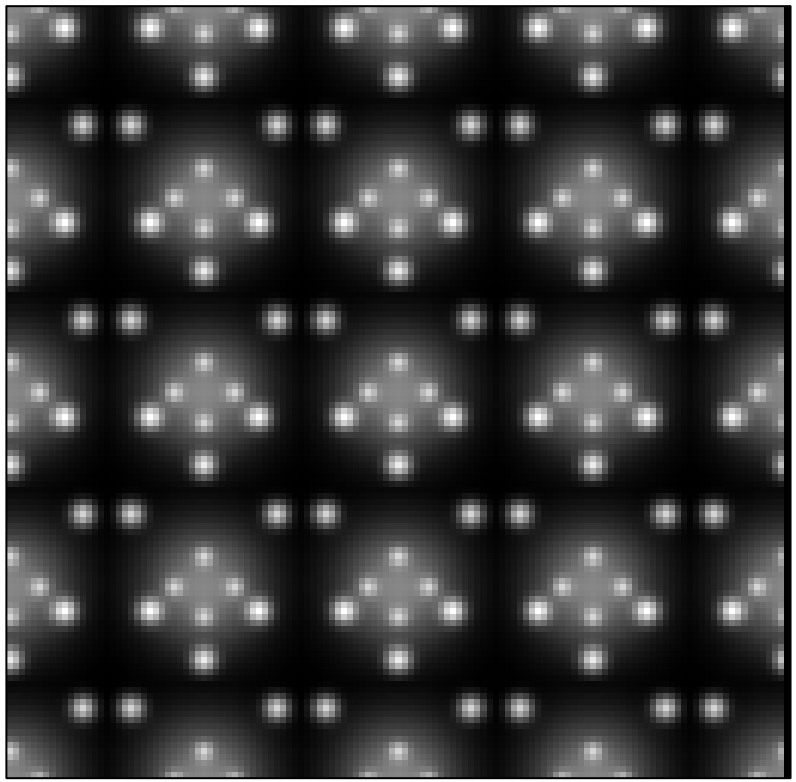
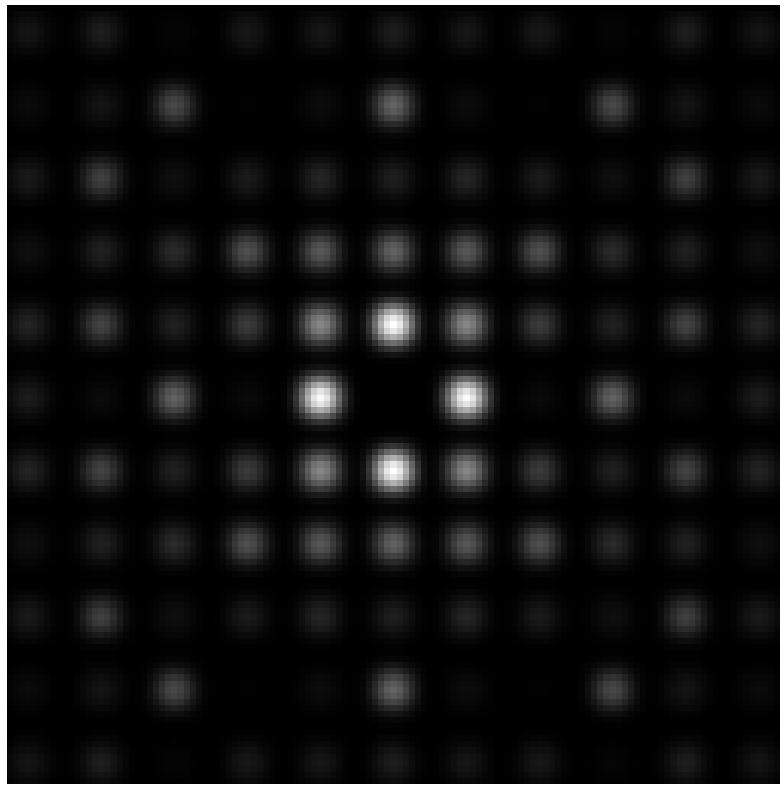
Demonstration: how resolution works in reciprocal space: If we can add beams at large distance from center of patterns *with the correct phase*, we can reconstruct the structure with very high definition:

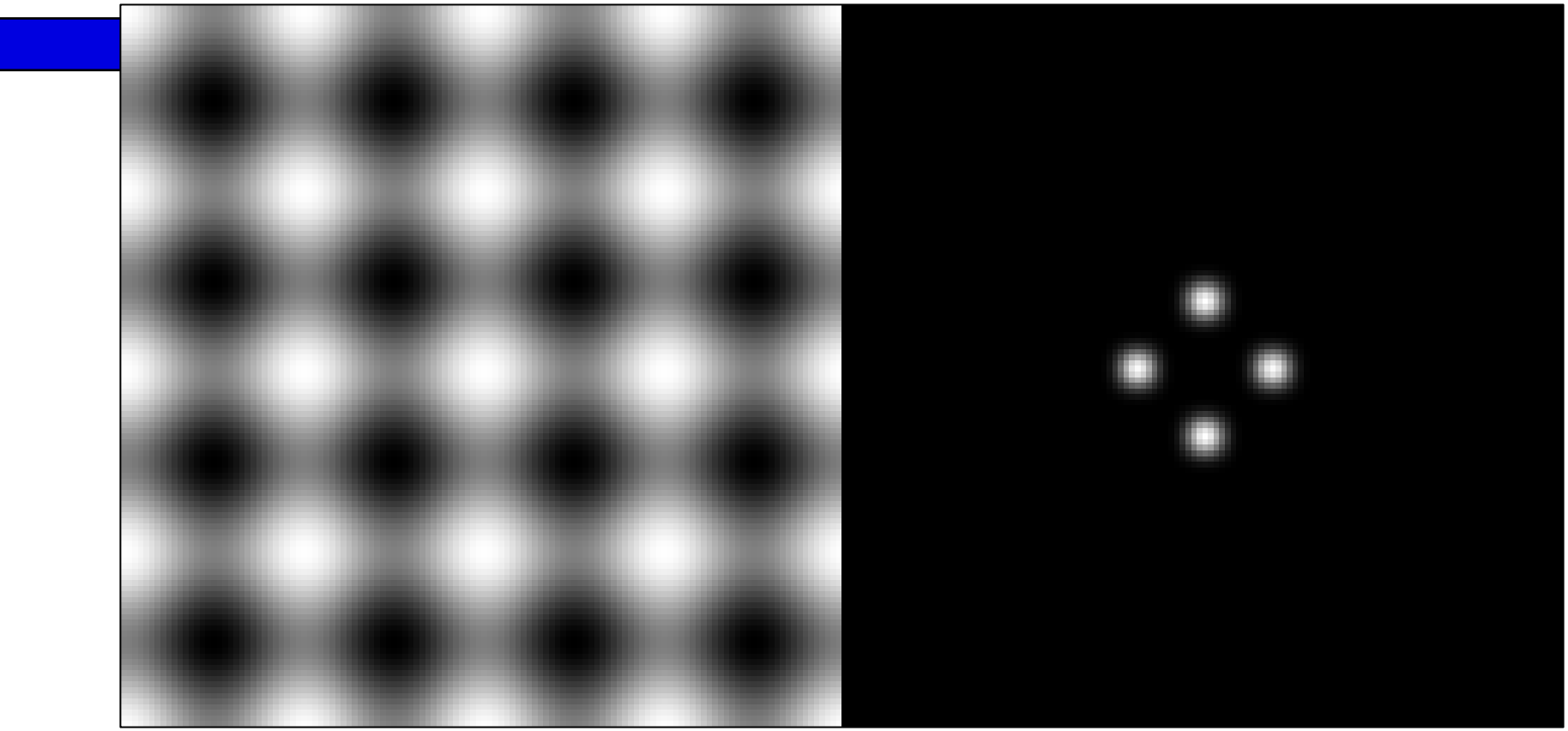
Example: simple test structure of repeated molecule



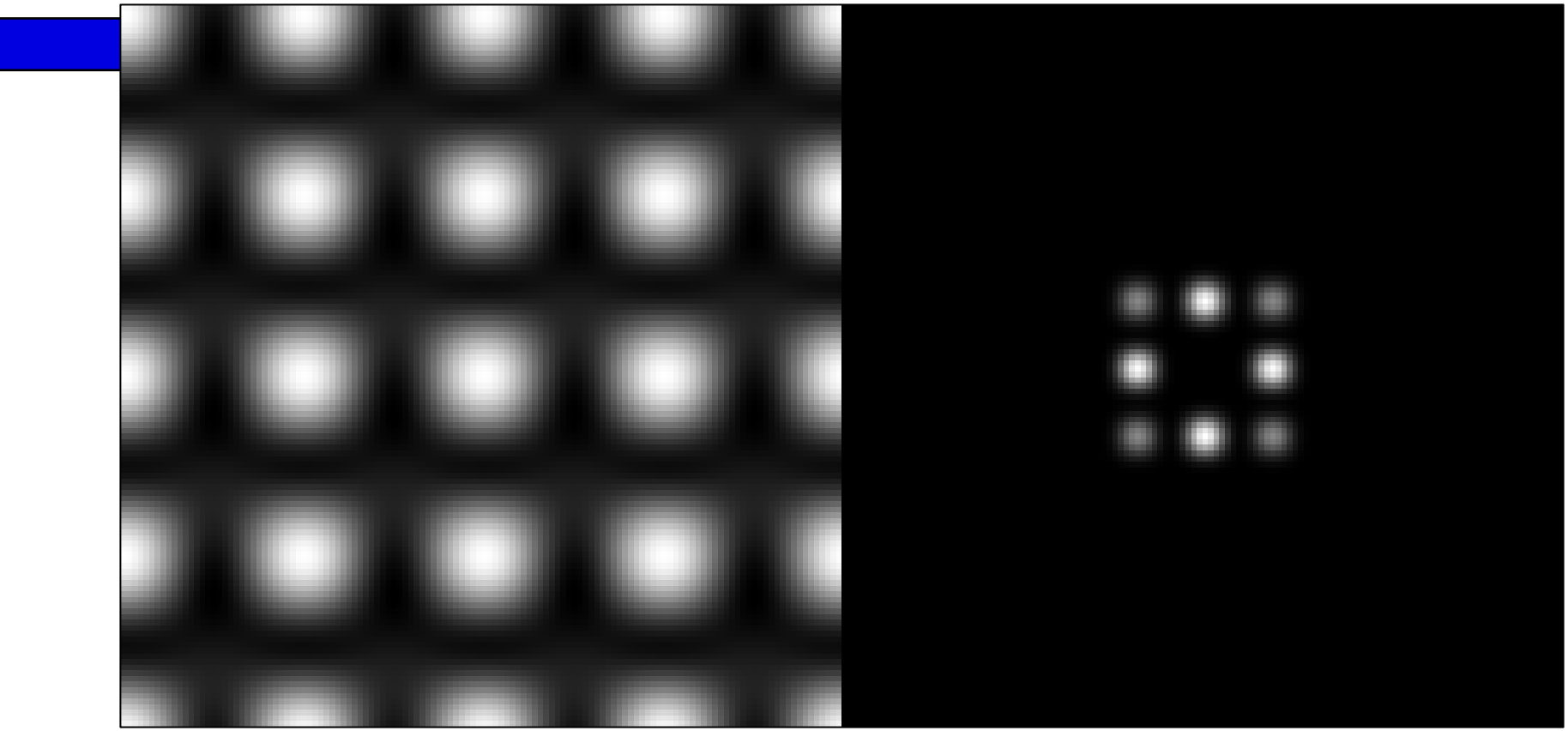
Computed diffraction pattern of structure at left.



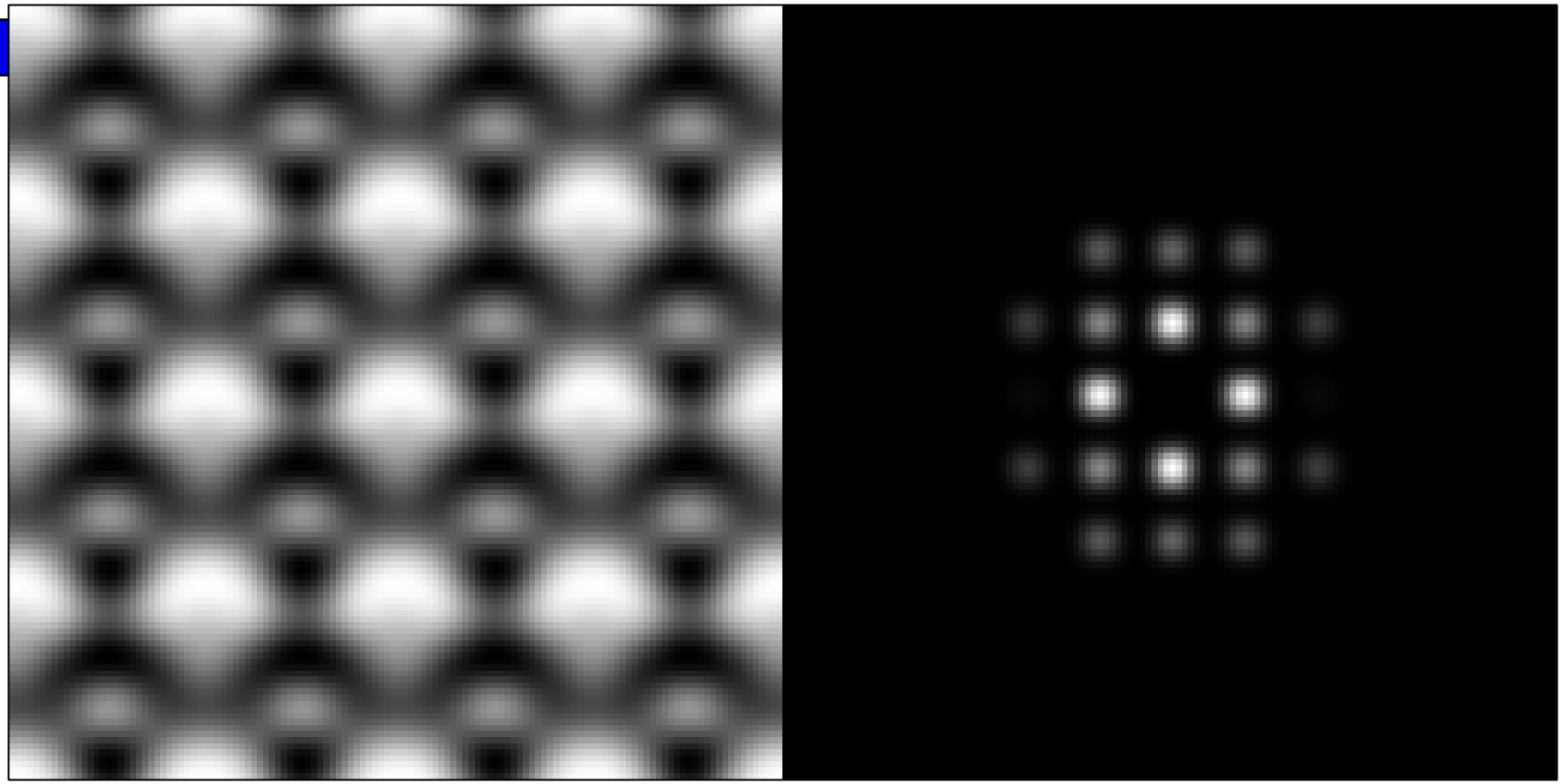
Low Resolution ...



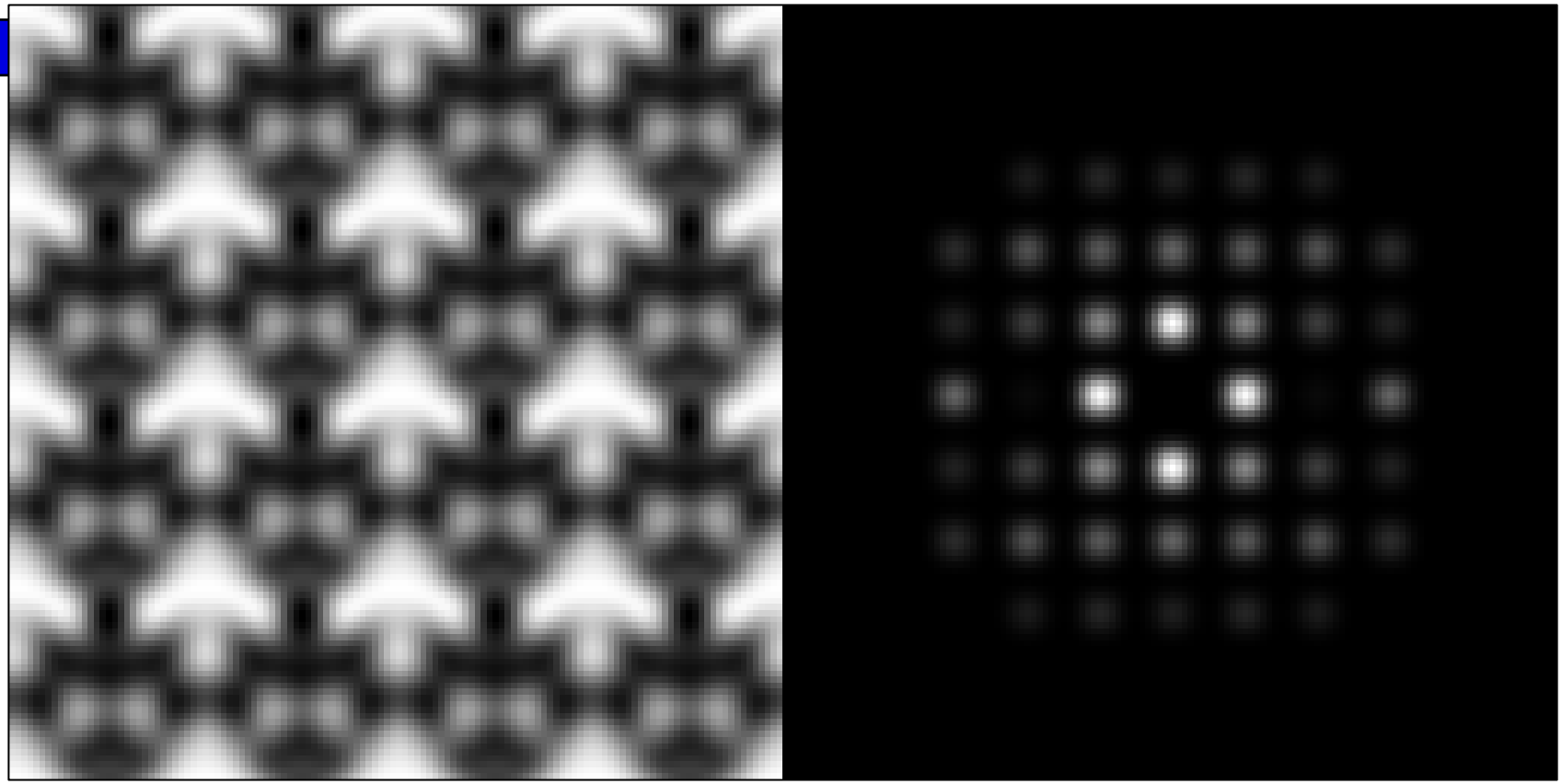
Higher resolution …



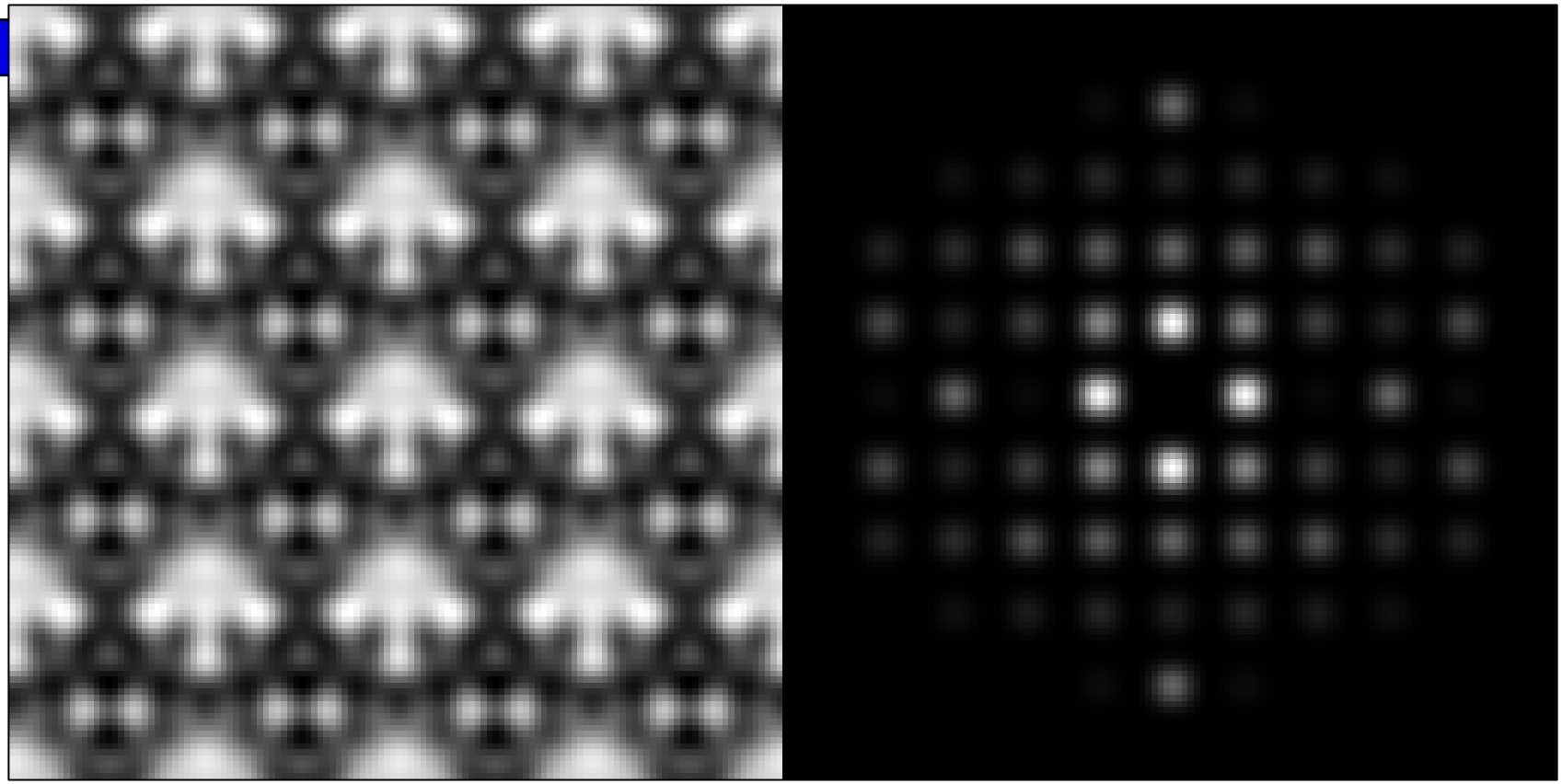
Higher resolution ...



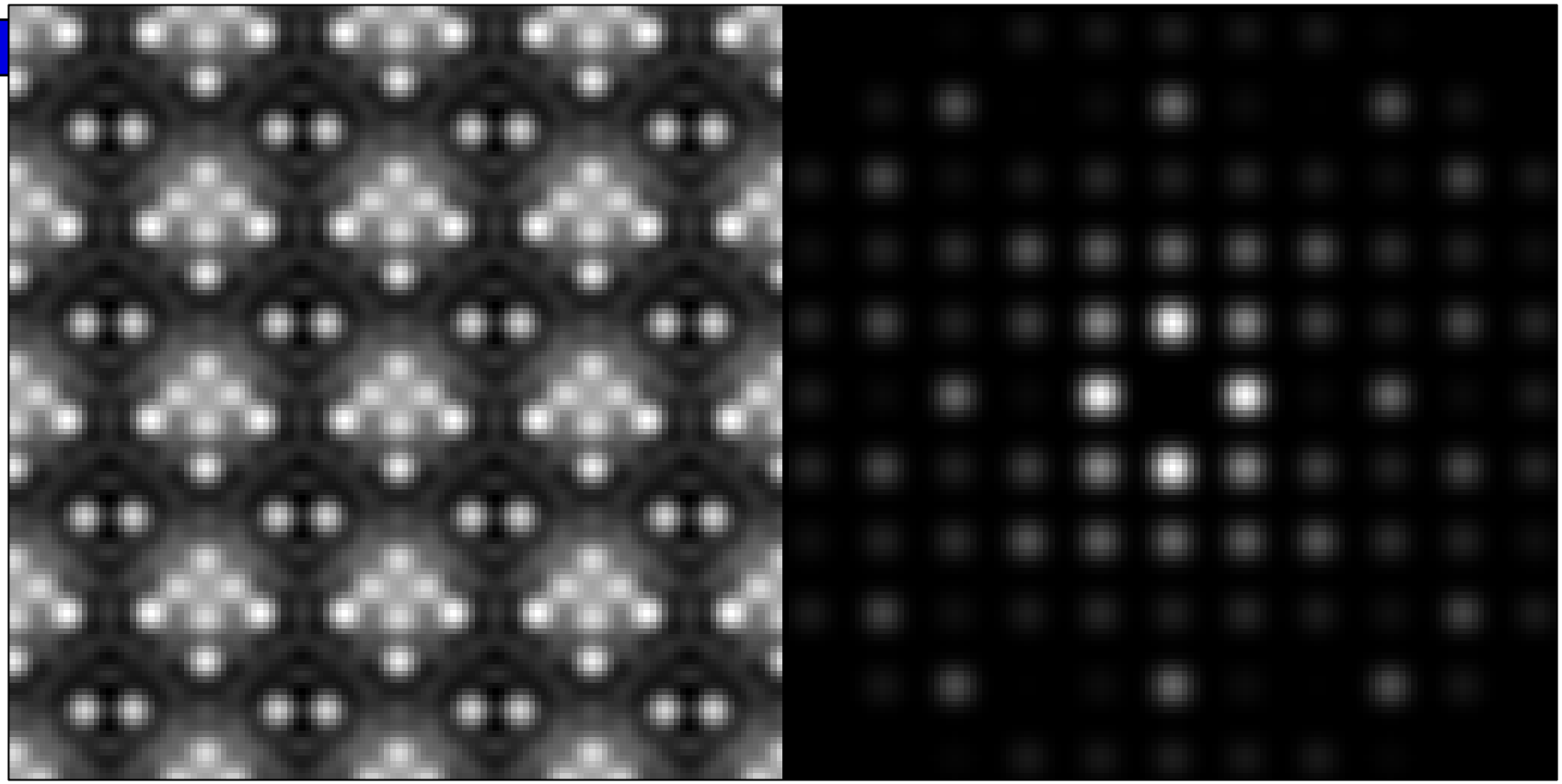
Higher resolution …



Higher resolution …



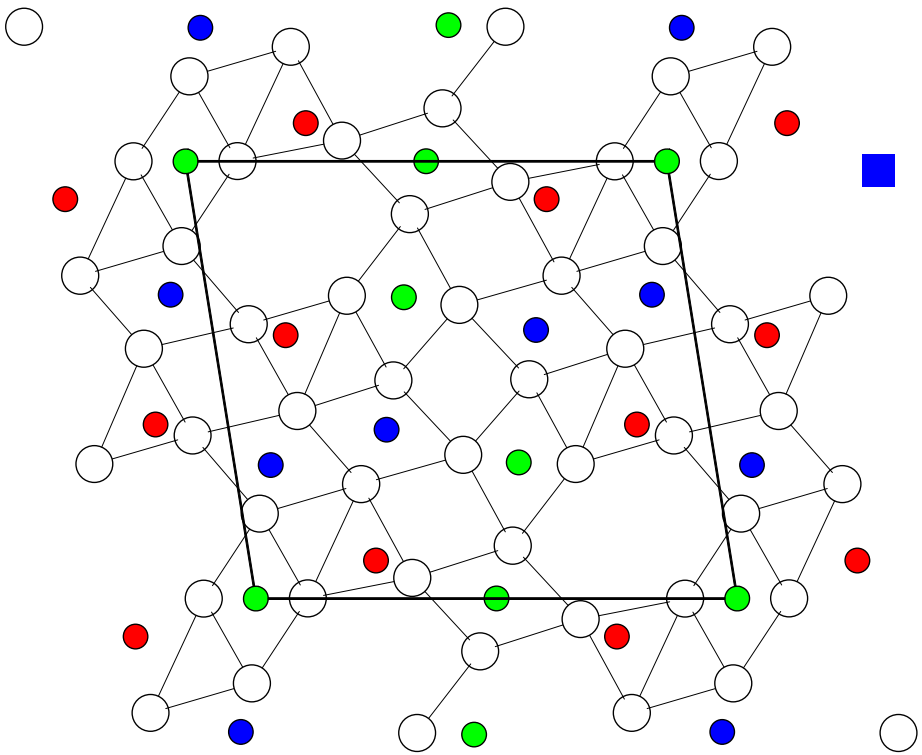
This is the goal of direct methods. Given measurement of amplitudes, obtain phases using educated guesswork. As illustrated, good phases give accurate representation of structure.



Methods

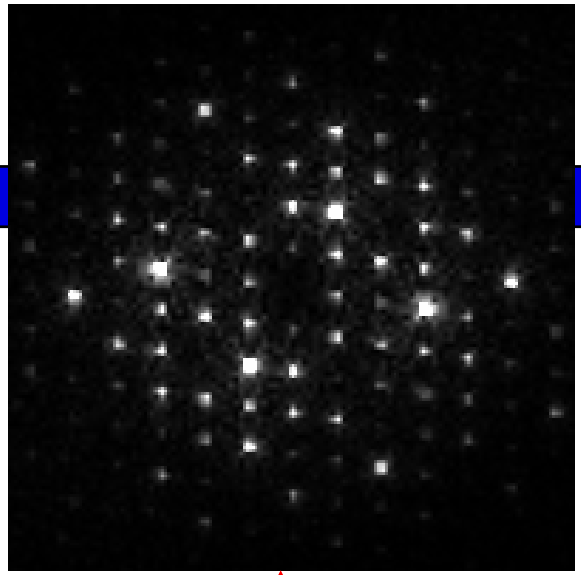
- “Classical” HREM
  - Yields low resolution image
  - Phases for some reflections directly determinable
- Diffraction using a small probe (~5 nm diameter)
  - Reduces perturbations from orientation/thickness changes
  - Dynamical Diffraction (NOT Kinematical)

Neutron Refined Final Structure

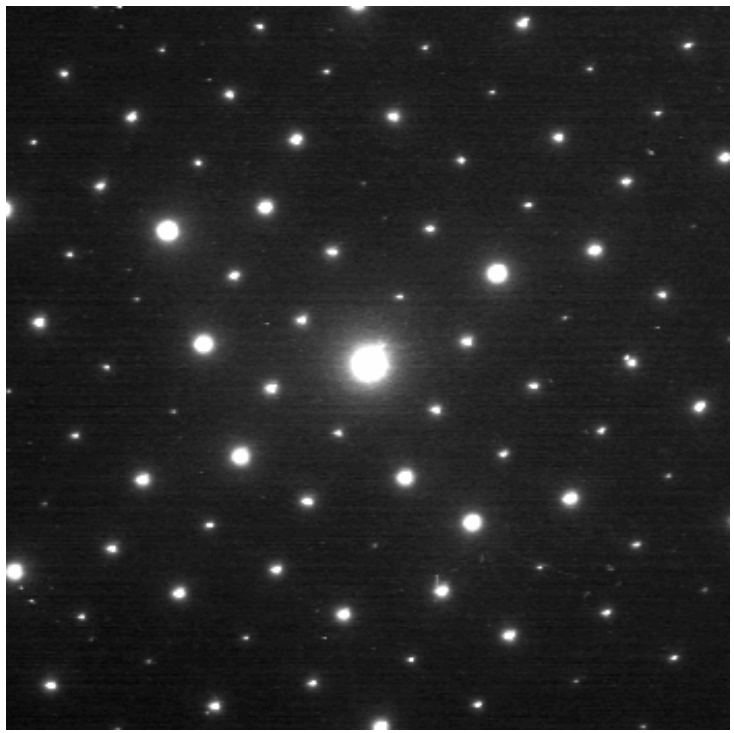


- Used HREM/TED sites as initial model
- Errors 0.01-0.02nm

Method

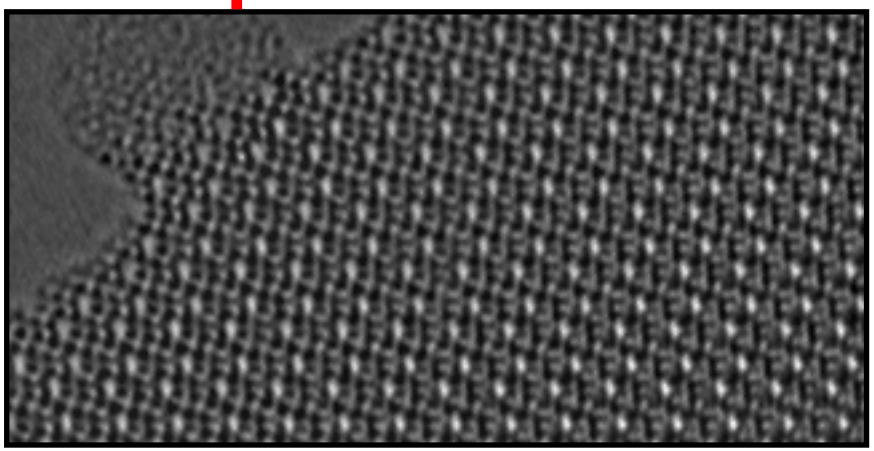


Initial
Phases



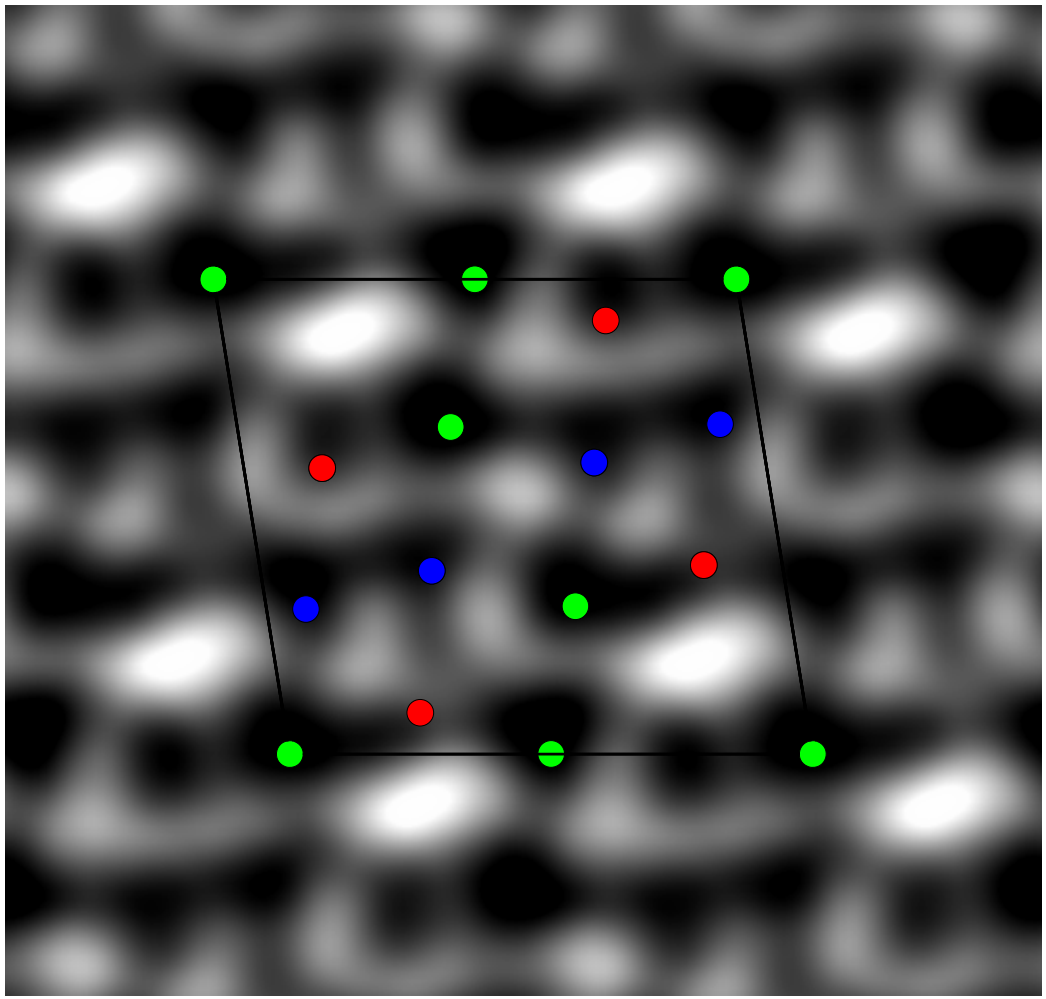
Nanoprobe
Diffraction Data

FFT

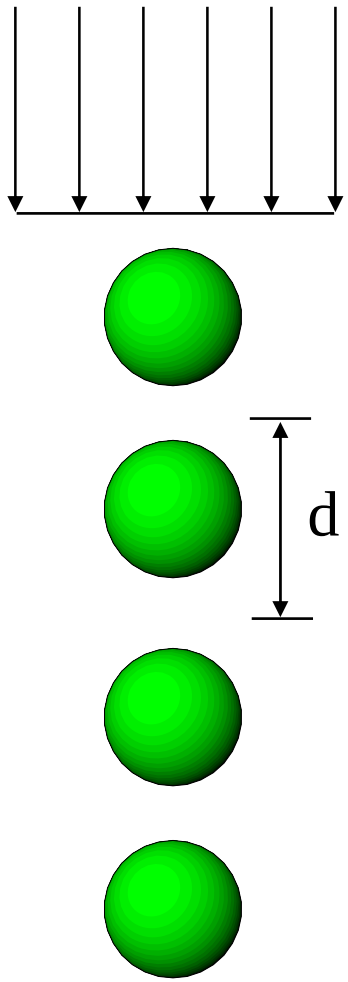


Conventional HREM Image

Cation sites in $(Ga,In)_2SnO_5$ determined from HREM.



Channeling Approximation



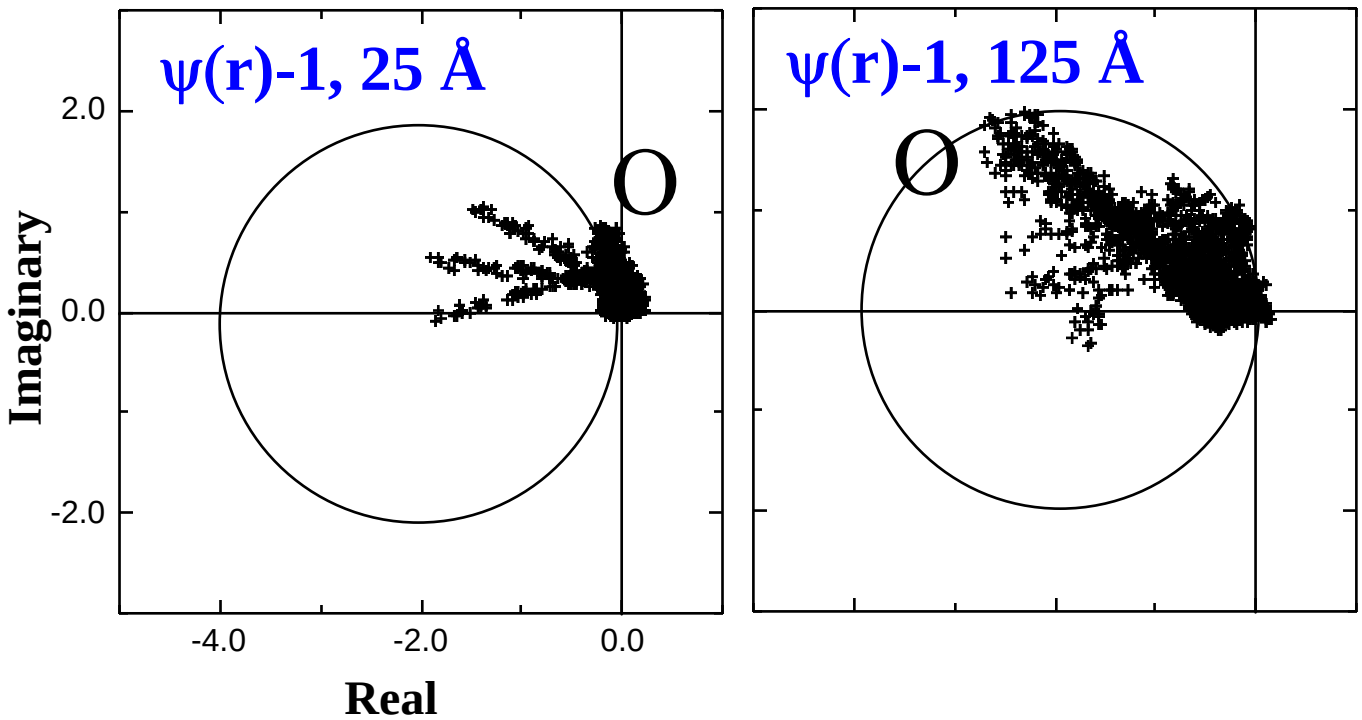
$$\mathbf{V}(\mathbf{x},\mathbf{y}) = \frac{1}{\mathbf{d}} \int_{-\infty}^{\infty} \mathbf{V}_0(\mathbf{r})\mathbf{dz}$$

$$\psi(\mathbf{r},\mathbf{z}) = \sum_{\mathbf{n}} \mathbf{C}_{\mathbf{n}} \Phi_{\mathbf{n}}(\mathbf{r}) \exp\{-\mathbf{i}\lambda_{\mathbf{n}}\mathbf{z}\}$$

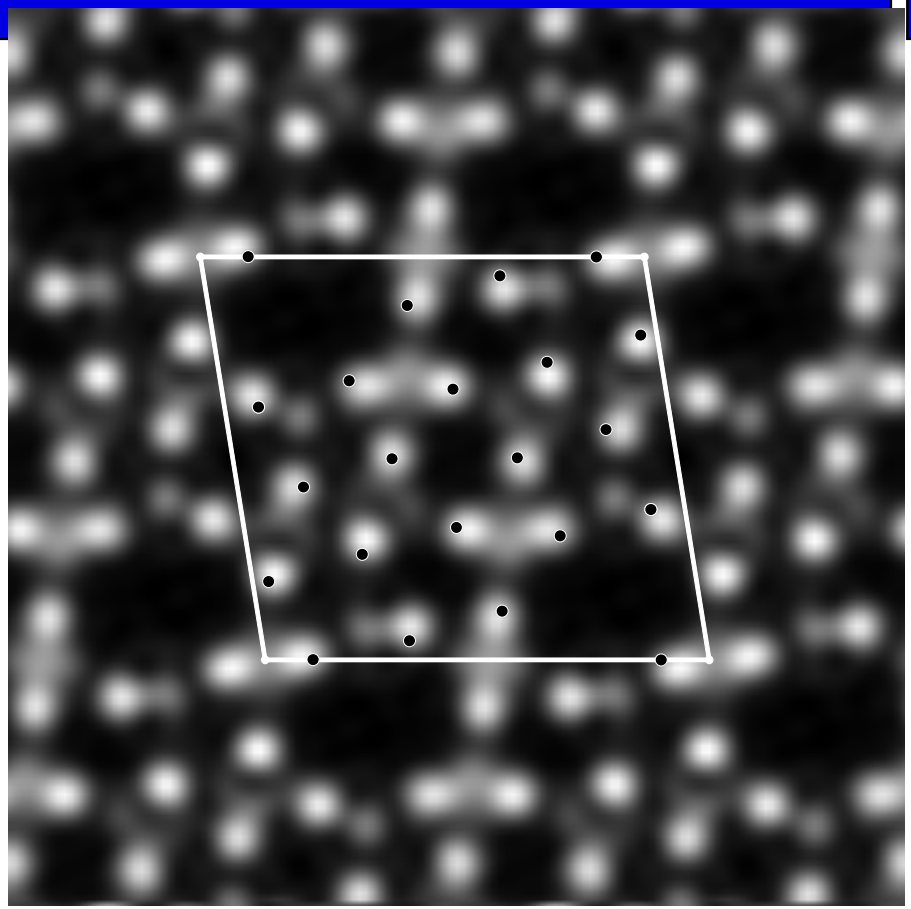
$\lambda_{\mathbf{n}}$ 2-D Eigenvalue

Quasi-Kinematical

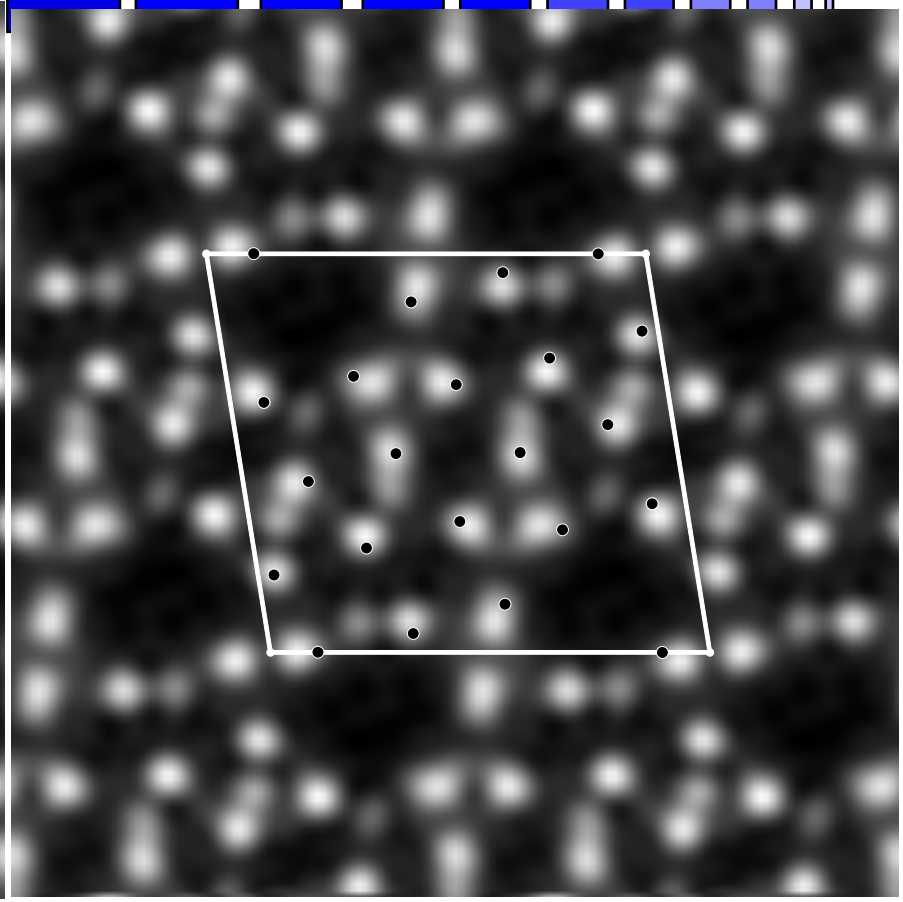
ψ(r)-1 has atom-like peaks with species-dependent oscillation



Calculated Wave



|ψ(r)-1| at 113 A thickness



|ψ(r)-1| at 202 A thickness

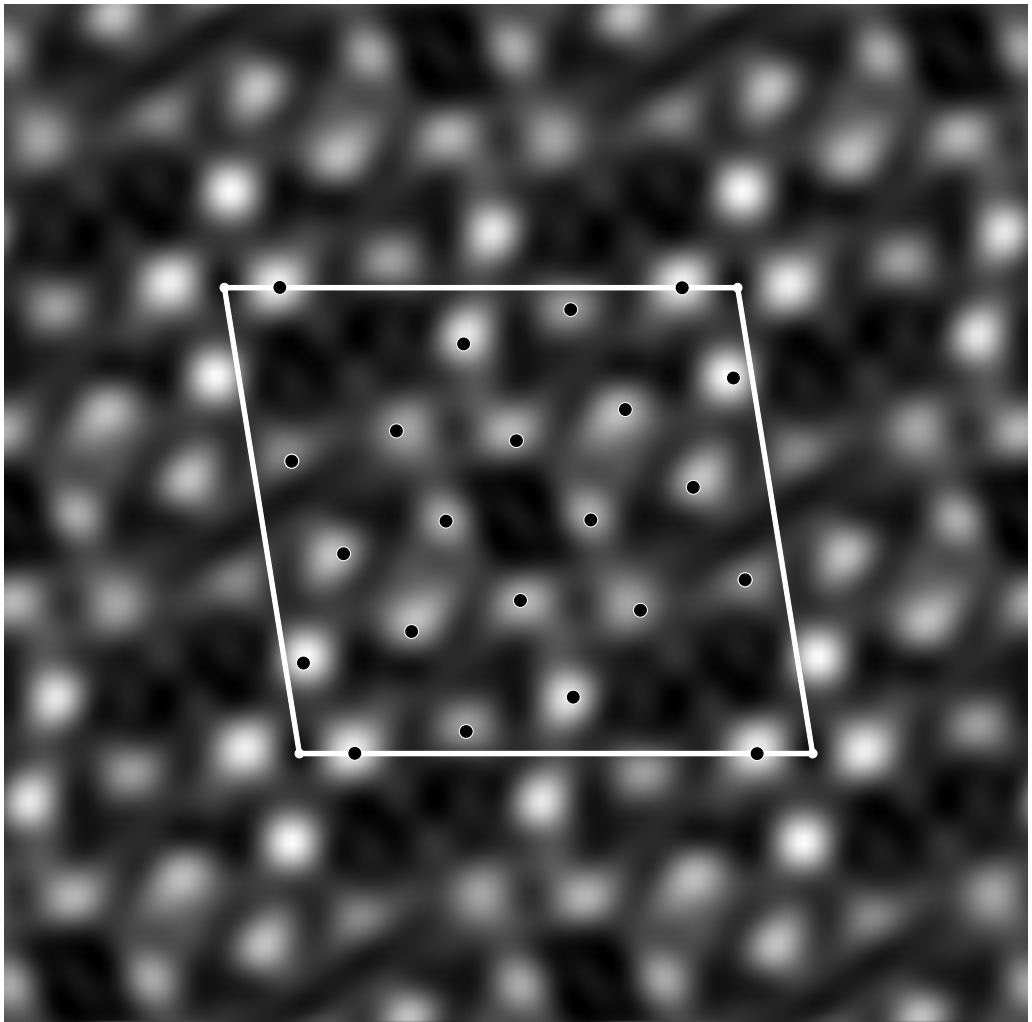
Peakiness

- Wave is sum of single-column solutions:

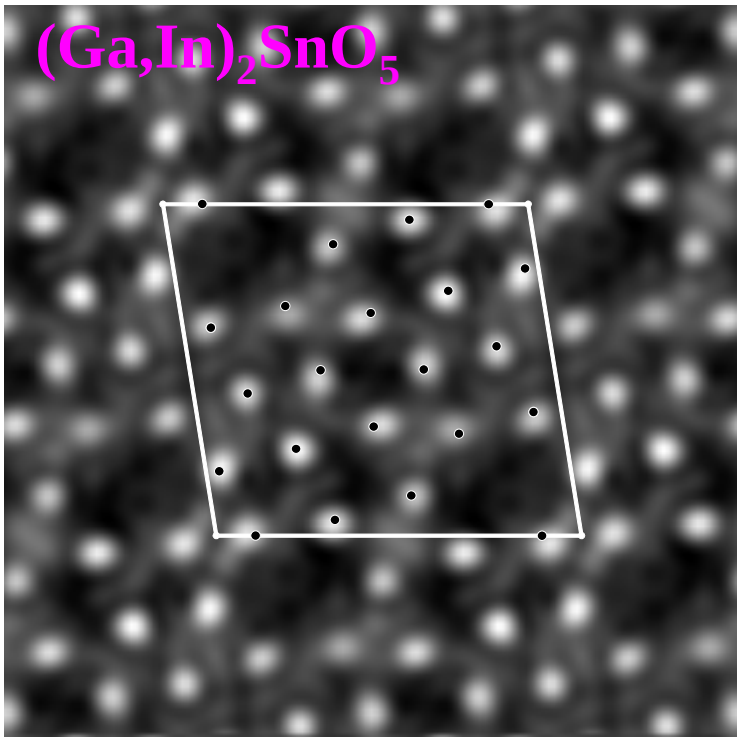
  $\psi(r,z) = \quad \Sigma\, \Phi_l(r-r_l,z)$

- Single column states $\Phi_l(r-r_l,z)$ confined close to atom sites
- Satisfies “atomicity” criteria of Direct Methods

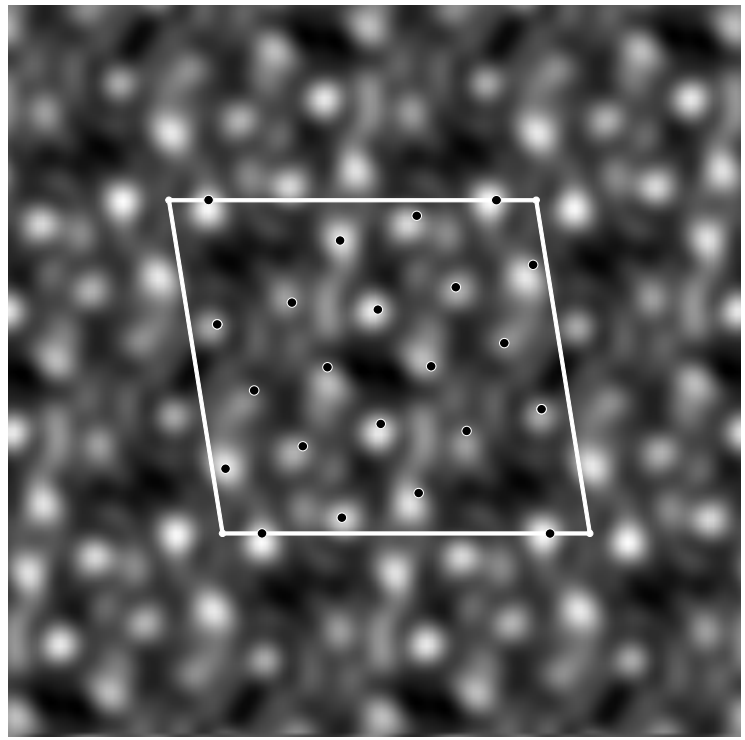
O sites in $(Ga,In)_2SnO_5$ determined using direct phasing of TED data.



Wedge Sample



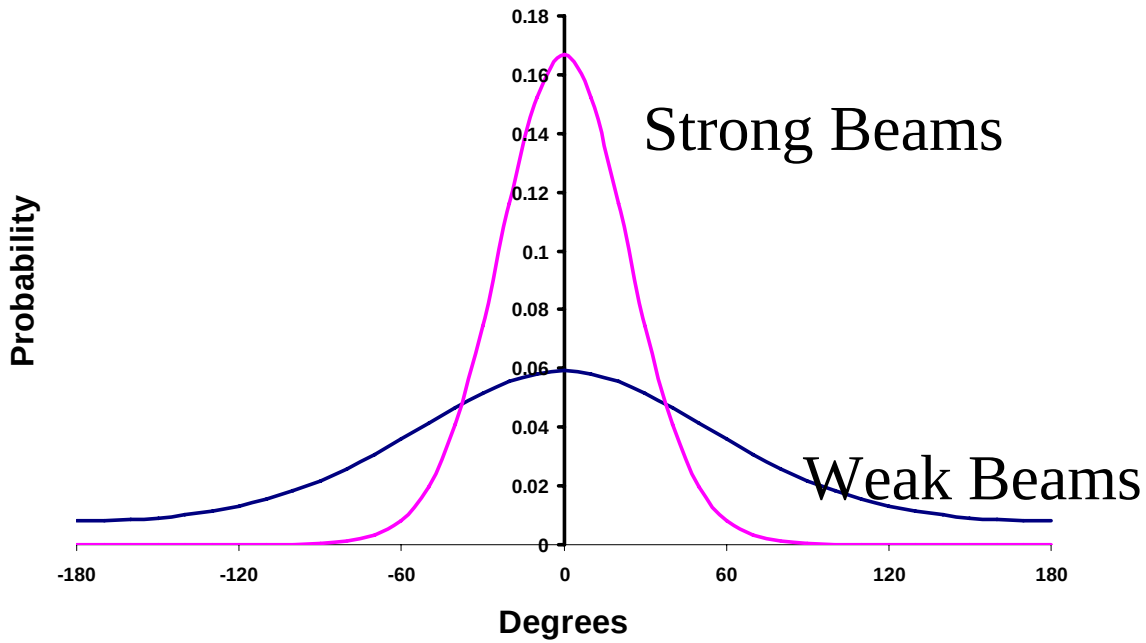
Inverted model data wedge sample 0 Å to 200 Å



Inverted experimental data, 50 nm probe

Statistical Approach (Cochran Distribution)

- This is a kinematical approach
- What happens if we consider a dynamical approach with (1st approximation) 1s channeling states?

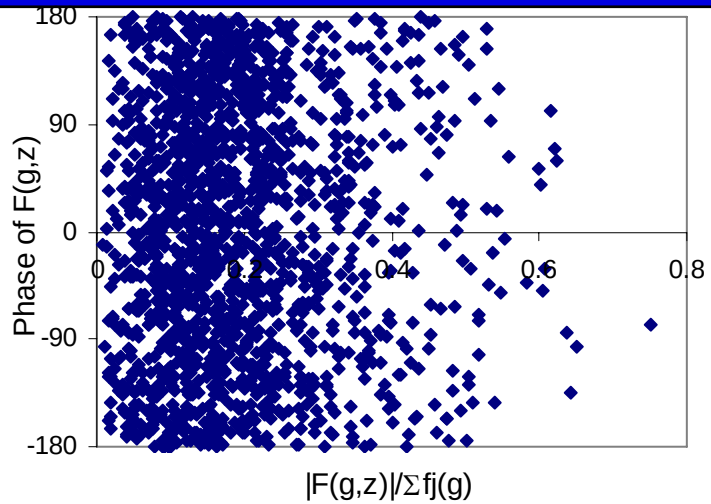


Statistics in a 1s model

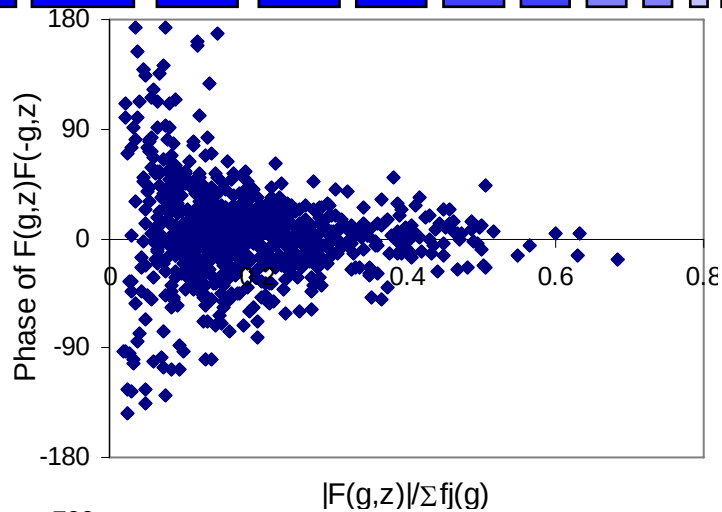
Kinematical	Dynamical
Σ_o:	Σ_o:
$\phi(g)+ \phi(-g) = 0$	$\phi(g)+ \phi(-g) \sim \omega$
$\|F(g)\| = \|F(-g)\|$	$\|F(g)\| \sim \|F(-g)\|$
Σ_2:	Σ_2:
$\phi(g)+\phi(h-g)+\phi(-h) \sim 2n\pi$	$\phi(g)+\phi(h-g)+\phi(-h) \sim \zeta$

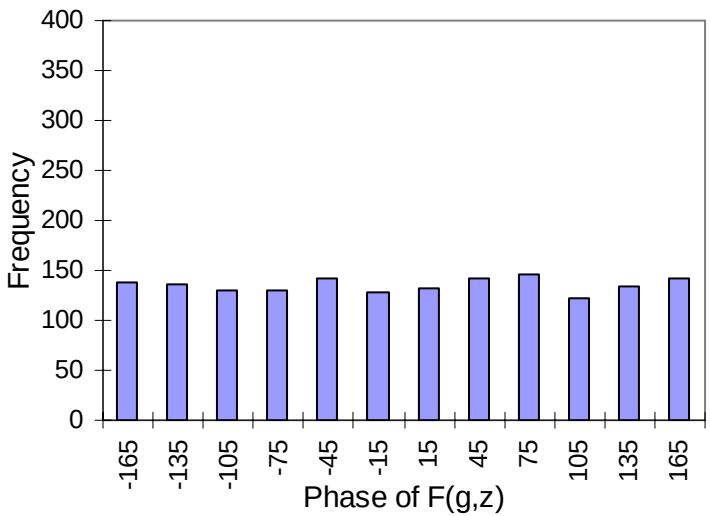
Note: derivation is tedious, uses existing theory for anomalous diffraction

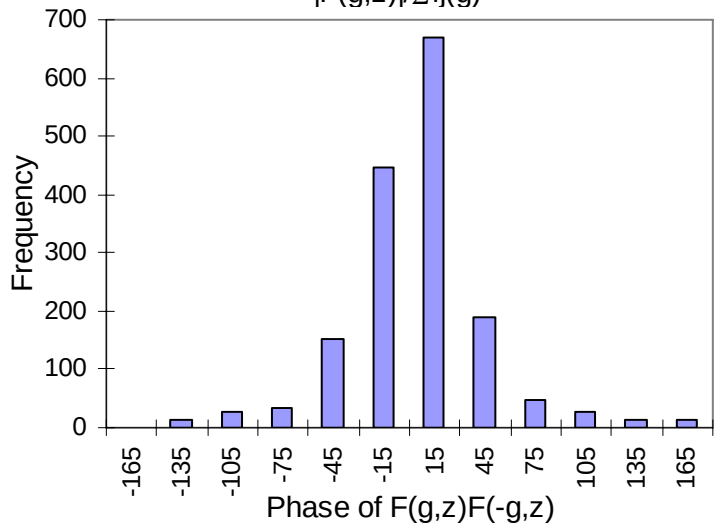
Σ_0 dynamical distribution



non-symm
5.264nm

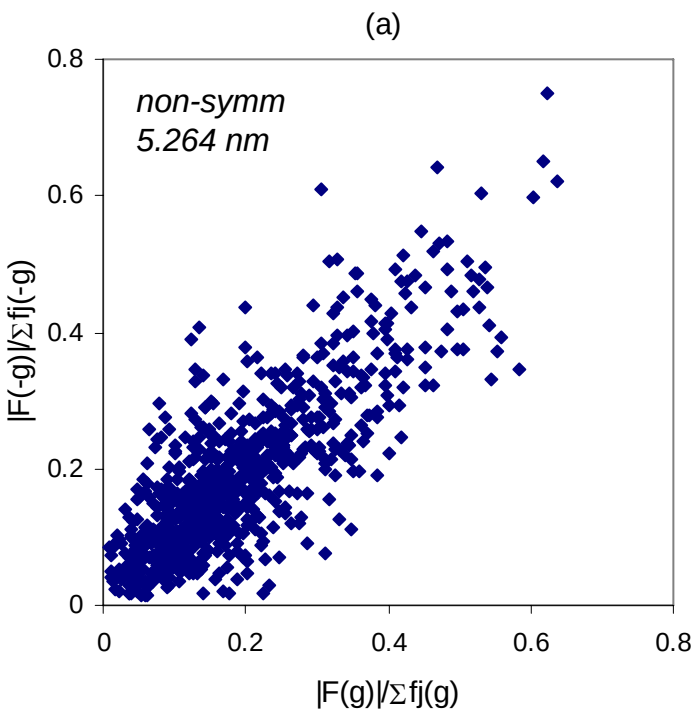


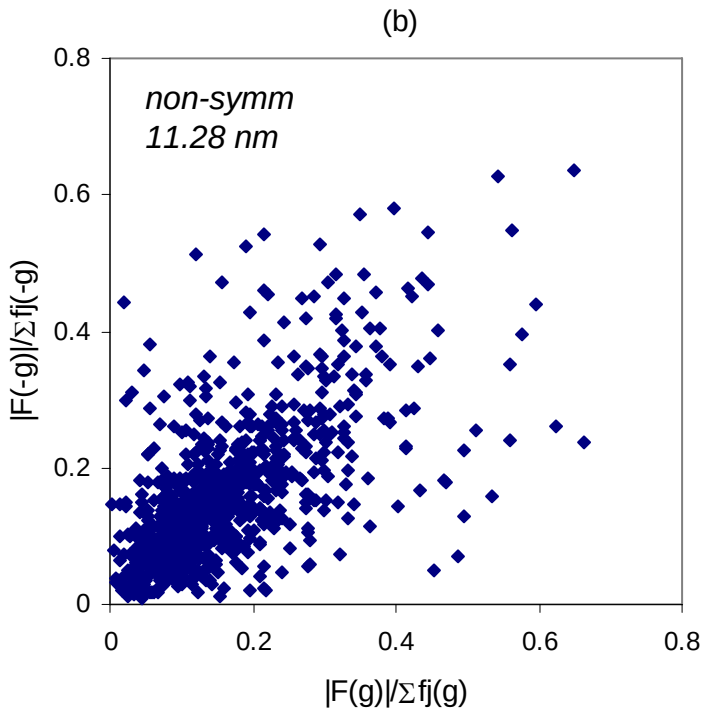




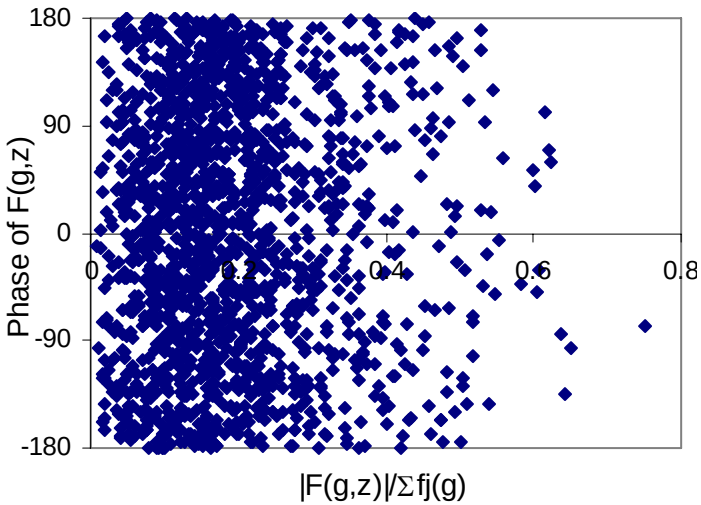
Σ_0 dynamical distribution

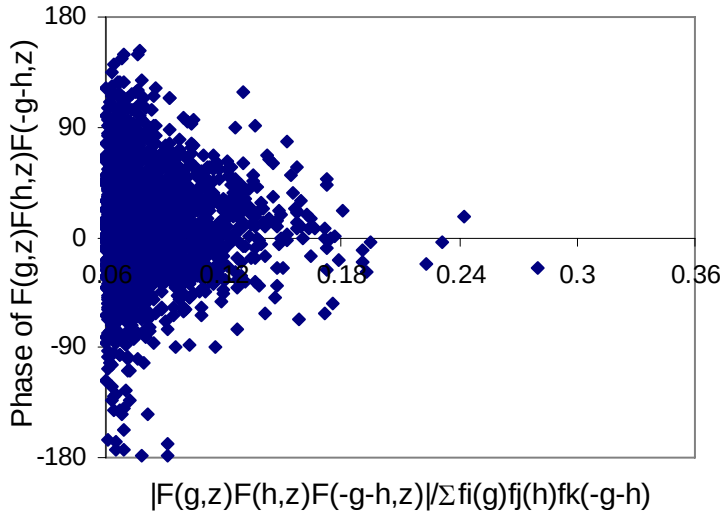
Freidel symmetry is statistical

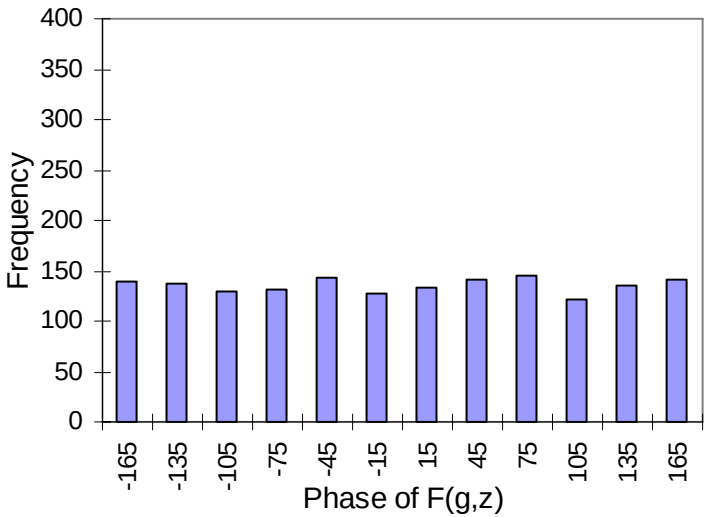


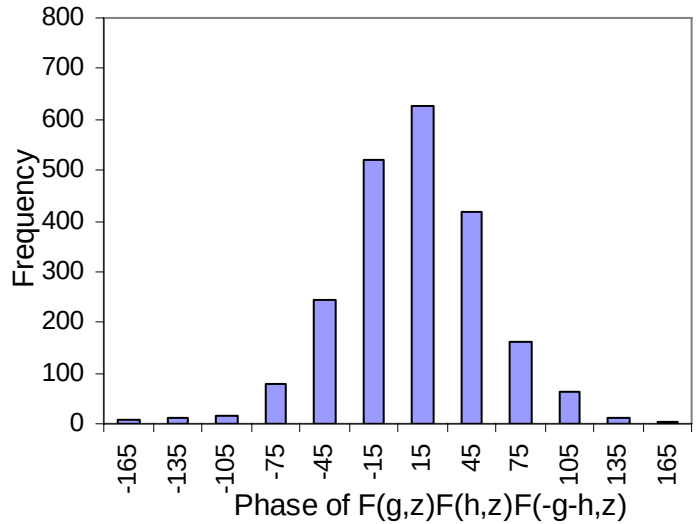


Σ_2 dynamical distribution









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Σ_2:	Σ_2:
$\phi(g)+\phi(h-g)+\phi(-h) \sim 2n\pi$	$\phi(g)+\phi(h-g)+\phi(-h) \sim \zeta$

Kinematical Theory is “wrong”, but statistically “right”

Sufficient Conditions

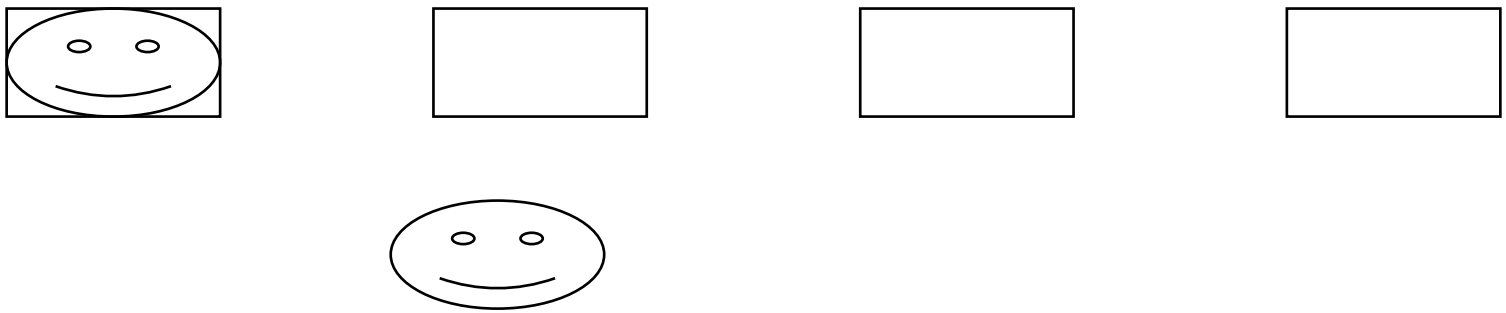
1. Kinematical Scattering
2. Intensity mapping:I(k)>I(k') iff |F(k)|>|F(k')
   - Small deviations from kinematical
   - Precession diffraction
3. Scattering dominated by one type of atom

Inversion

- I(r) -- Experimental Data
- $(d_1,d_2\ldots,d_n)$ Atom positions
- Iff
  - $M^{-1}\{I(r)\} \approx (d_1,d_2\ldots,d_n)$
- Mapping M^{-1} is a legitimate inversion

But….

- $M(d_1,d_2\ldots,d_n) \neq I(r)$
- This is not a problem
- Example
  - Fit circles into squares to find center



Kinematical PseudoInversion

- Peaks in image (or Direct Methods restoration)
- Center of peaks may be a legitimate pseudoinverse
- Iff Patterson Function for dynamical exit wave ~ Kinematical Patterson Function