The Phase Problem: Examples

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Kinematical

- Easy to interpret images
- Diffraction data can be inverted

When is image interpretation easy?

- Frequently
- Obscurations of dynamical diffraction & non-linear imaging often “don’t matter”
- Why?
  - Kinematical is violated statistically, not systematically in most cases
  - Hence it is a valid first approximation

When does it work?

- Exact problems (Diffractive Imaging)
- Kinematical Diffraction (surfaces)
- 1s-Channelling (HREM+HAADF)
- Intensity ordering (PED)

L. D. Marks, W. Sinkler, Sufficient conditions for direct methods with swift electrons. *Microsc. Microanal.* **9**, 399 (2003).

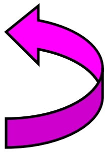
Exact Cases

- Suppose we have N pixels, and N/2 are known to be zero (compact support)
- Wave is described by N/2 moduli, N/2 phases (for a real wave) in reciprocal space
- Unkowns – N ; measurements N/2 ; contraints N/2
- Problem is in principle fully solveable

(It can be shown to be unique in 2 or more dimensions, based upon the fundamental theorem of algebra)

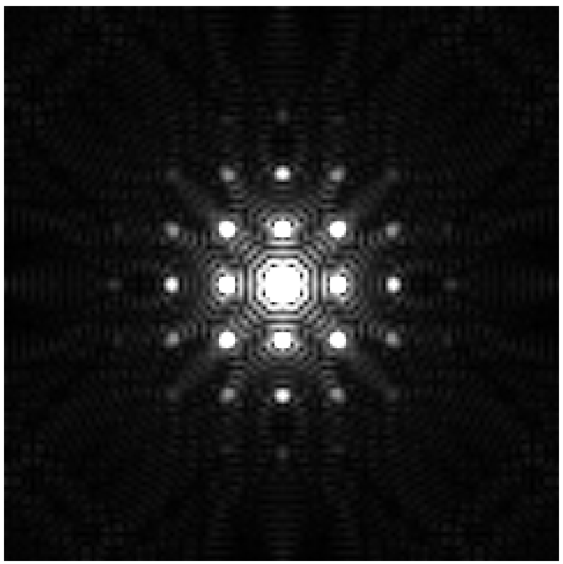
Example: Diffractive imaging

- Constraint: part of real-space x is zero (Convex constraint)
- Iteration
  - x = 0, part of map
  - $|X| = |X_{observed}|$



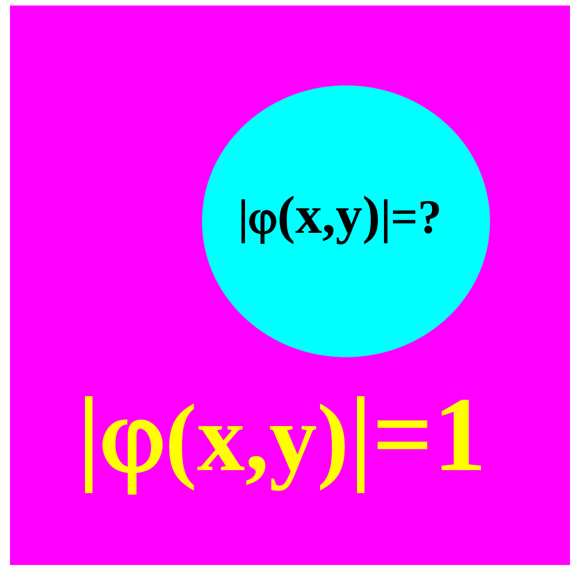
Iterate

Phase Recovery for a Small Particle



True diffraction pattern
for small particle model
(Non-Convex Constraint)

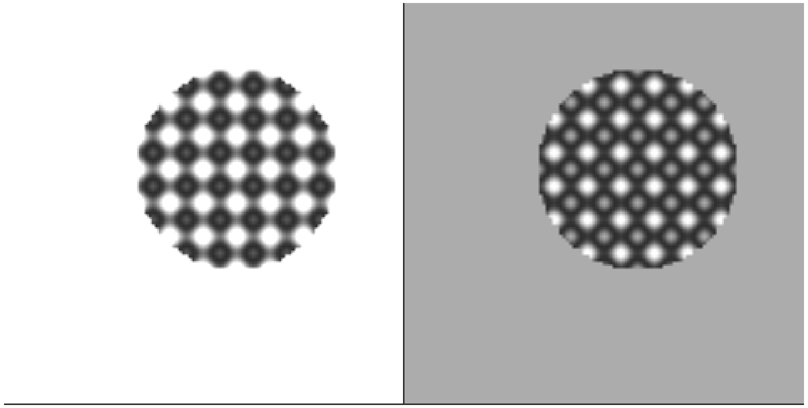
+



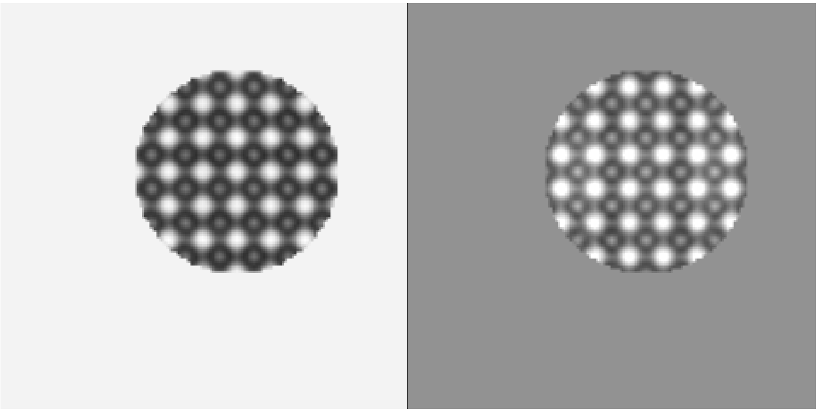
Convex Support
Constraint

= ?

Phase Recovery for a Small Particle

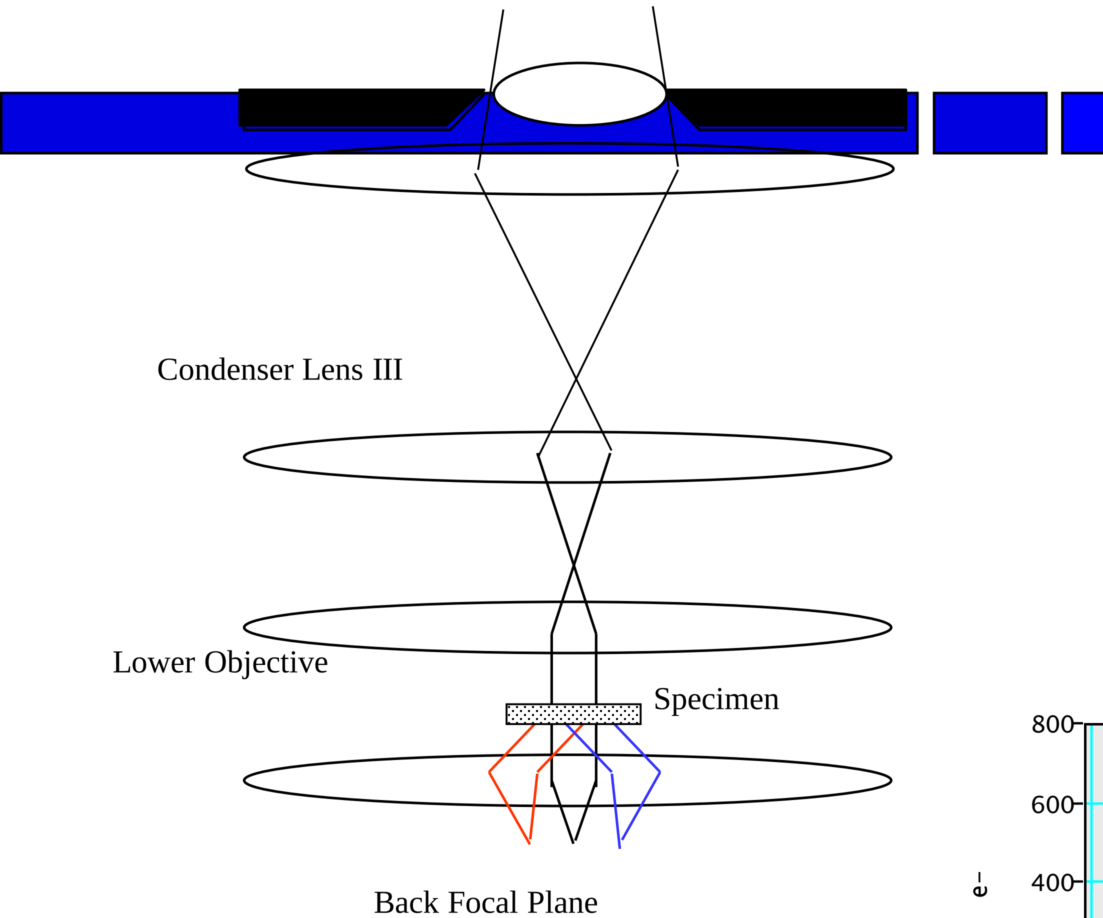


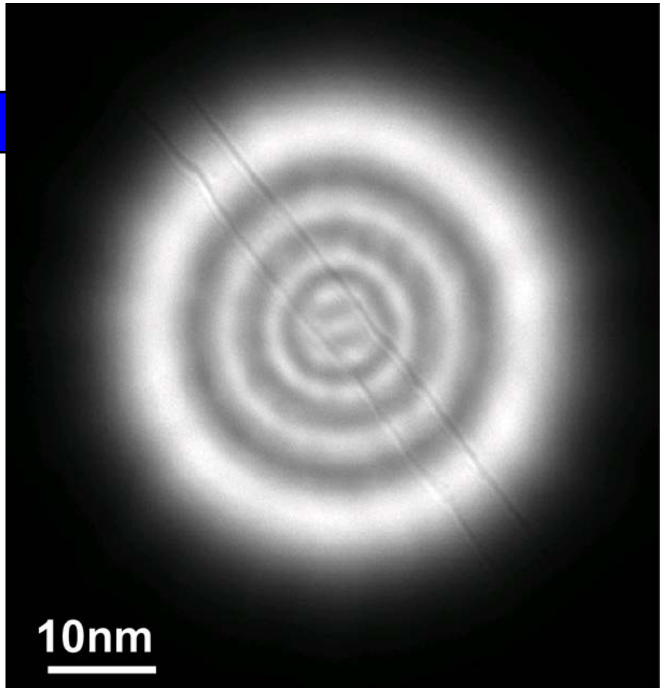
True real space exit wave for small particle model

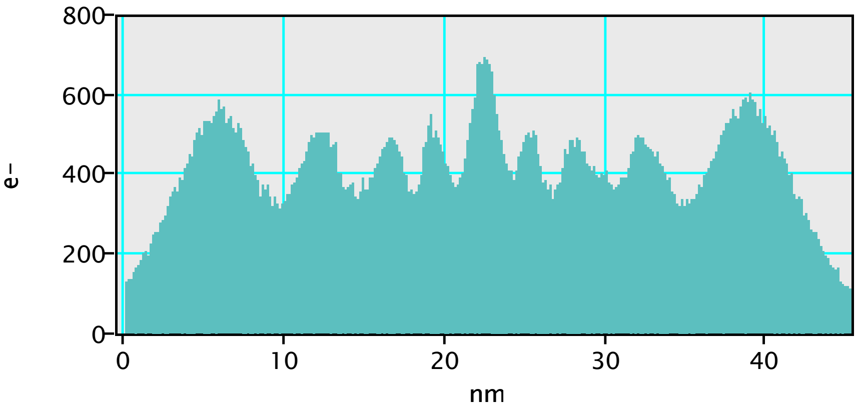


Reconstructed exit wave after 3000 iterations

Electron Nanoprobe formation

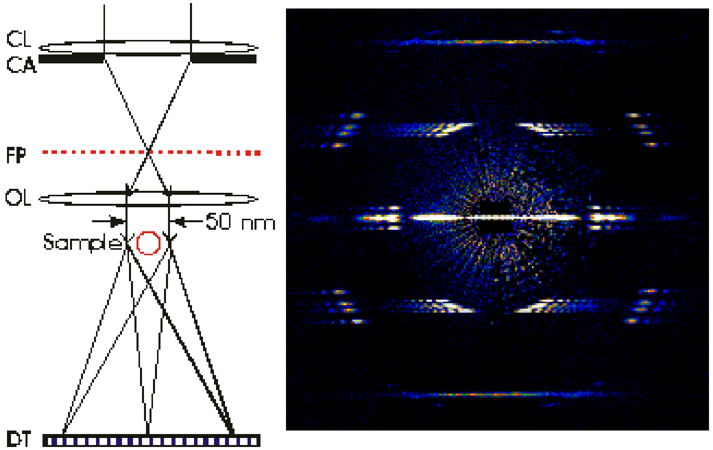




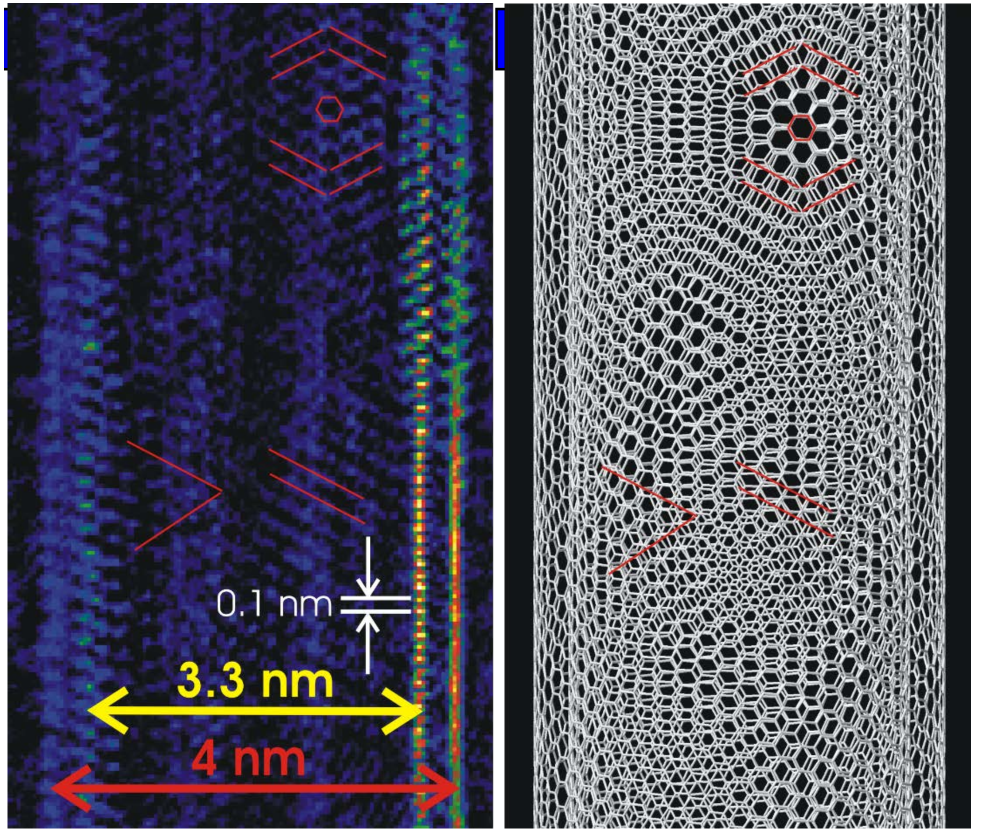


10 mm aperture -> 50 nm beam
M = 1/200

Diffractive Imaging and Phase Retrieval

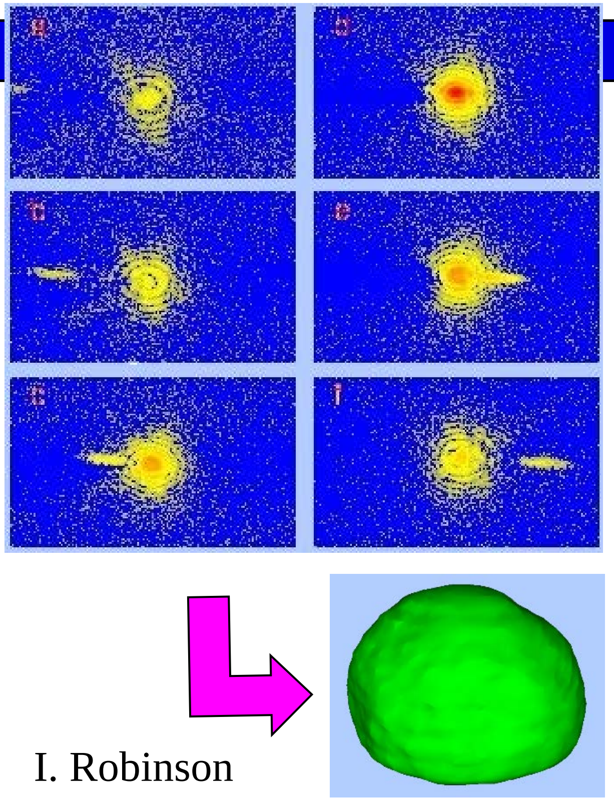


(left) A single double wall nanotube is illuminated with a narrow beam of electrons. (right) The diffraction pattern of the tube

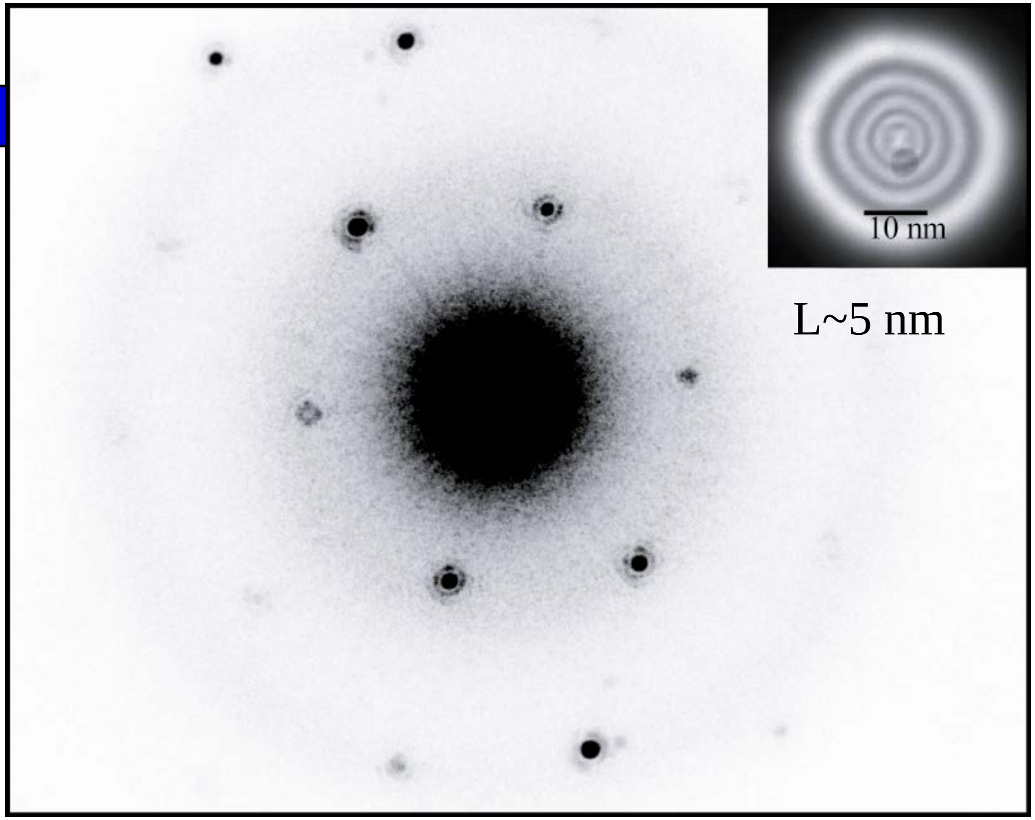


J.M. Zuo, I. Vartanyants, M. Gao, R. Zhang and L.A. Nagahara, *Science, 300,* 1419 *(2003)*

Single Particle Diffraction



I. Robinson
Synchrotron



- Atomic resolution
- Strong interaction of electrons

J. Tao, See Zuo et al, Microscopy Research Techniques, 2004

When does it work?

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- Kinematical Diffraction (surfaces)
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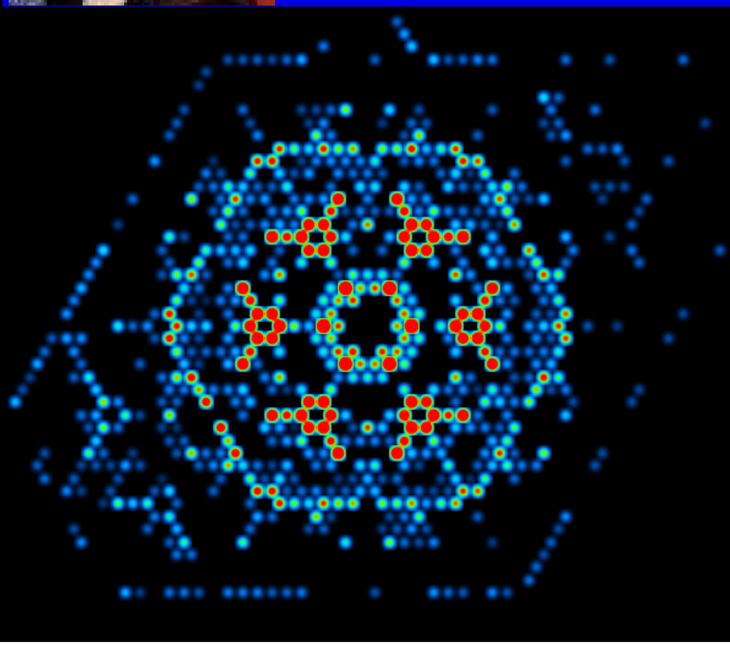
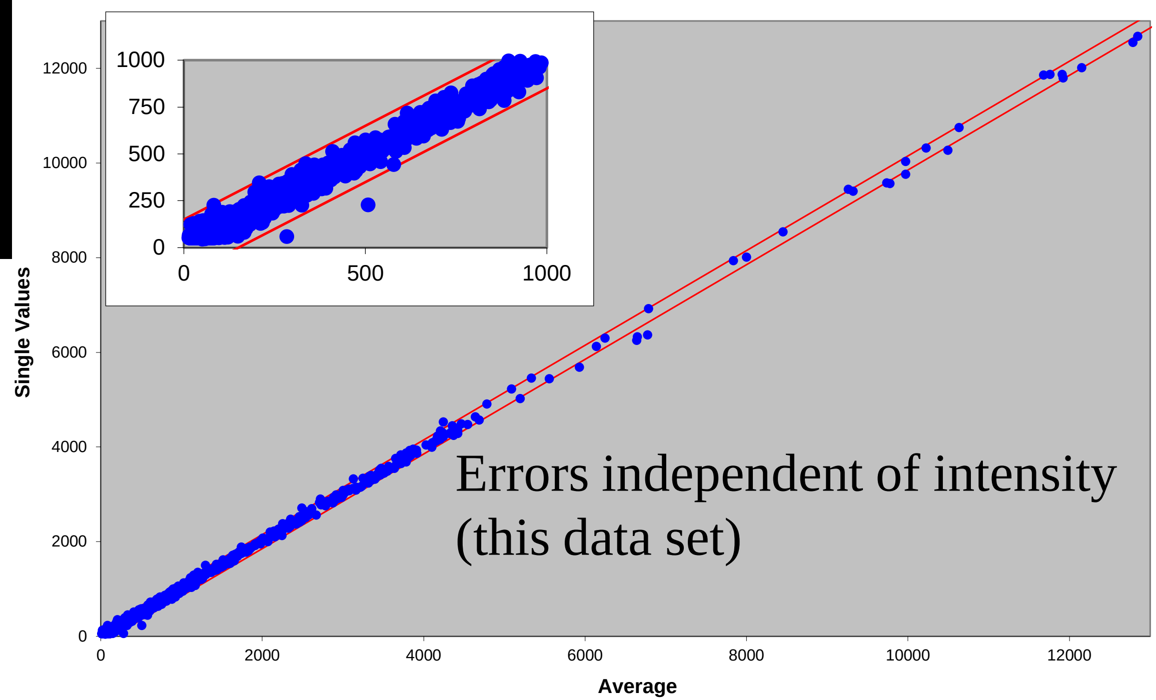
L. D. Marks, W. Sinkler, Sufficient conditions for direct methods with swift electrons. *Microsc. Microanal.* **9**, 399 (2003).

TED: Si (111) 7x7



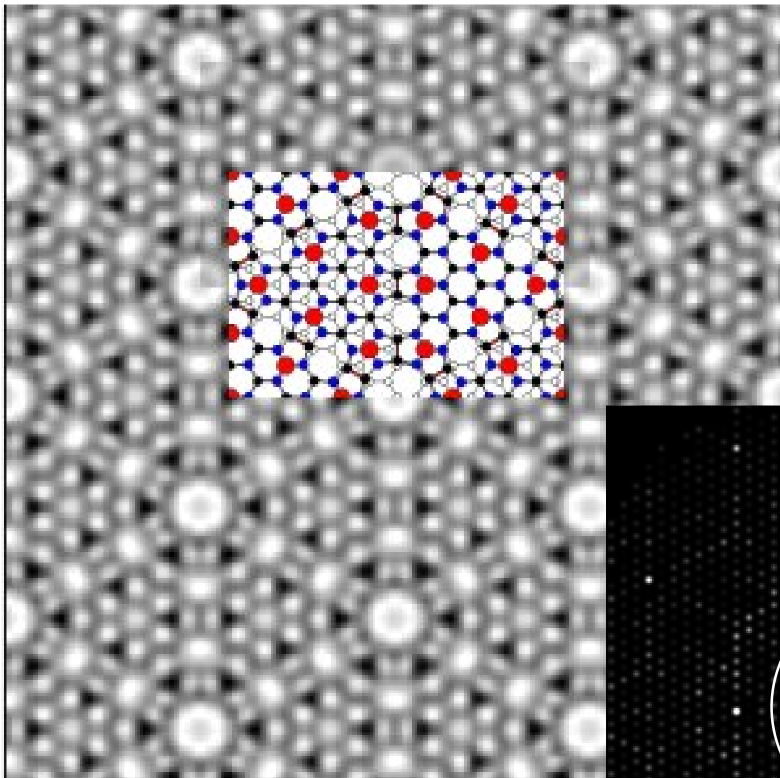
Method: Merge data for 6-20 different exposures to obtain accuracies of ~1% with statistical significance



Cross-Correlation Method

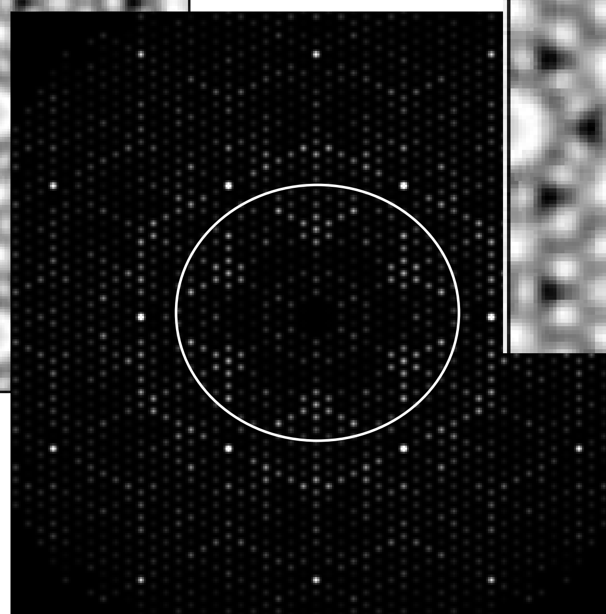
P. Xu, et al. *Ultramicroscopy* **53**, 15 (1994).

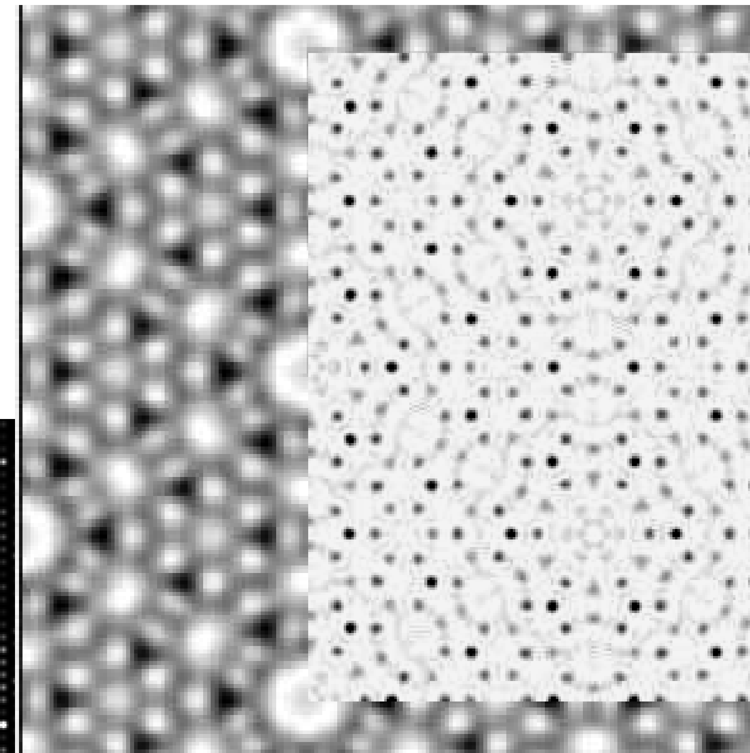
Restoration and Extension



0.3nm Image

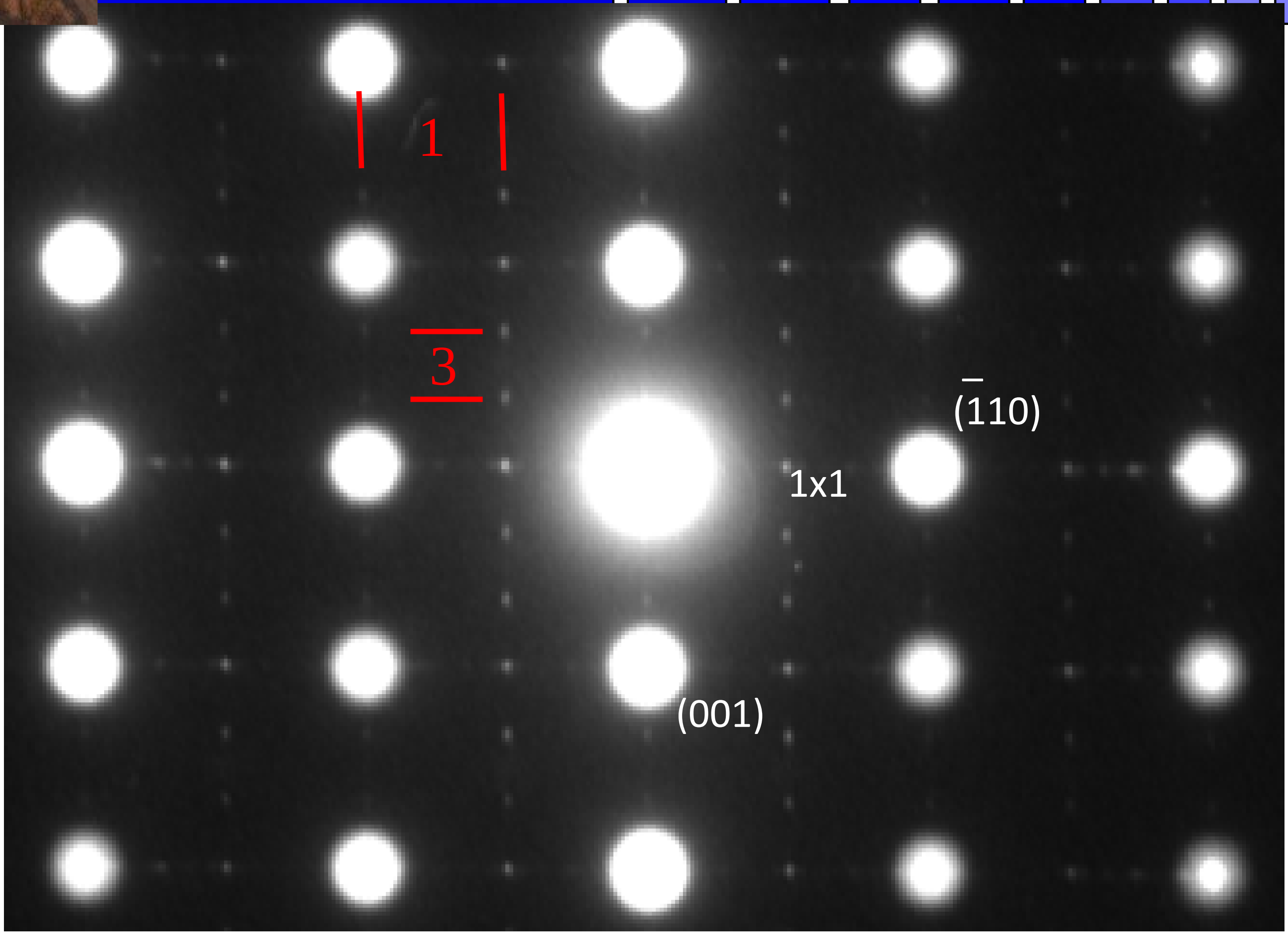
+DP



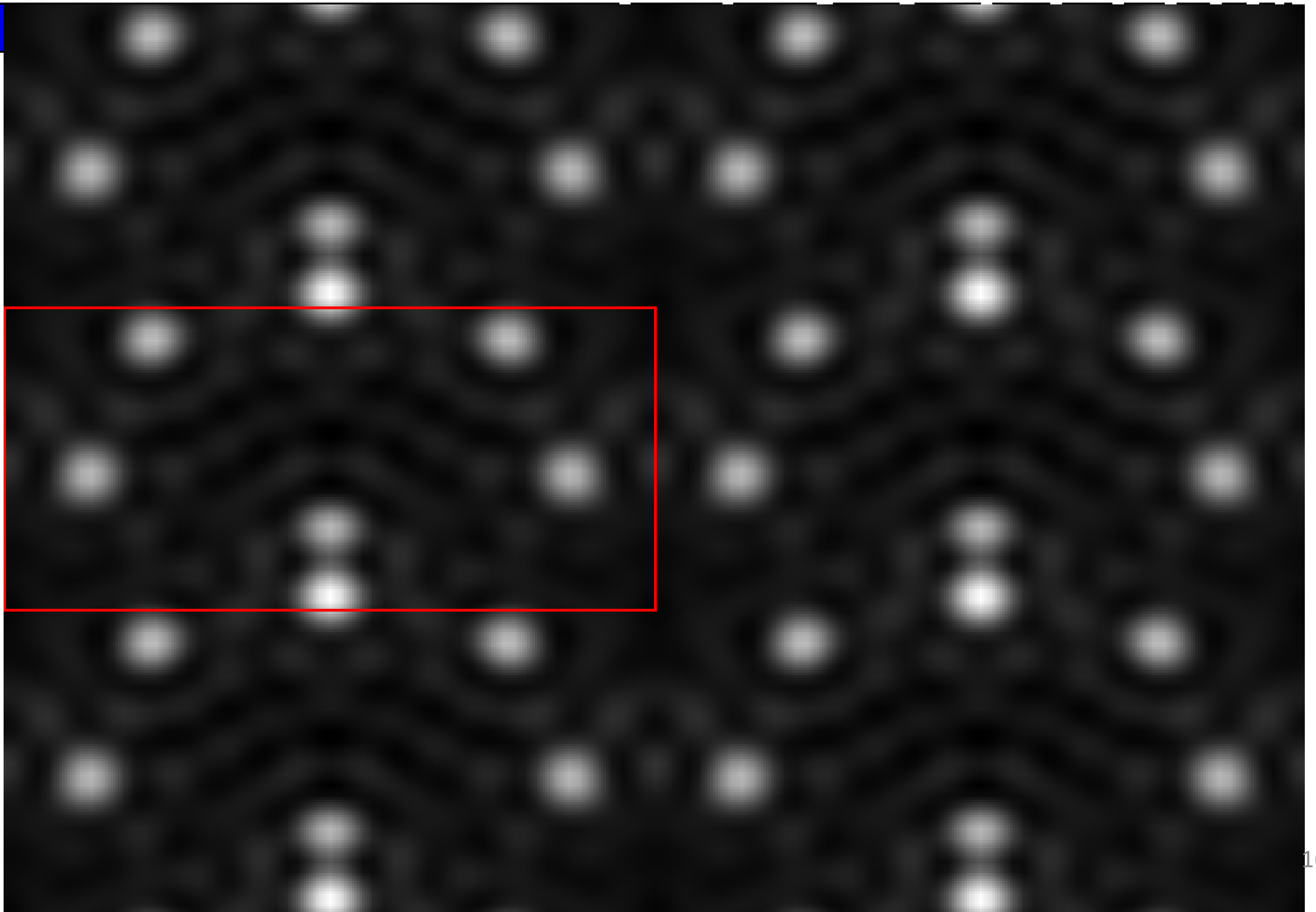


0.05nm Image

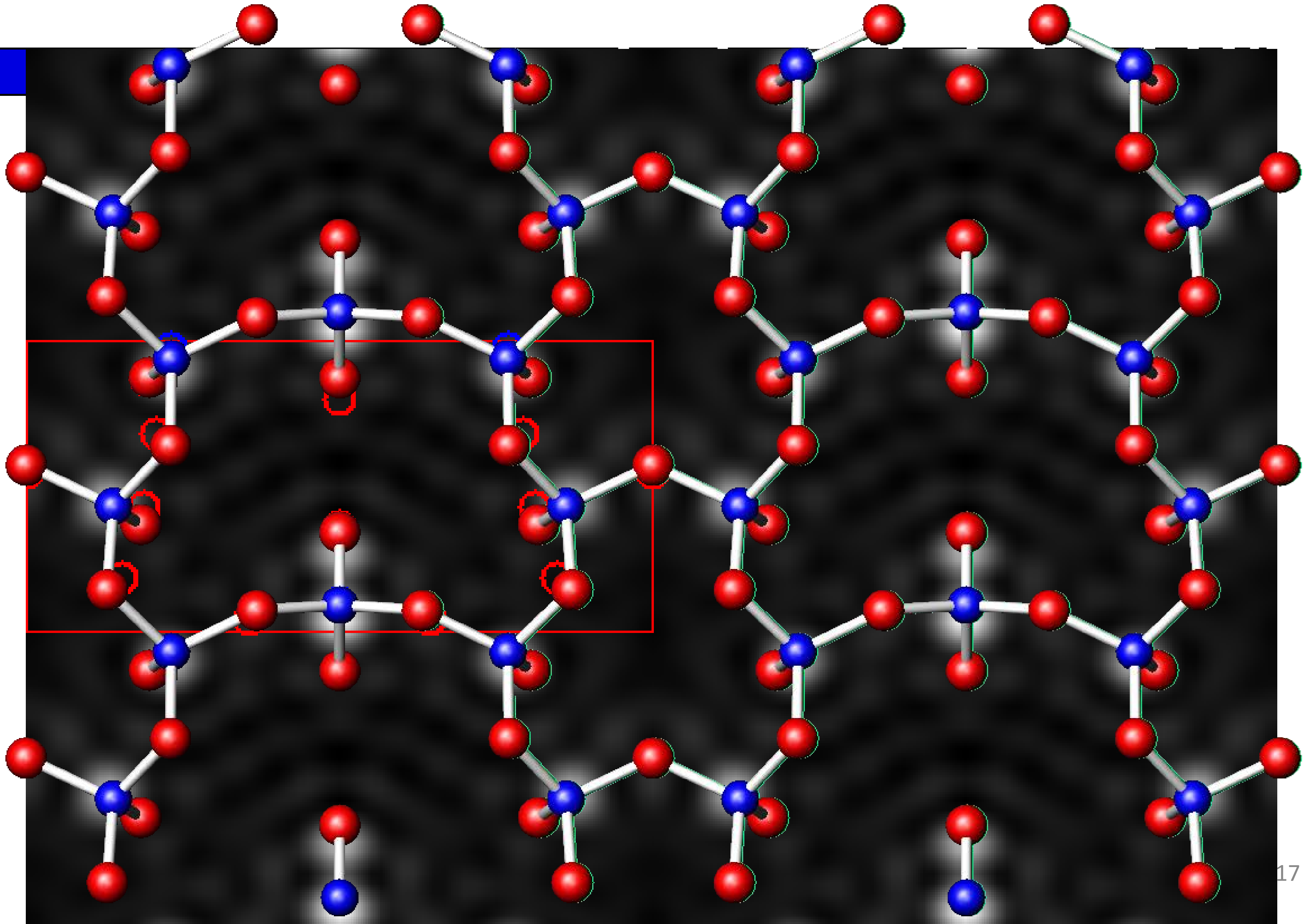
1000 °C in flowing O_2



Direct Methods Solution

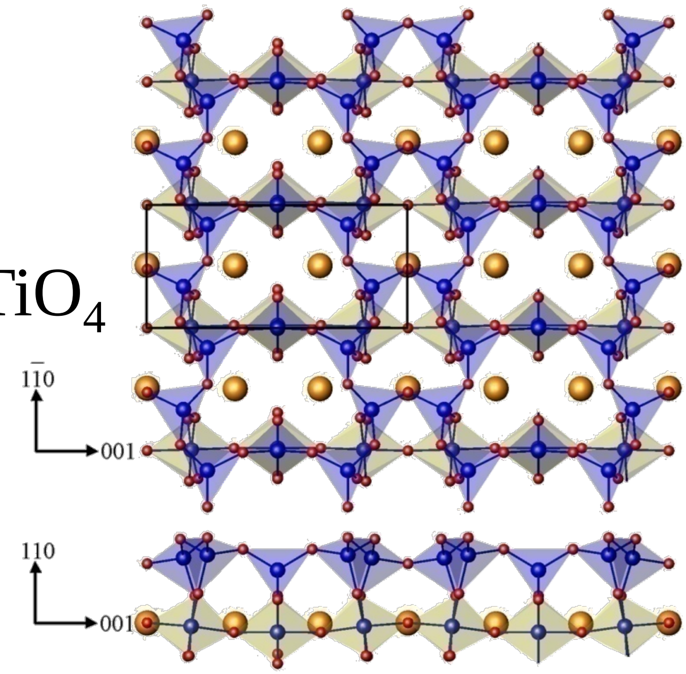


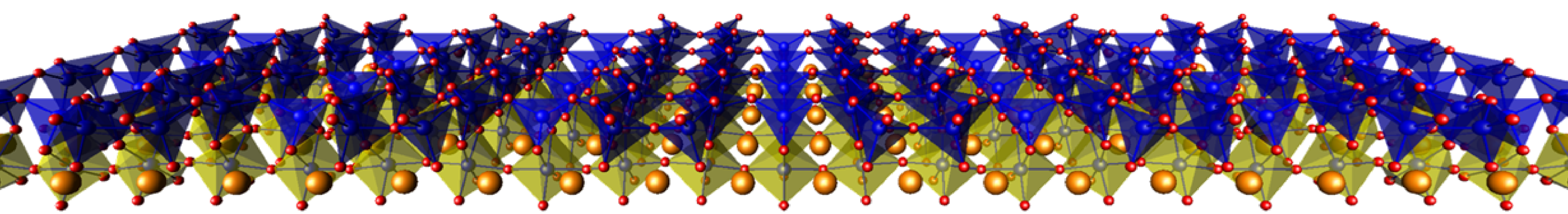
Atomic Positions Refined



$SrTiO_3$ (110) 3x1

- TiO_2 overall surface stoichiometry
  - Ti_5O_7 atop O_2 termination
  - Ti_5O_{13} atop SrTiO termination
- Surface composed of corner sharing TiO_4 tetrahedra
  - Arranged in rings of 6 or 8 tetrahedra
  - 4 corner share with bulk octahedra
  - 1 edge shares with bulk octahedron





Blue polyhedra are surface polyhedra, gold are bulk octahedra, orange spheres Sr, blue spheres Ti, red spheres O

Enterkin et. al., Nature Materials, 2010

Inversion

- $I(r) \sim \int \Psi(u) T(u) \exp(2\pi i u.r) du$ + noise

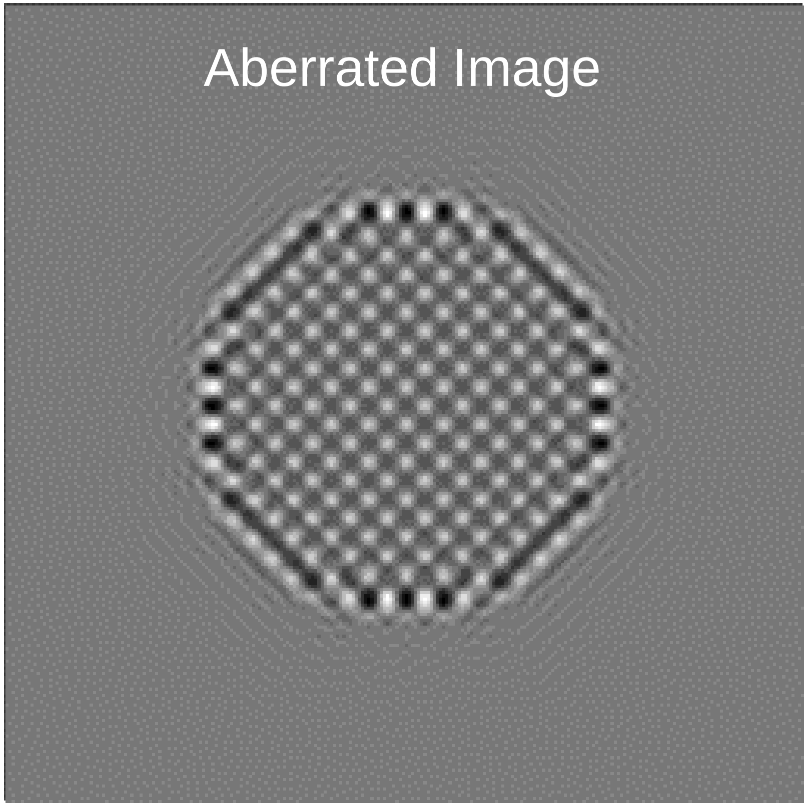
  write $A(u)=\Psi(u)T(u)$
- The optimal filter (L2) F(u) to apply is given by (Wiener, 1940)

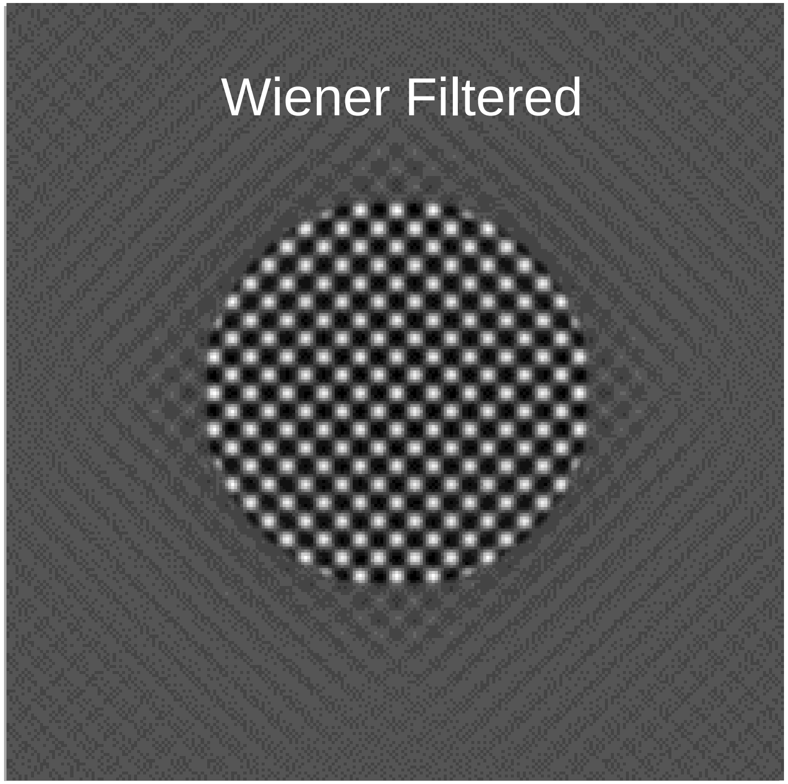
$$F(u) = T^*(u)/\{|T(u)|^2+n(u)^2/S(u)^2\}$$

n(u) = spectral distribution of noise

S(u) = estimate of signal

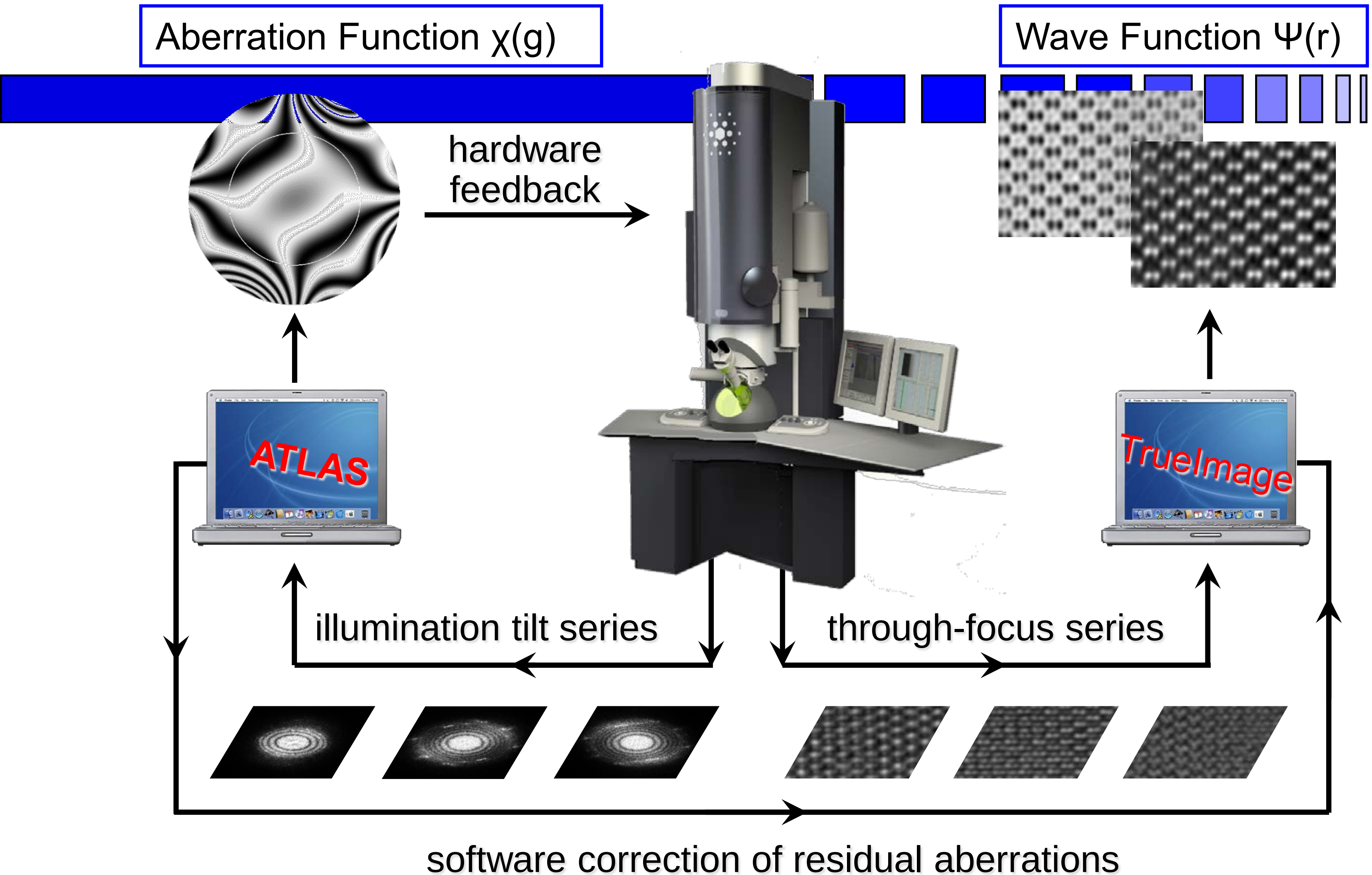
Wiener Filtering





Simple Example

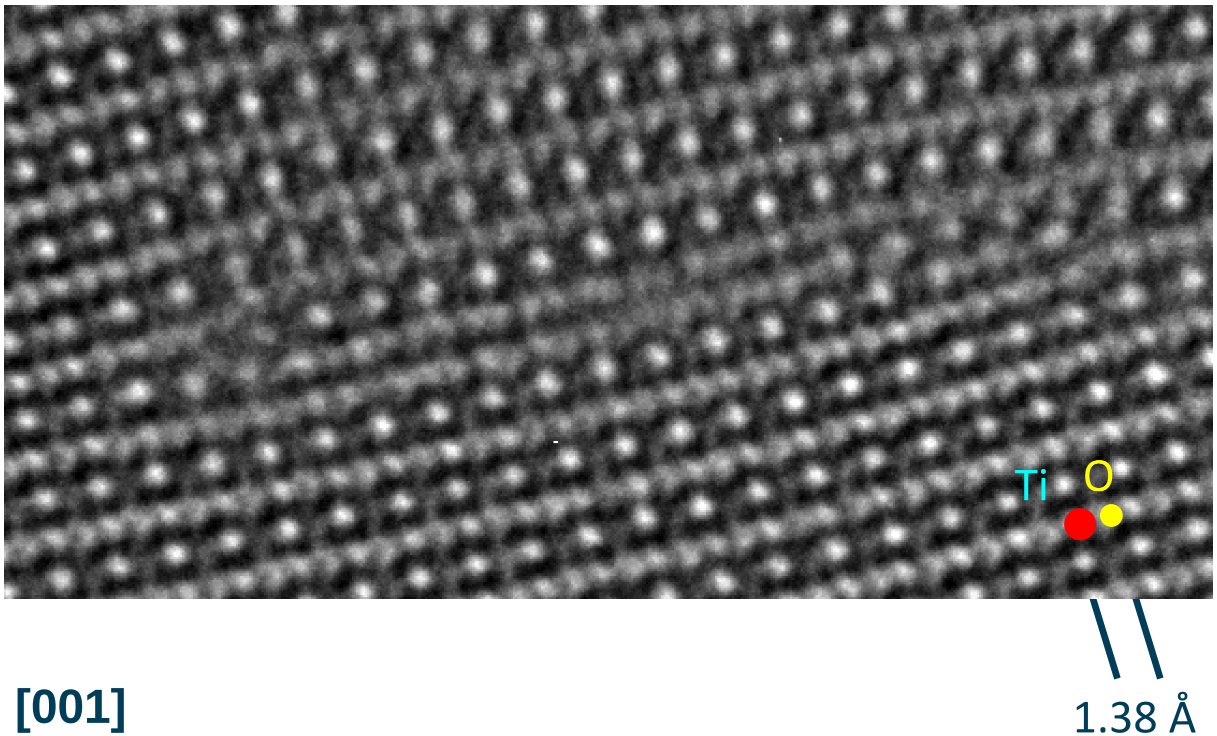
Aberration control & reconstruction of electron wave function



Curtesy Rafal Dunin-Borkowski

FZ Jülich

ATLAS & TrueImage:: Stacking Faults in $SrTiO_3$ (110)



Z_{opt} micrograph

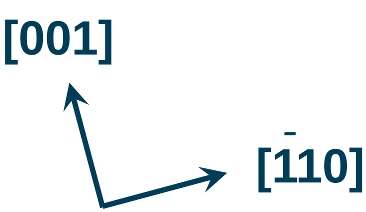
Titan 80-300

deficiencies:

shaded columns

inf. signal-to-noise ratio

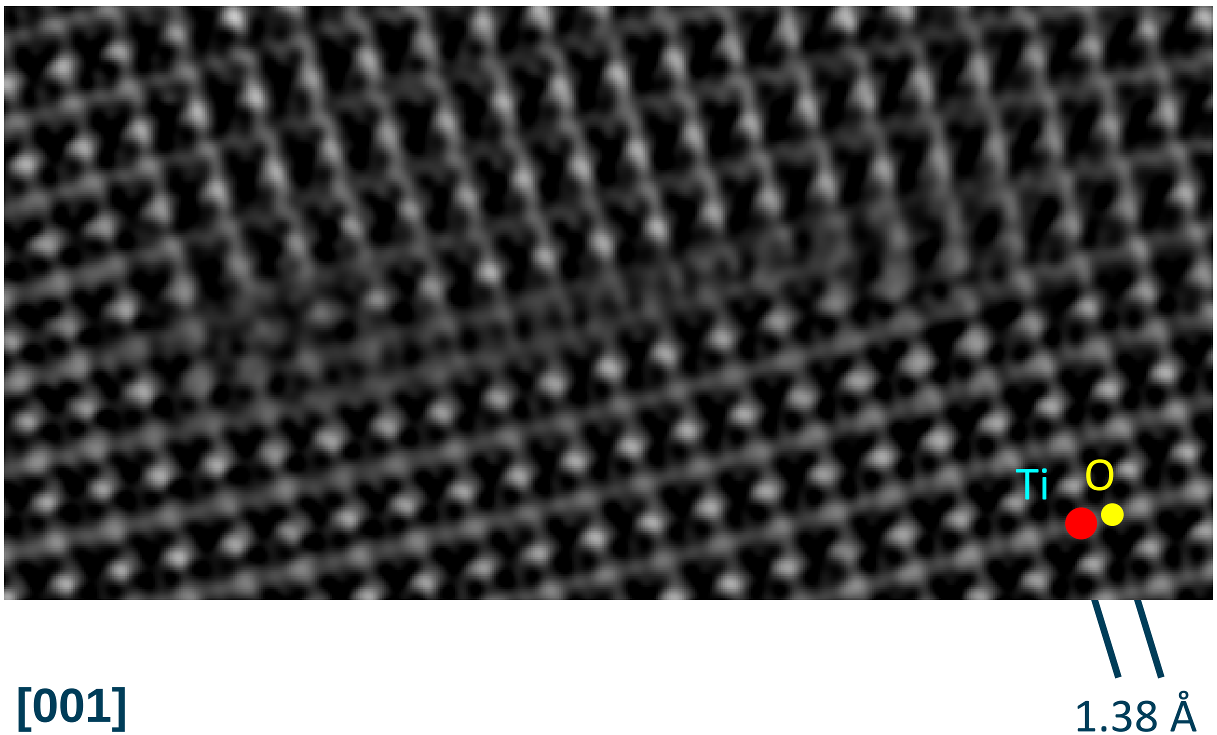
spurious contrast peaks



J. Barthel, PhD Thesis (2007)

Curtesy Rafal Dunin-Borkowski

ATLAS & TrueImage:: Stacking Faults in $SrTiO_3$ (110)

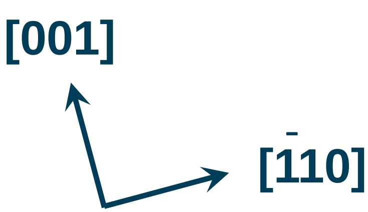


uncorrected phase image

Titan 80-300

deficiencies:

shaded columns



J. Barthel, PhD Thesis (2007)

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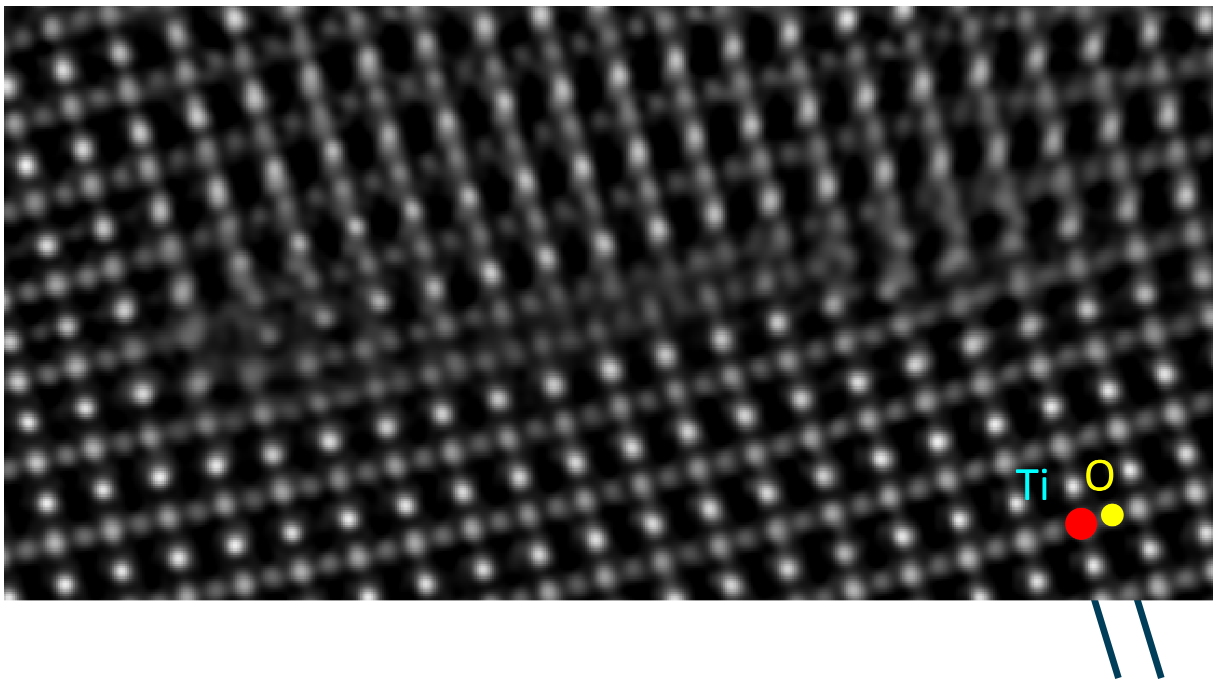
ATLAS & TrueImage:: Stacking Faults in $SrTiO_3$ (110)



1.38 Å

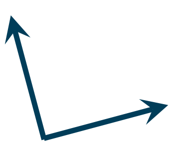
corrected phase image

Titan 80-300

deficiencies:

none

[001]



[$\bar{1}$10]

J. Barthel, PhD Thesis (2007)

Curtesy Rafal Dunin-Borkowski

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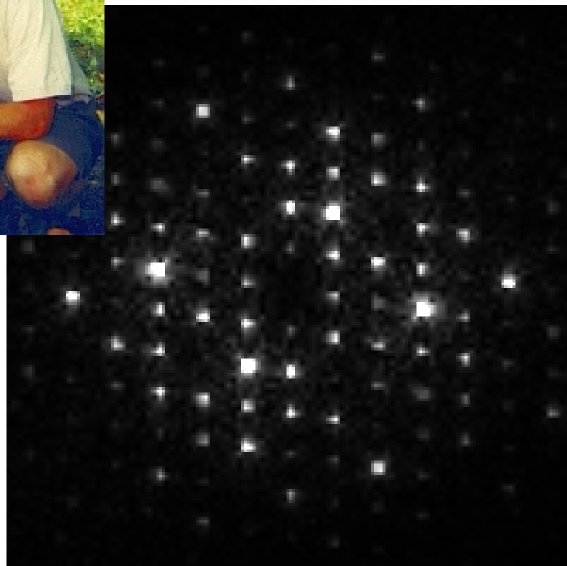
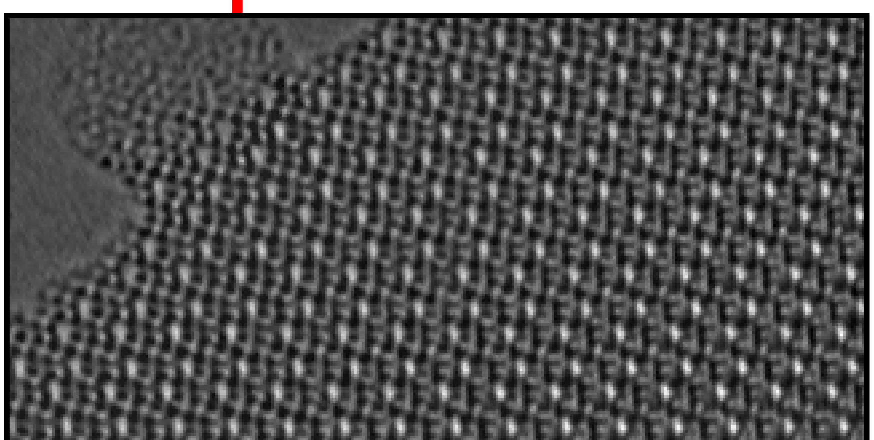
Method

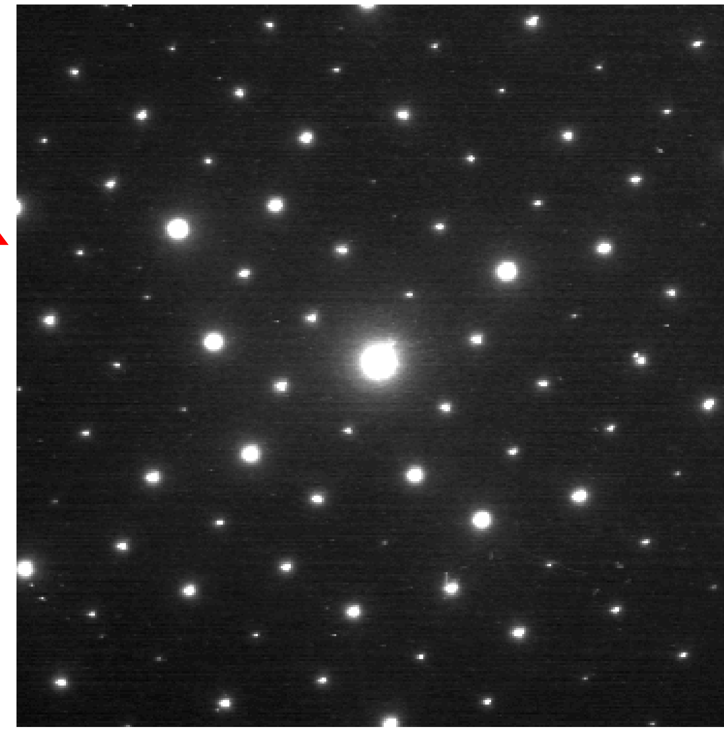


Initial Phases

FFT



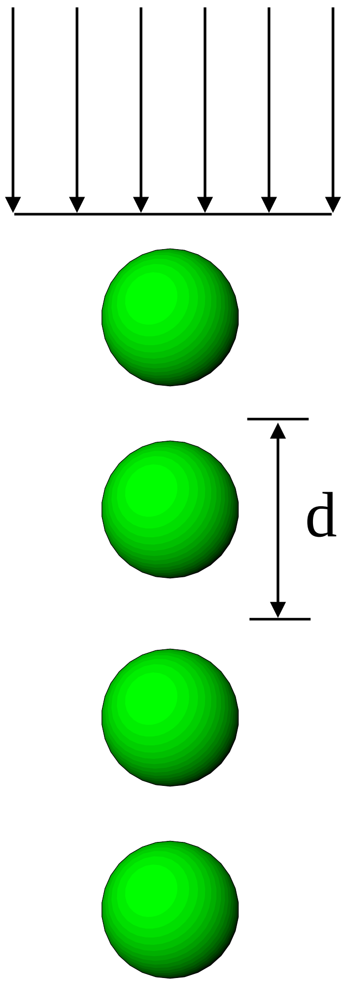
Conventional HREM Image



Nanoprobe Diffraction Data

W. Sinkler et al. *Acta Crystallogr. Sect. A* **54**, 591 (1998)

Channeling Approximation



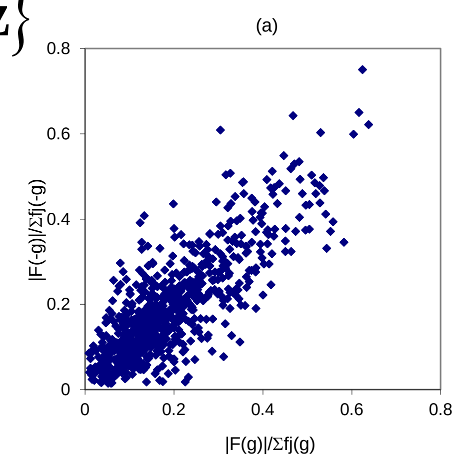
e^- $$\mathbf{V}(\mathbf{x},\mathbf{y}) = \frac{1}{\mathbf{d}}\int_{-\infty}^{\infty}\mathbf{V}_0(\mathbf{r})\mathbf{dz}$$

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

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

Talks later by Van Dyck and Chukhovskii will explain more details

Σ_0 distribution is statistically kinematical



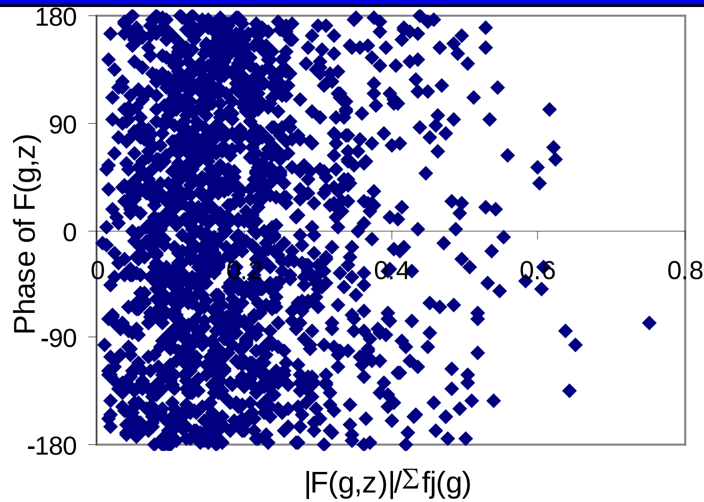
F. N. Chukhovskii, et al *Acta Cryst A* **57**, 231 (2001)

Statistics in a 1s model

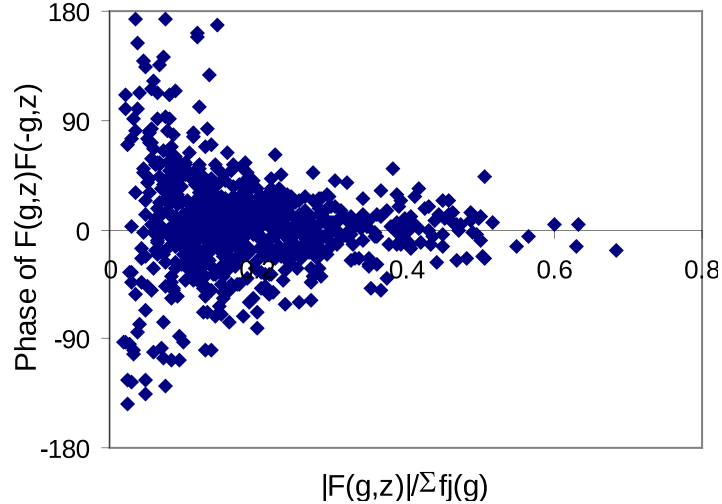
Kinematical	Dynamical
Σ_o:	Σ_o:
$\phi(g)+\phi(-g) = 0$	$\phi(g)+\phi(-g) \sim \omega$
$\lvert F(g)\rvert = \lvert F(-g)\rvert$	$\lvert F(g)\rvert \sim \lvert F(-g)\rvert$
Σ_2:	Σ_2:
$\phi(g)+\phi(h-g)+\phi(-h) \sim 2n\pi$	$\phi(g)+\phi(h-g)+\phi(-h) \sim \zeta$

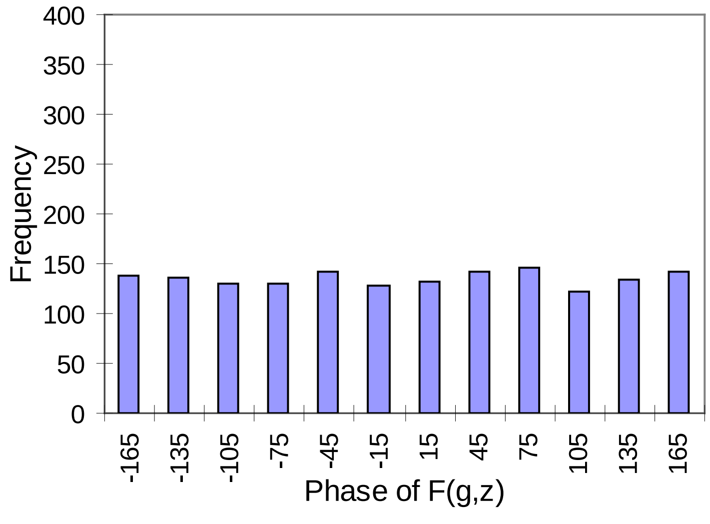
Kinematical Theory for electrons is "wrong", but statistically "right" in many cases, ***BUT NOT ALL*** (see C252)

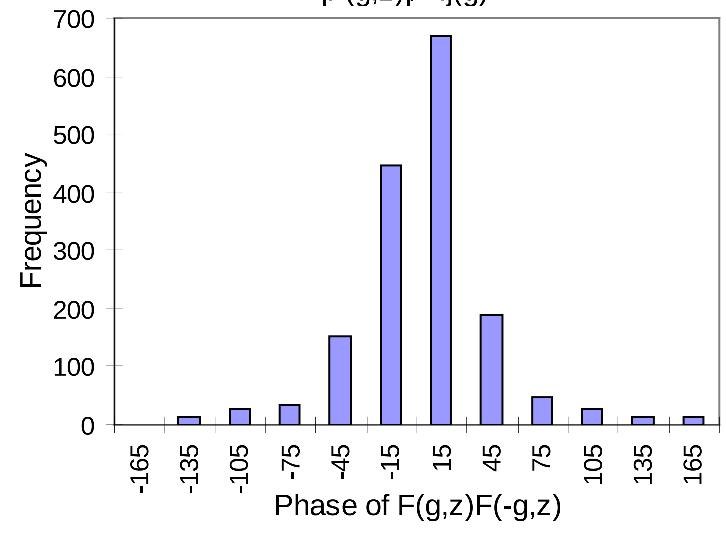
Σ_0 dynamical distribution



non-symm
5.264nm

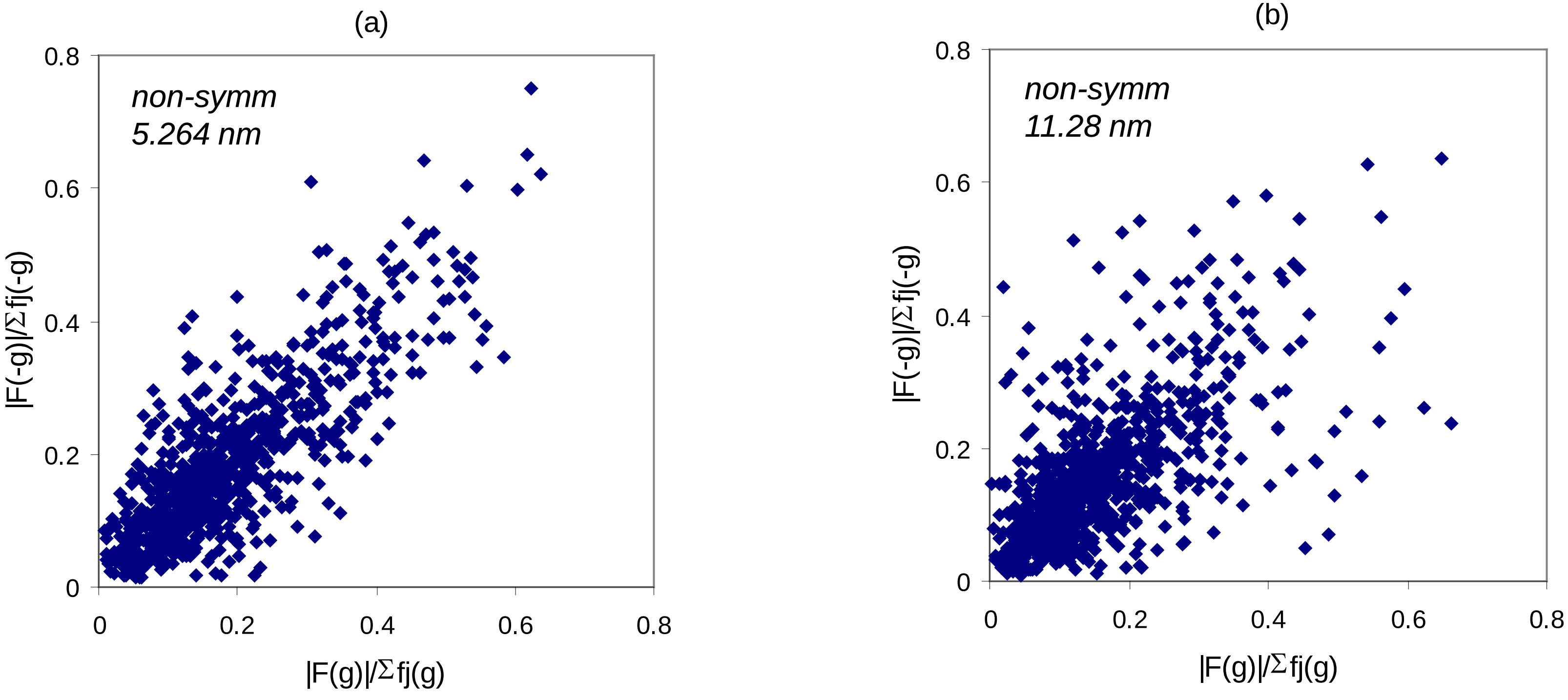




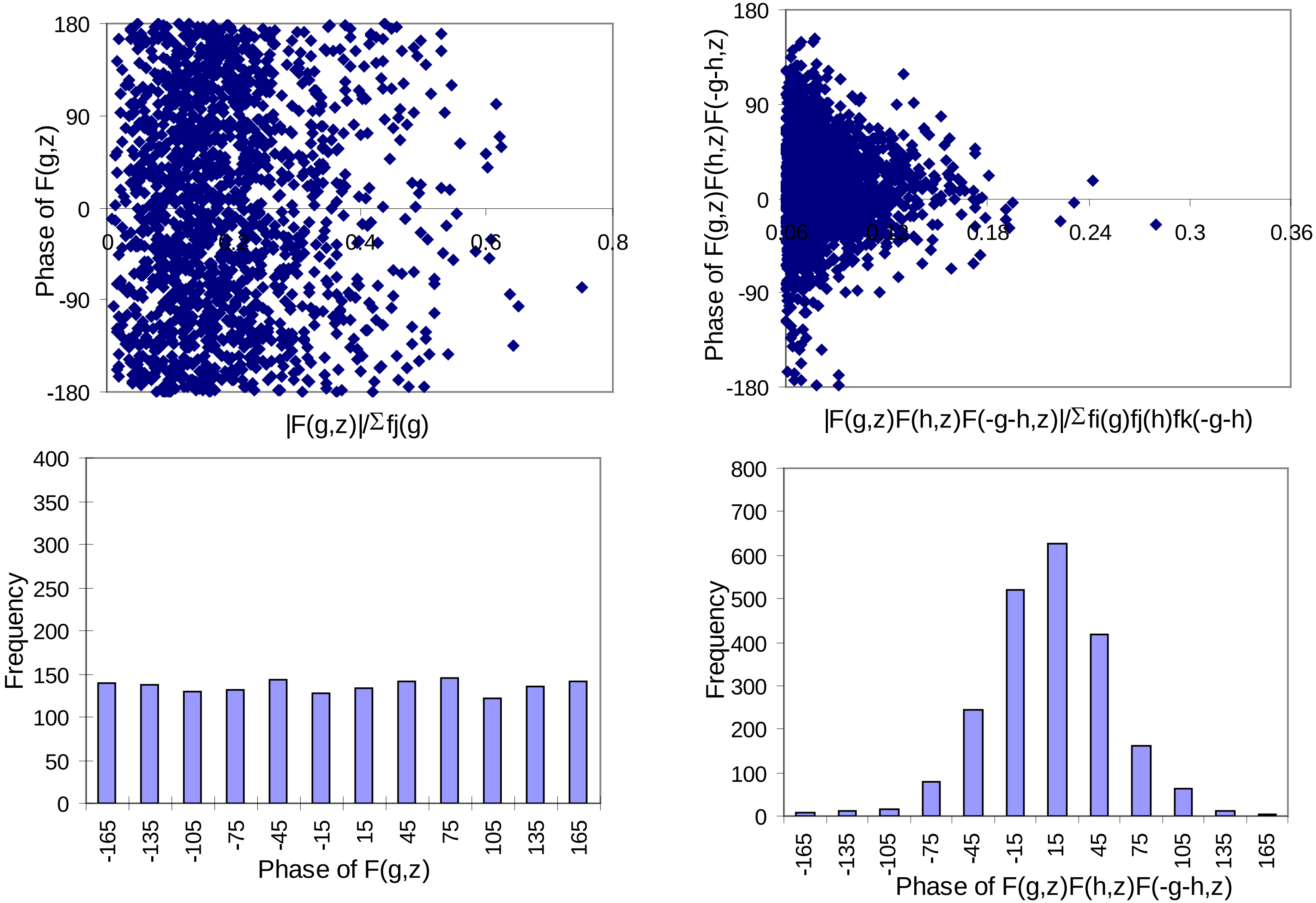


Σ_0 dynamical distribution

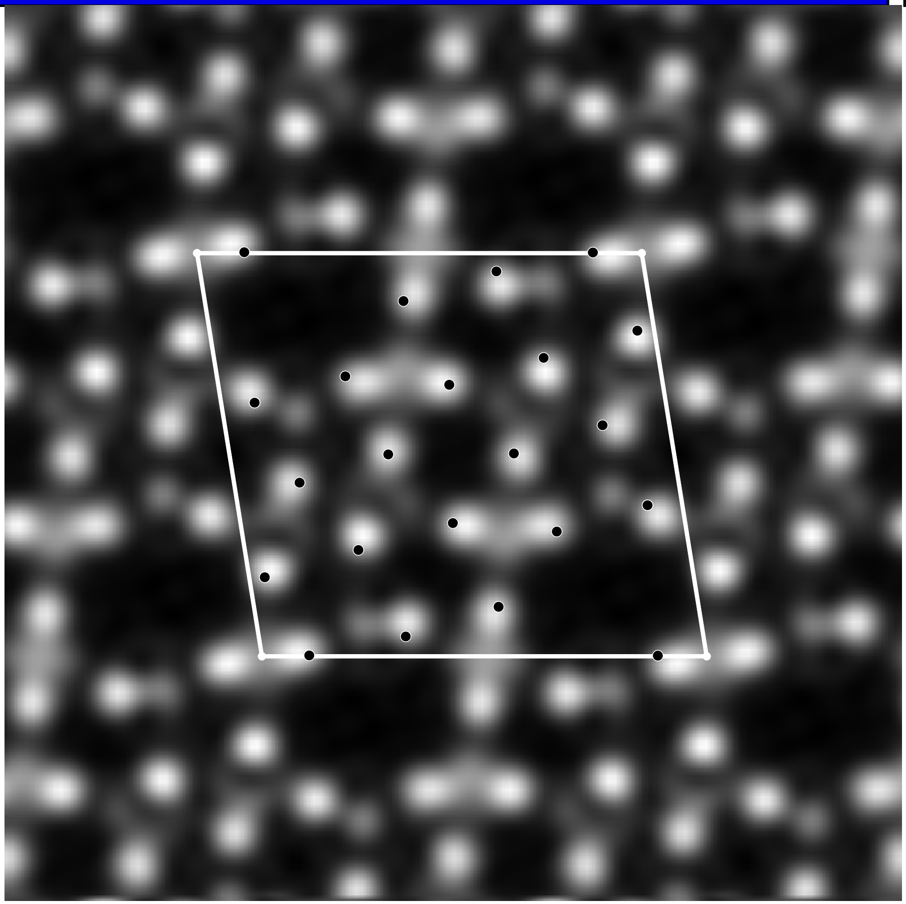
Freidel symmetry is statistical



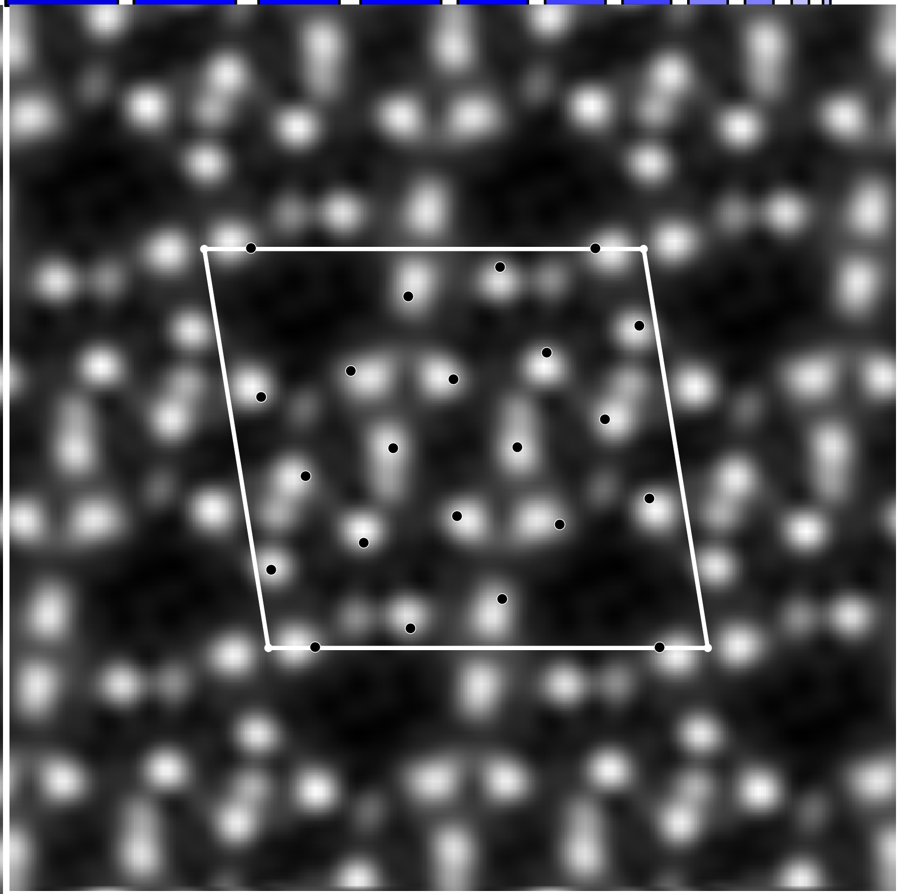
Σ_2 dynamical distribution



Calculated Wave

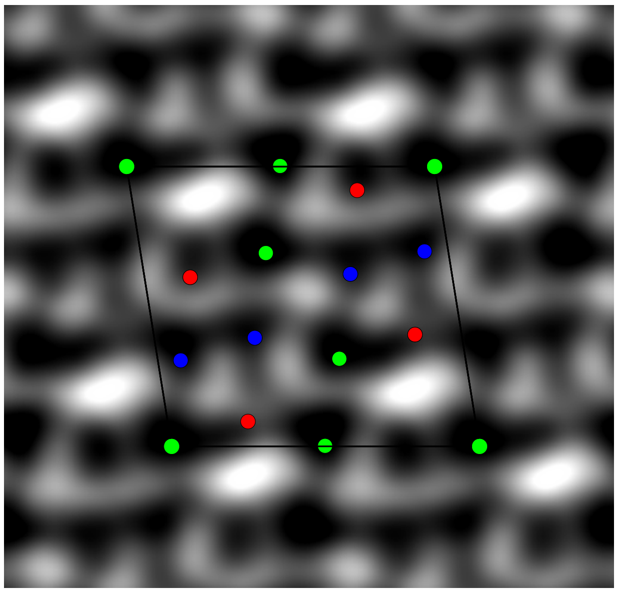


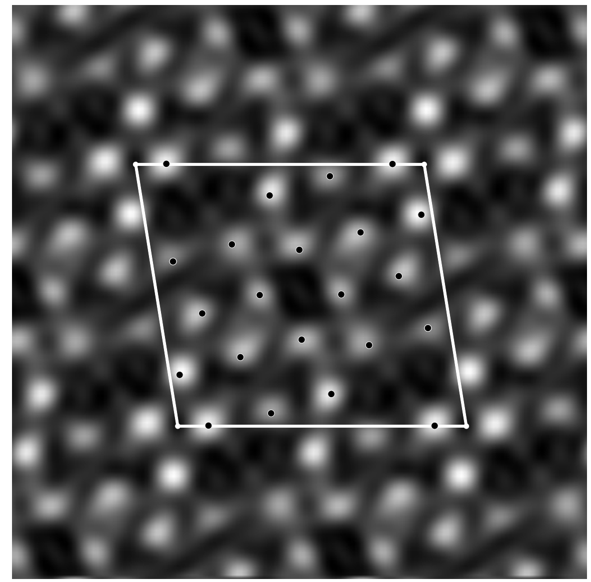
|ψ(r)-1| at 113 A thickness

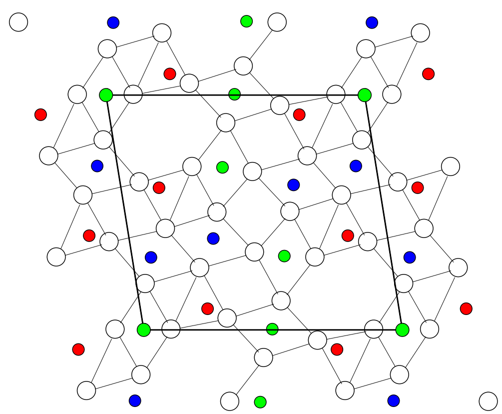


|ψ(r)-1| at 202 A thickness

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







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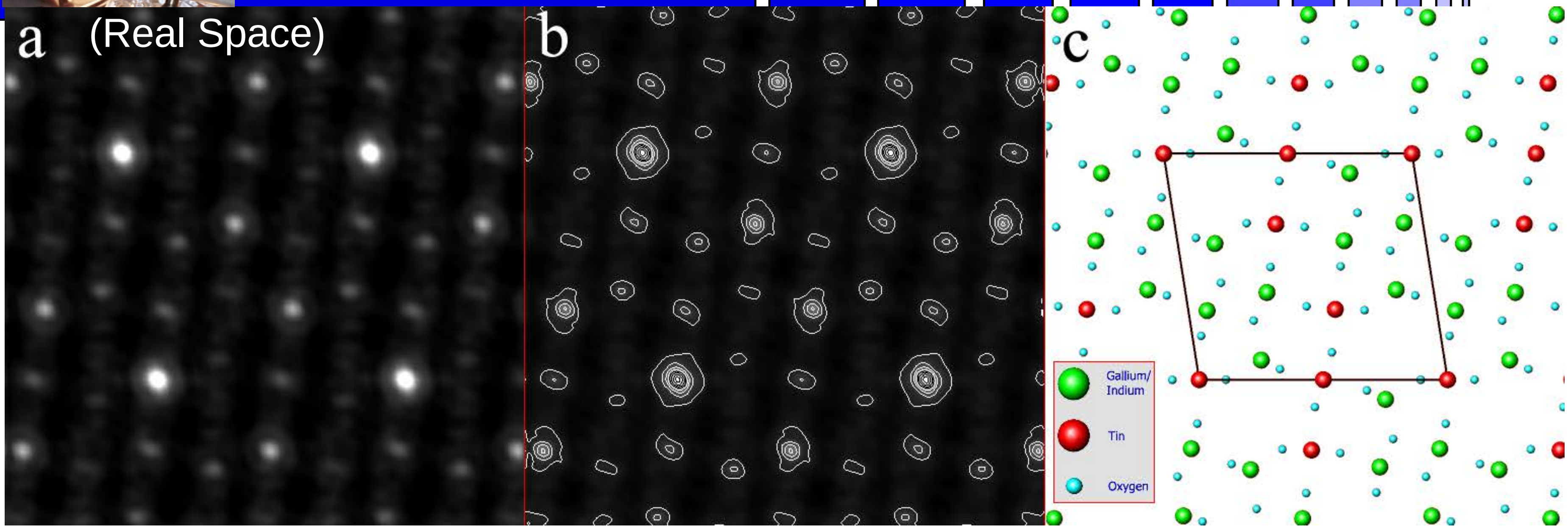
L. D. Marks, W. Sinkler, Sufficient conditions for direct methods with swift electrons. *Microsc. Microanal.* **9**, 399 (2003).

Precession Electron Diffraction

- Quasi-Kinematical Data
- Averaging over angle/phase (and thickness) damps dynamical contributions
- Intensities are close to monatomic with structure factors (statistically)

$(Ga,In)_2SnO_4$ precession data: Direct methods solution



	ΔR (Å)
Sn1	0.00E+00
Sn2	0.00E+00
Sn3	6.55E-03
In/Ga1	5.17E-02
In/Ga2	2.37E-03
Ga1	6.85E-02
Ga2	1.22E-01

Displacement $(R_{neutron} - R_{precession})$:

$\Delta R_{mean} <$ **$4*10^{-2}$ Å**

(Sinkler, et al. J. Solid State Chem, 1998).

(Own, Sinkler, & Marks, submitted.)

Conclusion

- The “Phase Problem” with electrons is no longer really a problem….assuming ideal data of course
- Many technique work most of the time
- Few techniques work all the time
- Some unresolved issues (proper dynamical refinement)
- Rember that we are solving an inversion problem, and these are susceptible to ill-conditioning

Four basic elements are required to solve a recovery problem

1. A data formation model

    Imaging/Diffraction/Measurement

2. A priori information

    The presence of atoms or similar

3. A recovery criterion:

    A numerical test of Goodness-of-Fit

4. A solution method.

    Mathematical details

Patrick Combettes, (1996). Adv. Imag. Elec. Phys. 95, 155