Electron ptychography: bringing imaging and diffraction into phase

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Outline

- 4D STEM
- The phase problem
- A wave-optical model of 4D STEM
  - The STEM imaging master equation
  - BF imaging (coherent) and ADF (incoherent imaging)
- Ptychographical retrieval of the phase image
  - Single side-band method – correction of aberrations – optical sectioning
  - Wigner distribution deconvolution – super-resolution
  - Iterative methods (ePIE)
- Quiz / Coffee
- Effect of partial coherence
- Dynamic range
- Sampling in real and reciprocal space
- Transfer functions, signal to noise and detective quantum efficiency

- Please do stop me to ask questions during the lecture

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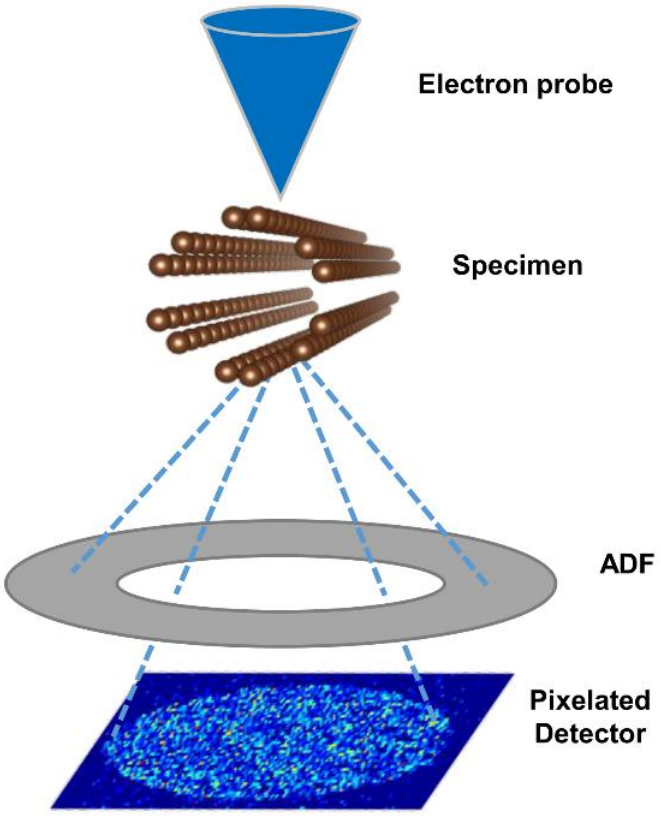
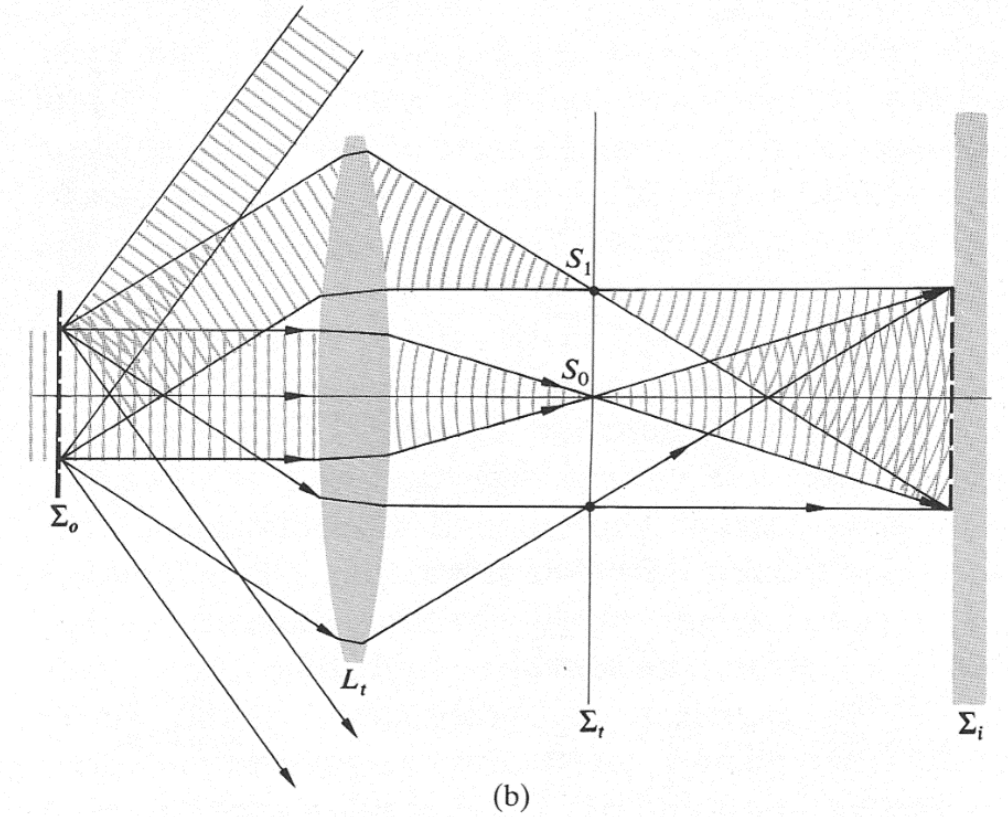
4D STEM



- The probe scans in 2D real space.
- The detector plane is a far-field diffraction plane and is a 2D reciprocal space
- Recording diffraction patterns as a function of probe position gives a 4D data set
  - Two dimensions in real space and two dimensions in reciprocal space.
- For a focused probe the structure in the detector plane can be quite slowly varying
  - Segmented detectors such as quadrants or octants can be effective.

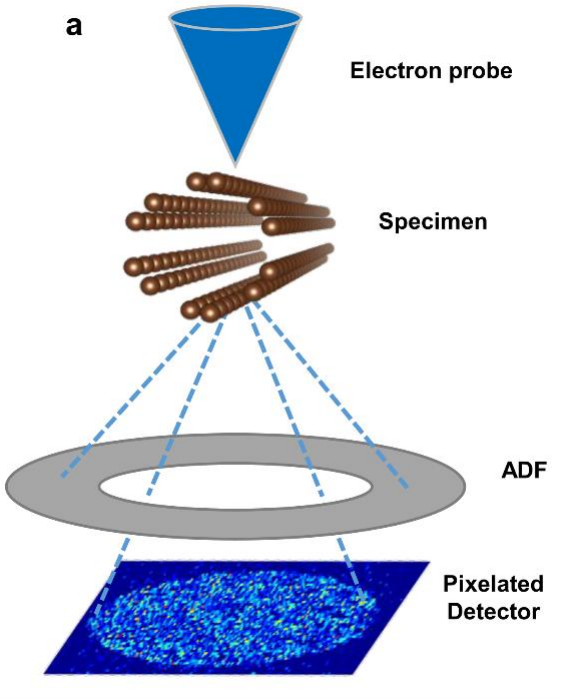
Abbe theory of image formation

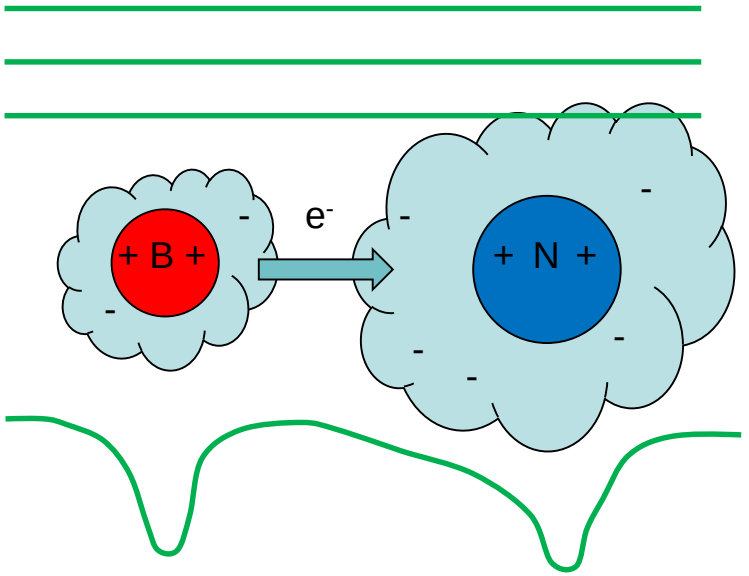
Ernst Abbe
(1840-1905)



If we solve the phase problem, then the distinction between imaging and diffraction vanishes.

Electric field detection or phase shift

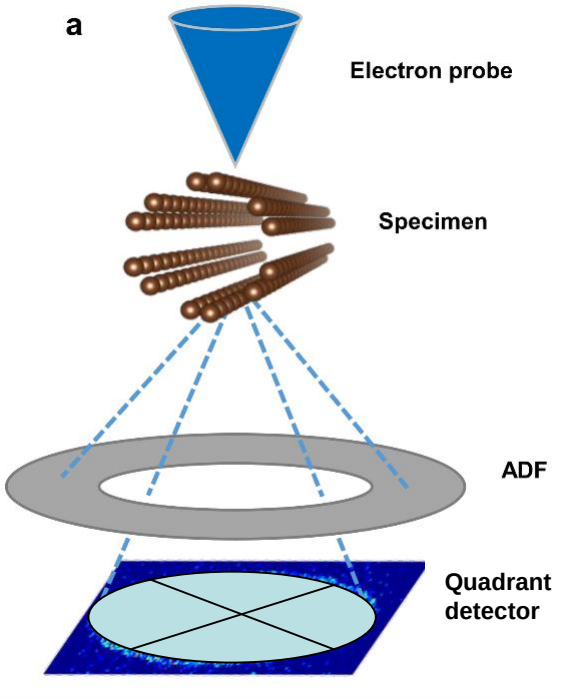


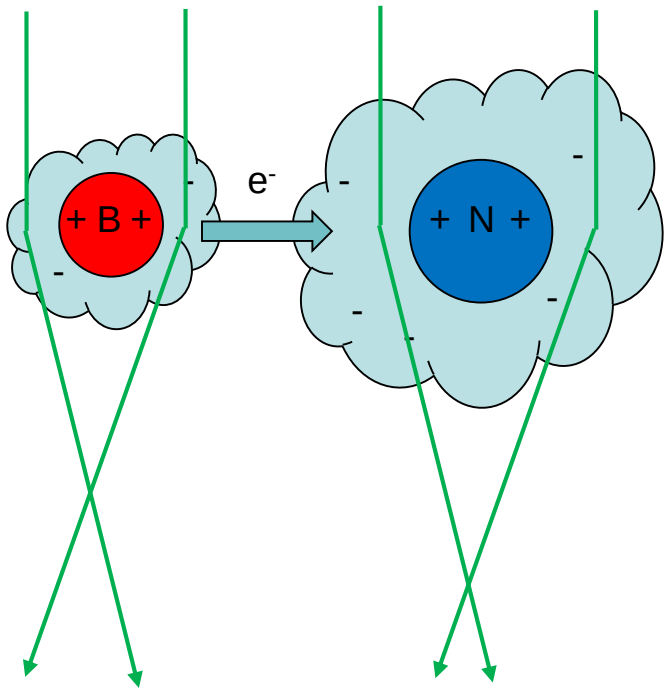


phase imaging

- In the phase object approximation, the effect of the electron scattering is to shift the phase of the transmitted electron wave by an amount that is proportional to the projected potential.

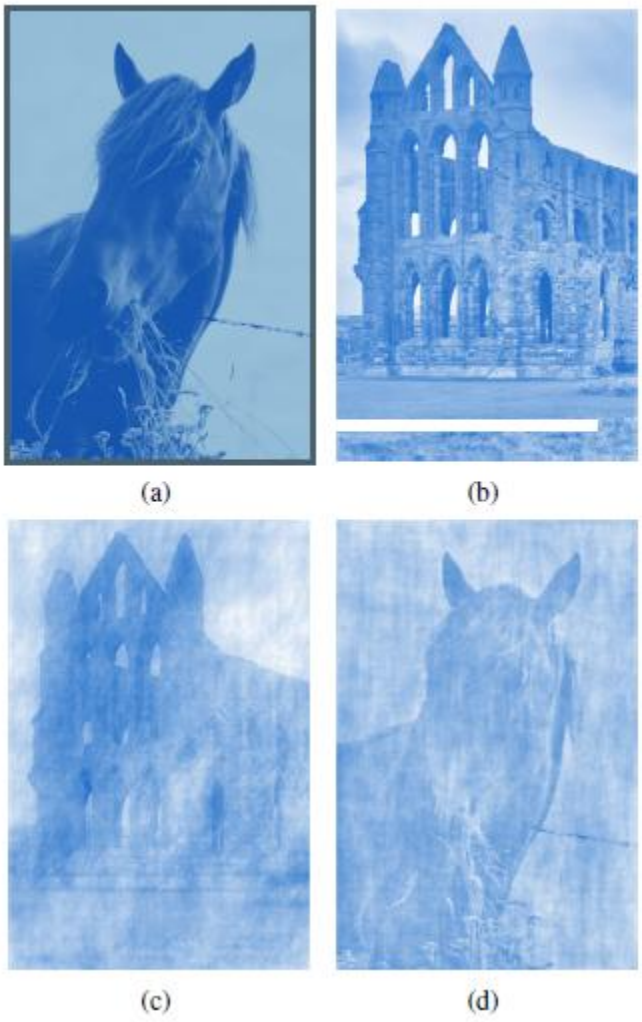
Electric field detection or phase shift





- First moment (or centre of mass) can be regarded as a measurement of field strength.
- Differential phase contrast can also be regarded as a measurement of field.
- Phase is proportion to projected potential, and field is the differential of potential, so ptychography, centre of mass, DPC and iDPC are very closely related and are measuring the same quantity.

Phase carries position information



1. The Fourier transform is taken of both (a) and (b)
2. The phase information in the Fourier transform is swapped to join the “wrong” modulus.
3. (c) has modulus of FT of (a), but phase of FT of (b)
4. (d) has modulus of FT of (b) but phase of FT of (a)

5. The phase is important for controlling positional information: “geometric phase”

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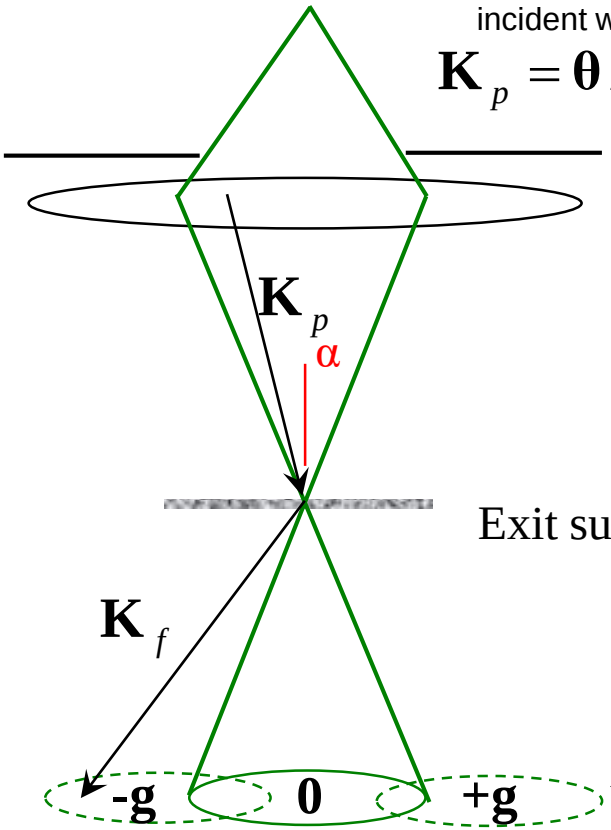
What do we see in the detector plane?



transverse component of incident wavevector

$$\mathbf{K}_p = \boldsymbol{\theta} / \lambda$$

objective aperture

aberration function

$$A(\mathbf{K}_p) = H(\mathbf{K}_p)\exp\left[-i\chi(\mathbf{K}_p)\right]$$

F.T.

$$\chi = \left(\frac{2\pi}{\lambda}\right)\left[\frac{C_{1,0}\lambda^2 K_i^2}{2} + \frac{C_{3,0}\lambda^4 K_i^4}{4}\right]$$

Exit surface

$$\psi_p(\mathbf{R})$$

probe position

$$\psi(\mathbf{R}) = \psi_p(\mathbf{R} - \mathbf{R}_p)\phi(\mathbf{R})$$

transmission function

F.T.

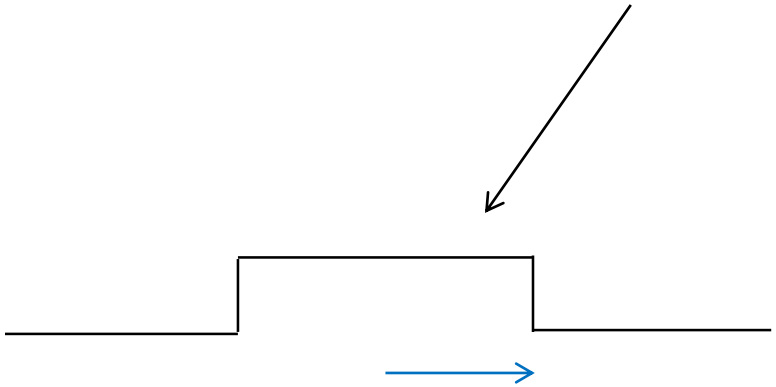
$$\Psi_f(\mathbf{K}_f, \mathbf{R}_p) = A(\mathbf{K}_f)\exp\left[i2\pi\mathbf{K}_f \cdot \mathbf{R}_p\right] \otimes \Phi(\mathbf{K}_f)$$

- For a crystalline sample, you see diffracted discs.
- If the discs overlap, you will see interference fringes.
- The position of the fringes depends on the aberrations, the probe position *and the relative phases of the diffracted beams*.

Hoppe, W. *Acta Cryst A* **25** (1969) 495 and 508.

Aperture function and point-spread function

$$A(\mathbf{K}) = H(\mathbf{K})\exp[-i\chi(\mathbf{K})]$$



$$\chi(\theta) = \left(\frac{2\pi}{\lambda}\right)\left[\frac{C_{1,0}\theta^2}{2} + \frac{C_{3,0}\theta^4}{4}\right]$$

$$\theta = |\mathbf{K}|\lambda$$

$$\chi(\mathbf{K}) = \left(\frac{2\pi}{\lambda}\right)\sum_{n,m}\left[C_{n.m,a}\theta^n \cos(m\phi) + C_{n.m,b}\theta^n \sin(m\phi)\right]$$

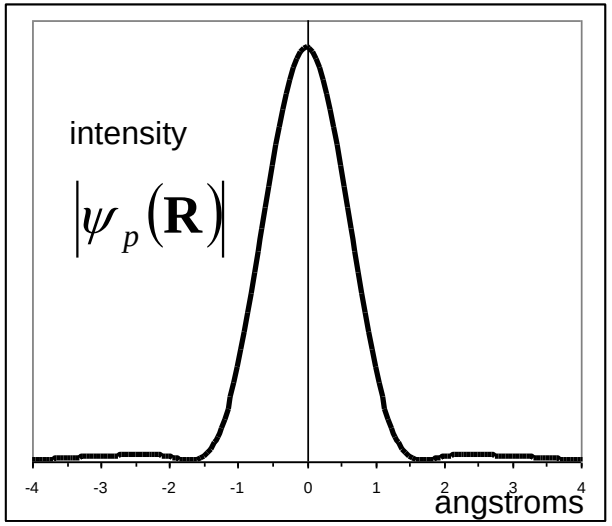
α = ~ 9 mrad (uncorrected)
20-30 mrad (corrected)

Krivanek aberration notation:

Ultramicroscopy, 78 (1999) 1-11

STEM diffraction limited probe intensity

$\psi_p(\mathbf{R})$ is the inverse Fourier transform of the aperture function. In the absence of aberrations, it become the inverse Fourier transform of a disc function, the intensity of which is the Airy disc.

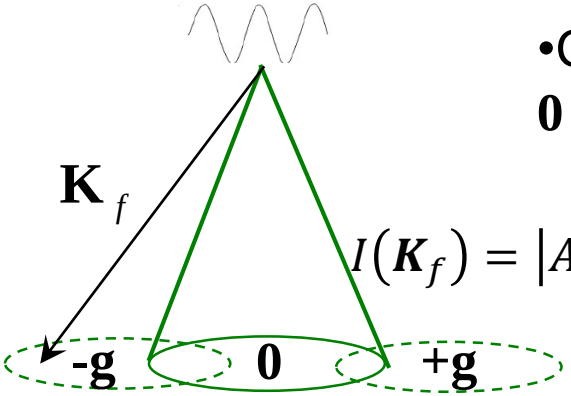


(V=300 kV, C_S=1 mm, z=-40 nm)

The FWHM of the intensity of the Airy disc is given approximately by

$$d_{\text{diff}} = \frac{0.61\lambda}{\alpha}$$

Consider sinusoidal phase grating



•Consider intensity in overlap region between **0** and +**g** discs.

$$I(\boldsymbol{K}_f) = \left|A(\boldsymbol{K}_f)\exp\left[i2\pi\boldsymbol{K}_f.\boldsymbol{R}_p\right] + F_{\boldsymbol{g}}A(\boldsymbol{K}_f-\boldsymbol{g})\exp\left[i2\pi(\boldsymbol{K}_f-\boldsymbol{g}).\boldsymbol{R}_p\right]\right|^2$$

•Expand square modulus.

$$I(\boldsymbol{K}_f) = \left|A(\boldsymbol{K}_f)\right|^2 + \left|F_{\boldsymbol{g}}\right|^2\left|A(\boldsymbol{K}_f-\boldsymbol{g})\right|^2 + A(\boldsymbol{K}_f)$$
$$F_{\boldsymbol{g}}^*A^*(\boldsymbol{K}_f-\boldsymbol{g})\exp\left[i2\pi\boldsymbol{g}.\boldsymbol{R}_p\right] + F_{\boldsymbol{g}}A^*(\boldsymbol{K}_f)\,A(\boldsymbol{K}_f-\boldsymbol{g})$$
$$\exp\left[-i2\pi\boldsymbol{g}.\boldsymbol{R}_p\right]$$

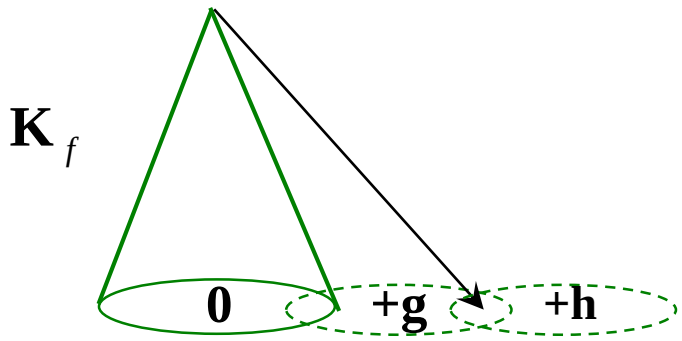
•Assume no lens aberrations.

$$I(\boldsymbol{K}_f) = \left|A(\boldsymbol{K}_f)\right|^2 + \left|A(\boldsymbol{K}_f-\boldsymbol{g})\right|^2 +$$
$$2\left|F_{\boldsymbol{g}}\right|A(\boldsymbol{K}_f)A(\boldsymbol{K}_f-\boldsymbol{g})\cos\left[i2\pi\boldsymbol{g}.\boldsymbol{R}_p - \angle F_{\boldsymbol{g}}\right]$$

Structure factor phase

The intensity in the overlap region oscillates as the probe scans with the periodicity of the sinusoidal grading (ie with a spatial frequency of ***g***).

Interference between diffracted beams



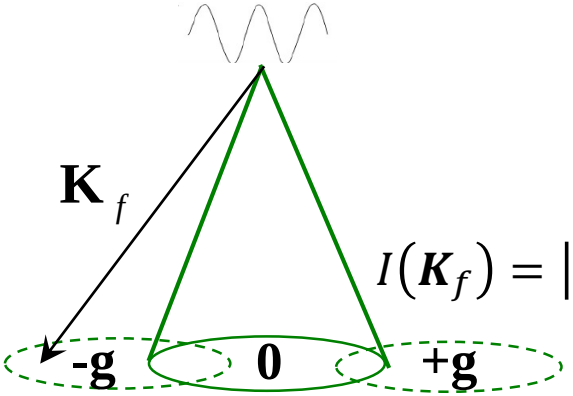
$$I(\boldsymbol{K}_f) = \left|F_{\boldsymbol{g}} A(\boldsymbol{K}_f - \boldsymbol{g}) \exp\left[i2\pi(\boldsymbol{K}_f - \boldsymbol{g}).\boldsymbol{R}_p\right] + F_{\boldsymbol{h}} A(\boldsymbol{K}_f - \boldsymbol{h}) \exp\left[i2\pi(\boldsymbol{K}_f - \boldsymbol{h}).\boldsymbol{R}_p\right]\right|^2$$

- Assume no lens aberrations.

$$I(\boldsymbol{K}_f) = \left|F_{\boldsymbol{g}} A(\boldsymbol{K}_f - \boldsymbol{g})\right|^2 + \left|F_{\boldsymbol{h}} A(\boldsymbol{K}_f - \boldsymbol{h})\right|^2 + 2\left|F_{\boldsymbol{g}}\right|\left|F_{\boldsymbol{h}}\right| A(\boldsymbol{K}_f - \boldsymbol{g}) A(\boldsymbol{K}_f - \boldsymbol{h}) \cos\left[i2\pi(\boldsymbol{g} - \boldsymbol{h}).\boldsymbol{R}_p - \angle F_{\boldsymbol{g}} + \angle F_{\boldsymbol{h}}\right]$$

The intensity in the overlap region oscillates as the probe scans with the periodicity of the difference of the scattering angle (ie with a spatial frequency of ***g-h***). **Note does not depend on absolute scattering angle**.

With lens aberrations



•Consider intensity in overlap region between **0** and +**g** discs.

$$I(\boldsymbol{K}_f) = \left|A(\boldsymbol{K}_f)\exp\left[i2\pi\boldsymbol{K}_f.\boldsymbol{R}_p\right] + F_{\boldsymbol{g}}A(\boldsymbol{K}_f-\boldsymbol{g})\exp\left[i2\pi(\boldsymbol{K}_f-\boldsymbol{g}).\boldsymbol{R}_p\right]\right|^2$$

$$I(\boldsymbol{K}_f) = \left|A(\boldsymbol{K}_f)\right|^2 + \left|F_{\boldsymbol{g}}\right|^2\left|A(\boldsymbol{K}_f-\boldsymbol{g})\right|^2 + F_{\boldsymbol{g}}^*A(\boldsymbol{K}_f)\,A^*(\boldsymbol{K}_f-\boldsymbol{g})\exp\left[i2\pi\boldsymbol{g}.\boldsymbol{R}_p\right] + F_{\boldsymbol{g}}\,A^*(\boldsymbol{K}_f)\,A(\boldsymbol{K}_f-\boldsymbol{g})\exp\left[-i2\pi\boldsymbol{g}.\boldsymbol{R}_p\right]$$

•Add some defocus. Set $\boldsymbol{R}_p$=0 for simplicity. Look at interference term: $F_{\boldsymbol{g}}^*A(\boldsymbol{K}_f)\,A^*(\boldsymbol{K}_f-\boldsymbol{g}) + F_{\boldsymbol{g}}A^*(\boldsymbol{K}_f)\,A(\boldsymbol{K}_f-\boldsymbol{g})$

$$= \left|F_{\boldsymbol{g}}\right|H(\boldsymbol{K}_f)\,H(\boldsymbol{K}_f-\boldsymbol{g})\{\exp i\left[\chi(\boldsymbol{K}_f)-\chi(\boldsymbol{K}_f-\boldsymbol{g})-\angle F_{\boldsymbol{g}}\right] + \exp i\left[-\chi(\boldsymbol{K}_f)+\chi(\boldsymbol{K}_f-\boldsymbol{g})+\angle F_{\boldsymbol{g}}\right]\}$$

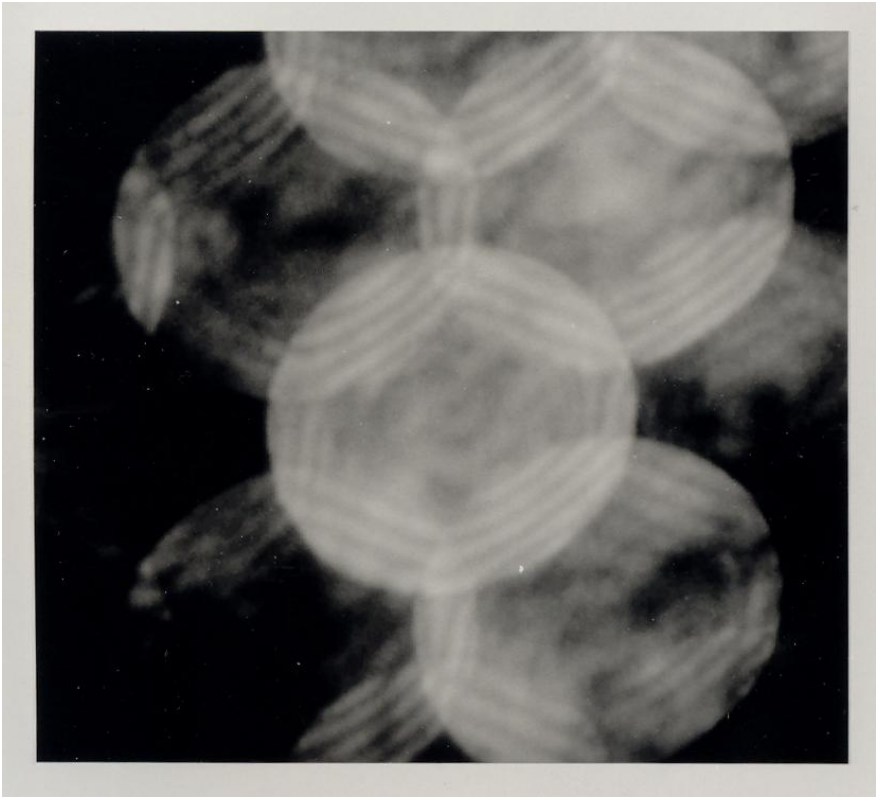
$$= 2\left|F_{\boldsymbol{g}}\right|H(\boldsymbol{K}_f)\,H(\boldsymbol{K}_f-\boldsymbol{g})\cos\left[\chi(\boldsymbol{K}_f)-\chi(\boldsymbol{K}_f-\boldsymbol{g})-\angle F_{\boldsymbol{g}}\right]$$

$$= 2\left|F_{\boldsymbol{g}}\right|H(\boldsymbol{K}_f)\,H(\boldsymbol{K}_f-\boldsymbol{g})\cos\{\pi C_{1,0}\lambda\left[2\boldsymbol{K}_f.\boldsymbol{g}-\boldsymbol{g}^2-\angle F_{\boldsymbol{g}}\right]\}$$

$$\chi(\boldsymbol{K}_f) = \pi C_{1,0}\lambda K_f^2$$

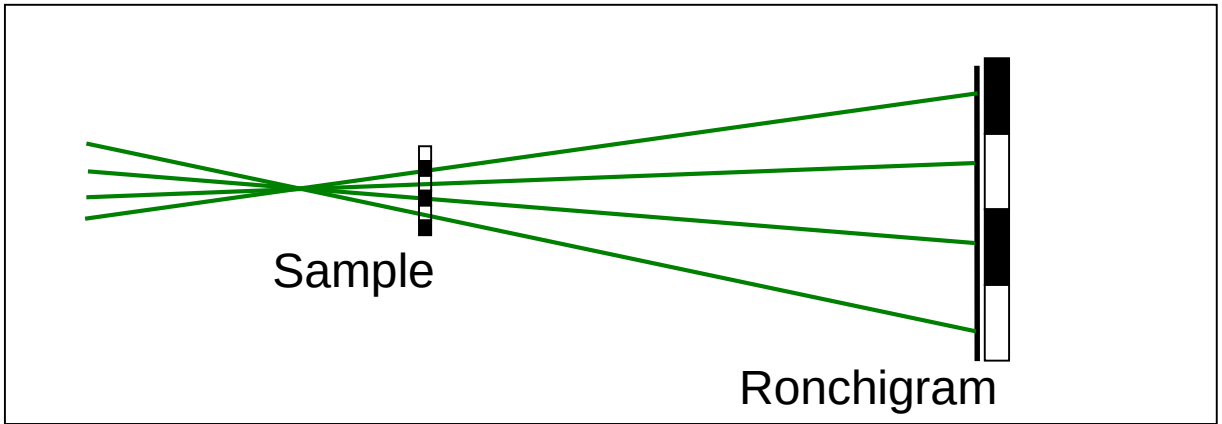
A uniform set of fringes that can be thought of as a shadow of the lattice.

Coherent CBED



With a large aperture you see what is sometimes referred to as a shadow image or Ronchigram.

Test your understanding: Show that the spacing of the fringes is exactly that you would expect for a shadow of the corresponding lattice spacing illuminated by a point source at a distance given by the defocus.



$$M = \frac{L}{C_{1,0}}$$

Go into 4D reciprocal space

Rodenburg and Bates, Phil. Trans. R. Soc London A, **339** (1992) 521.

We measure the intensity in the detector plane to form the 4D STEM data set.

$$\left|M\left(\boldsymbol{K}_f, \boldsymbol{R}_p\right)\right|^2 = \left|A\left(\boldsymbol{K}_f\right)\exp\left[i2\pi\boldsymbol{K}_f.\boldsymbol{R}_p\right]\otimes\Phi\left(\boldsymbol{K}_f\right)\right|^2$$

Expanding the modulus-squared and the two resulting convolutions gives:

$$\left|M\left(\boldsymbol{K}_f, \boldsymbol{R}_p\right)\right|^2 = \int A\left(\boldsymbol{K}'_f\right)\exp\left[i2\pi\boldsymbol{K}'_f.\boldsymbol{R}_p\right]\Phi\left(\boldsymbol{K}_f-\boldsymbol{K}'_f\right)d\boldsymbol{K}'_f$$

$$\times\int A^*\left(\boldsymbol{K}''_f\right)\exp\left[-i2\pi\boldsymbol{K}''_f.\boldsymbol{R}_p\right]\Phi^*\left(\boldsymbol{K}_f-\boldsymbol{K}''_f\right)d\boldsymbol{K}''_f$$

The magic part is now to take the Fourier transform with respect to $\boldsymbol{R}_p$. Performing the Fourier transform and only including the factors above that include $\boldsymbol{R}_p$ gives

FT kernel

$$\int\exp\left[i2\pi\boldsymbol{R}_p.\left(\boldsymbol{K}'_f-\boldsymbol{K}''_f+\boldsymbol{Q}_p\right)\right]d\boldsymbol{R}_p=\delta\left(\boldsymbol{K}'_f-\boldsymbol{K}''_f+\boldsymbol{Q}_p\right)$$

Substituting in the delta function into the $\boldsymbol{K}''_f$ integral only allows values when

$$\boldsymbol{K}''_f=\boldsymbol{K}'_f+\boldsymbol{Q}_p$$

The *G* data set

$$G\left(\boldsymbol{K}_f, \boldsymbol{Q}_p\right) = \int A\left(\boldsymbol{K}'_f\right)\Phi\left(\boldsymbol{K}_f - \boldsymbol{K}'_f\right)A^*\left(\boldsymbol{K}'_f + \boldsymbol{Q}_p\right)\Phi^*\left(\boldsymbol{K}_f - \boldsymbol{K}'_f - \boldsymbol{Q}_p\right)d\boldsymbol{K}'_f$$

Which can be written using convolution notation:

$$G\left(\boldsymbol{K}_f, \boldsymbol{Q}_p\right) = A\left(\boldsymbol{K}_f\right)A^*\left(\boldsymbol{K}_f + \boldsymbol{Q}_p\right)\otimes_{\boldsymbol{K}_f}\Phi\left(\boldsymbol{K}_f\right)\Phi^*\left(\boldsymbol{K}_f - \boldsymbol{Q}_p\right)$$

So far we are not defining any one method (eg ptychography, DPC, COM etc), this is simply a statement of the physics of 4D STEM.

How should we interpret $G\left(\boldsymbol{K}_f, \boldsymbol{Q}_p\right)$?

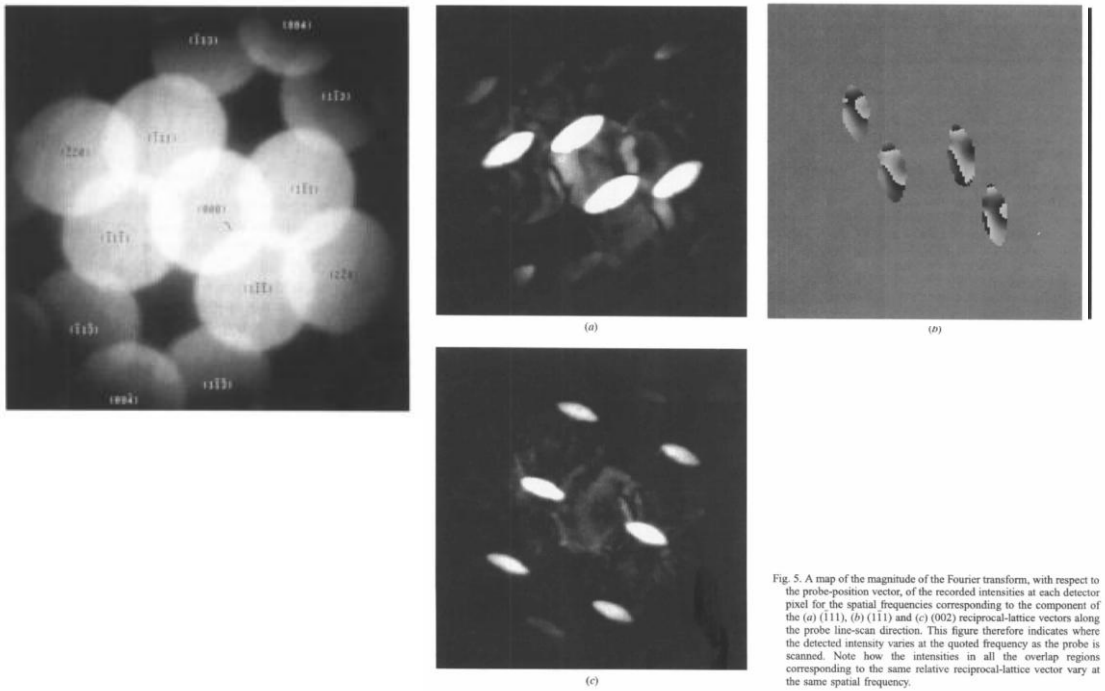


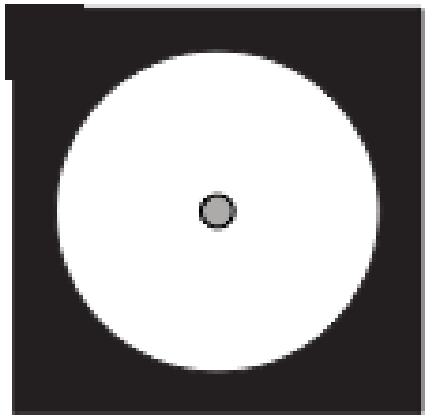
Fig. 5. A map of the magnitude of the Fourier transform, with respect to the probe-position vector, of the recorded intensities at each detector pixel for the spatial frequencies corresponding to the component of the (*a*) (111), (*b*) (11$\bar{1}$) and (*c*) (002) reciprocal-lattice vectors along the probe line-scan direction. This figure therefore indicates where the detected intensity varies at the quoted frequency as the probe is scanned. Note how the intensities in all the overlap regions corresponding to the same relative reciprocal-lattice vector vary at the same spatial frequency.

At a particular spatial frequency in the image, $\boldsymbol{Q}_p$, the *G* data set contains the amplitude and phase of the variation of the intensity in the overlap function as a function of probe position for all diffracted beams separated by $\boldsymbol{Q}_p$.

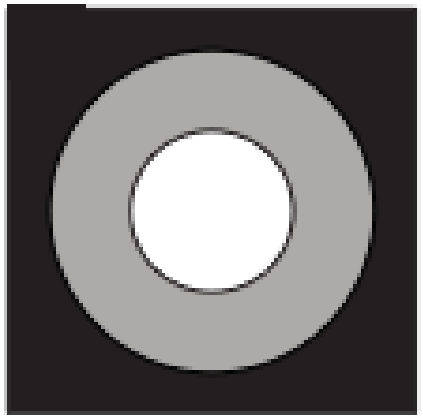
Nellist and Rodenburg, *Acta Cryst.* **54** (1998) 49

Can consider integrating over detector geometries

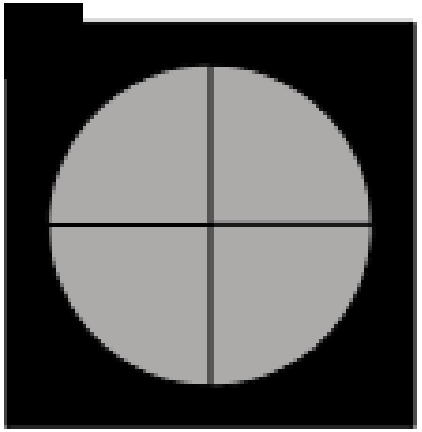
Conventional Bright Field



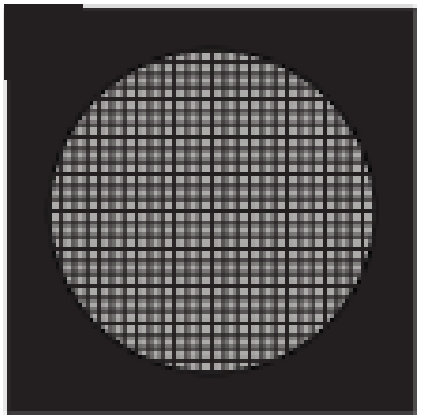
Annular Bright Field



Quadrants (DPC)

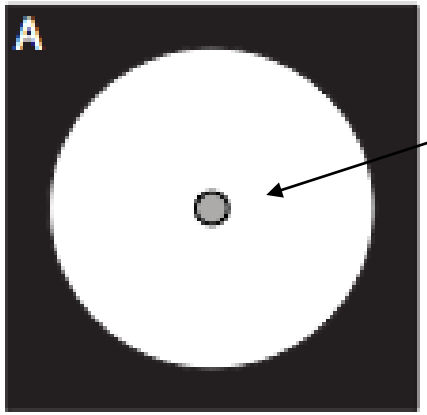


Ptychography Centre of Mass



Derivation of conventional bright-field

$$G\left(\boldsymbol{K}_f\ ,\boldsymbol{Q}_p\right) = A\left(\boldsymbol{K}_f\ \right)A^*\left(\boldsymbol{K}_f\ +\boldsymbol{Q}_p\right)\otimes_{\boldsymbol{K}_f}\Phi\left(\boldsymbol{K}_f\right)\Phi^*\left(\boldsymbol{K}_f-\boldsymbol{Q}_p\right)$$



Assume a small detector at the origin, therefore set $\boldsymbol{K}_f = 0$.

Double check my maths here!

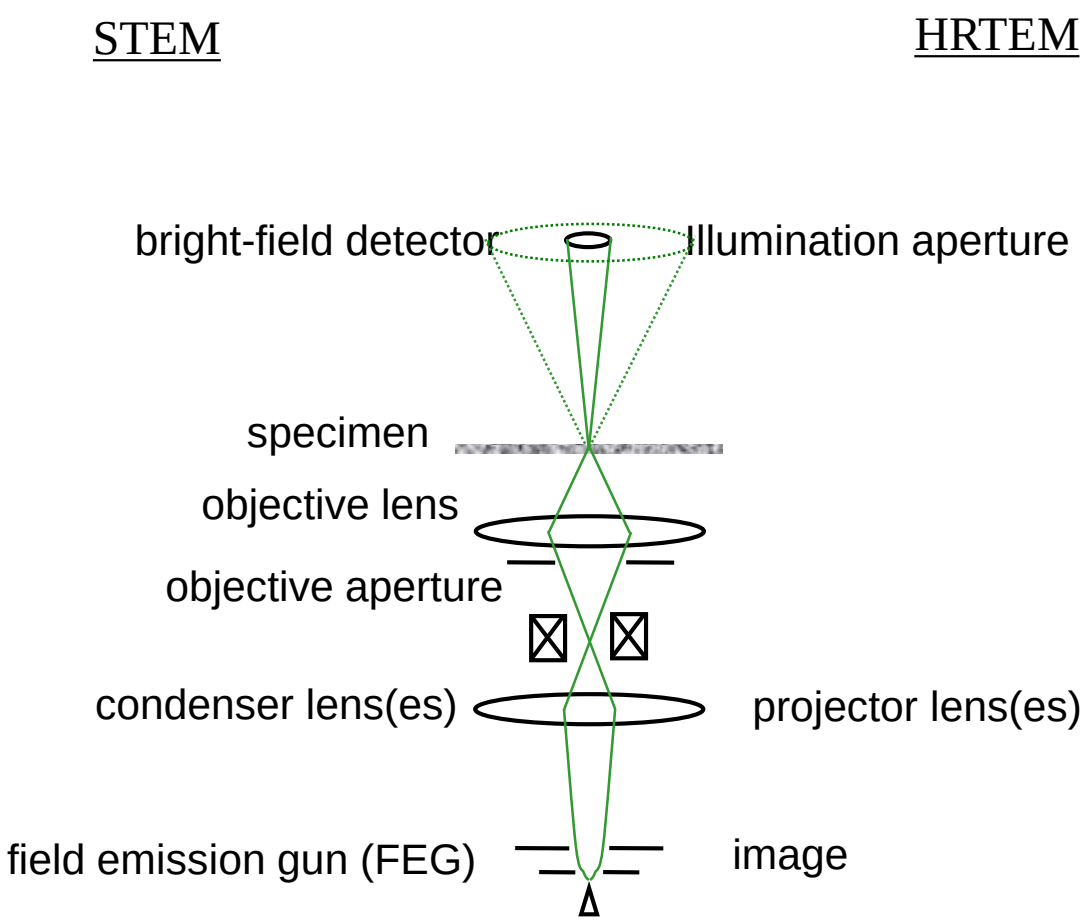
$$G\left(\boldsymbol{K}_f\ ,\boldsymbol{Q}_p\right) = \int A\left(\boldsymbol{K}'_f\right)\Phi\left(\boldsymbol{K}_f-\boldsymbol{K}'_f\right)A^*\left(\boldsymbol{K}'_f+\boldsymbol{Q}_p\right)\Phi^*\left(\boldsymbol{K}_f-\boldsymbol{K}'_f-\boldsymbol{Q}_p\right)d\boldsymbol{K}'_f$$

$$G\left(0,\boldsymbol{Q}_p\right) = \int A\left(\boldsymbol{K}'_f\right)\Phi\left(-\boldsymbol{K}'_f\right)A^*\left(\boldsymbol{K}'_f+\boldsymbol{Q}_p\right)\Phi^*\left(-\boldsymbol{K}'_f-\boldsymbol{Q}_p\right)d\boldsymbol{K}'_f$$

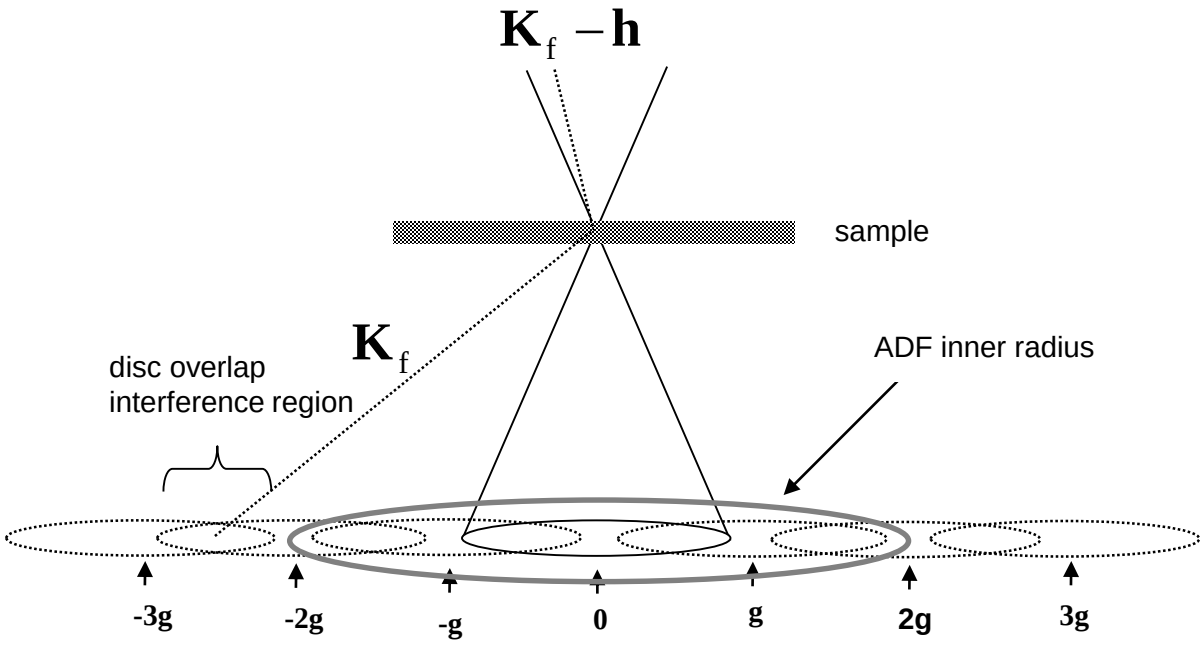
$$I(\boldsymbol{R_p}) = \left|P(\boldsymbol{R_p})\otimes\phi(-\boldsymbol{R_p})\right|^{\boldsymbol{2}}$$

Coherent imaging. Same formula lies at the heart of HRTEM.

Reciprocity



Summing many overlaps



- Assume a thin sample and ignoring the effects of thermal diffuse scattering for the moment.

Large detector leads to incoherence

$$I_{ADF}(\mathbf{Q}) = \int \sum_{\mathbf{g}} D_{ADF}(\mathbf{K}_p + \mathbf{g}) \phi_{\mathbf{g}} \phi^*_{\mathbf{g}-\mathbf{Q}} A(\mathbf{K}_p) A^*(\mathbf{K}_p + \mathbf{Q}) d\mathbf{K}_p$$

Ignore the $\mathbf{K}_p$ dependence of D_{ADF}, thus allowing separation of the integral and summation,

$$I_{ADF}(\mathbf{Q}) = \sum_{\mathbf{g}} D_{ADF}(\mathbf{g}) \phi_{\mathbf{g}} \phi^*_{\mathbf{g}-\mathbf{Q}} \int A(\mathbf{K}_p) A^*(\mathbf{K}_p + \mathbf{Q})\, d\mathbf{K}_p$$

F.T.

$$I_{ADF}(\mathbf{R}_p) = \left|\psi_p(\mathbf{R}_p)\right|^2 \otimes O(\mathbf{R}_p)$$

- This is the definition of incoherent imaging.
- The image is the object blurred by the intensity of the illuminating probe.
  - No need to use aberrations to generate contrast.
- Compare with coherent BF imaging $I(\boldsymbol{R_p}) = \left|P(\boldsymbol{R_p}) \otimes \phi(\boldsymbol{R_p})\right|^2$

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- Please do stop me to ask questions during the lecture

The *G* data set

$$G\left(\boldsymbol{K}_f, \boldsymbol{Q}_p\right) = \int A\left(\boldsymbol{K}'_f\right)\Phi\left(\boldsymbol{K}_f - \boldsymbol{K}'_f\right)A^*\left(\boldsymbol{K}'_f + \boldsymbol{Q}_p\right)\Phi^*\left(\boldsymbol{K}_f - \boldsymbol{K}'_f - \boldsymbol{Q}_p\right)d\boldsymbol{K}'_f$$

Which can be written using convolution notation:

$$G\left(\boldsymbol{K}_f, \boldsymbol{Q}_p\right) = A\left(\boldsymbol{K}_f\right)A^*\left(\boldsymbol{K}_f + \boldsymbol{Q}_p\right)\otimes_{\boldsymbol{K}_f}\Phi\left(\boldsymbol{K}_f\right)\Phi^*\left(\boldsymbol{K}_f - \boldsymbol{Q}_p\right)$$

So far we are not defining any one method (eg ptychography, DPC, COM etc), this is simply a statement of the physics of 4D STEM.

How should we interpret $G\left(\boldsymbol{K}_f, \boldsymbol{Q}_p\right)$?

Ptychography algorithms are all essentially different approaches to the deconvolution problem posed by the *G* function.

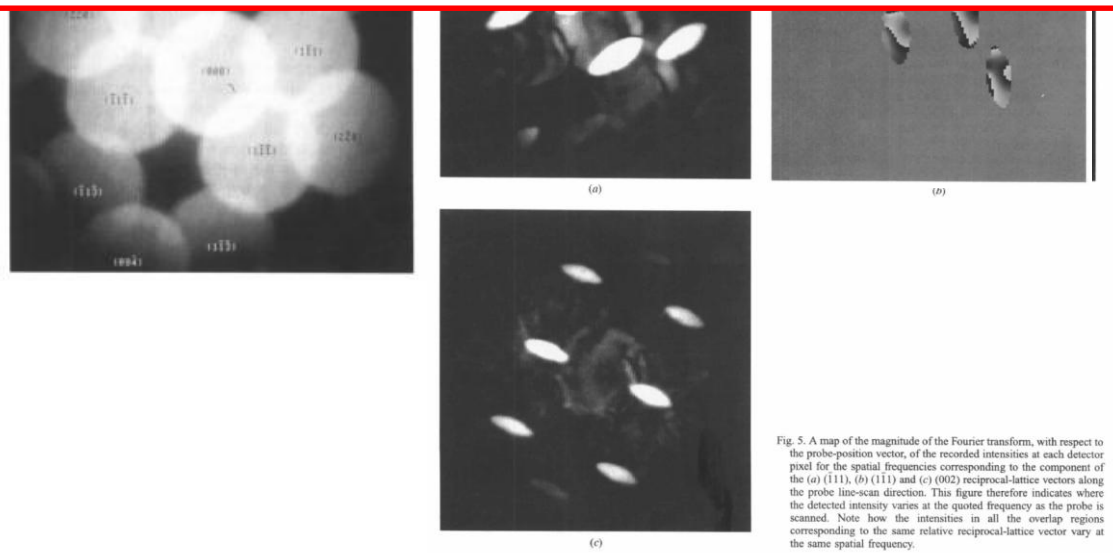


Fig. 5. A map of the magnitude of the Fourier transform, with respect to the probe-position vector, of the recorded intensities at each detector pixel for the spatial frequencies corresponding to the component of the (*a*) (111), (*b*) (11$\bar{1}$) and (*c*) (002) reciprocal-lattice vectors along the probe line-scan direction. This figure therefore indicates where the detected intensity varies at the quoted frequency as the probe is scanned. Note how the intensities in all the overlap regions corresponding to the same relative reciprocal-lattice vector vary at the same spatial frequency.

the *G* data set contains the amplitude and phase of the variation of the intensity in the overlap function as a function of probe position for all diffracted beams separated by $\boldsymbol{Q}_p$.

Nellist and Rodenburg, *Acta Cryst.* **54** (1998) 49

Retrieving an image from *G*

1. The weak phase approximation

$$\Phi(\boldsymbol{R}) = \exp\left[i\sigma V_p(\boldsymbol{R})\right] \approx 1 + i\sigma V_p(\boldsymbol{R})$$

phase object

weak phase object

projected potential

$$\Phi(\boldsymbol{K}) = \delta(\boldsymbol{K}) + i\sigma V_p(\boldsymbol{K})$$

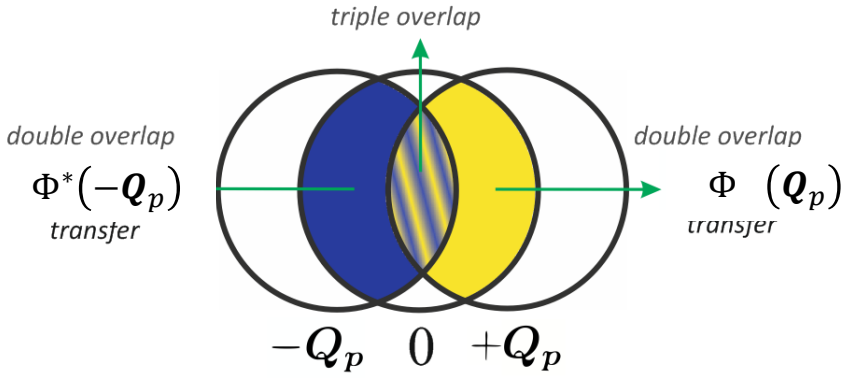
$$G\left(\boldsymbol{K}_f, \boldsymbol{Q}_p\right) = A\left(\boldsymbol{K}_f\right)A^*\left(\boldsymbol{K}_f + \boldsymbol{Q}_p\right) \otimes_{\boldsymbol{K}_f} \Phi\left(\boldsymbol{K}_f\right)\Phi^*\left(\boldsymbol{K}_f - \boldsymbol{Q}_p\right)$$

gives

$$G\left(\boldsymbol{K}_f, \boldsymbol{Q}_p\right) = \left|A\left(\boldsymbol{K}_f\right)\right|^2 \delta(\boldsymbol{Q}_p)$$

$$+A\left(\boldsymbol{K}_f - \boldsymbol{Q}_p\right)A^*\left(\boldsymbol{K}_f\right)i\sigma V_p(\boldsymbol{Q}_p)$$

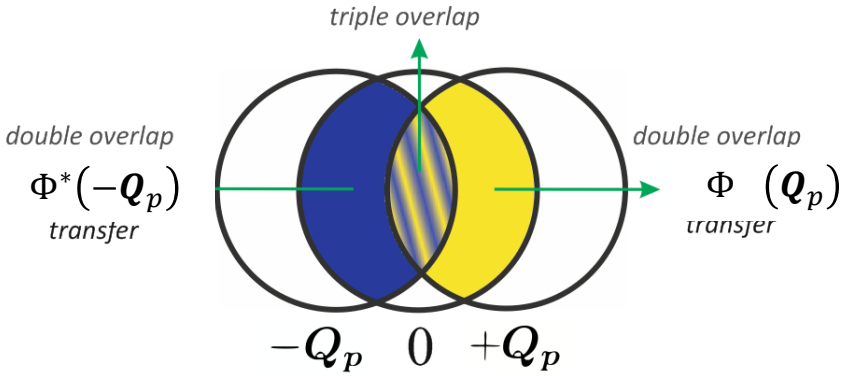
$$+A\left(\boldsymbol{K}_f\right)A^*\left(\boldsymbol{K}_f + \boldsymbol{Q}_p\right).-i\sigma V_p{}^*(-\boldsymbol{Q}_p)$$

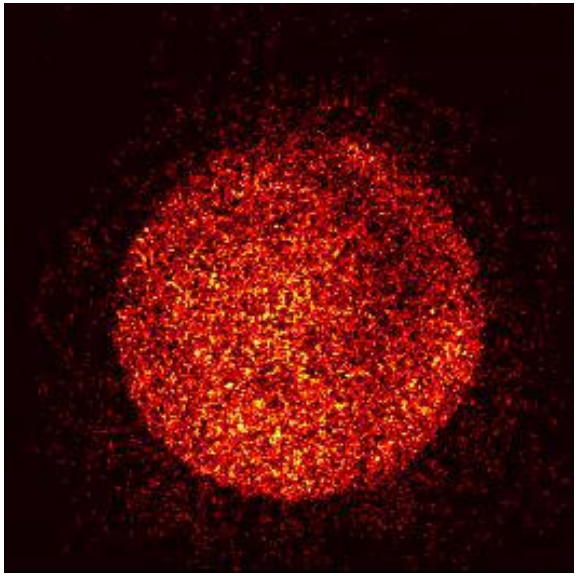


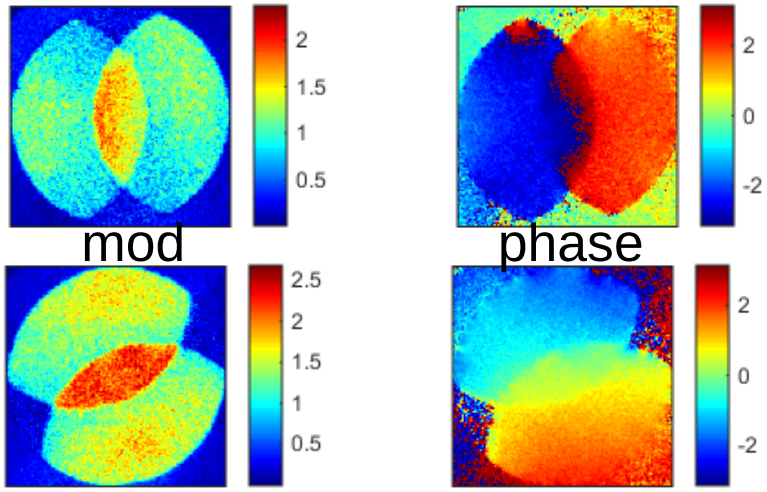
The weak phase approximation

Pennycook et al. *Ultramicroscopy* **151** (2015) 160.

The data appears to be a bright-field disc with fluctuations in intensity:







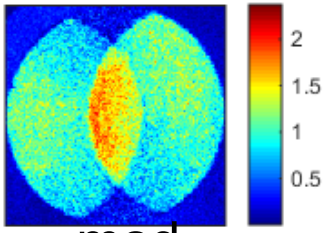
$G\left(\boldsymbol{K}_f, \boldsymbol{Q}_p\right)$ as a function of $\boldsymbol{K}_f$ for two different values of $\boldsymbol{Q}_p$

$$G\left(\boldsymbol{K}_f, \boldsymbol{Q}_p\right) = \left|A\left(\boldsymbol{K}_f\right)\right|^2 \delta(\boldsymbol{Q}_p) + A\left(\boldsymbol{K}_f - \boldsymbol{Q}_p\right)A^*\left(\boldsymbol{K}_f\right) i\sigma V_p(\boldsymbol{Q}_p) + A\left(\boldsymbol{K}_f\right)A^*\left(\boldsymbol{K}_f + \boldsymbol{Q}_p\right). -i\sigma V_p{}^*\left(-\boldsymbol{Q}_p\right)$$

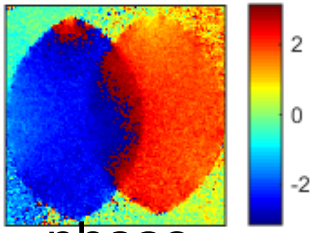
- Note that the phase in the aperture overlap functions is flat – aberration corrector.
- The two side-bands are exactly in anti-phase – shift of C of M.

The single side-band method of ptychography

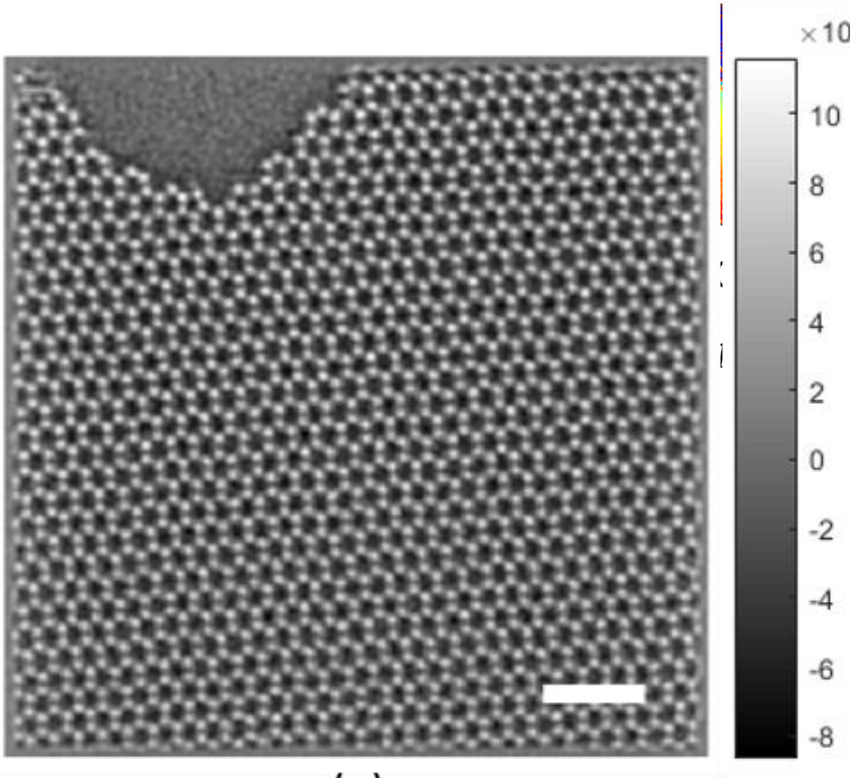
Pennycook et al. *Ultramicroscopy* **151** (2015) 160.
Yang et al. *Ultramicroscopy* **151** (2015) 232.



mod



phase



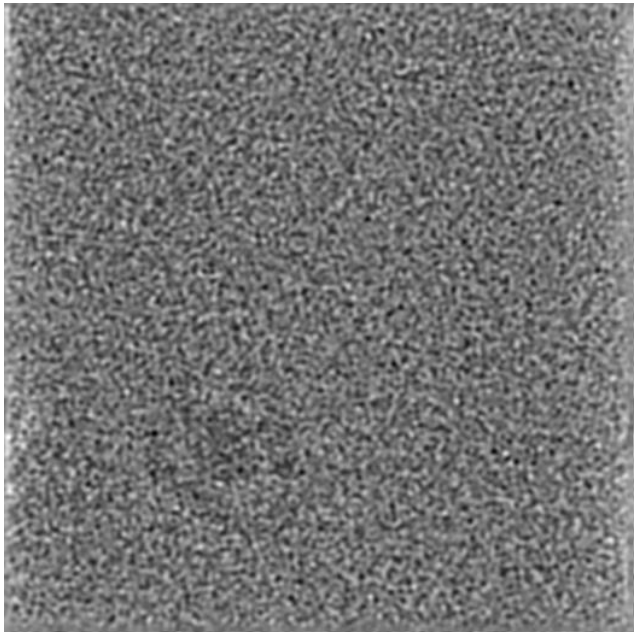
(a)

$$G\left(\boldsymbol{K}_f, \boldsymbol{Q}_p\right) = \left|A\left(\boldsymbol{K}_f\right)\right|^2 \delta(\boldsymbol{Q}_p) + A\left(\boldsymbol{K}_f - \boldsymbol{Q}_p\right)A^*\left(\boldsymbol{K}_f\right) i\sigma V_p(\boldsymbol{Q}_p) + A\left(\boldsymbol{K}_f\right)A^*\left(\boldsymbol{K}_f + \boldsymbol{Q}_p\right). -i\sigma {V_p}^* (-\boldsymbol{Q}_p)$$

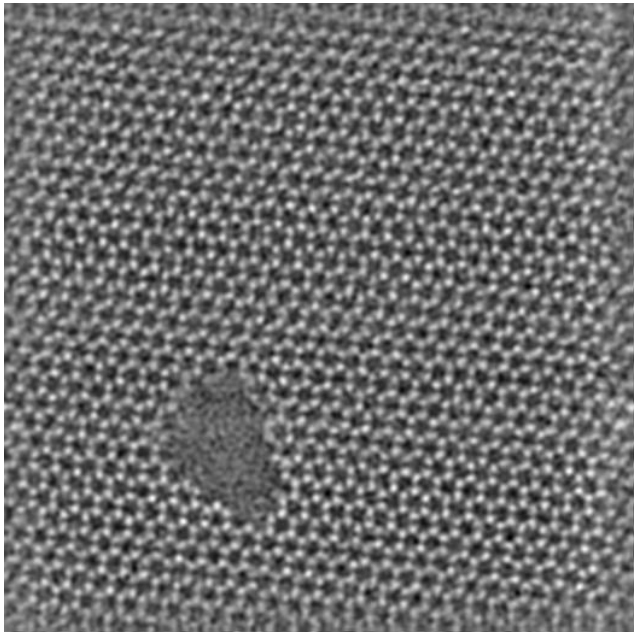
- For a particular spatial frequency, $\boldsymbol{Q}_p$, integrate over one of the double overlap regions (ie one side band) to get a value for $i\sigma V_p(\boldsymbol{Q}_p)$.
- No point in using both side bands, you simply double the signal and double the noise.
- For zero aberrations, there is no information in the triple overlap region

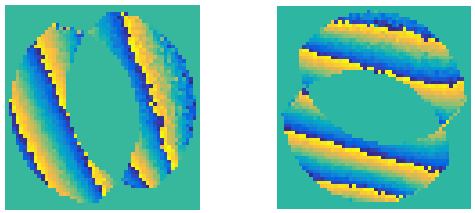
Correction of residual aberrations

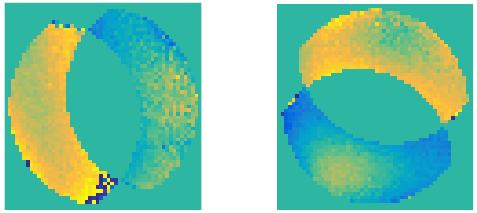
Uncorrected phase image



Corrected phase image

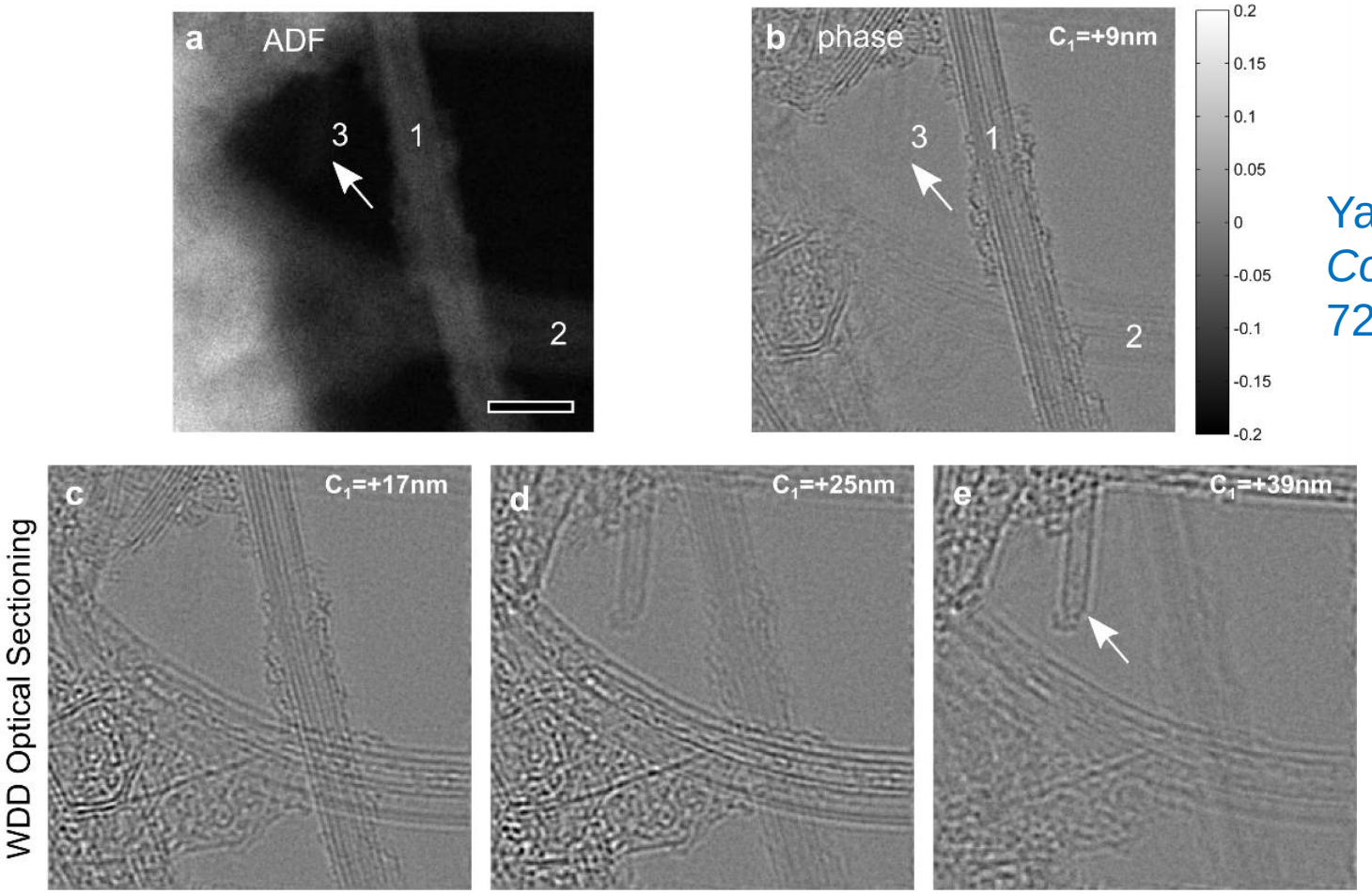






- If the phase is not flat in the overlap regions of $G\left(\boldsymbol{K}_f\ ,\boldsymbol{Q}_p\right)$ then you know you have some residual aberrations.
- Can fit the phase variation to a set of aberration coefficients and correct the data in post-processing
- Challenge problem: Show that defocus gives a linear phase ramp in *G*.

Optical sectioning

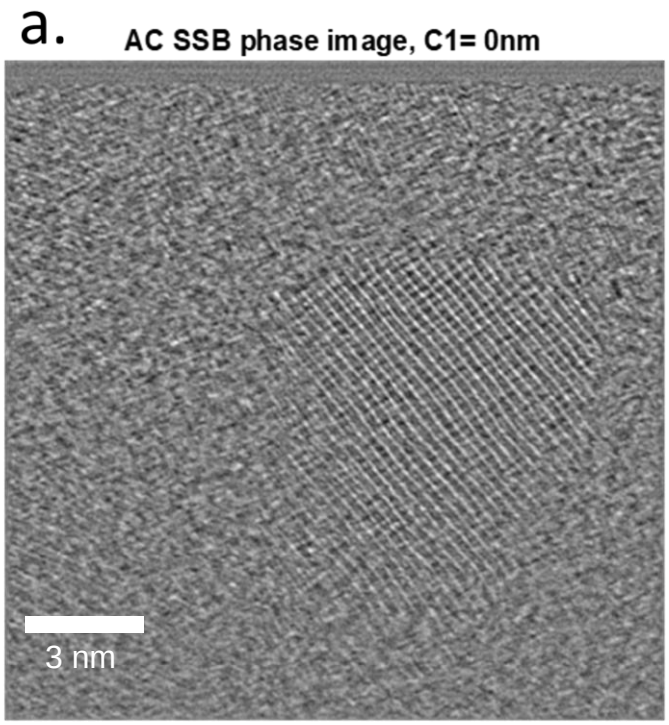


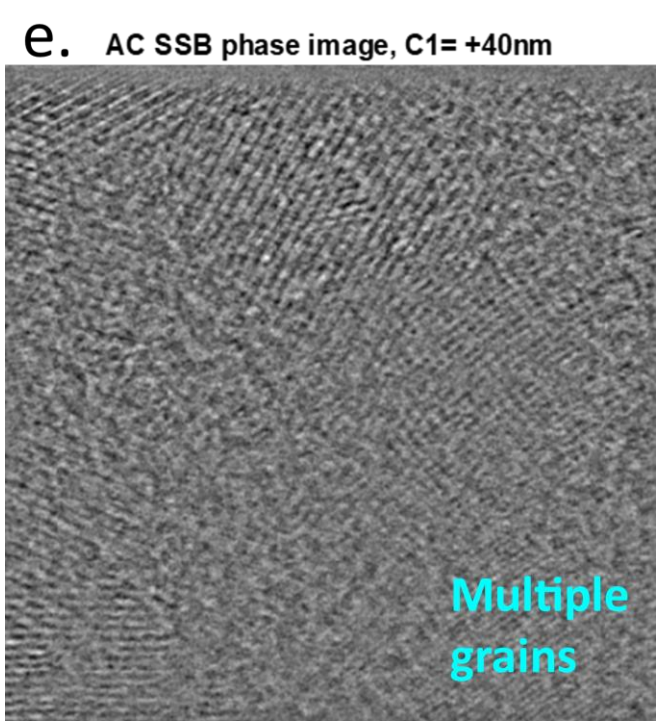
Yang et al. *Nat. Comm.* **6** (2015) 7266

These reconstructions are all from a single scan. It is not the case that some tubes are blurred when not at focal plane, their contribution to the image is reduced.

See also Wang et al. *Nat. Comm.* **6** (2015) 7266 for an ePIE demonstration

Ptychography optical sectioning of crystalline domains in polymers





poly (ethylene 2,6-naphthalate) (PEN)

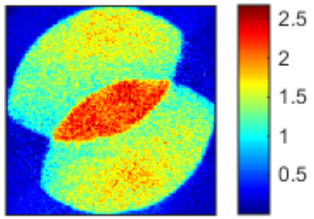
Unpublished work by Hui Luo.

Different layers sectioned from a single scan.

Quiz (just for fun!)

- The “ptych” in ptychography has a Greek root. Is the English equivalent:
  - A. “to overlap”
  - B. “to fold”
  - C. “to confuse”

Quiz (just for fun!)



- The disc overlap feature in *G* is sometimes referred to as a “pig’s trotter” (pied de cochon). Is it because:
  - A. The modulus of the *G* data set in 3D looks a bit like a pigs trotter.
  - B. The early pioneers of ptychography enjoyed discussing the topic over nice pork dinners.
  - C. Because early attempts at ptychography were so inept we said we were making a “pig’s ear” of it.

Quiz (just for fun!)

- The “PIE” name of the algorithm comes from:
  - A. Ptychography Is Everything (PIE)
  - B. Ptychographic Iterative Engine (PIE).
  - C. The baked food involving pastry, as in “This ****ing ptychography stuff is a load of pie in the sky!” [“pie in the sky” means unlikely to work]

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- Please do stop me to ask questions during the lecture

Retrieving an image from *G*

2. Stronger objects

Rodenburg and Bates, Phil. Trans. R. Soc London A, **339** (1992) 521.

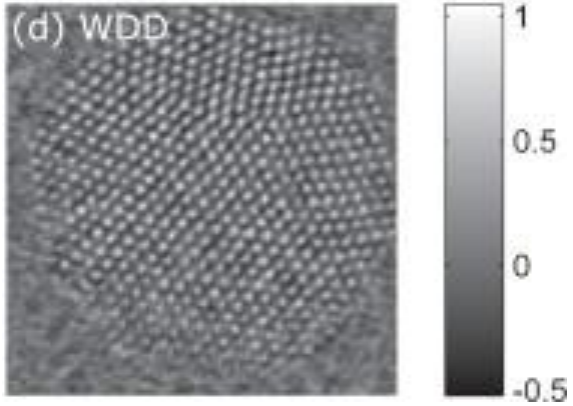
$$G\left(\boldsymbol{K}_f\ ,\boldsymbol{Q}_p\right)=A\left(\boldsymbol{K}_f\ \right)A^*\left(\boldsymbol{K}_f\ +\boldsymbol{Q}_p\right)\otimes_{\boldsymbol{K}_f}\Phi\left(\boldsymbol{K}_f\right)\Phi^*\left(\boldsymbol{K}_f-\boldsymbol{Q}_p\right)$$

Let us simply deconvolve the aperture functions from the transmission functions:

1. Fourier transform $G\left(\boldsymbol{K}_f\ ,\boldsymbol{Q}_p\right)$ with respect to $\boldsymbol{K}_f$.
2. The convolution becomes a product between two functions, which are known as Wigner distribution functions.

$$H\left(\boldsymbol{R_f},\boldsymbol{Q}_p\right)=\chi_A\left(\boldsymbol{R_f},-\boldsymbol{Q}_p\right).\,\chi_\Phi\left(\boldsymbol{R_f},\boldsymbol{Q}_p\right)$$

3. Then divide by the $\chi_A\left(\boldsymbol{R_f},-\boldsymbol{Q}_p\right)$ function using a Wiener deconvolution



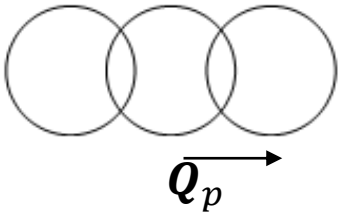
$$\chi_\Phi\left(\boldsymbol{R_f},\boldsymbol{Q}_p\right)=\frac{\chi_A^*\left(\boldsymbol{R_f},-\boldsymbol{Q}_p\right)H\left(\boldsymbol{R_f},\boldsymbol{Q}_p\right)}{\left|\chi_A^*\left(\boldsymbol{R_f},-\boldsymbol{Q}_p\right)\right|^2+\varepsilon}$$

1. Now set $\boldsymbol{K}_f=0$ in the retrieved $\Phi\left(\boldsymbol{K}_f\right)\Phi^*\left(\boldsymbol{K}_f-\boldsymbol{Q}_p\right)$ function.

Super-resolution

$$G\left(\boldsymbol{K}_f\ ,\boldsymbol{Q}_p\right) = A\left(\boldsymbol{K}_f\ \right)A^*\left(\boldsymbol{K}_f\ +\boldsymbol{Q}_p\right)\otimes_{\boldsymbol{K}_f}\Phi\left(\boldsymbol{K}_f\right)\Phi^*\left(\boldsymbol{K}_f-\boldsymbol{Q}_p\right)$$

The *G* function has non-zero values for $\left|\boldsymbol{Q}_p\right| < \frac{2\alpha}{\lambda}$ ie smaller than the diameter of the BF disc.

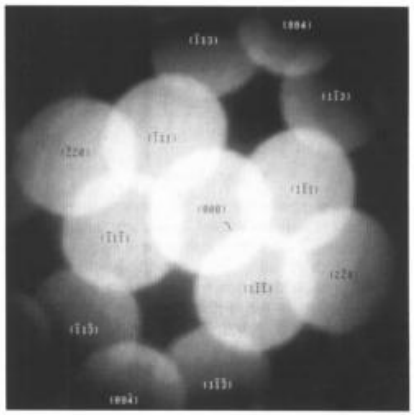


"Now set $\boldsymbol{K}_f = 0$ in the retrieved $\Phi\left(\boldsymbol{K}_f\right)\Phi^*\left(\boldsymbol{K}_f-\boldsymbol{Q}_p\right)$ function."
This allows reconstruction of the sample up to a resolution given by the diameter of the bright-field disc (twice resolution of conventional bright-field imaging, but the same resolution as ADF).

"Now set $\boldsymbol{K}_f = -\boldsymbol{Q}_p$in the retrieved $\Phi\left(\boldsymbol{K}_f\right)\Phi^*\left(\boldsymbol{K}_f-\boldsymbol{Q}_p\right)$ function" – resolution beyond the conventional diffraction limit.

"Stepping out"

Super-resolution using ptychography

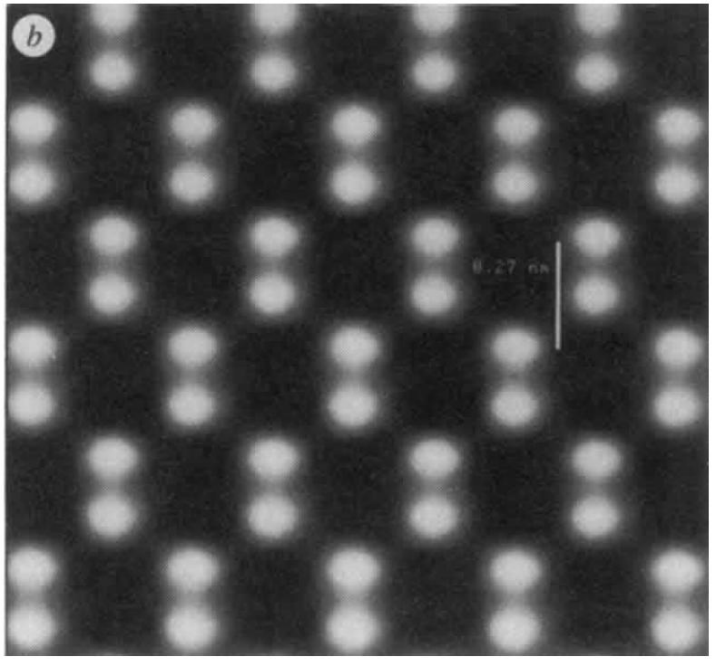


(a)

(b)

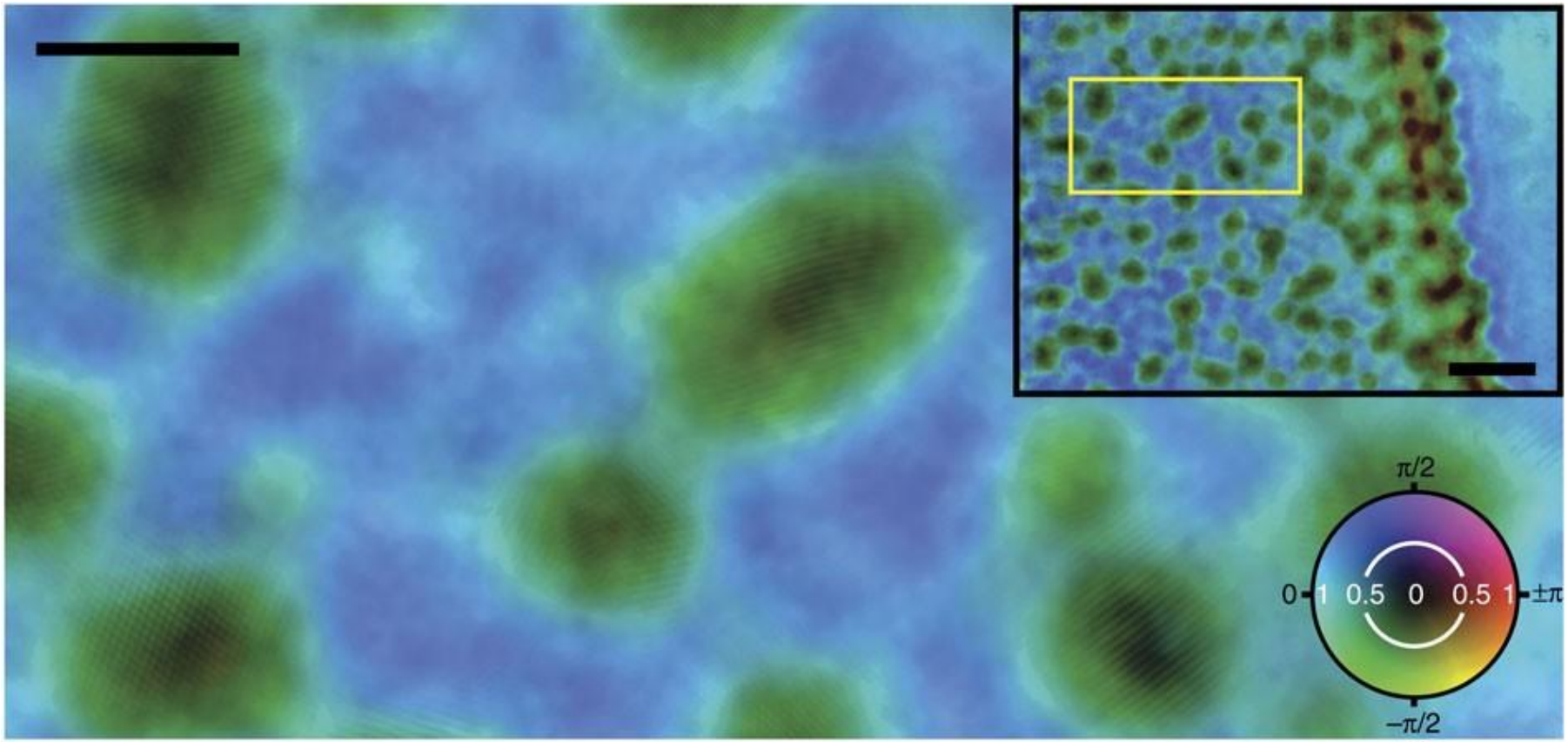
(c)

Fig. 5. A map of the magnitude of the Fourier transform, with respect to the probe-position vector, of the recorded intensities at each detector pixel for the spatial frequencies corresponding to the component of the (*a*) (111), (*b*) (111) and (*c*) (002) reciprocal-lattice vectors along the probe line-scan direction. This figure therefore indicates where the detected intensity varies at the quoted frequency as the probe is scanned. Note how the intensities in all the overlap regions corresponding to the same relative reciprocal-lattice vector vary at the same spatial frequency.



Nellist et al., *Nature* (1995): 1.36 Å
Jiang et al., *Nature (*2018): 0.39 Å
Chen et al., *Nature* (2021): 0.20 Å

Atomic resolution in an SEM



Humphry et al., *Nat. Comm.* **3** (2012) 730.

The resolution can be much higher than the stability of the microscope!

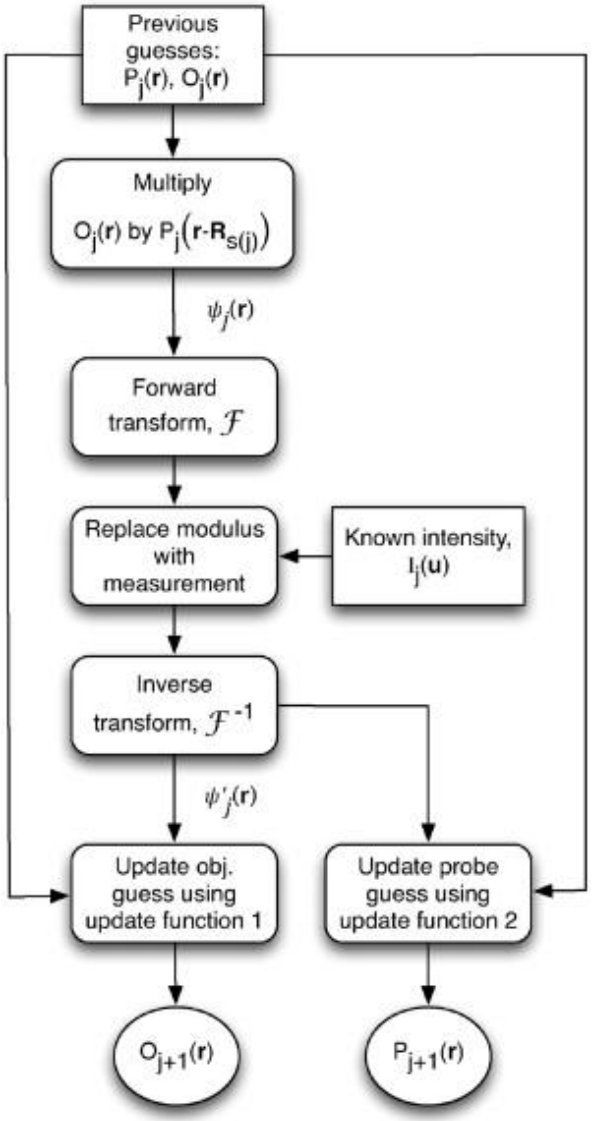
3. Iterative methods - ePIE

Maiden and Rodenburg, *Ultramicroscopy* **109** (2009) 1256

Using forward convolutions rather than deconvolutions avoids the risks associated with deconvolutions.

Solves for illumination even for strong objects.

Gives super-resolution.



Data processing in 4D STEM

$$G\left(\boldsymbol{K}_f\ ,\boldsymbol{Q}_p\right) = A\left(\boldsymbol{K}_f\ \right)A^*\left(\boldsymbol{K}_f\ +\boldsymbol{Q}_p\right)\otimes_{\boldsymbol{K}_f}\Phi\left(\boldsymbol{K}_f\right)\Phi^*\left(\boldsymbol{K}_f-\boldsymbol{Q}_p\right)$$

- For a weak phase object the convolution separates itself
  - Single side-band approximation
- Deconvolve the aperture function part directly
  - Wigner distribution deconvolution – super-resolution [Rodenburg and Bates]
- Do the deconvolution through forward iterative matching
  - (extended) Ptychographic Iterative Engine (e)PIE [Rodenburg et al.]
- Integrate weighted by the diffraction vector

$$\int \boldsymbol{K}_f G\left(\boldsymbol{K}_f,\boldsymbol{Q_p}\right)\,d\boldsymbol{K}_f$$

  - Centre of Mass or First Moment [Müller-Caspary]

Centre of Mass (CoM) or First Moment Approach

$$G\left(\boldsymbol{K}, \boldsymbol{Q_p}\right) = A(\boldsymbol{K})A^*\left(\boldsymbol{K} + \boldsymbol{Q_p}\right) \otimes_{\boldsymbol{K}} \Phi(\boldsymbol{K})\Phi^*\left(\boldsymbol{K} - \boldsymbol{Q_p}\right)$$

Integrate weighted by the diffraction vector $\int \boldsymbol{K} G\left(\boldsymbol{K}, \boldsymbol{Q_p}\right) d\boldsymbol{K}$

$$\boldsymbol{I_{CoM}}\left(\boldsymbol{Q_p}\right) = \int \boldsymbol{K}\, G\left(\boldsymbol{K}, \boldsymbol{Q_p}\right) d\boldsymbol{K} = \iint \boldsymbol{K} A(\boldsymbol{K'})A(\boldsymbol{K'} + \boldsymbol{Q_p})\Phi(\boldsymbol{K} - \boldsymbol{K'})\Phi^*\left(\boldsymbol{K} - \boldsymbol{K'} - \boldsymbol{Q_p}\right) d\boldsymbol{K'} d\boldsymbol{K}$$

The $\boldsymbol{K'}$ integral has a limited domain (the disc overlap). If we assume that the $\boldsymbol{K}$ integral is over the entire detector, then we can ignore the $\boldsymbol{K'}$ in the specimen transmission function part.

$$\boldsymbol{I_{CoM}}\left(\boldsymbol{Q_p}\right) = \int A(\boldsymbol{K'})A\left(\boldsymbol{K'} + \boldsymbol{Q_p}\right) d\boldsymbol{K'} \times \int \boldsymbol{K}\, \Phi(\boldsymbol{K})\Phi^*\left(\boldsymbol{K} - \boldsymbol{Q_p}\right) d\boldsymbol{K}$$

$$\boldsymbol{I_{CoM}}\left(\boldsymbol{R_p}\right) = \left|P(\boldsymbol{R_p})\right|^2 \otimes \boldsymbol{E}(\boldsymbol{R_p})$$

Müller-Caspary, K., et al. *Ultramicroscopy* **178** (2017).

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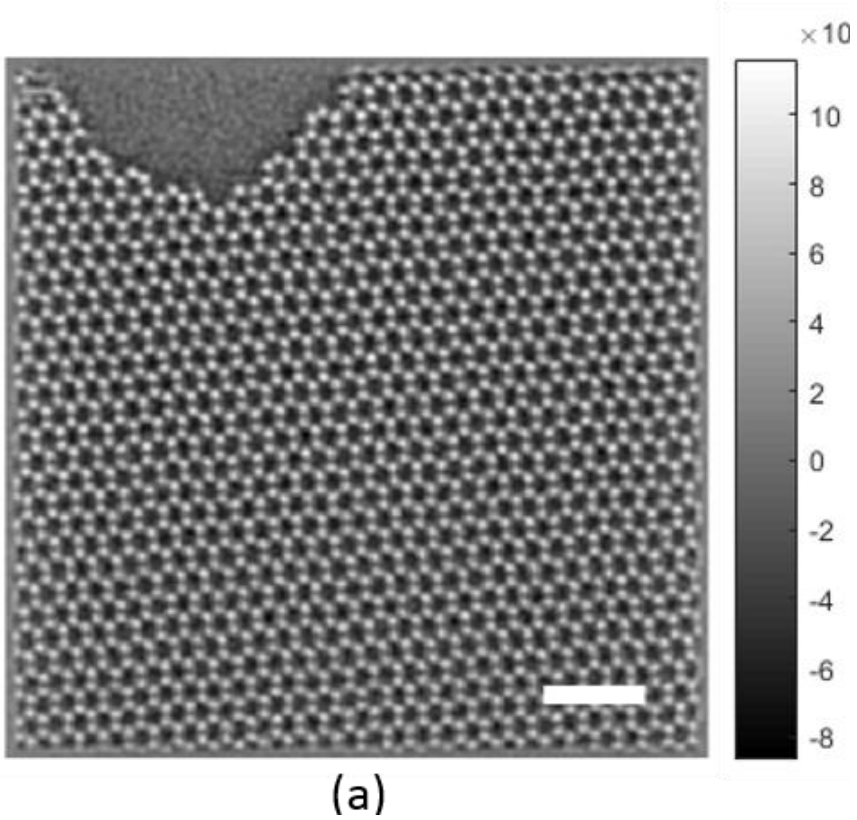
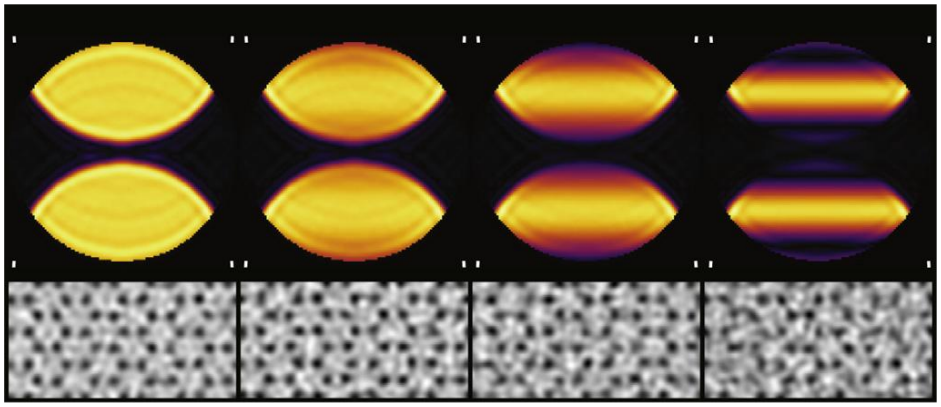
Effect of chromatic defocus spread



(a)



The centres of the overlap regions are achromatic (not attenuated by defocus spread).

No need for monochromators or Cc correctors.

Pennycook, T. J. et al. *Ultramicroscopy* **196** (2019) 131.

Nellist and Rodenburg, *Ultramicroscopy* **54** (1994) 61.

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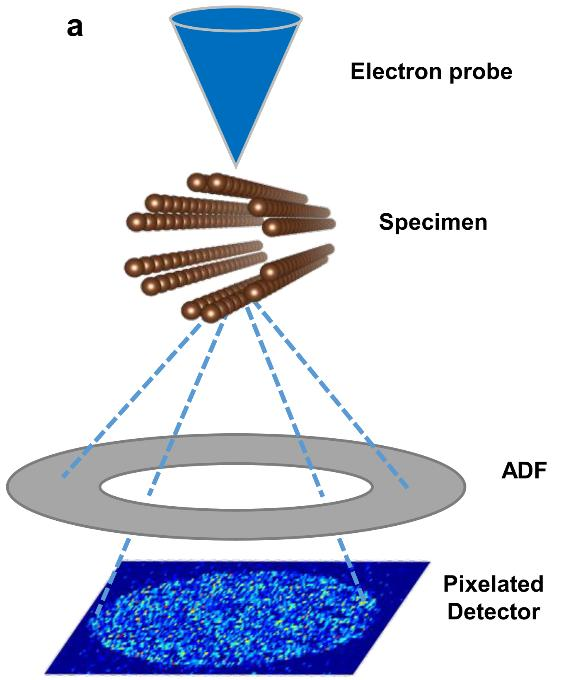
Camera frame speed dictates STEM probe dwell time



- Many cameras operate at a few kHz frame speed, so the corresponding probe dwell time is >100 μs.
- Conventional STEM dwell times may be ~ a few μs.
- Difficulty with operating the microscope with very low beam currents.

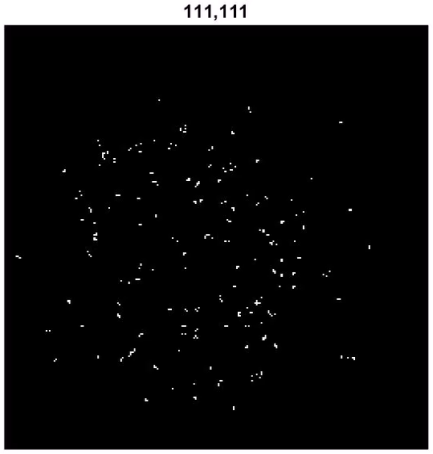
Speed or dynamic range?

- 1 kHz frame rate – 1+ minute acquisition time
  - Scan distortions
  - Drift in aberrations
  - Sample drift
  - Large dose

Run in 1-bit mode to increase frame rate - does ptychography still work?

Medipix3 detector speeds:

- 24-bit: 0.6 kHz
- 12-bit: 1.2 kHz
- 6-bit: 2.4 kHz
- **1-bit: 14.4 kHz**



C. M. O'Leary, et al., *APL* (2020)

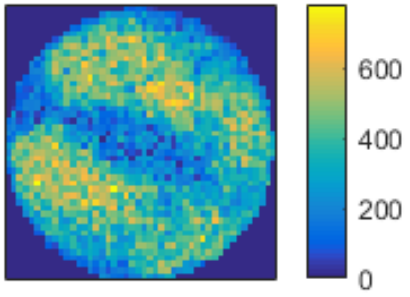
OxfordMaterials

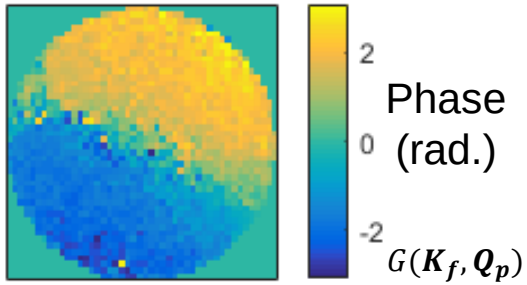
Image reconstruction from binary data

aluminosilicate zeolite (ZSM-5)

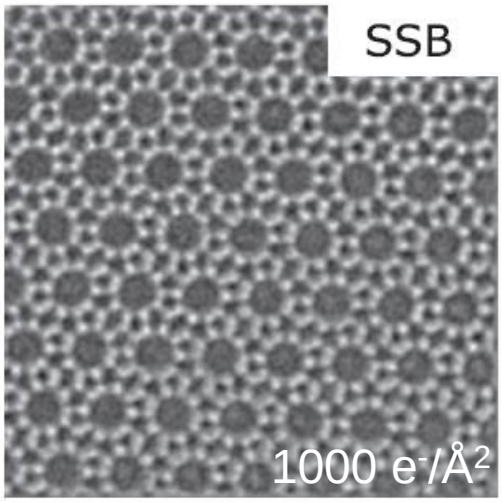
Enough statistics from sparse CBED patterns to get a meaningful phase measurement.

Amplitude

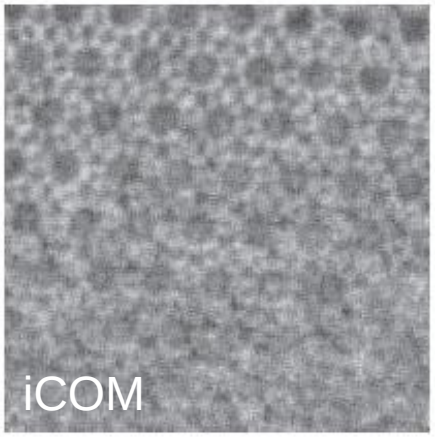




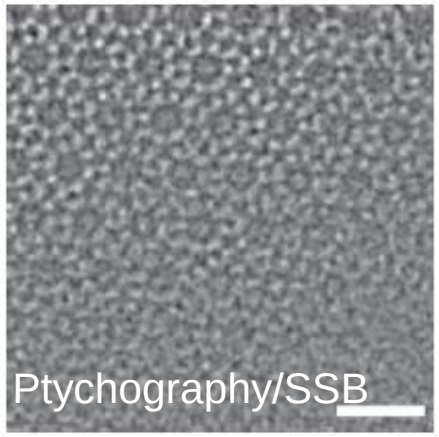
O'Leary et al. *APL* (2020)



(a)



(b)



(c)

Outline

- 4D STEM
- The phase problem
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  - Single side-band method – correction of aberrations – optical sectioning
  - Wigner distribution deconvolution – super-resolution
  - Iterative methods (ePIE)
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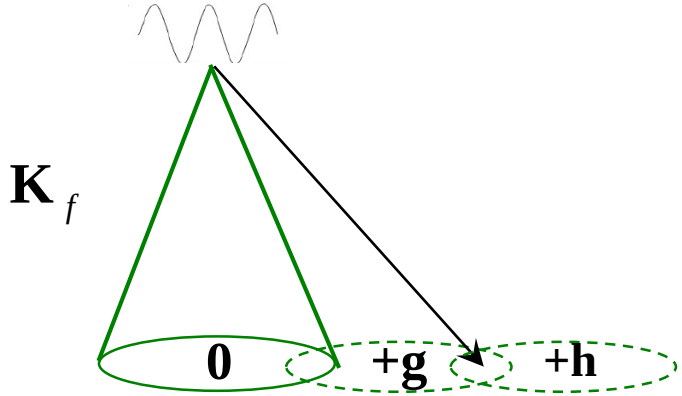
- Effect of partial coherence
- Dynamic range
- Sampling in real and reciprocal space
- Transfer functions, signal to noise and detective quantum efficiency

- Please do stop me to ask questions during the lecture

Interference between diffracted beams



$$I(\boldsymbol{K}_f) = \left|F_{\boldsymbol{g}} A(\boldsymbol{K}_f - \boldsymbol{g}) \exp\left[i2\pi(\boldsymbol{K}_f - \boldsymbol{g}).\boldsymbol{R}_p\right] + F_{\boldsymbol{h}} A(\boldsymbol{K}_f - \boldsymbol{h}) \exp\left[i2\pi(\boldsymbol{K}_f - \boldsymbol{h}).\boldsymbol{R}_p\right]\right|^2$$

- Assume no lens aberrations.

$$I(\boldsymbol{K}_f) = \left|F_{\boldsymbol{g}} A(\boldsymbol{K}_f - \boldsymbol{g})\right|^2 + \left|F_{\boldsymbol{h}} A(\boldsymbol{K}_f - \boldsymbol{h})\right|^2 + 2\left|F_{\boldsymbol{g}}\right|\left|F_{\boldsymbol{h}}\right| A(\boldsymbol{K}_f - \boldsymbol{g}) A(\boldsymbol{K}_f - \boldsymbol{h}) \cos\left[i2\pi(\boldsymbol{g} - \boldsymbol{h}).\boldsymbol{R}_p + \angle F_{\boldsymbol{g}}\right]$$

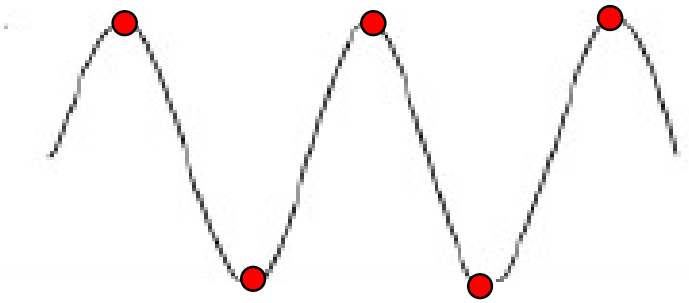
The maximum spatial frequency with which an overlap can oscillate as the probe scans is given by the aperture diameter.

Sampling in real space

The maximum spatial frequency with which an overlap can oscillate as the probe scans is given by the aperture diameter.

The *G* function has non-zero values for $\left|\boldsymbol{Q}_p\right| < \frac{2\alpha}{\lambda}$ ie smaller than the diameter of the BF disc.

Nyquist says we need two samples per cycle of the intensity

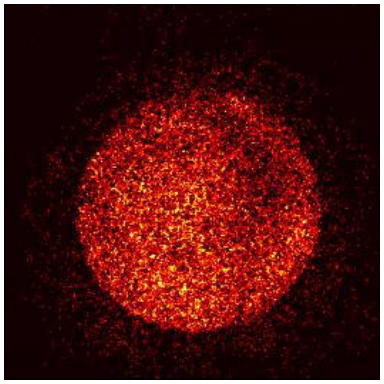


For focused probe ptychography, the probe sampling must be smaller than $\frac{\lambda}{4\alpha}$ (same as ADF imaging).

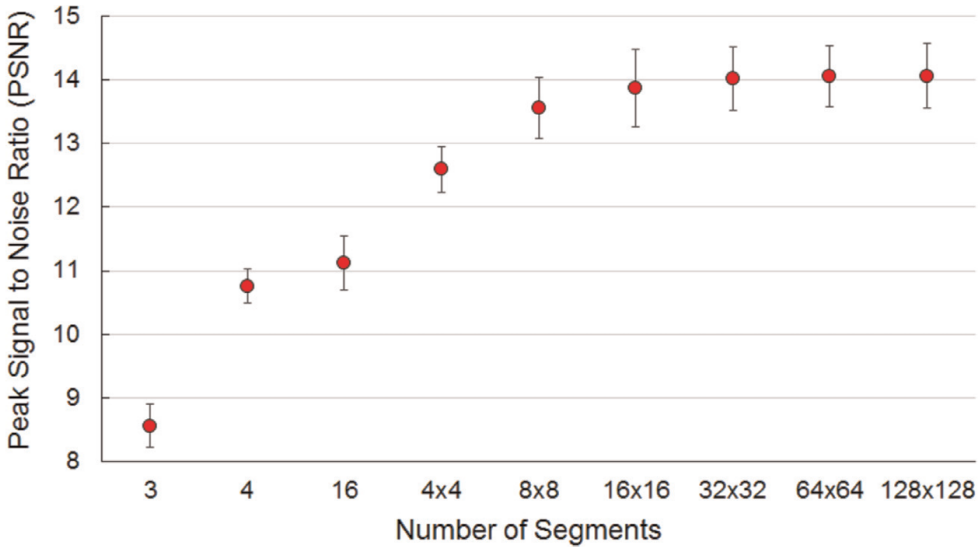
Defocused probe ptychography has a bigger probe and can have larger step size, but still need sufficient overlap of illumination areas.

Sampling in reciprocal space

- For in-focus 4D STEM, the intensity fluctuations are on a large scale.
  - Can use large pixels or segmented detectors
    - DPC



H. Yang et al. / Ultramicroscopy 151 (2015) 232–239

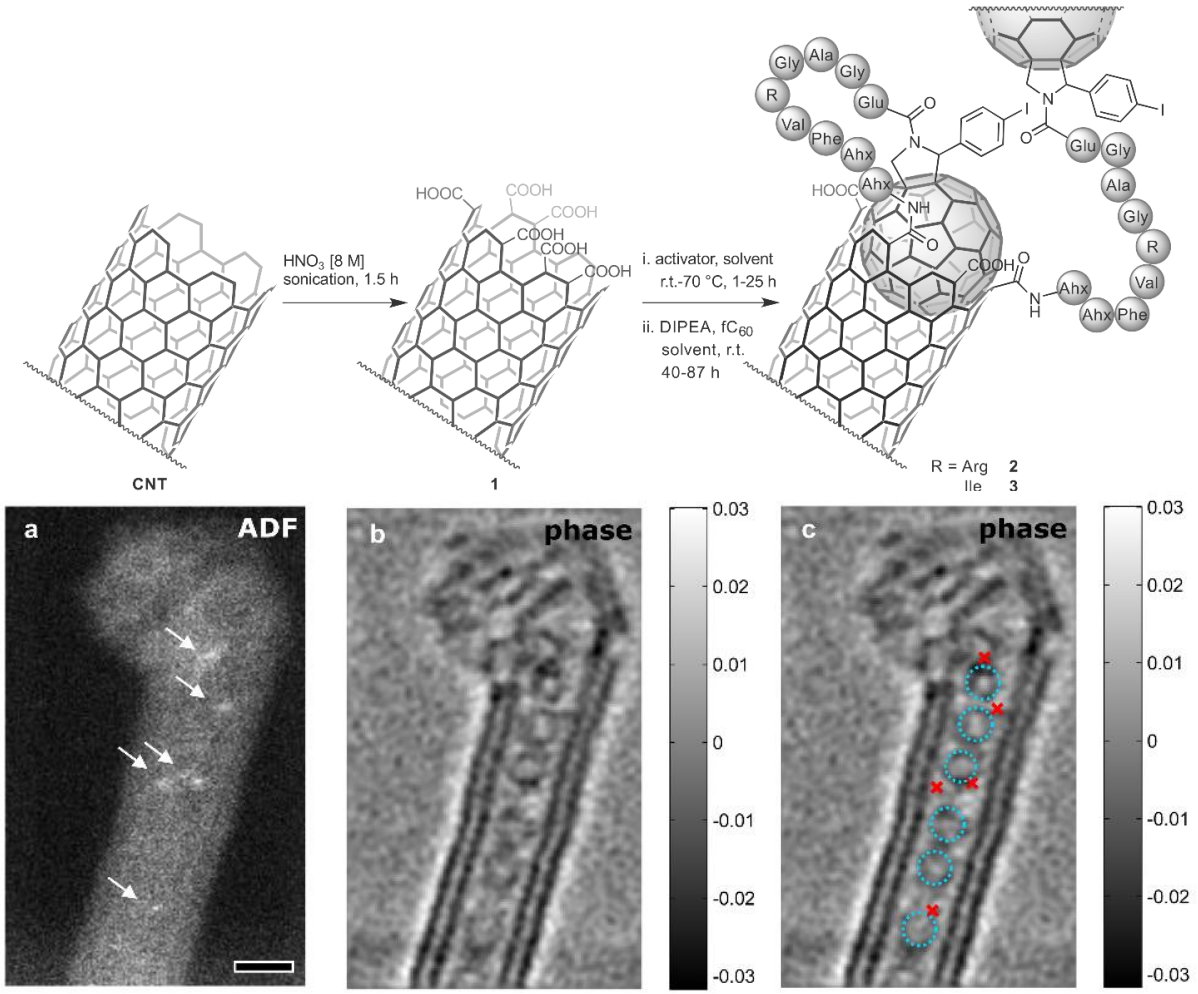


For defocused probe ptychography, you have a bigger probe, so can sample less frequency, but have small features in the diffraction pattern, so need more pixels (see slide 15). Sampling required is:

$$\Delta\theta = \frac{\lambda}{4\alpha C_{1,0}}$$

Application 1
Simultaneous light and heavy element detection

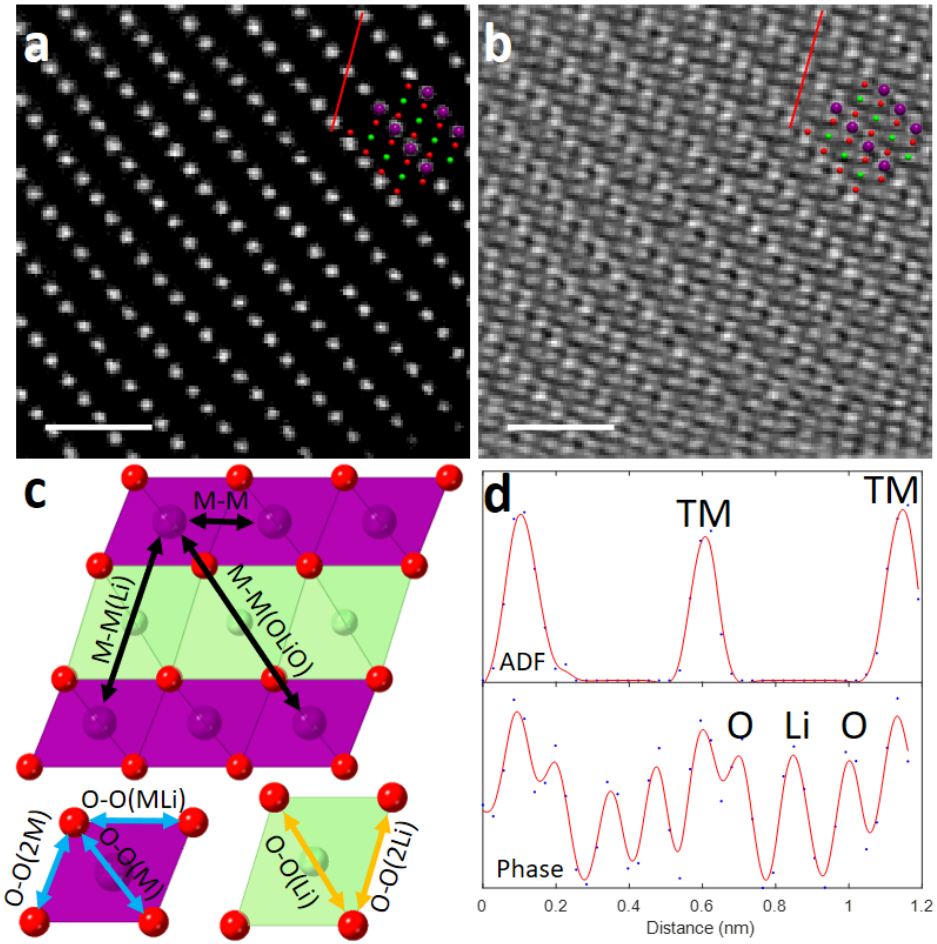
Ptychography give phase imaging at zero aberrations (no need for Scherzer conditions), so ideal for simultaneous imaging with ADF.



Yang et al. *Nat. Comm.* **6** (2015) 7266

Application 1:
Simultaneous light and heavy element detection

Simultaneous ADF and phase imaging is ideal for battery materials.



Non-rigid registered fast-scan ADF images used for TM distance measurement.

TM locations in ADF image simultaneous with ptychography allows distortion correction of ptychography images and accurate measurements of O octahedral distortions.

Song et al. *Joule* **(2022)**

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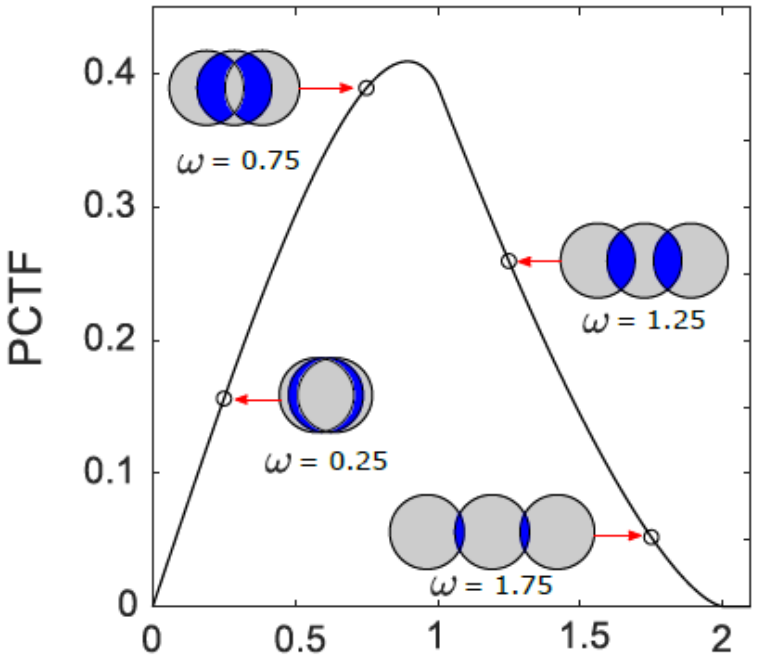
Can we define a transfer function for ptychography?

Take PCTF to be area of signal on detector

- At zero aberrations:
  - The two double overlap regions oscillate in antiphase
  - There is no variation in intensity wrt to probe in triple overlap region.
  - No phase contrast for a centrosymmetric detector

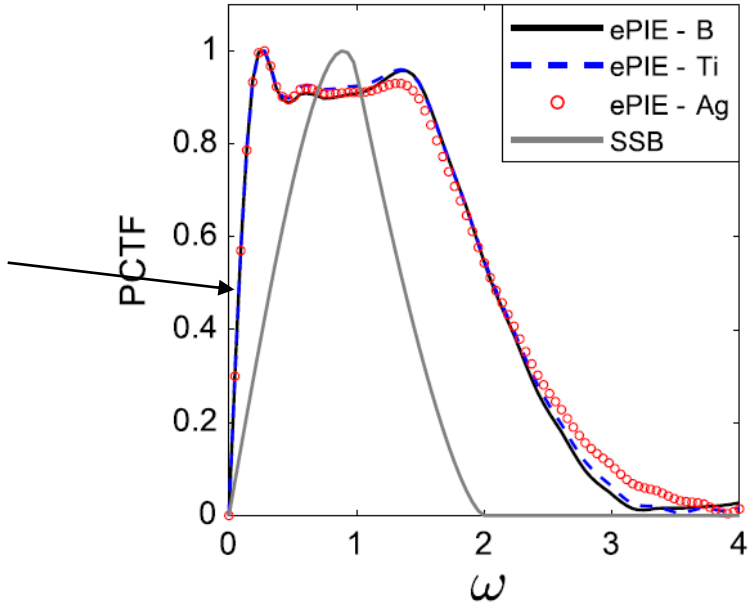


$$\omega = Q_p/\alpha$$

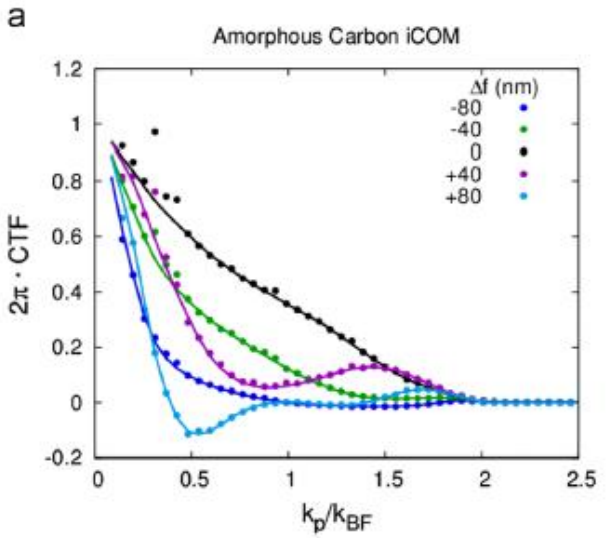
Rodenburg et al. *Ultramicroscopy* (1993)
Pennycook et al. *Ultramicroscopy* (2015)
Yang et al. *Ultramicroscopy* (2015)

Other reconstruction methods

Danger of noise amplification?



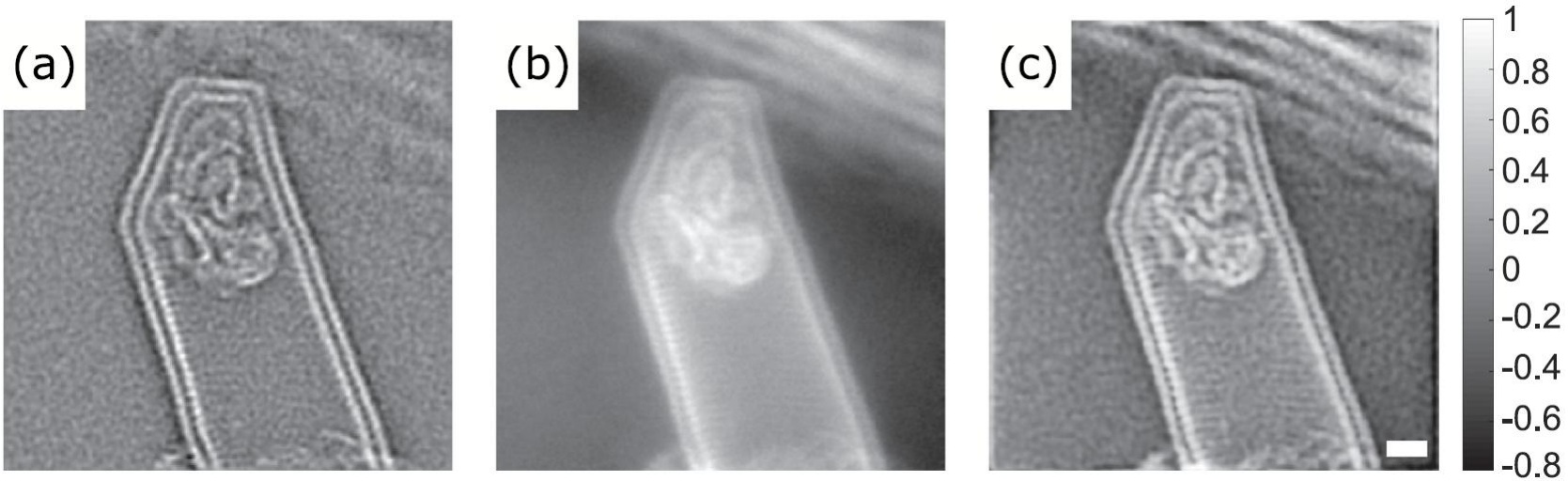
O'Leary et al.
Ultramicroscopy (*2020)*



iDPC [Lazic et al. Ultramicroscopy (2016)]

Does the reconstruction method dictate the amount of information?
How does 4D STEM phase imaging compare with TEM?

Images from different reconstruction methods



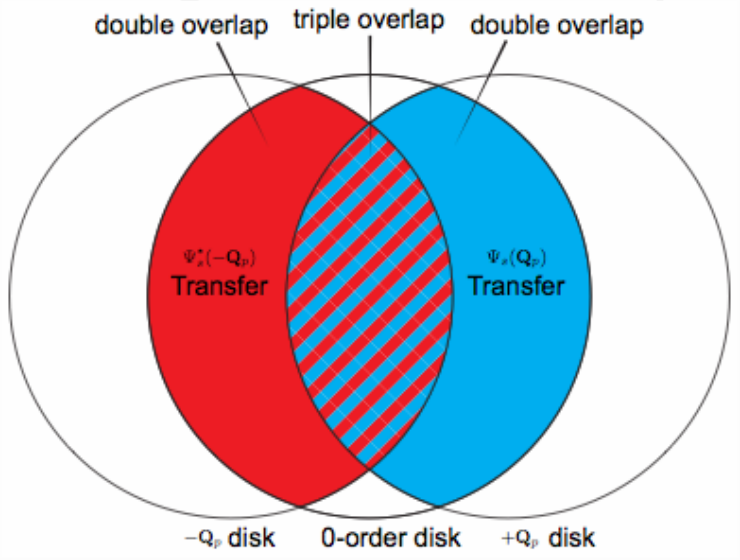
SSB

ePIE

Fourier filtered ePIE

All from the same 4D-STEM data set

Signal to noise spectrum – inherent denoising



4D STEM allows for a detector geometry that adapts as a function of spatial frequency in the image

Noise contribution to ptychography: variance is proportion to overlap area, so SD is square root of overlap area = sqrt(PCTF). Normalised PCTF is therefore = sqrt (PCTF).

Quantitative comparison of different electron imaging methods methods

- Common measures for image quality: peak signal to noise ratio, PCTF, resolution, etc…
- The ultimate quantity for microscope quality is
  - Sample independent
  - Dose independent

$\Rightarrow$ Detective Quantum Efficiency (DQE)

$\Rightarrow$ "The **detective quantum efficiency** (often abbreviated as **DQE**) is a measure of the combined effects of the signal (related to image contrast) and noise performance of an imaging system, generally expressed as a function of spatial frequency."

Acknowledgement to Felix Bennemann and discussions with David Muller

Detective quantum efficiency (DQE)

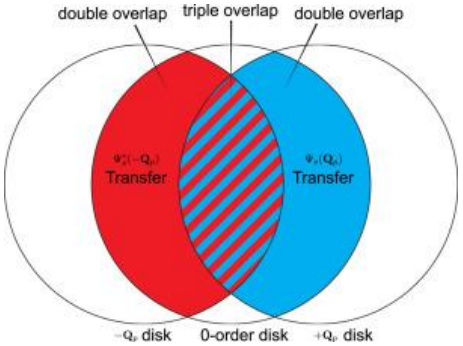
- SNR_{out}: final signal to noise ratio, ptychographic reconstruction SNR
- SNR_{in}: initial signal to noise ratio, ideal signal to noise ratio, Zernike phase plate microscopy

$$DQE = \frac{SNR_{out}^2}{SNR_{in}^2}$$

See also Dwyer and Paginin, *PRB (2024).*

Assume linear imaging (ie weak phase)

Analytical DQE – SSB ptychography



See also Dwyer and Paginin, *arXiv.*

$$DQE_{SSB}(\mathbf{Q_p}) = \frac{SNR_{SSB}(\mathbf{Q_p})^2}{SNR_{Z,\mathrm{ideal}}(\mathbf{Q_p})^2} = \frac{2PCTF(\mathbf{Q_p})N_xN_yDN_\alpha}{4N_xN_yDN_\alpha} = \frac{PCTF(\mathbf{Q_p})}{2}$$

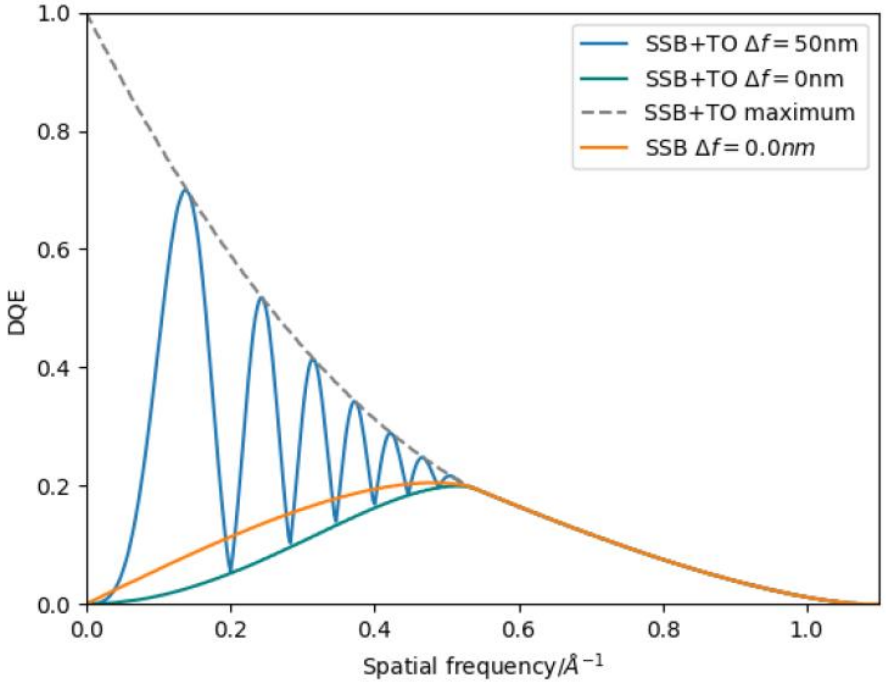
4D STEM

$$DQE_Z(\mathbf{Q_p}) = \frac{SNR_Z(\mathbf{Q_p})^2}{SNR_{Z,\mathrm{ideal}}(\mathbf{Q_p})^2} = \frac{4|A(\mathbf{Q_p})|^2|E(\mathbf{Q_p})|^2|\sin\chi(\mathbf{Q_p})|^2N_xN_yDN_\alpha}{4N_xN_yDN_\alpha}$$
$$= |A(\mathbf{Q_p})|^2|E(\mathbf{Q_p})|^2|\sin\chi(\mathbf{Q_p})|^2$$

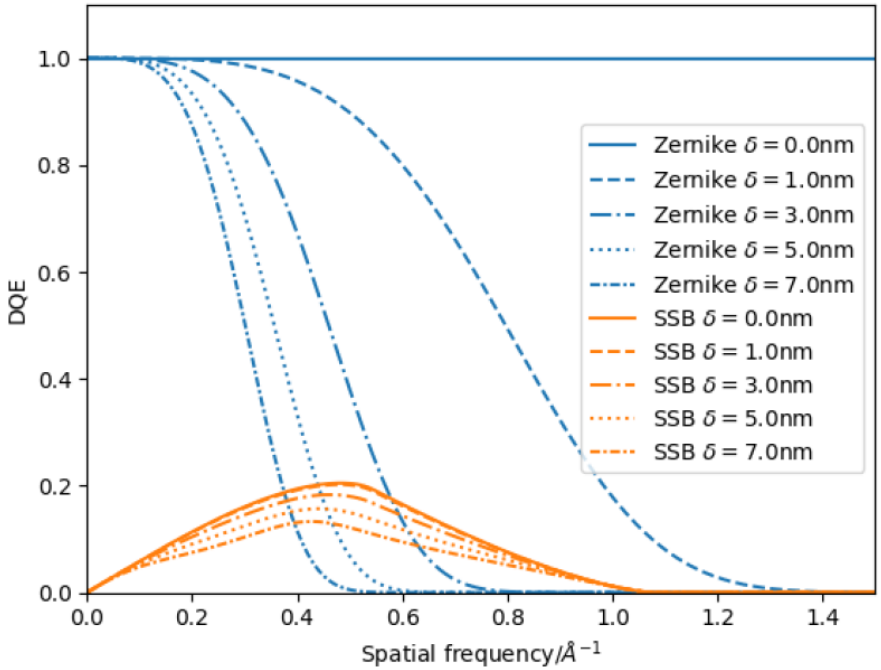
Zernike

Bennemann et al. *in preparation*

DQE of in-focus and defocused ptychography



Perfect coherence



Partial coherence

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