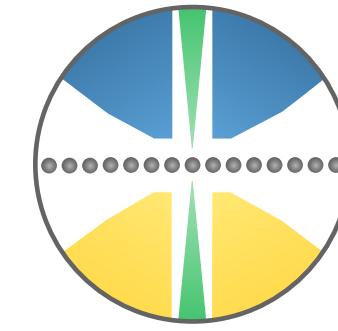




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# Open-Source 4D-STEM Phase Reconstructions of Focused-Probe Data with Near-Ideal Direct-Electron Detection

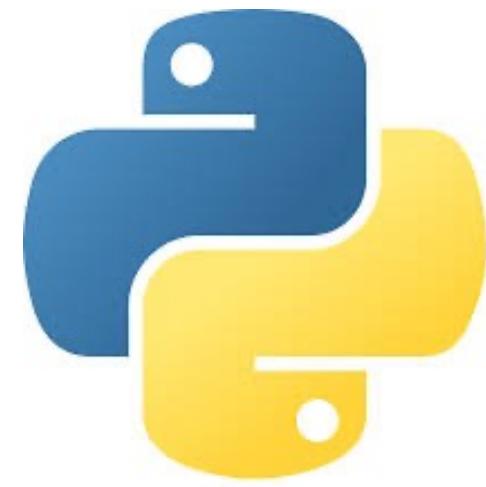
**Toma Susi**, Jani Kotakoski, Clemens Mangler (U. Vienna)  
Niklas Dellby, Russ Hayner, Tracy Lovejoy, Andreas Mittelberger, Benjamin Plotkin-Swing (Nion Co. R&D / Bruker AXS)

Christoph Hofer, Timothy Pennycook (U. Antwerp)



# Open-source software

## Language



Python  
(3.11.9)

## Data Acquisition

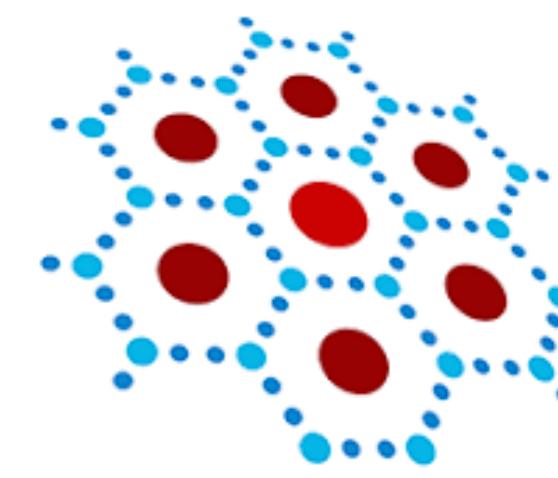


Nion Swift  
(0.16.10)

## Phase Reconstructions



PyPtychoSTEM  
(20d3be1f)



py4DSTEM  
(0.14.16)



Stephanie  
Ribet



Georgios  
Varnavides

## Reconstruction Algorithms:

single-sideband (**SSB**), Wigner distribution deconvolution (**WDD**),  
(iterative) differential phase contrast (**DPC**),  
parallax imaging ie. tilt-corrected bright-field STEM (**parallax**),  
iterative gradient-descent ptychography (**GD**)

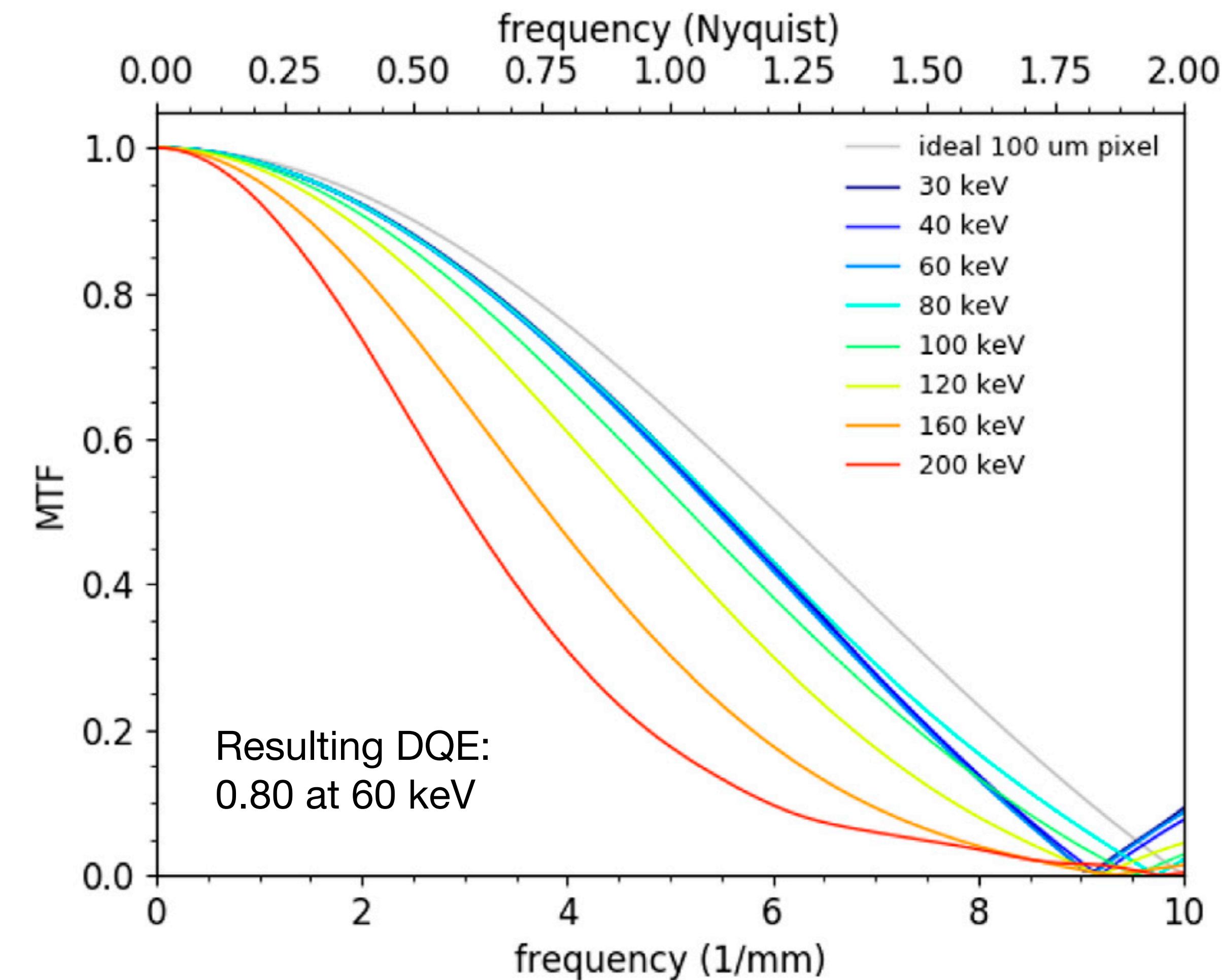
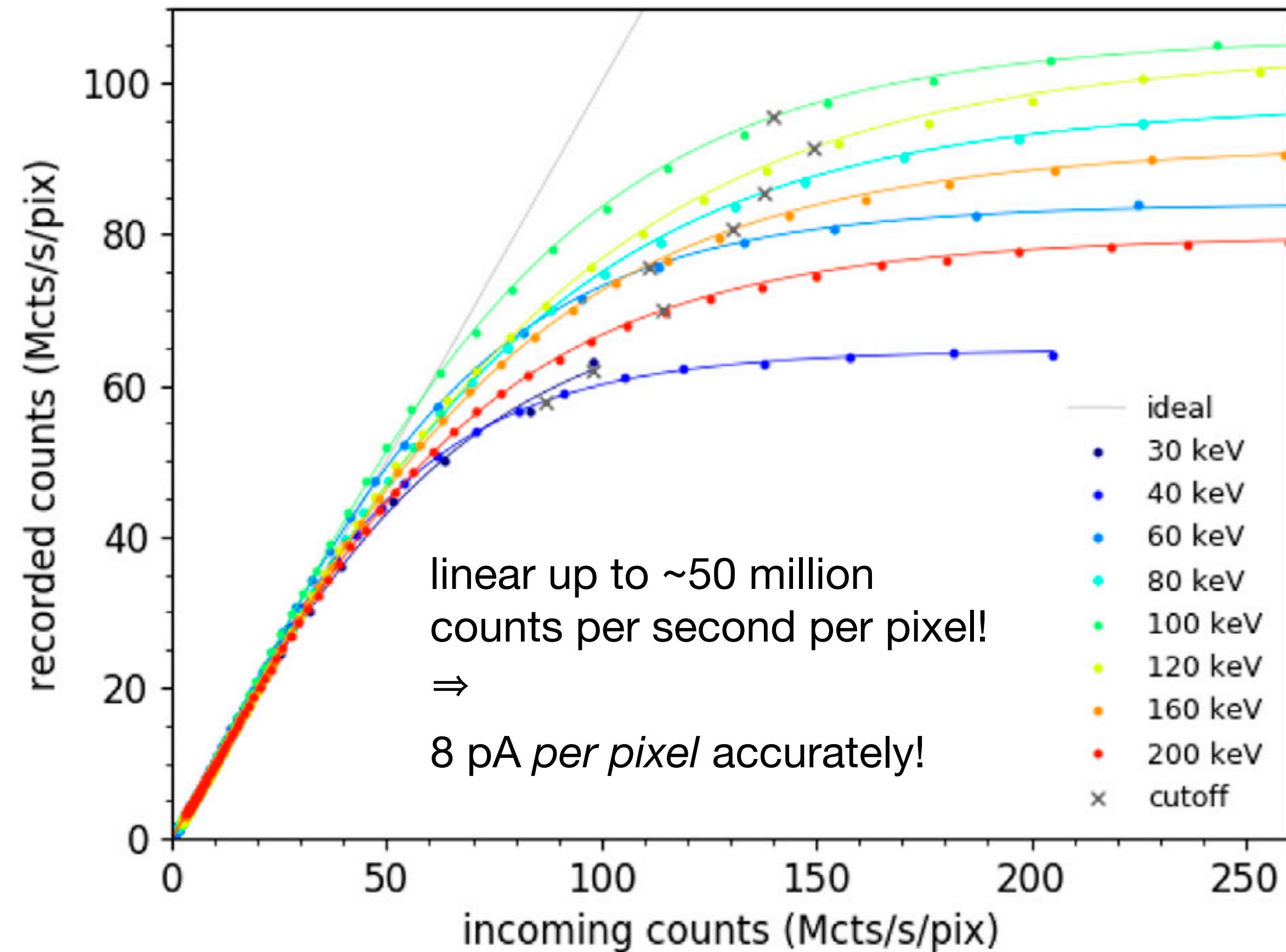
# Near-ideal direct-electron 4D-STEM

<b>Number of pixels (W x H)</b>	<b>192 × 192</b>
<b>Active area (W x H) [mm<sup>2</sup>]</b>	<b>20 × 20</b>
<b>Pixel size (W x H) [μm<sup>2</sup>]</b>	<b>100 × 100</b>
<b>Sensor material</b>	<b>Silicon (Si) or high-Z</b>
<b>Energy range [keV]</b>	<b>30–300</b>
<b>Frame rate (max.) [Hz]</b>	<b>120,000</b>
<b>Count rate (max.) [el/s/pixel]</b>	<b>10<sup>8</sup></b>
<b>Detective Quantum Efficiency, DQE(0)</b>	<b>0.82 @ 80 keV 0.75 @ 200 keV, 0.75 @ 300 keV</b>
<b>Detector mounting</b>	<b>Retractable</b>



Near-ideal 4D-STEM performance at 60 keV

**Installation of production unit #001  
in Vienna at the start of 2024**



# 4D-STEM data volumes

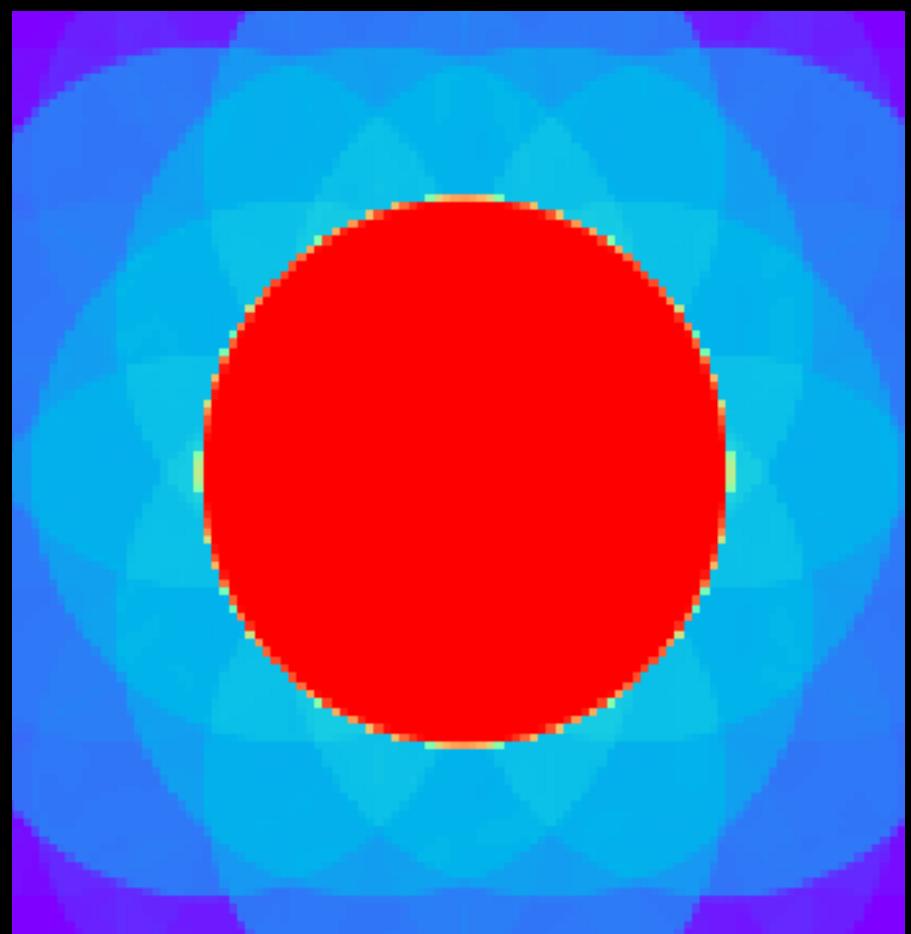
Data volumes collected during typical 4D-STEM experiments (assuming a  $512 \times 512$  px scan).

Mode	Pixels	Bit depth	Bits / CBED	Frame rate	MB / s	GB/scan
Saved file (hdf5)	192×192	12	442368	20000	1106	
Numpy (fp16)	192×192	16	589824	-	-	39
Reduced file (hdf5)	96×96	8	73728	120000	1106	
Numpy (fp16)	96×96	16	147456	-	-	9.7

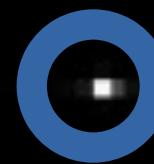
Huge data volumes can be collected in seconds (challenge for storage, processing...)

# Ptychography can correct aberrations post-acquisition

PACBED

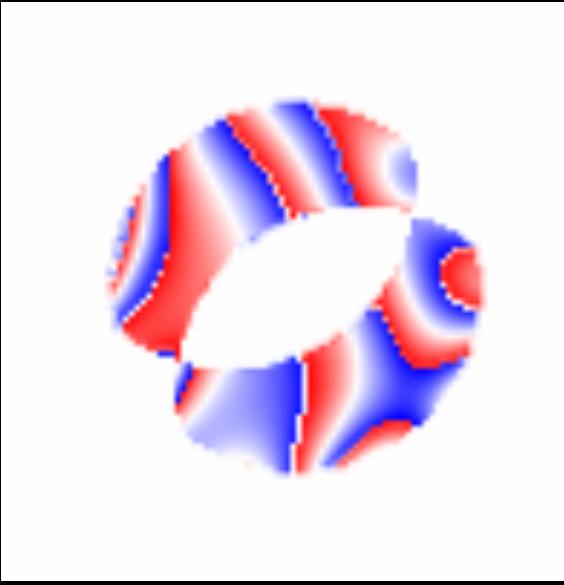


Spatial frequencies

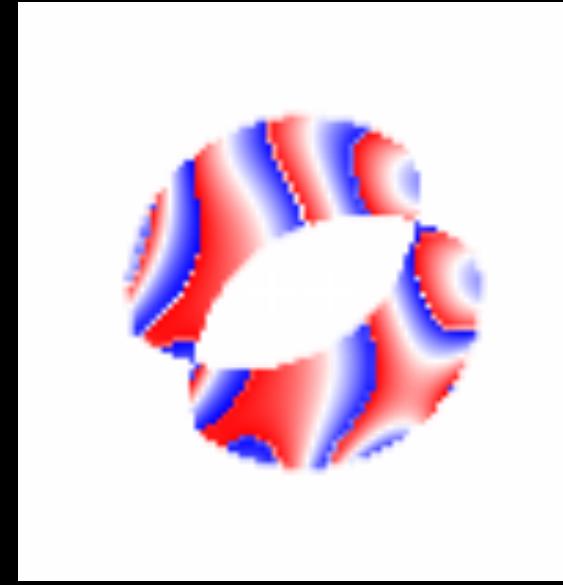


$A(K)^* A(K-Q)$

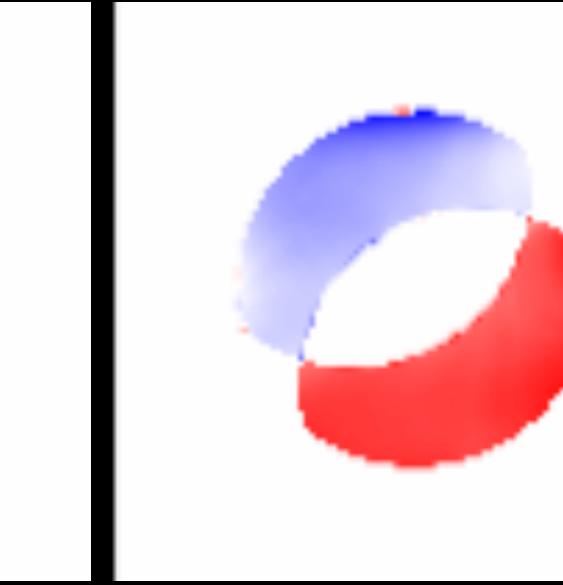
'Exp'



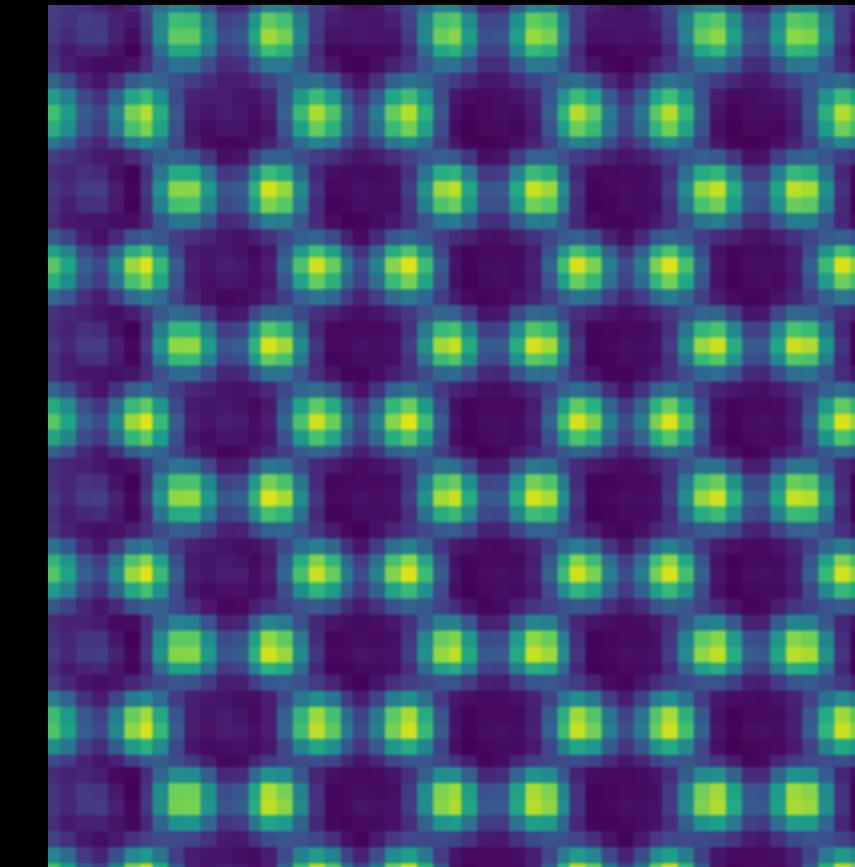
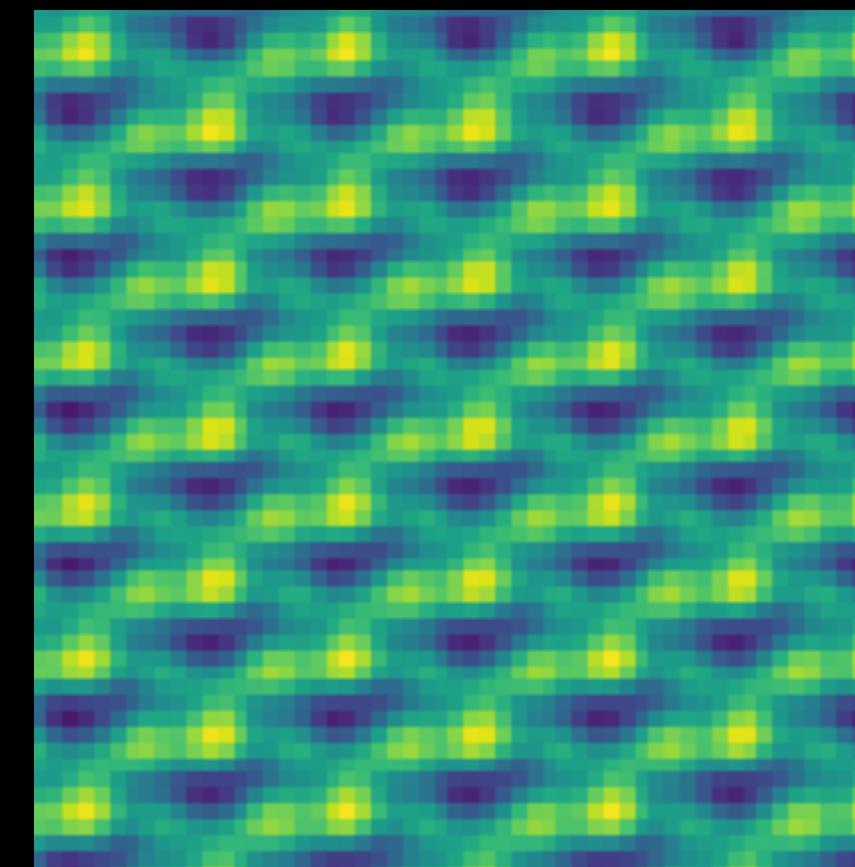
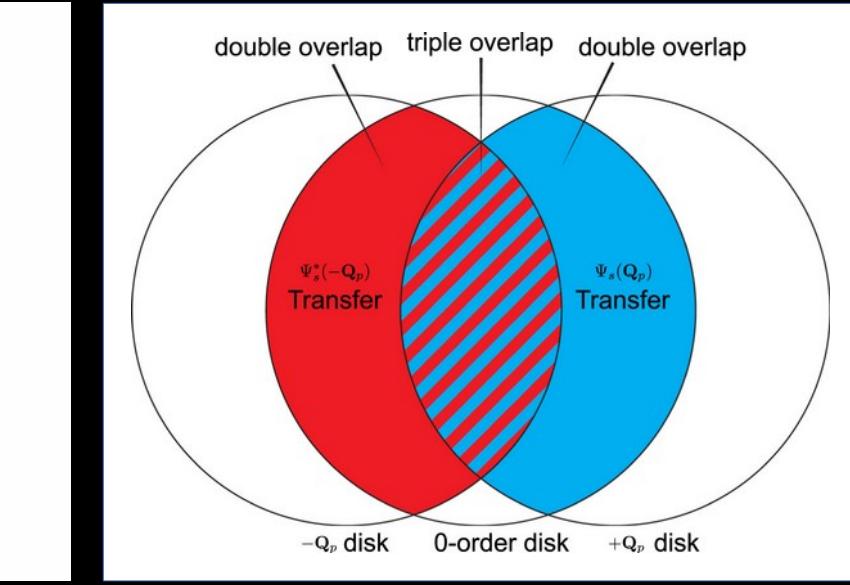
SVD



corrected



Theory



SVD = singular value decomposition



Timothy  
Pennycook

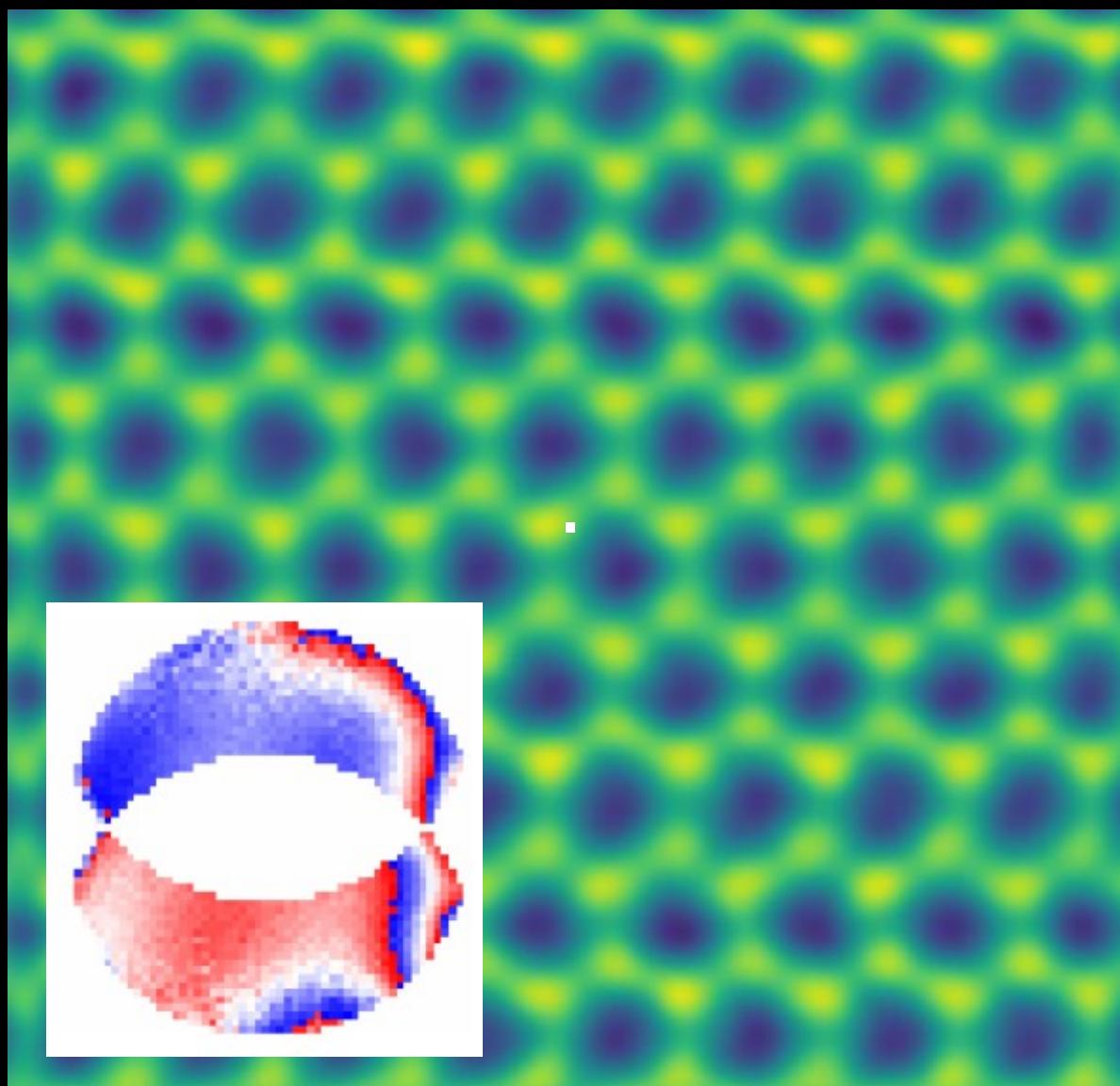


Christoph  
Hofer

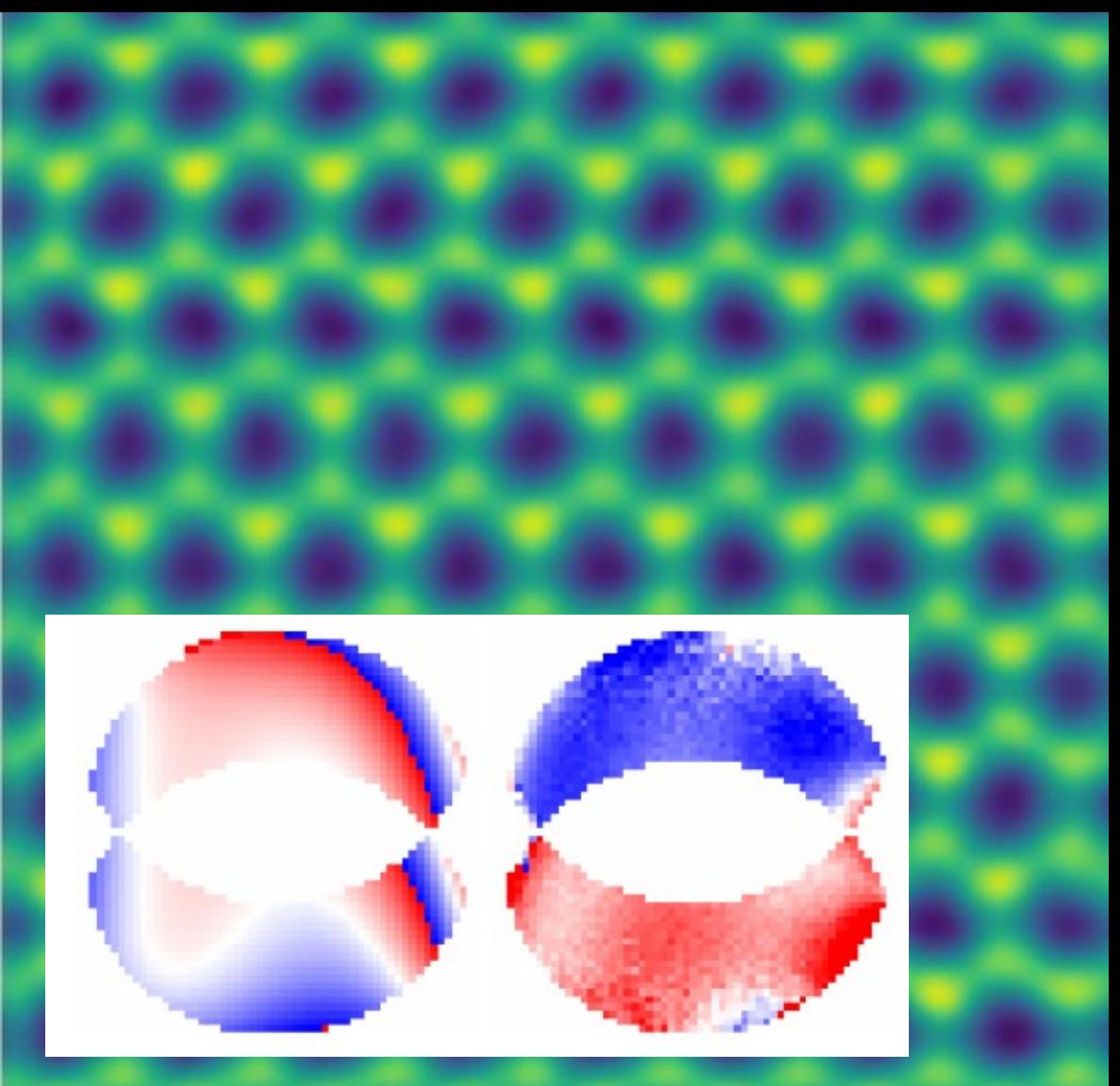
# 3<sup>rd</sup> order aberration correction might not be sufficient

Experimental data of monolayer graphene, 35 mrad (Vienna)

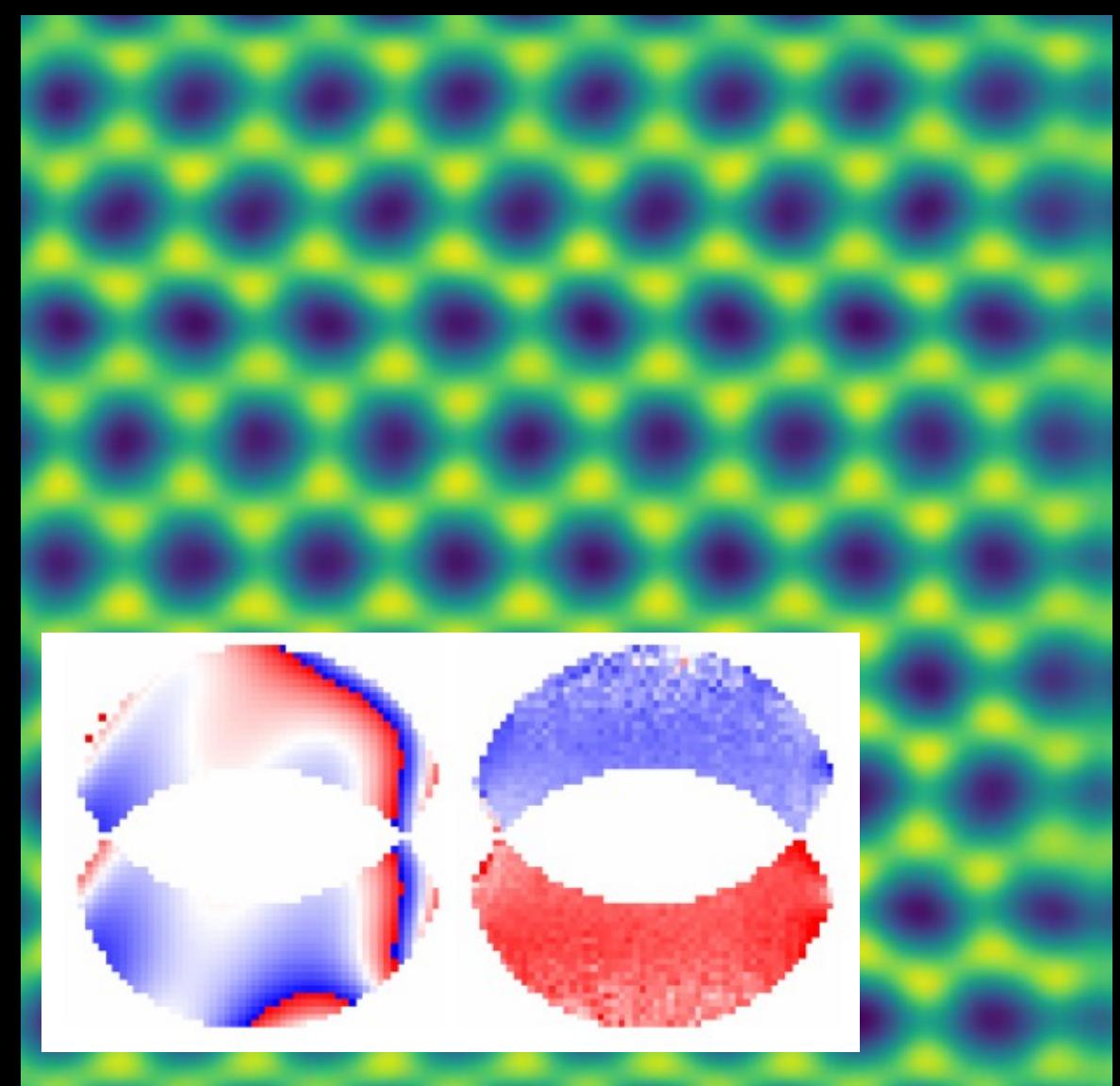
uncorrected



3<sup>rd</sup> order corrected



5<sup>th</sup> order corrected

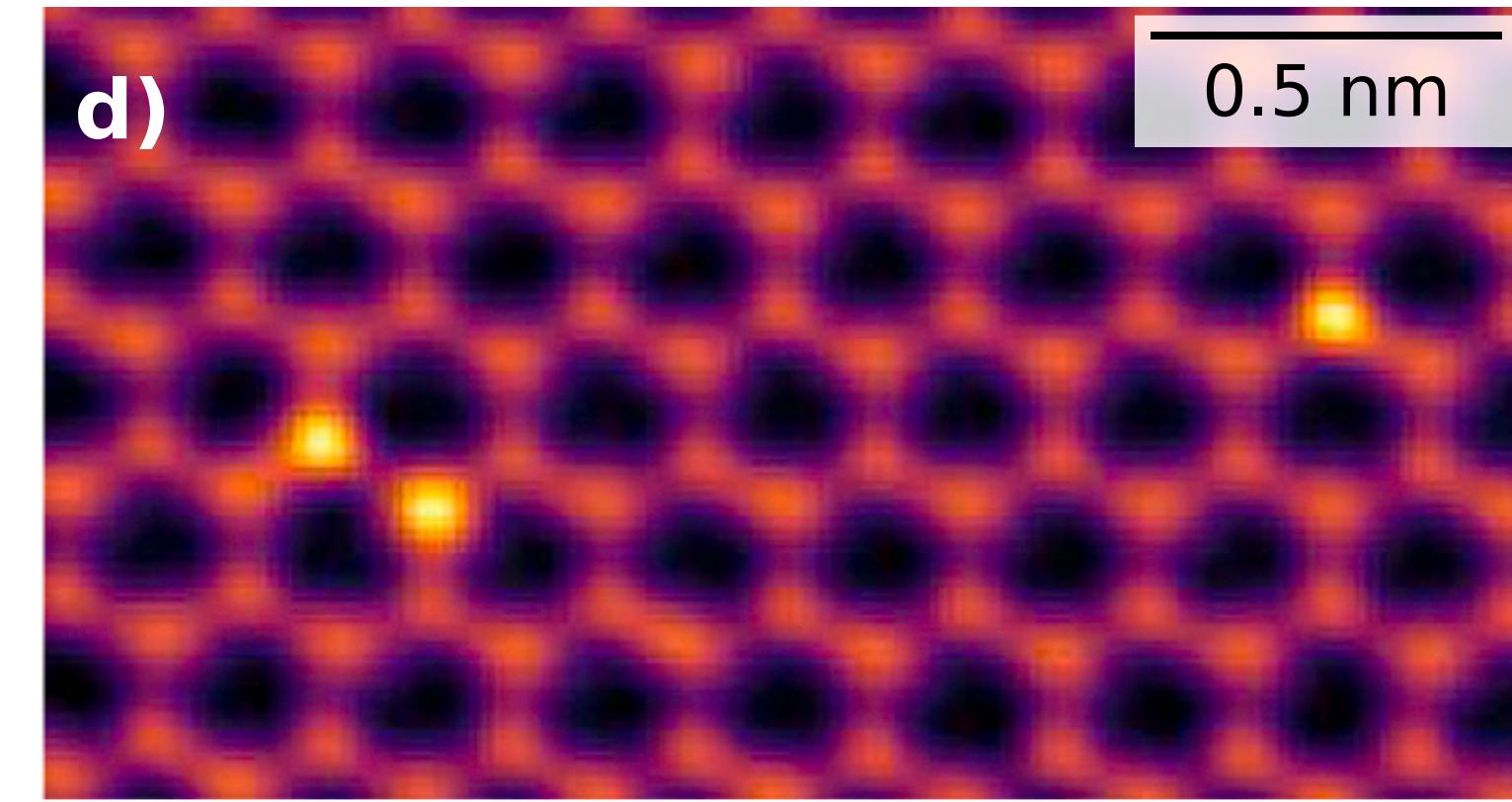
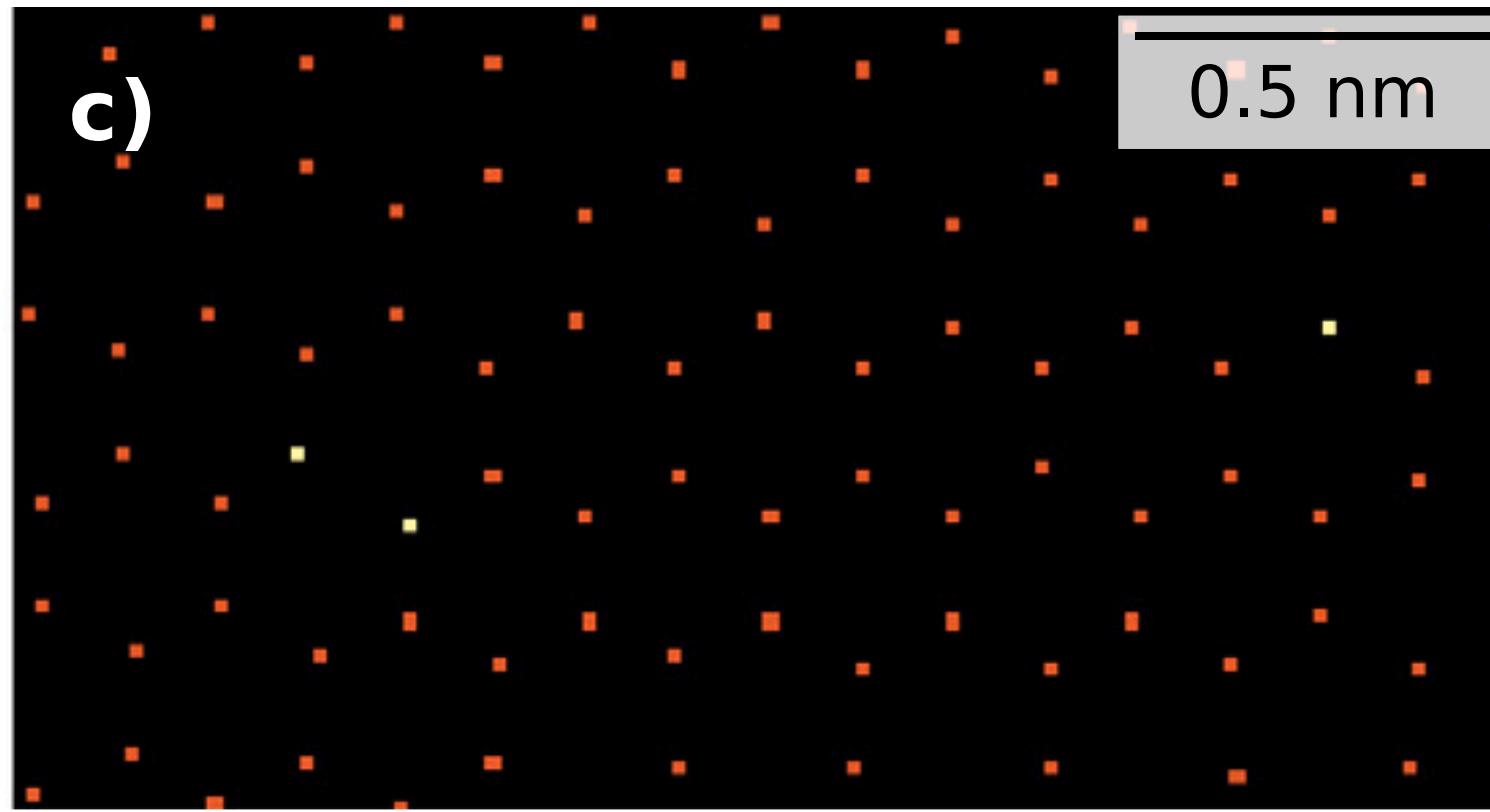
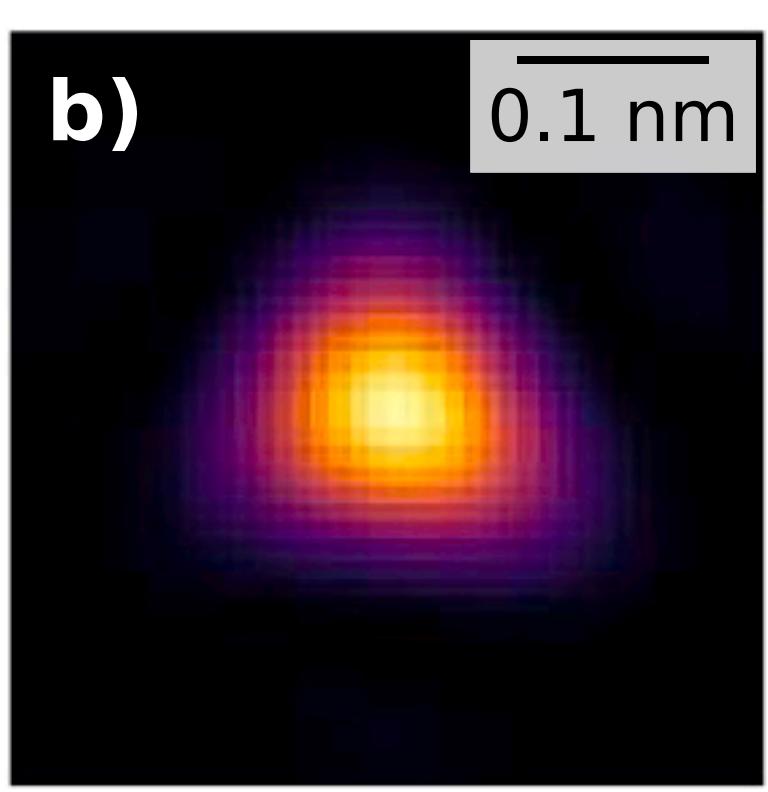


Timothy  
Pennycook



Christoph  
Hofer

# Site-specific phase via iterative optimization



Maximize correlation between experimental and simulated image

$$R = \frac{\sum_{i=1}^N \left( (\mu^{sim} - I_i^{sim}) (\mu^{exp} - I_i^{exp}) \right)}{\sigma^{sim} \sigma^{exp} (N - 1)}$$

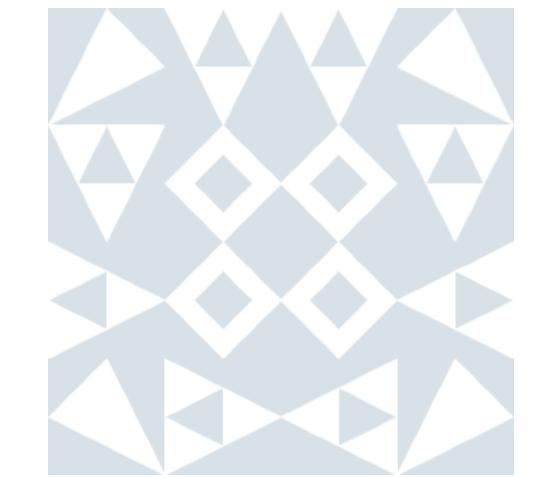
$I^{exp}$  and  $I^{sim}$  = image intensities

$\mu^{exp}$  and  $\mu^{sim}$  = mean values

$\sigma^{sim}$  and  $\sigma^{exp}$  = standard deviations

- Can account for:
- aberrations
  - incoherence
  - scan distortions
  - sample drift
  - sample tilt
  - etc...

Phase analysis

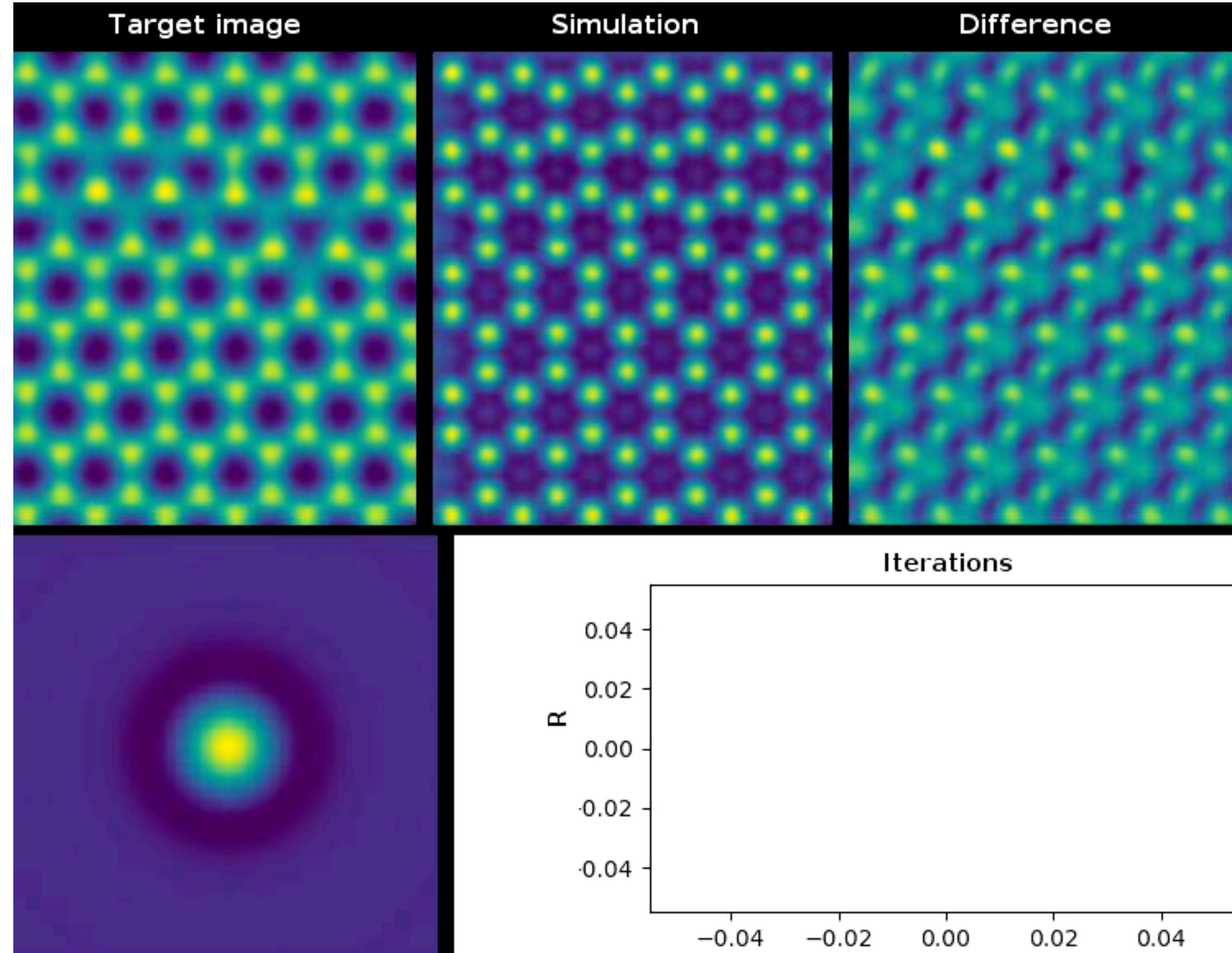


STEM\_Optimization\*  
(commit 6c549ba9)

## Defective $WS_2$

Can account for:

- aberrations
- incoherence
- scan distortions
- sample drift
- sample tilt
- etc...

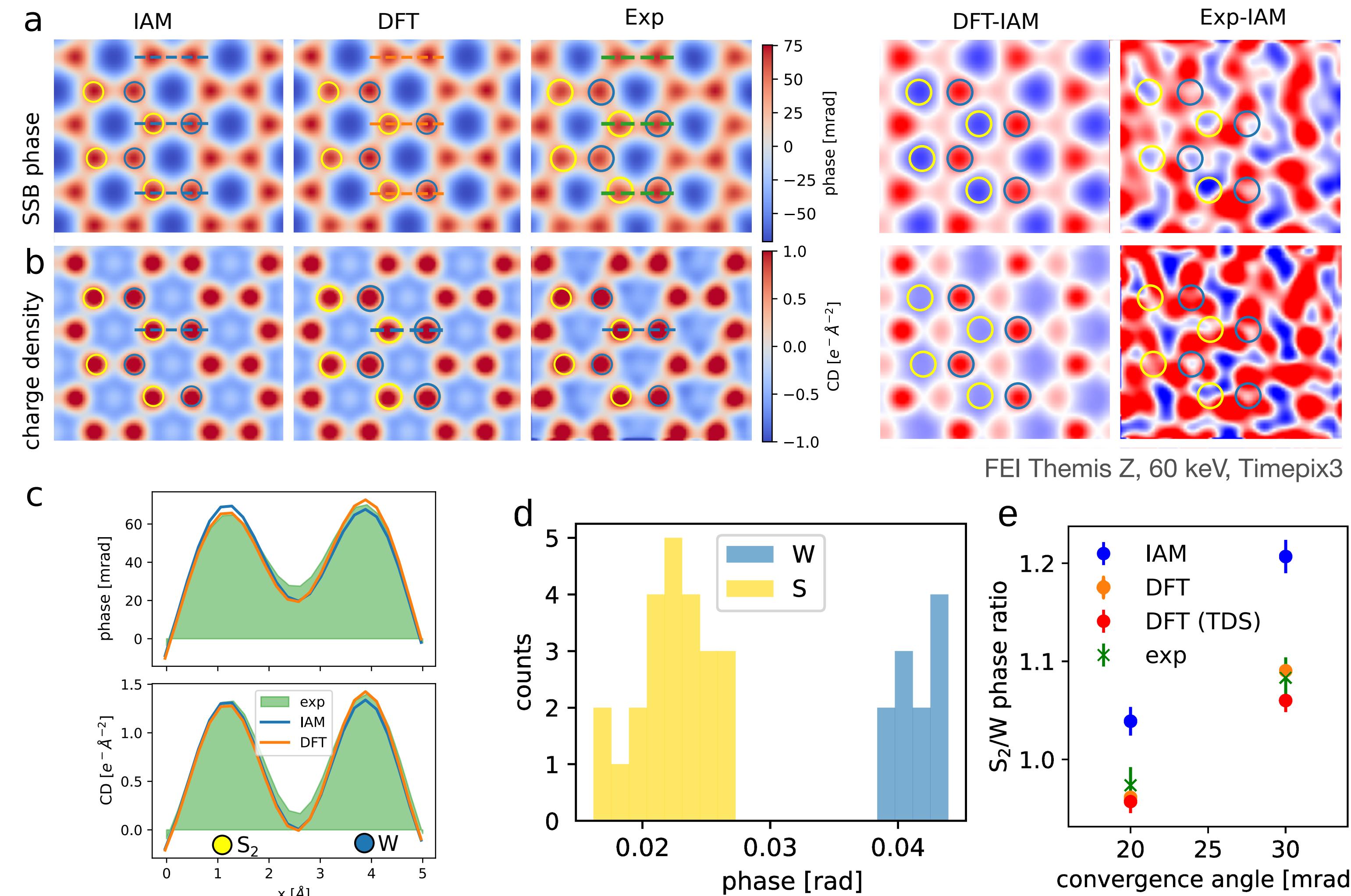
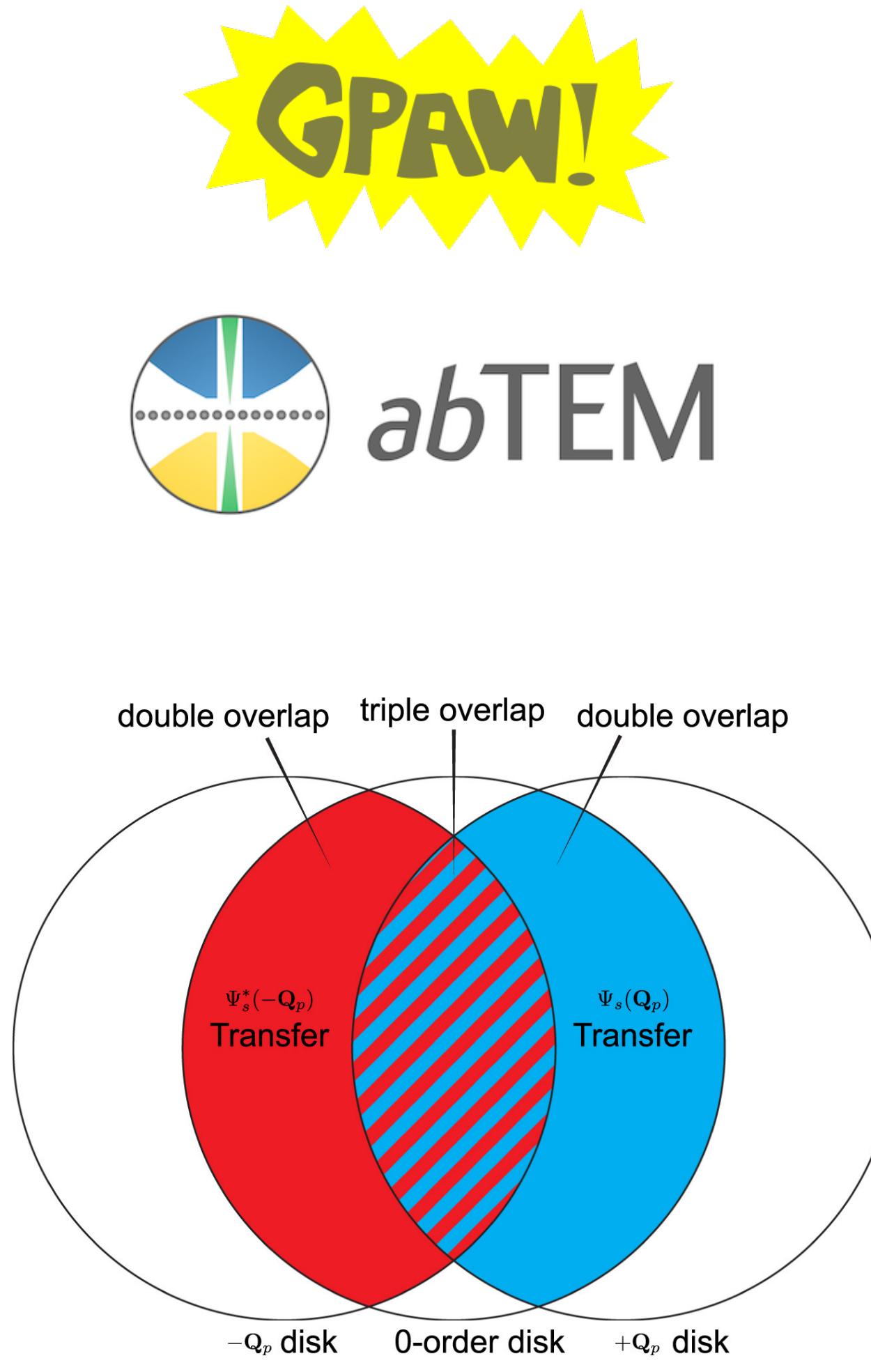


Timothy  
Pennycook



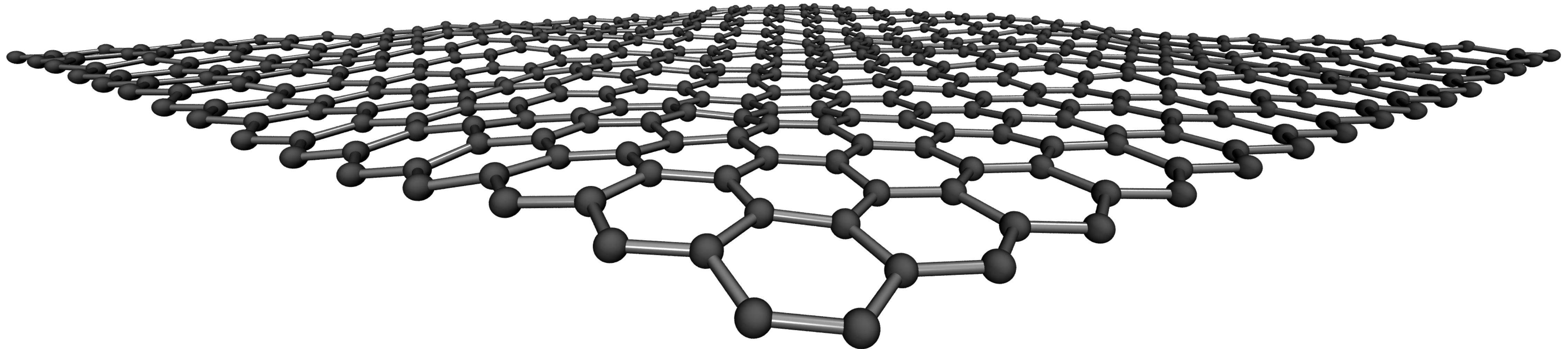
Christoph  
Hofer

# Ptychography for charge transfer in defective WS<sub>2</sub>

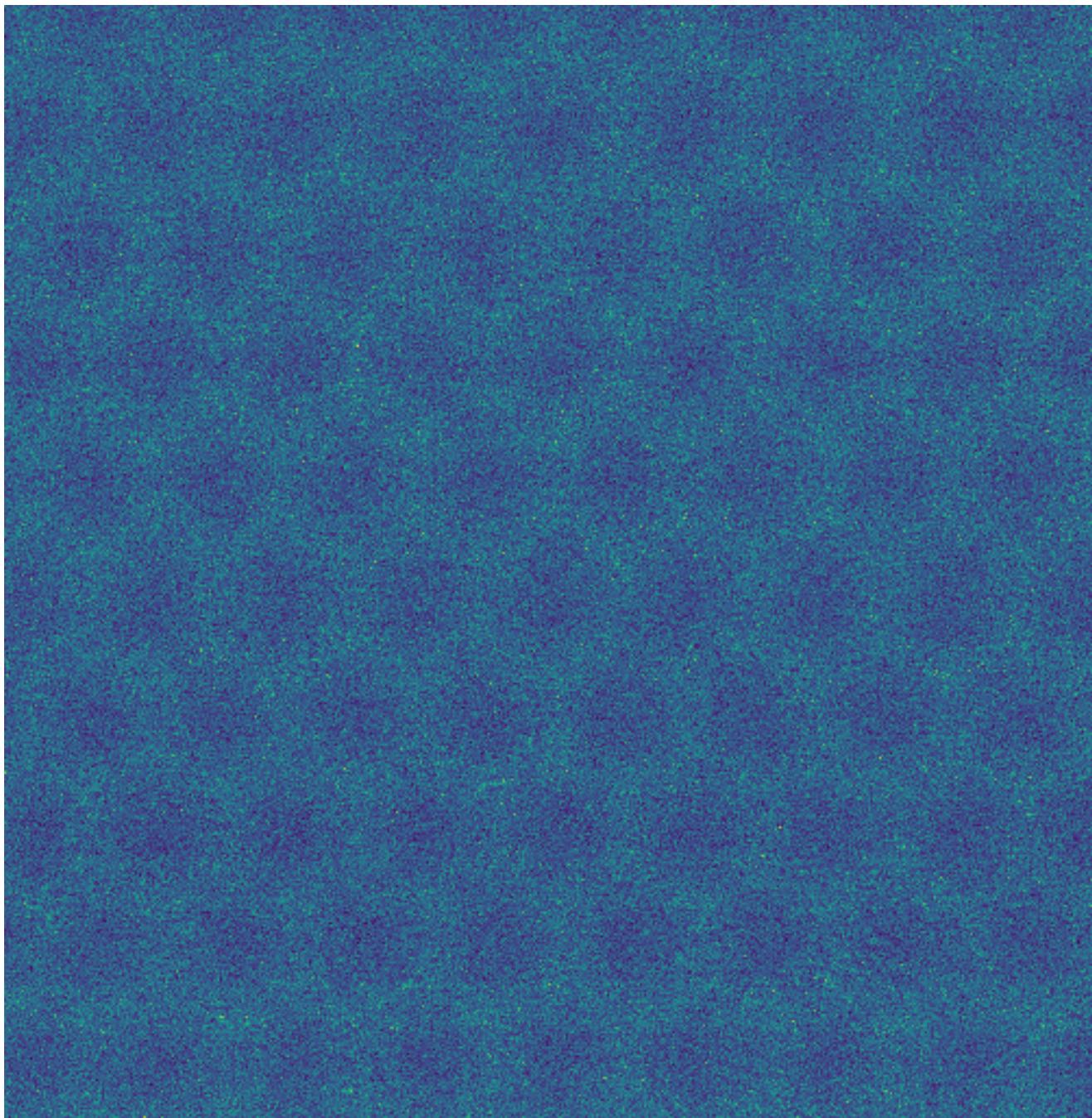


# Graphene: ideal uniform phase object

- ✓  $\infty$  radiation hardness  $<80$  keV  $\Rightarrow$  no dose limitation
- ✓ low- $Z$ , one-atom-thick  $\Rightarrow$  perfect weak phase object
- ✓ only one element  $\Rightarrow$  each atomic site identical



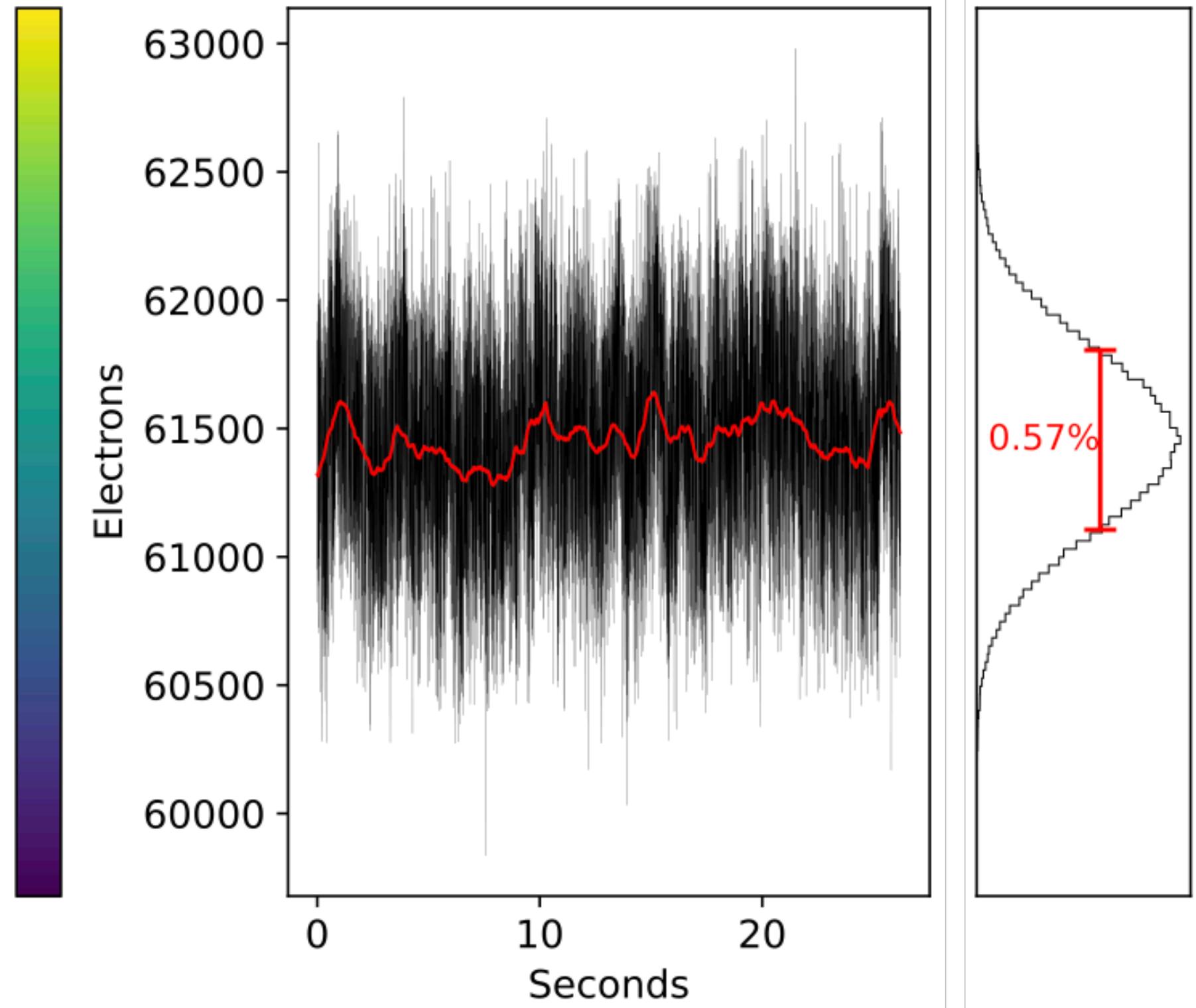
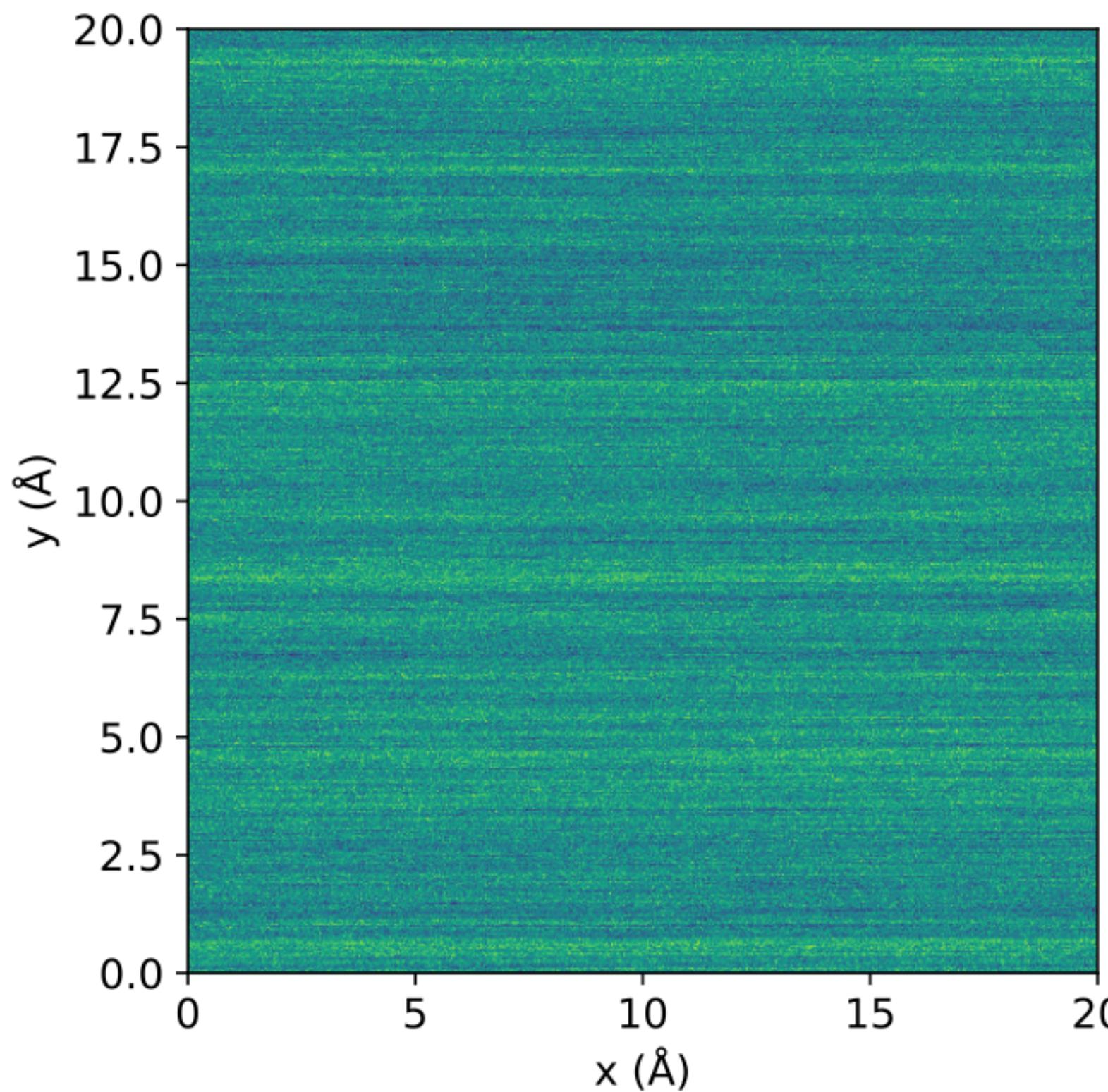
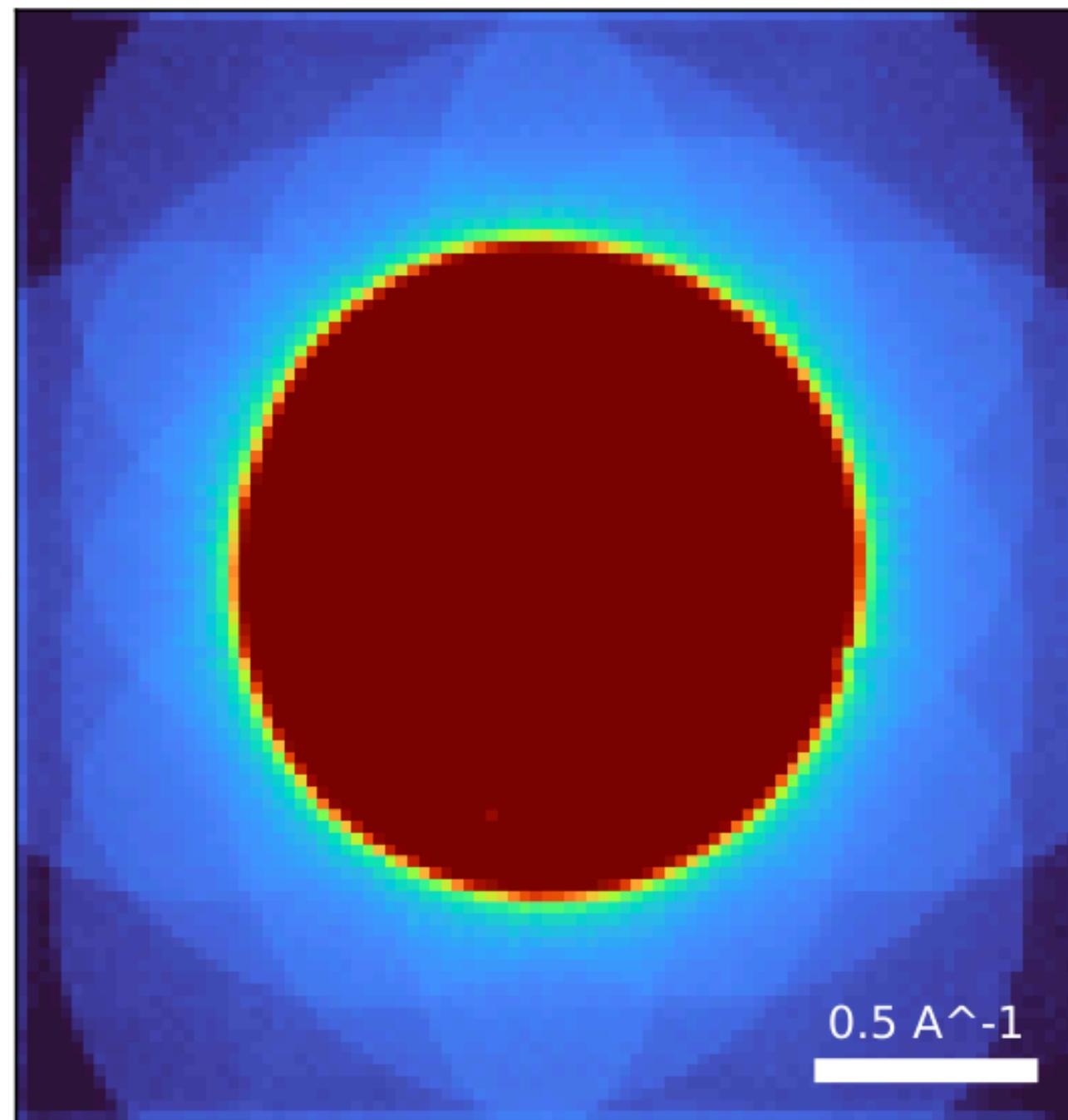
# Nion + ARINA 4D-STEM data collection (original)



HAADF (80–300 mrad)

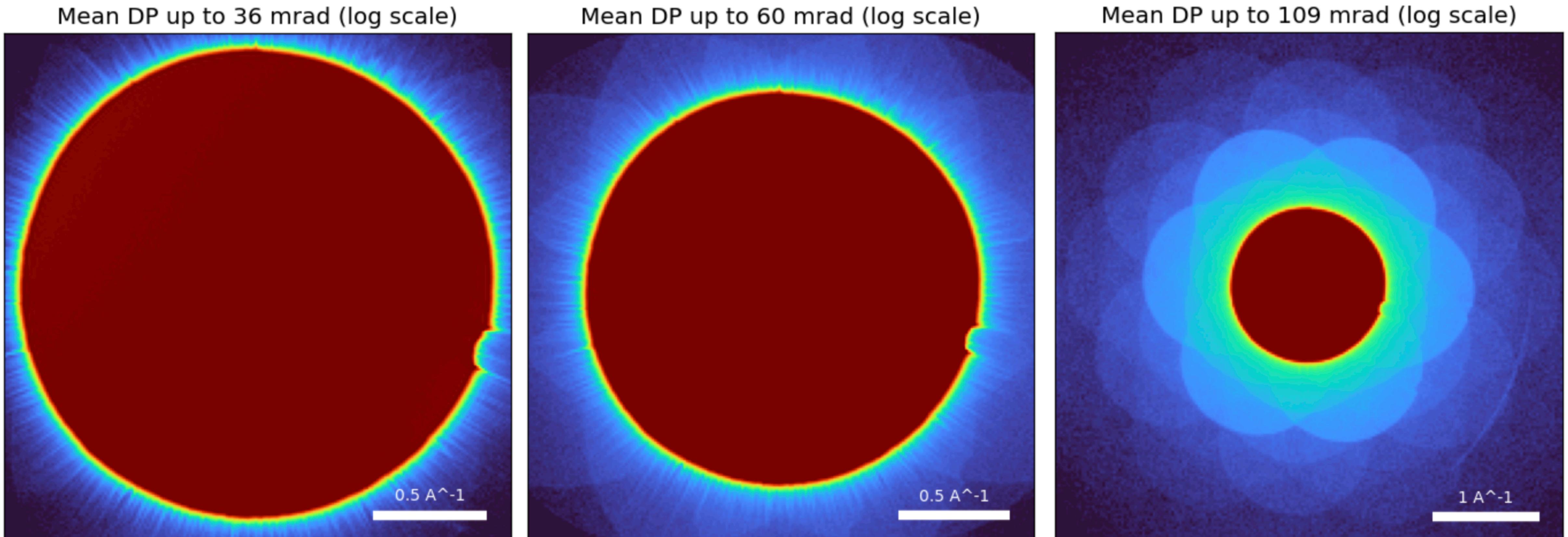
# Nion + ARINA 4D-STEM data collection (original)

512 × 512 px scan (**Rpix**), 96 × 96 px CBED (**Qpix**; binned by 4 for analysis), 100 us dwell time



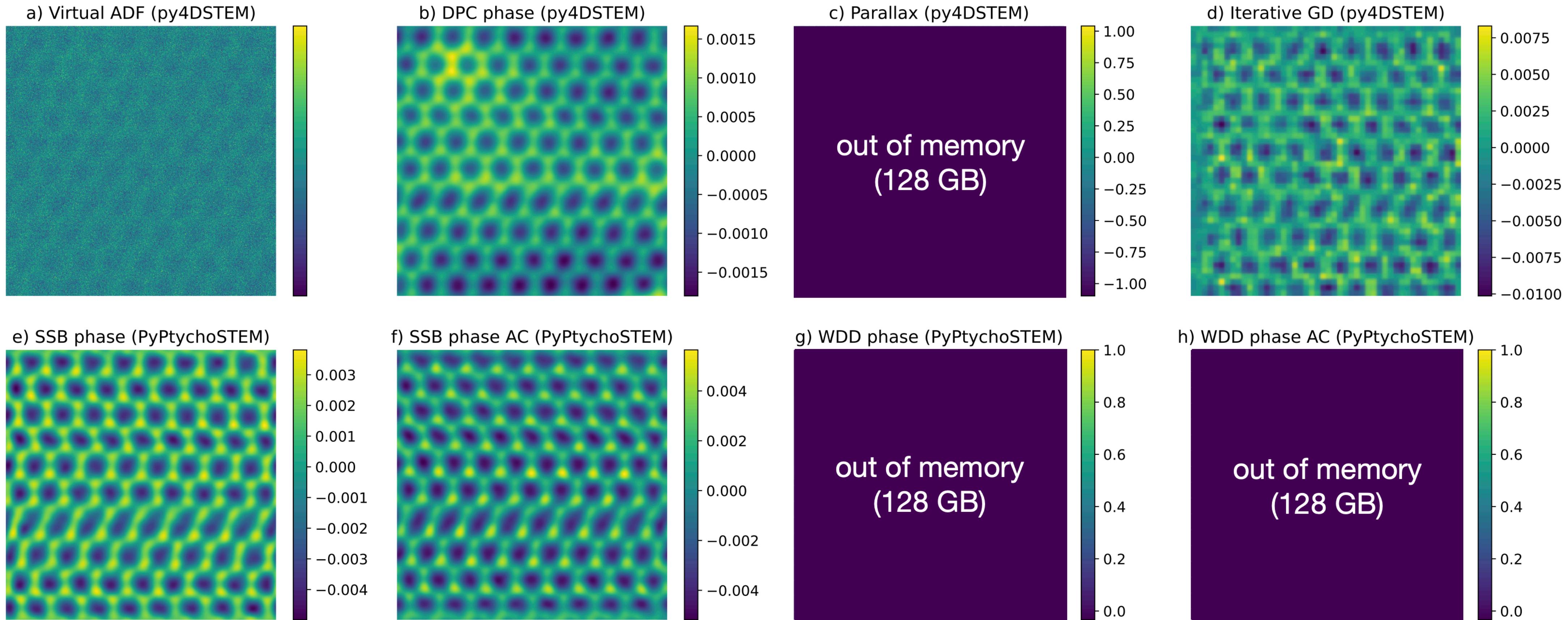
- 34 mrad semi-convergence angle, ~70 mrad max collection angle, ~8 pA
- counting every electron, corrected dose per pixel can be normalized

# Nion + ARINA 4D-STEM data collection (final)



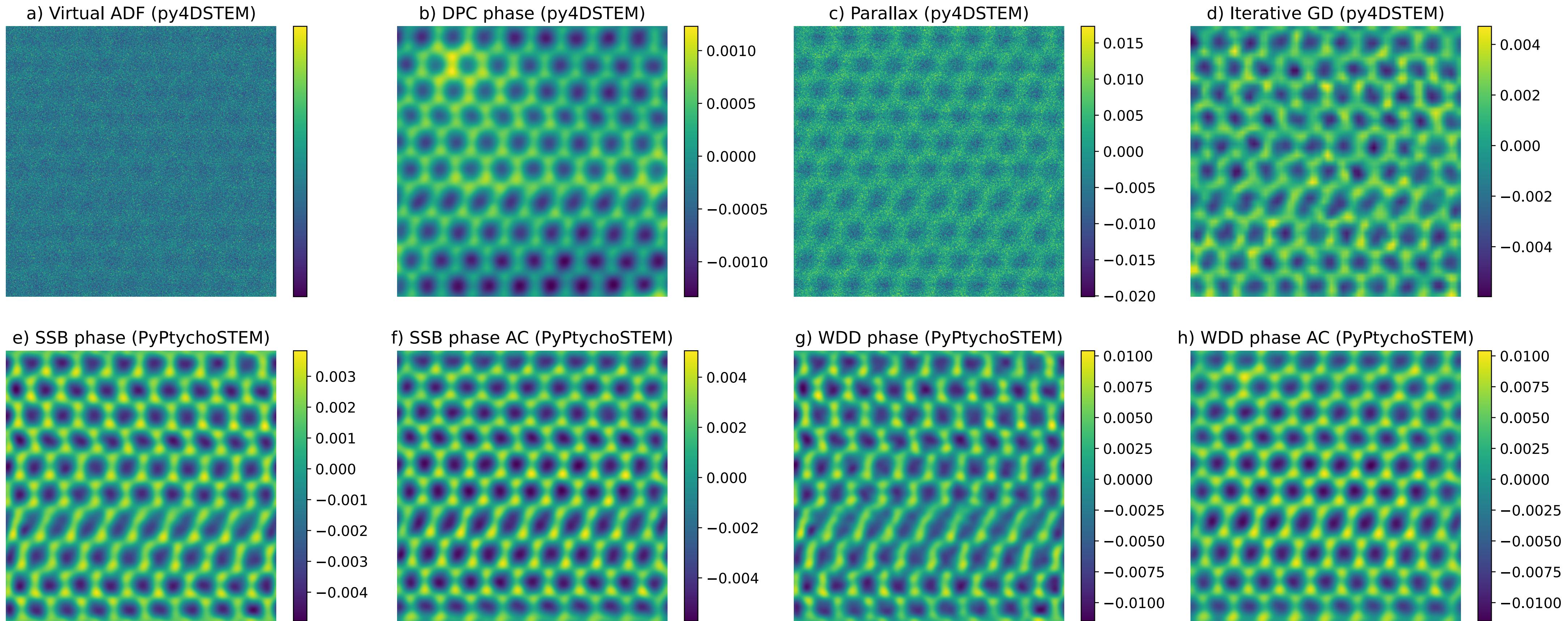
$\sim 200 \text{ pA current!}$

# Comparing phase reconstruction algorithms (Q binning)



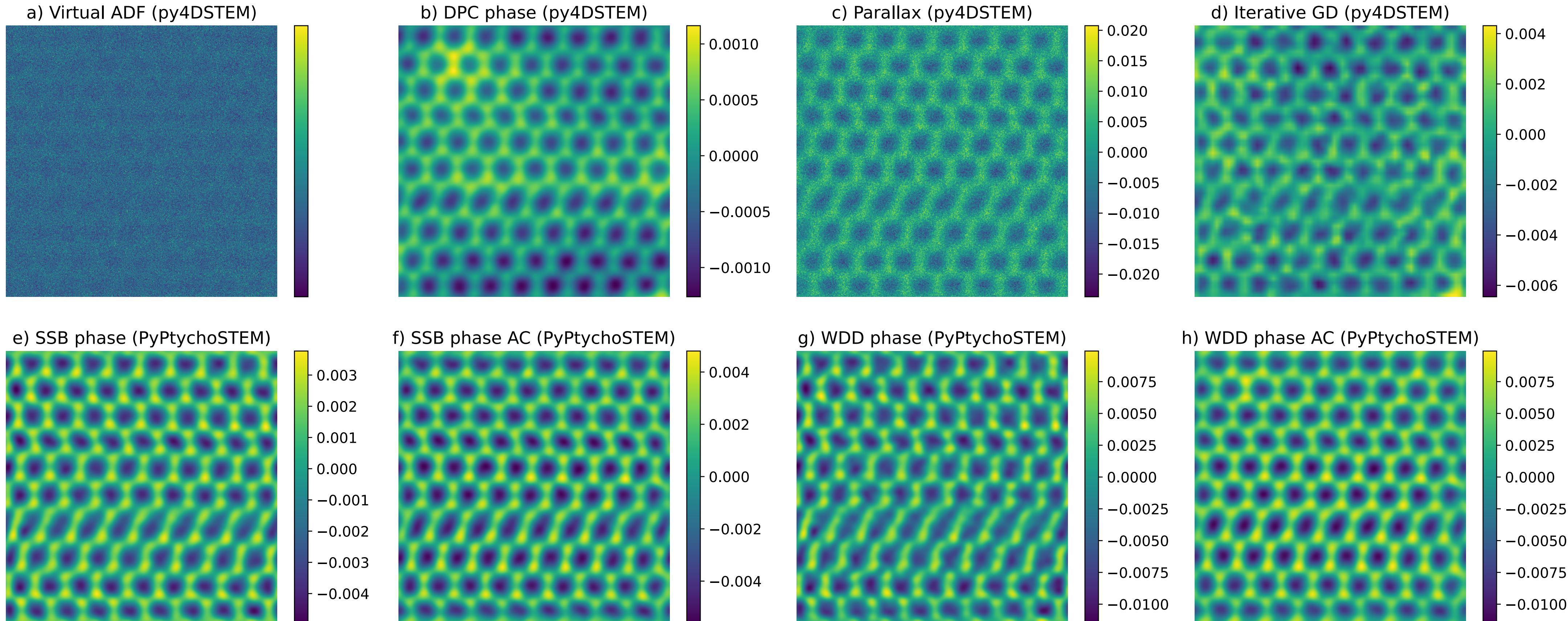
36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 192×192 Qpix  
1× binning in Q

# Comparing phase reconstruction algorithms (Q binning)



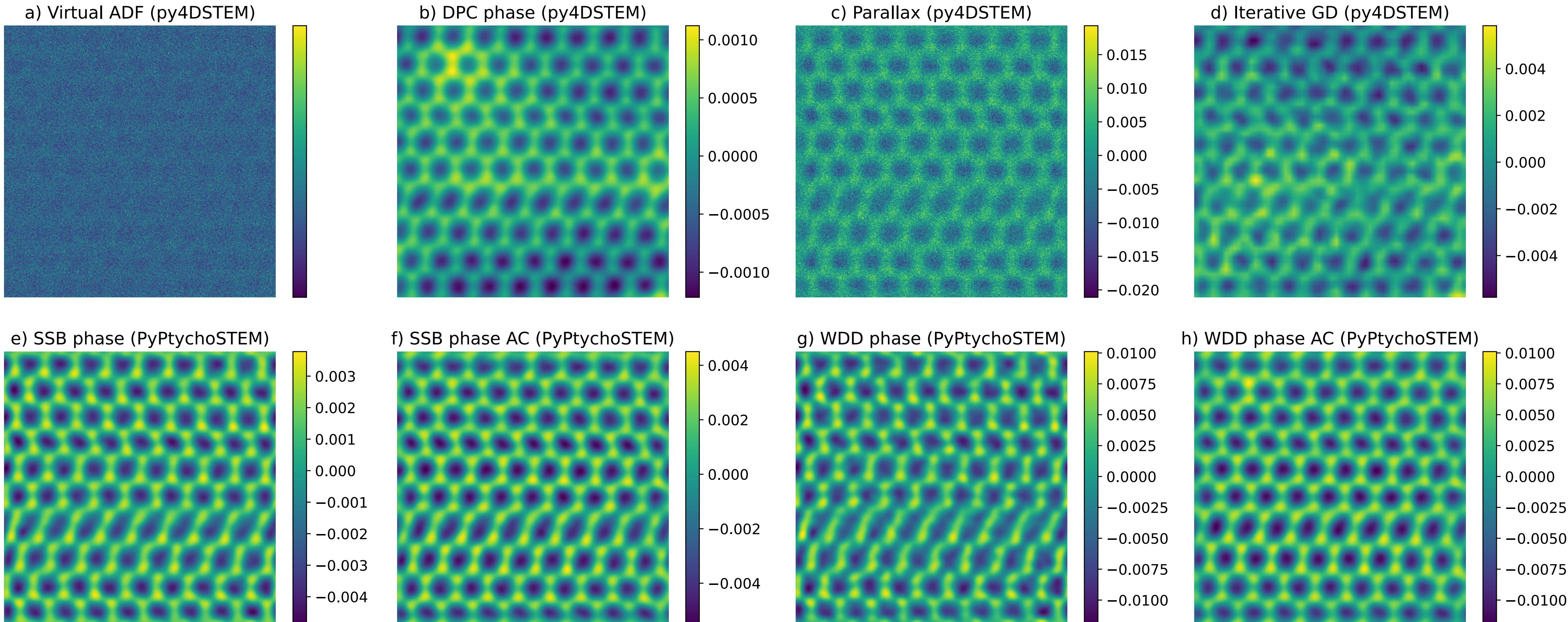
36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 192×192 Qpix  
2× binning in Q

# Comparing phase reconstruction algorithms (Q binning)



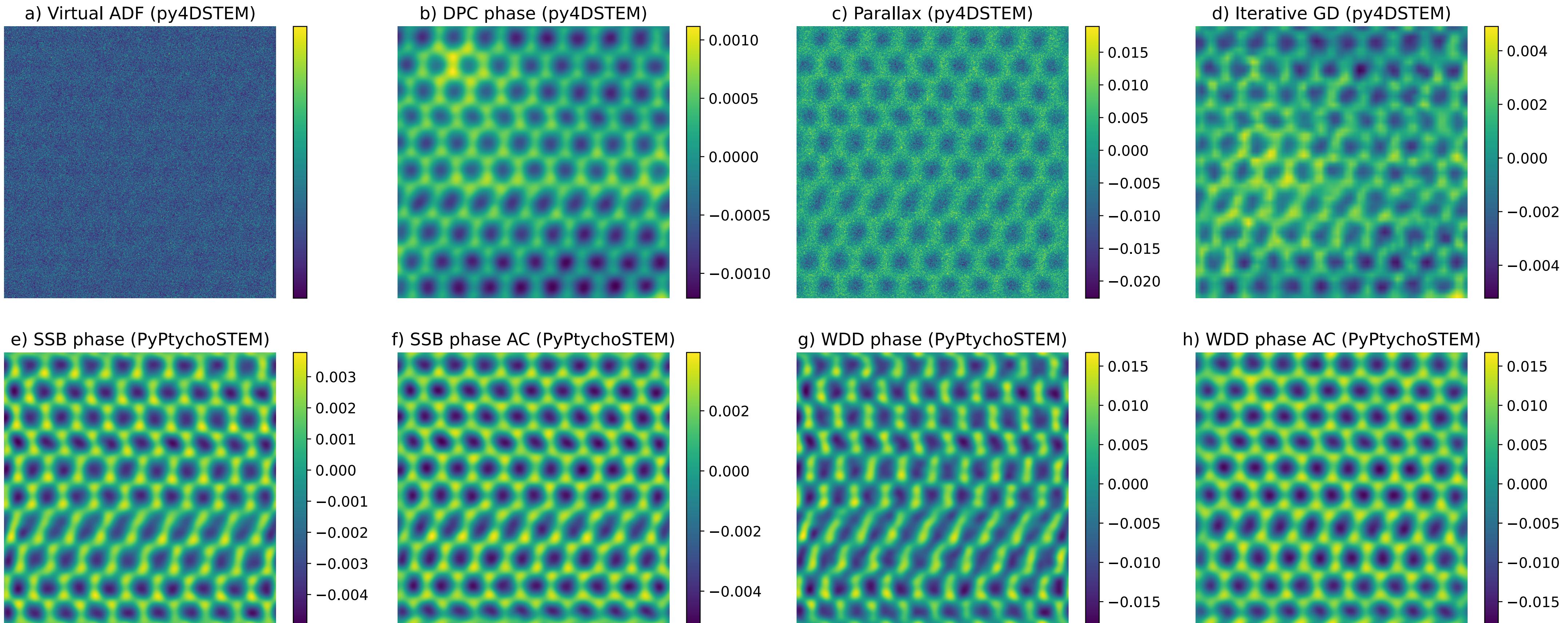
36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 192×192 Qpix  
4× binning in Q

# Comparing phase reconstruction algorithms (Q binning)



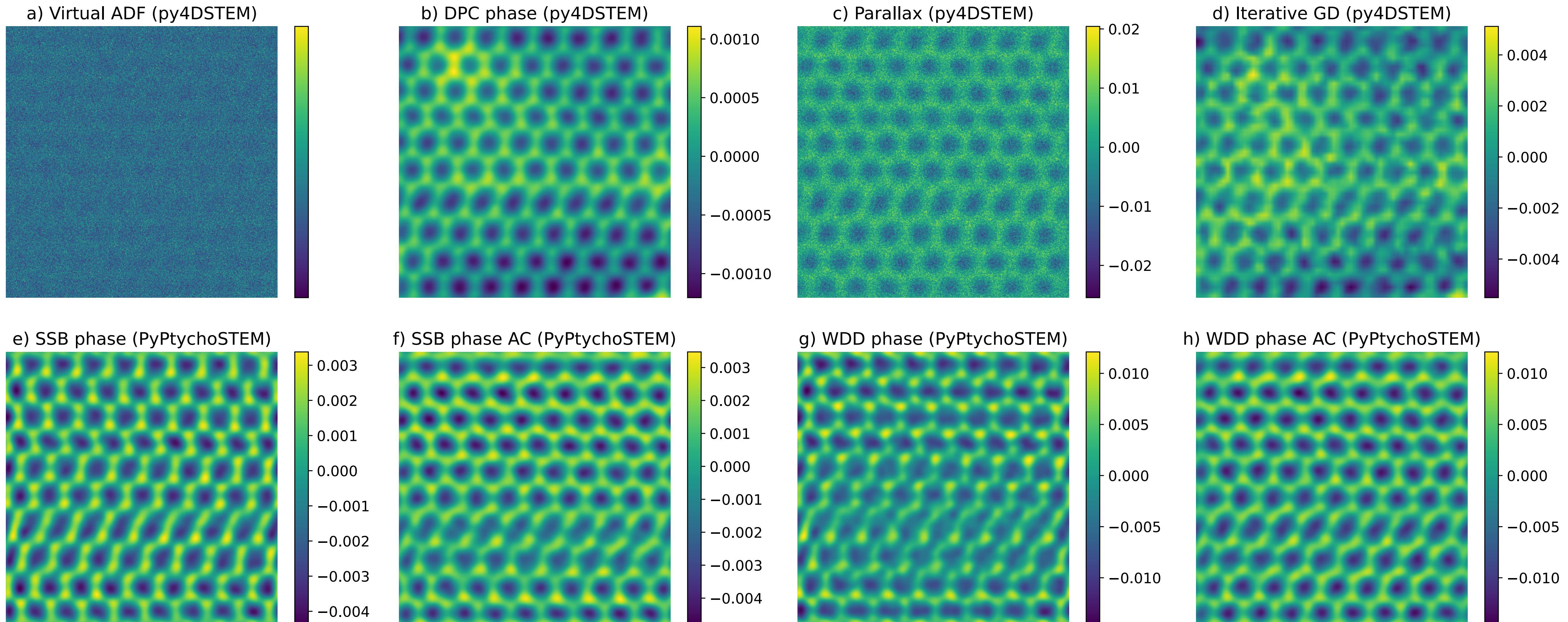
36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 192×192 Qpix  
8× binning in Q

# Comparing phase reconstruction algorithms (Q binning)



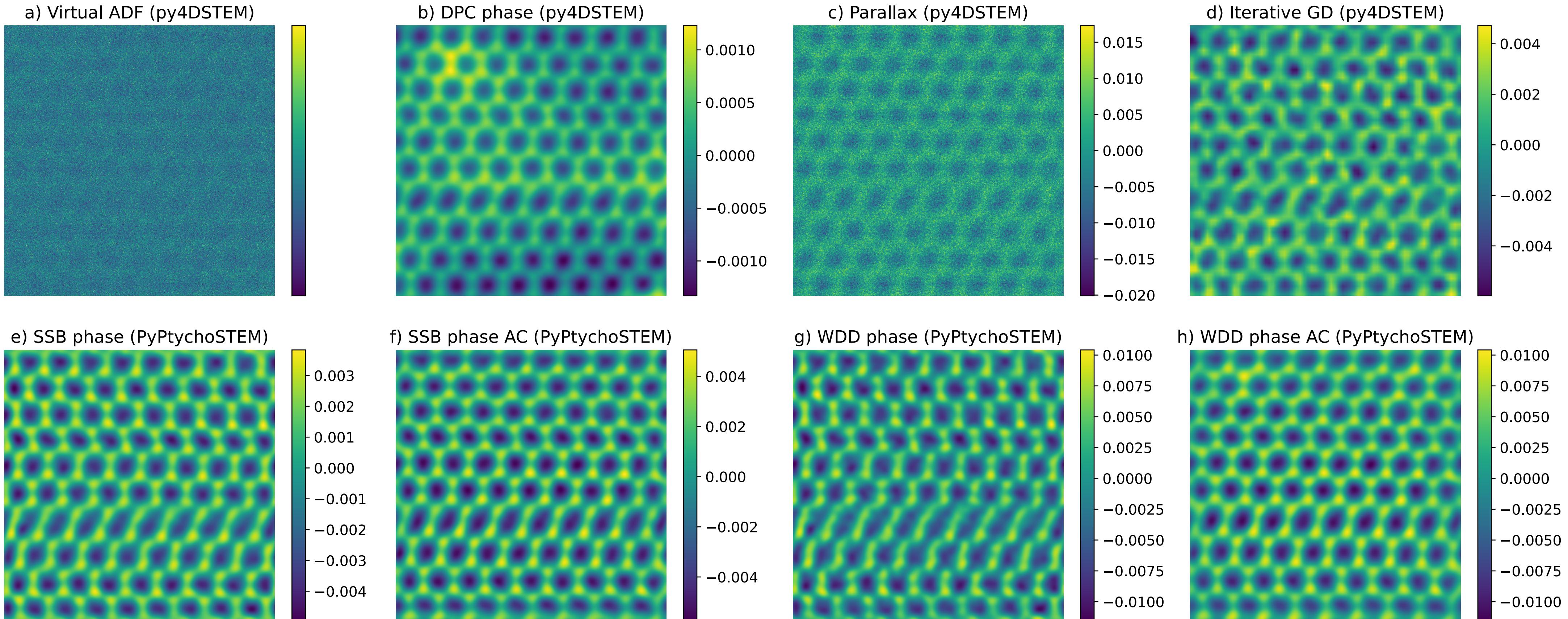
36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 192×192 Qpix  
16× binning in Q

# Comparing phase reconstruction algorithms (Q binning)



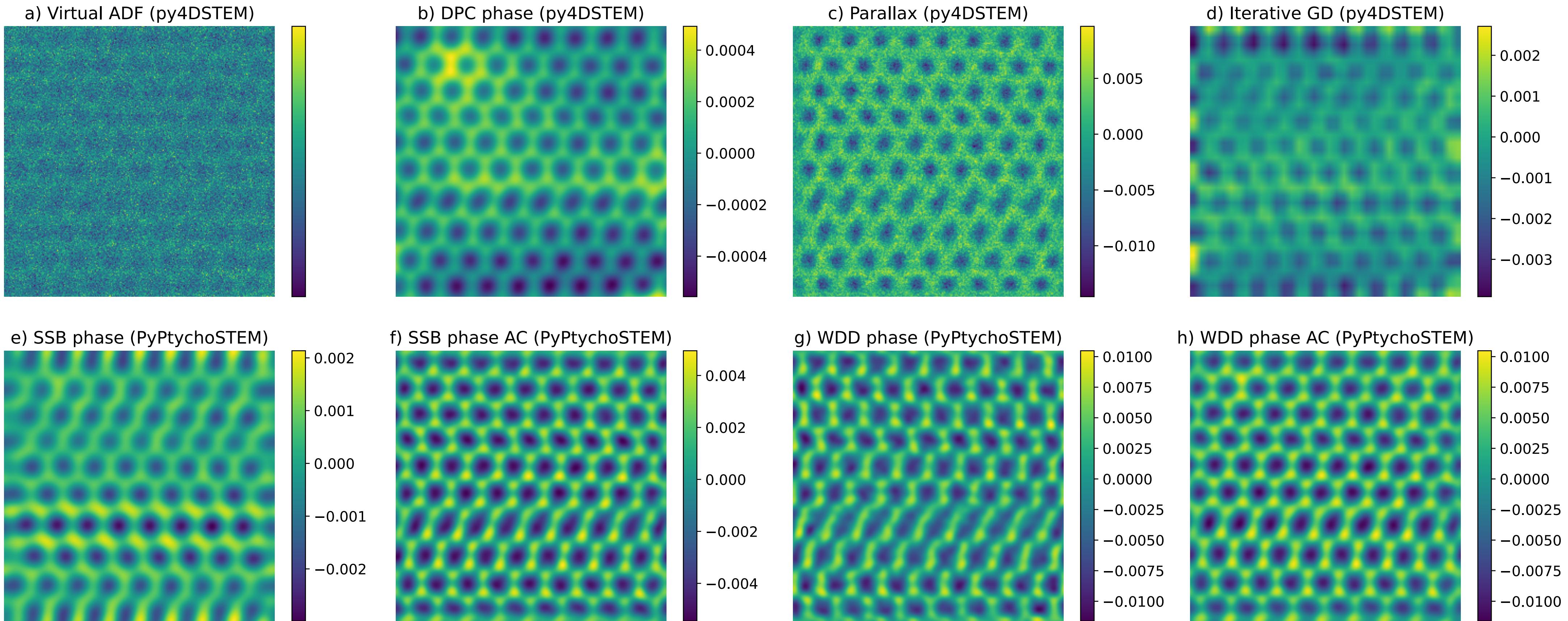
36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 192×192 Qpix  
32× binning in Q

# Comparing phase reconstruction algorithms (R binning)



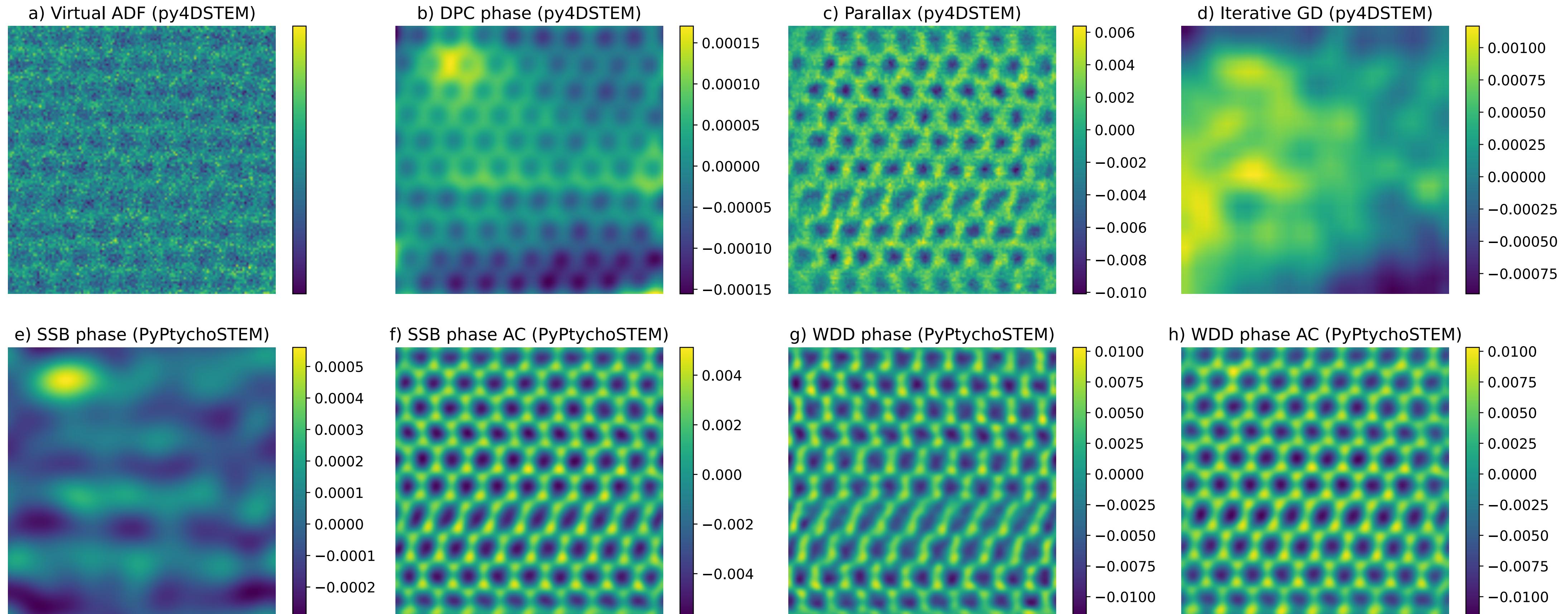
36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 96×96 Qpix (HW binQ by 2)  
1× binning in R

# Comparing phase reconstruction algorithms (R binning)



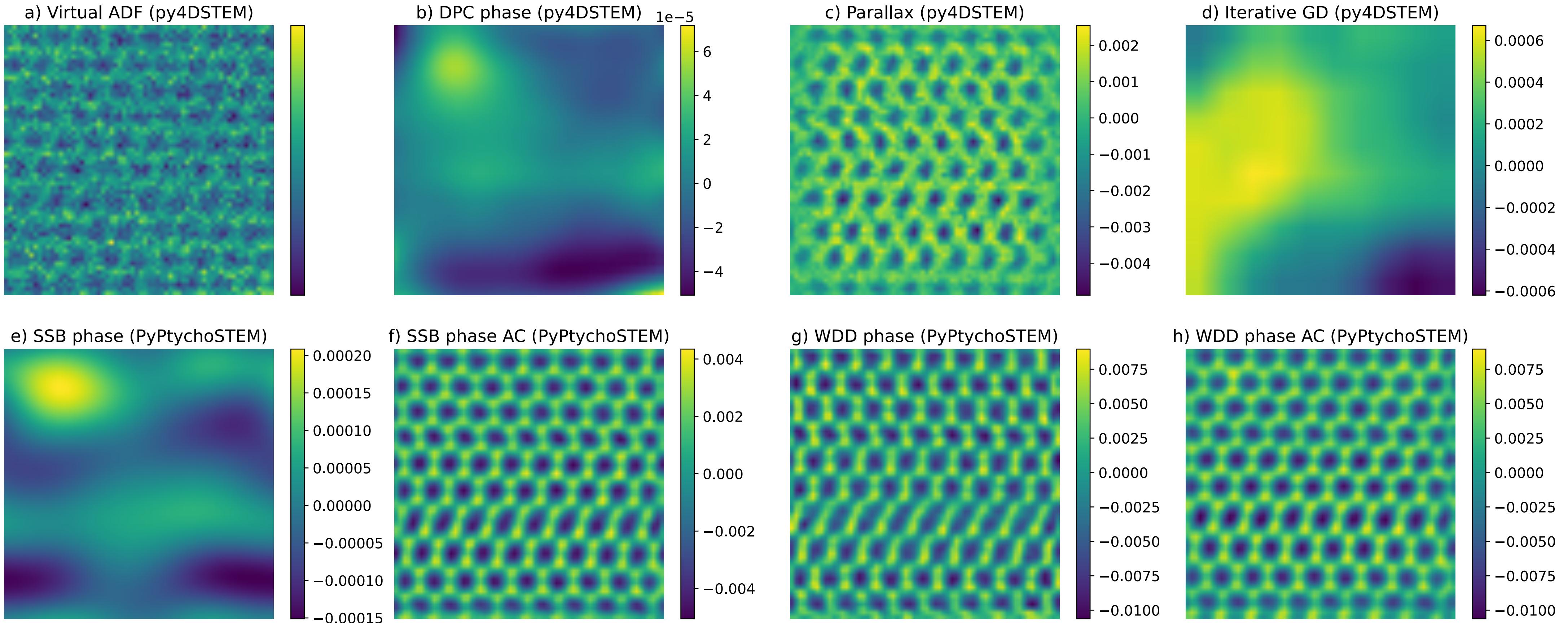
36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 96×96 Qpix (HW binQ by 2)  
2× binning in R

# Comparing phase reconstruction algorithms (R binning)



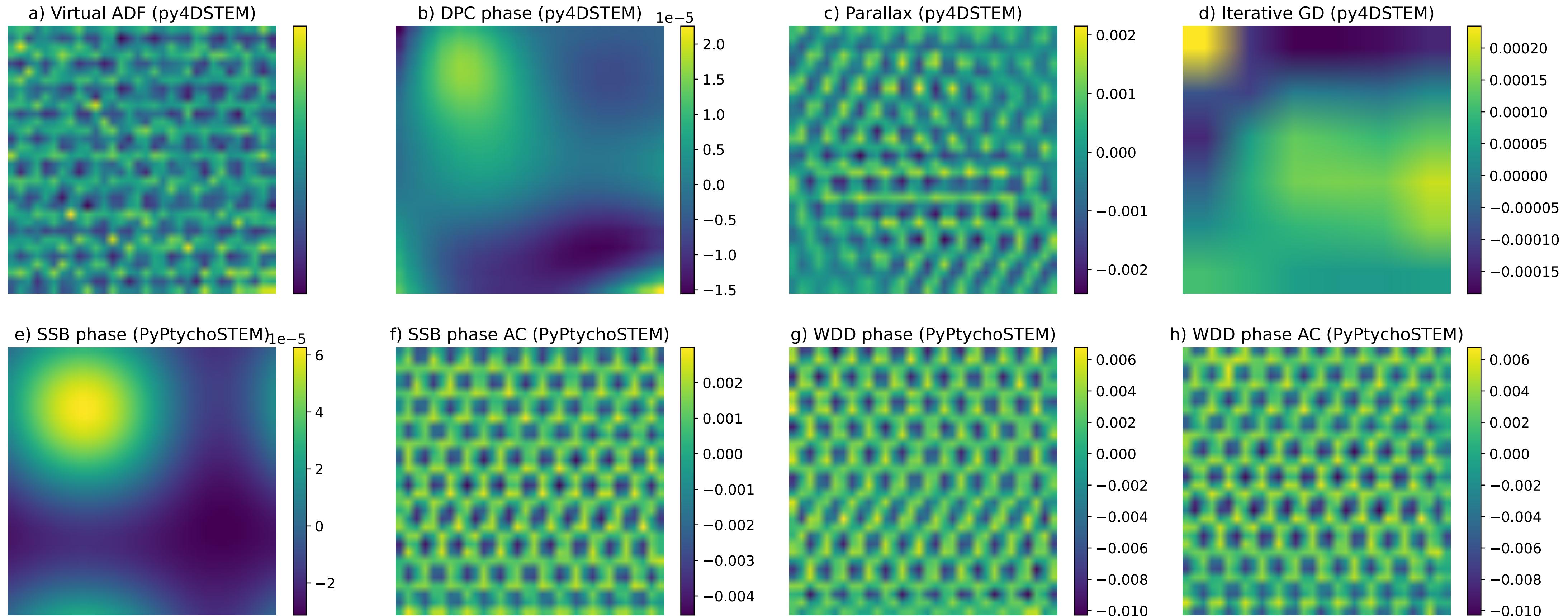
36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 96×96 Qpix (HW binQ by 2)  
4× binning in R

# Comparing phase reconstruction algorithms (R binning)



36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 96×96 Qpix (HW binQ by 2)  
8× binning in R

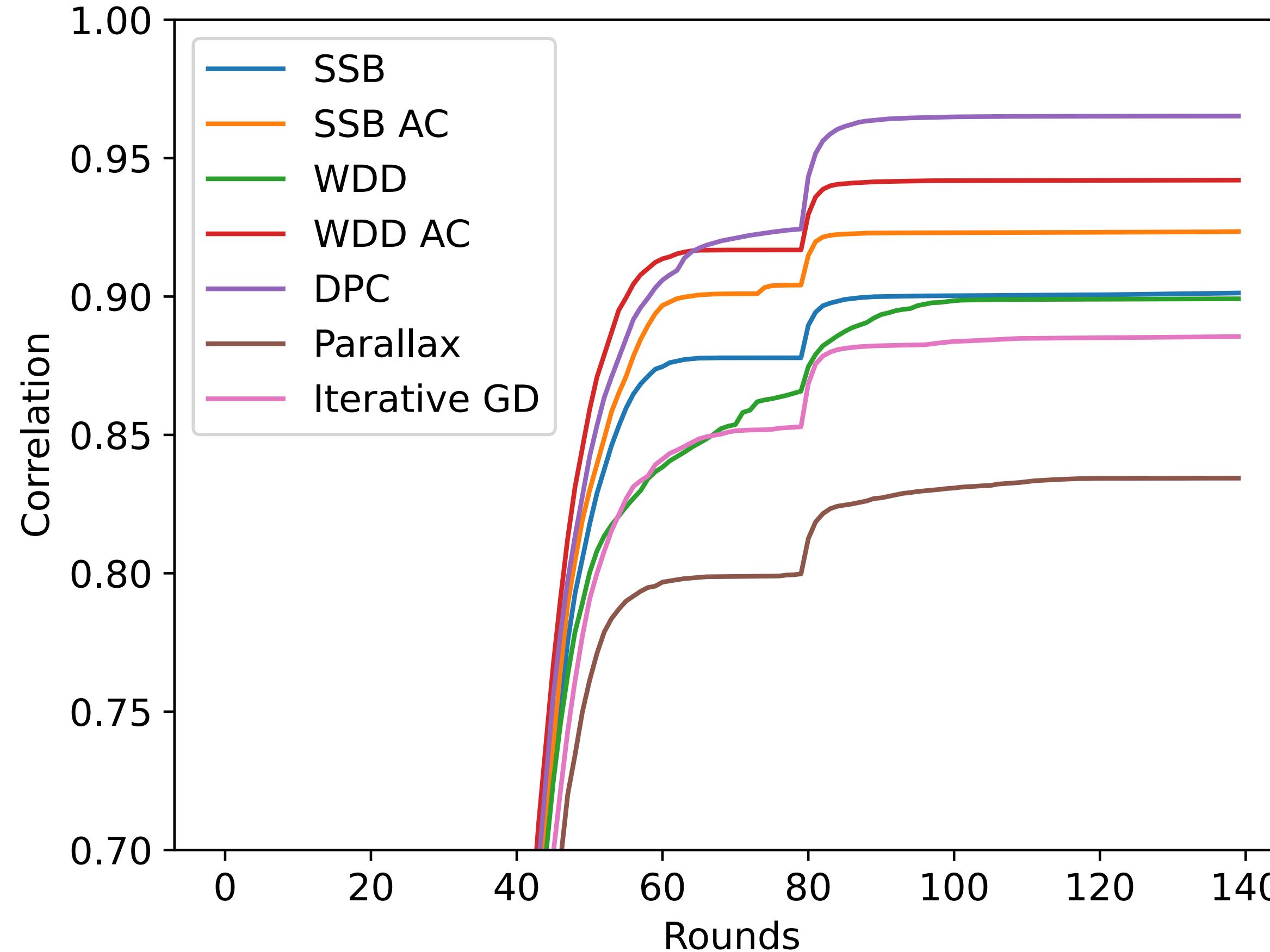
# Comparing phase reconstruction algorithms (R binning)



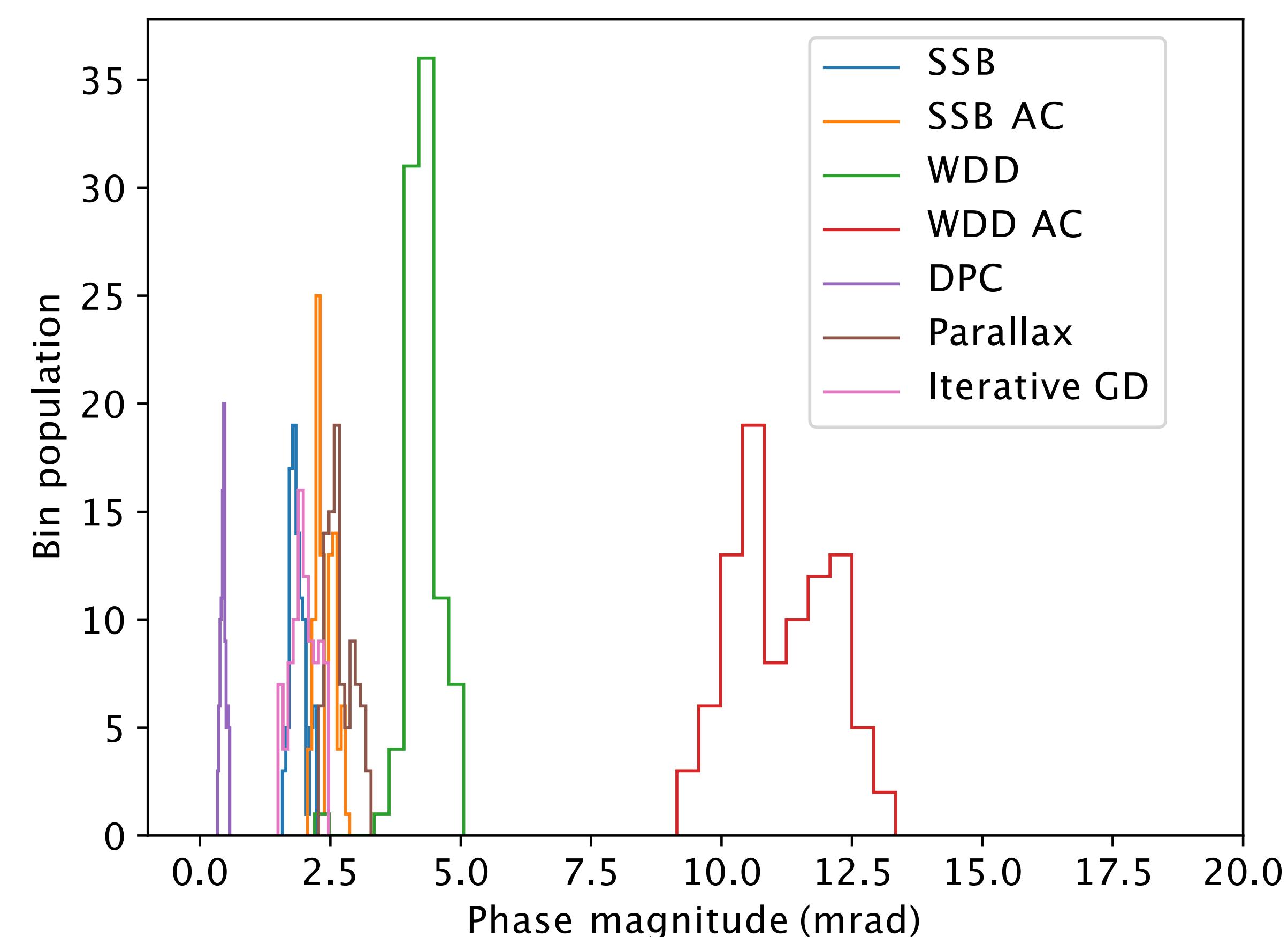
36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 96×96 Qpix (HW binQ by 2)  
16x binning in R

# Atomic phase assignment

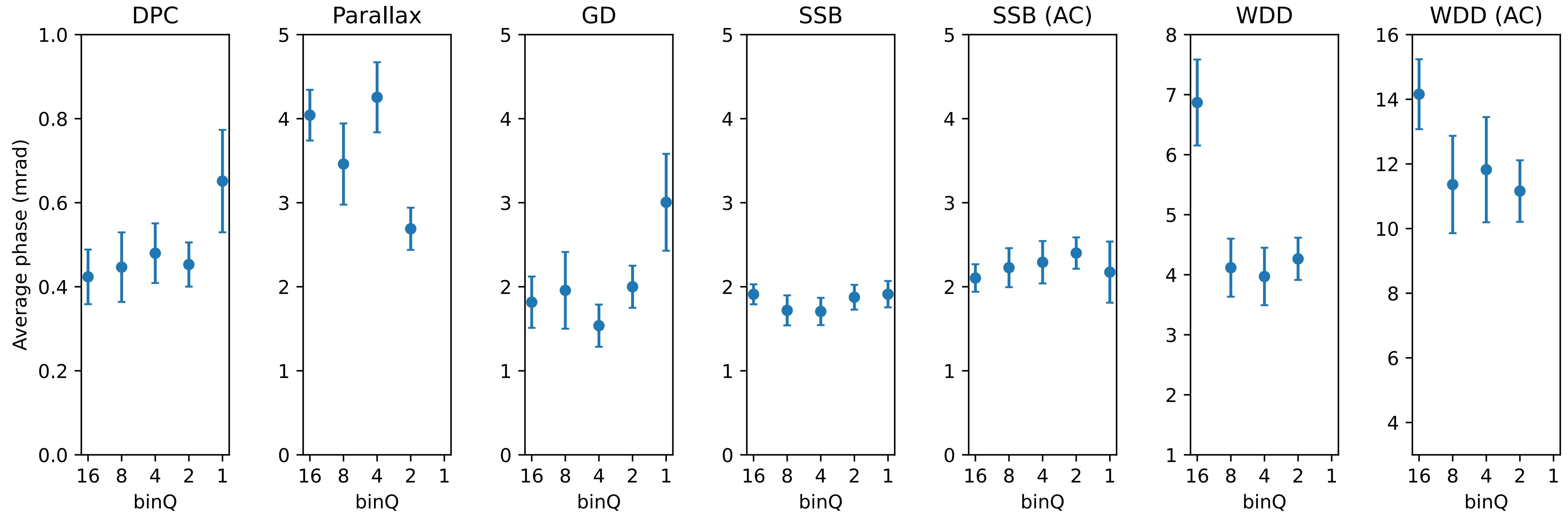
Optimization of atomic phases for reconstructions



Phase variation over graphene C atoms

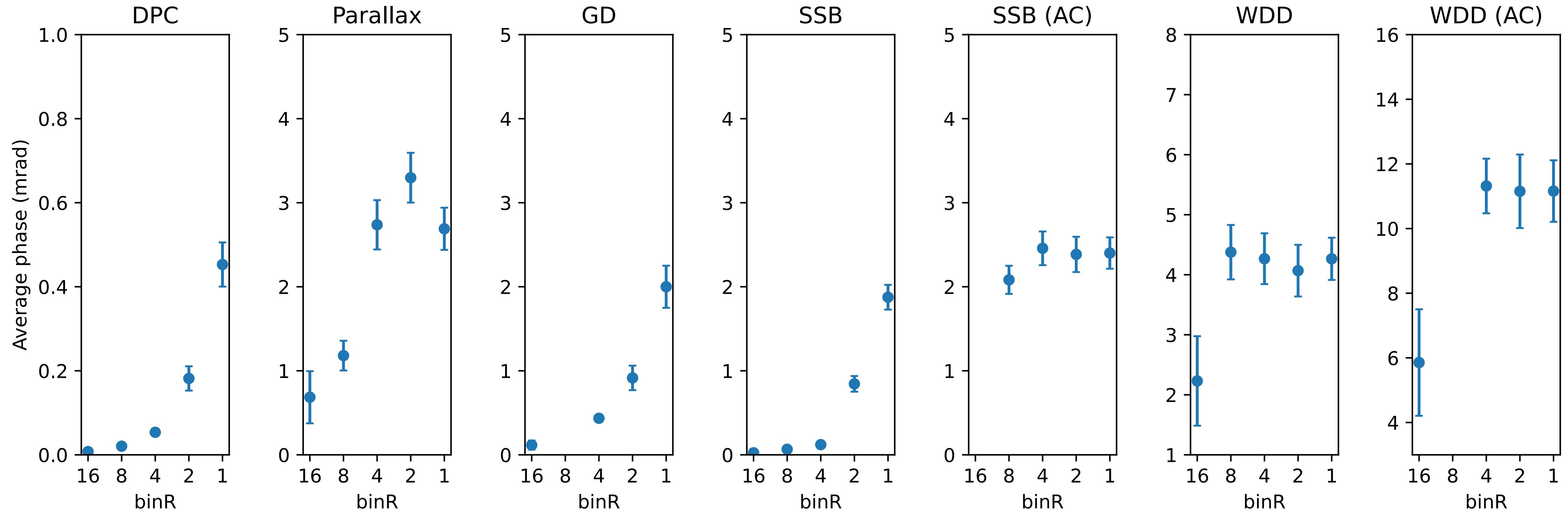


# Quantification of phase variation



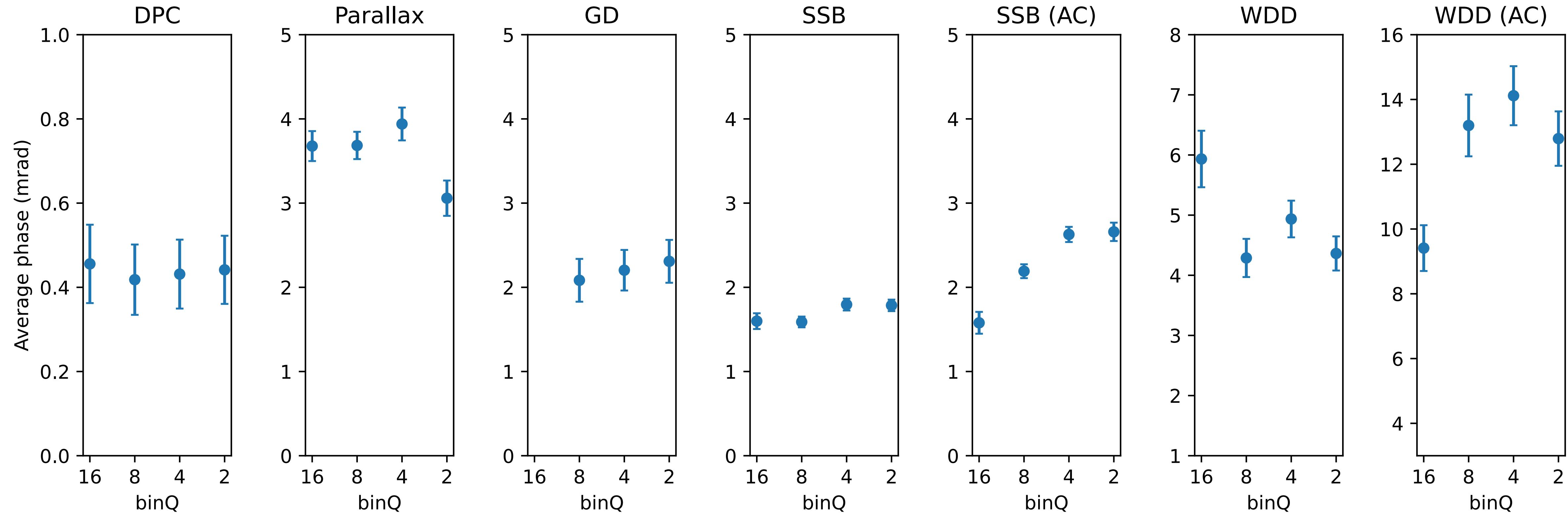
36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 192×192 Qpix (**binQ**)

# Quantification of phase variation



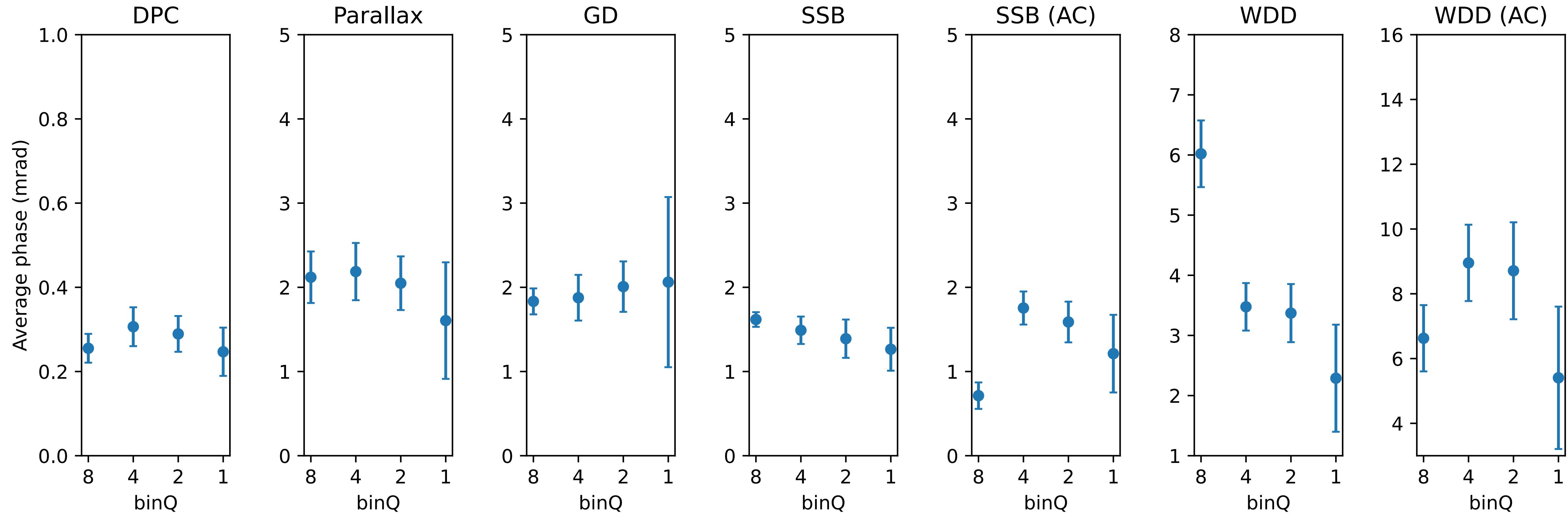
36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 192×192 Qpix (**binR**)

# Quantification of phase variation



60 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 192×192 Qpix

# Quantification of phase variation

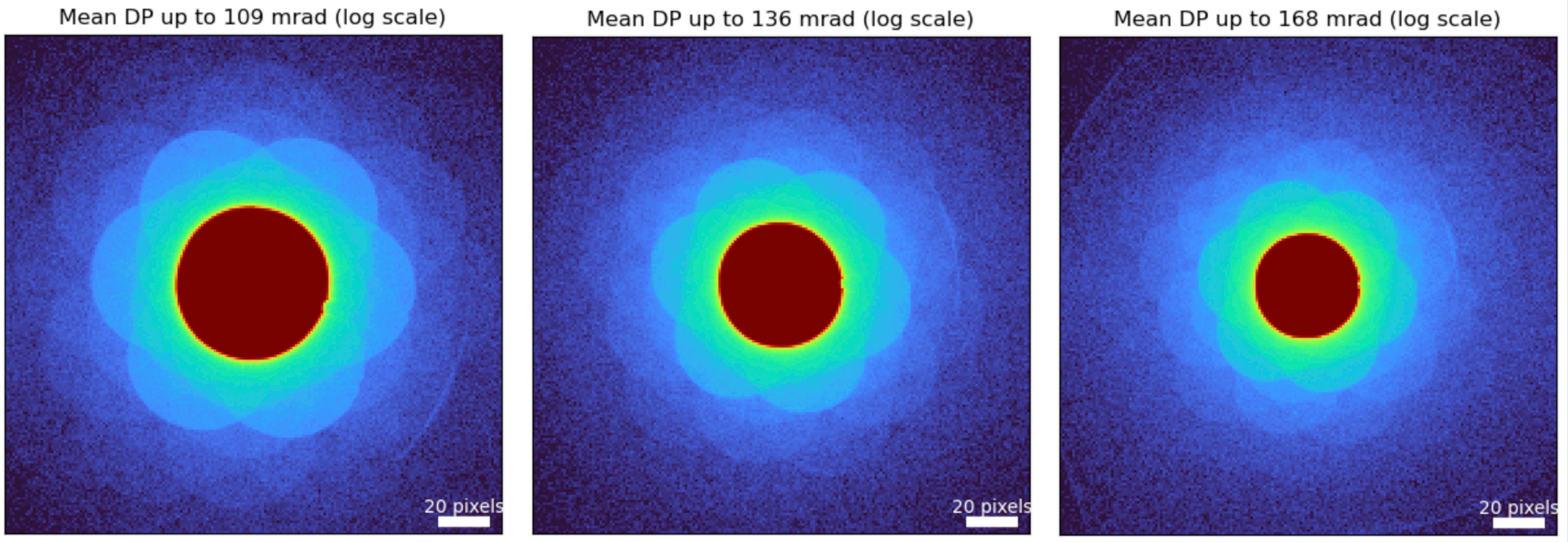


109 mrad maximum scattering angle, 100 us dwell time, 256×256 Rpix, 192×192 Qpix

# Computational cost (36 mrad datasets)

Scan (px)	binQ	CBED (Qpix)	Array (MB)	DPC (s)	Parallax <sup>†</sup> (s)	Iter. GD <sup>‡</sup> (s)	SSB (s)	SSB AC (s)	WDD (s)	WDD AC (s)
512×512	1	192×192	38656	16	–	578	236	18097	–	–
	2	96×96	9664	6.1	855	517	59	888	1938	2375
	4	48×48	2416	6.9	728	195	34	272	714	784
	8	24×24	604	5.4	68	146	13	87	306	317
	16	12×12	151	5.0	24	126	7.2	41	163	165

# (Some) Future prospects



- Defocused-probe ptychography with iterative reconstructions
- Variable camera lengths (for super-resolution up to  $\sim 4 \times$  alpha)
- Live virtual detectors / (semi-)live single-sideband ptychography?
- Structure & charge transfer in various materials... *and much more*

# Upcoming Japan stay (Sep 2024 – Feb 2025)



Nagashima (TUS, Sep)



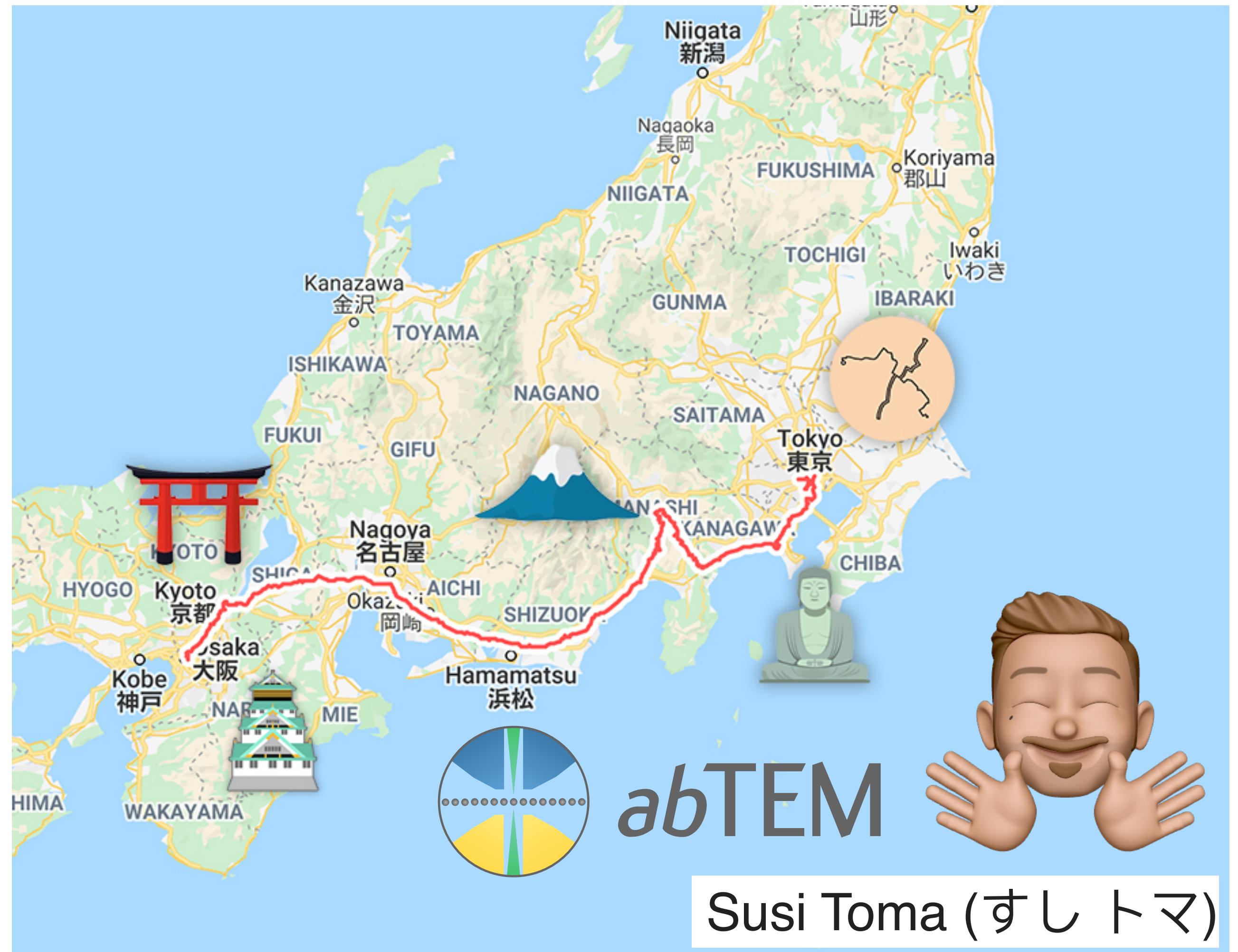
Suenaga (Osaka, Oct)



Shibata (Tokyo, Nov–)



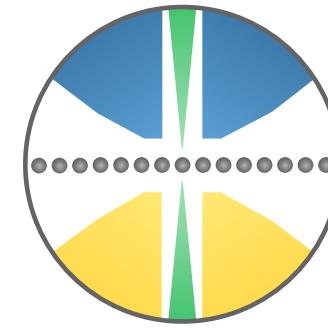
you? (???)





universität  
wien

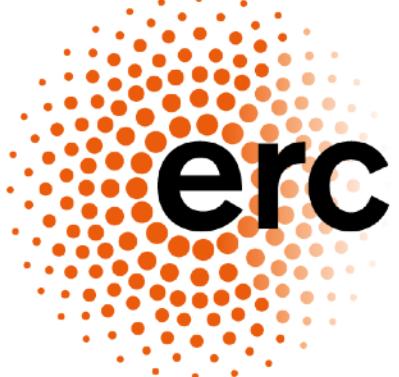
Φnm



abTEM

FWF

Der Wissenschaftsfonds.



Thank you!

Questions, collaborations: [toma.susi@univie.ac.at](mailto:toma.susi@univie.ac.at)



**Open code & talk slides:**  
<https://github.com/TomaSusi/arina-ptycho>

**Open datasets (click "Open link"):**  
<https://phaidra.univie.ac.at/o:2081765>

