



universität
wien

nion

DECTRIS
detecting the future

emc2+24
COPENHAGEN

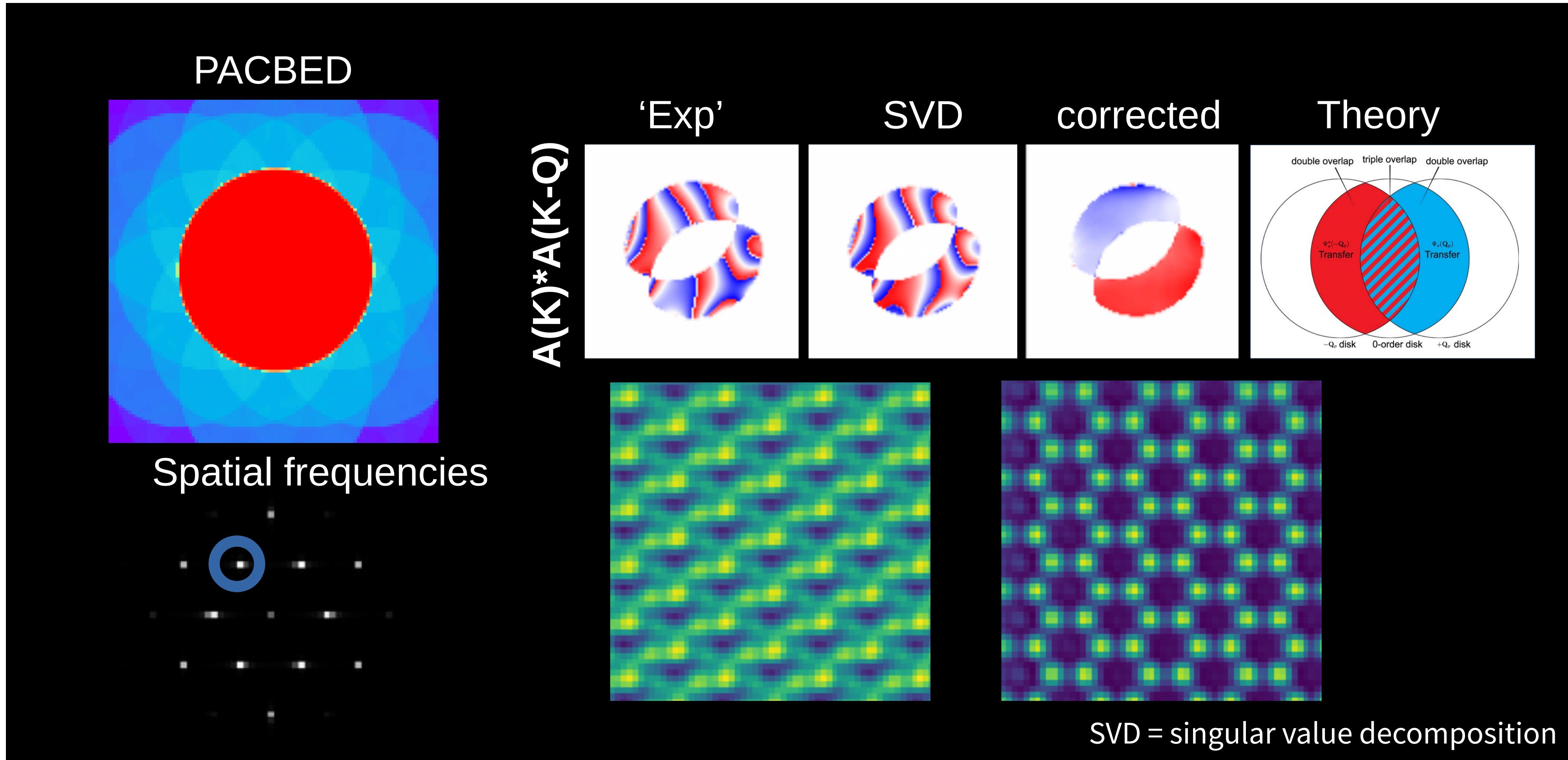
Near-Ideal Direct-Electron Focused-Probe 4D-STEM Data for **Open-Source Phase Reconstructions**

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)



(Bright-field) ptychography & aberration correction



Timothy
Pennycook

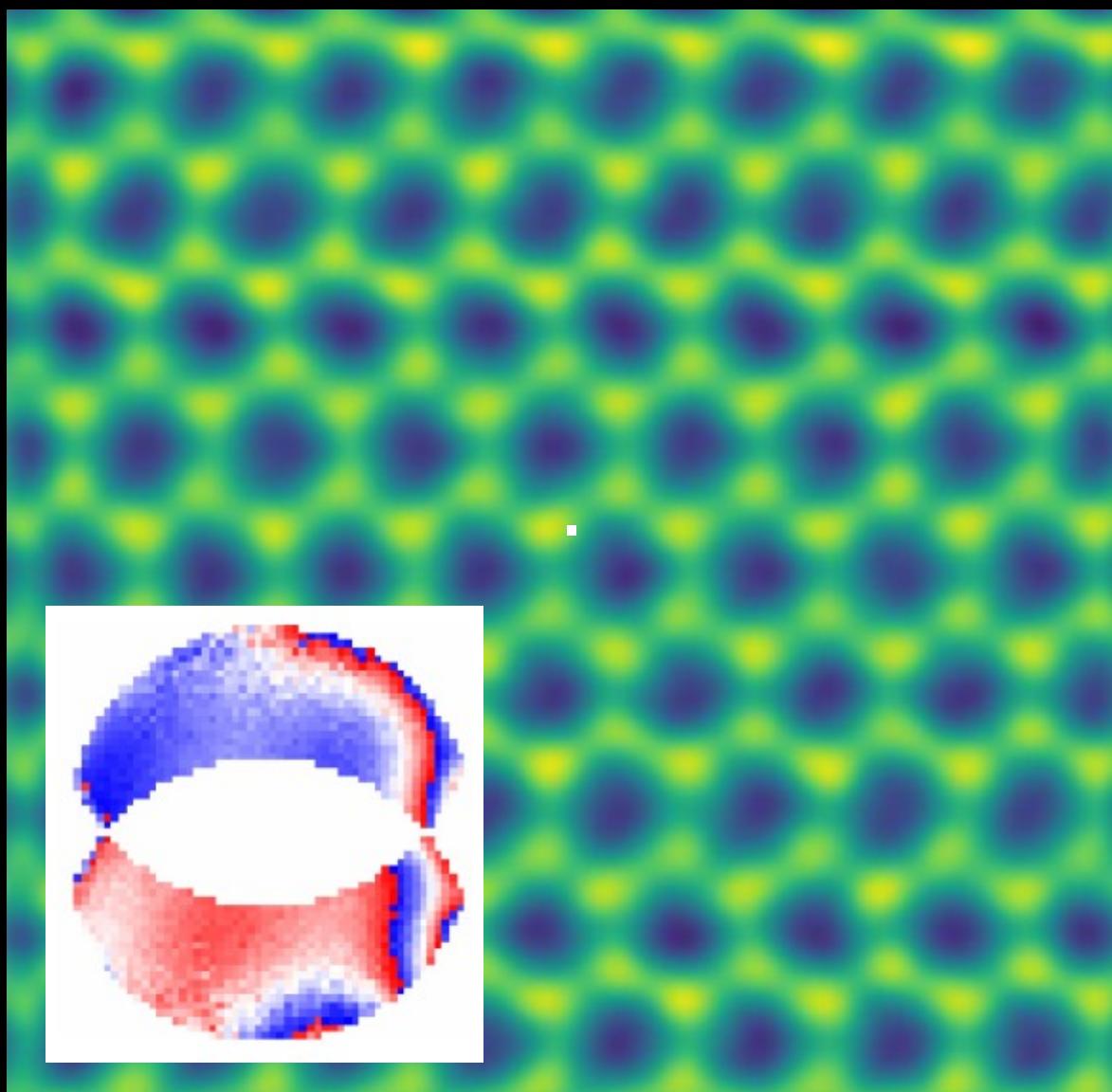


Christoph
Hofer

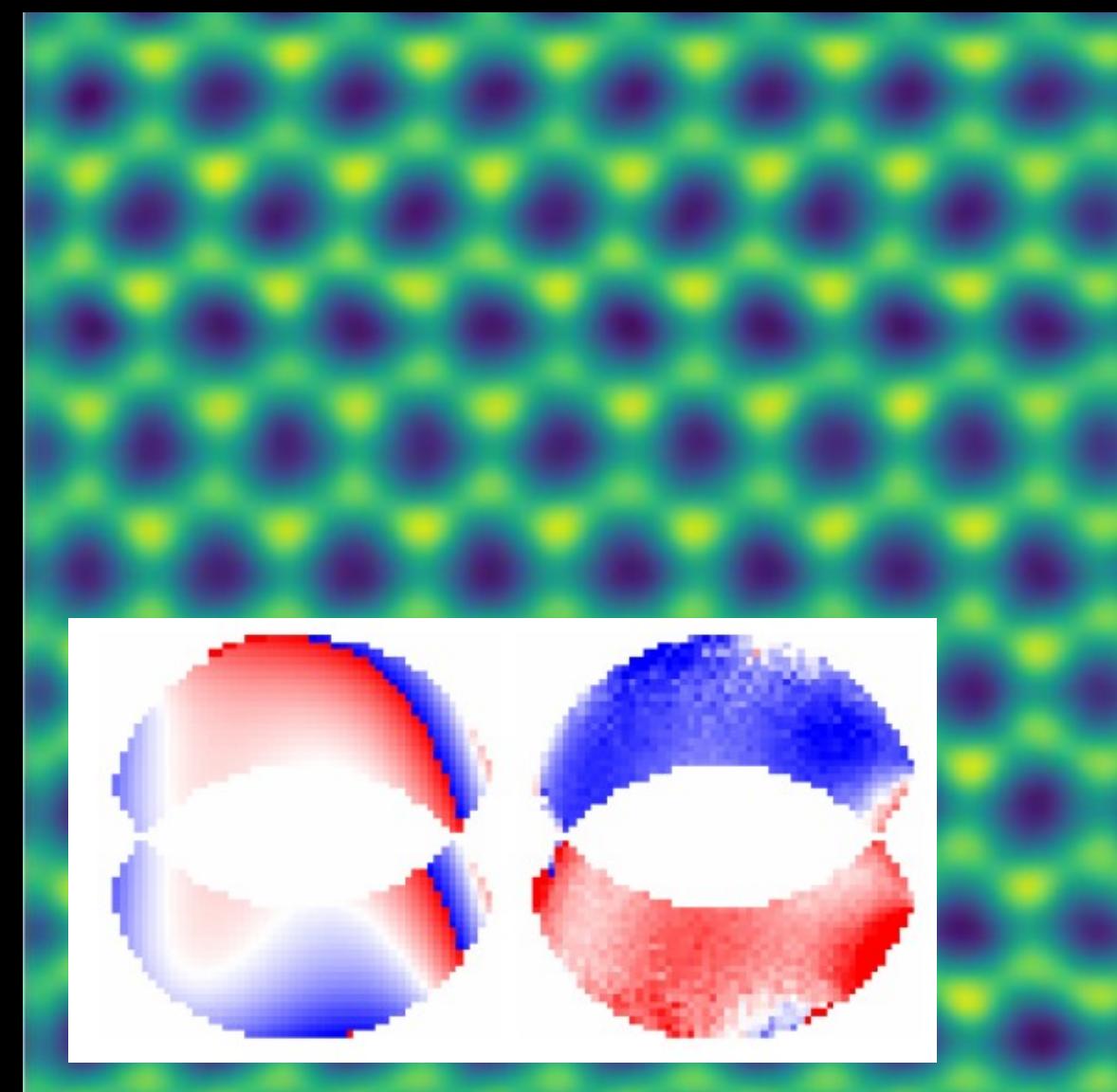
Post-acquisition aberration correction (5th order)

Experimental data of monolayer graphene, 35 mrad (Vienna)

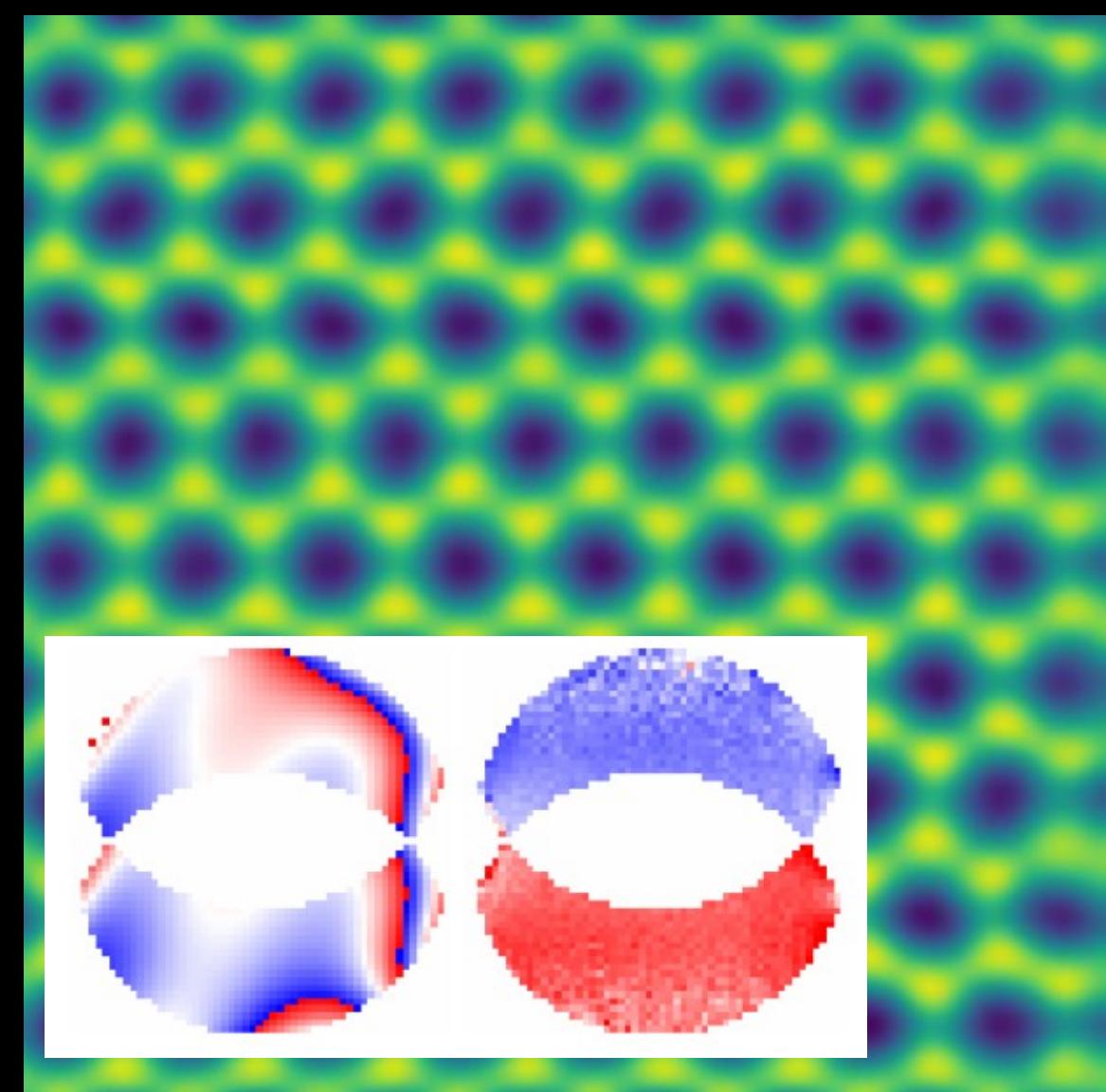
uncorrected



3rd order corrected



5th order corrected

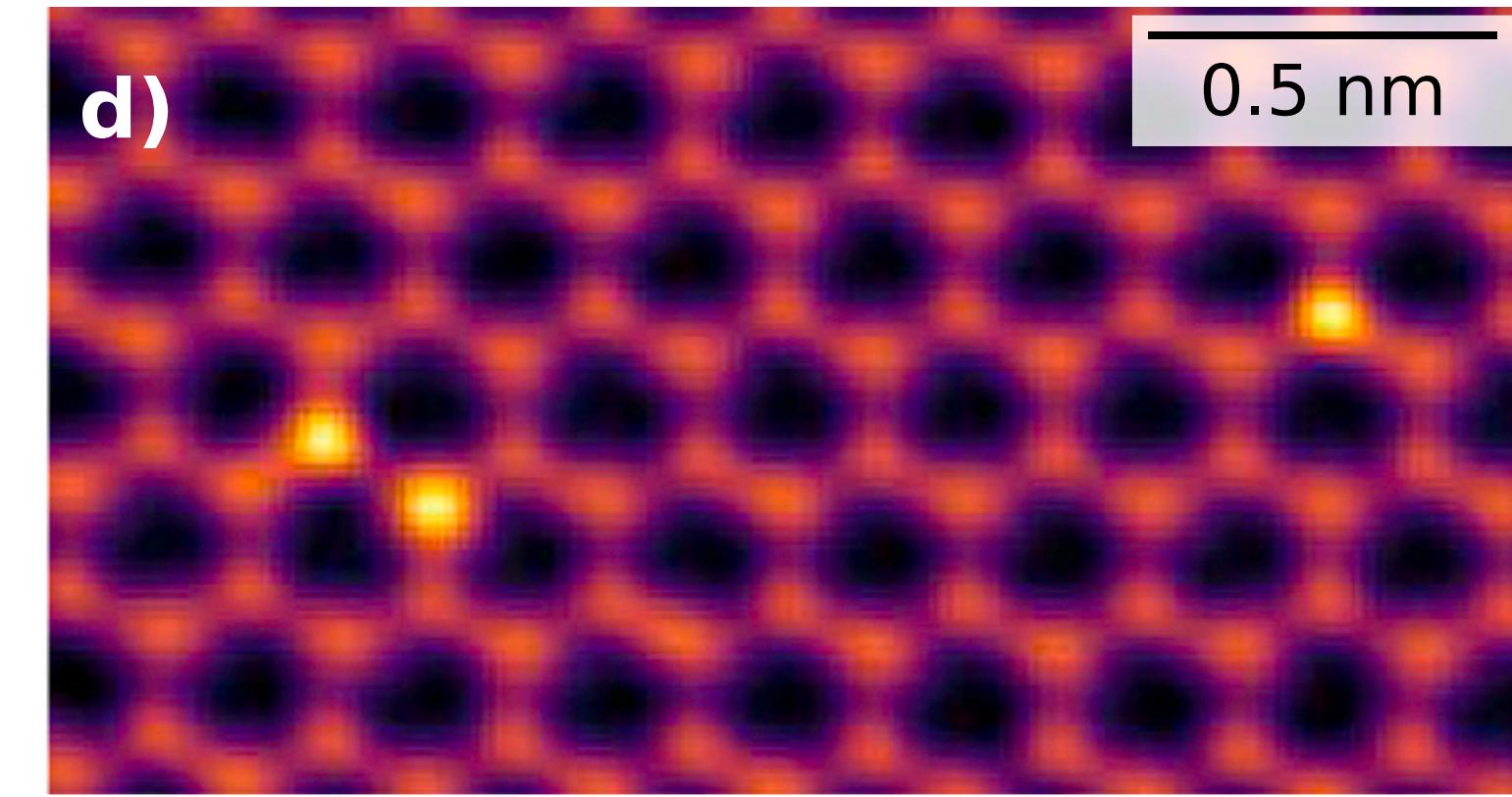
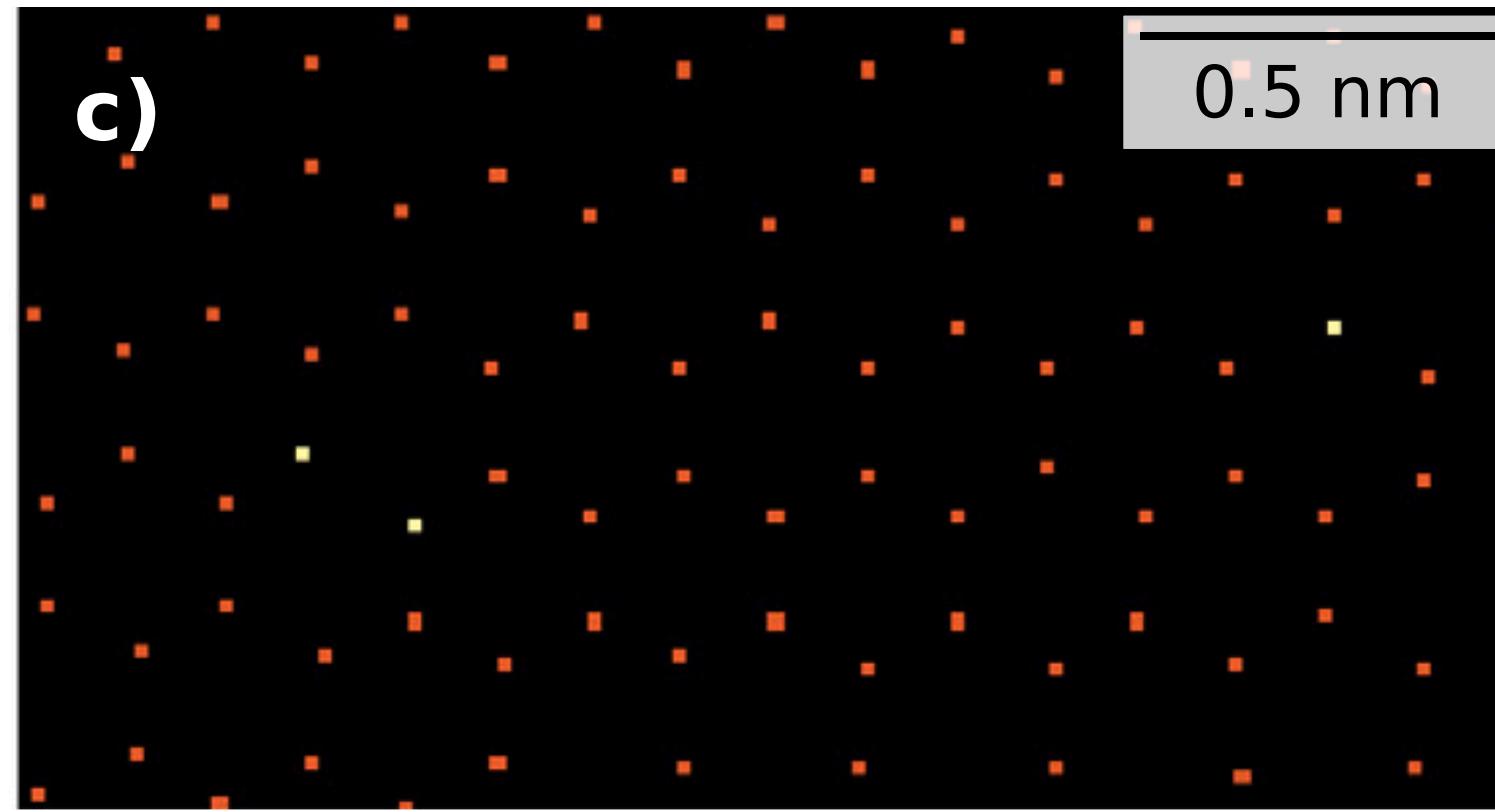
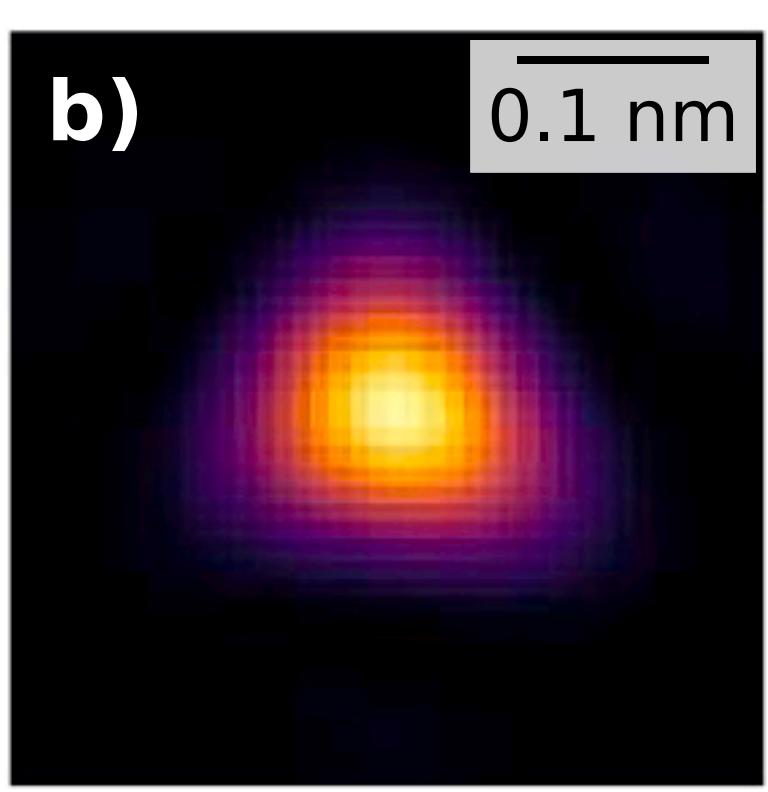


Timothy
Pennycook



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

μ^{exp} and μ^{sim} = mean values

σ^{sim} and σ^{exp} = standard deviations

Can account for:

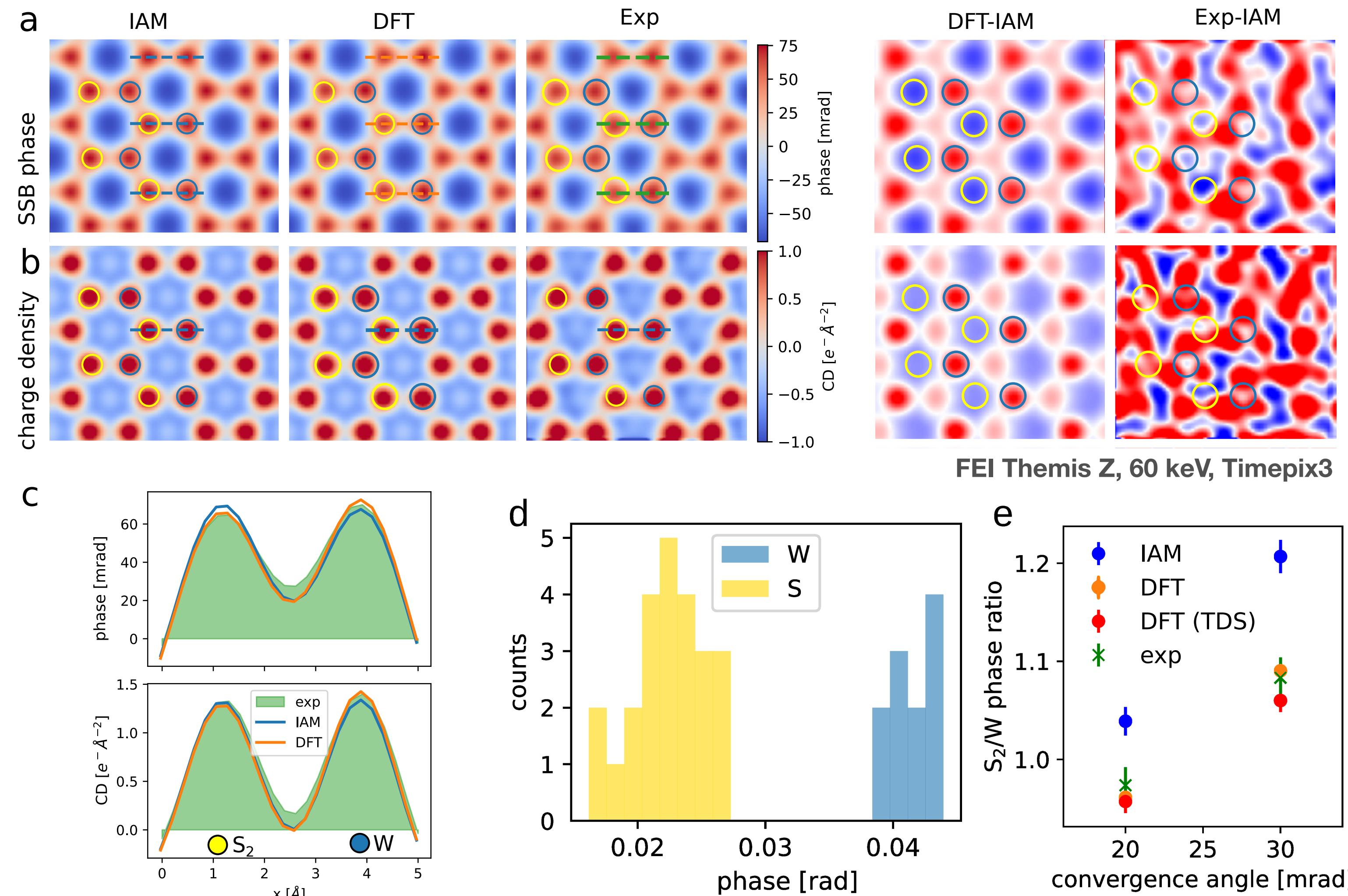
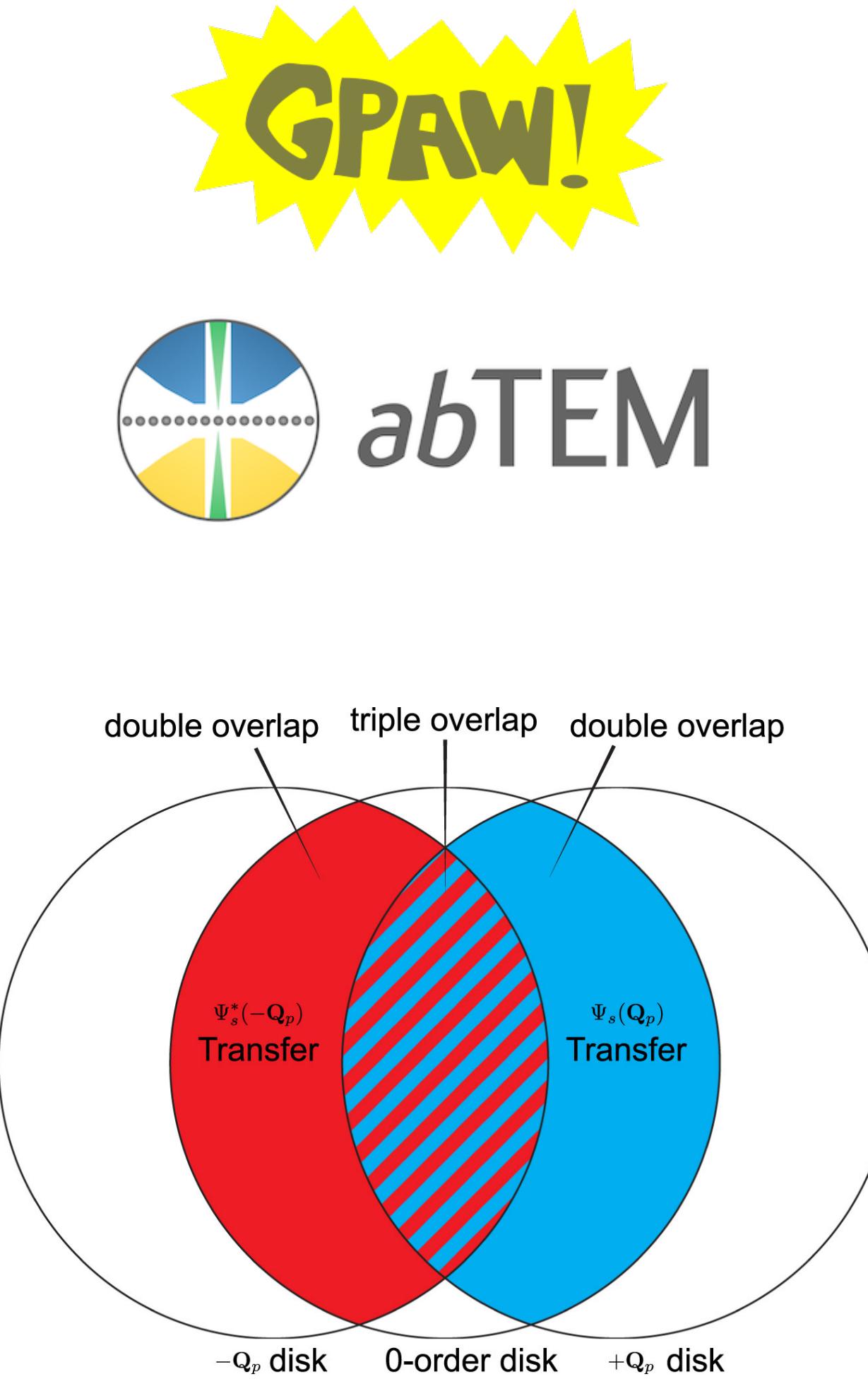
- aberrations
(not used later in this talk!)
- incoherence
- scan distortions
- sample drift
- sample tilt
- etc...

Phase analysis



STEM_Optimization*
(commit 6c549ba9)

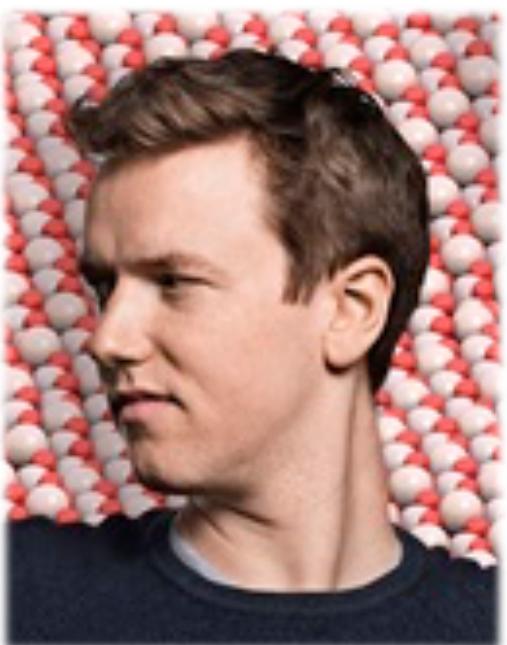
Collaboration: detecting charge transfer in defective WS₂



My favourite hobby project:



abTEM



Simulation challenge for phase-contrast

Current limitations

- Sample: quality, stability
- Microscope: stability, cleanliness, automation
- Electron beam induced charging, damage, contamination
- Need for DFT calculations: speed, size, integration with image simulations
- Strong dynamical effects
- Difficulty of determining specimen & imaging parameters incl. thickness

 JÜLICH
Forschungszentrum

Our multislice solution

1. Integration with *ab initio* codes



Jacob
Madsen

2. Python, but *fast*



CuPy



3. Promote notebook/scripted workflow



4. Access to low-level API: Easy to extend



5. Open source (huge ecosystem!)



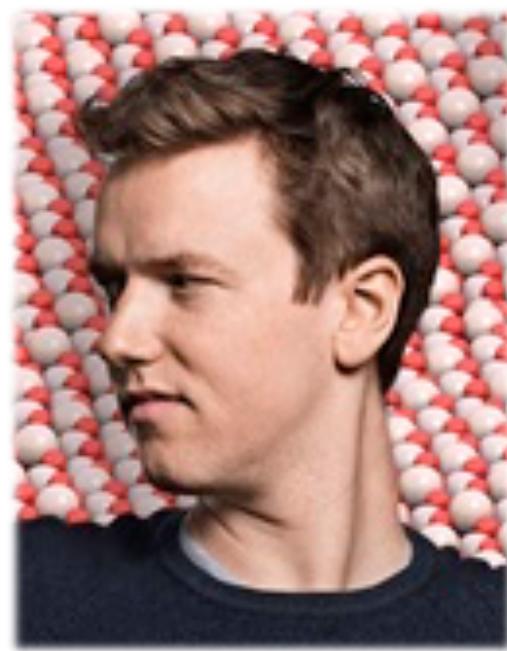
6. High-performance parallel GPUs & file access

Rafal Dunin-Borkowski @ Salve 2D22

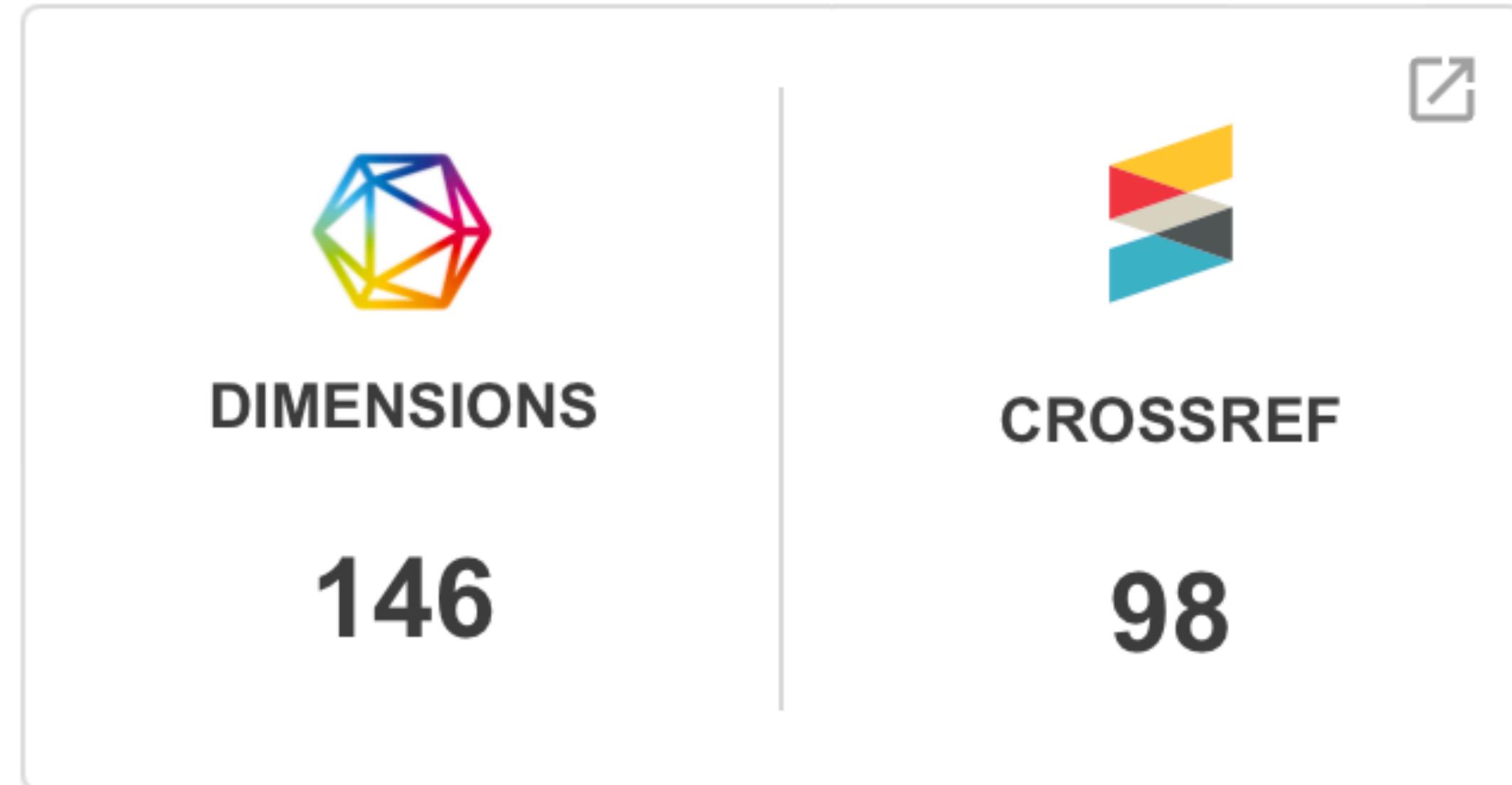
My favourite hobby project:



abTEM



Jacob
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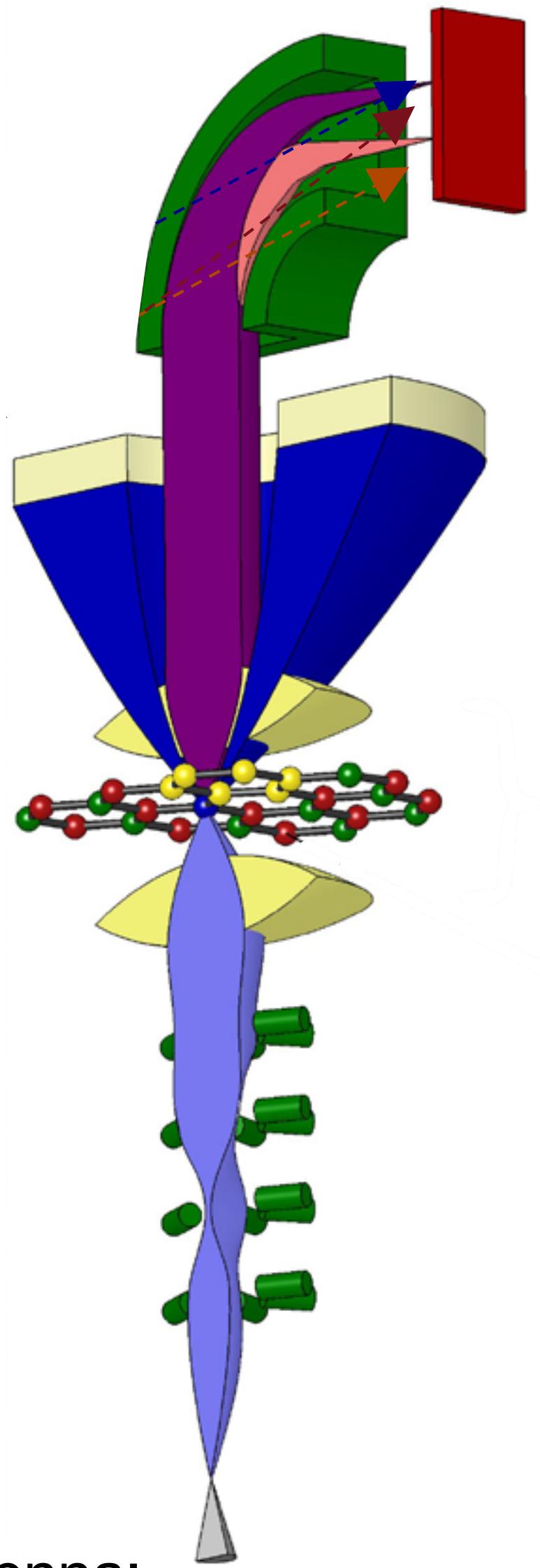


6. High-performance parallel GPUs & file access



Major upcoming & planned features:

- Arbitrary tilts for 3DED / microED
- Plasmon loss for incoherence
- Pauli multislice for magnetic scattering
- True frozen phonon displacements (& vibrational EELS...)



Vienna:

Nion UltraSTEM100

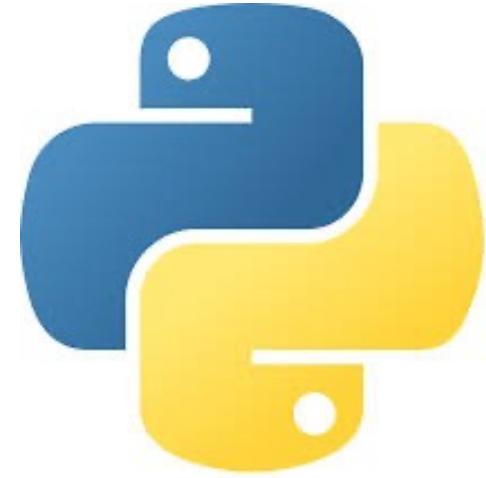
Ptycho powered by (Python) open source

Data acquisition



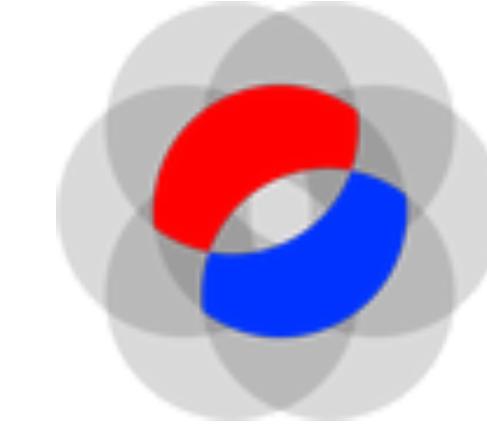
Nion Swift
(0.16.10)

Language

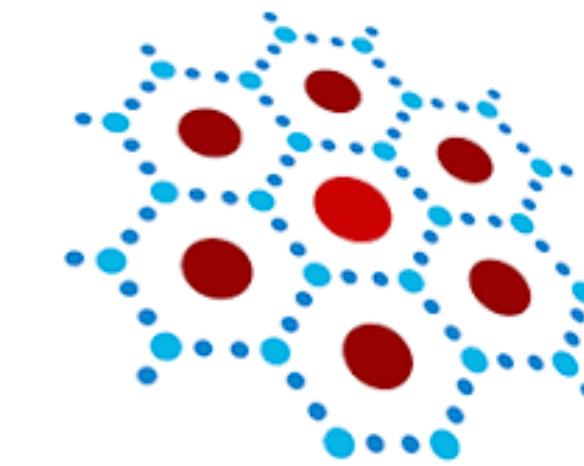


Python
(3.11.9)

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 single-slice ptychography (**GD**)

Direct-electron detector for 4D-STEM

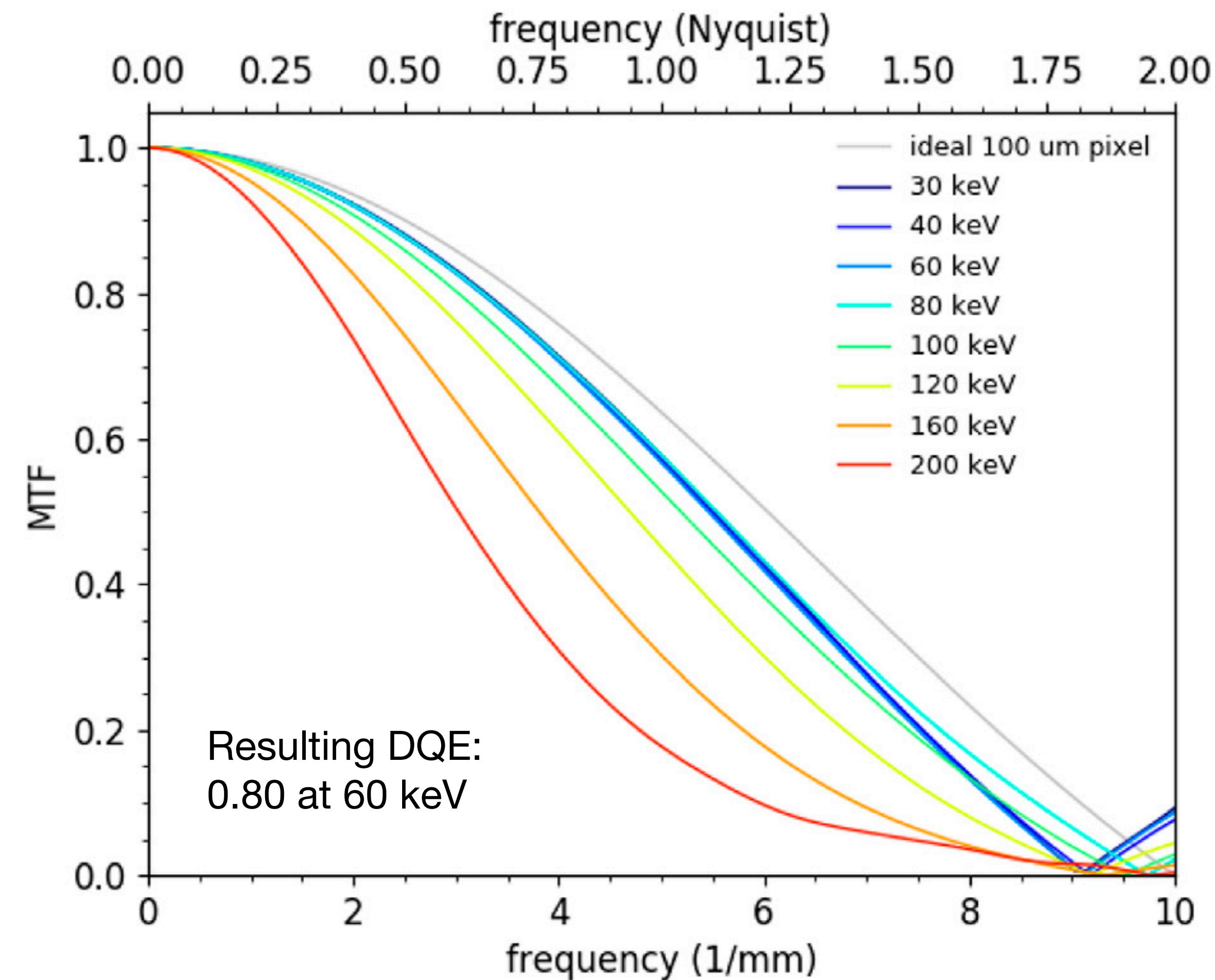
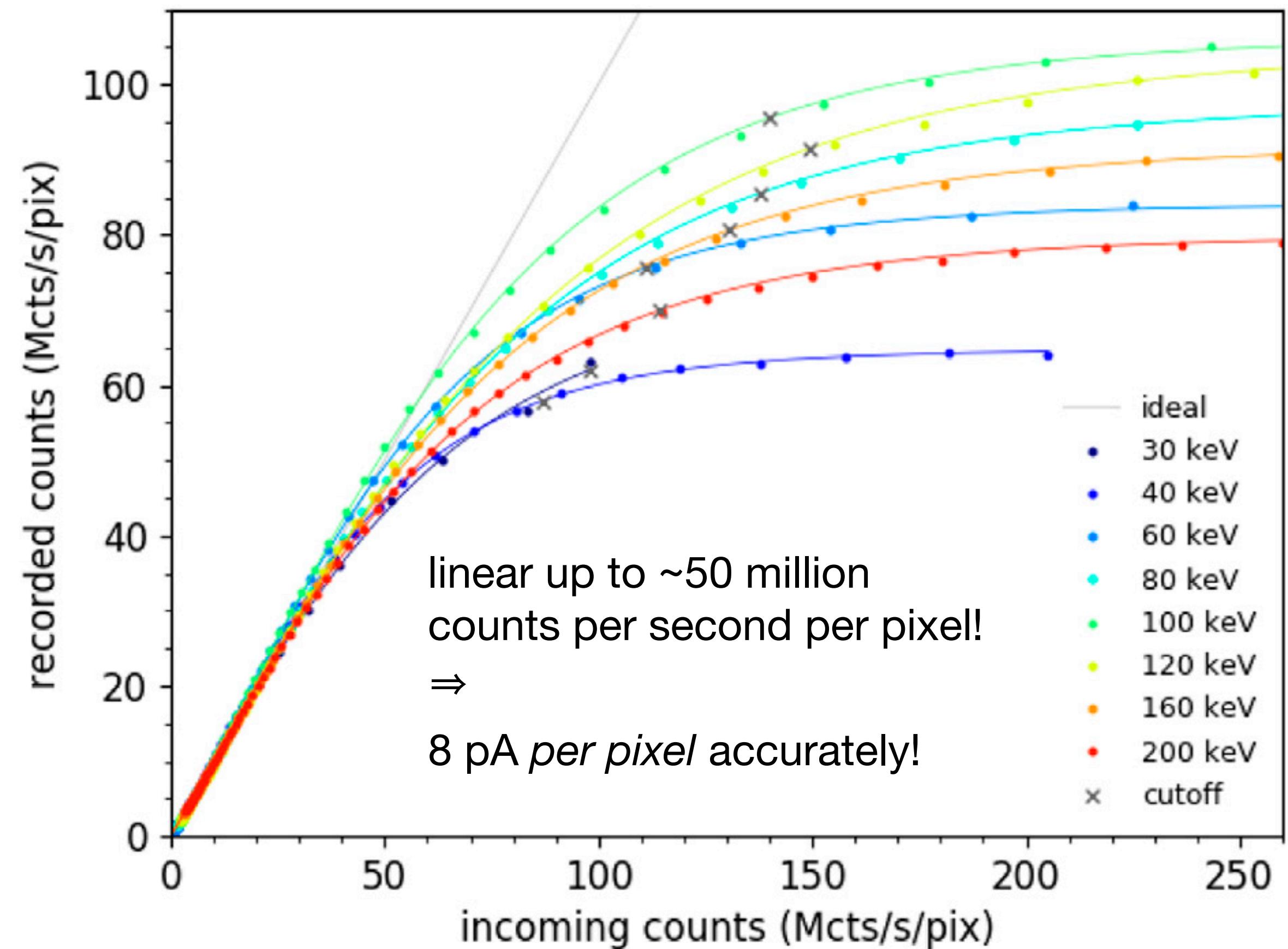
Number of pixels (Q_{px})	192 × 192
Active area [mm 2]	20 × 20
Pixel size [μm^2]	100 × 100
Sensor material	Silicon (Si) or high-Z
Energy range [keV]	30–300
Frame rate (max.) [Hz]	120,000
Count rate (max.) [el/s/pixel]	10^8
Detective Quantum Efficiency, DQE(0)	0.82 @ 80 keV 0.75 @ 200 keV, 0.75 @ 300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



On-axis (btw.
HAADF & MAADF)

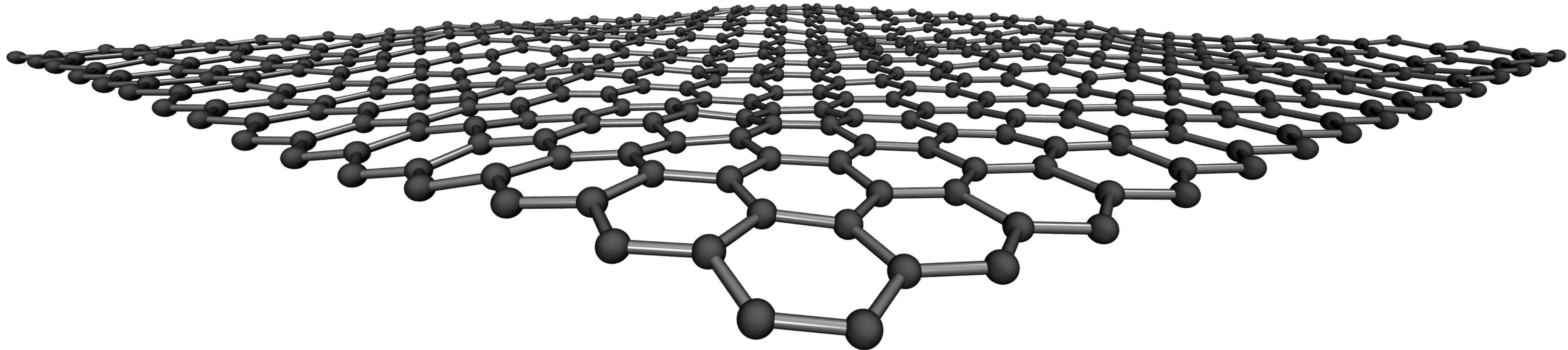
Near-ideal 4D-STEM performance at 60 keV

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

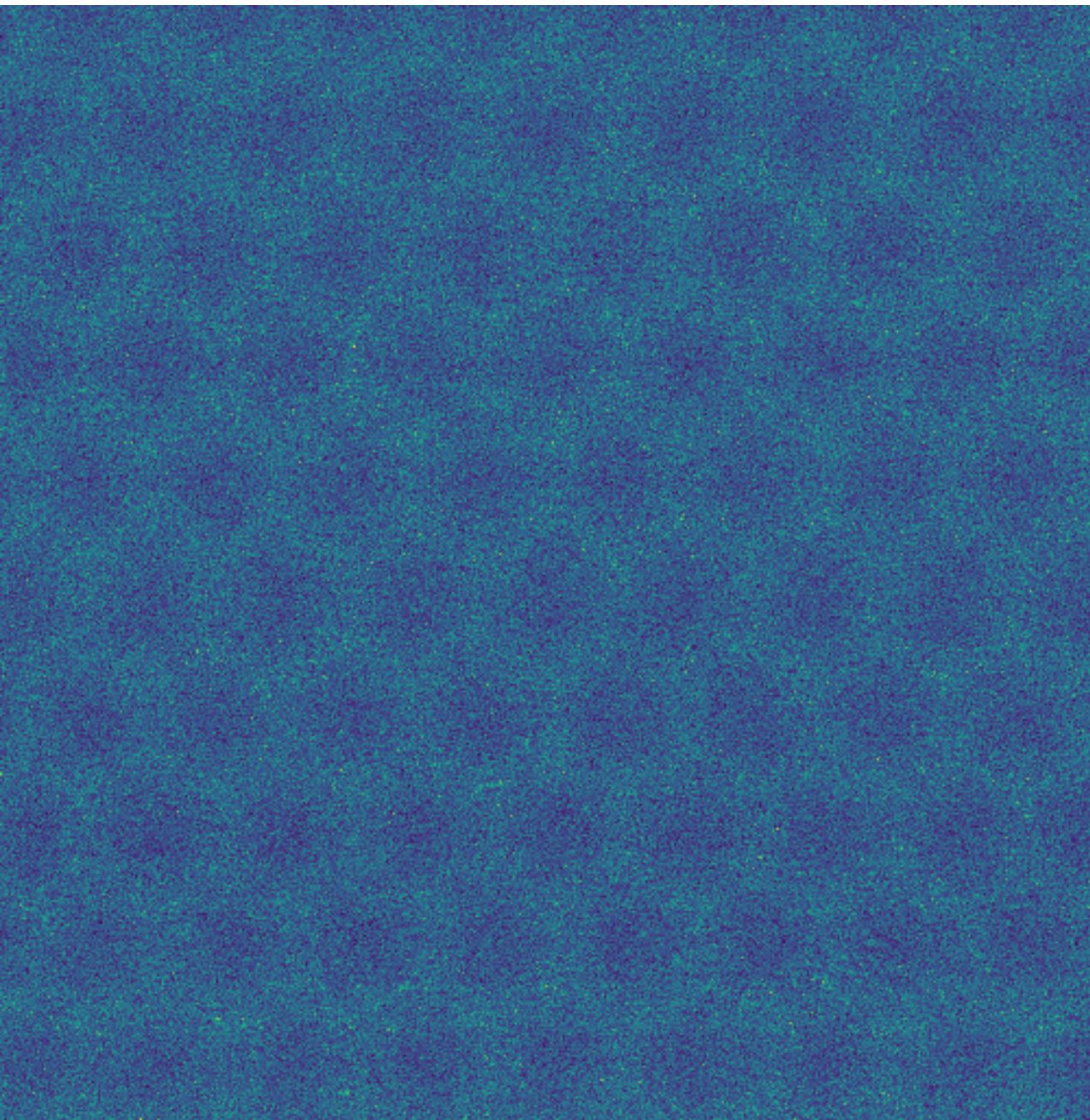


Graphene as ideal uniform phase object

- ✓ ∞ 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



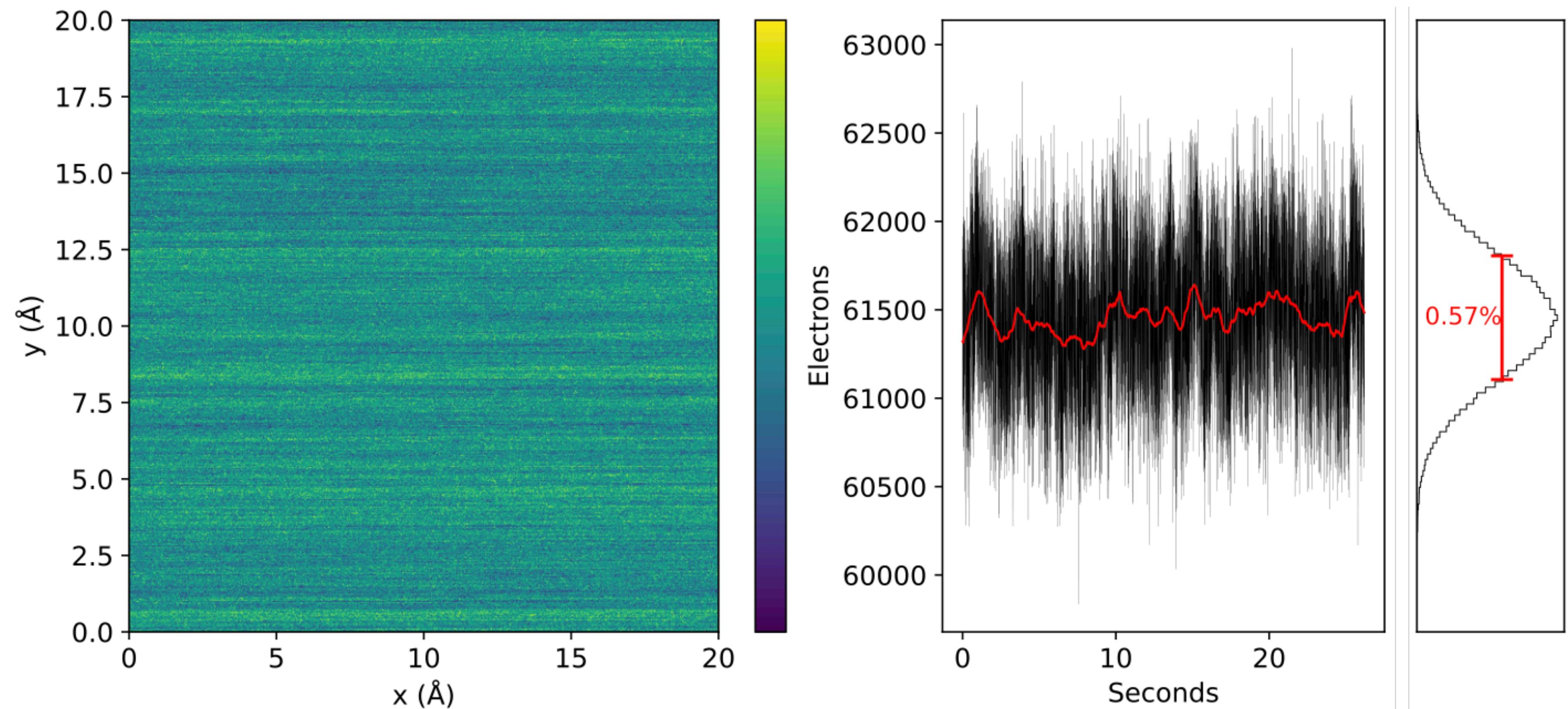
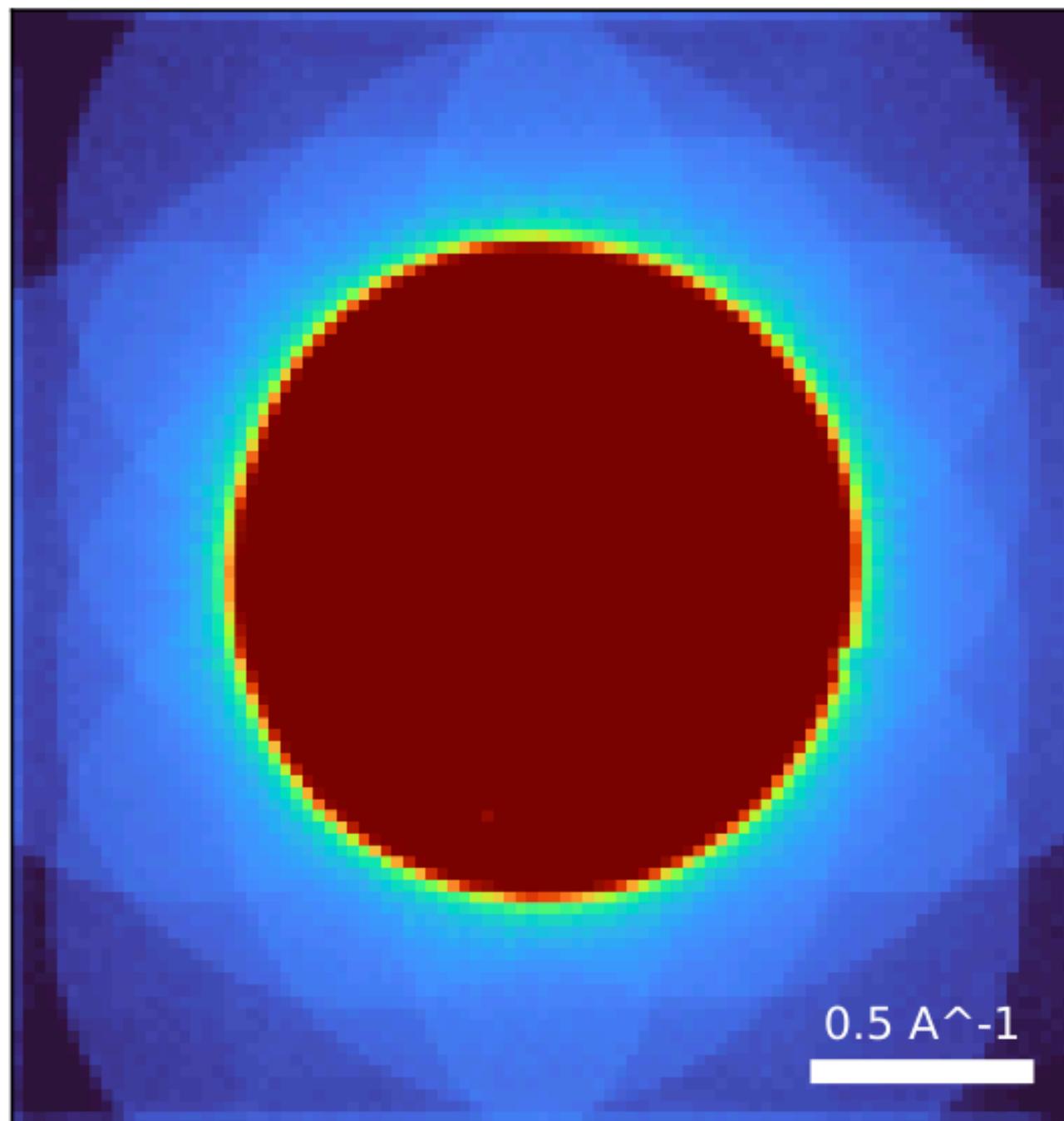
Nion + ARINA 4D-STEM data collection



HAADF (80–300 mrad)

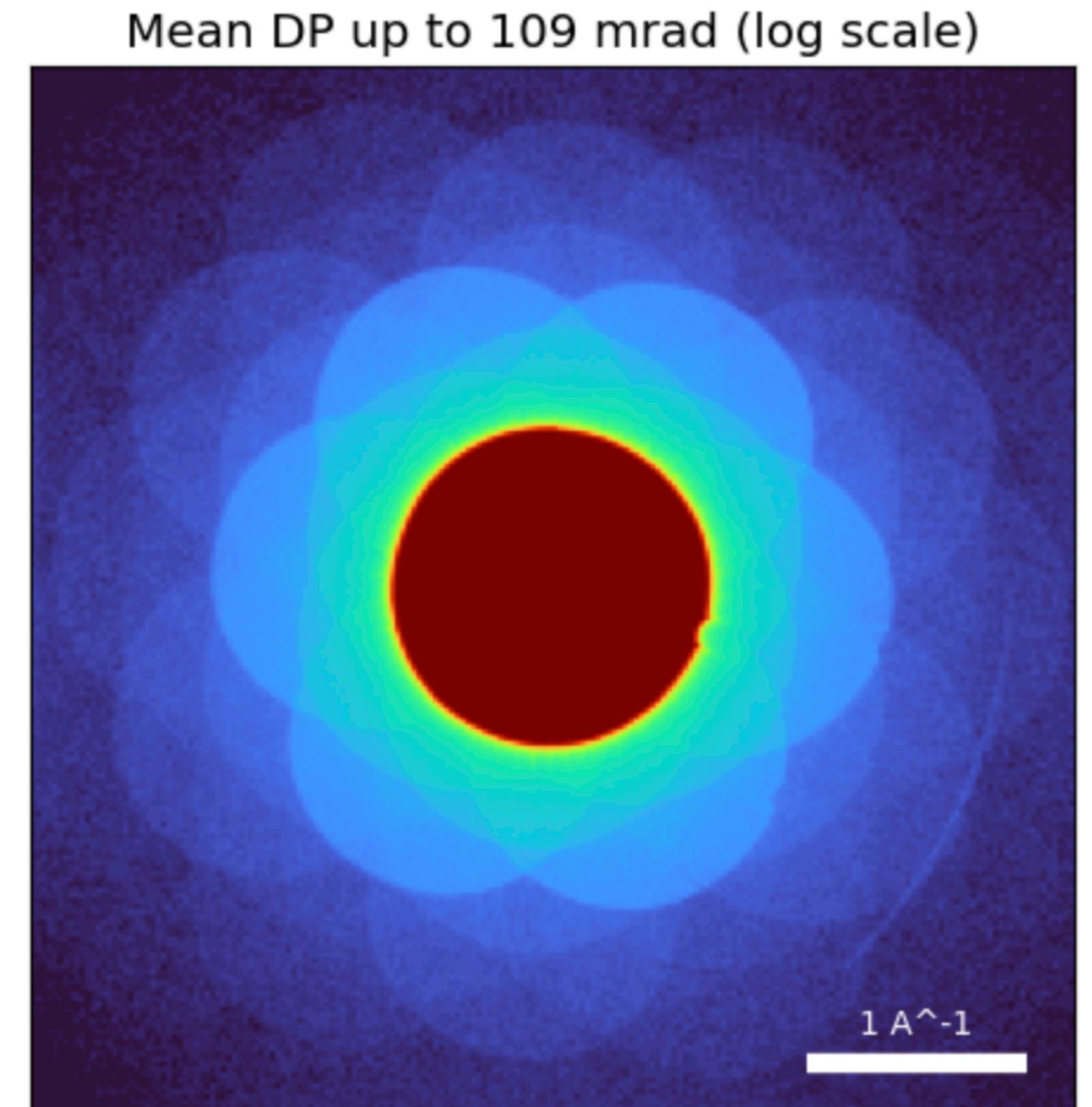
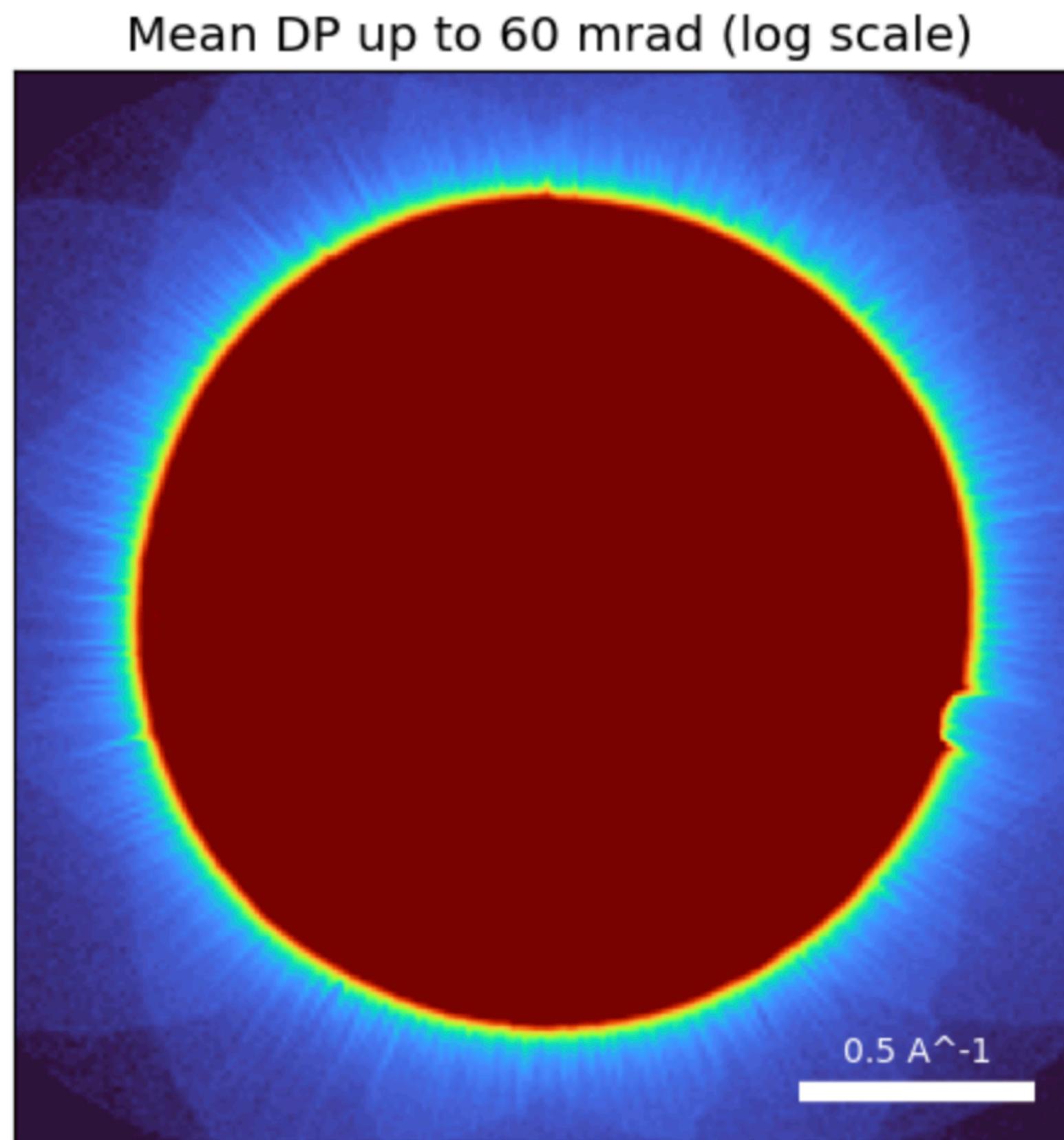
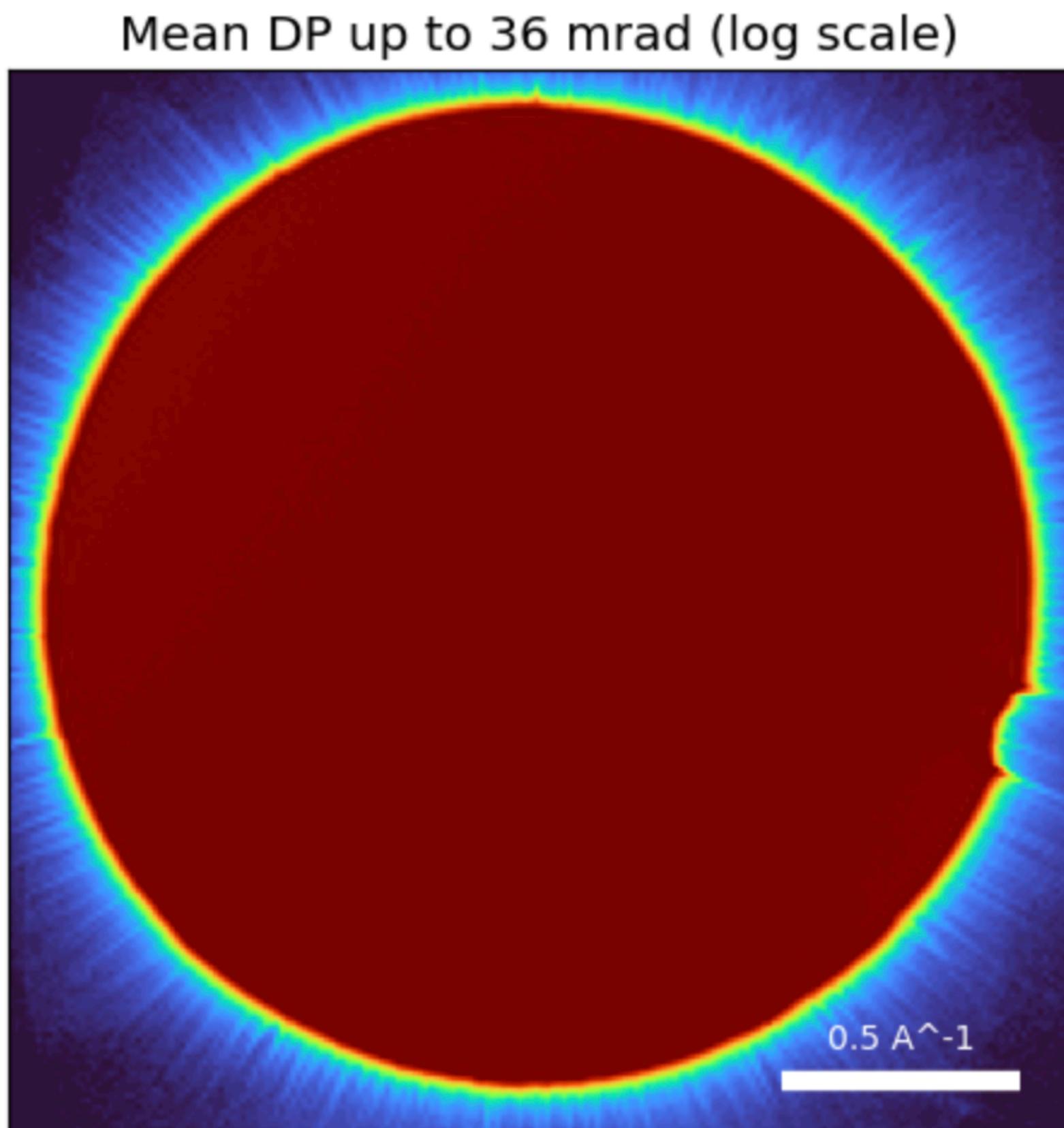
Nion + ARINA 4D-STEM data collection

Example: 512×512 px scan (R_{px}), 96×96 px CBED (Q_{px}), 100 us dwell time (8 pA)



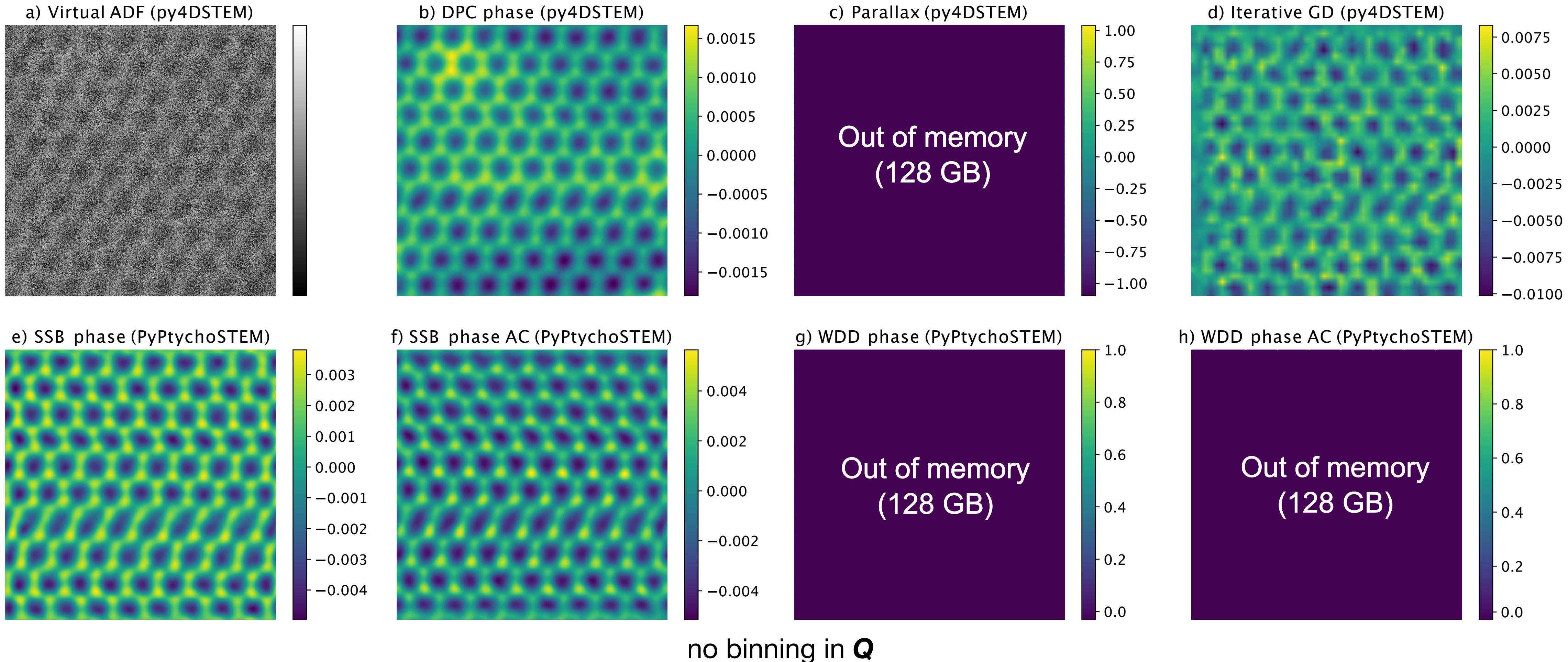
- 34 mrad semi-convergence angle, variable camera length (max collection angle)
- counting electrons: accurate dose (& dose per pixel can be normalized)

Nion + ARINA variable camera lengths



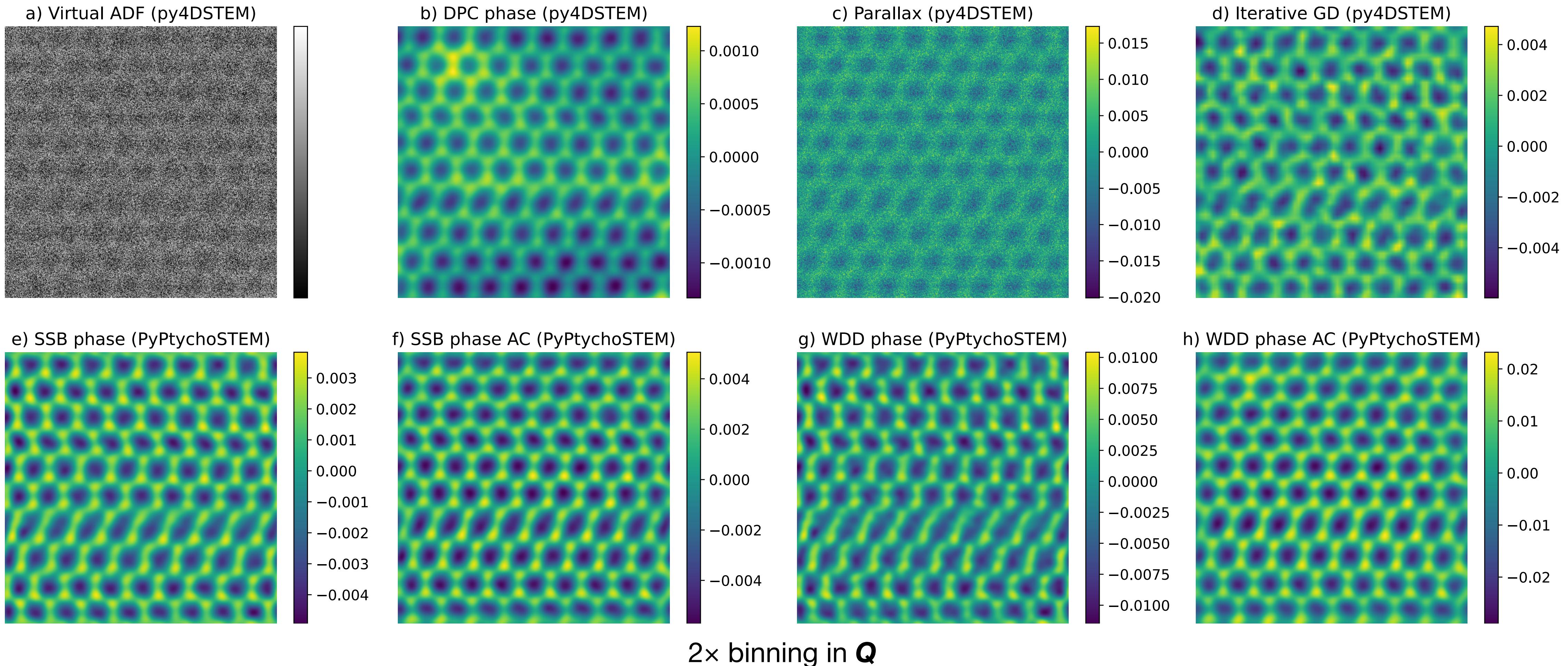
$\sim 200 \text{ pA current!}$

Comparing phase reconstruction algorithms (\mathbf{Q} binning)



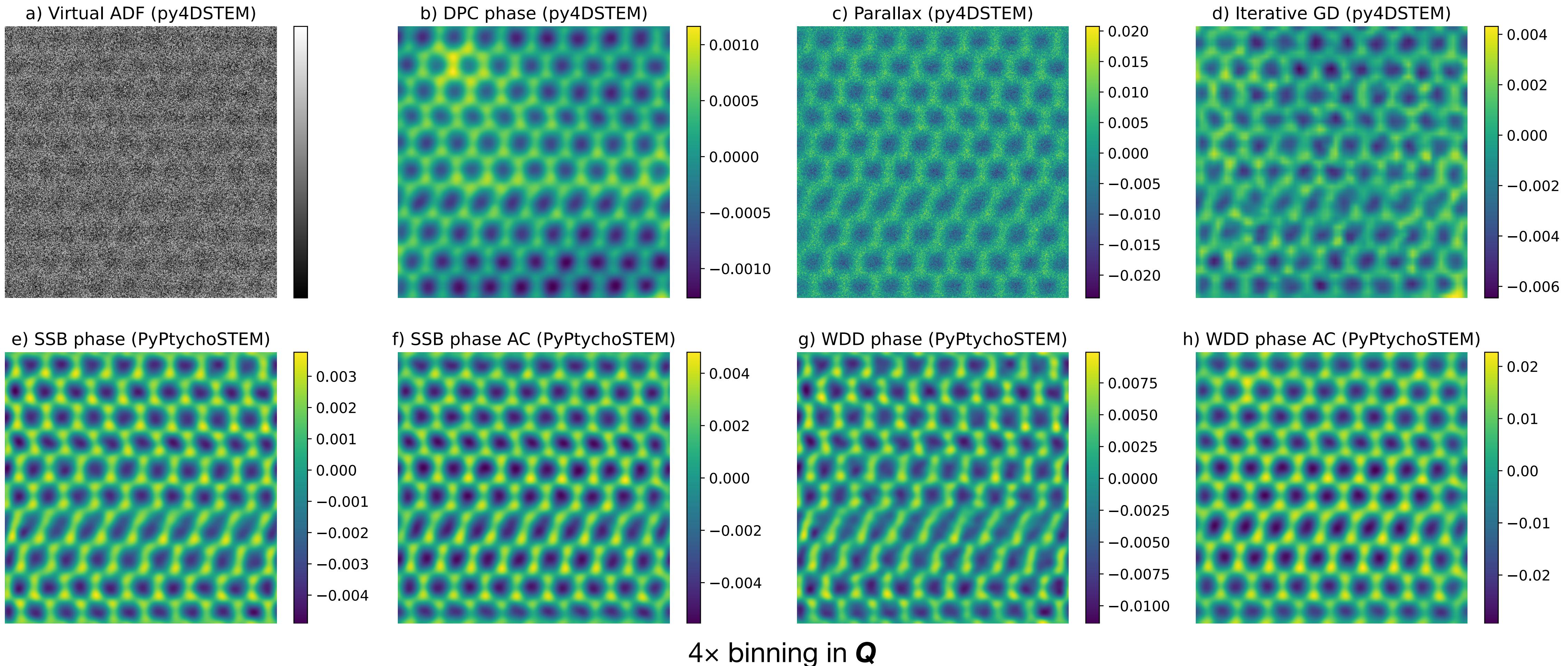
Binning in \mathbf{Q} : **36 mrad** max. scattering angle, 33.3 us dwell time, 512×512 \mathbf{R} px, 192×192 \mathbf{Q} px

Comparing phase reconstruction algorithms (\mathbf{Q} binning)



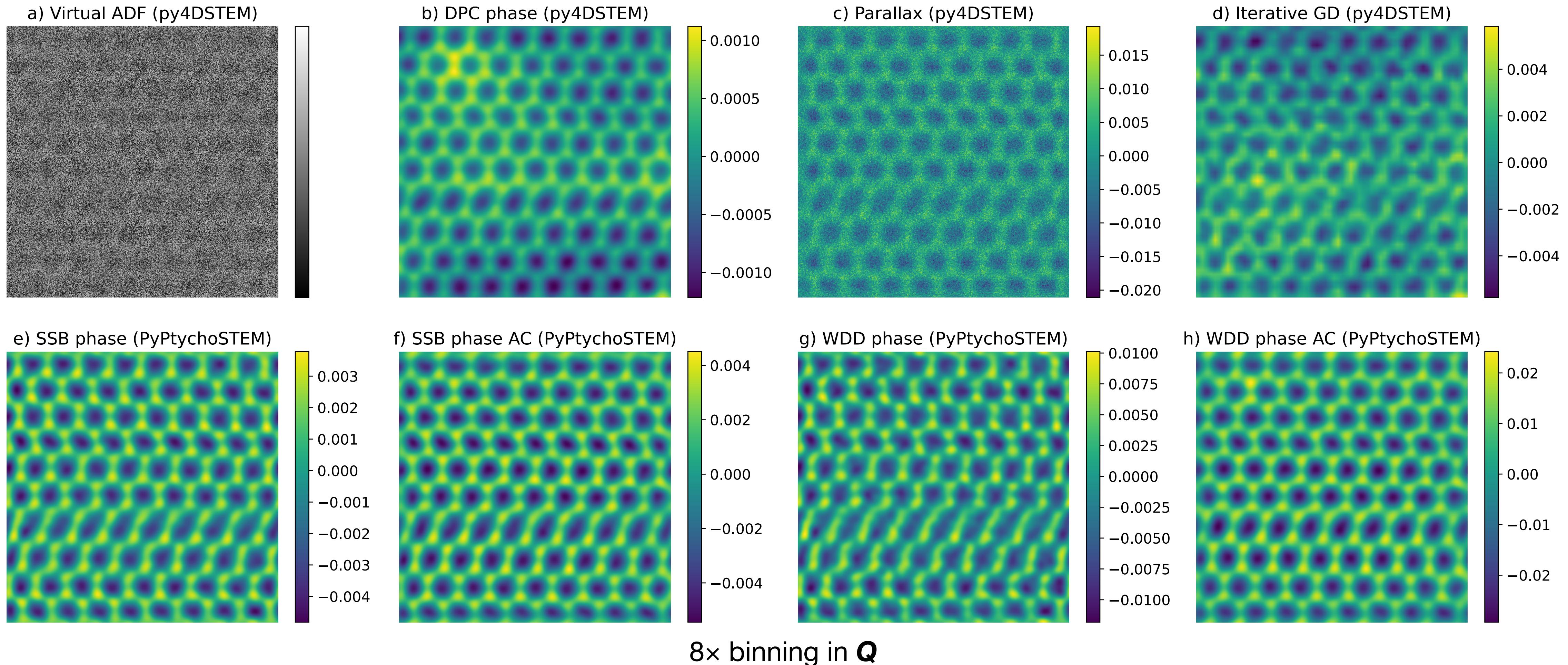
Binning in \mathbf{Q} : **36 mrad** max. scattering angle, 33.3 us dwell time, 512×512 \mathbf{R} px, 192×192 \mathbf{Q} px

Comparing phase reconstruction algorithms (\mathbf{Q} binning)



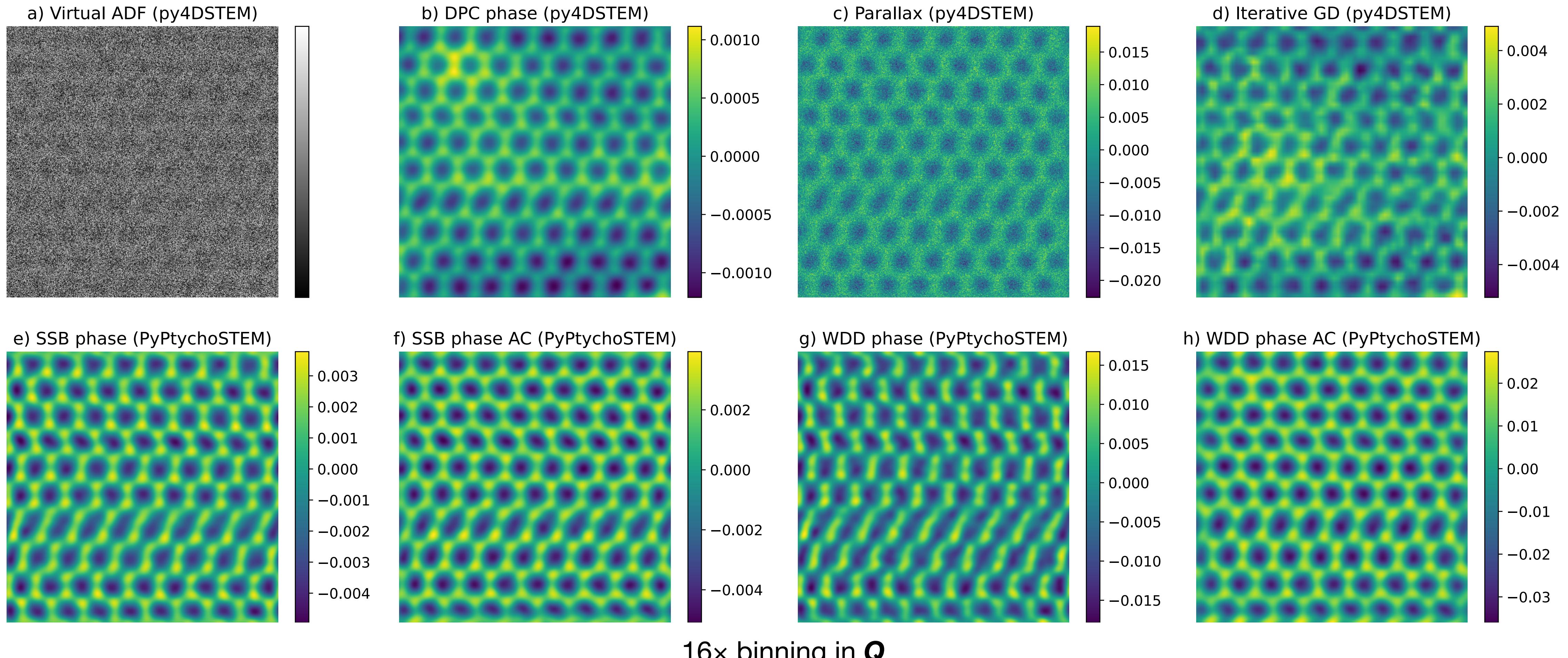
Binning in \mathbf{Q} : **36 mrad** max. scattering angle, 33.3 us dwell time, 512×512 \mathbf{R} px, 192×192 \mathbf{Q} px

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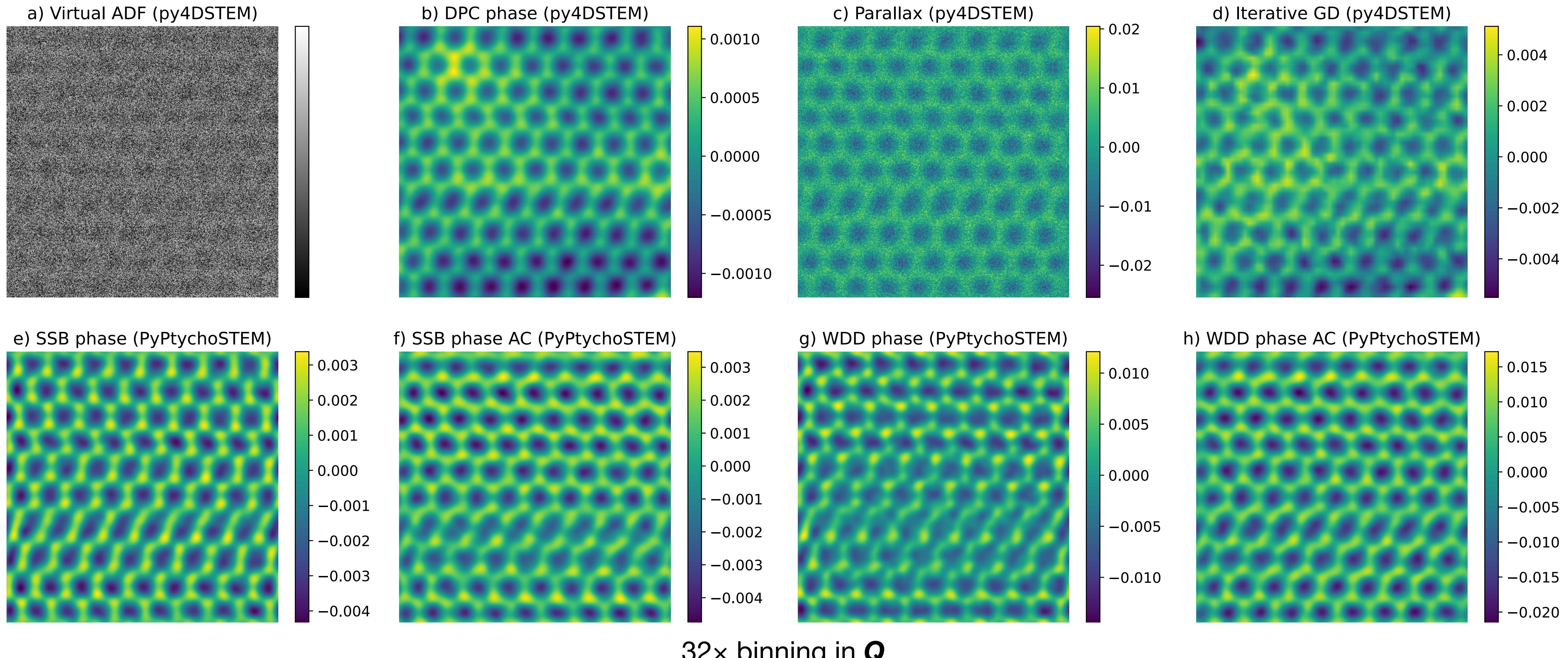
Binning in \mathbf{Q} : **36 mrad** max. scattering angle, 33.3 us dwell time, 512×512 \mathbf{R} px, 192×192 \mathbf{Q} px

Comparing phase reconstruction algorithms (\mathbf{Q} binning)



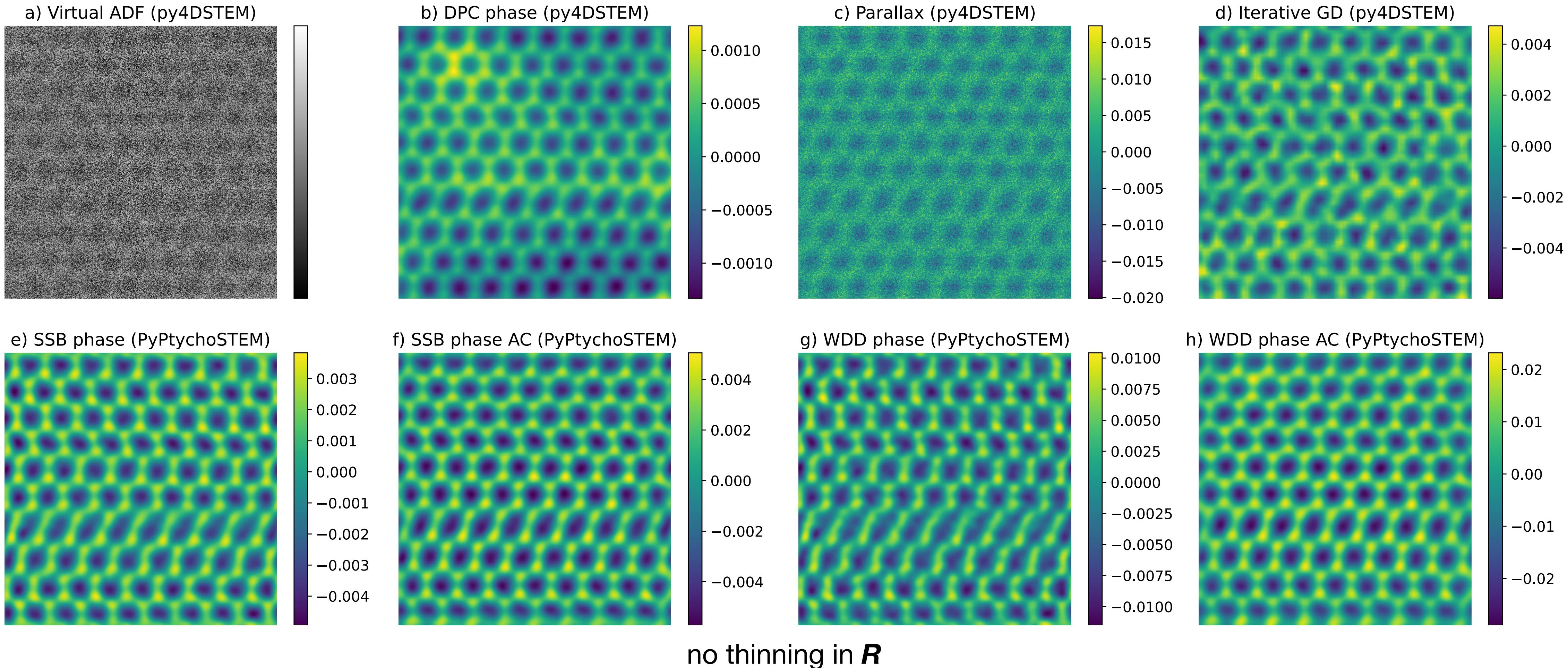
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Comparing phase reconstruction algorithms (\mathbf{Q} binning)



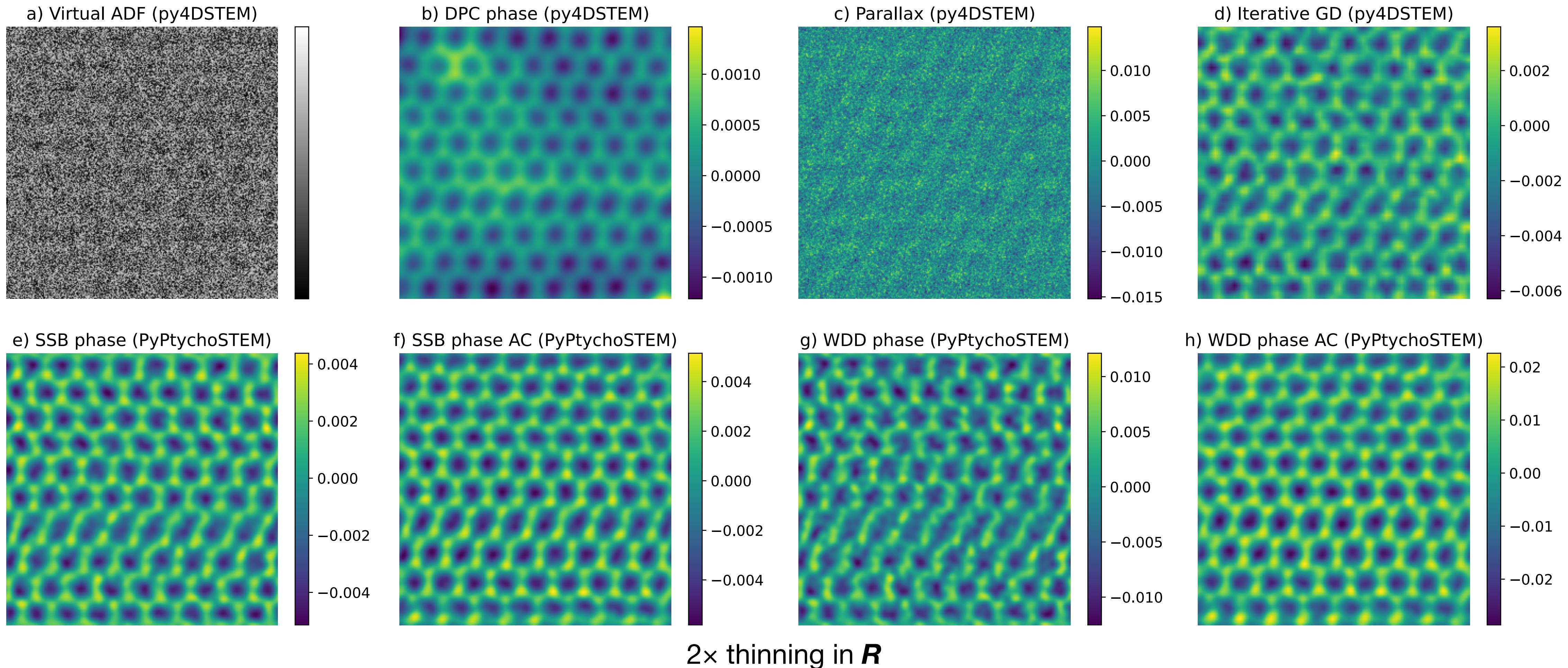
Binning in \mathbf{Q} : **36 mrad** max. scattering angle, 33.3 us dwell time, 512×512 \mathbf{R} px, 192×192 \mathbf{Q} px

Comparing phase reconstruction algorithms (R thinning)



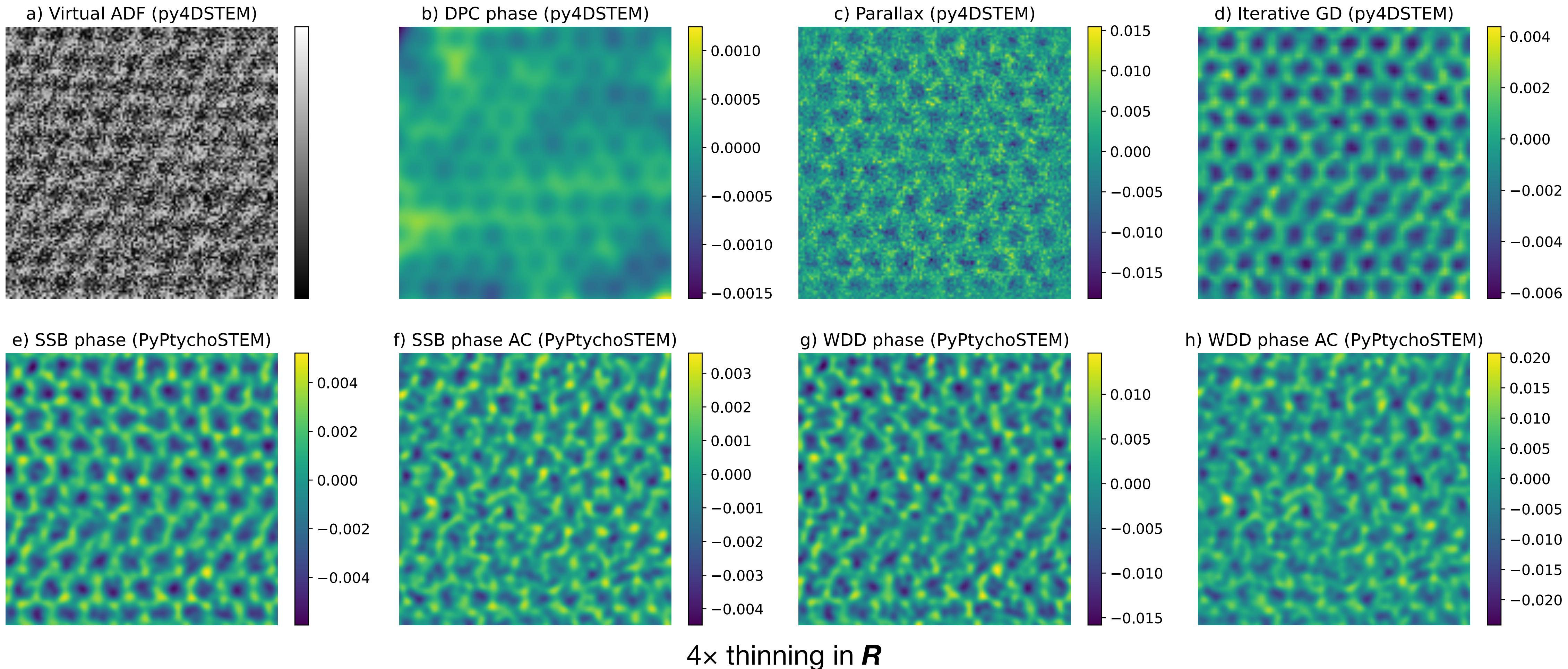
Thinning in R : **36 mrad** max. scattering angle, 33.3 us dwell time, 512×512 R px, 96×96 Q px (binned by 2)

Comparing phase reconstruction algorithms (R thinning)



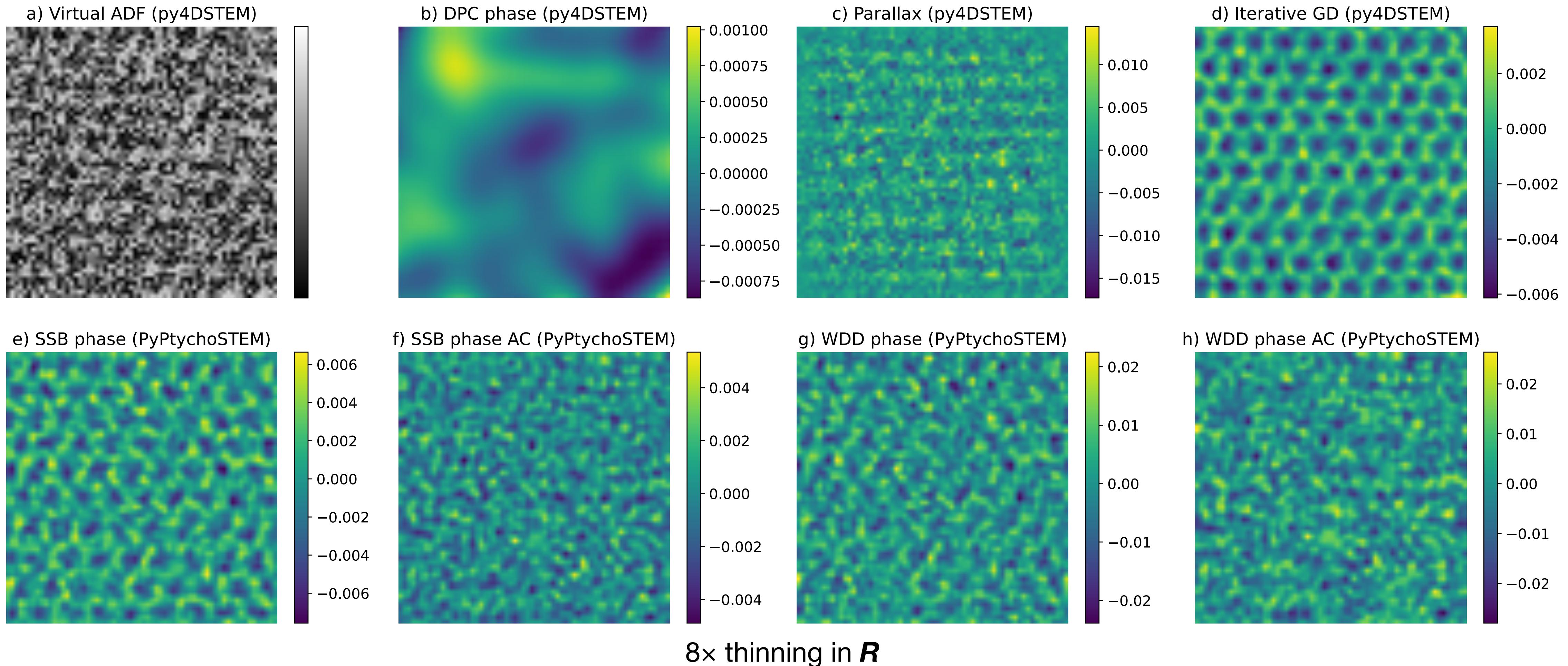
Thinning in R : **36 mrad** max. scattering angle, 33.3 us dwell time, 512×512 R px, 96×96 Q px (binned by 2)

Comparing phase reconstruction algorithms (R thinning)



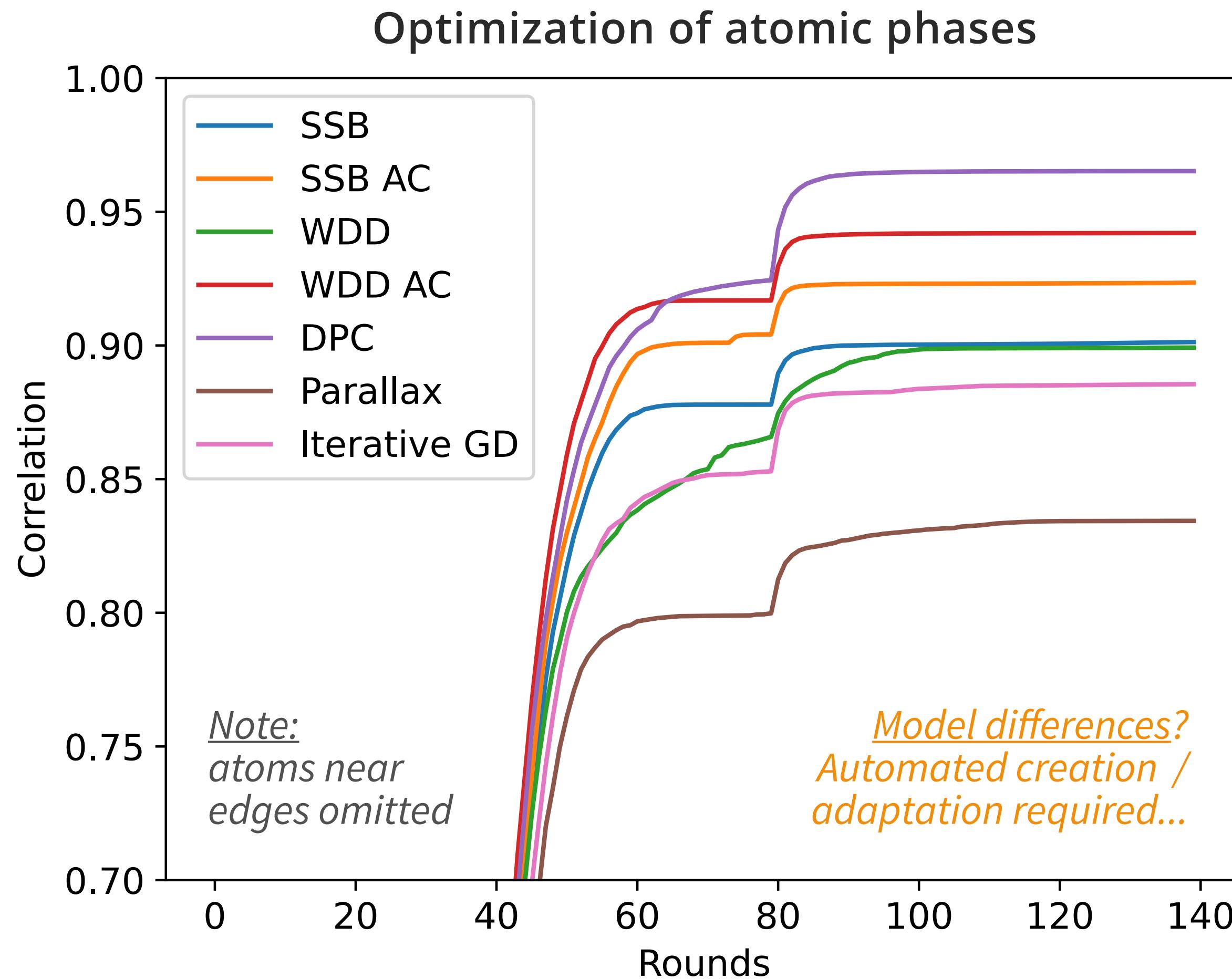
Thinning in R : **36 mrad** max. scattering angle, 33.3 us dwell time, 512×512 R px, 96×96 Q px (binned by 2)

Comparing phase reconstruction algorithms (R thinning)



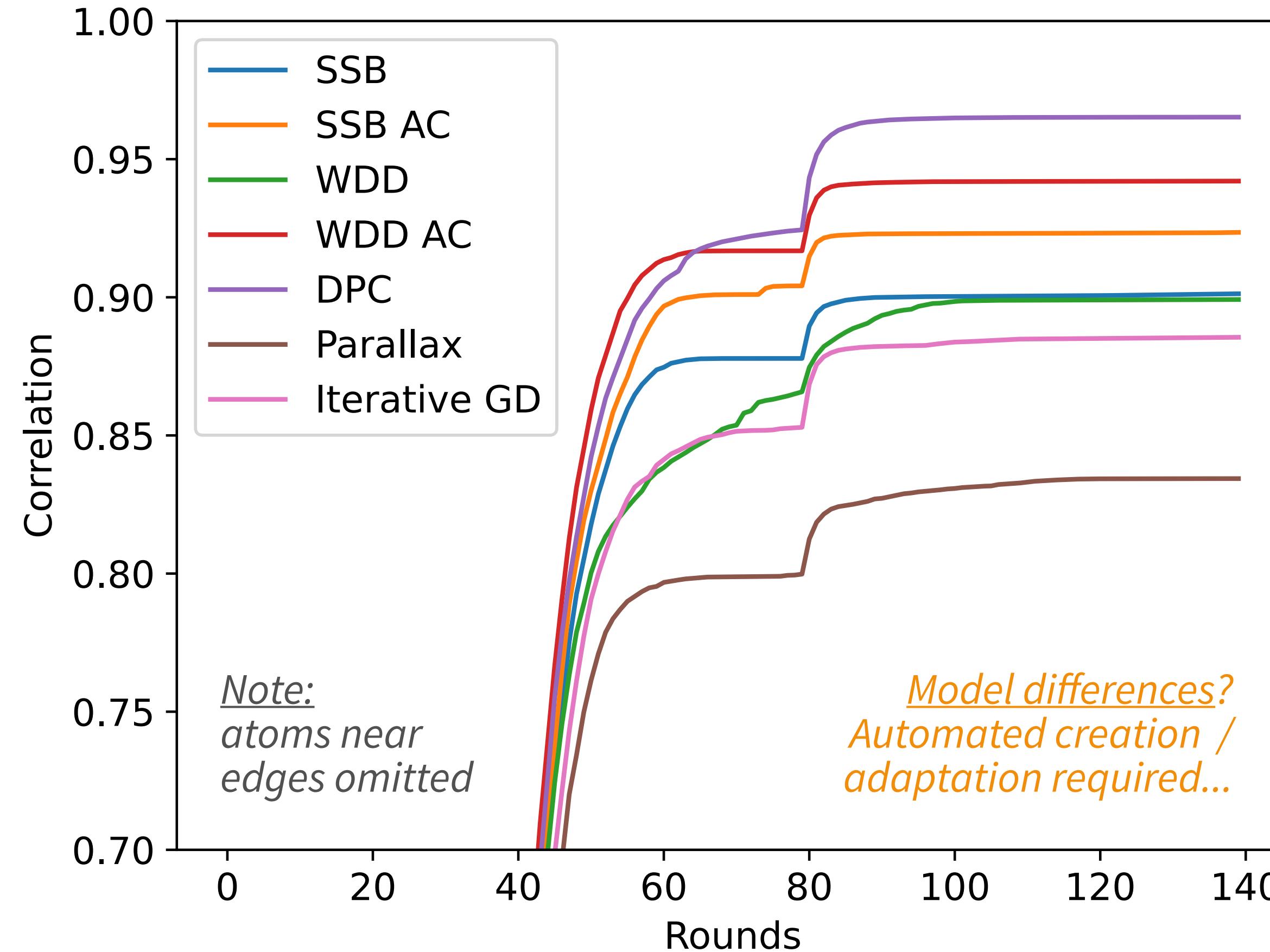
Thinning in R : **36 mrad** max. scattering angle, 33.3 us dwell time, 512×512 R px, 96×96 Q px (binned by 2)

Atomic phase assignment

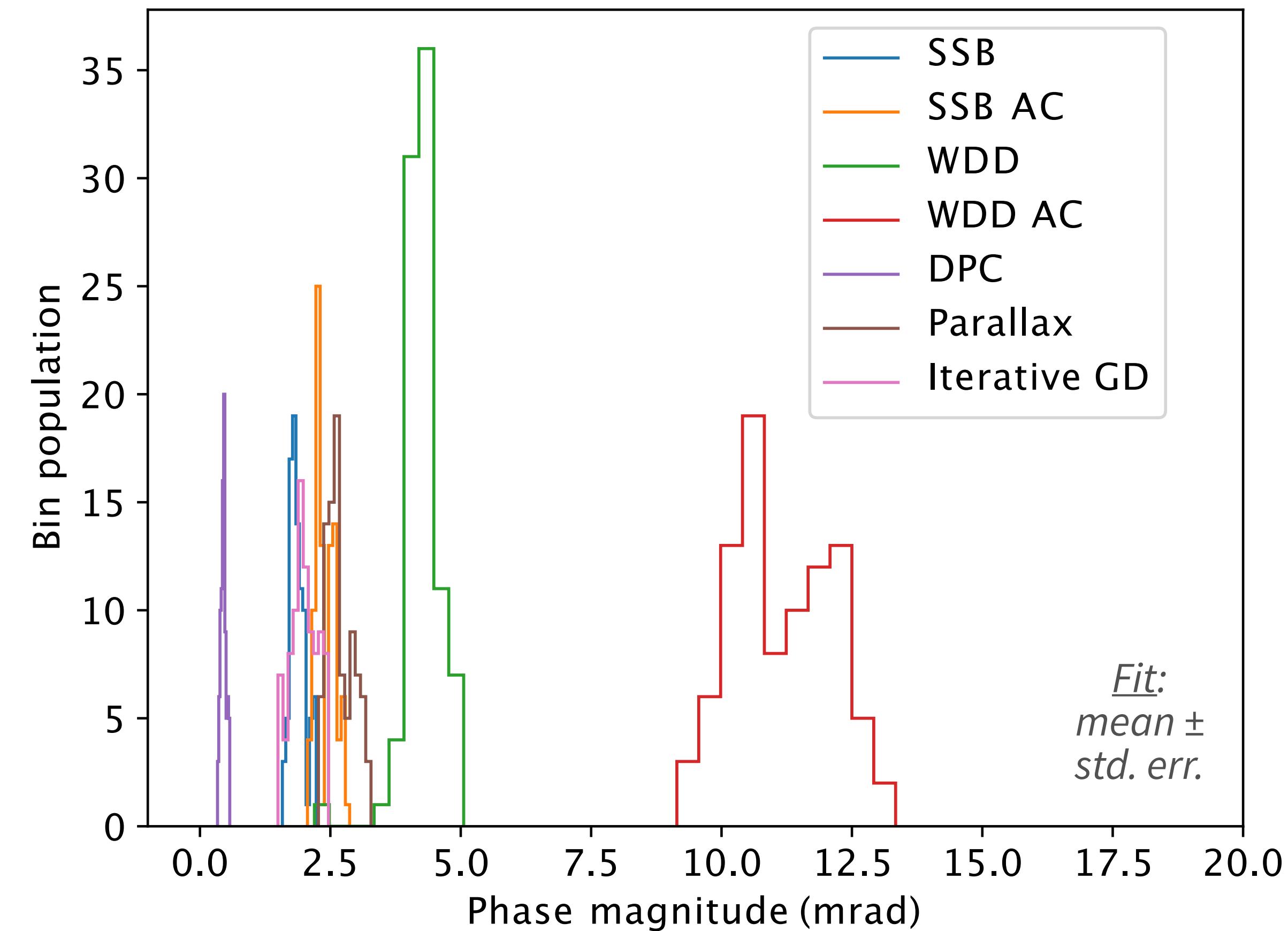


Atomic phase assignment

Optimization of atomic phases

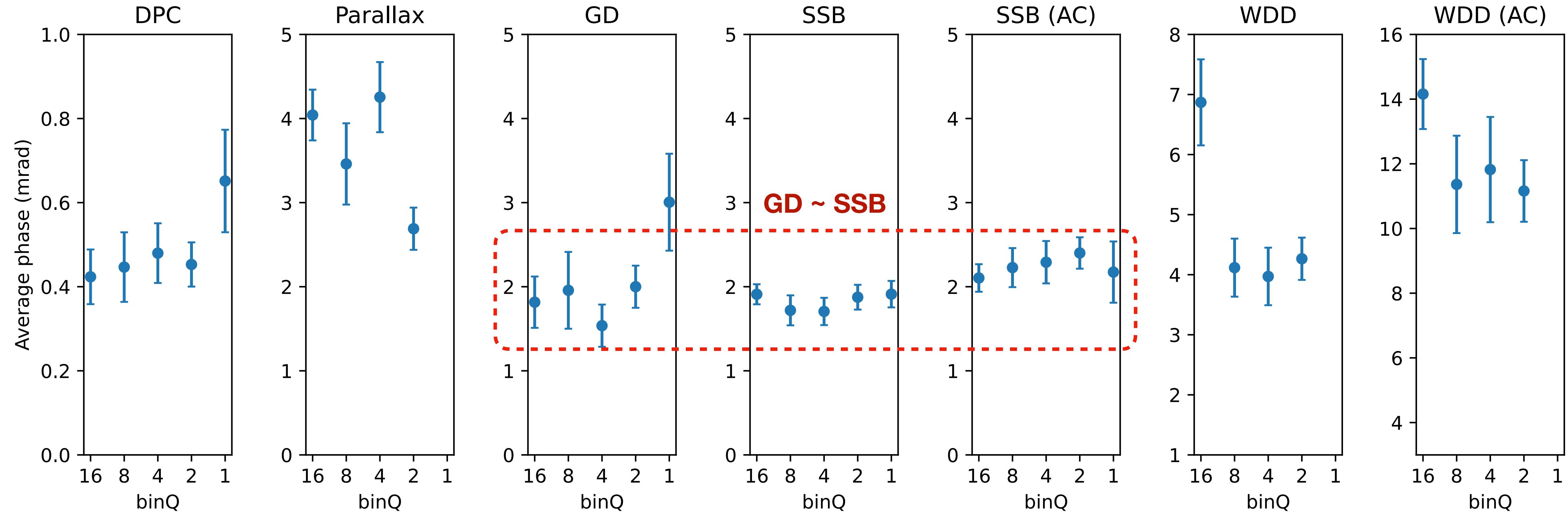


Phase variation over graphene C atoms



Quantification of phase variation

Note: different y-scales!

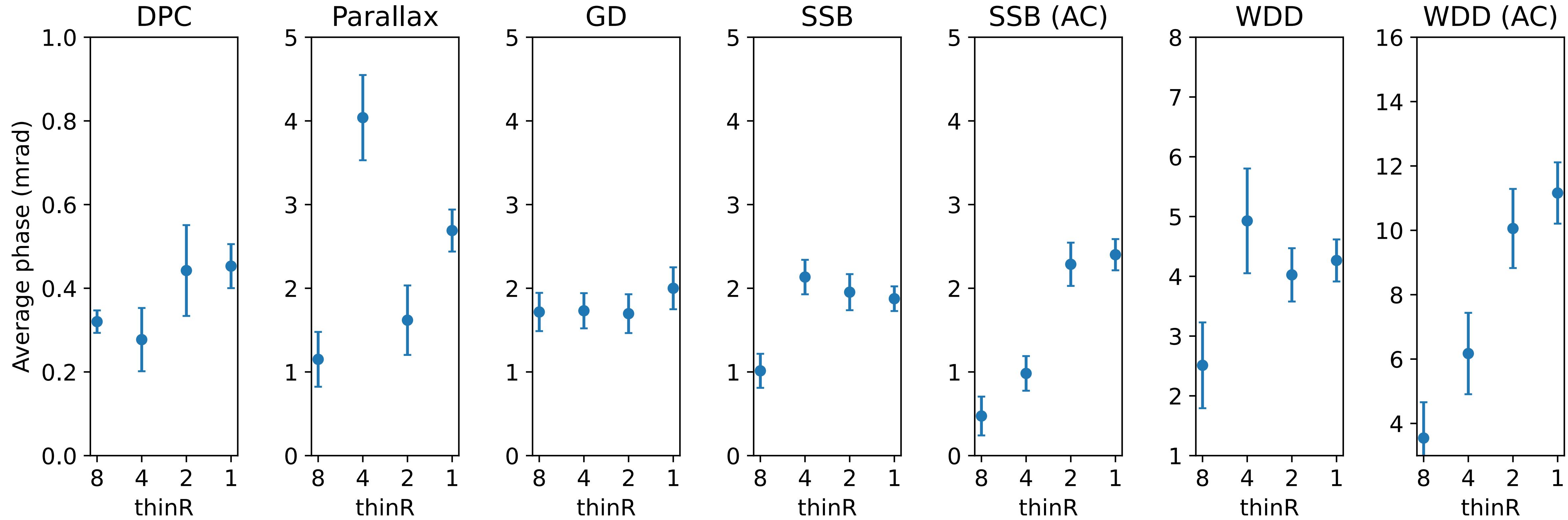


Plot:
mean \pm
std. err.

Binning in **Q**: 36 mrad max. scattering angle, 33.3 us dwell time, 512×512 **R** px, 192×192 **Q** pix

Quantification of phase variation

Note: different y-scales!

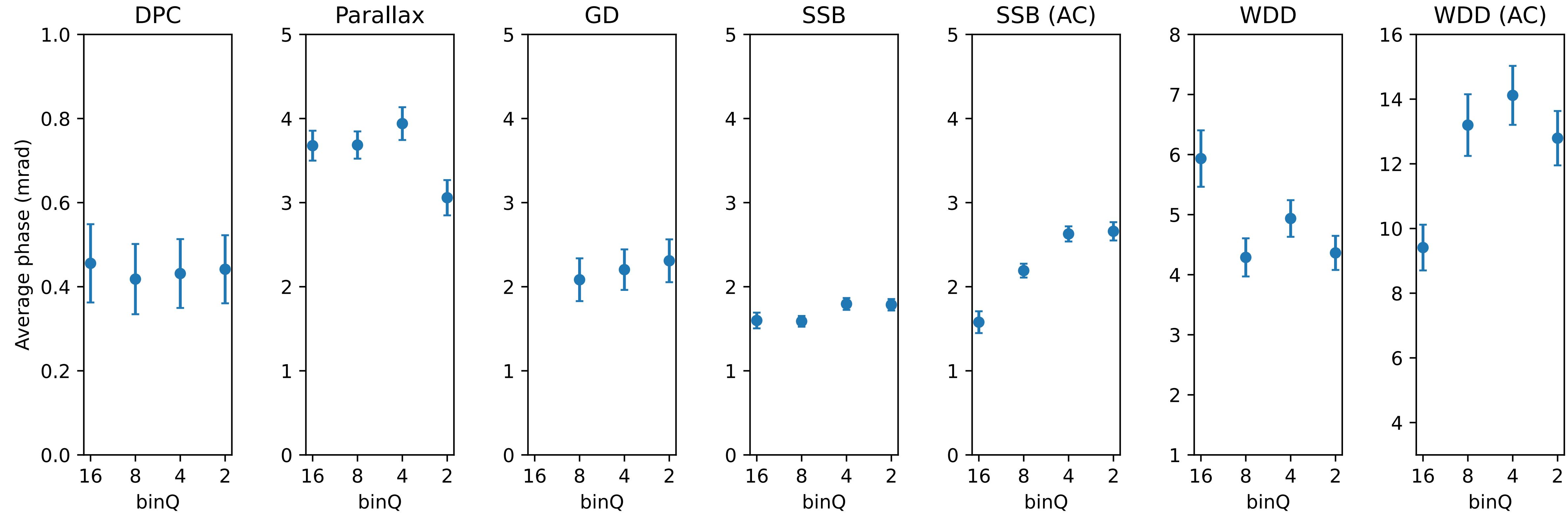


Plot:
mean \pm
std. err.

Thinning in R : 36 mrad max. scattering angle, 33.3 us dwell time, 512×512 R px, 96×96 Q pix

Quantification of phase variation

Note: different y-scales!

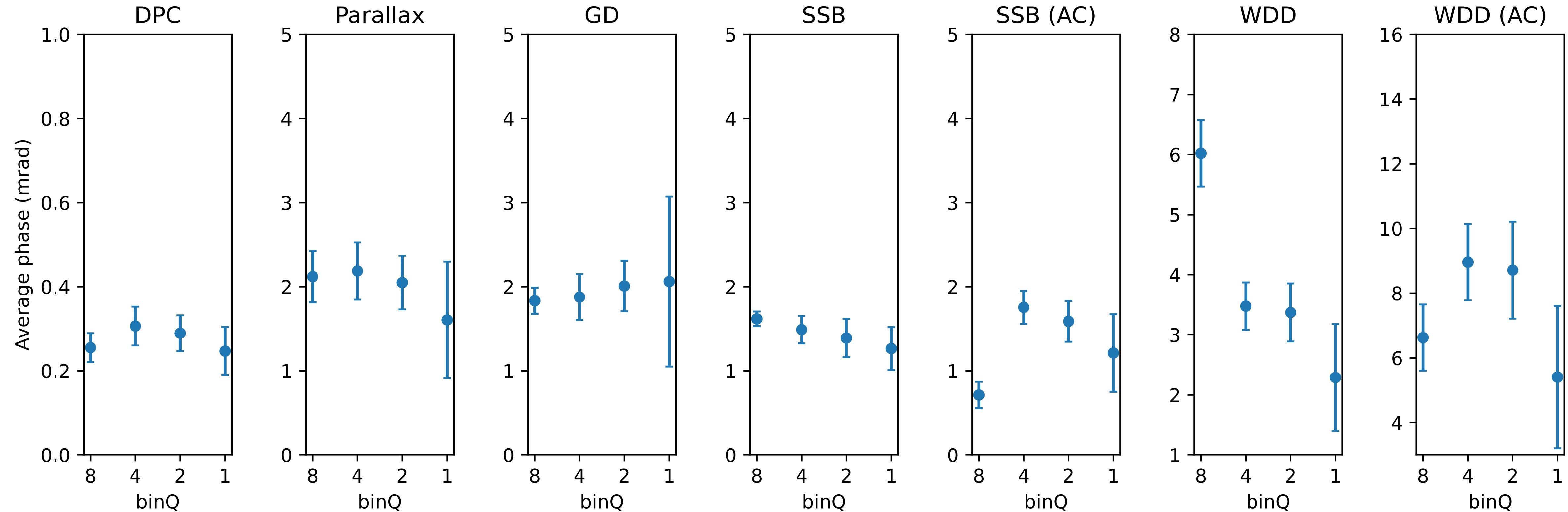


Plot:
mean \pm
std. err.

Binning in **Q**: **60 mrad** max. scattering angle, 33.3 us dwell time, 512×512 **R** px, 192×192 **Q** pix

Quantification of phase variation

Note: different y-scales!



Plot:
mean \pm
std. err.

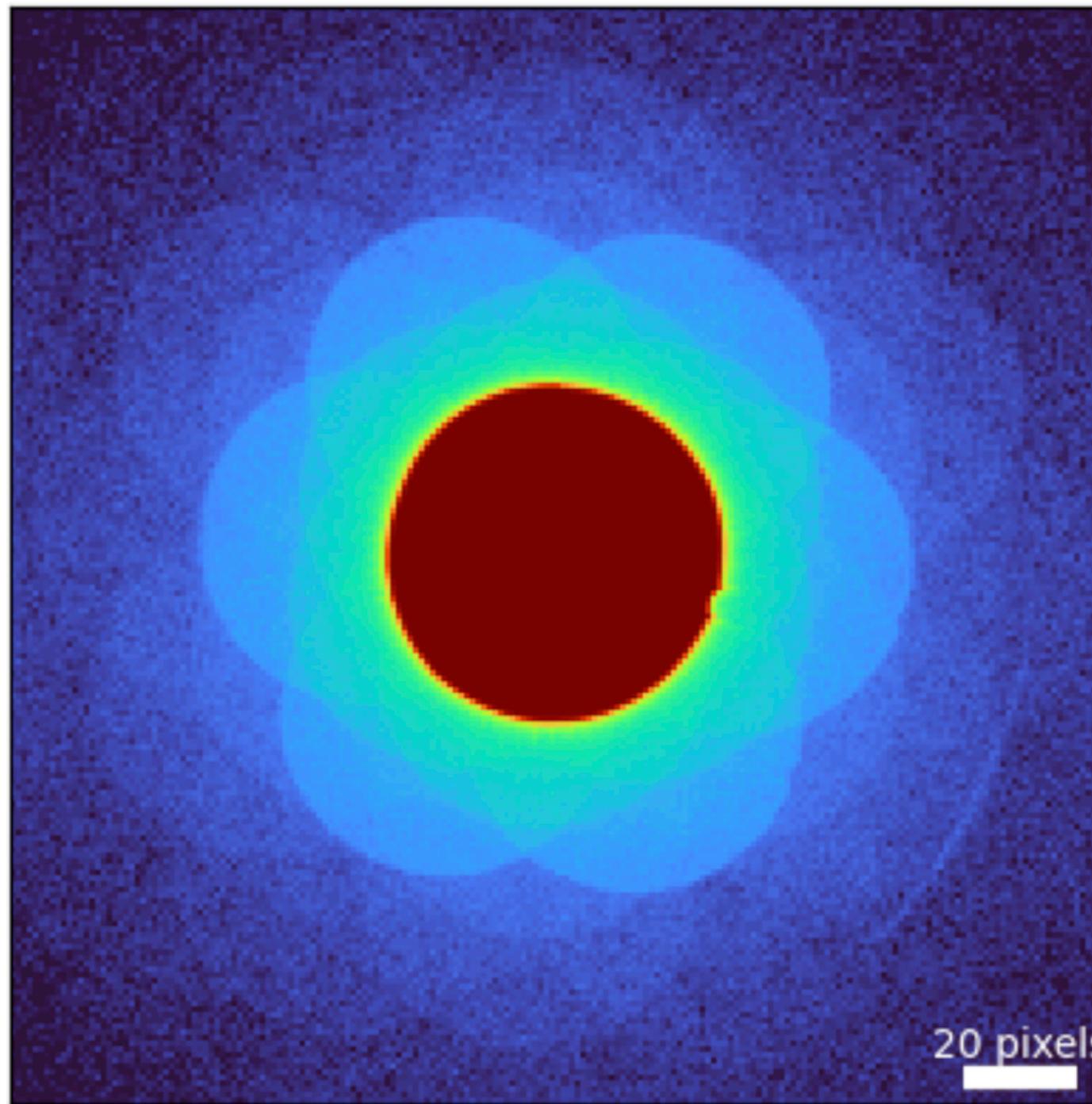
Binning in **Q**: 109 mrad max. scattering angle, 100 us dwell time, 256×256 **R** px, 192×192 **Q** pix

Computational cost (36 mrad datasets)

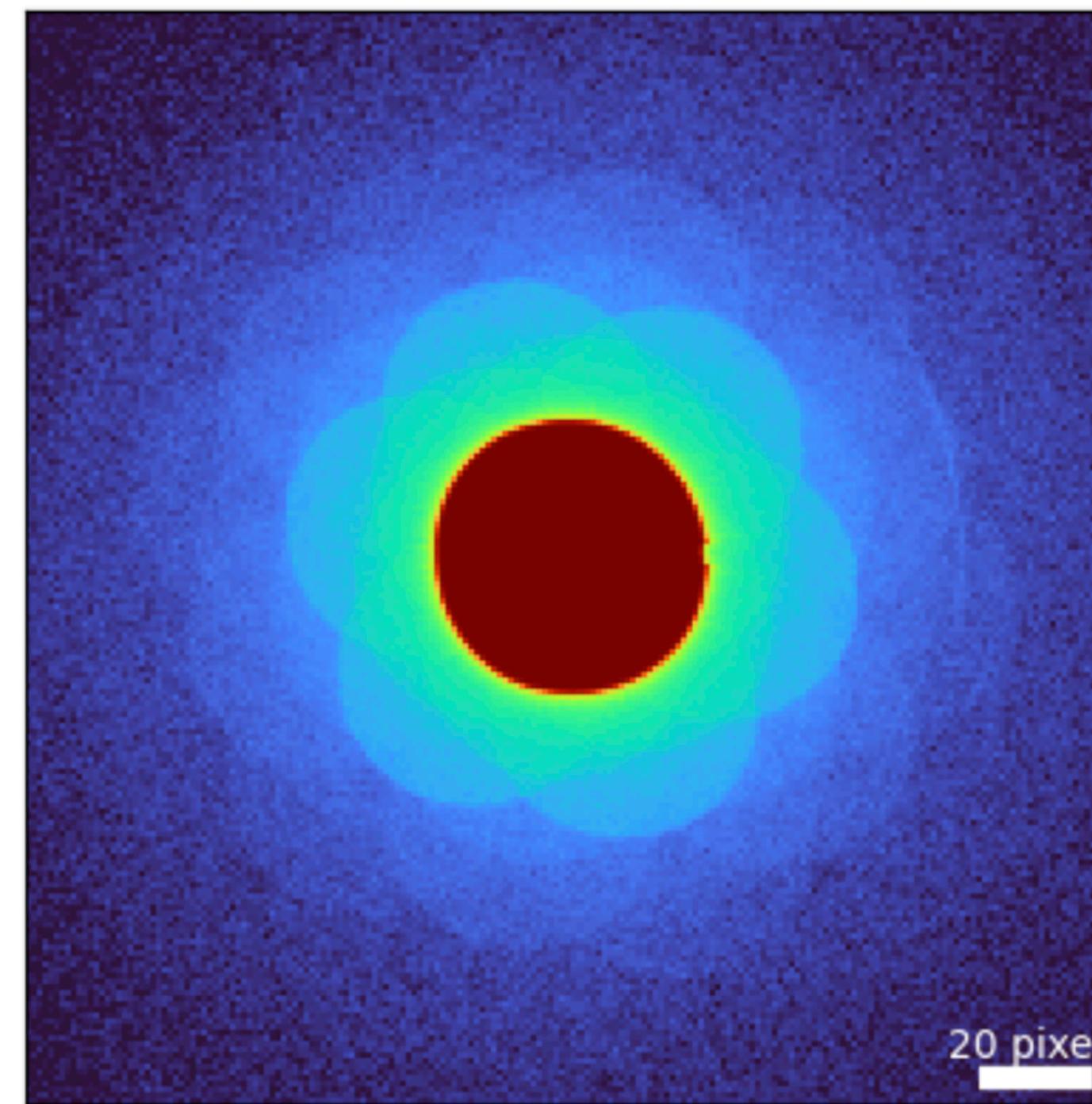
Scan (R px)	Bin in Q	CBED (Q px)	Array (MB)	DPC (s)	Parallax [†] (s)	Iter. GD‡ (s)	SSB (s)	SSB AC (s)	WDD (s)	WDD AC (s)
512×512	1	192×192	38656	16	–	578	236	18097	–	–
	2	96×96	9664	6.1	855	517	59	888	1938	2375
	4	48×48	2416	6.9	728	195	34	272	714	784
	8	24×24	604	5.4	68	146	13	87	306	317
	16	12×12	151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

Conclusions & future prospects

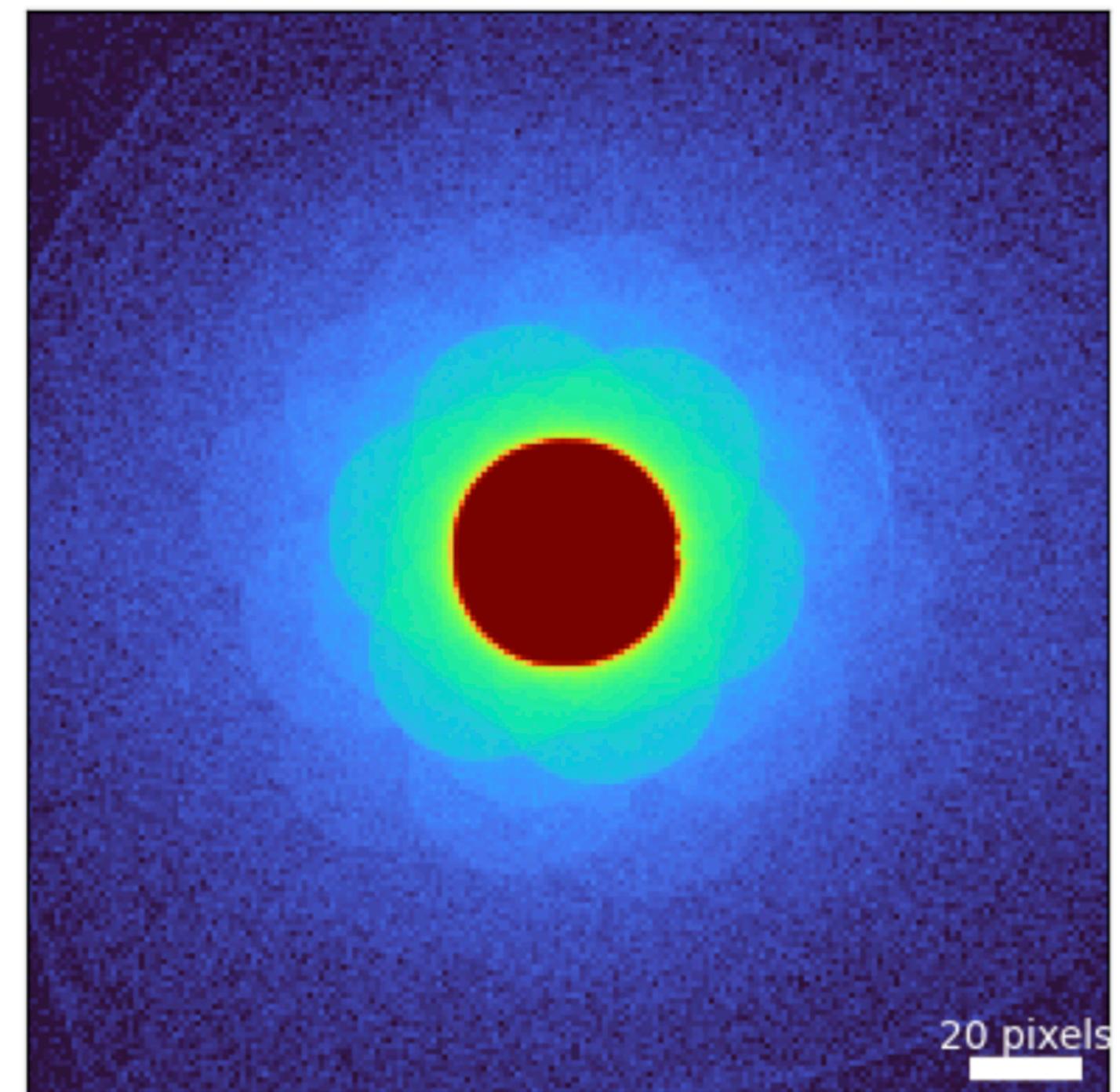
Mean DP up to 109 mrad (log scale)



Mean DP up to 136 mrad (log scale)



Mean DP up to 168 mrad (log scale)



- Quantitative absolute phase values?
 - SSB and GD seem to ~agree (WDD?!)
 - GD good even for focused (& fast on GPU)
- Super-resolution iterative ptycho up to $\sim 4 \times$ alpha
- Live virtual detectors / (semi-)live SSB?
- Structure & charge transfer in various materials...

Upcoming Japan stay (Sep 2024 – Feb 2025)



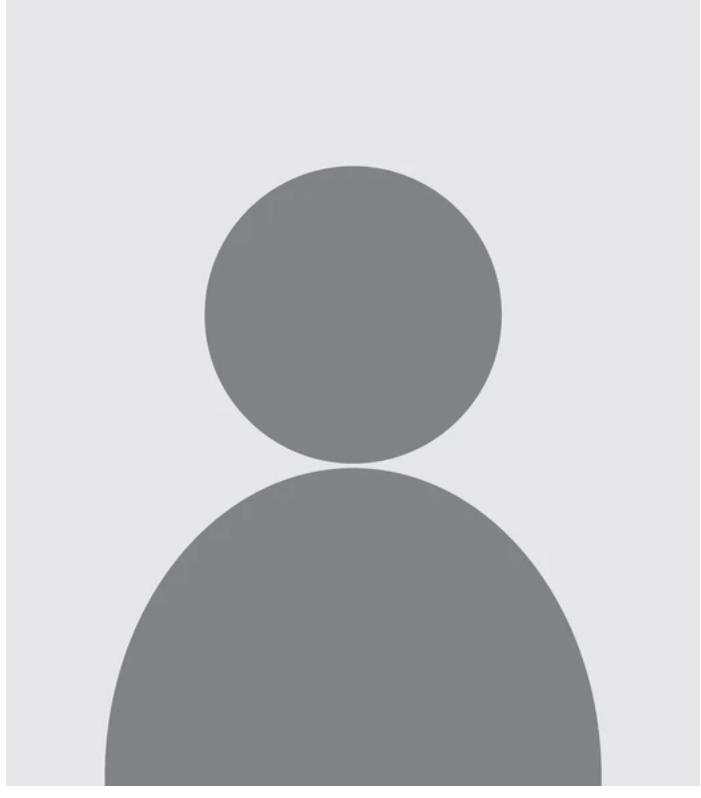
Nagashima (TUS, Sep)



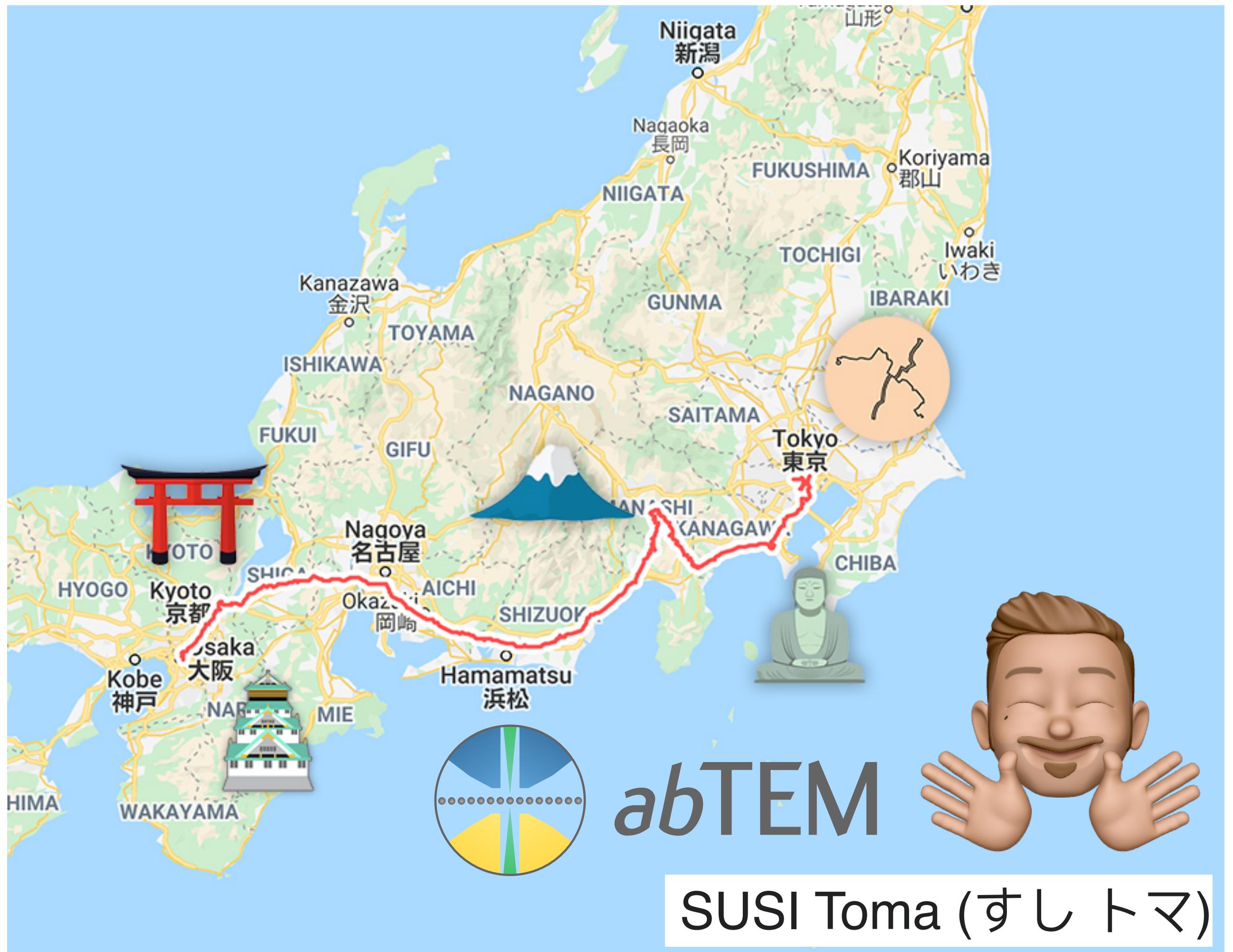
Suenaga (Osaka, Oct)



Shibata (Tokyo, Nov–)



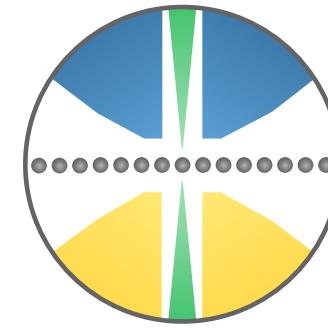
you?





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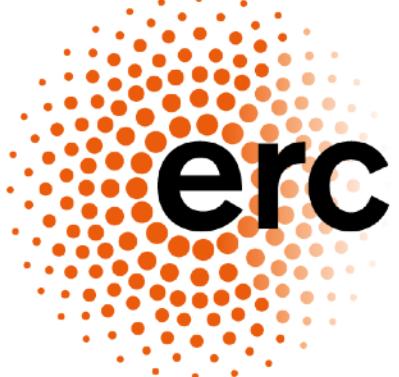
Φnm



abTEM

FWF

Der Wissenschaftsfonds.



Thank you!

Questions, collaborations: *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>

