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

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Niklas Dellby, Russ Hayner, Tracy Lovejoy, Andreas Mittelberger, Benjamin Plotkin-Swing (Nion Co. R&D / Bruker AXS)

Open-source software

Language

Python
(3.11.9)

Data Acquisition

Nion Swift
(0.16.10)

Phase Reconstructions

PyPtychoSTEM
(0.14.16)

py4DSTEM
(0.14.16)

Reconstruction Algorithms:

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

Stephanie Ribet

Georgios Varnavides

SSB & WDD: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167
DPC, Parallax, Iterative GD: G. Varnavides *et al.*, arXiv:2309.05250v2 (2024)

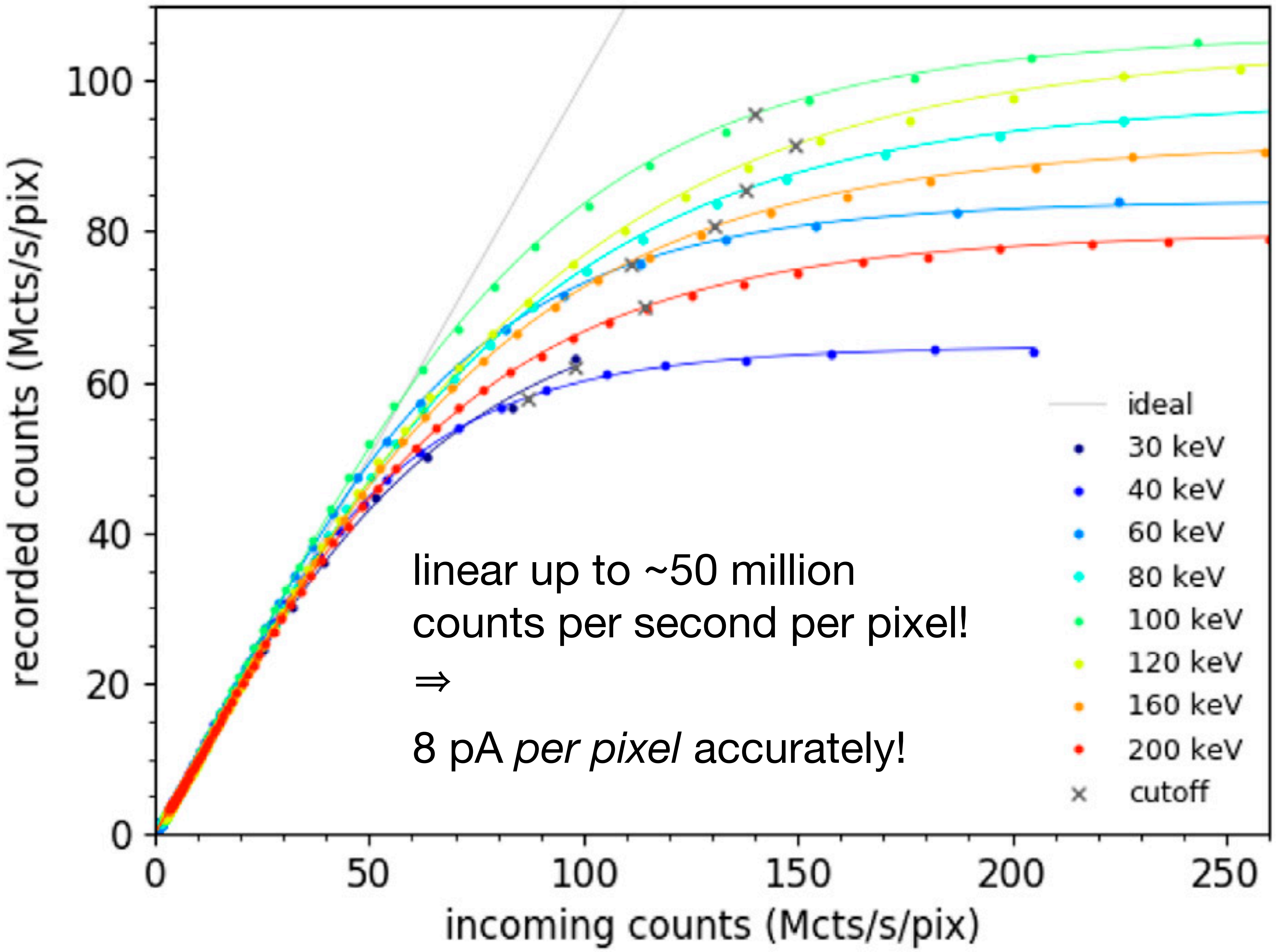
Near-ideal direct-electron 4D-STEM

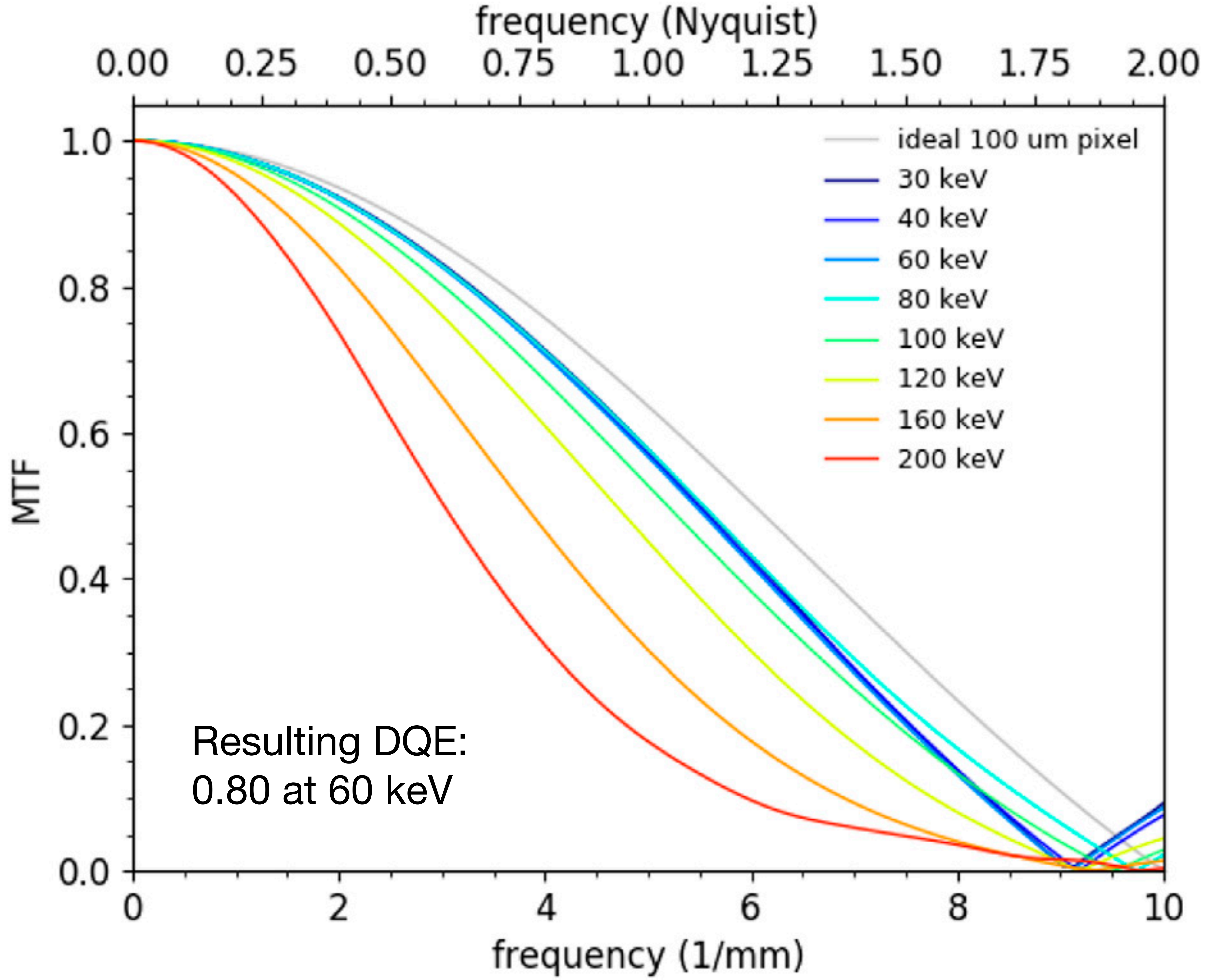
Number of pixels (W x H)	**192 × 192**
Active area (W x H) [mm²]	20 × 20
Pixel size (W x H) [µm²]	100 × 100
Sensor material	Silicon (Si) or high-Z
Energy range [keV]	30–300
Frame rate (max.) [Hz]	**120,000**
Count rate (max.) [el/s/pixel]	10^8
Detective Quantum Efficiency, DQE(0)	0.82 @ 80 keV 0.75 @ 200 keV, 0.75 @ 300 keV
Detector mounting	Retractable

Near-ideal 4D-STEM performance at 60 keV

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

Near-ideal direct-electron 4D-STEM





P. Zambon *et al.*, Front. Phys. 11:1308321 (2023)

4D-STEM data volumes

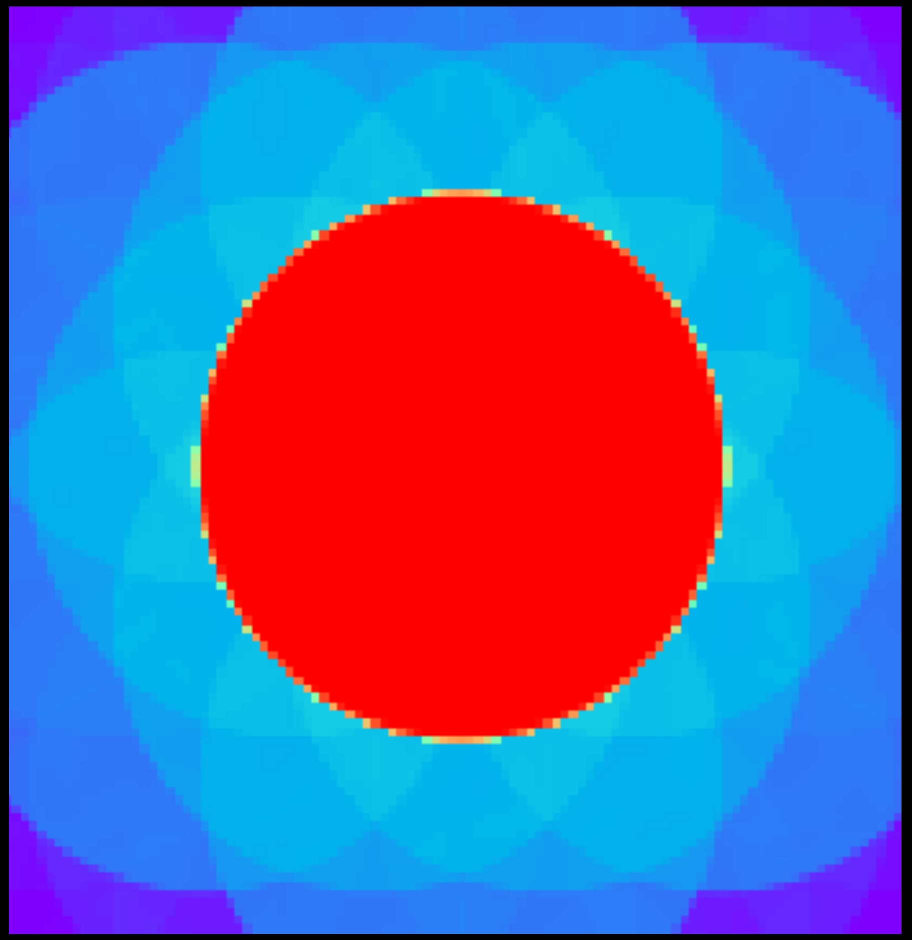
Data volumes collected during typical 4D-STEM experiments (assuming a 512 × 512 px scan).

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

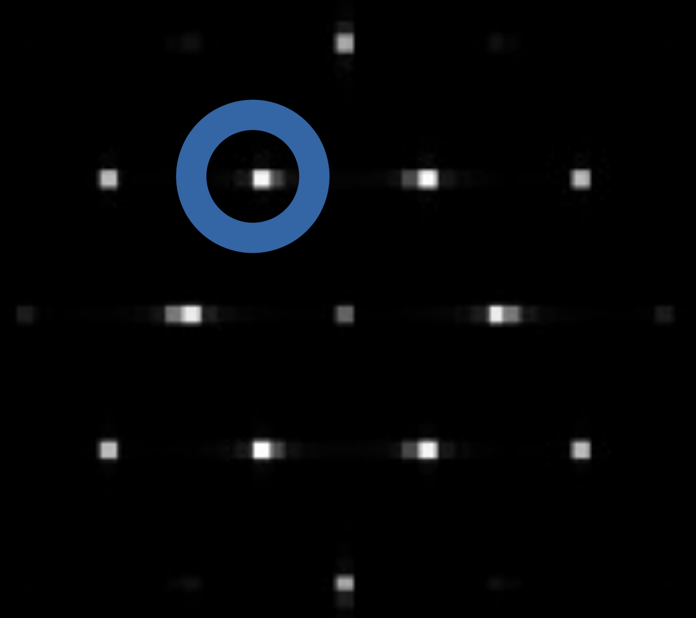
- Huge data volumes can be collected in seconds (challenge for storage, processing…)

Ptychography can correct aberrations post-acquisition

PACBED

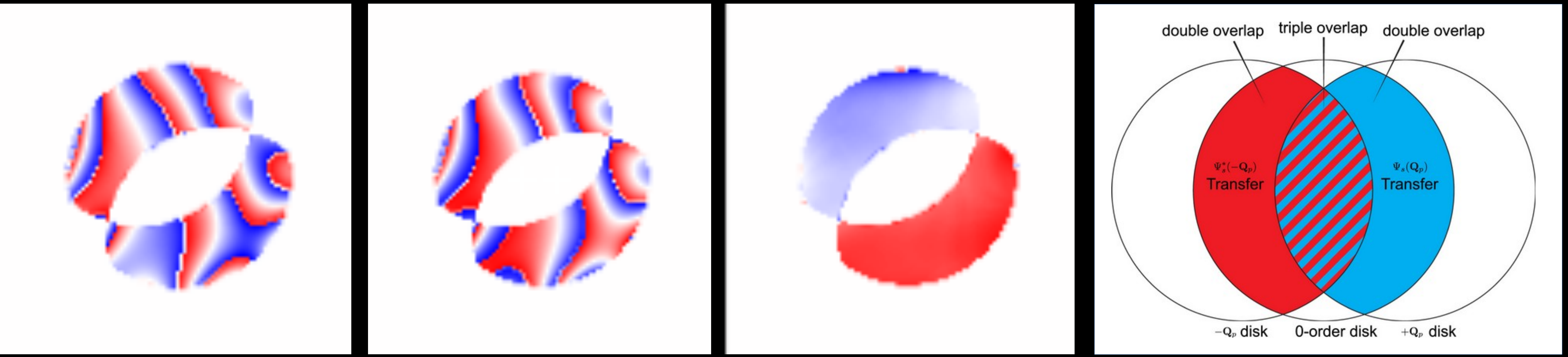


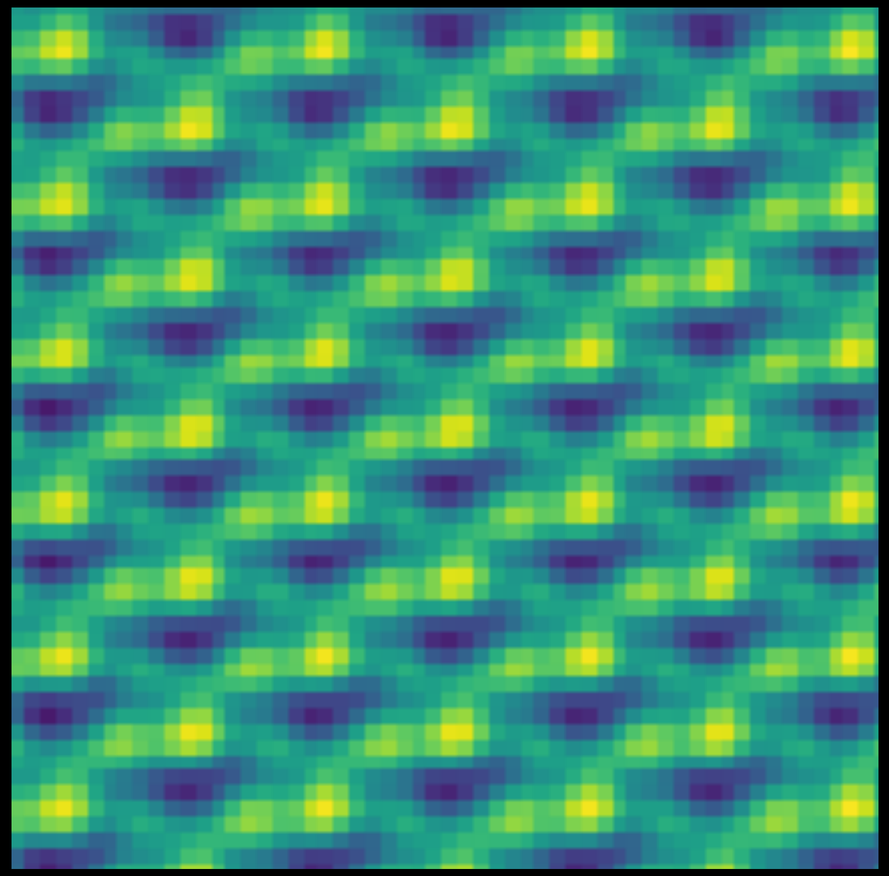
Spatial frequencies

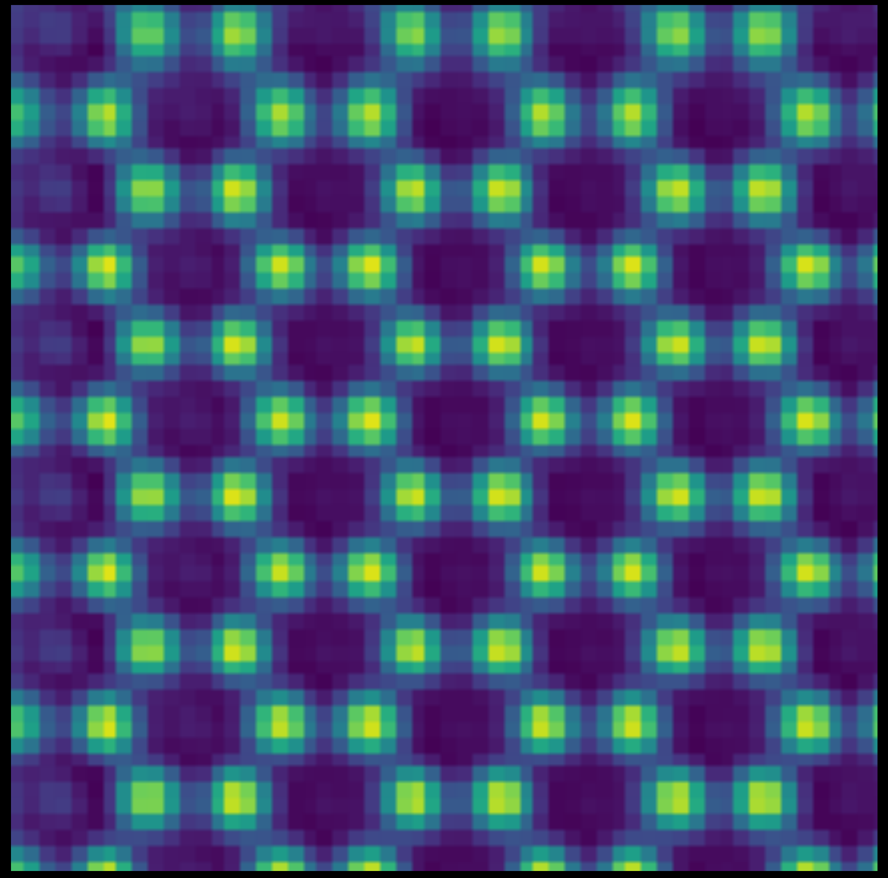


A(K)*A(K-Q)

'Exp' SVD corrected Theory







SVD = singular value decomposition

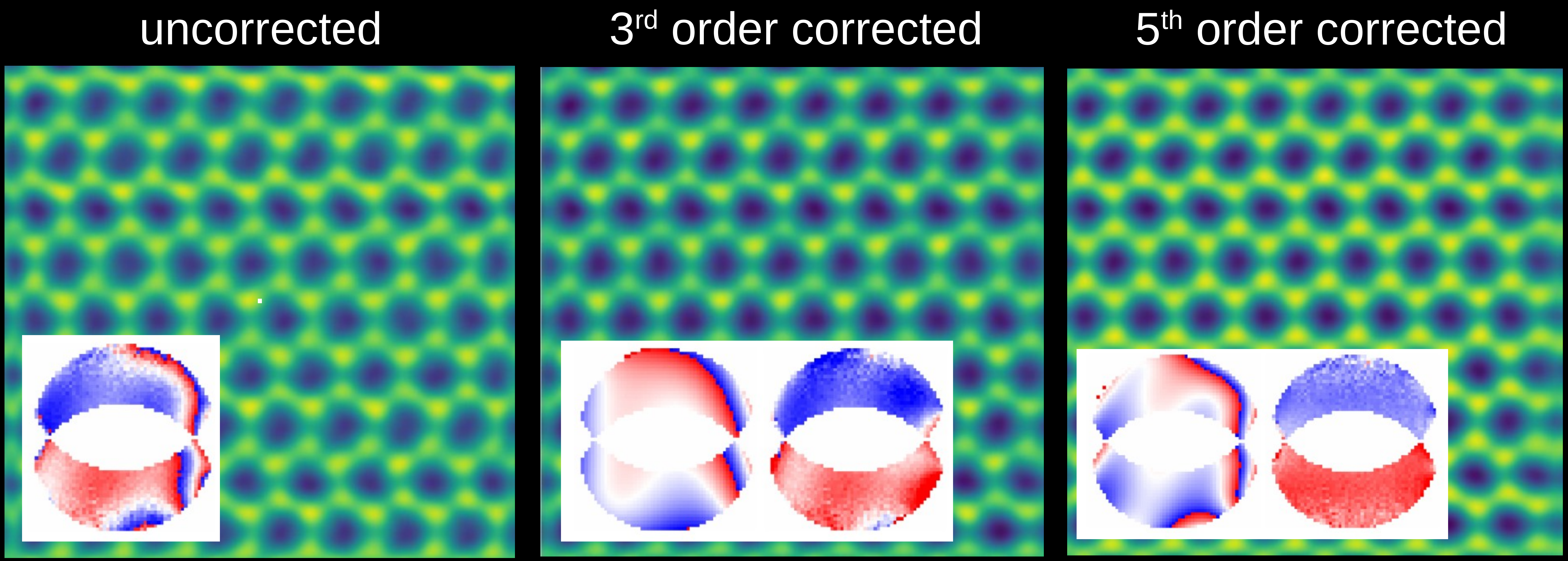
Timothy Pennycook

Christoph Hofer

Courtesy of Christoph Hofer

3rd order aberration correction might not be sufficient

Experimental data of monolayer graphene, 35 mrad (Vienna)

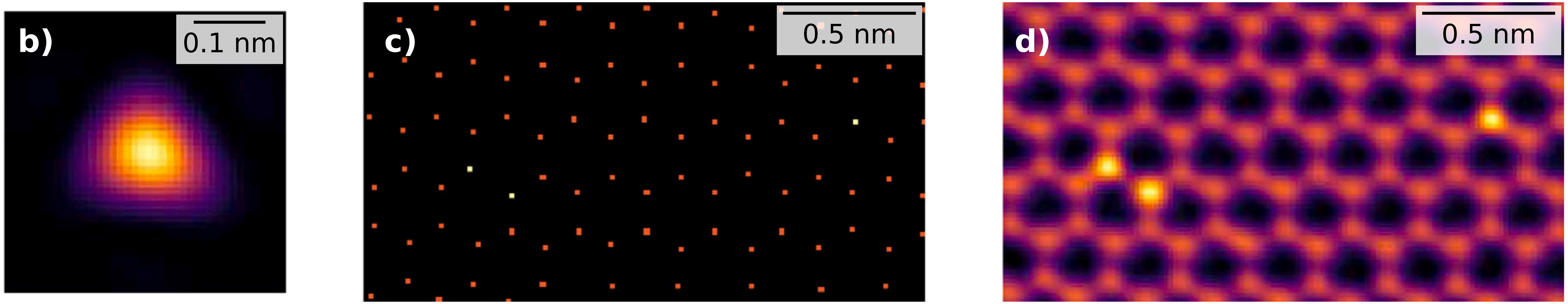



Timothy Pennycook


Christoph Hofer

Courtesy of Christoph Hofer

Site-specific phase via iterative optimization



Maximize correlation between experimental and simulated image

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

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

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

μ^{exp} and μ^{sim} = mean values

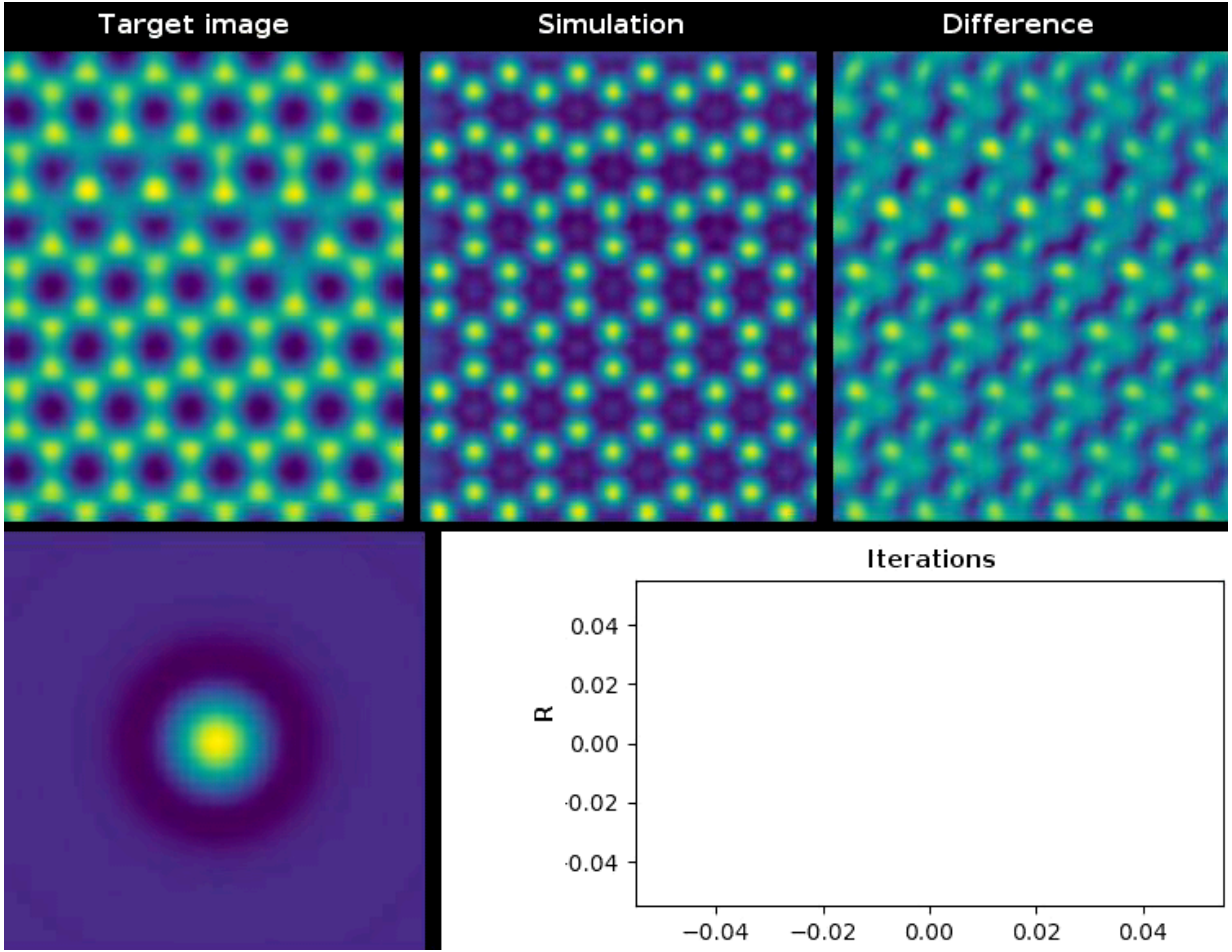
σ^{sim} and σ^{exp} = standard deviations

Phase analysis

STEM_Optimization*
(commit 6c549ba9)

C. Hofer et al., Ultramicroscopy 227 (2021) 113292
* https://gitlab.com/christoph_hofer/stem_optimization

Defective
WS_2



Timothy Pennycook

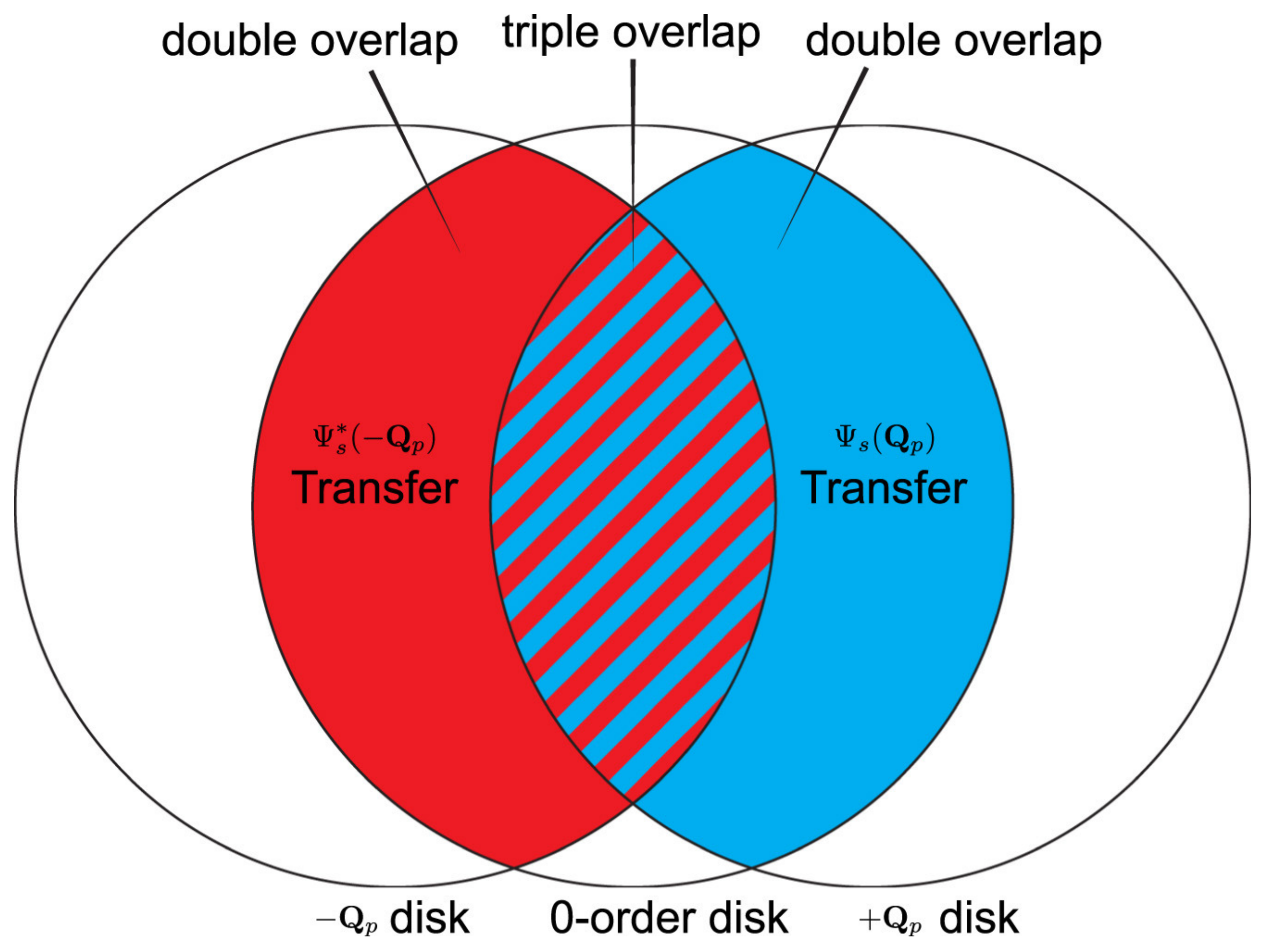
Christoph Hofer

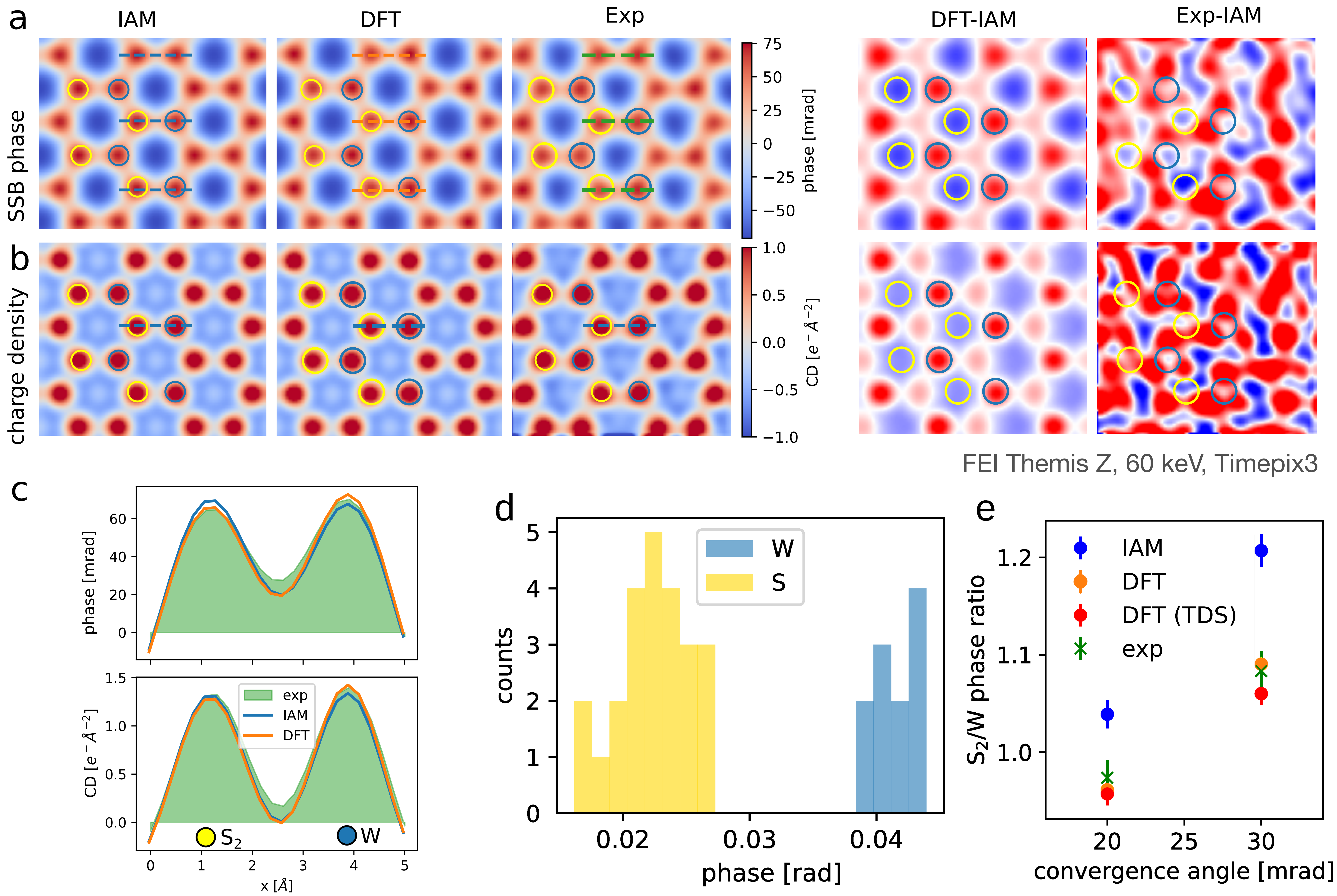
Can account for:

- aberrations
- incoherence
- scan distortions
- sample drift
- sample tilt
- *etc…*

C. Hofer et al., Ultramicroscopy 227 (2021) 113292

Ptychography for charge transfer in defective WS_2



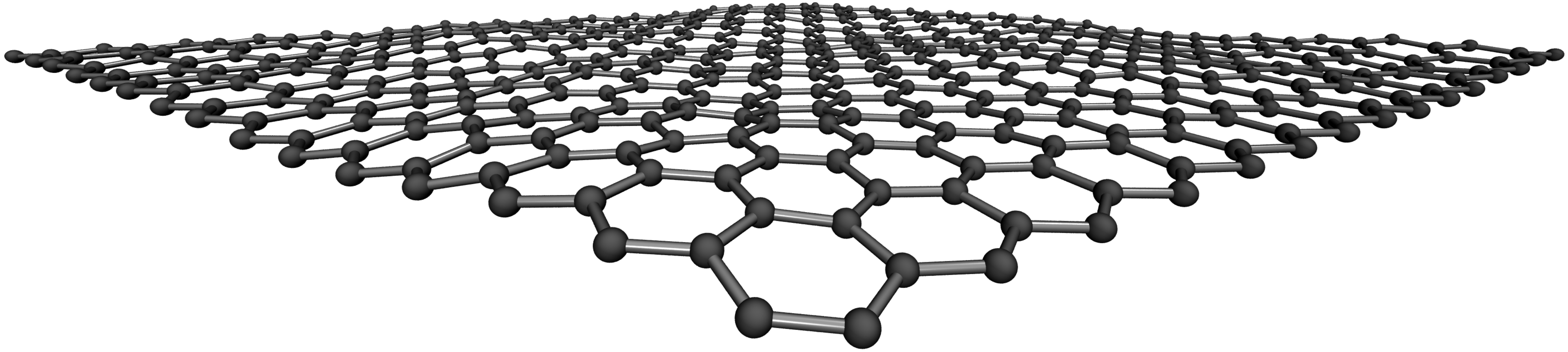


FEI Themis Z, 60 keV, Timepix3

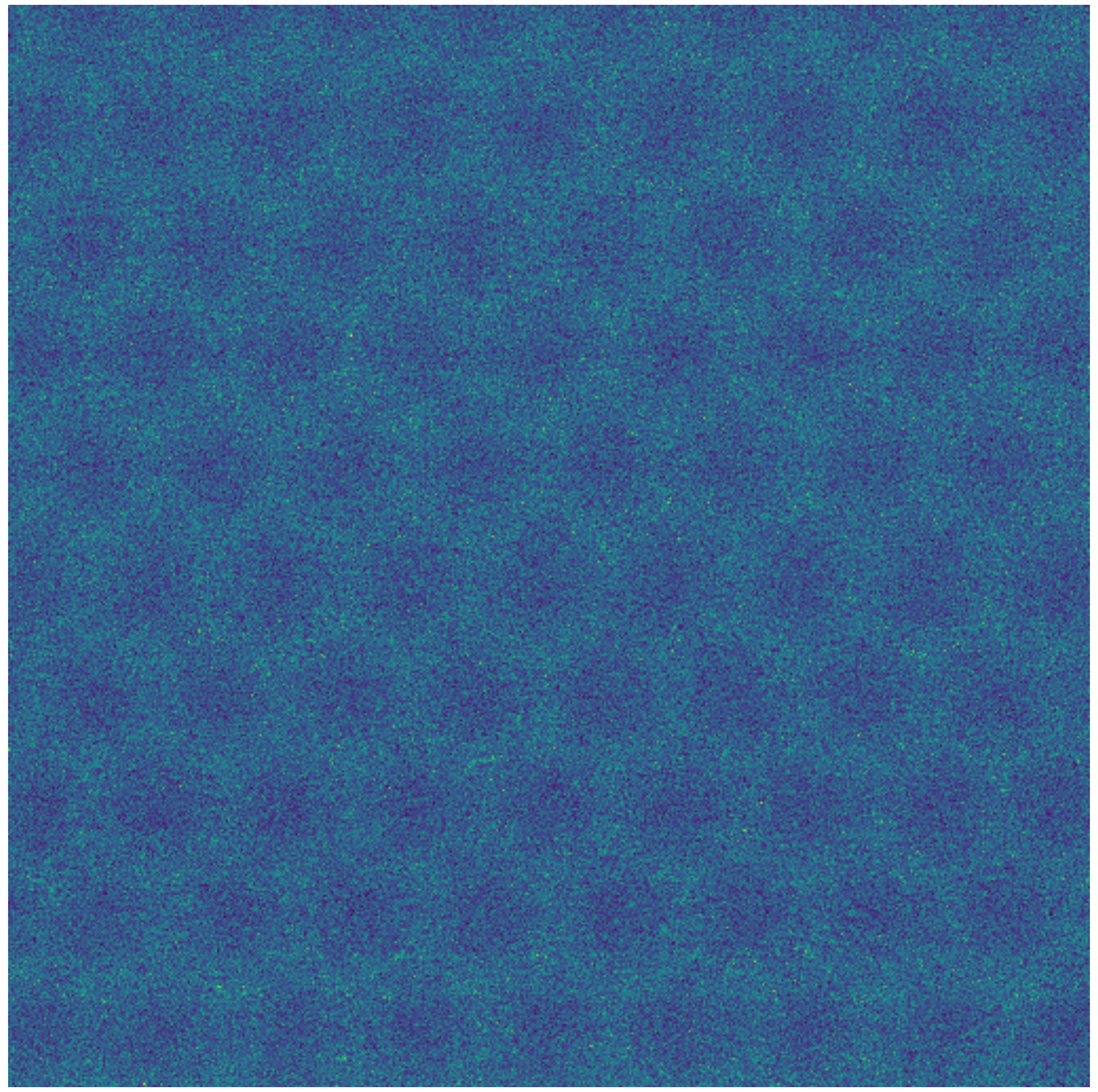
Timepix3 limited to ~2 pA…

Graphene: ideal uniform phase object

- ✅ ∞ radiation hardness <80 keV ⇒ no dose limitation
- ✅ low-Z, one-atom-thick ⇒ perfect weak phase object
- ✅ only one element ⇒ each atomic site identical



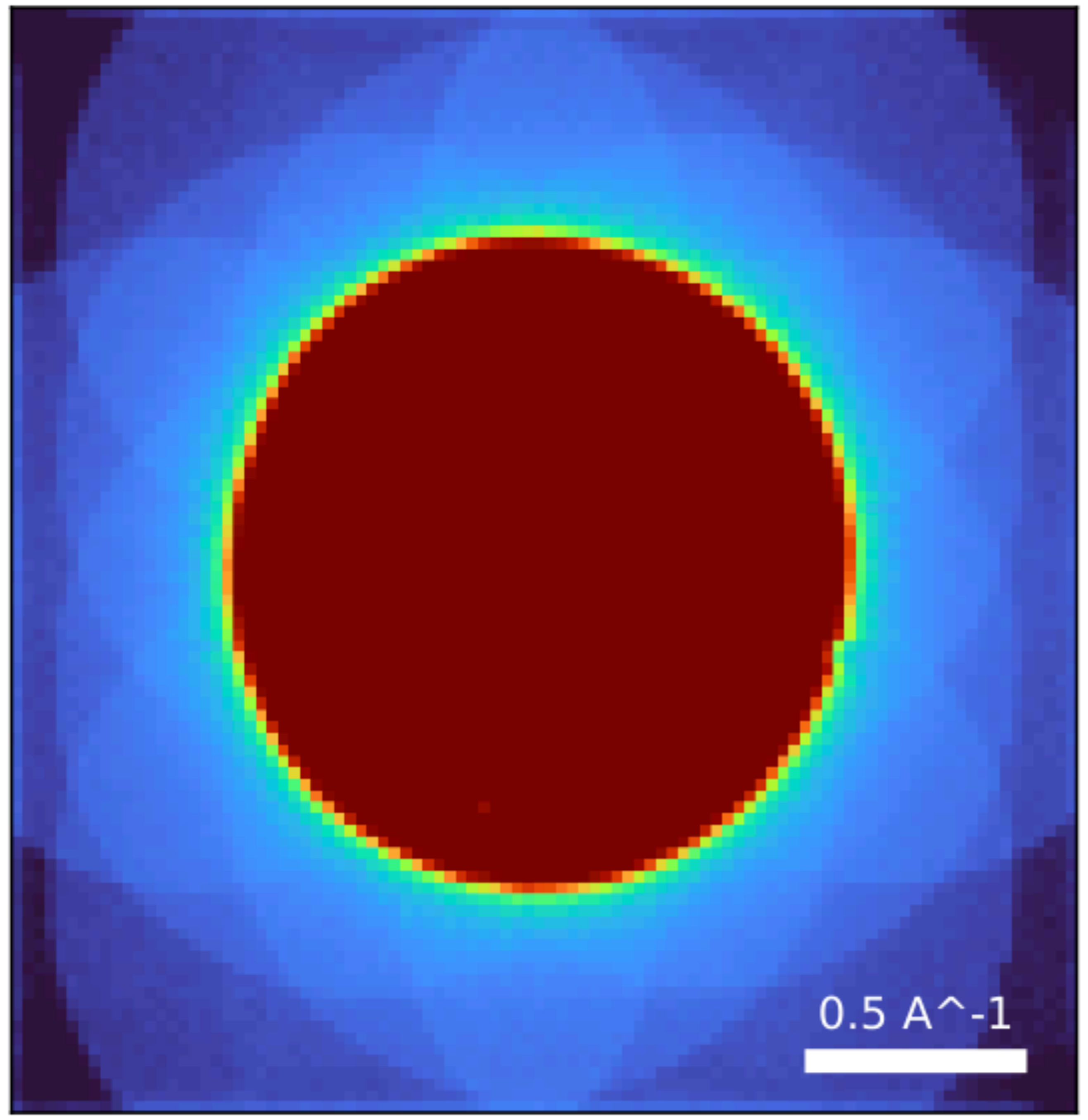
Nion + ARINA 4D-STEM data collection (original)

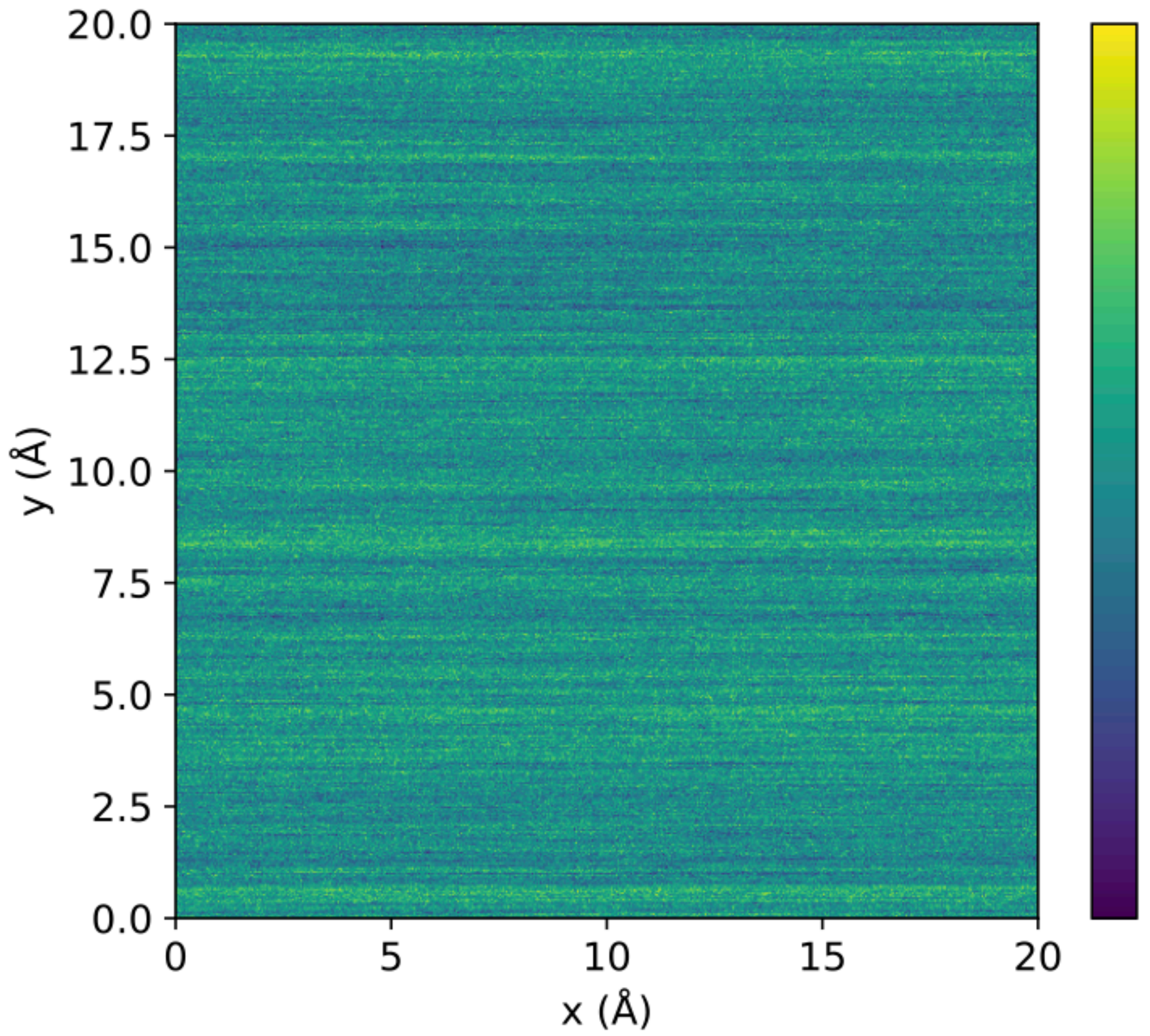


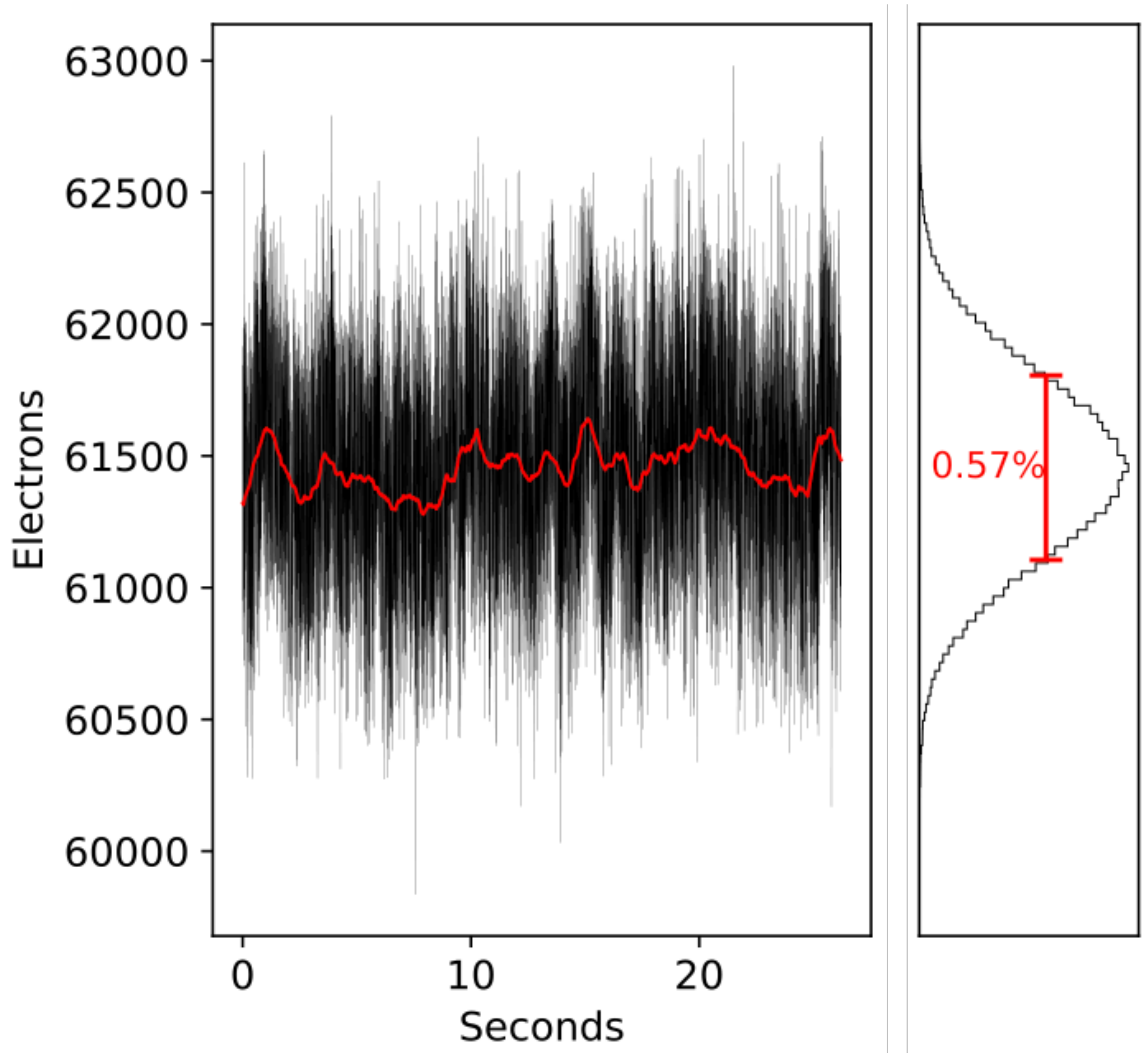
HAADF (80–300 mrad)

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

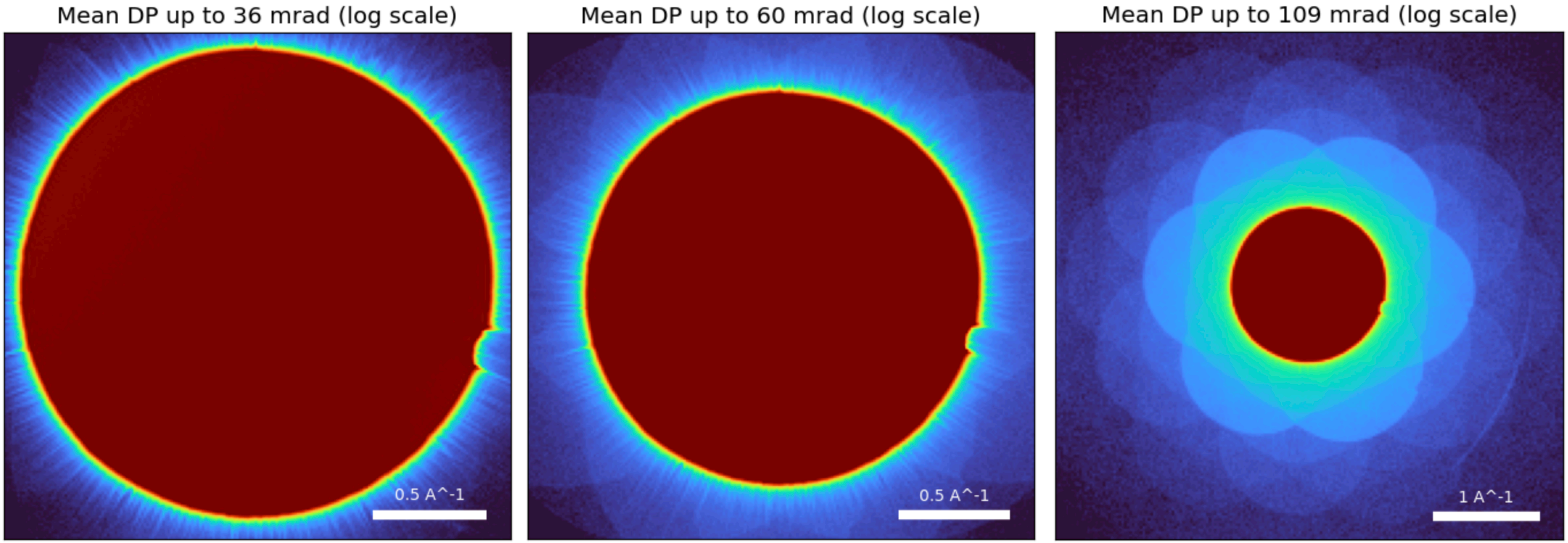






- 34 mrad semi-convergence angle, ~70 mrad max collection angle, ~8 pA
- counting every electron, corrected dose per pixel can be normalized

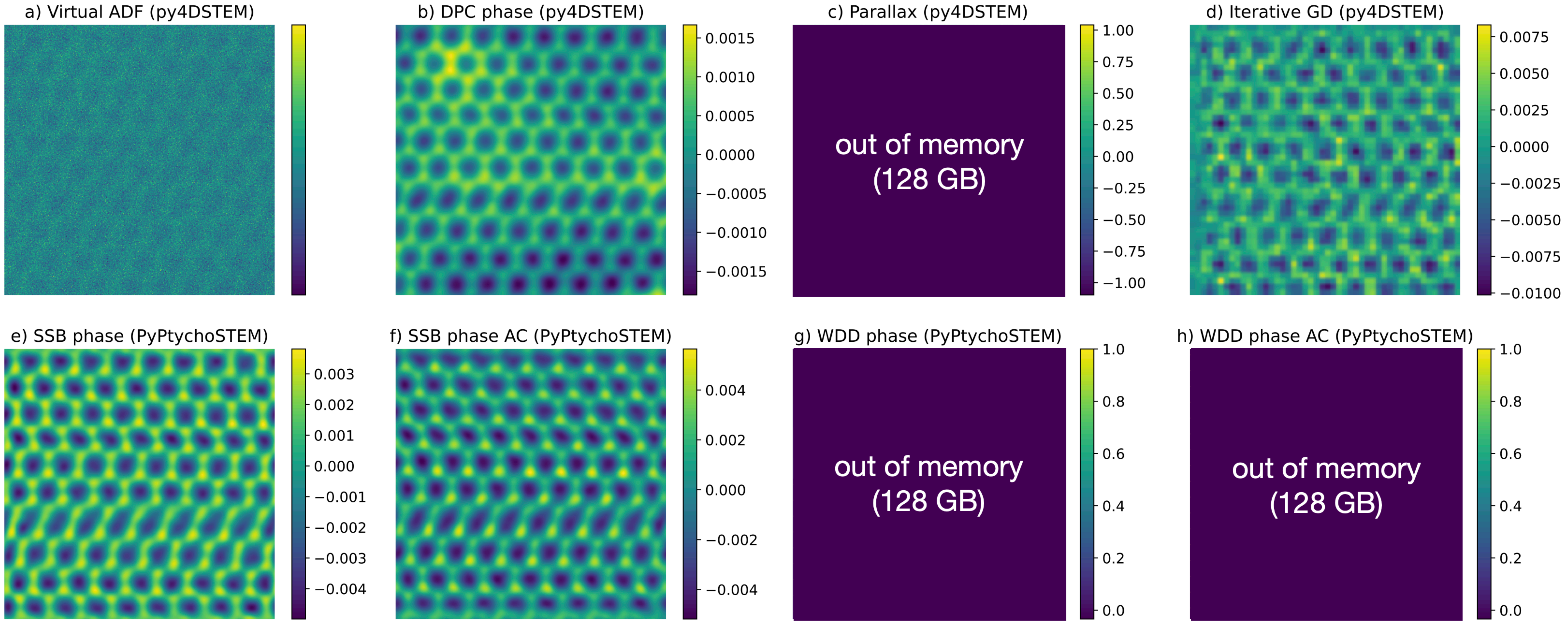
Nion + ARINA 4D-STEM data collection (final)



~200 pA current!

T. Susi, unpublished

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

SSB & WDD: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167
DPC, Parallax, Iterative GD: G. Varnavides *et al.*, arXiv:2309.05250v2 (2024)

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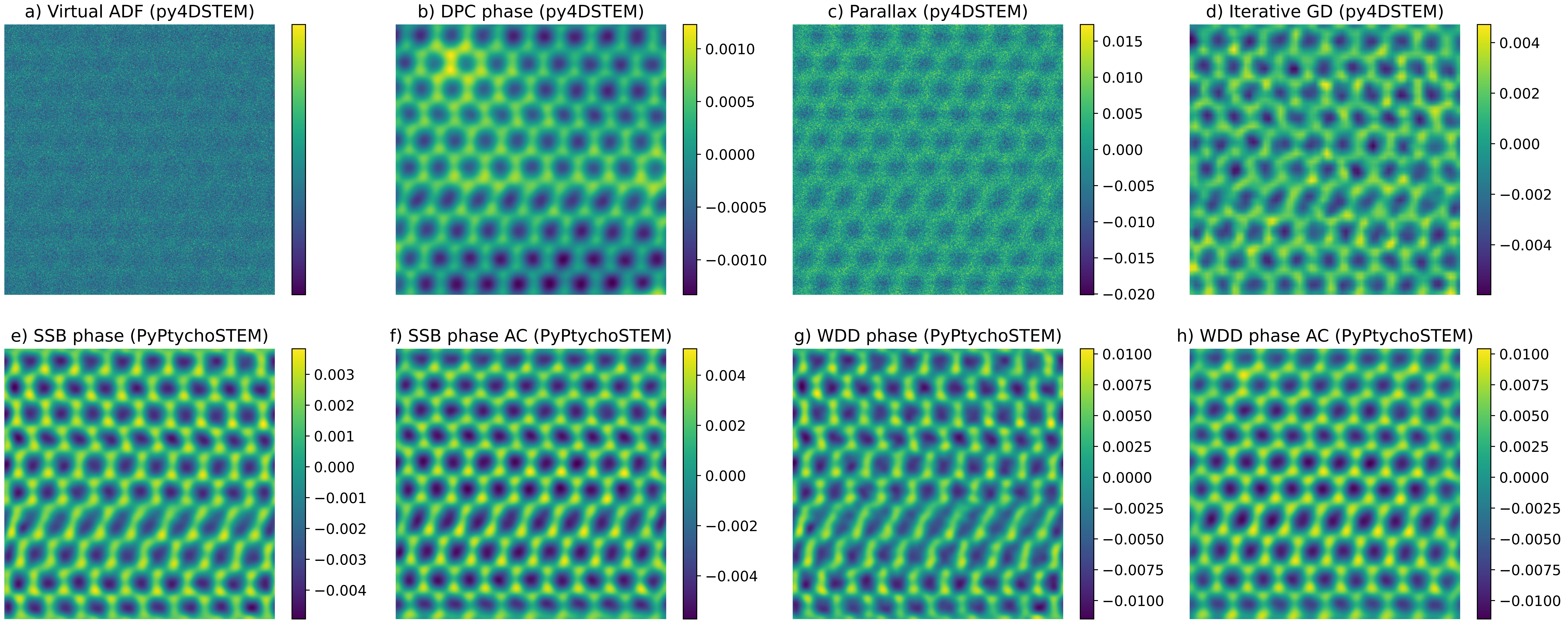

36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 192×192 Qpix
2× binning in Q

SSB & WDD: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167
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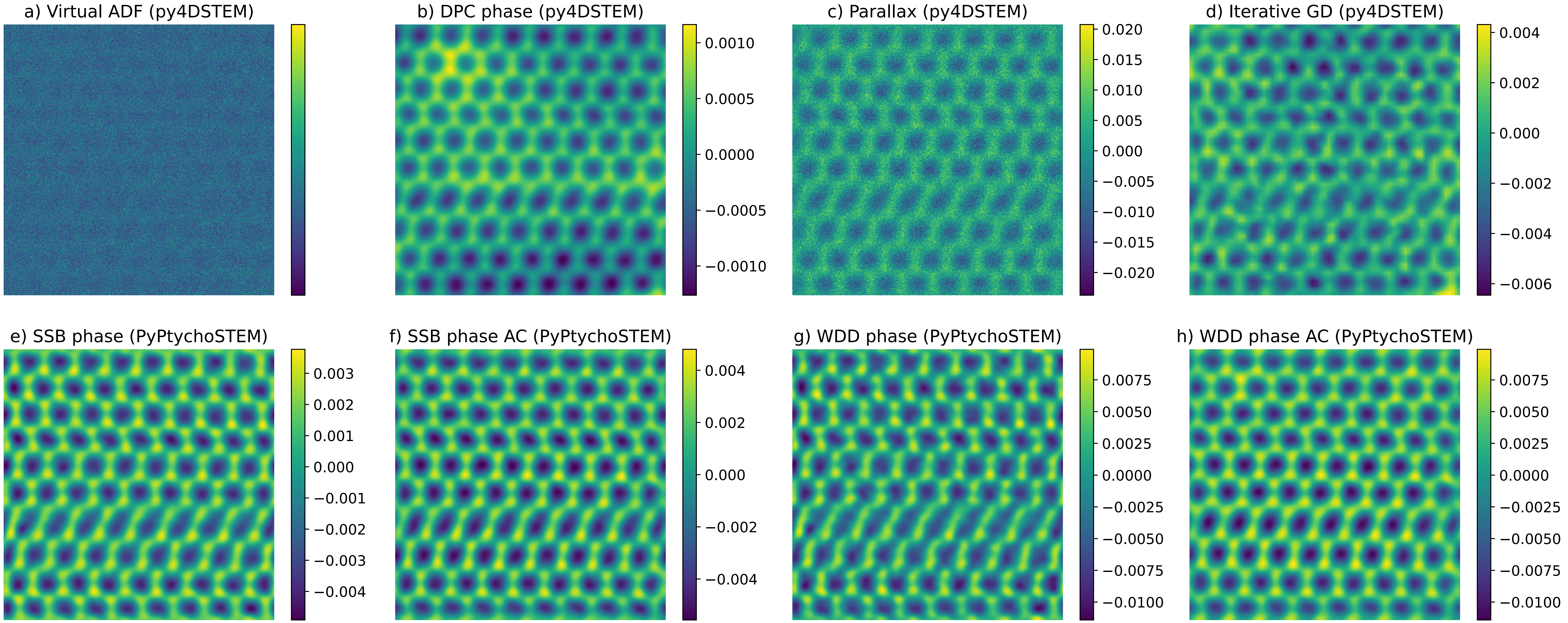

36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 192×192 Qpix

4× binning in Q

SSB & WDD: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167

DPC, Parallax, Iterative GD: G. Varnavides *et al.*, arXiv:2309.05250v2 (2024)

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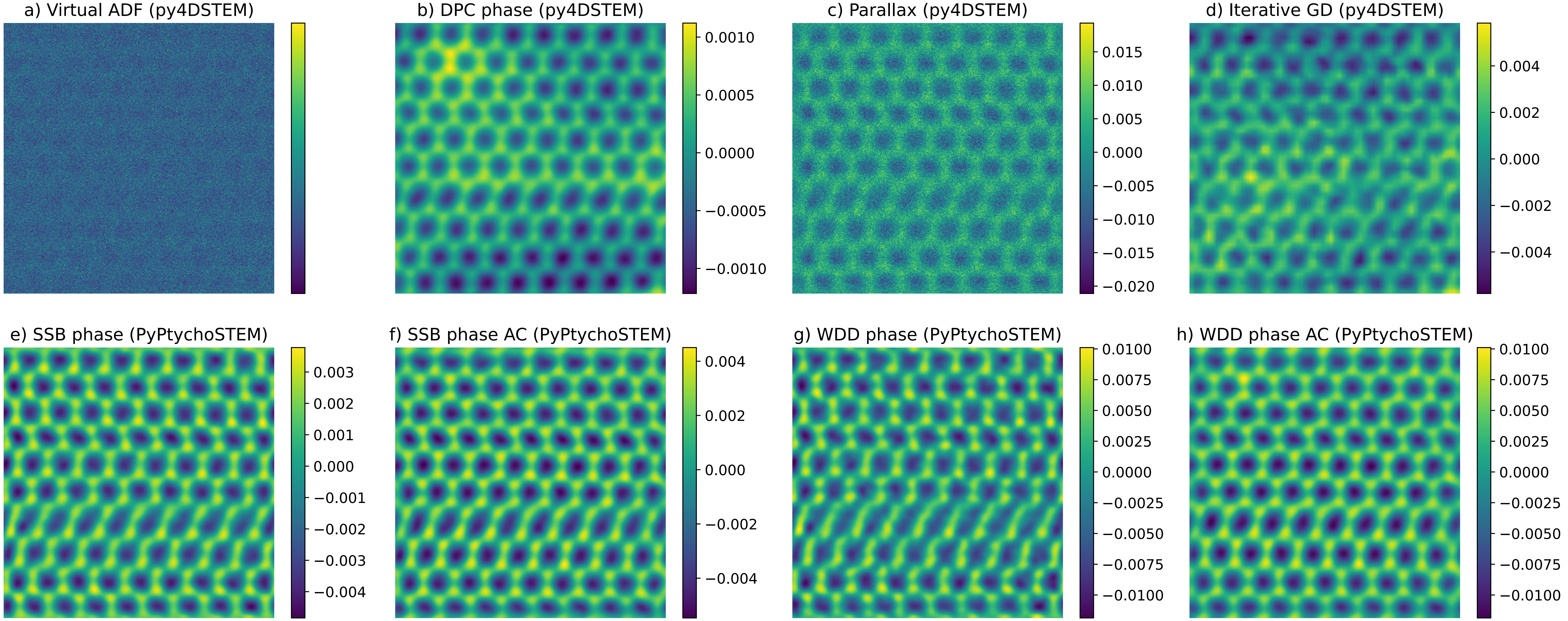

36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 192×192 Qpix
8× binning in Q

SSB & WDD: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167
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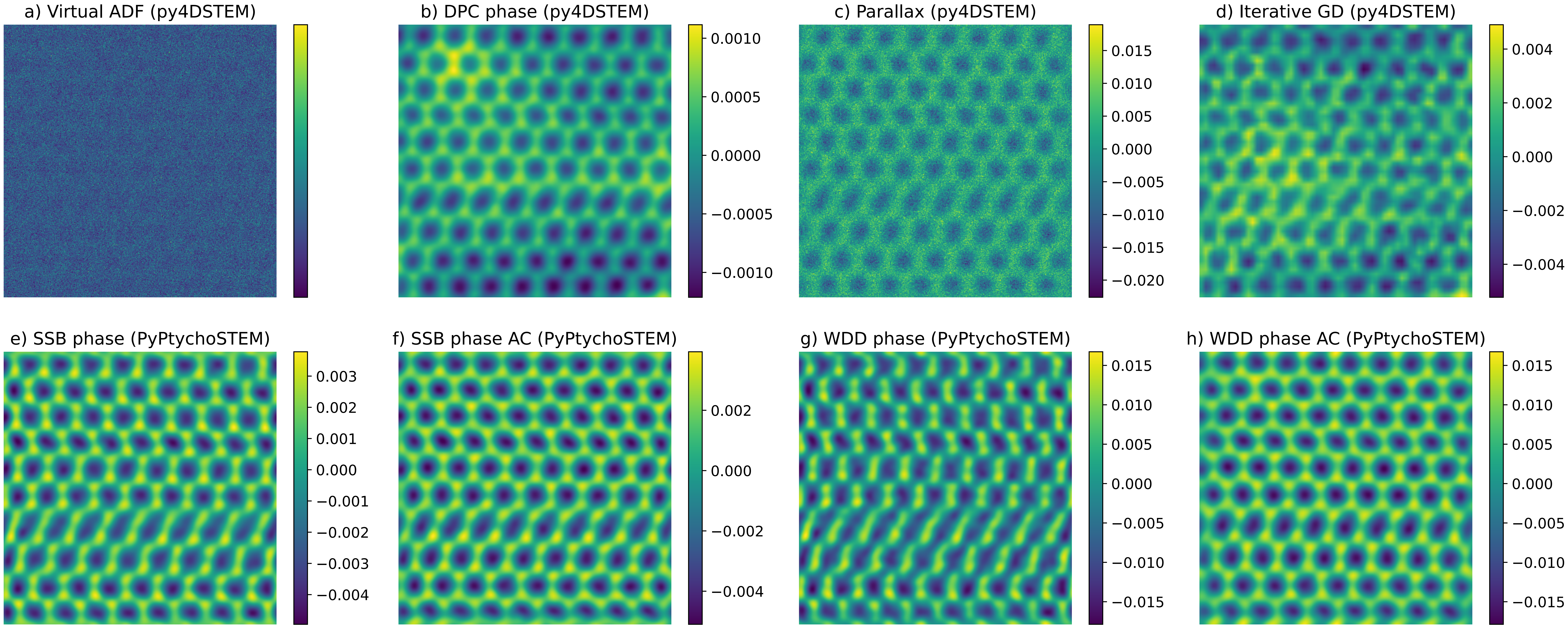

36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 192×192 Qpix

16× binning in Q

SSB & WDD: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167

DPC, Parallax, Iterative GD: G. Varnavides *et al.*, arXiv:2309.05250v2 (2024)

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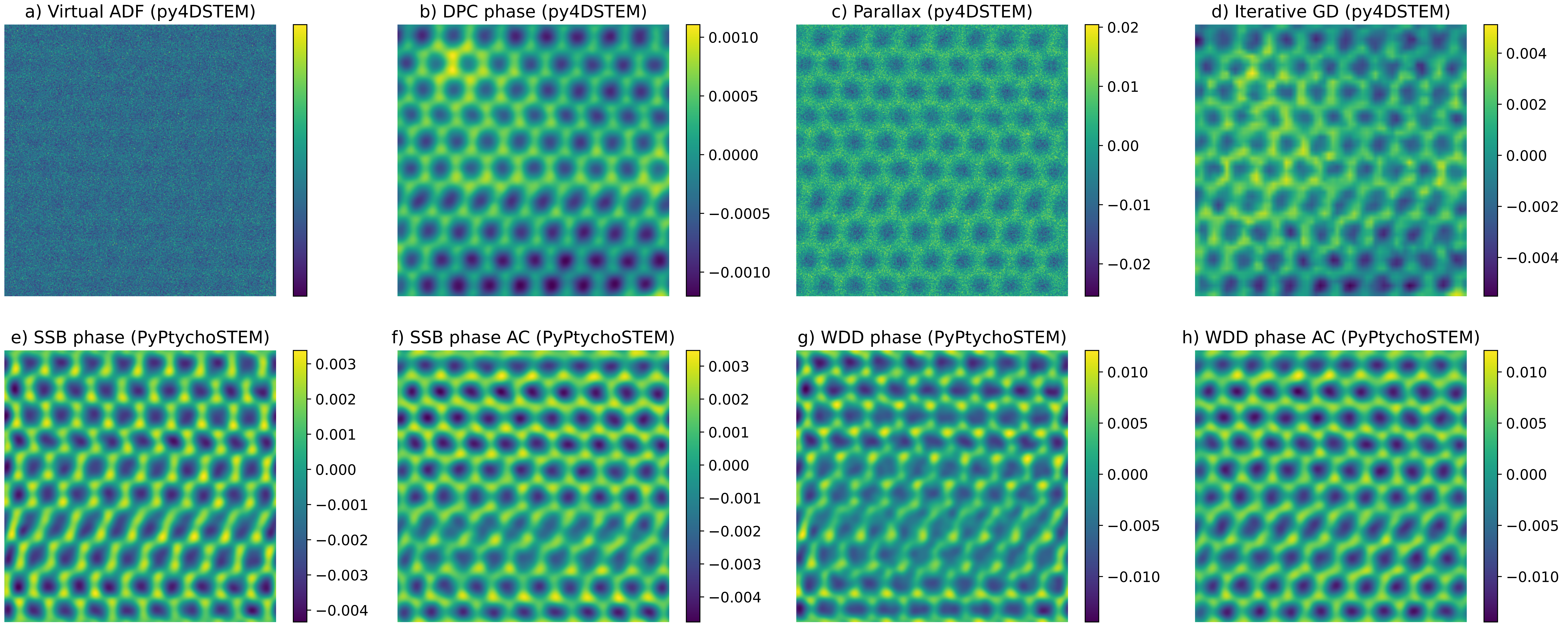
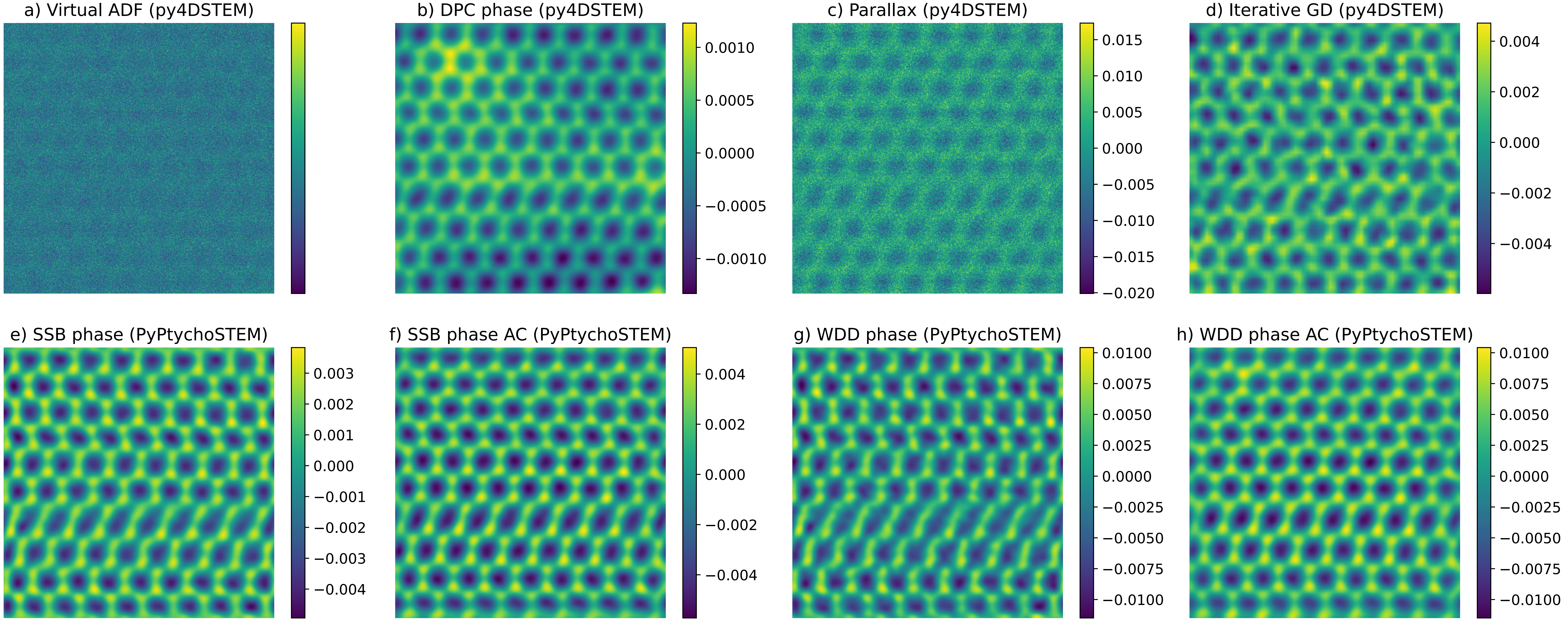


36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 192×192 Qpix
32× binning in Q

SSB & WDD: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167
DPC, Parallax, Iterative GD: G. Varnavides *et al.*, arXiv:2309.05250v2 (2024)

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

SSB & WDD: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167
DPC, Parallax, Iterative GD: G. Varnavides *et al.*, arXiv:2309.05250v2 (2024)

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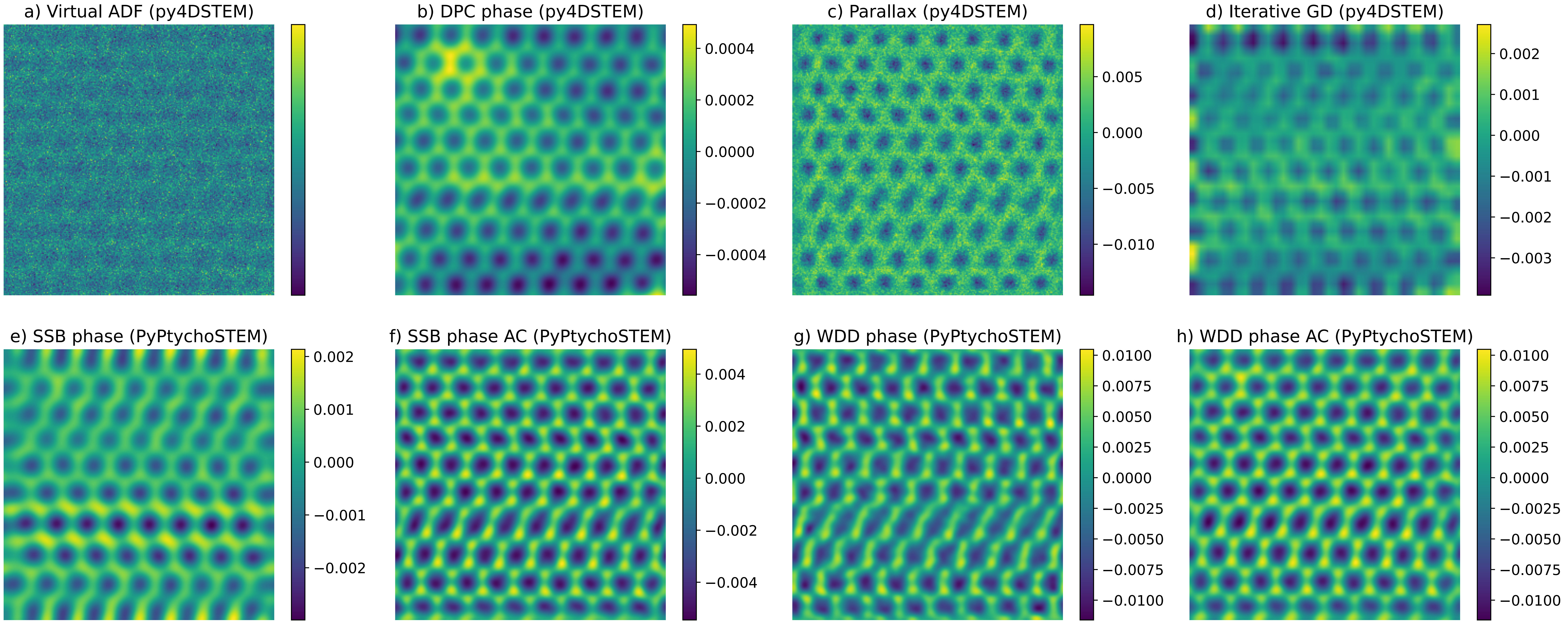

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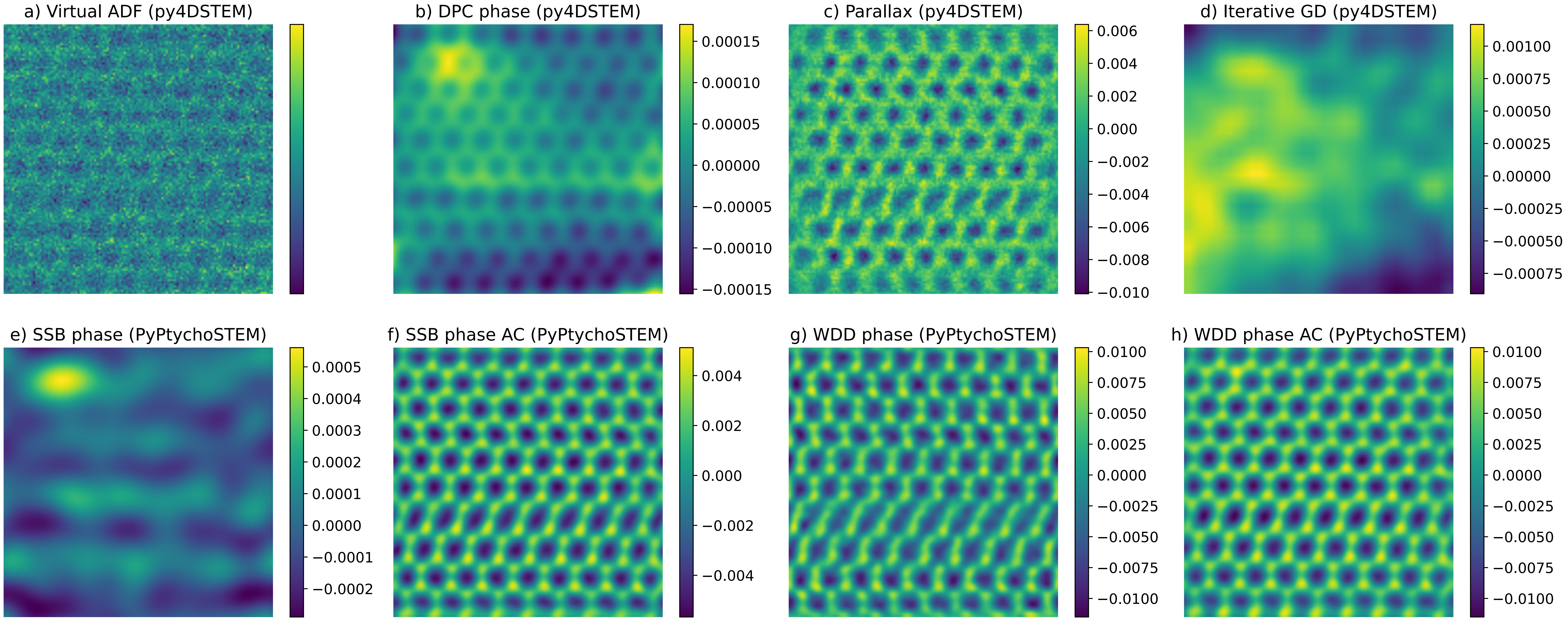

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4× binning in R

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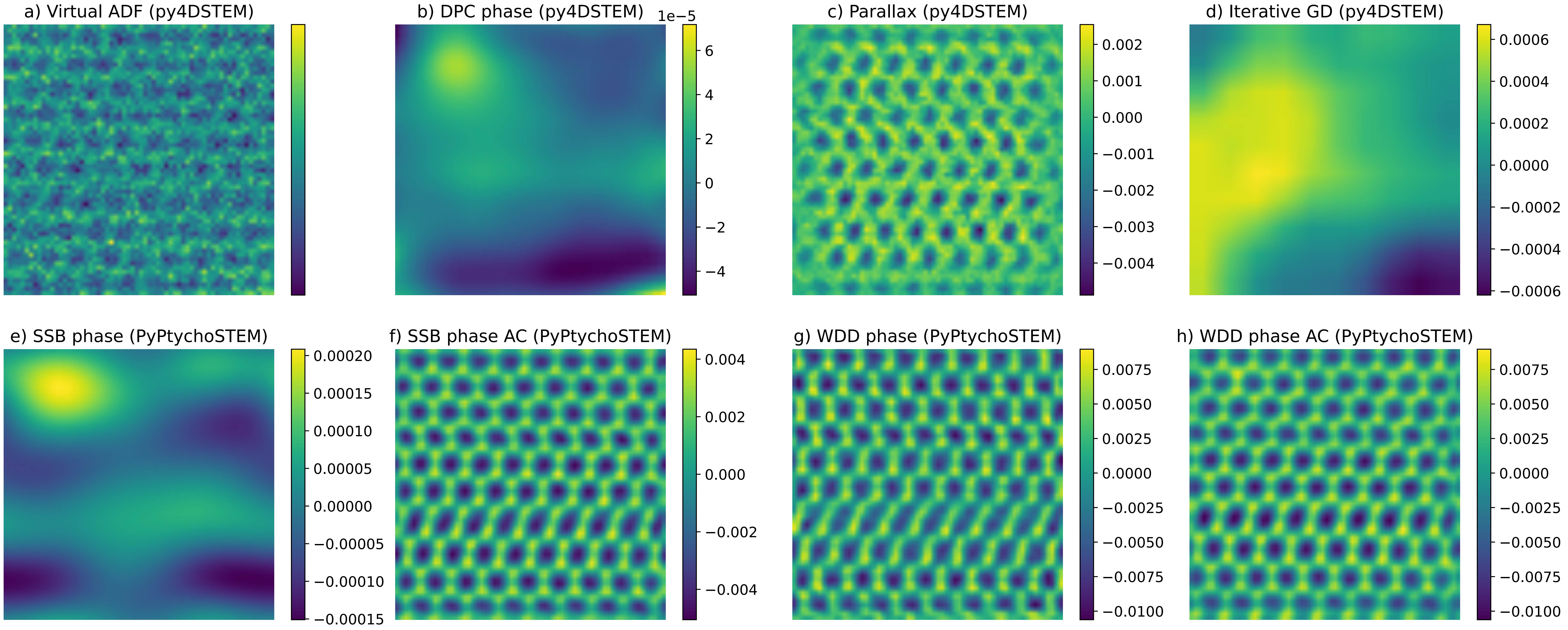

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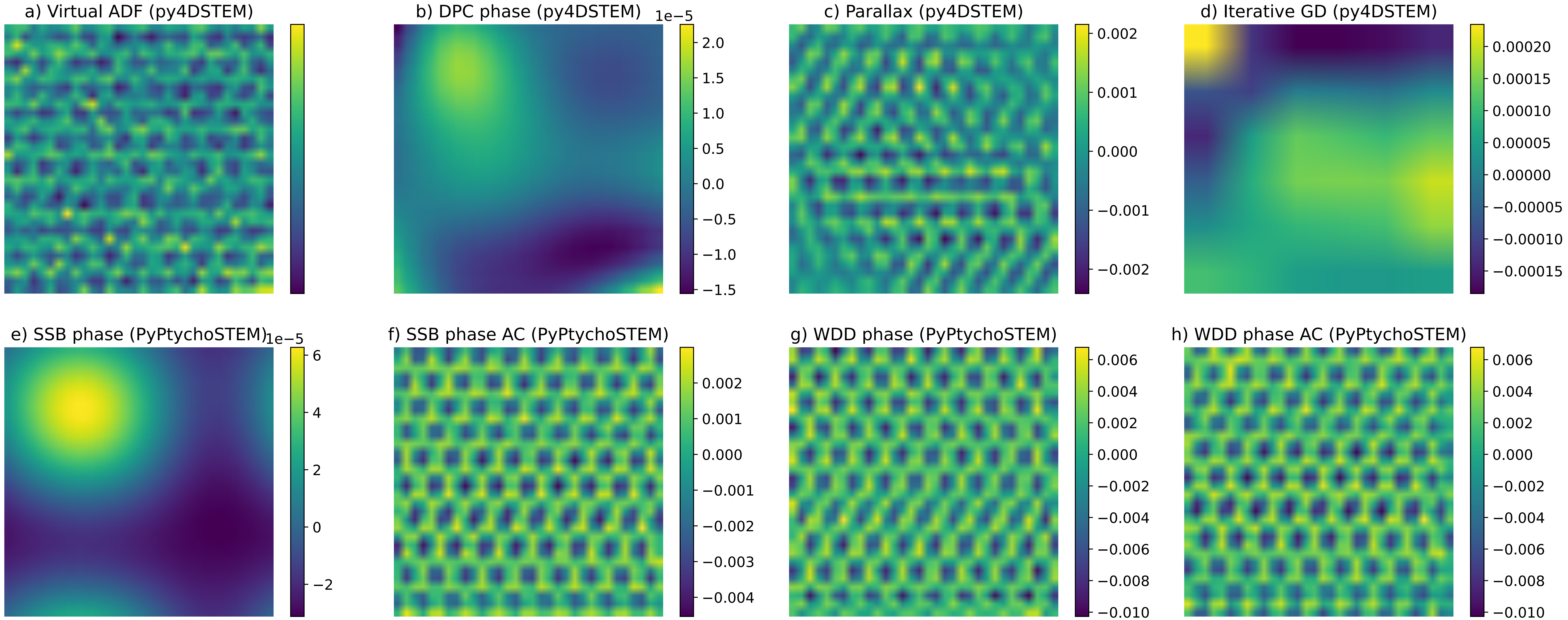

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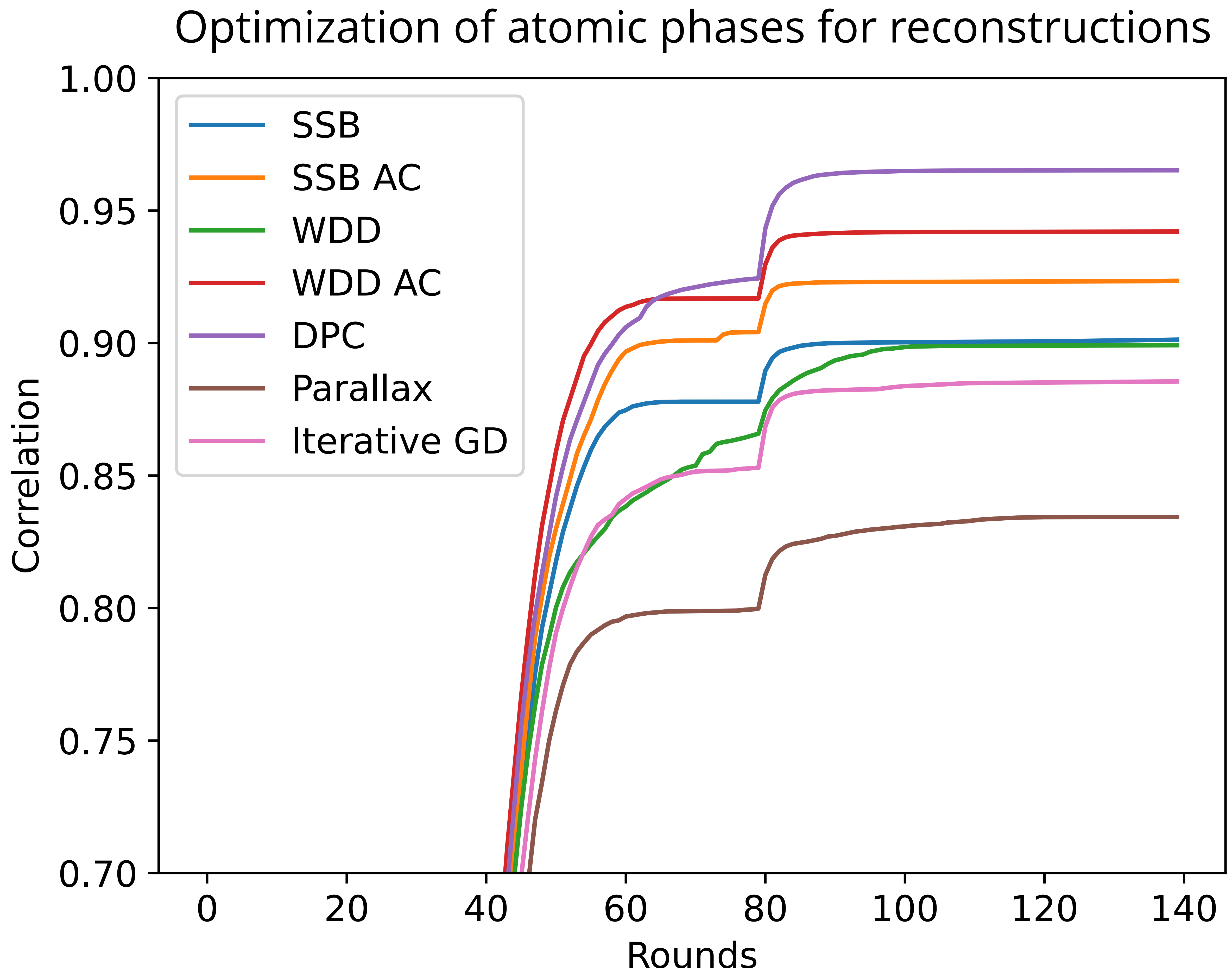
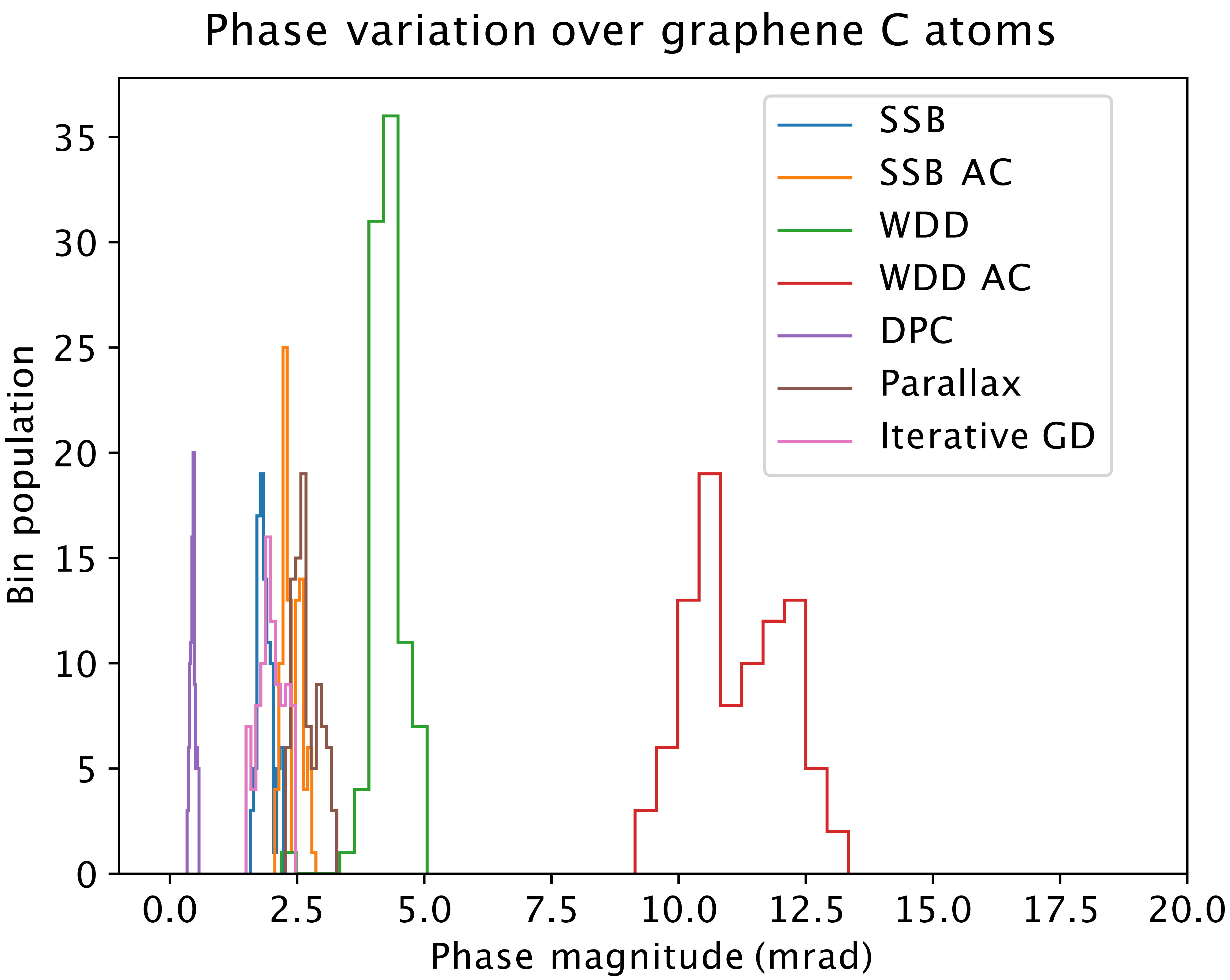
DPC, Parallax, Iterative GD: G. Varnavides *et al.*, arXiv:2309.05250v2 (2024)

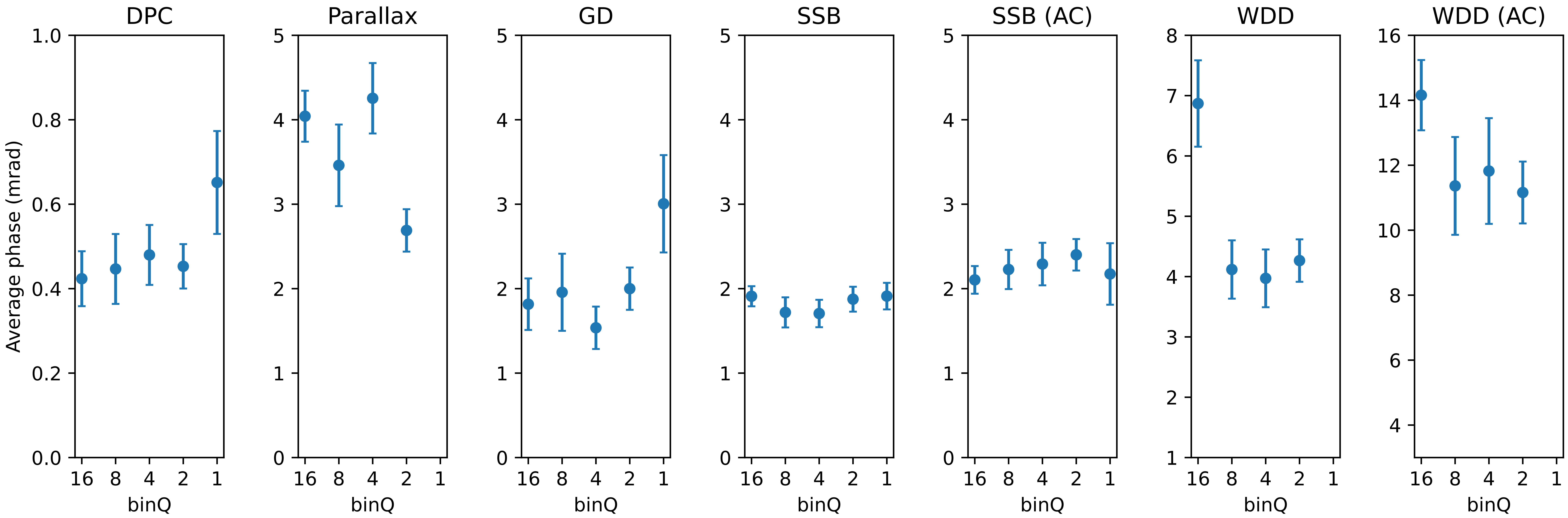
Atomic phase assignment





T. Susi, unpublished

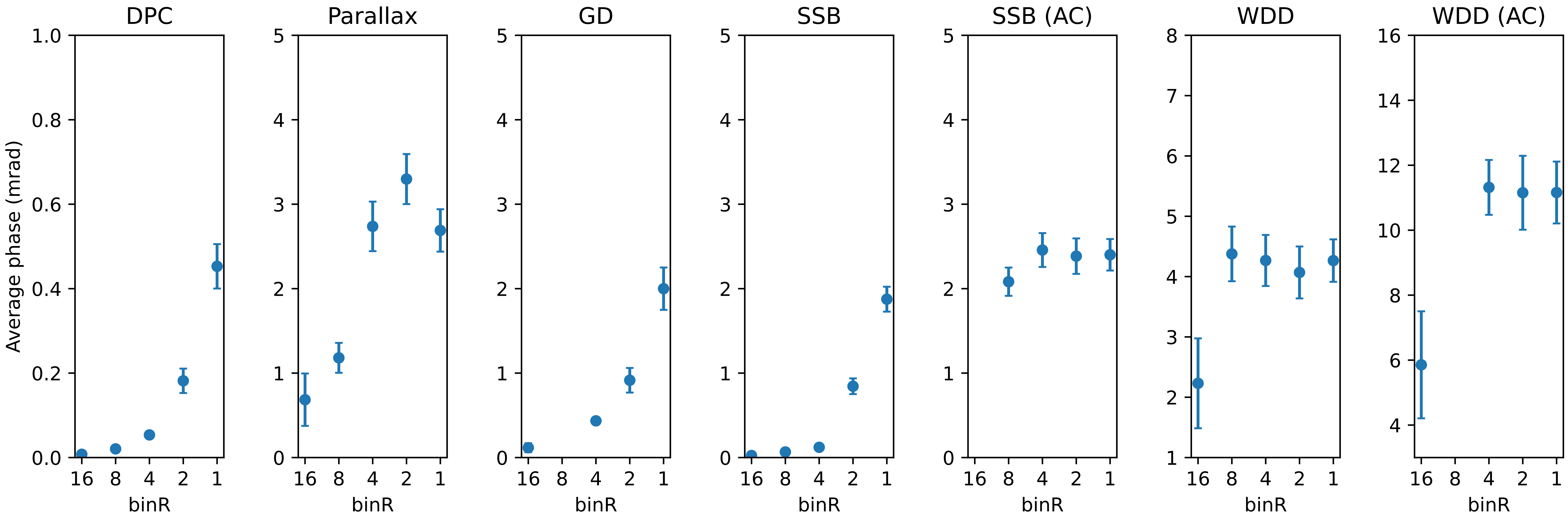
Quantification of phase variation



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

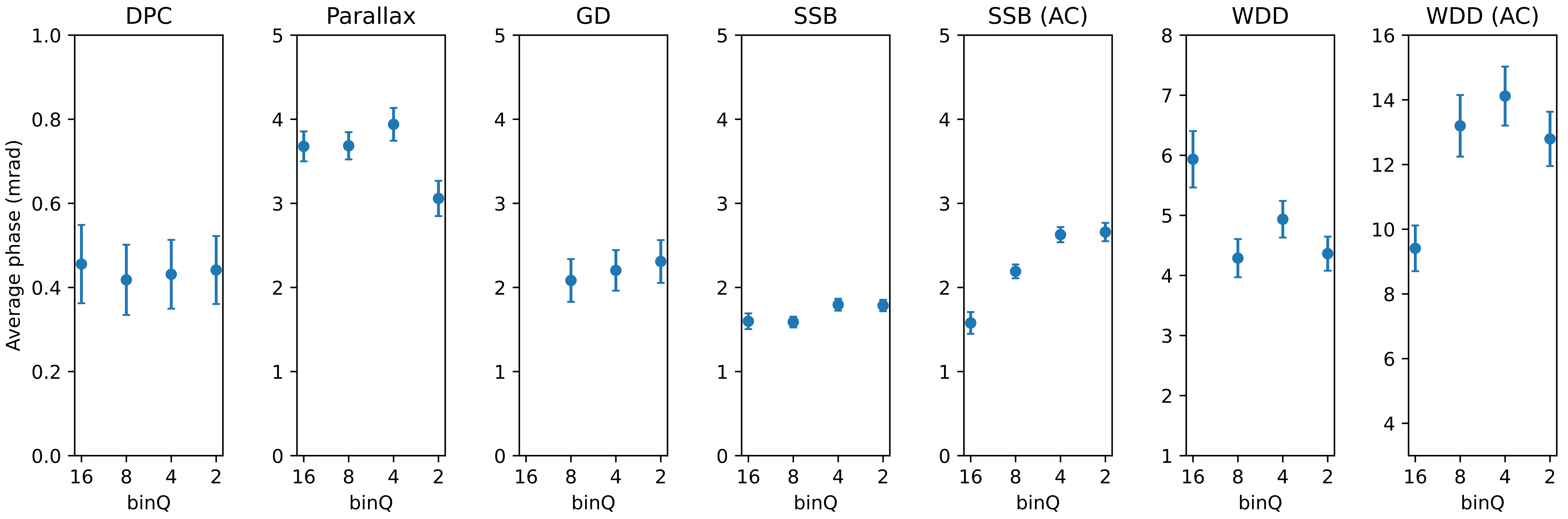
T. Susi, unpublished

Quantification of phase variation



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

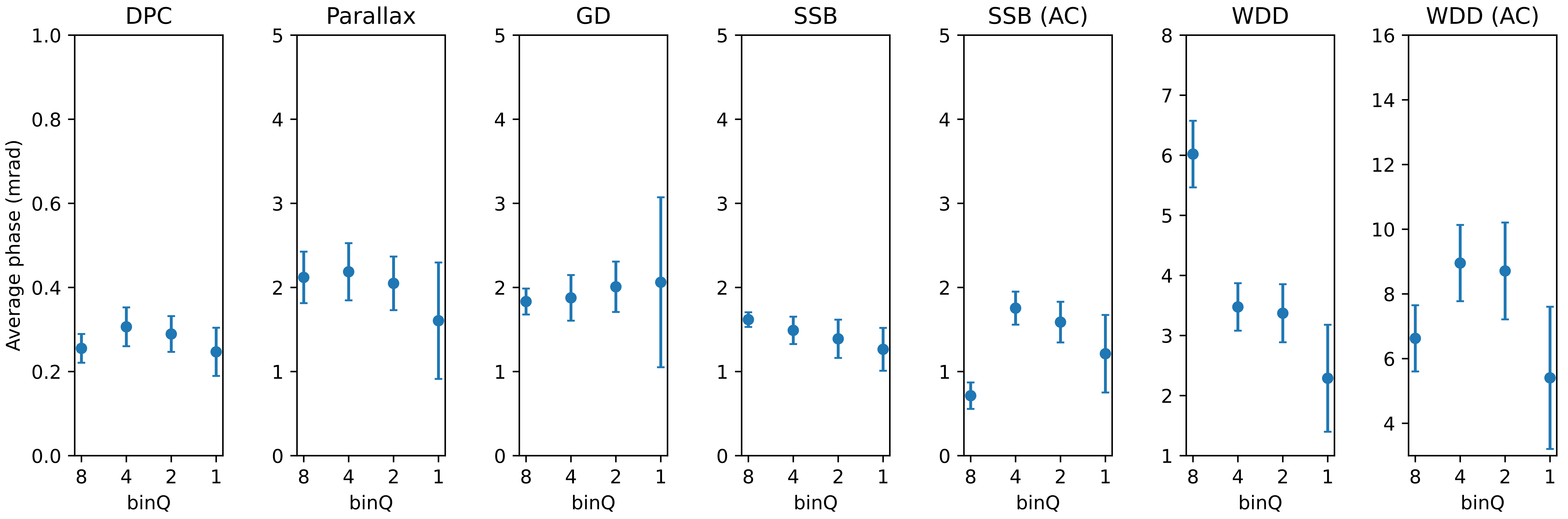
Quantification of phase variation



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

T. Susi, unpublished

Quantification of phase variation



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

universität wien

T. Susi, unpublished

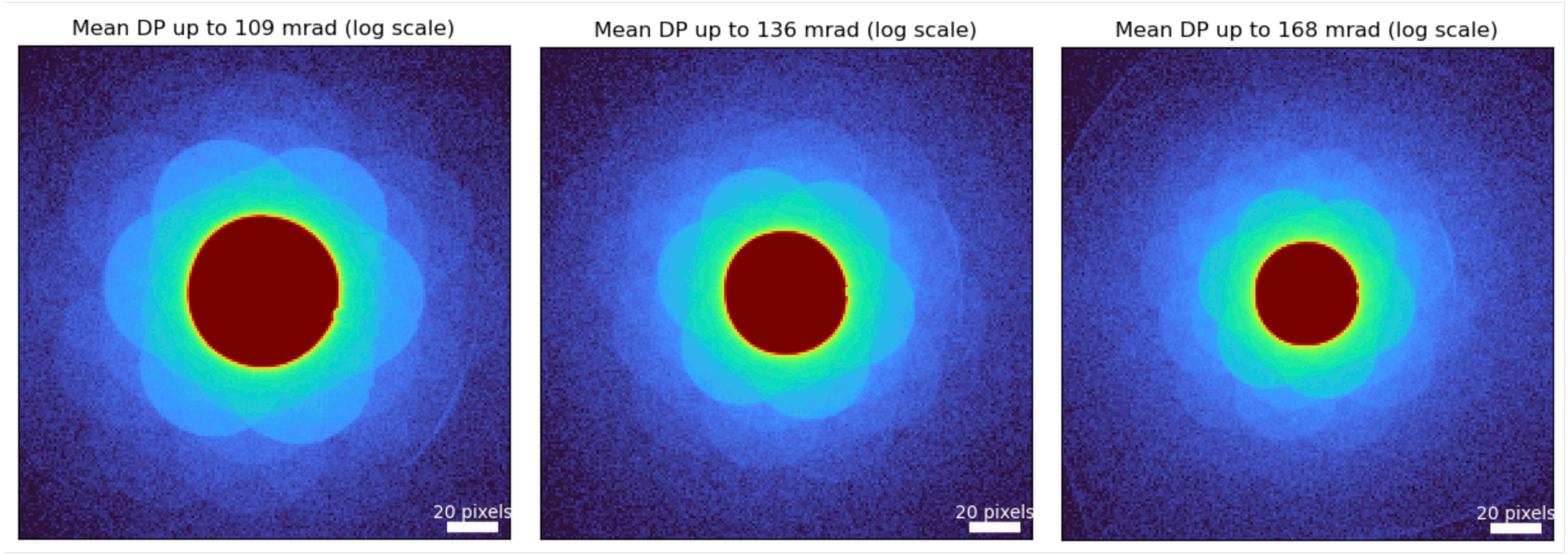
Computational cost

Scan (px)	binQ	CBED (Qpix)	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
	32	6×6	38	4.4	22	122	7.1	35	144	147

* GPU-accelerated (NVidia RTX4090)

T. Susi, unpublished

Future prospects



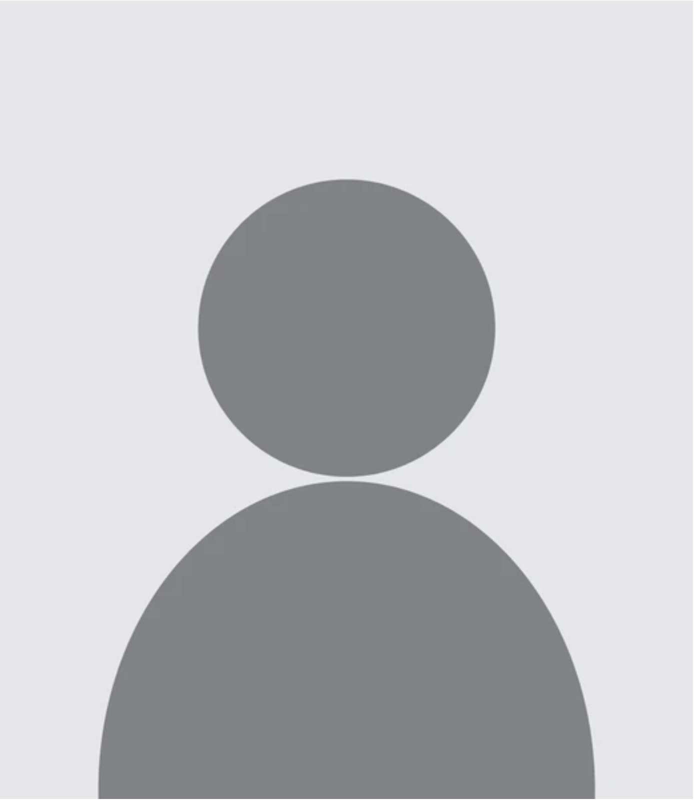
- Defocused-probe ptychography with iterative reconstructions
- Variable camera lengths (for super-resolution up to ~4 × alpha)
- Live virtual detectors / (semi-)live single-sideband ptychography?
- Structure & charge transfer in various materials… *and much more*

Upcoming Japan stay (Sep 2024 – Feb 2025)

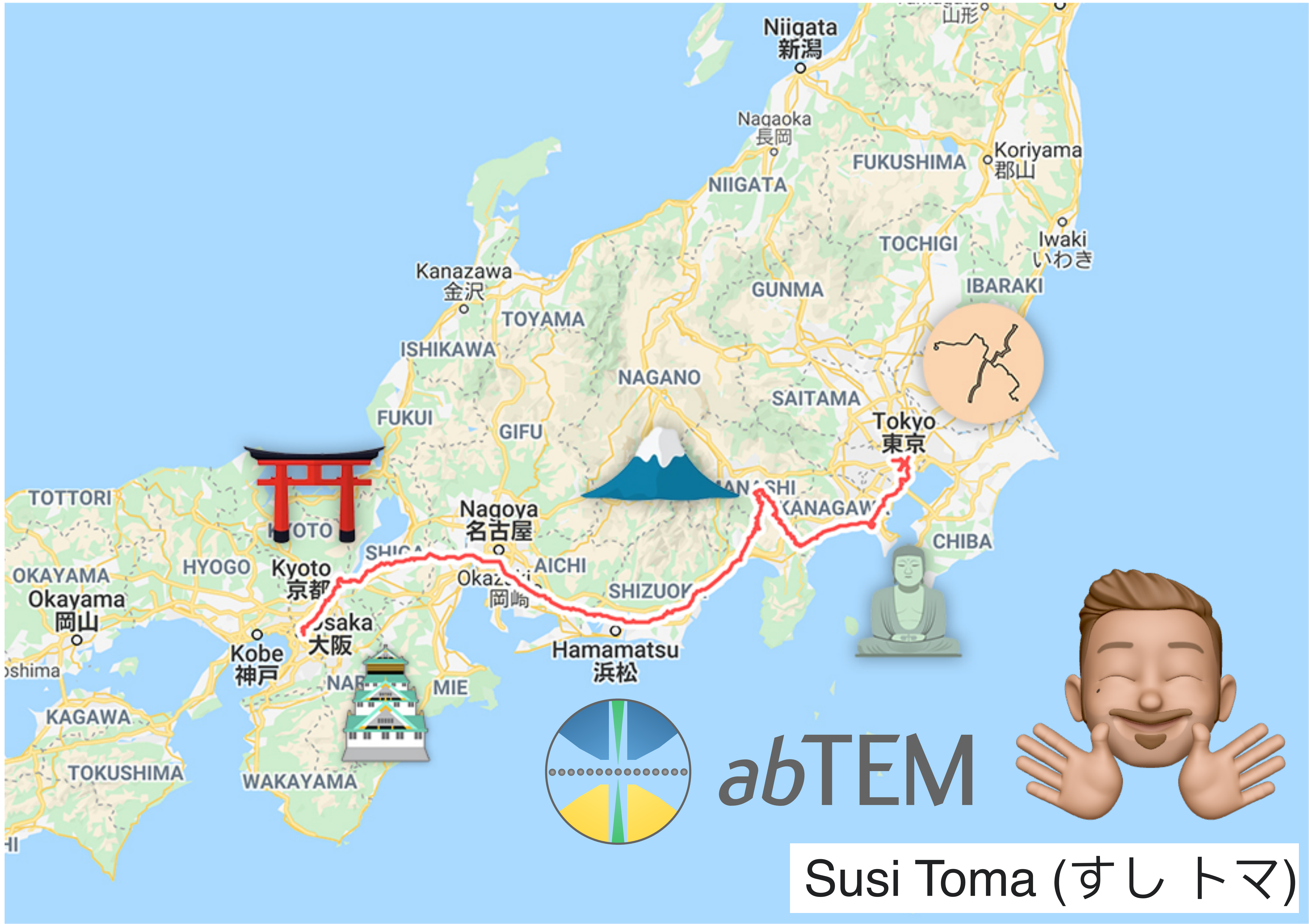
Nagashima (TUS)

Suenaga (Osaka)

Shibata (Tokyo)



you? (???)



Thank you!

Questions, collaborations: *toma.susi@univie.ac.at*

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

Open datasets:
https://phaidra.univie.ac.at/o:2081765