Open-Source 4D-STEM Phase Reconstructions

of Focused-Probe Data with Near-Ideal Direct-Electron Detection

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

Open-source software

MOLECULAR FOUNDRY

Language

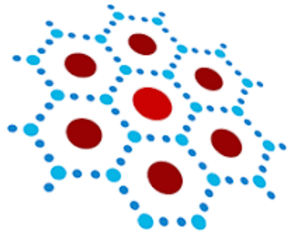
Python
(3.11.9)

Data Acquisition

Nion Swift
(0.16.10)

Phase Reconstructions

PyPtychoSTEM
(20d3be1f)



py4DSTEM
(0.14.16)

Stephanie Ribet

Georgios Varnavides

<u>Reconstruction Algorithms</u>:

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**)

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<u>SSB & WDD</u>: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167
<u>DPC, parallax, iterative GD</u>: G. Varnavides *et al.*, arXiv:2309.05250v2 (2024)

Near-ideal direct-electron 4D-STEM

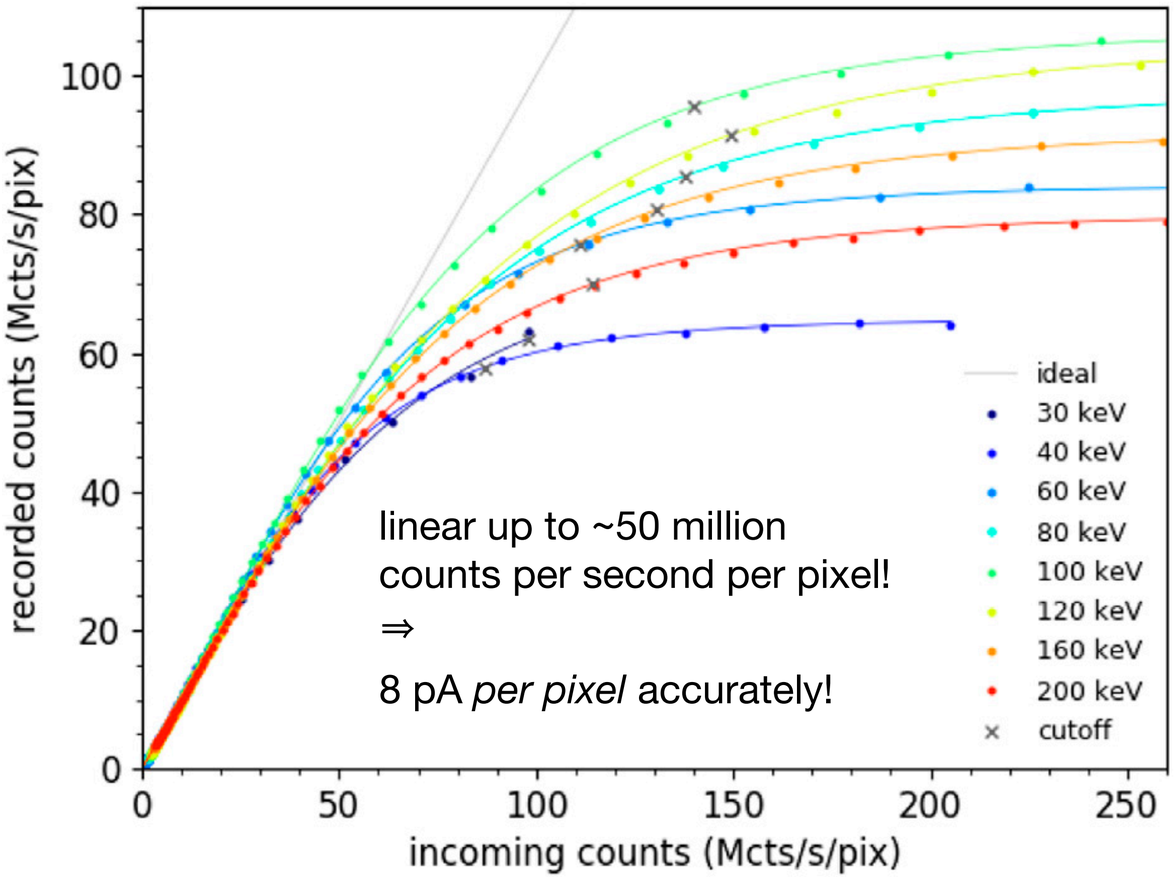
Number of pixels (W x H)	**192 × 192**
Active area (W x H) [mm²]	20 × 20
Pixel size (W x H) [µm²]	100 × 100
Sensor material	Silicon (Si) or high-Z
Energy range [keV]	30–300
Frame rate (max.) [Hz]	**120,000**
Count rate (max.) [el/s/pixel]	10^8
Detective Quantum Efficiency, DQE(0)	0.82 @ 80 keV 0.75 @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

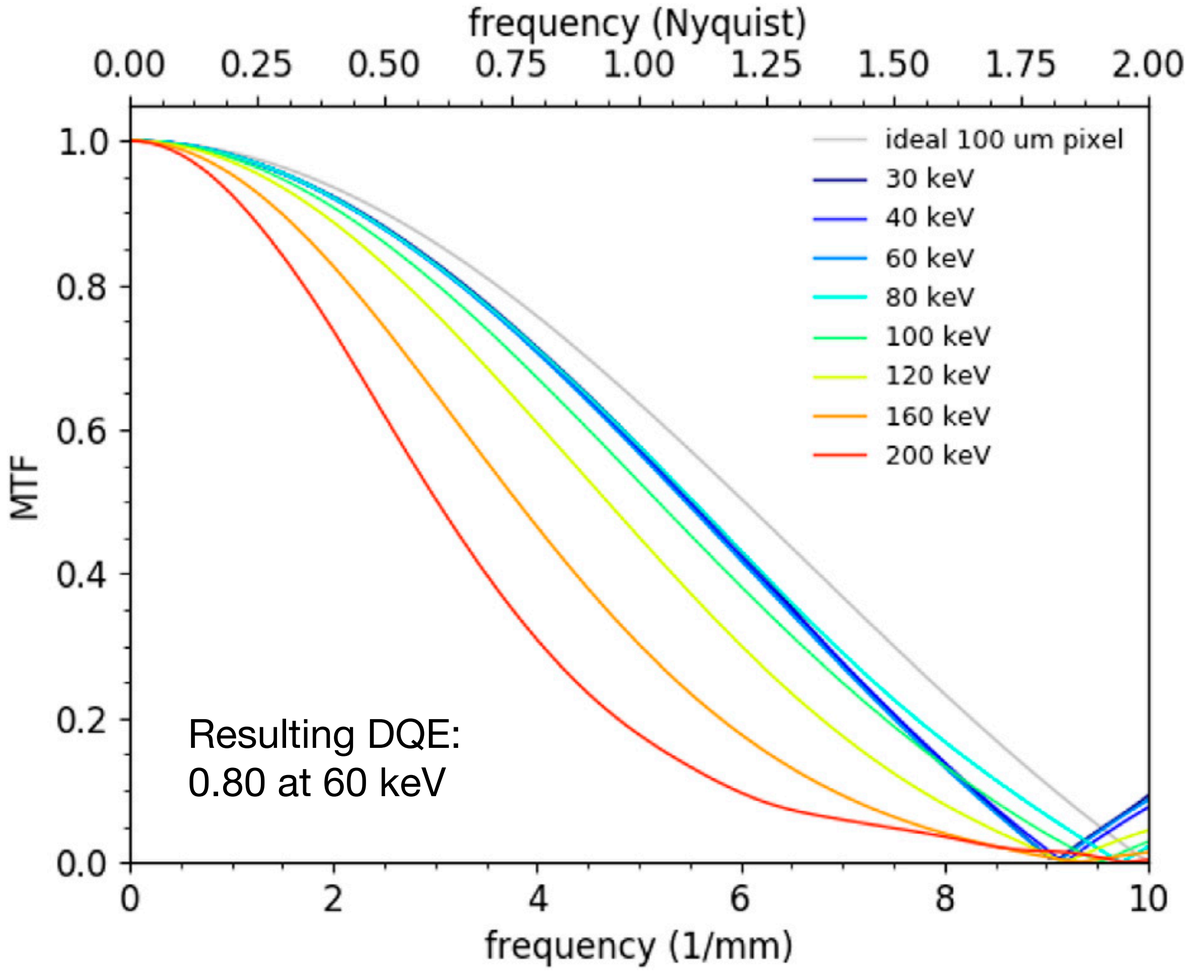
Near-ideal 4D-STEM performance at 60 keV

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

https://www.dectris.com/detectors/electron-detectors/for-materials-science/arina/

Near-ideal direct-electron 4D-STEM





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

4D-STEM data volumes

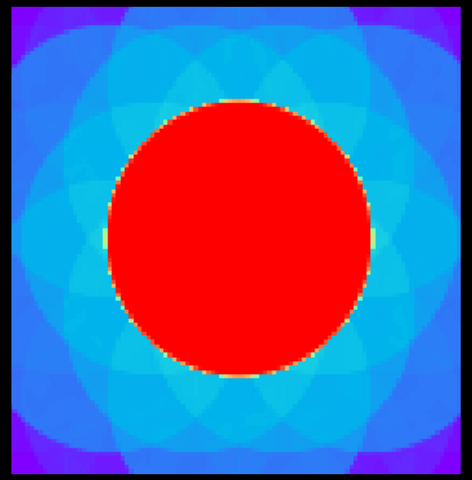
Data volumes collected during typical 4D-STEM experiments (assuming a 512 × 512 px scan).

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**

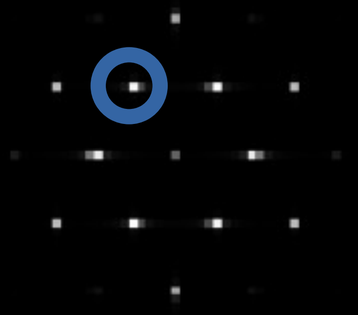
Huge data volumes can be collected in seconds (challenge for storage, processing…)

Ptychography can correct aberrations post-acquisition

PACBED

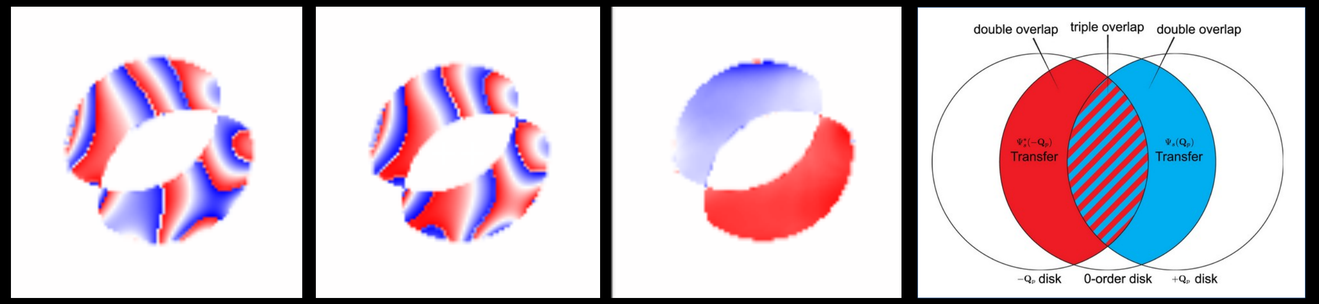


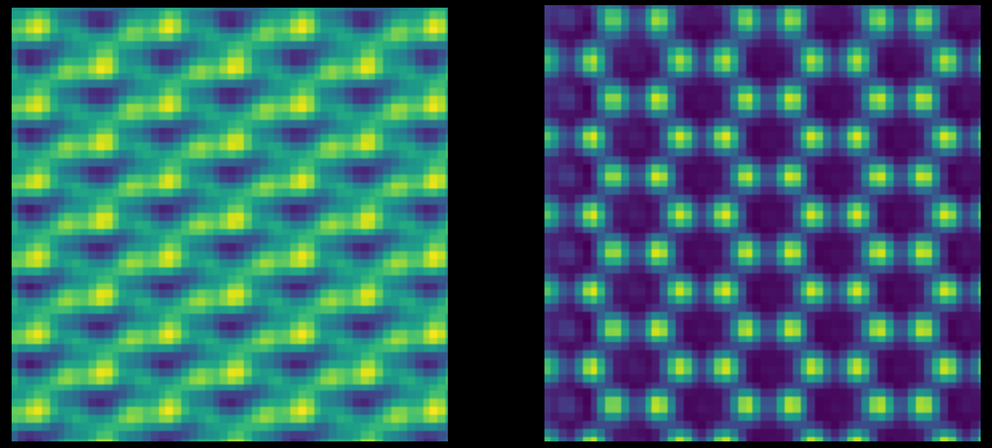
Spatial frequencies



A(K)*A(K-Q)

‘Exp’ SVD corrected Theory





SVD = singular value decomposition

Timothy Pennycook

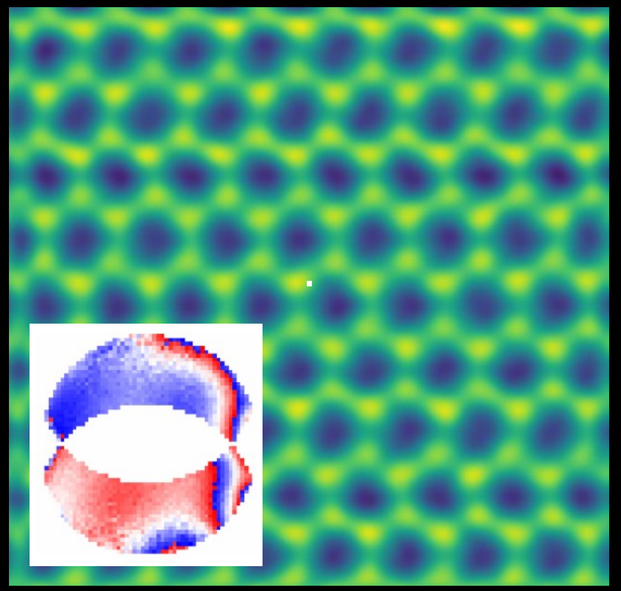
Christoph Hofer

Courtesy of Christoph Hofer

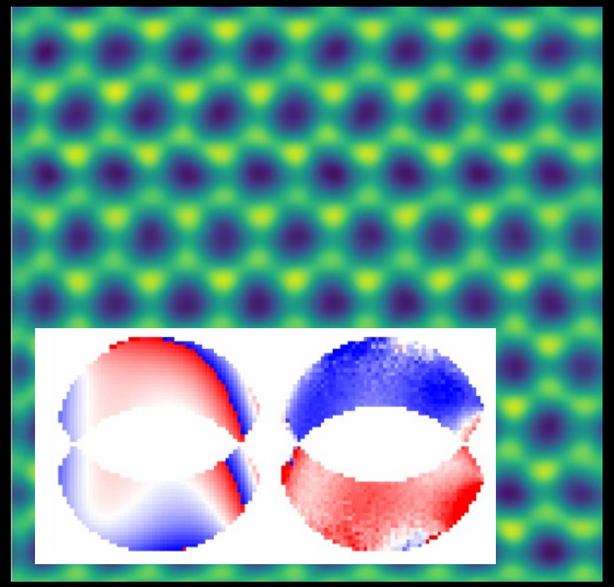
3rd order aberration correction might not be sufficient

Experimental data of monolayer graphene, 35 mrad (Vienna)

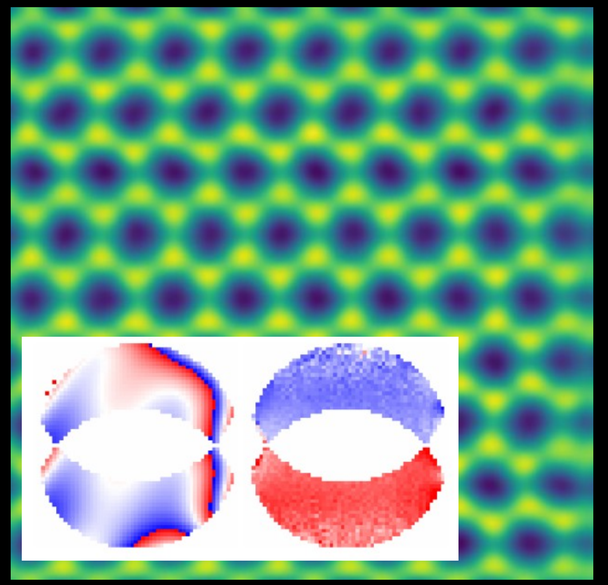
uncorrected



3rd order corrected



5th order corrected

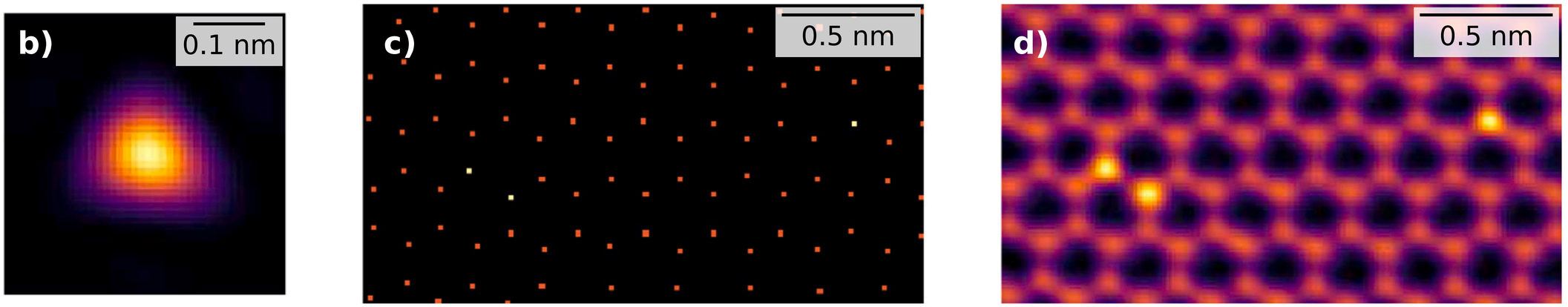


Timothy Pennycook

Christoph Hofer

Courtesy of Christoph Hofer

Site-specific phase via iterative optimization



Maximize correlation between experimental and simulated image

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

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

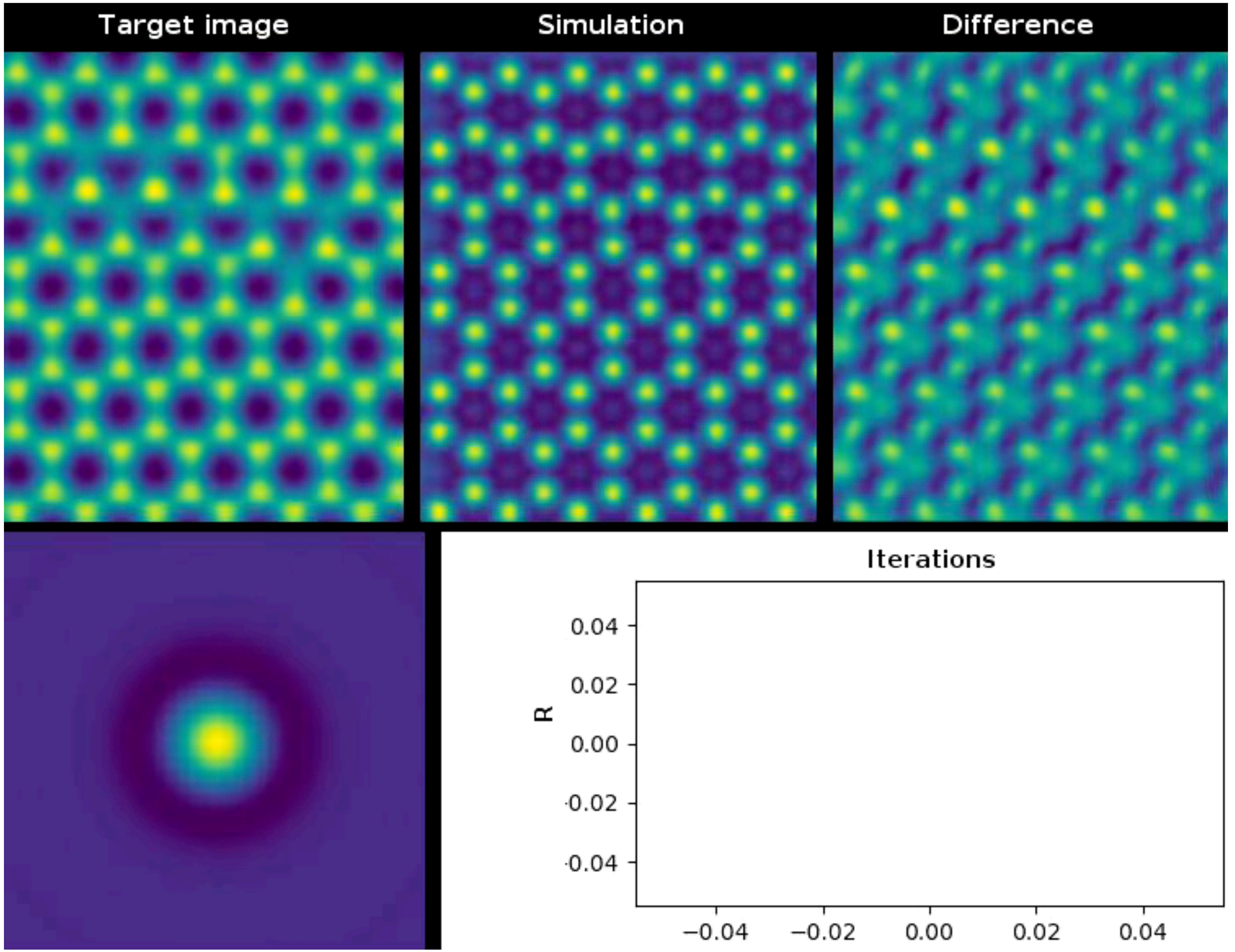
Phase analysis

STEM_Optimization*
(commit 6c549ba9)

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

* https://gitlab.com/christoph_hofer/stem_optimization

Defective WS_2



Can account for:

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

Universiteit Antwerpen

Timothy Pennycook

Christoph Hofer

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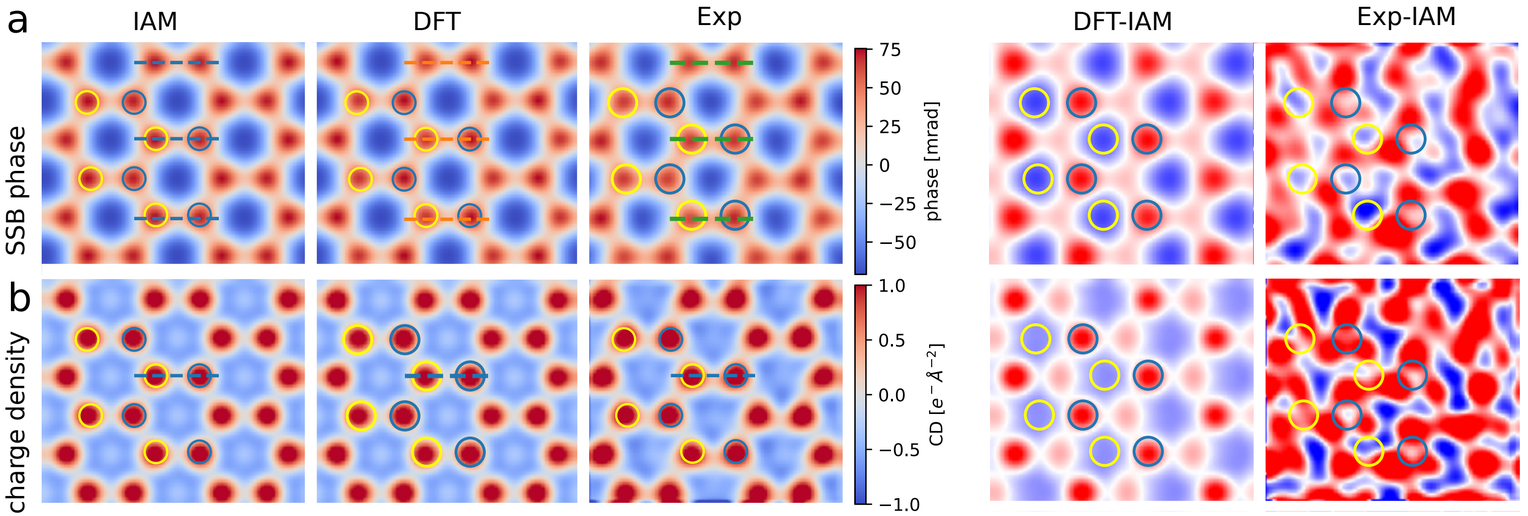
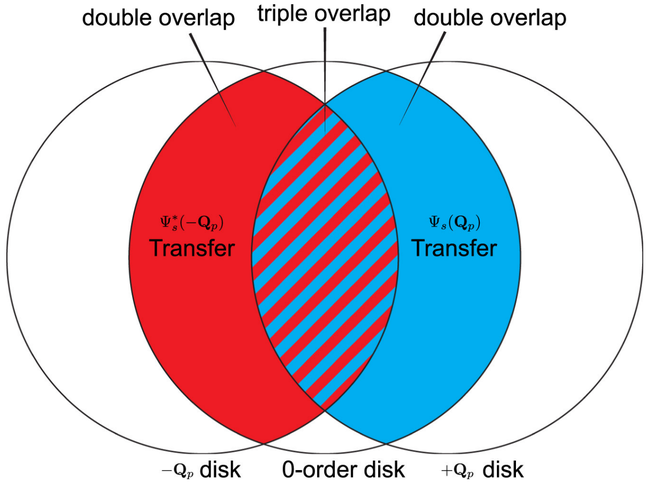
C. Hofer et al., Ultramicroscopy 227 (2021) 113292

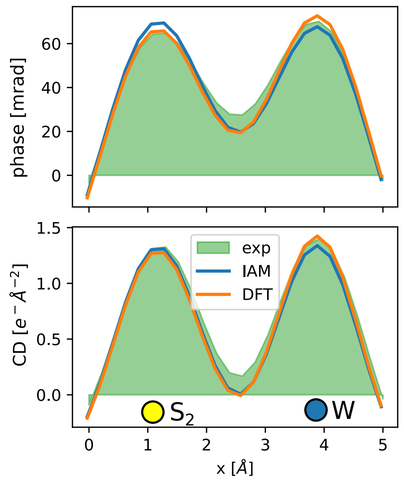
Ptychography for charge transfer in defective WS_2

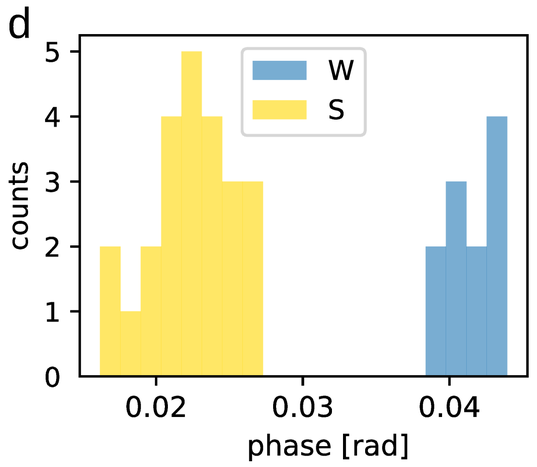


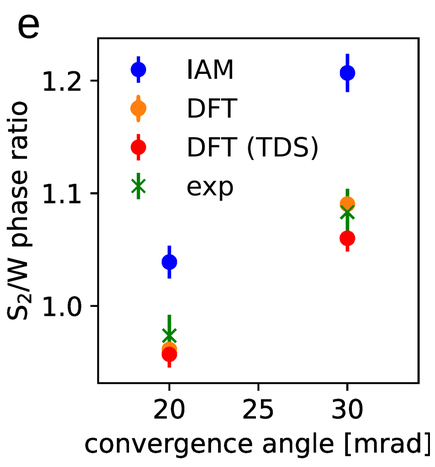
FEI Themis Z, 60 keV, Timepix3

Timepix3 limited to ~2 pA...









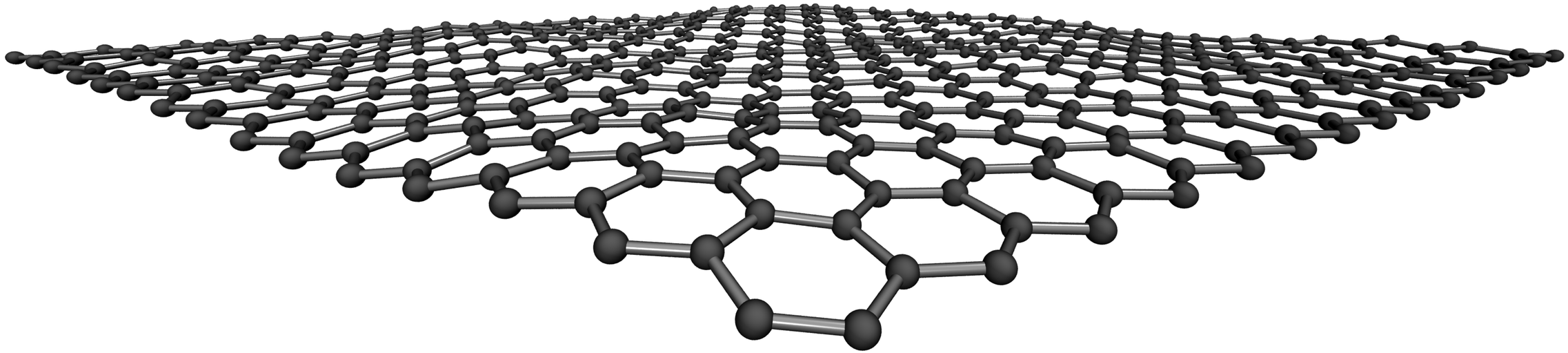
C. Hofer, J. Madsen, **TS**, T. Pennycook, arXiv:2301.04469v4 (2024)

Graphene: ideal uniform phase object

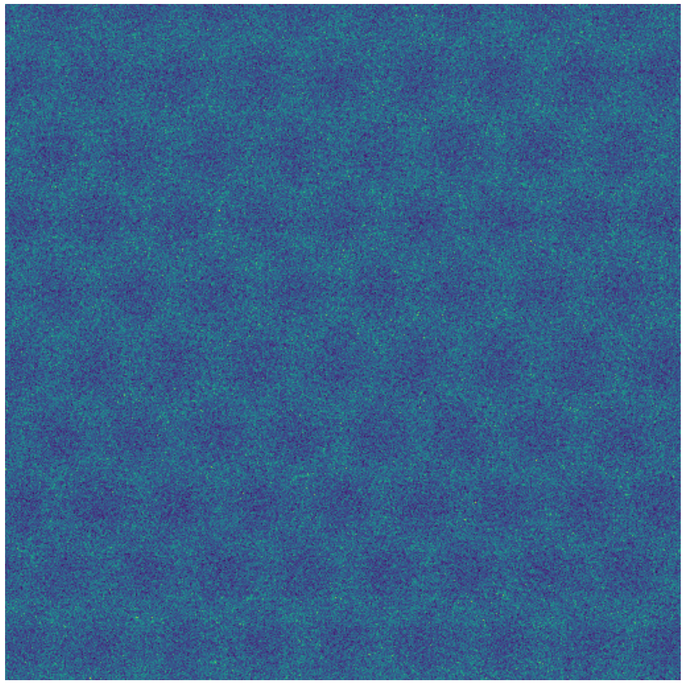
✅ ∞ radiation hardness <80 keV ⇒ no dose limitation

✅ low-*Z*, one-atom-thick ⇒ perfect weak phase object

✅ only one element ⇒ each atomic site identical



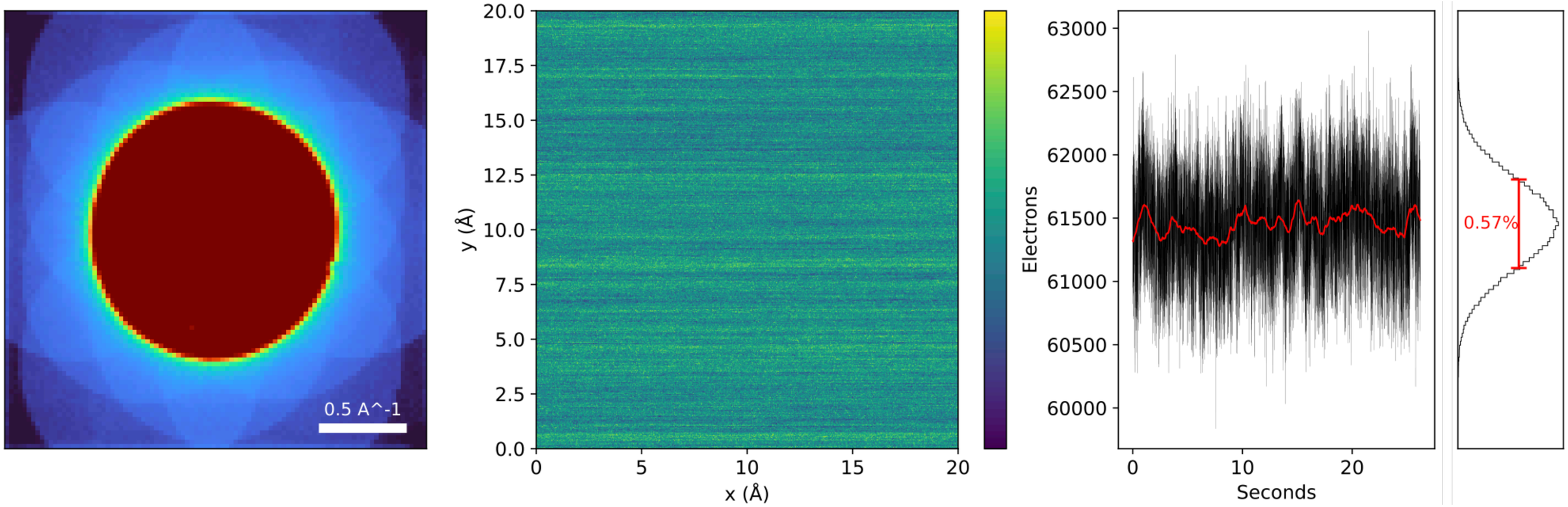
Nion + ARINA 4D-STEM data collection (original)



HAADF (80–300 mrad)

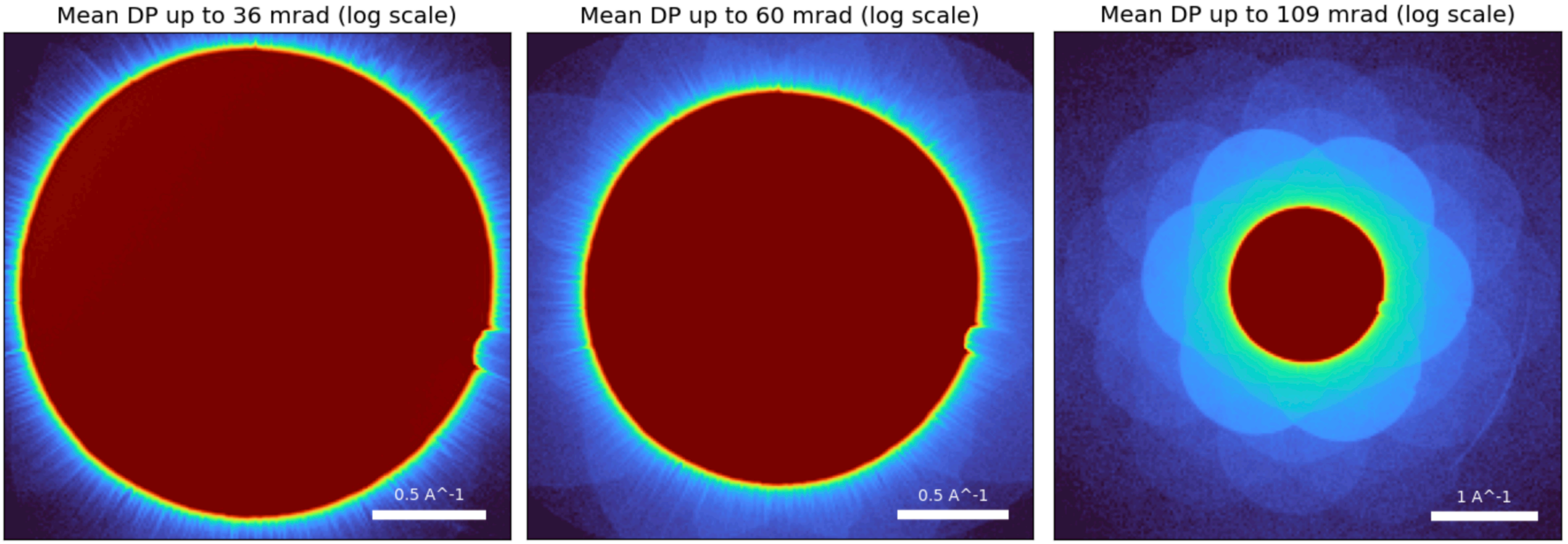
Nion + ARINA 4D-STEM data collection (original)

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



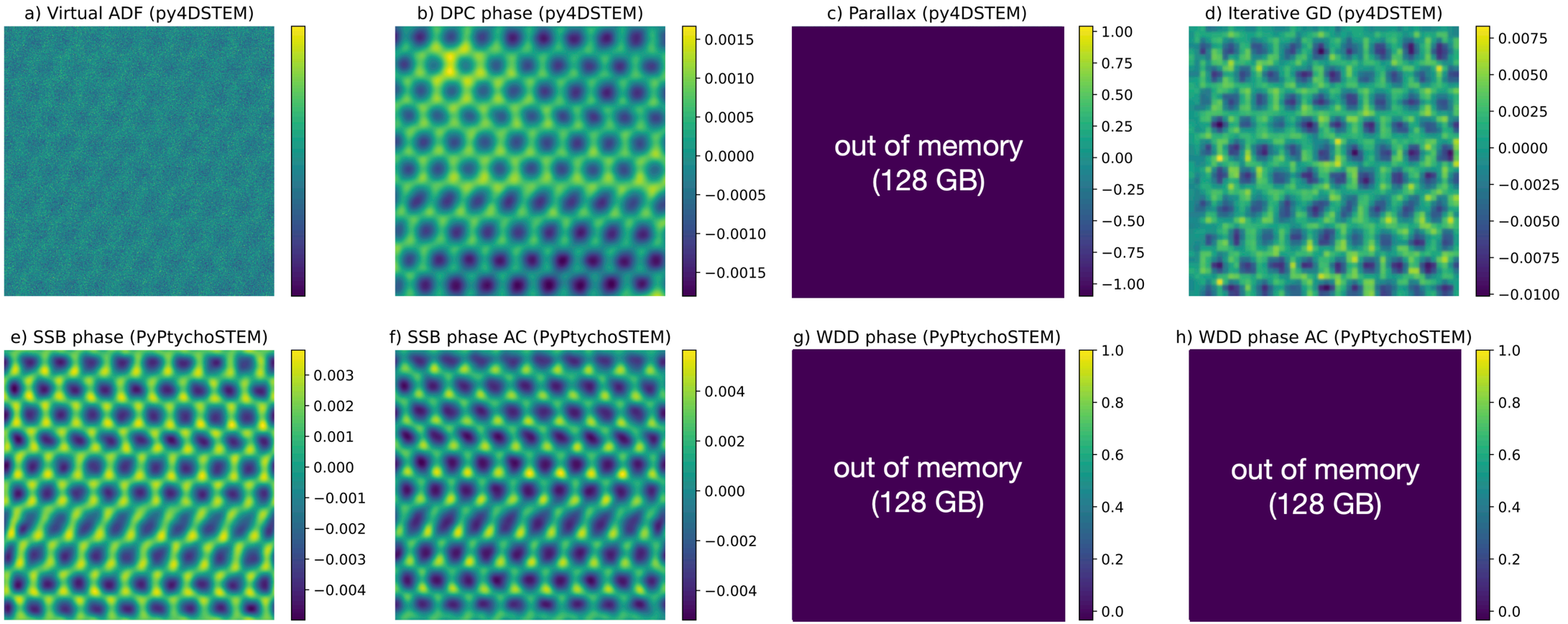
- 34 mrad semi-convergence angle, ~70 mrad max collection angle, ~8 pA
- counting every electron, corrected dose per pixel can be normalized

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



~200 pA current!

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

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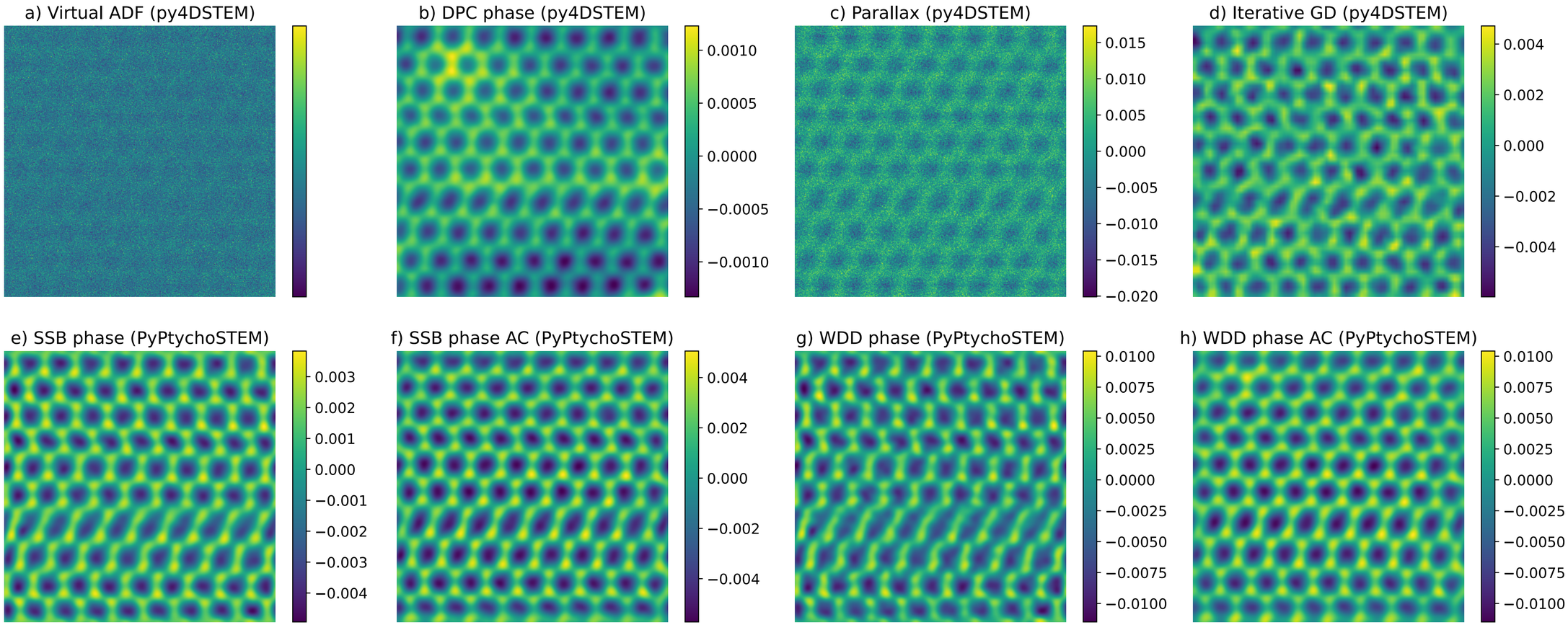
SSB & WDD: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167
DPC, Parallax, Iterative GD: G. Varnavides *et al.*, arXiv:2309.05250v2 (2024)

Comparing phase reconstruction algorithms (Q binning)



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

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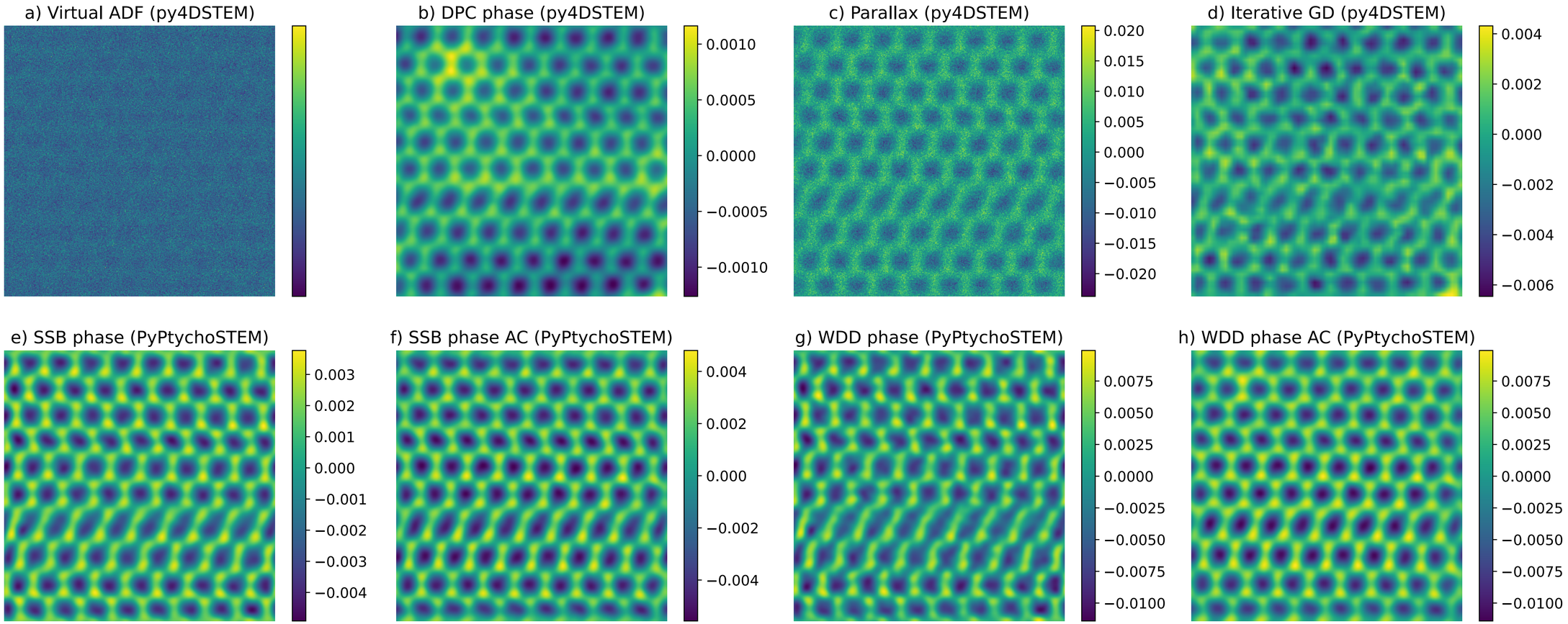

36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 192×192 Qpix
4× binning in Q

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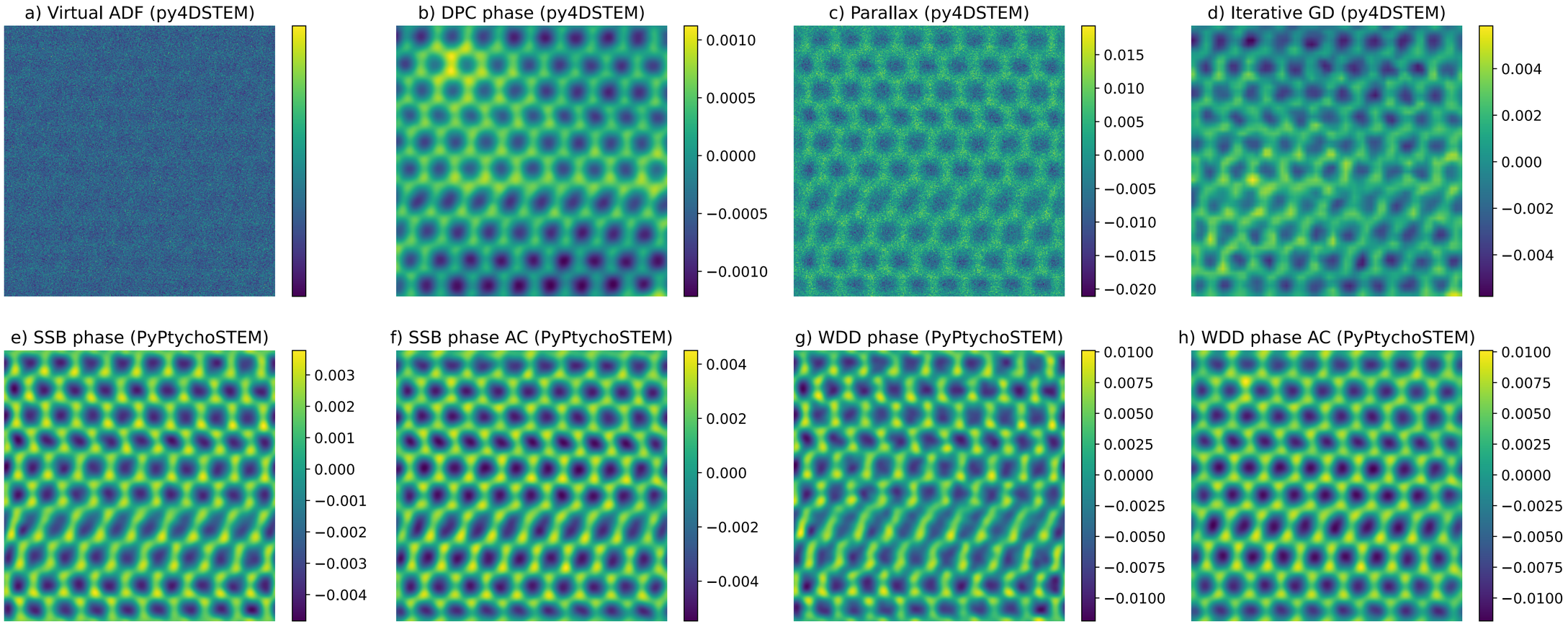
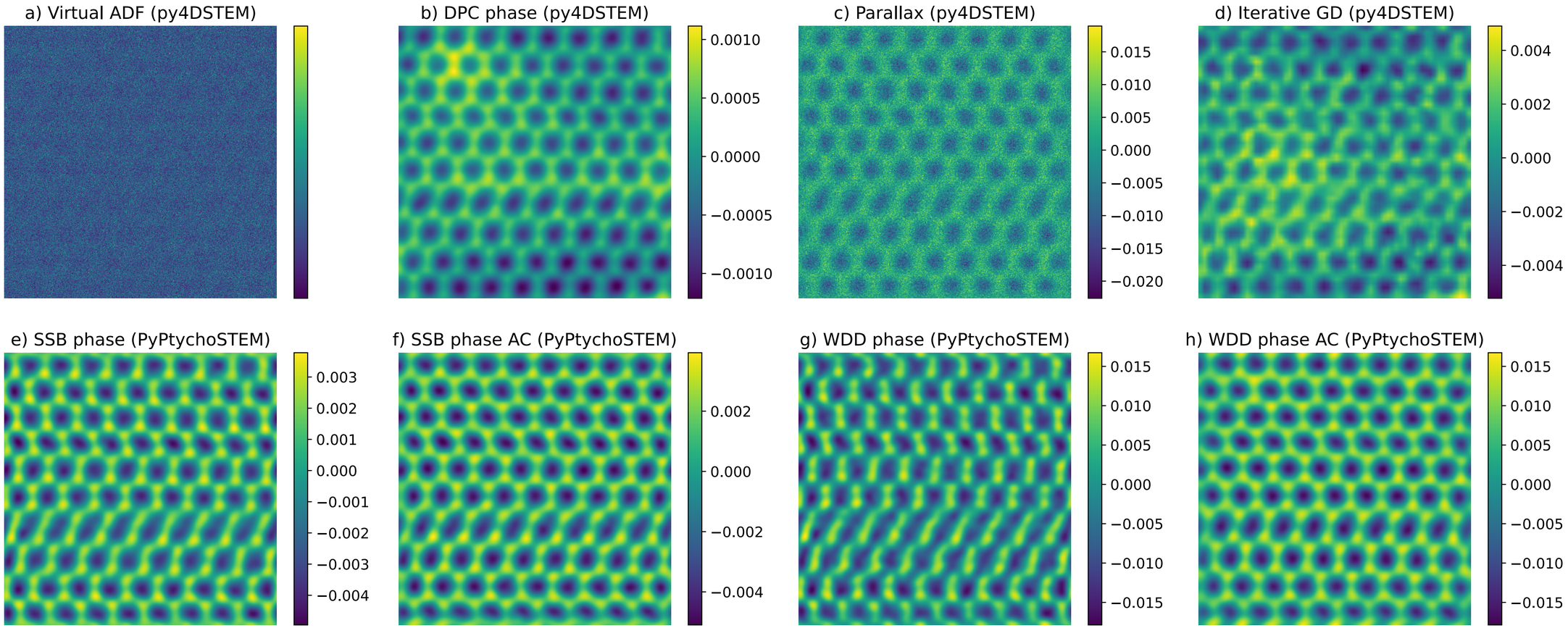
Comparing phase reconstruction algorithms (Q binning)



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

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 (Q binning)



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

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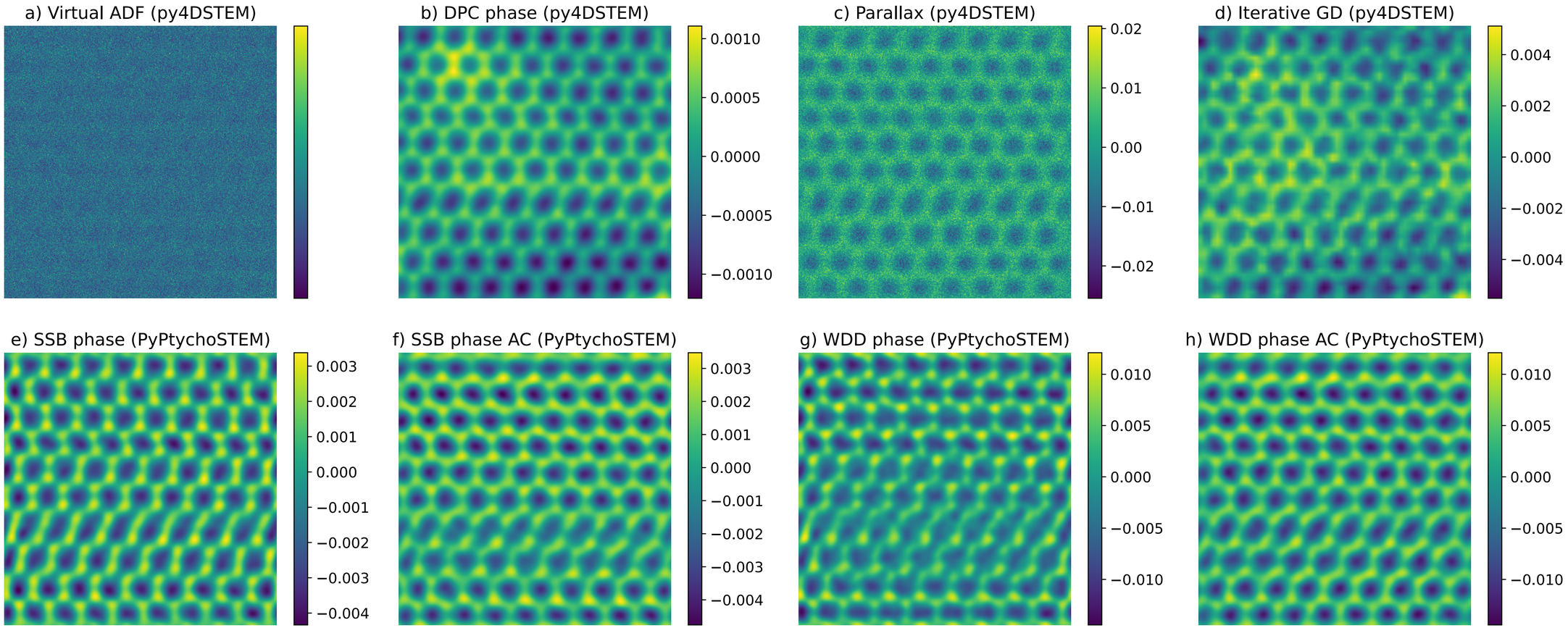
Comparing phase reconstruction algorithms (Q binning)



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

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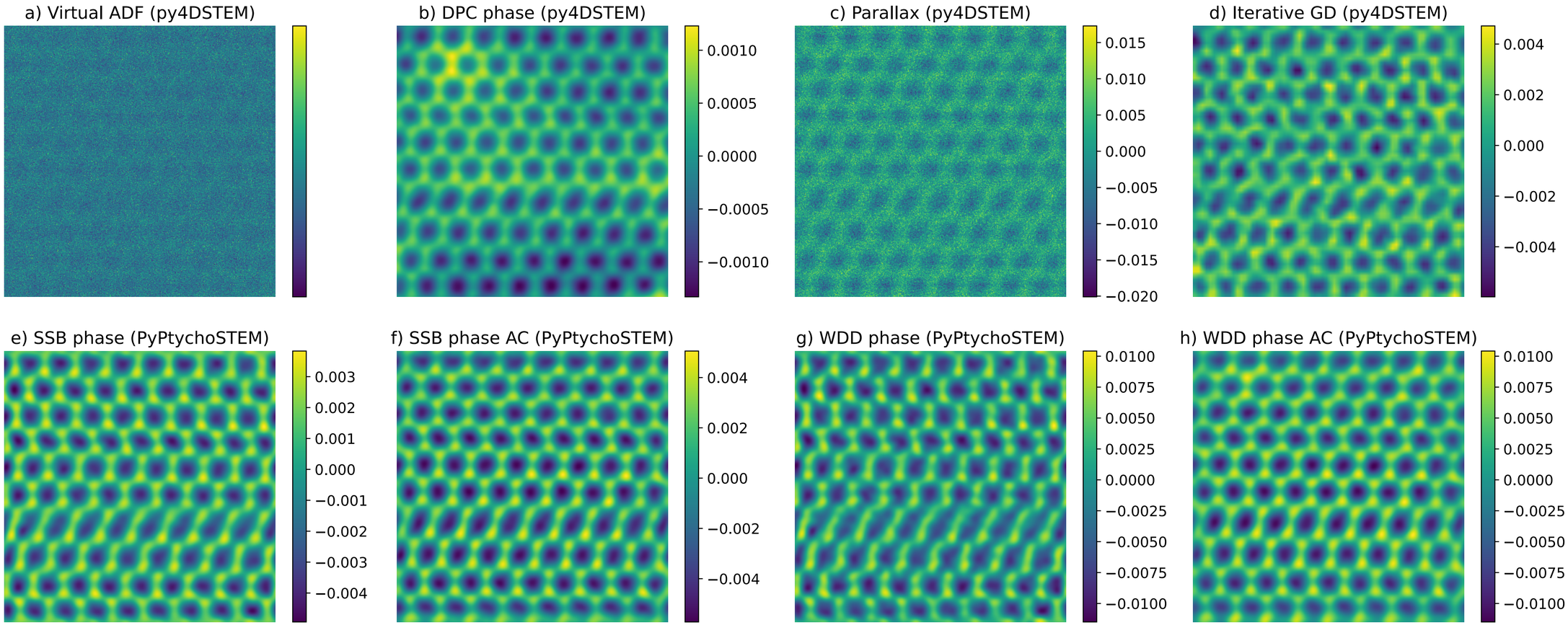
SSB & WDD: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167
DPC, Parallax, Iterative GD: G. Varnavides *et al.*, arXiv:2309.05250v2 (2024)

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

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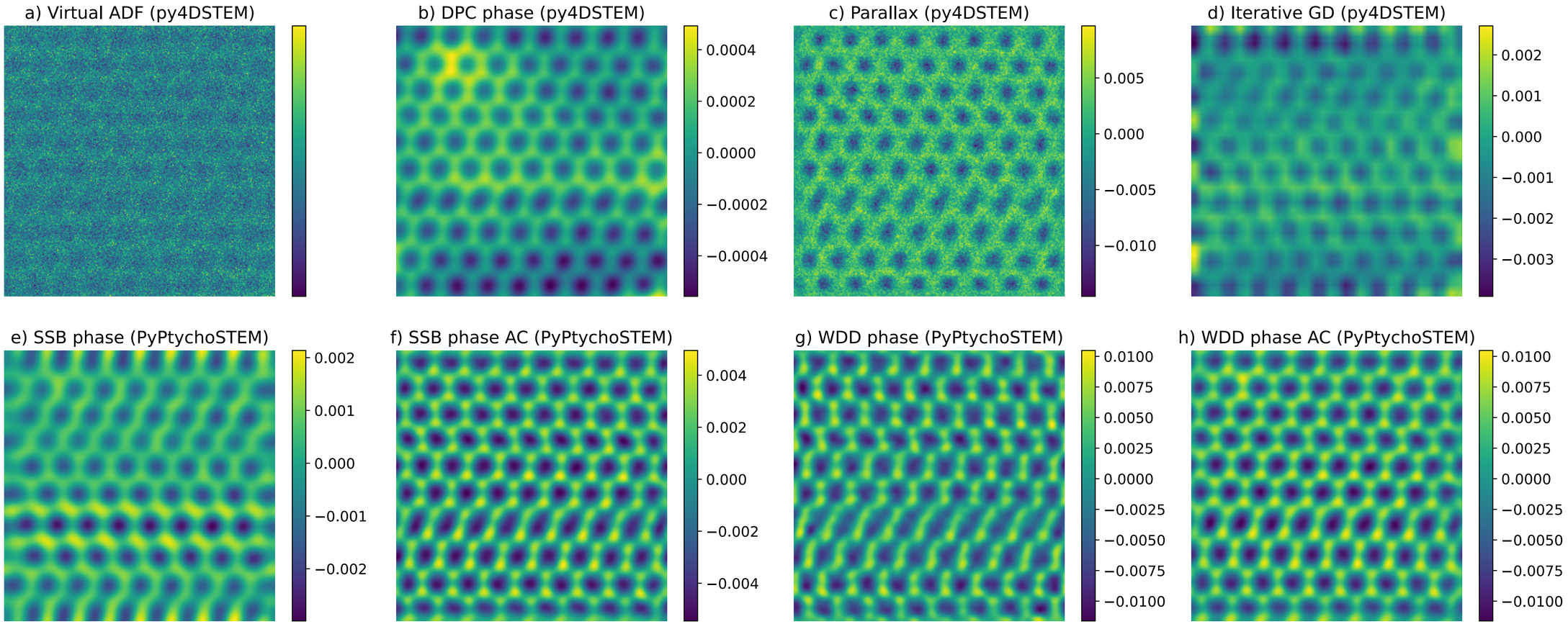
SSB & WDD: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167
DPC, Parallax, Iterative GD: G. Varnavides *et al.*, arXiv:2309.05250v2 (2024)

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)
2× binning in R

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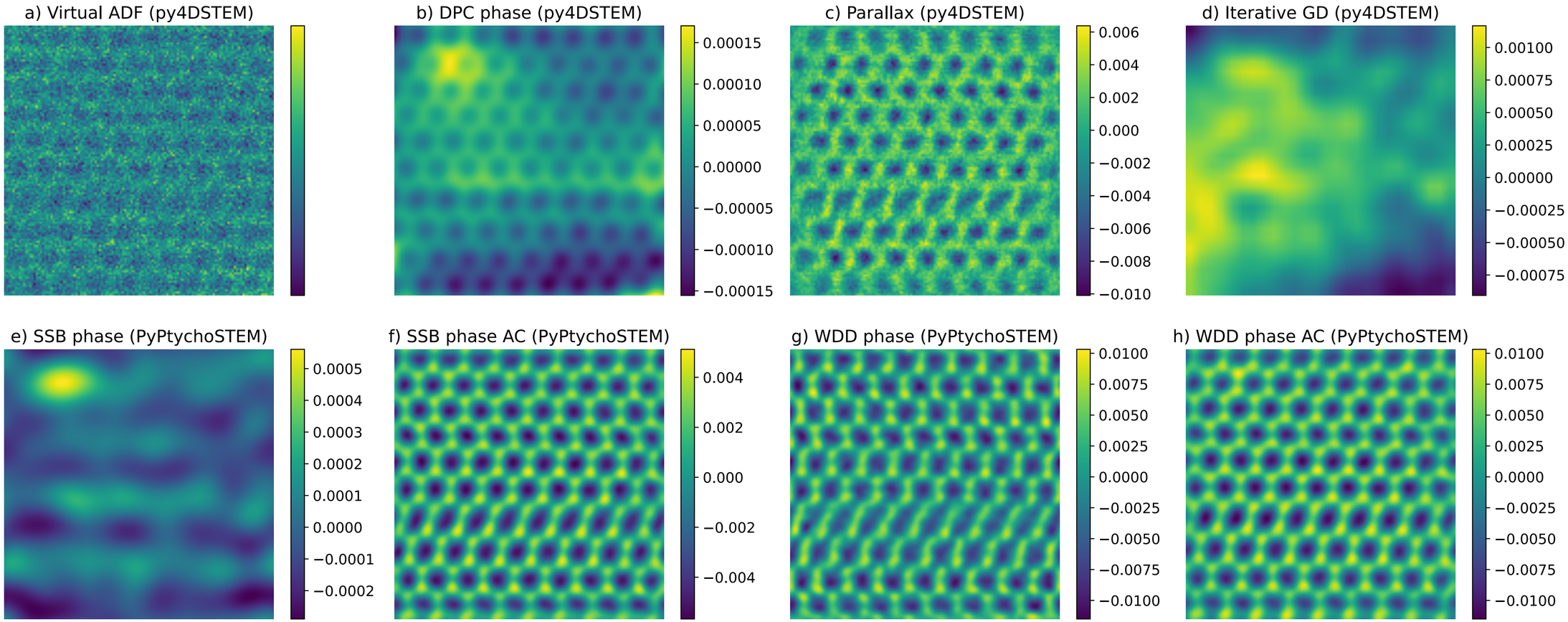
Comparing phase reconstruction algorithms (R binning)



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4× binning in R

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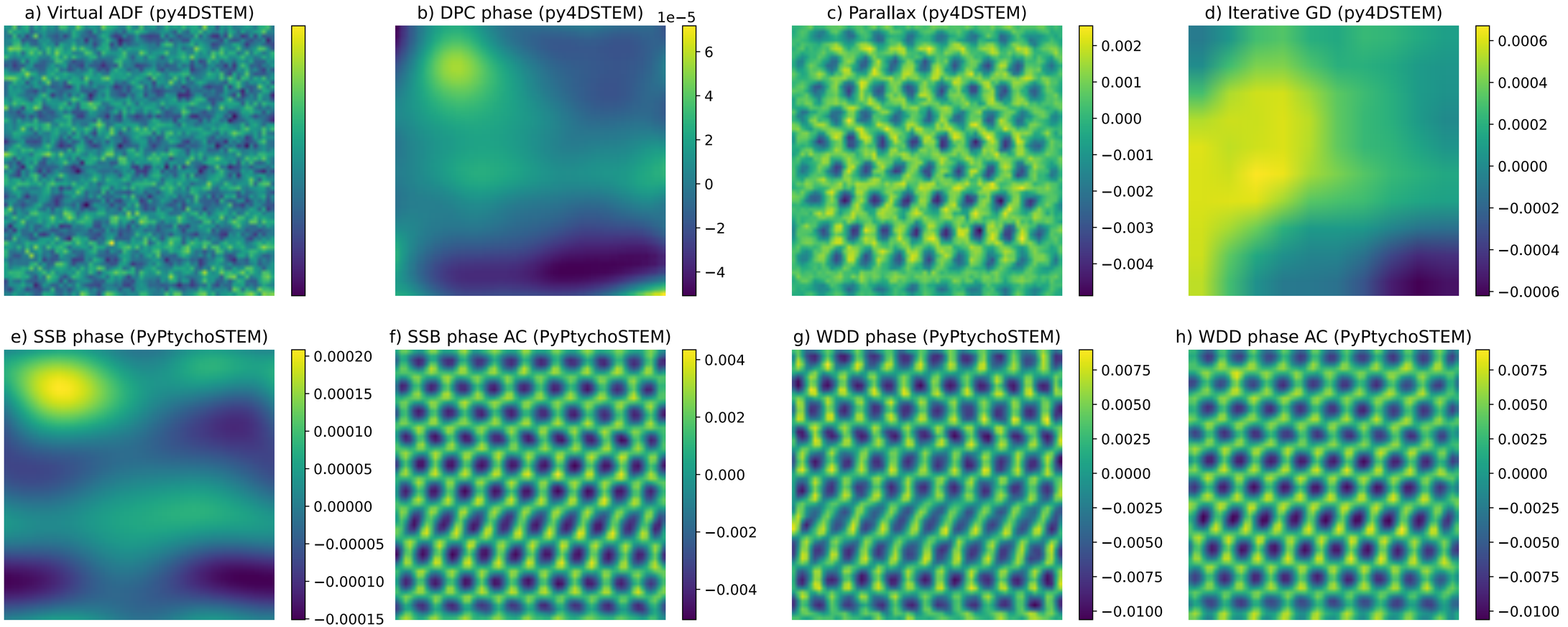
DPC, Parallax, Iterative GD: G. Varnavides *et al.*, arXiv:2309.05250v2 (2024)

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)
8× binning in R

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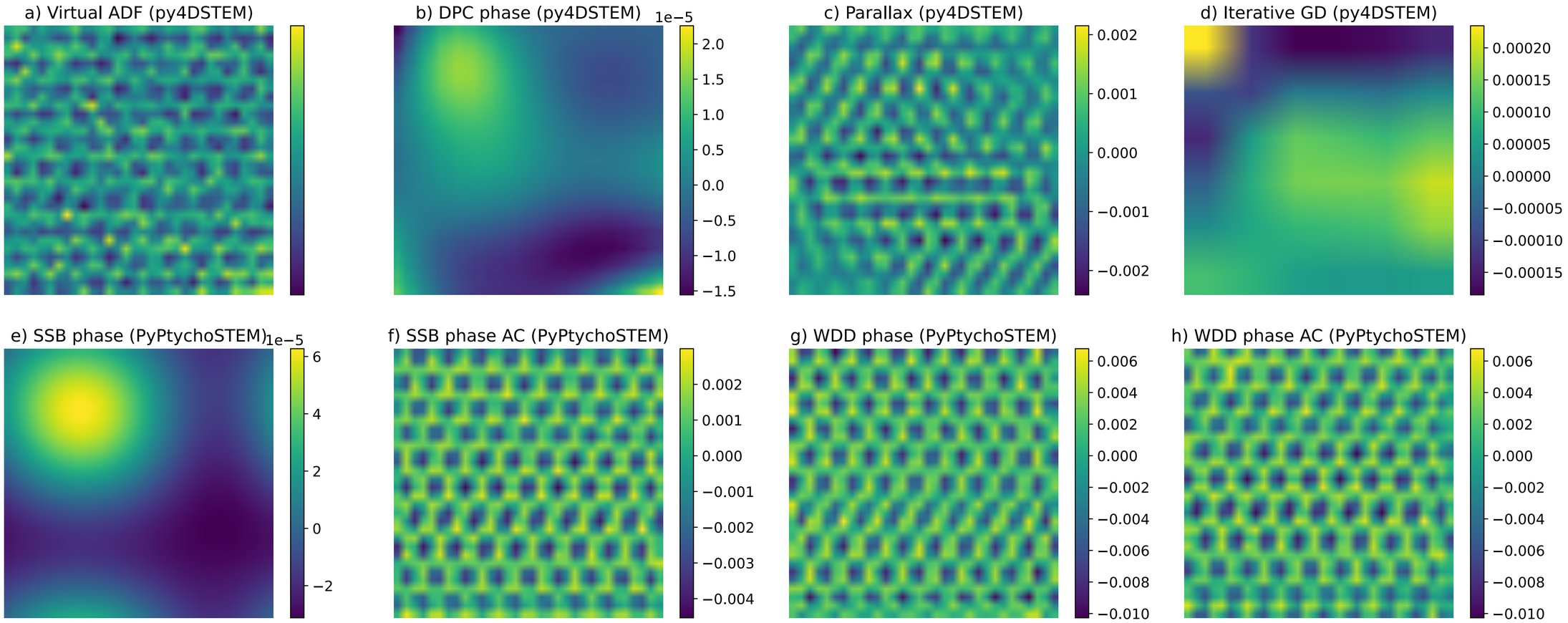
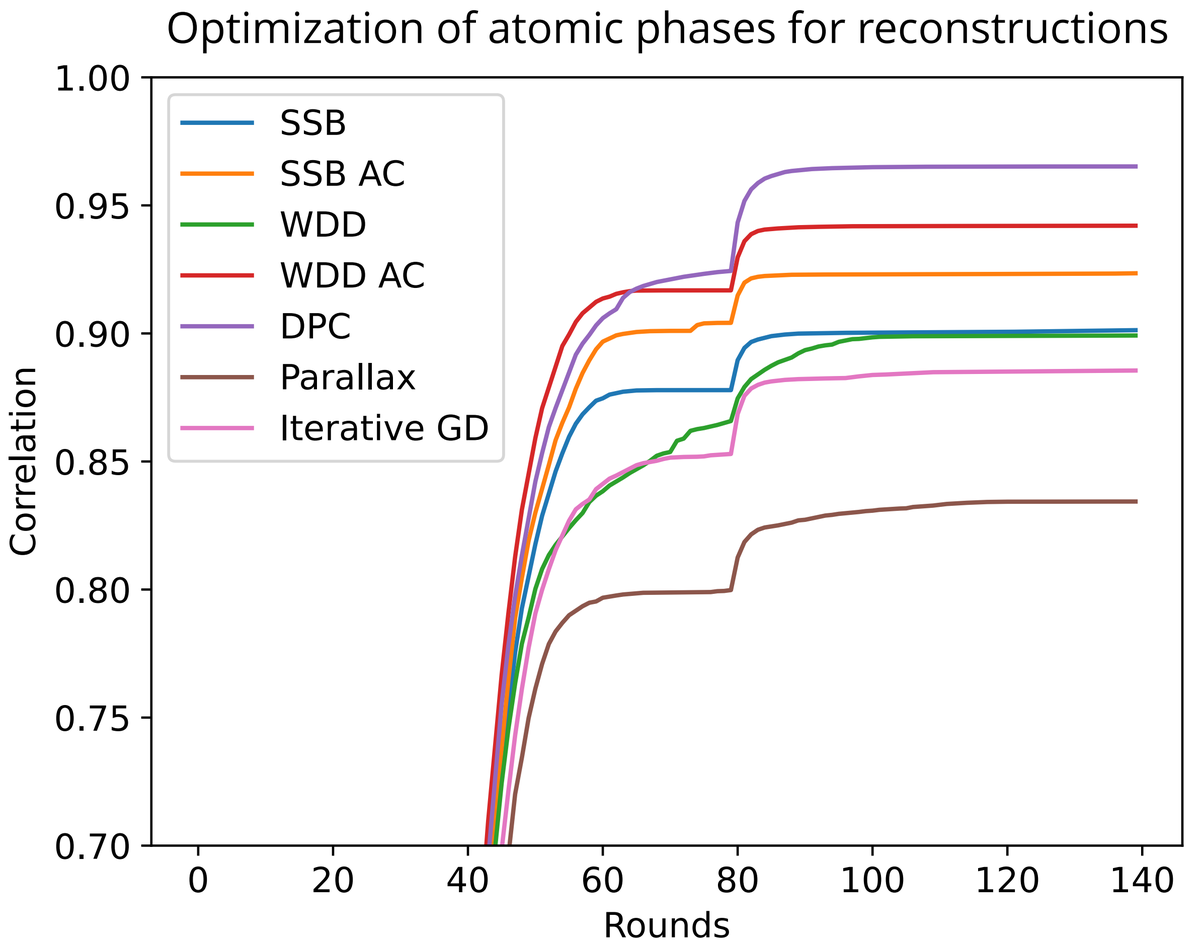
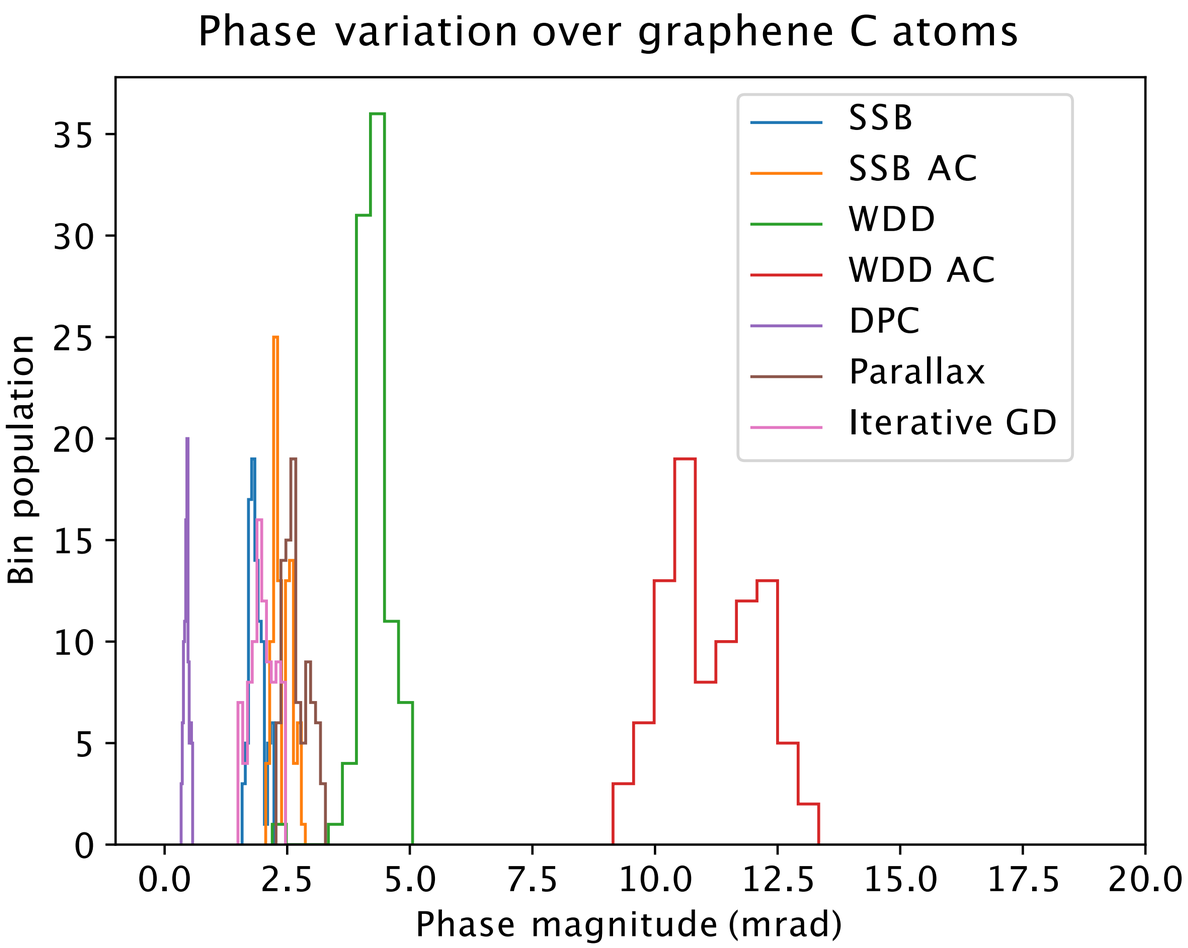


36 mrad maximum scattering angle, 33.3 us dwell time, 512×512 Rpix, 96×96 Qpix (HW binQ by 2)
16× binning in R

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

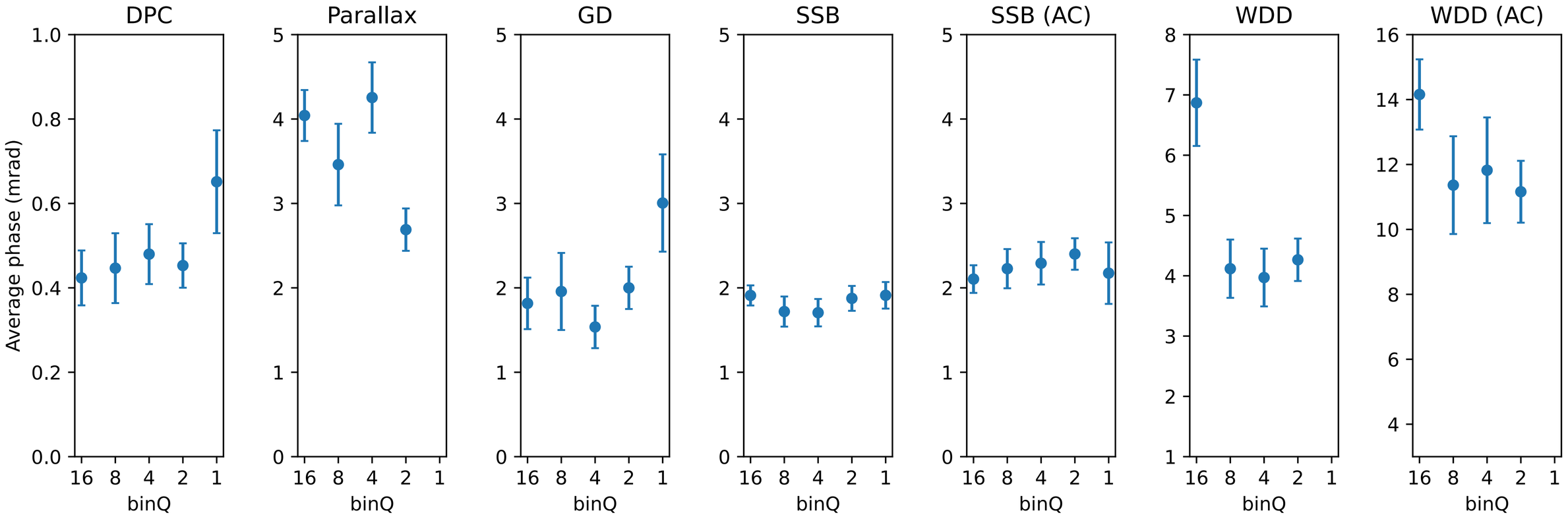
Atomic phase assignment





T. Susi, unpublished

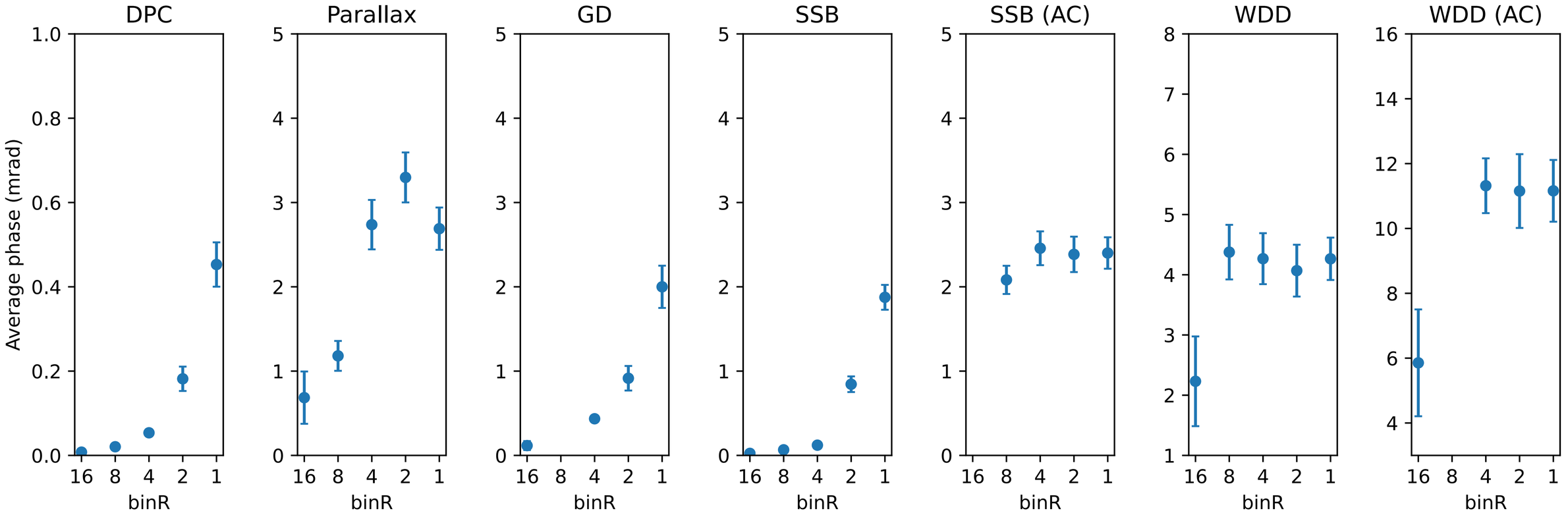
Quantification of phase variation



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

T. Susi, unpublished

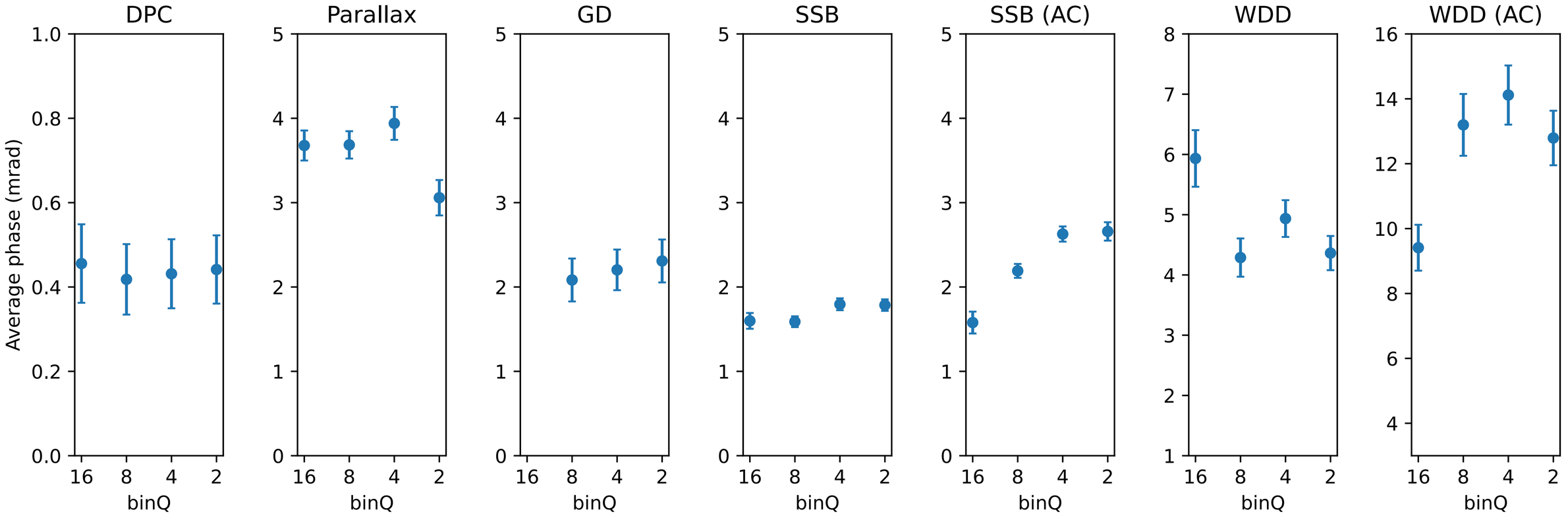
Quantification of phase variation



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

T. Susi, unpublished

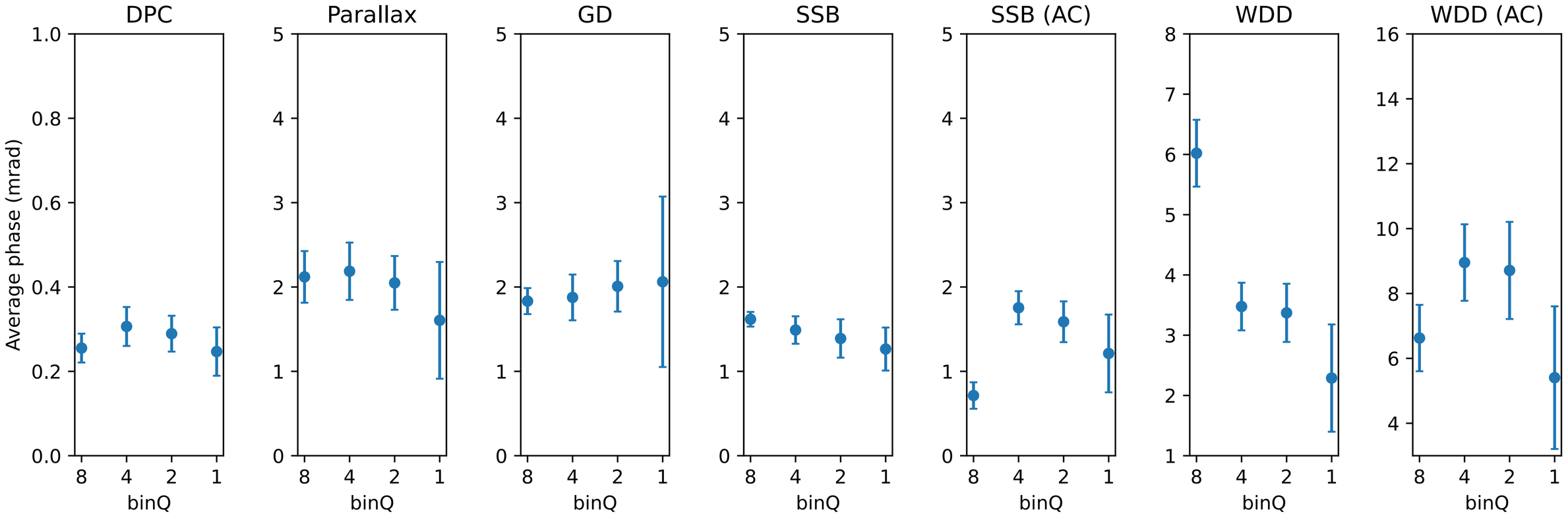
Quantification of phase variation



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

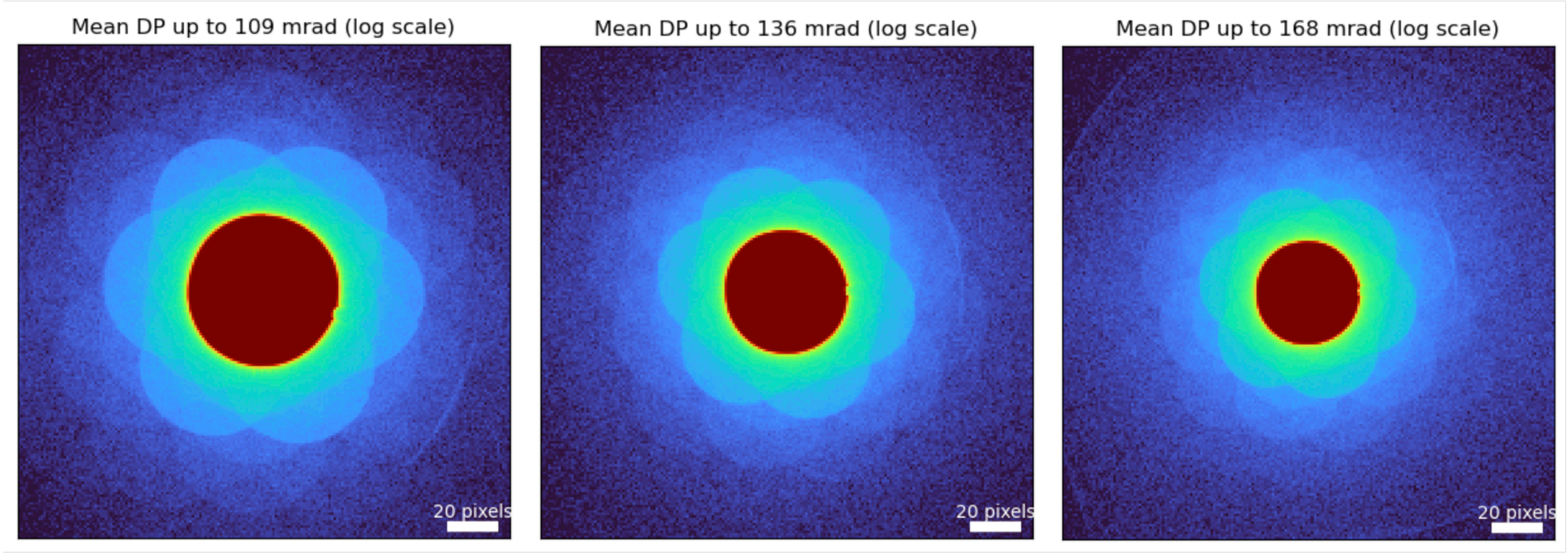
T. Susi, unpublished

Computational cost (36 mrad datasets)

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

† GPU-accelerated (NVidia RTX4090); varying Q-padding
‡ Slightly different numbers of alignment iterations

(Some) 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*

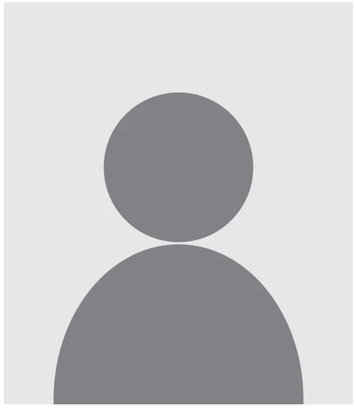
T. Susi, unpublished

Upcoming Japan stay (Sep 2024 – Feb 2025)

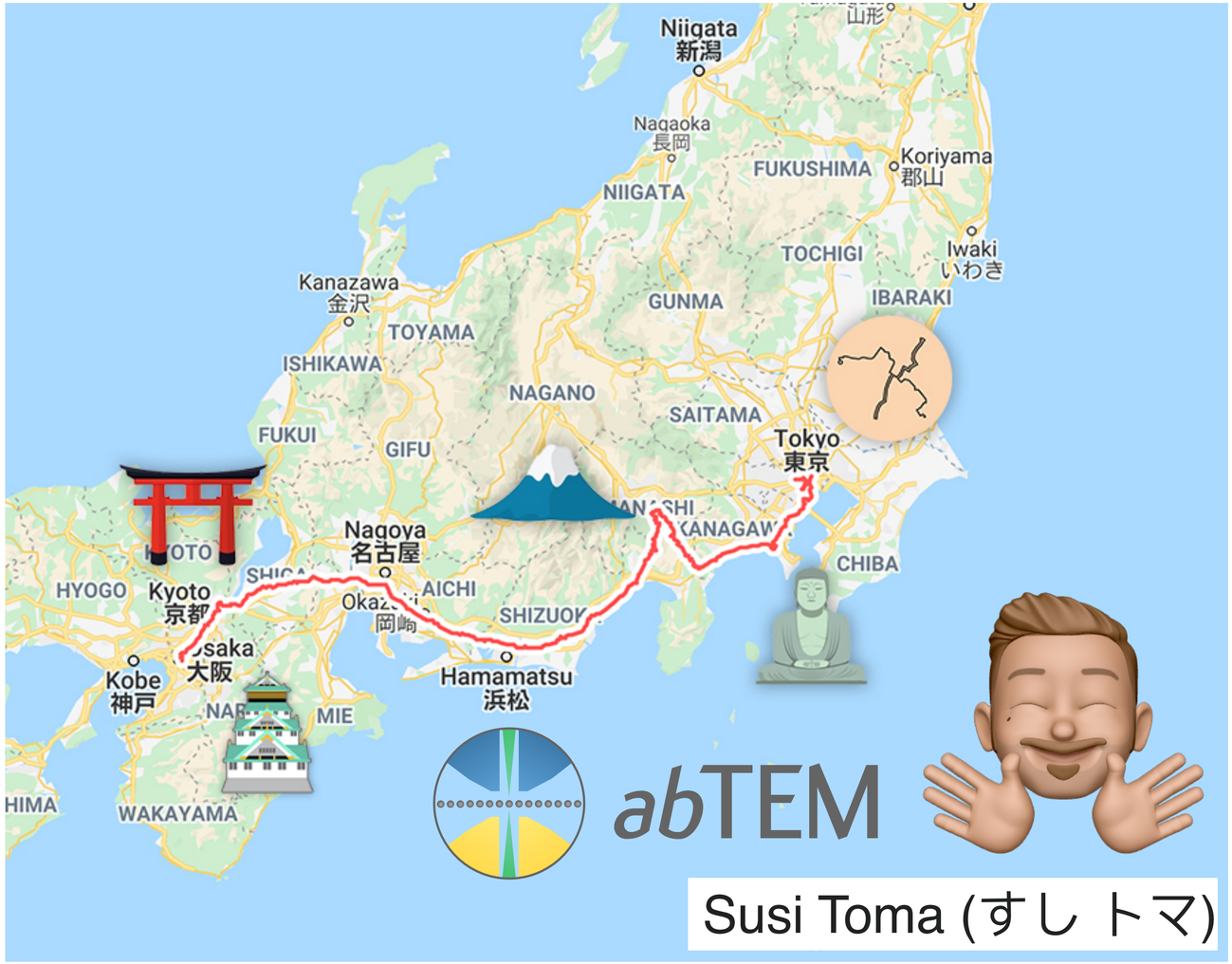
Nagashima (TUS, Sep)

Suenaga (Osaka, Oct)

Shibata (Tokyo, Nov–)



you? (???)



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FWF Der Wissenschaftsfonds.

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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