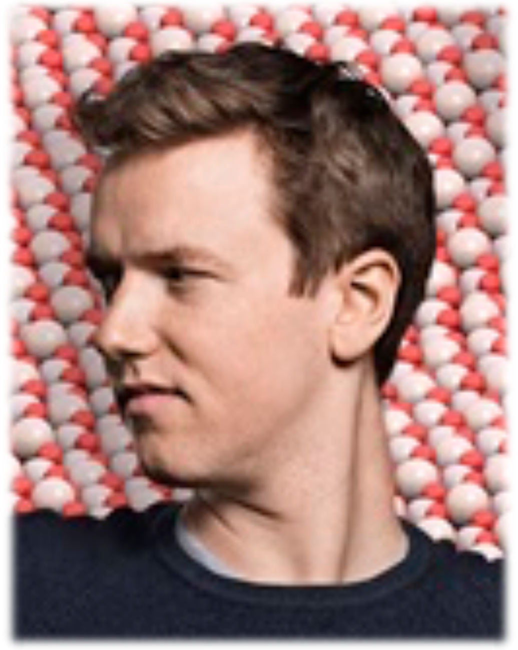
Near-Ideal Direct-Electron Focused-Probe 4D-STEM Data for
Open-Source Phase Reconstructions

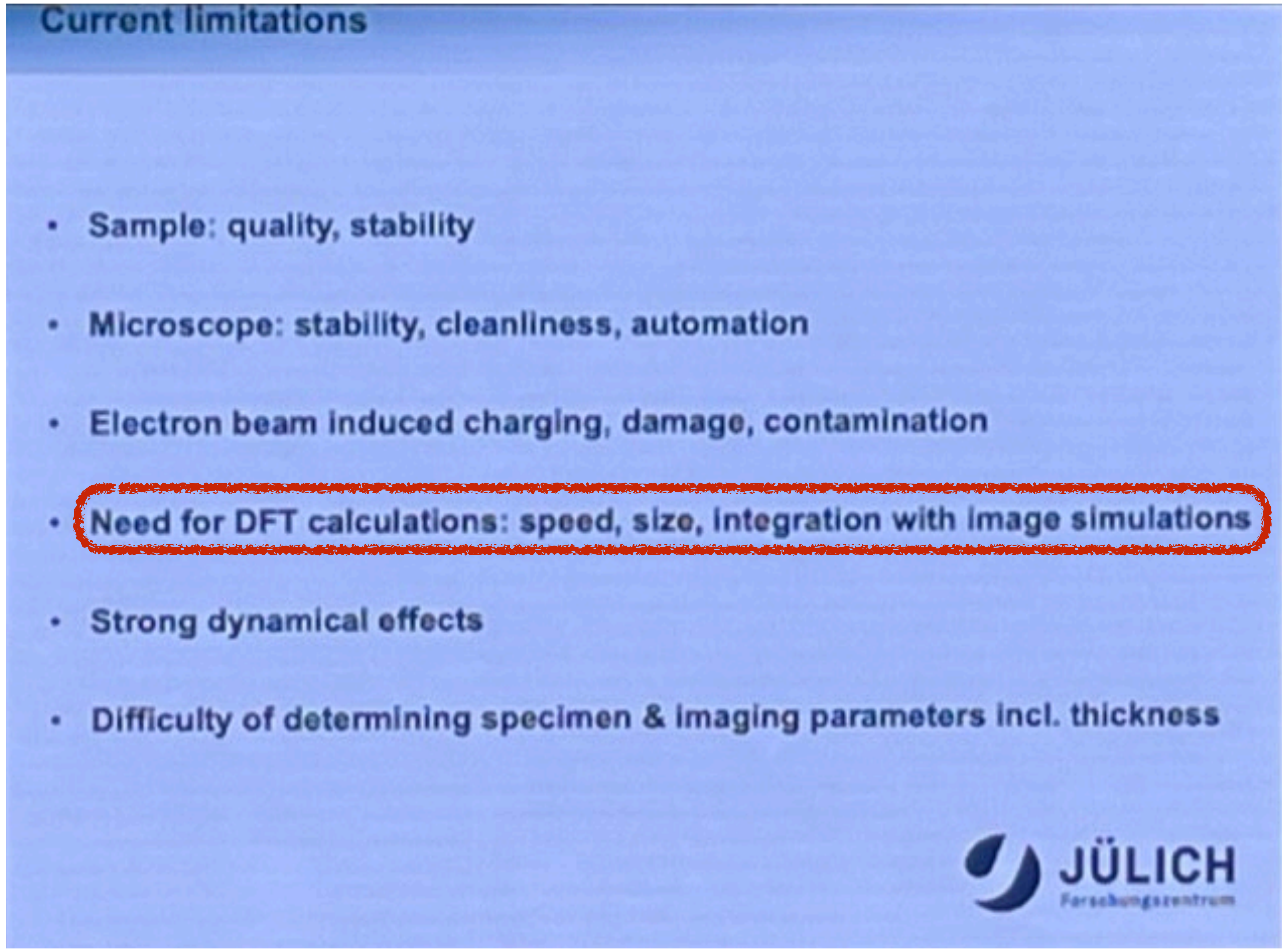
Toma Susi, Jani Kotakoski, Clemens Mangler (U. Vienna)
Christoph Hofer, Timothy Pennycook (U. Antwerp)
Niklas Dellby, Russ Hayner, Tracy Lovejoy, Andreas Mittelberger, Benjamin Plotkin-Swing (Nion Co. R&D / Bruker AXS)

My favourite hobby project: abTEM



Jacob Madsen

Simulation challenge for phase-contrast



Rafal Dunin-Borkowski @ Salve 2D22

Our multislice solution

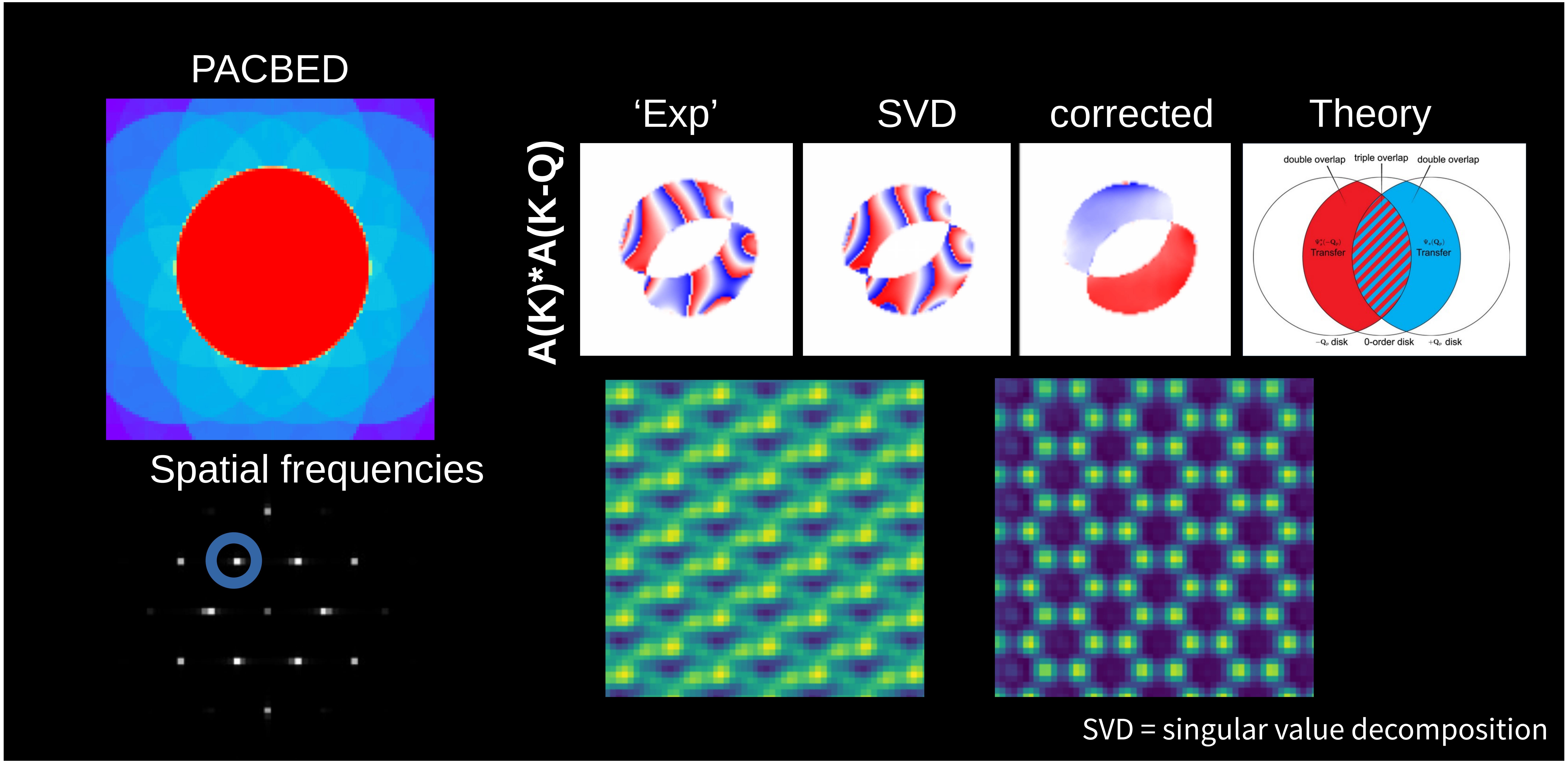
1. Integration with *ab initio* codes (ASE, GPAW!)
2. Python, but *fast* (CuPy, Numba)
3. Promote notebook/scripted workflow (jupyter)
4. Access to low-level API: Easy to extend
5. Open source (huge ecosystem!) (DASK)
6. High-performance parallel GPUs & file access (NVIDIA CUDA, Zarr)

Major upcoming & planned features:

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

(Bright-field) ptychography & aberration correction

Universiteit Antwerpen

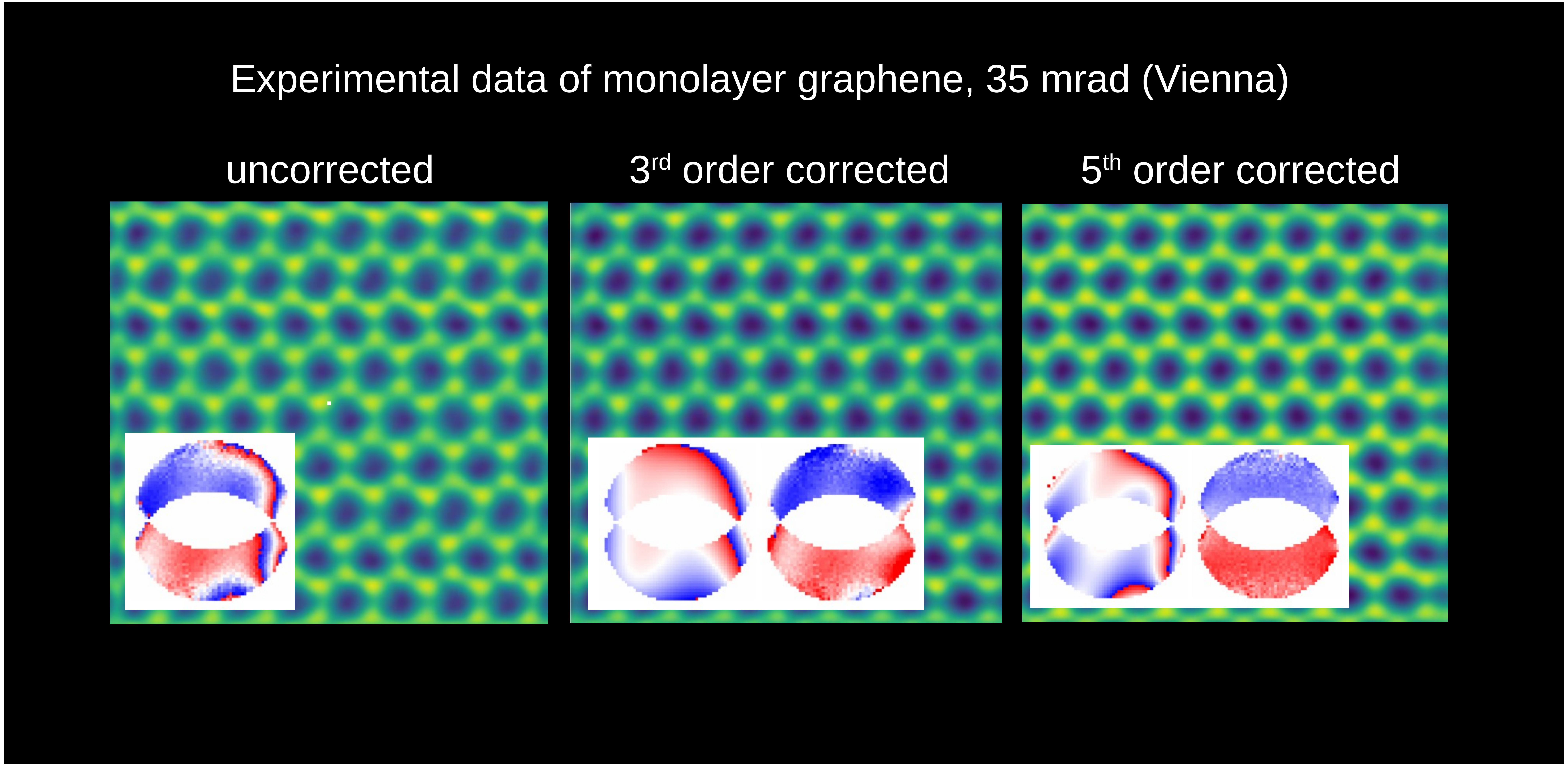


Timothy Pennycook

Christoph Hofer

Post-acquisition aberration correction (5th order)

Universiteit Antwerpen



Timothy Pennycook

Christoph Hofer

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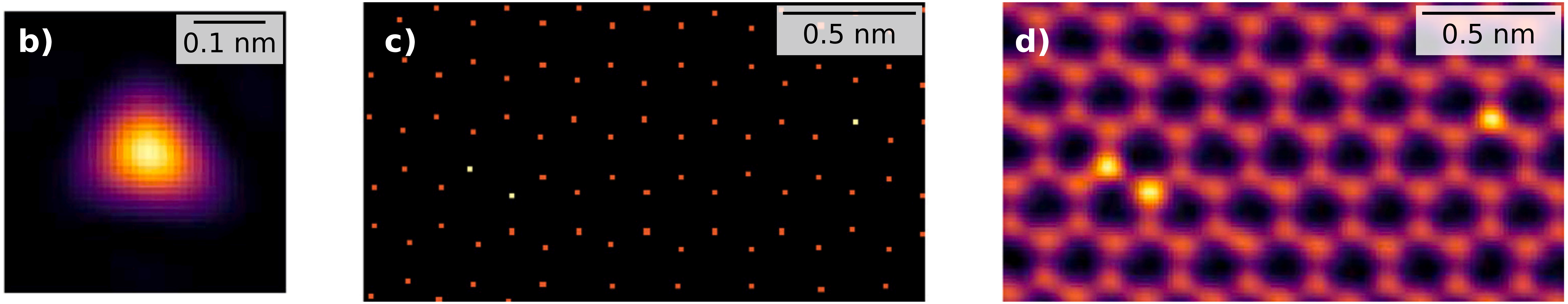
Courtesy of Christoph Hofer

Site-specific phase via iterative optimization



Maximize correlation between experimental and simulated image

Can account for:

- ~~aberrations~~
  (not used later in this talk!)
- 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

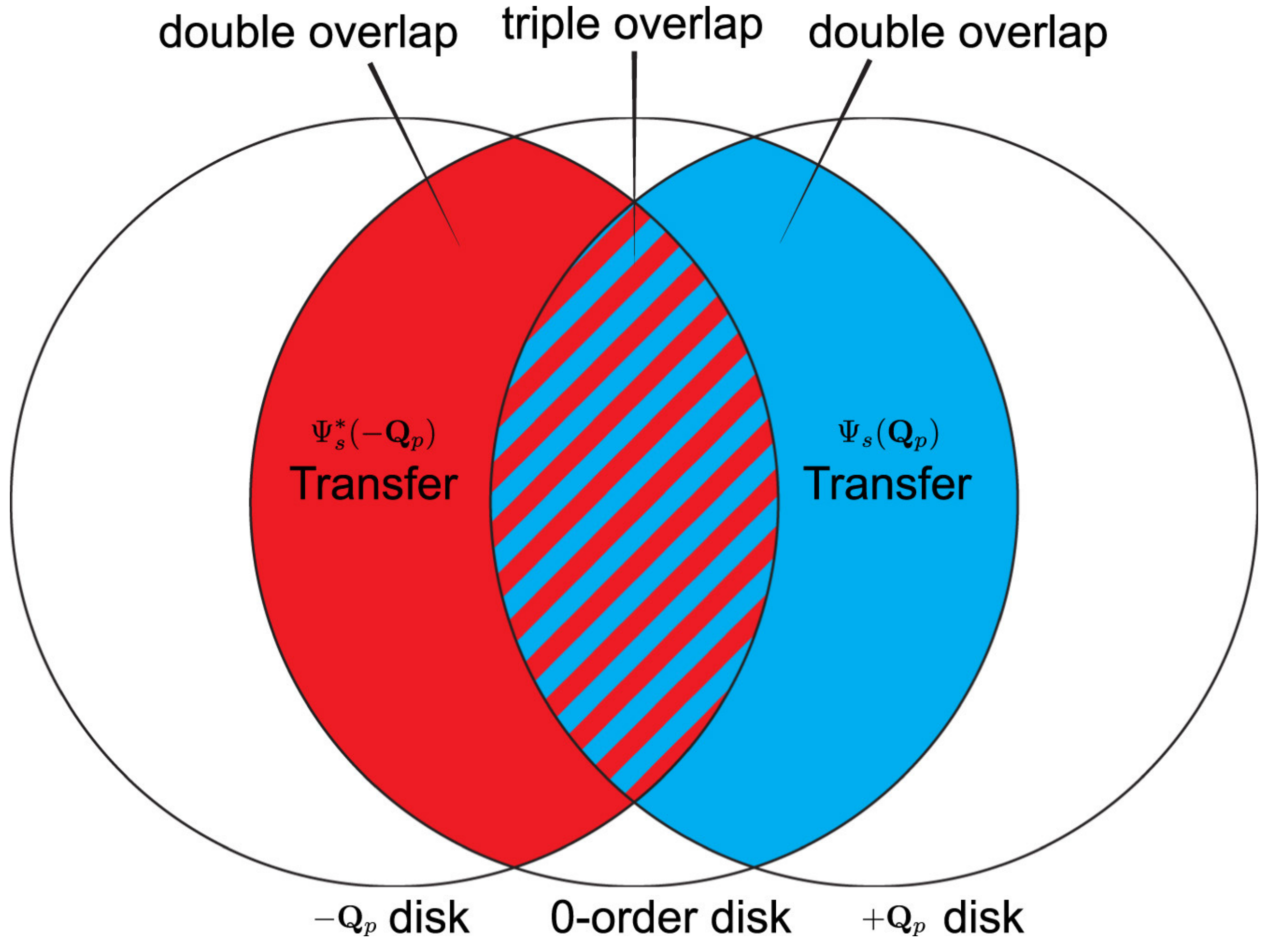
σ^{sim} and σ^{exp} = standard deviations

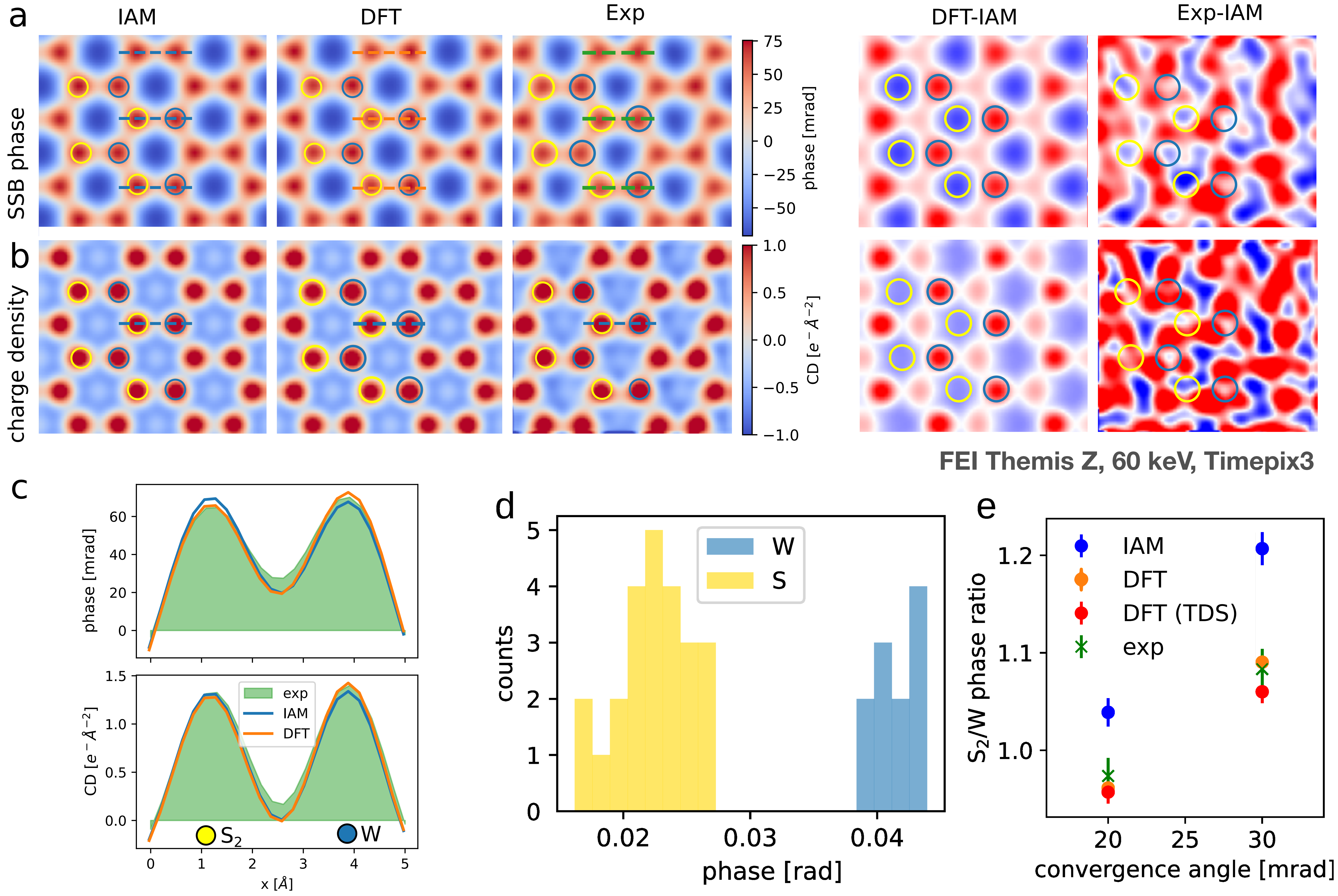
Phase analysis

STEM_Optimization*
(commit 6c549ba9)

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

Detecting charge transfer in defective WS_2



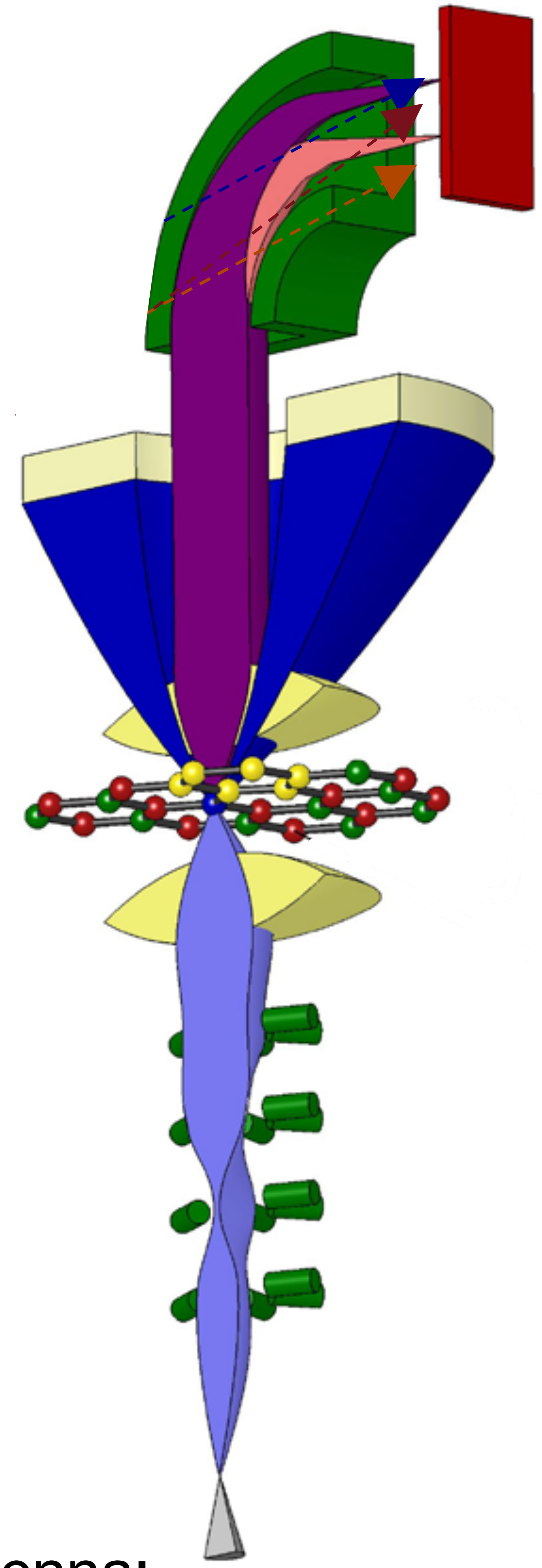


FEI Themis Z, 60 keV, Timepix3

Timepix3 limited to ~2 pA...

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

Ptycho powered by (Python) open source



Vienna:
Nion UltraSTEM100

Data acquisition	Language	Phase reconstructions	
Nion Swift (0.16.10)	Python (3.11.9)	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**)

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

Direct-electron detector for 4D-STEM

nion

DECTRIS
detecting the future

Number of pixels (*Q* px)	**192 × 192**
Active area [mm²]	20 × 20
Pixel size [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

39 GB for
512×512 *R* px

1106 MB/s!

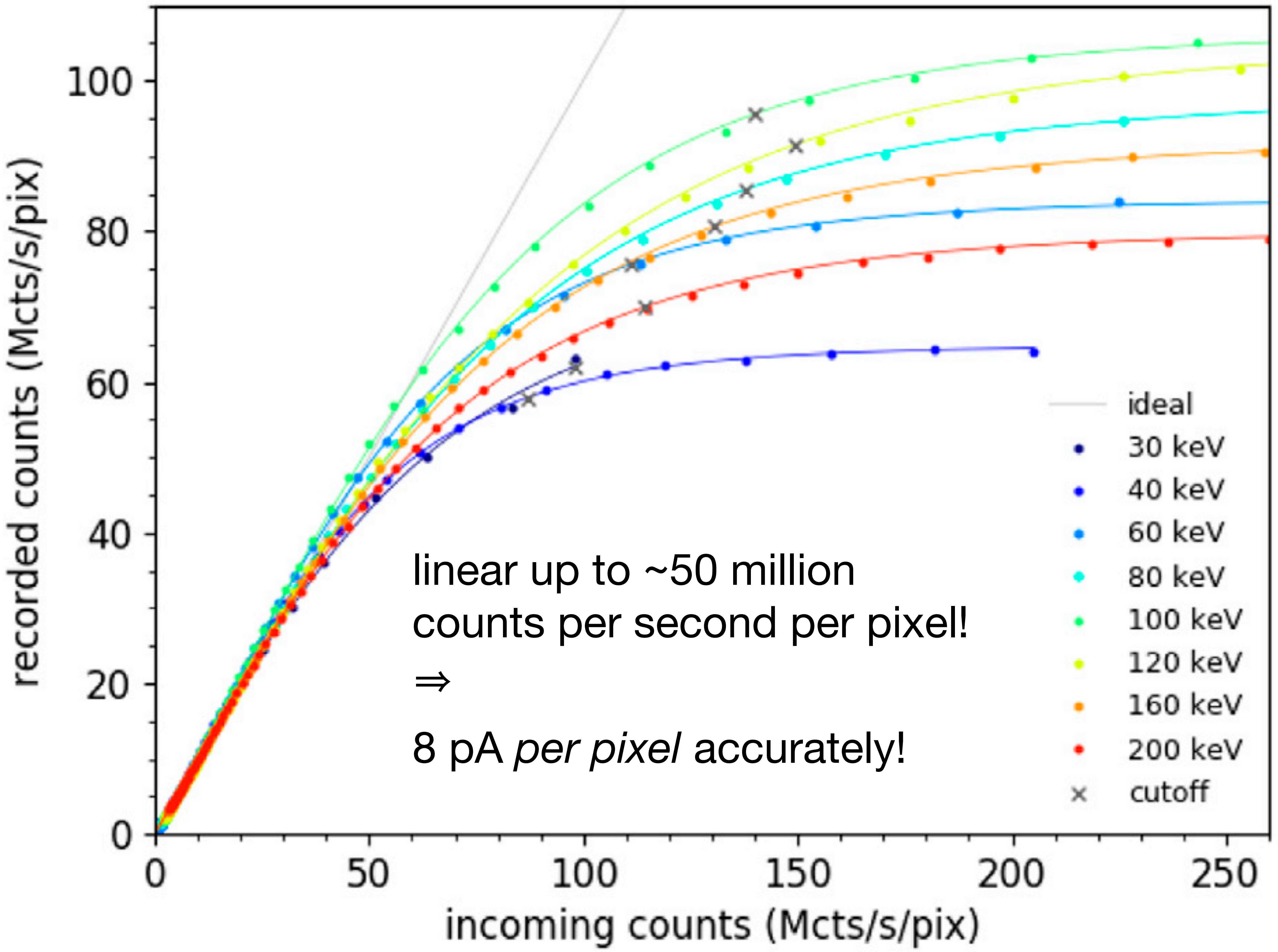
On-axis (btw.
HAADF & MAADF)

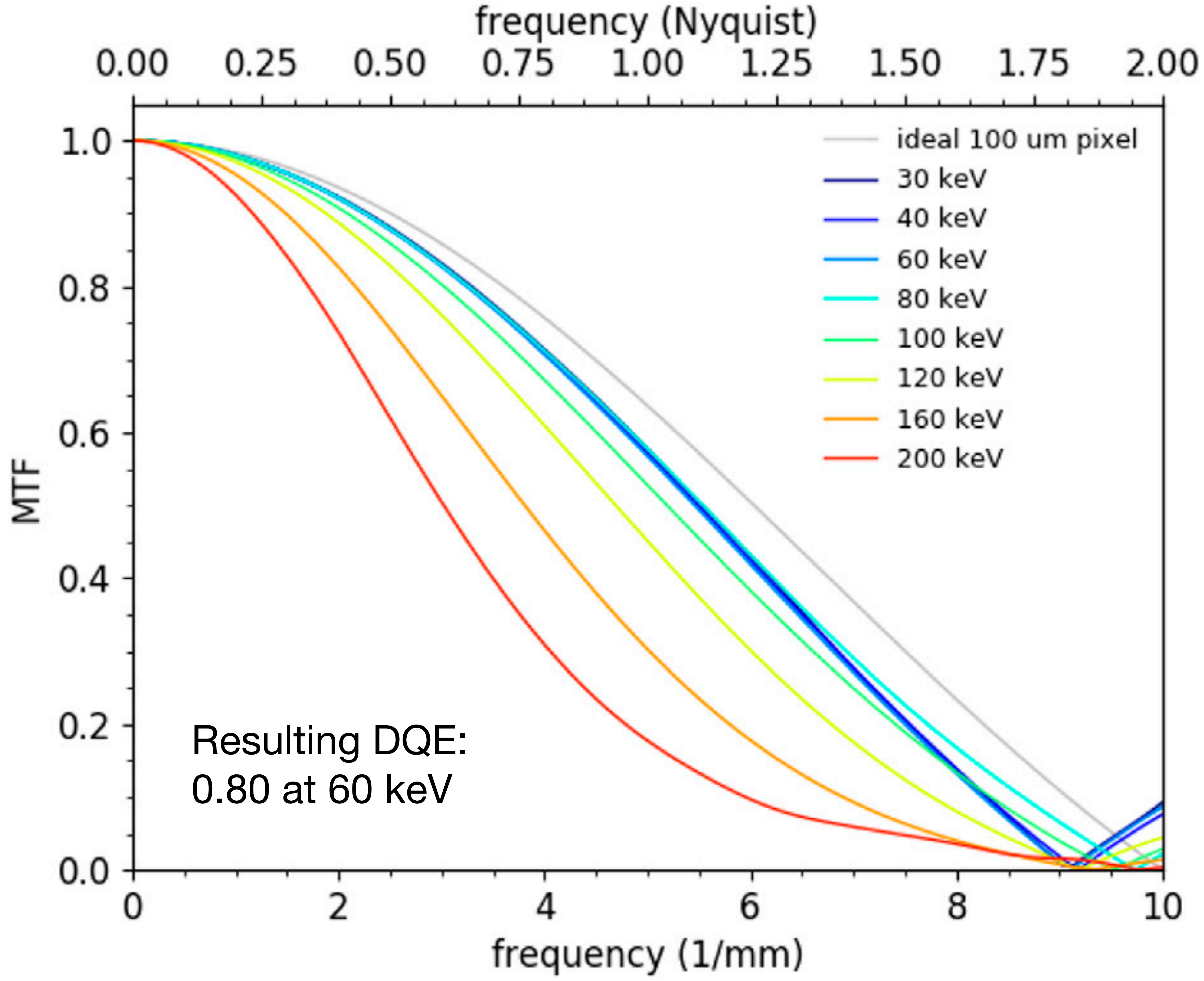
Near-ideal 4D-STEM performance at 60 keV

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

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

Near-ideal electron counting

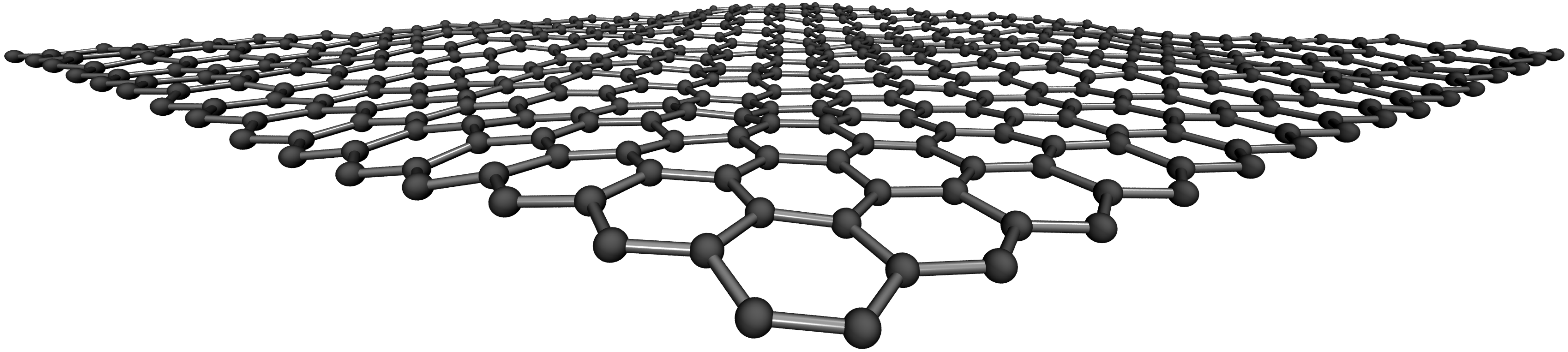




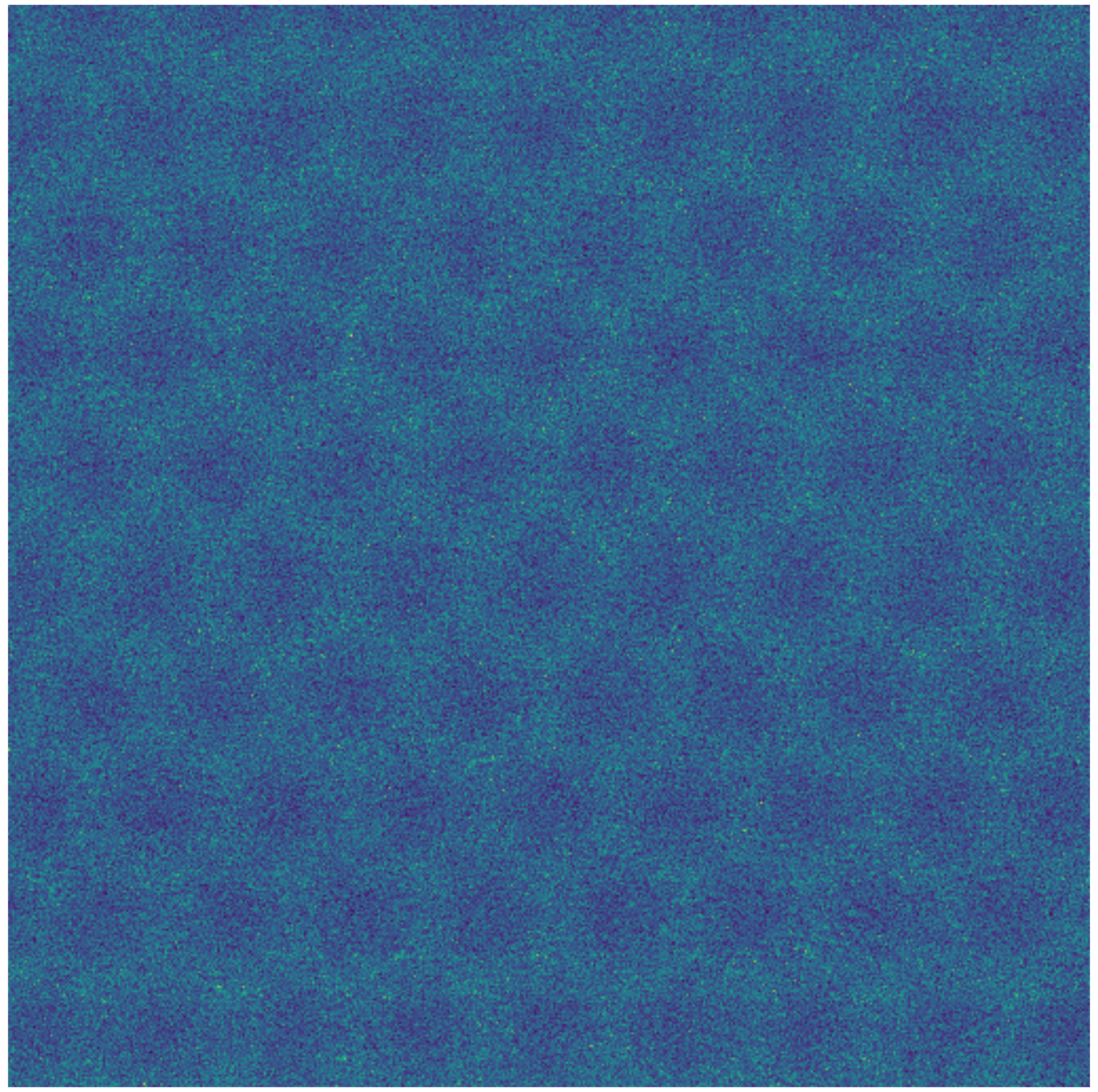
P. Zambon *et al.*, Front. Phys. 11:1308321 (2023)

Graphene as 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



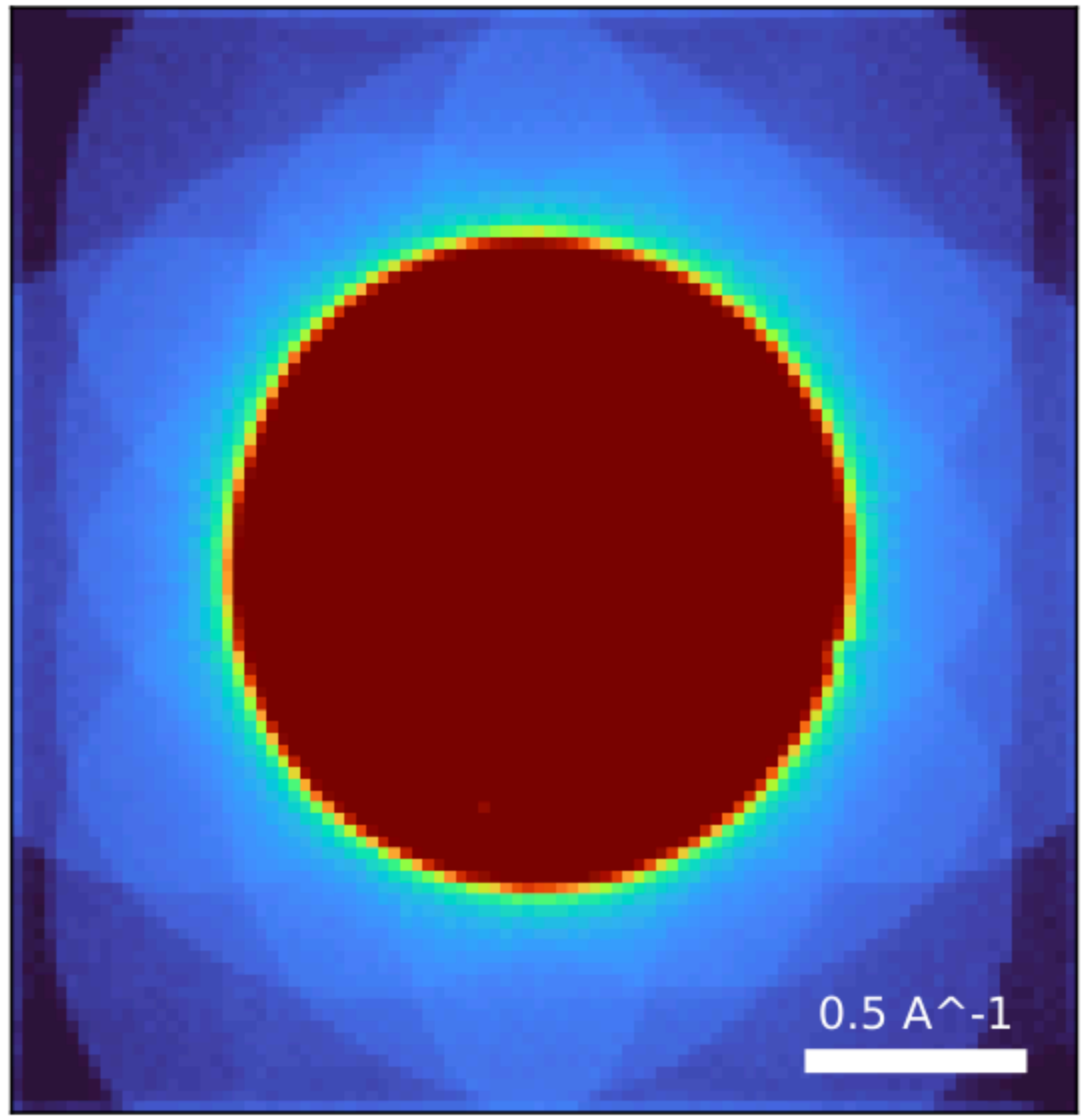
Nion + ARINA 4D-STEM data collection

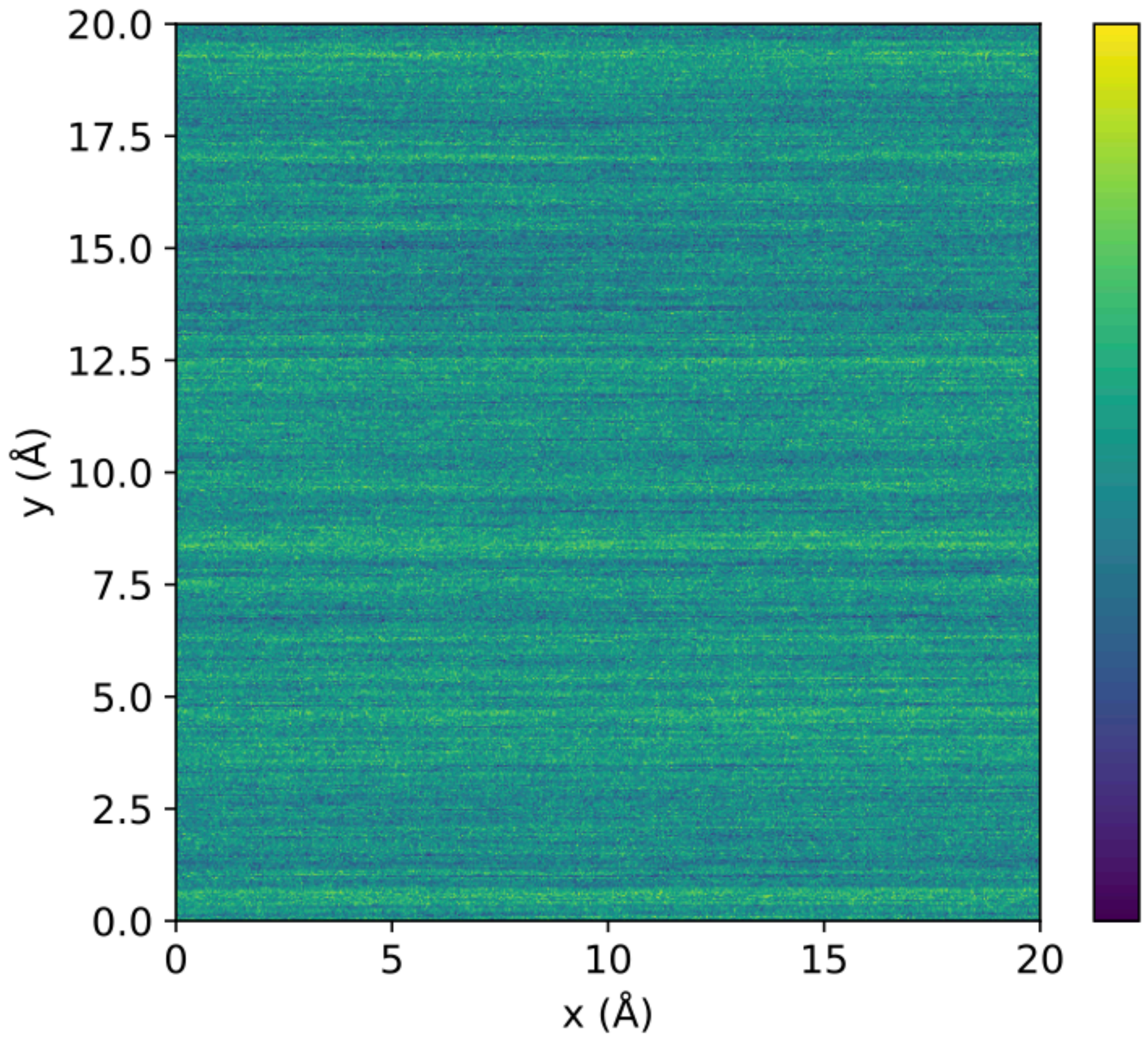


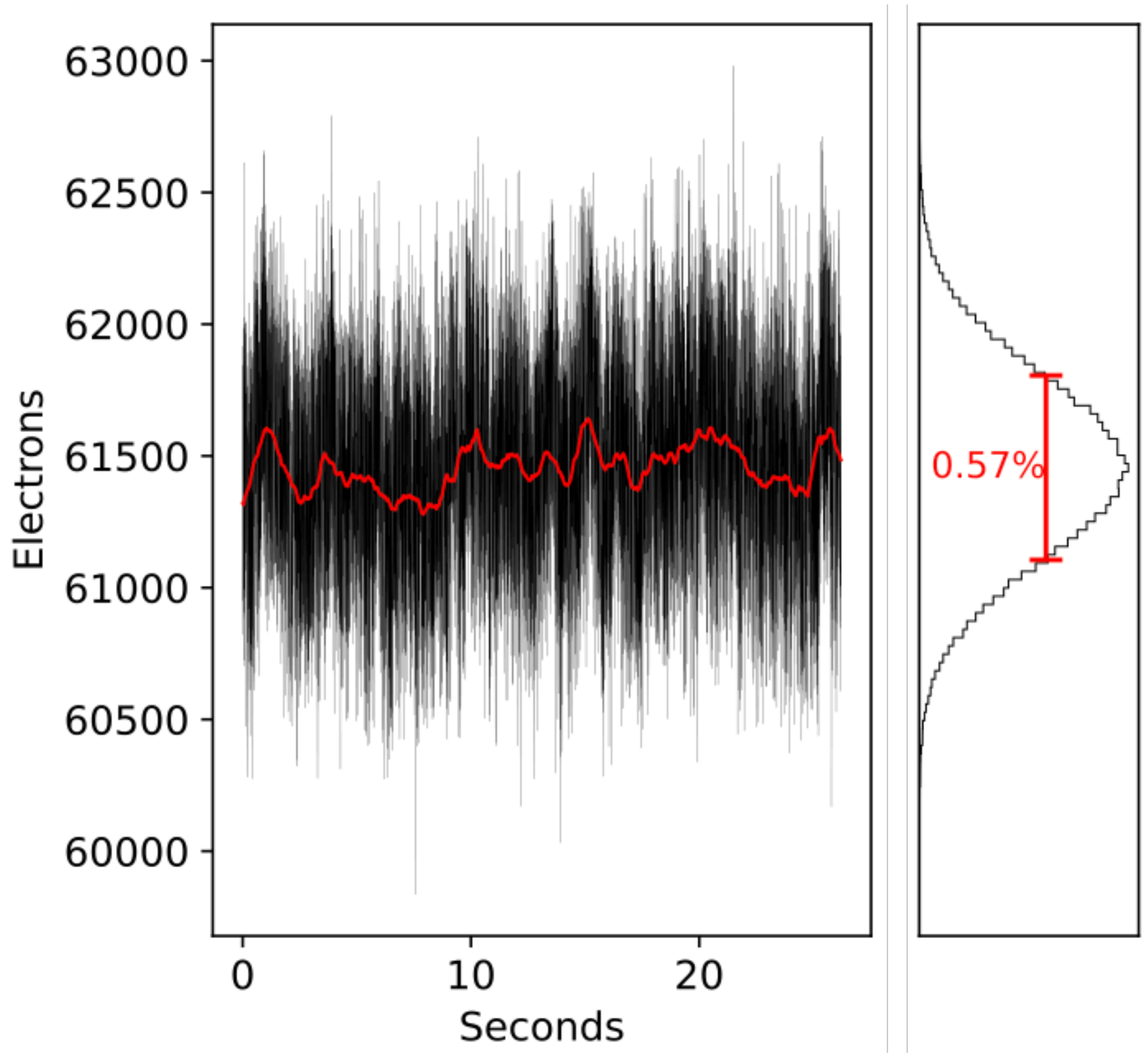
HAADF (80–300 mrad)

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)

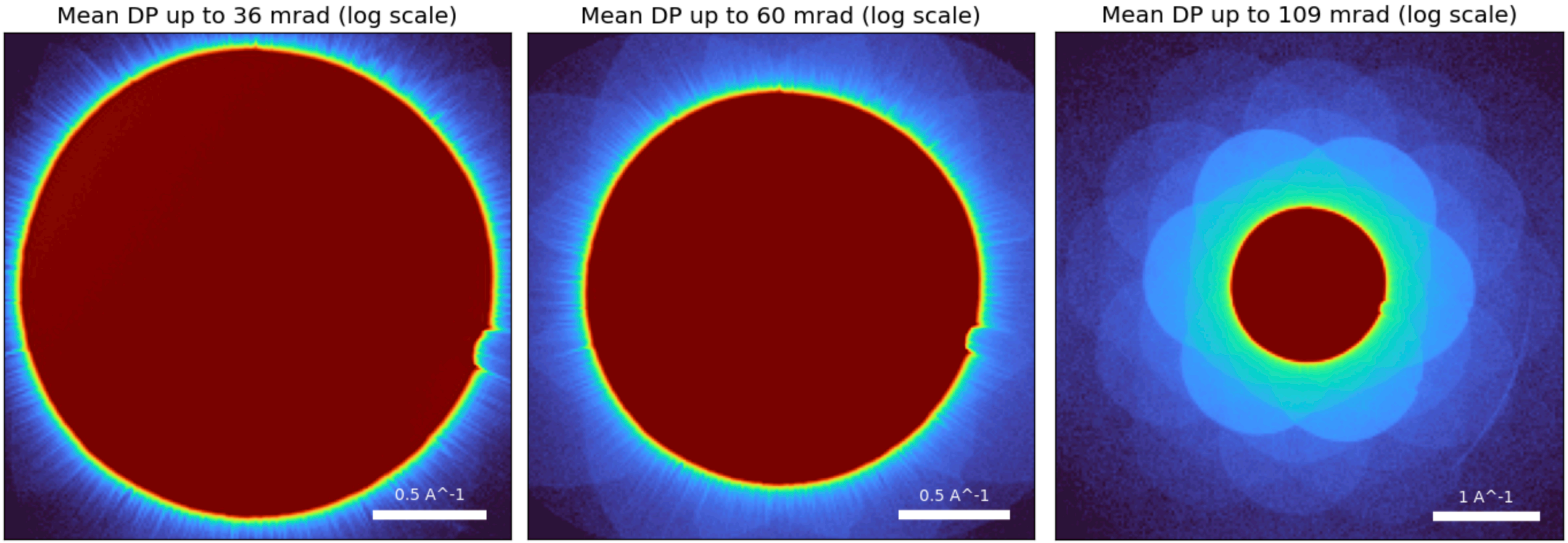






- 34 mrad semi-convergence angle, variable camera length (max collection angle)
- counting electrons: accurate dose (& dose per pixel can be normalized)

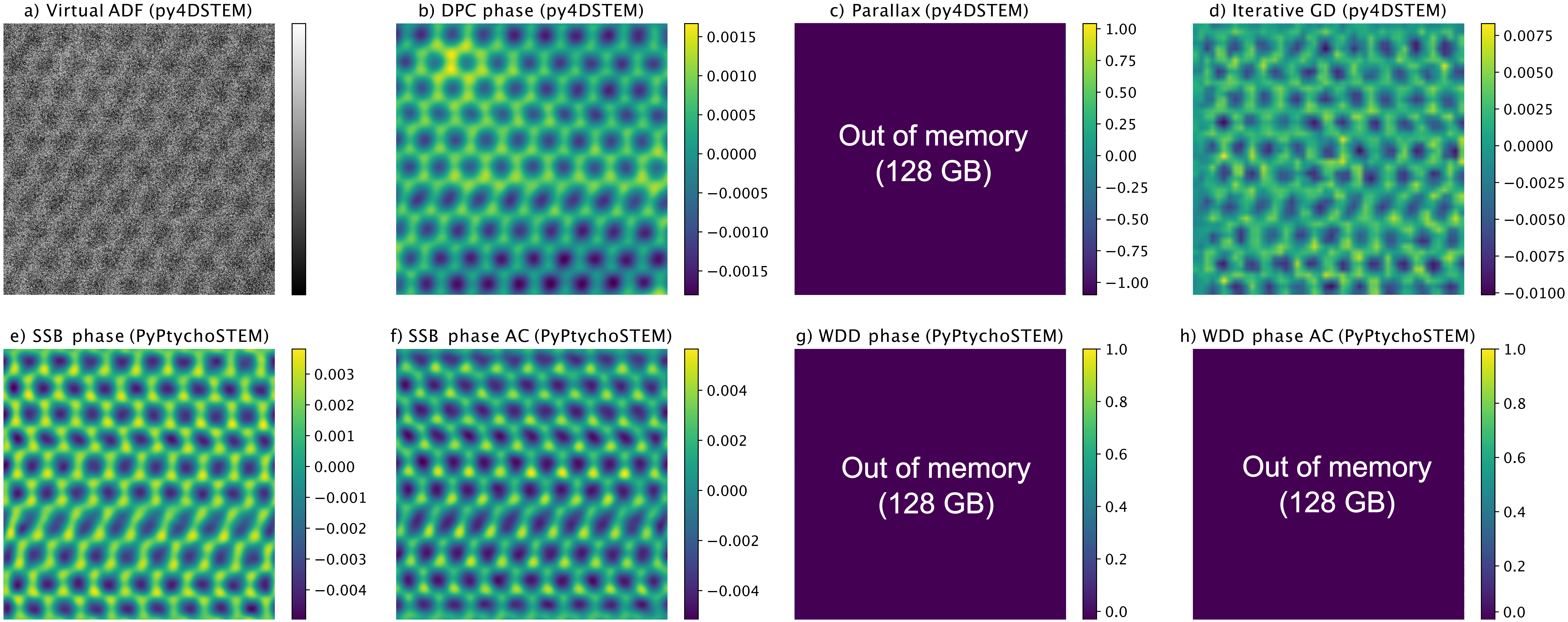
Nion + ARINA variable camera lengths



~200 pA current!

T. Susi, unpublished

Comparing phase reconstruction algorithms (***Q*** *binning*)

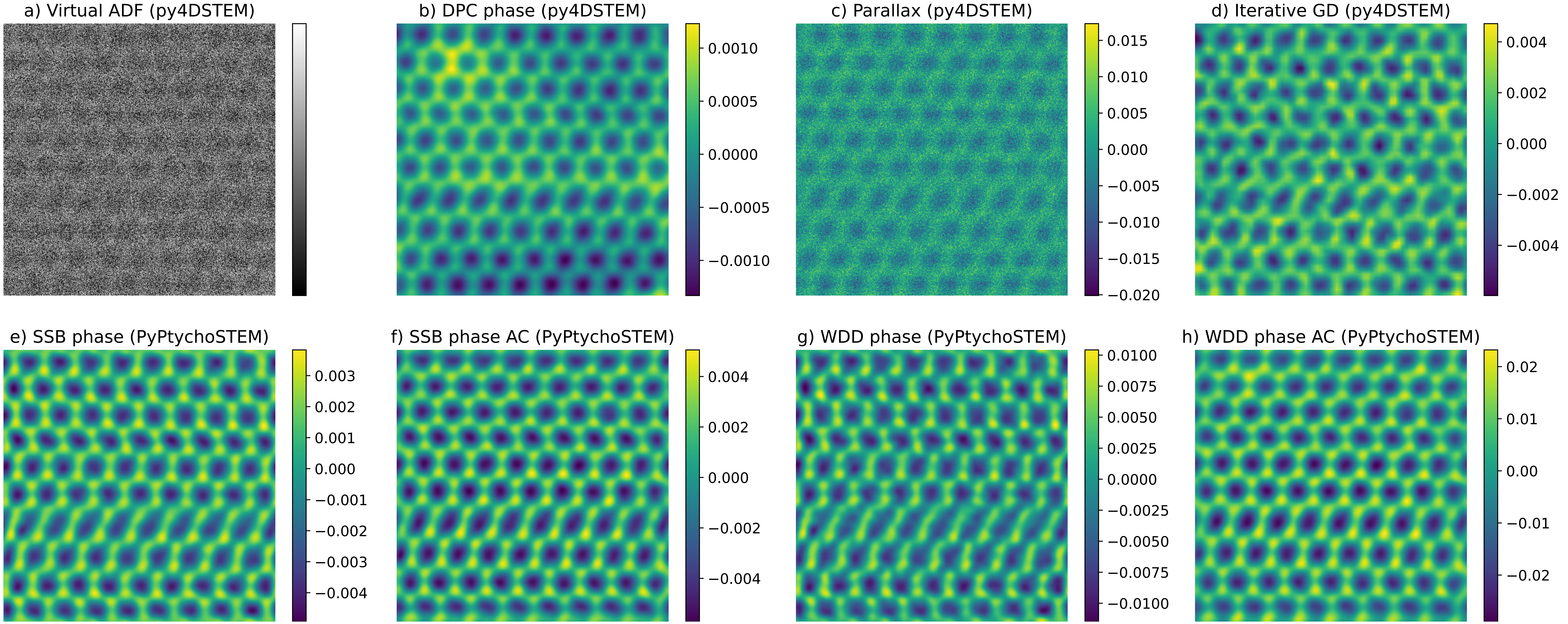


no binning in ***Q***

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

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*)



2× binning in ***Q***

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

SSB & WDD: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167
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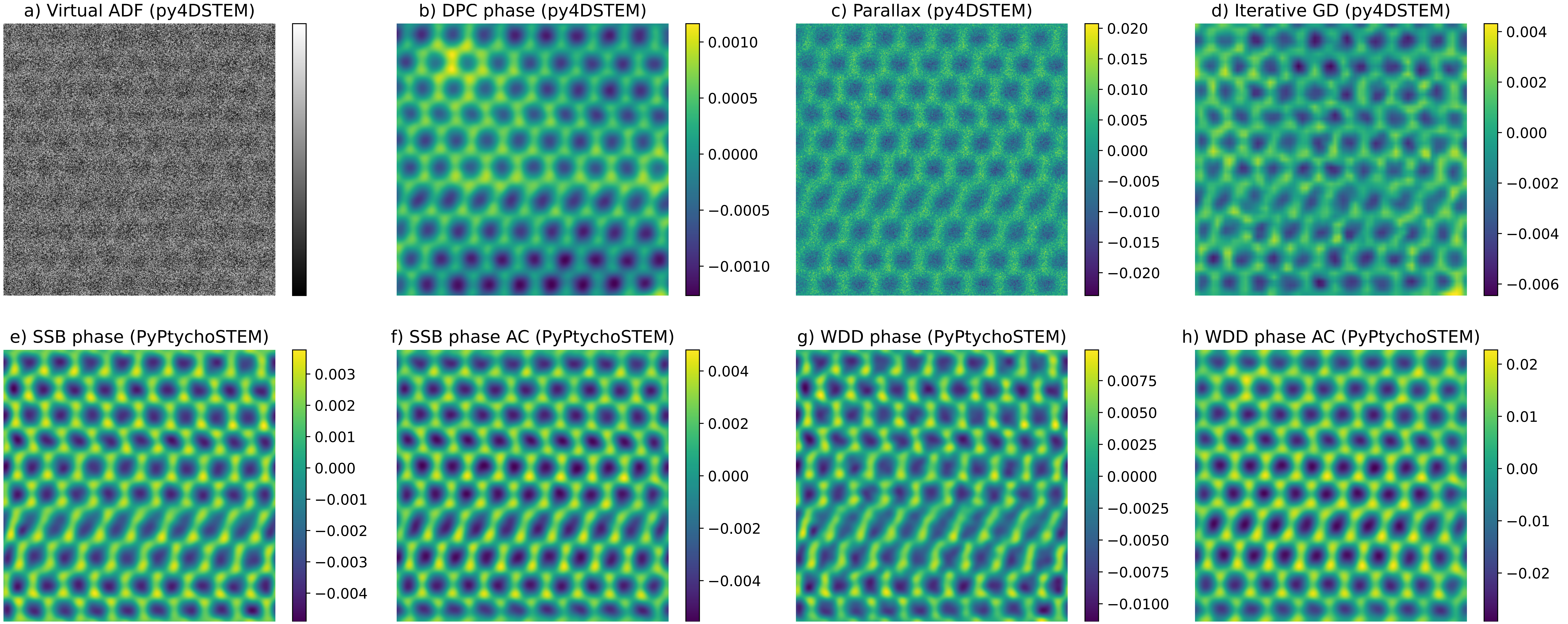
Comparing phase reconstruction algorithms (*Q binning*)

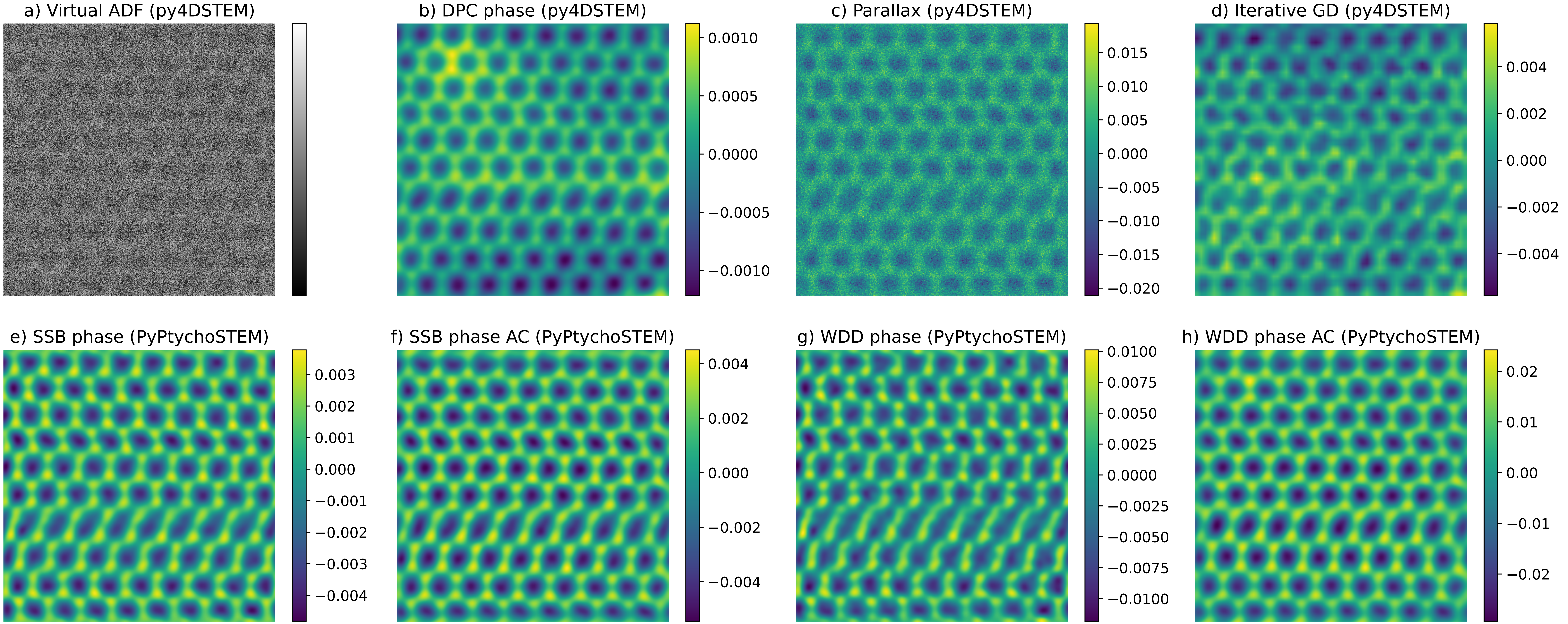


4× binning in ***Q***

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

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*)

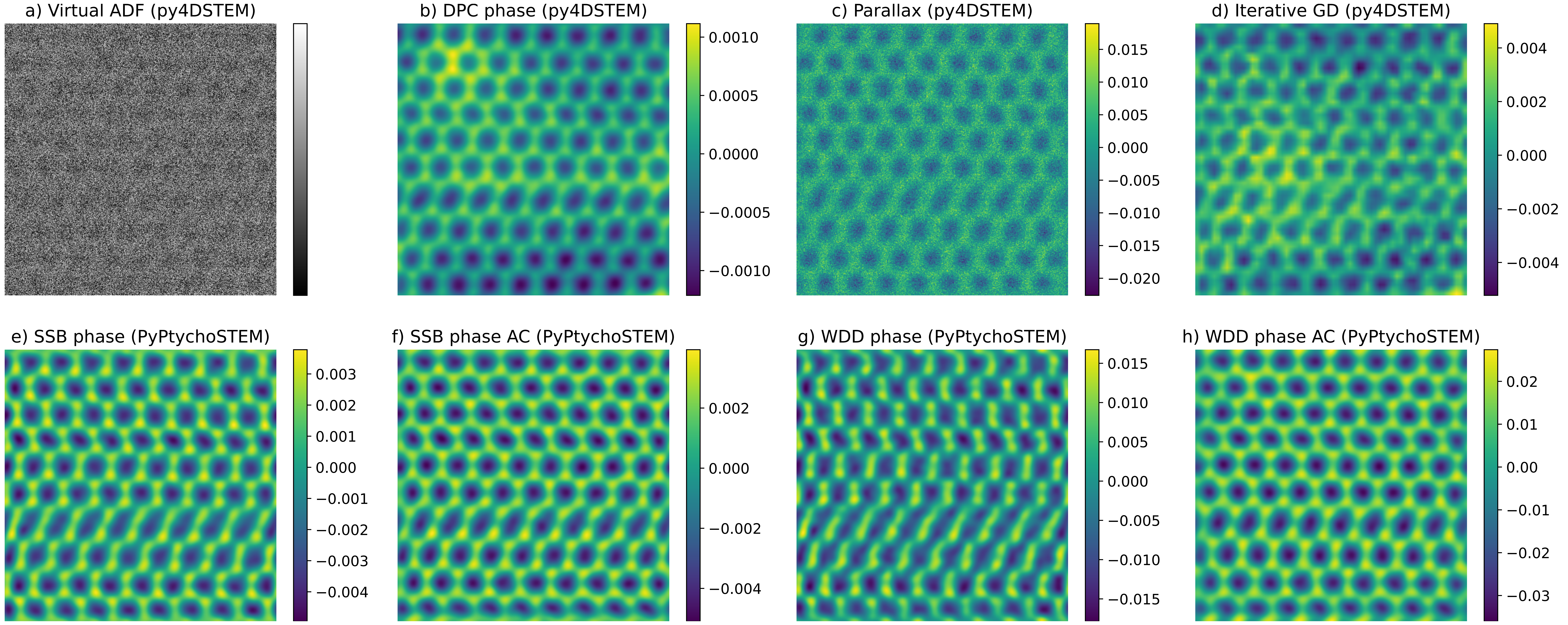


8× binning in ***Q***

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

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*)

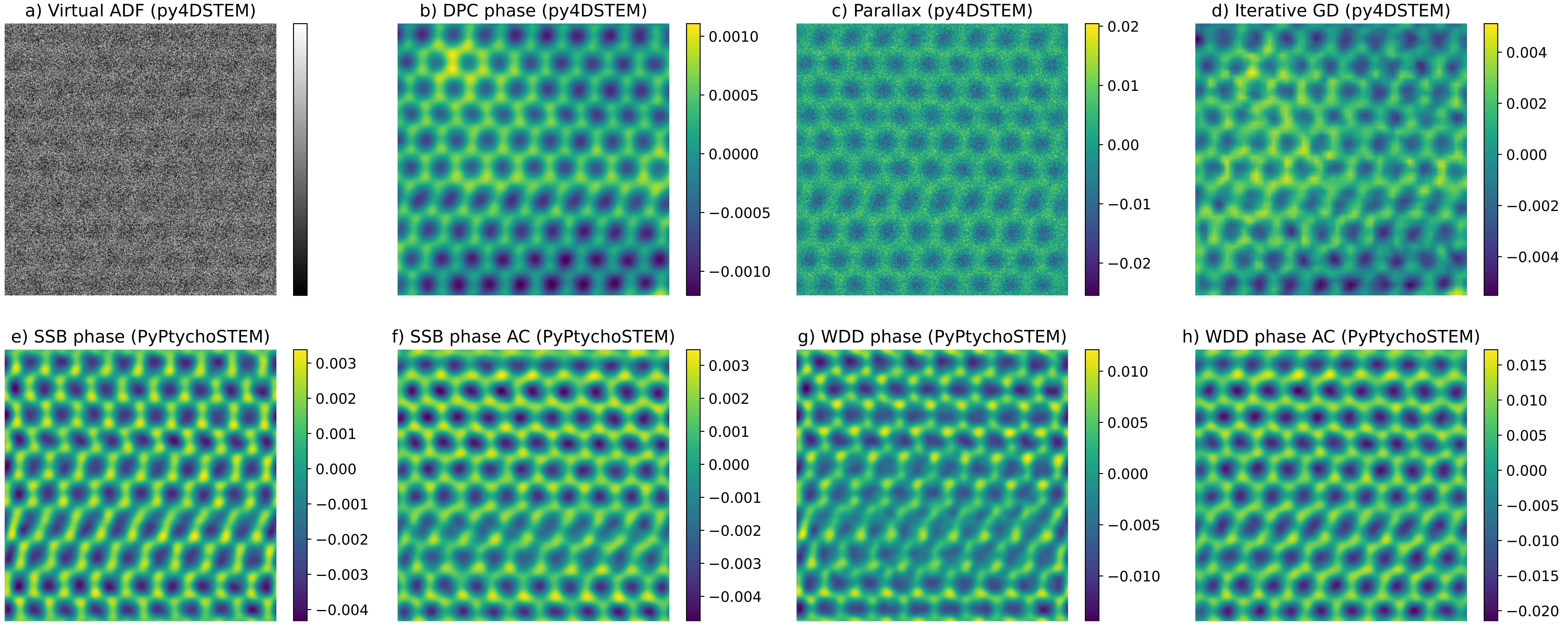


16× binning in ***Q***

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

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*)



32× binning in ***Q***

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

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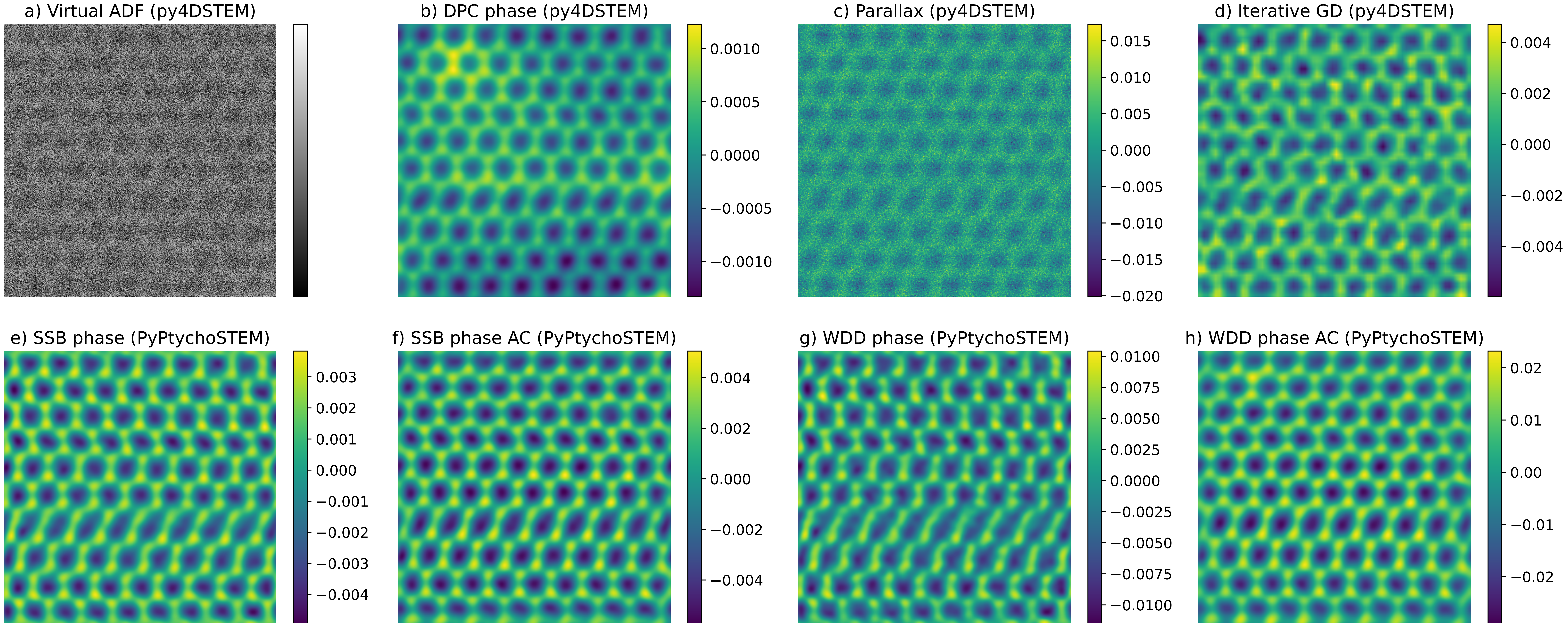
SSB & WDD: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167
DPC, Parallax, Iterative GD: G. Varnavides *et al.*, arXiv:2309.05250v2 (2024)

Comparing phase reconstruction algorithms (*R thinning*)



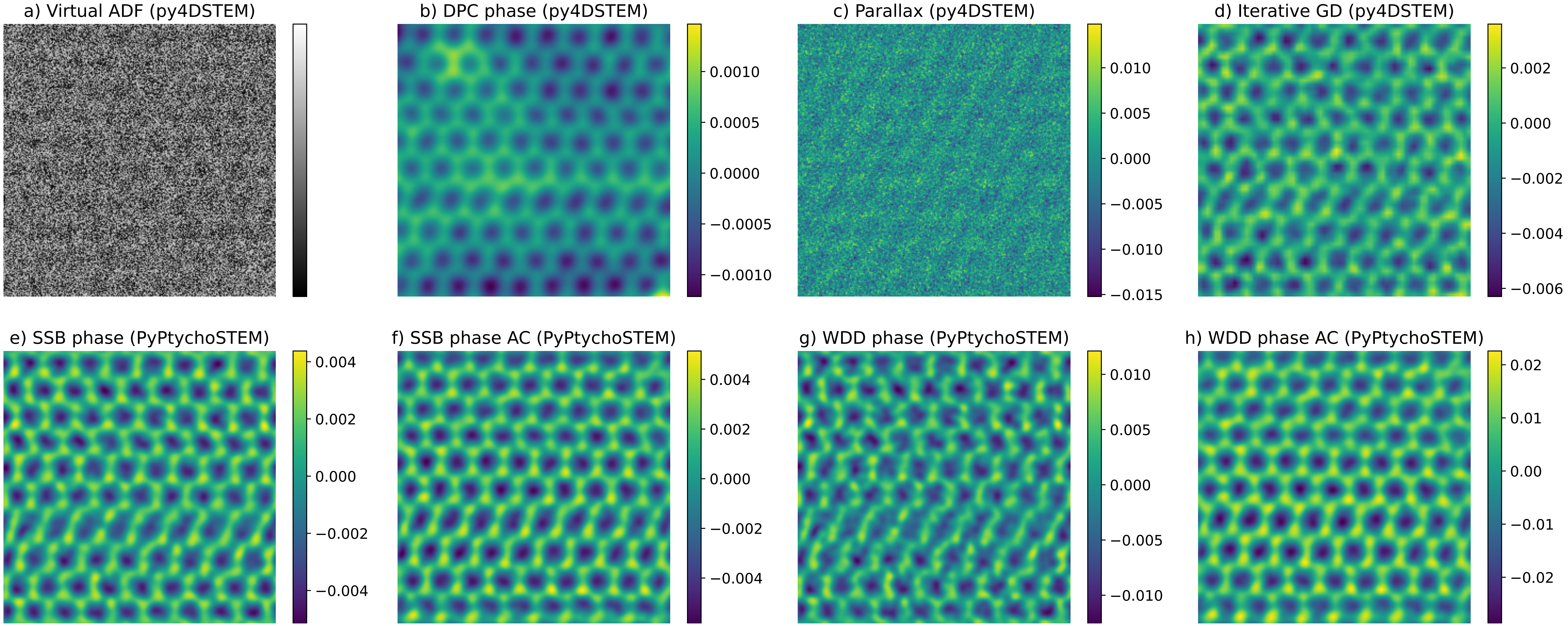
no thinning in ***R***

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

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 thinning*)

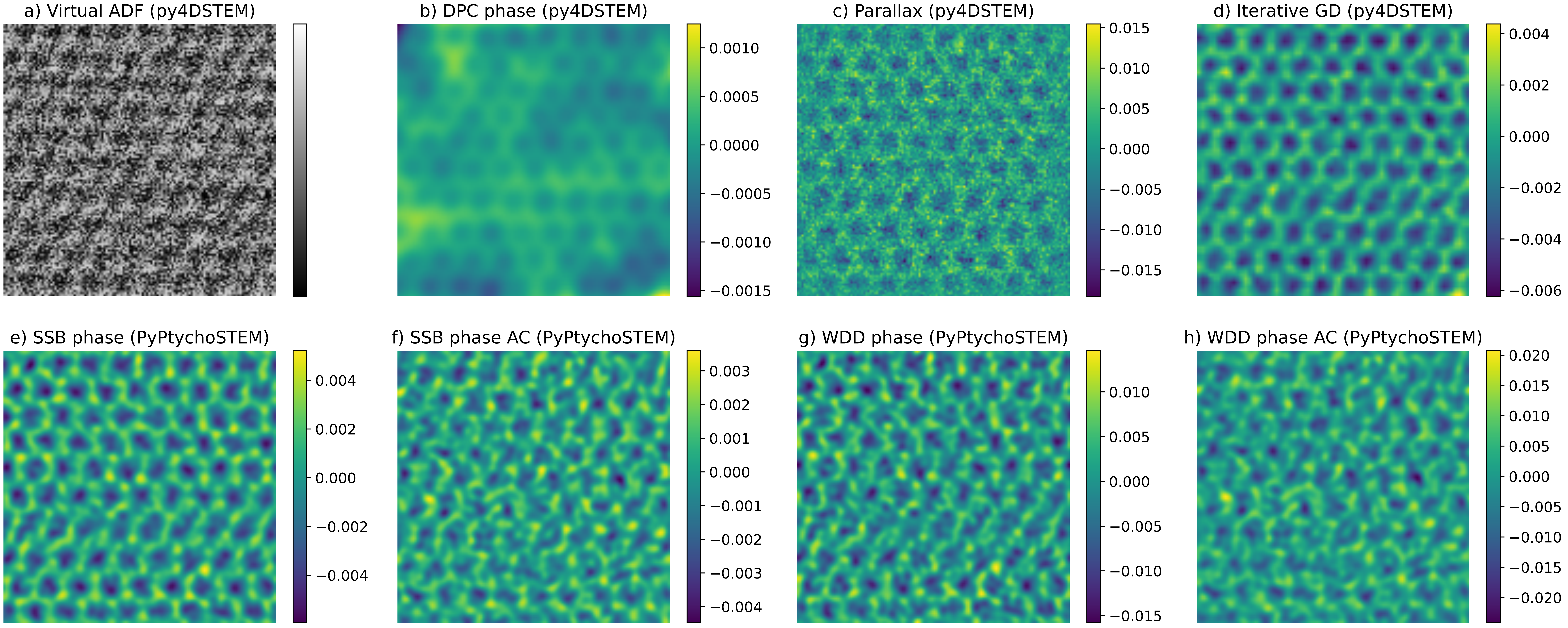


2× thinning in ***R***

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

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 thinning*)



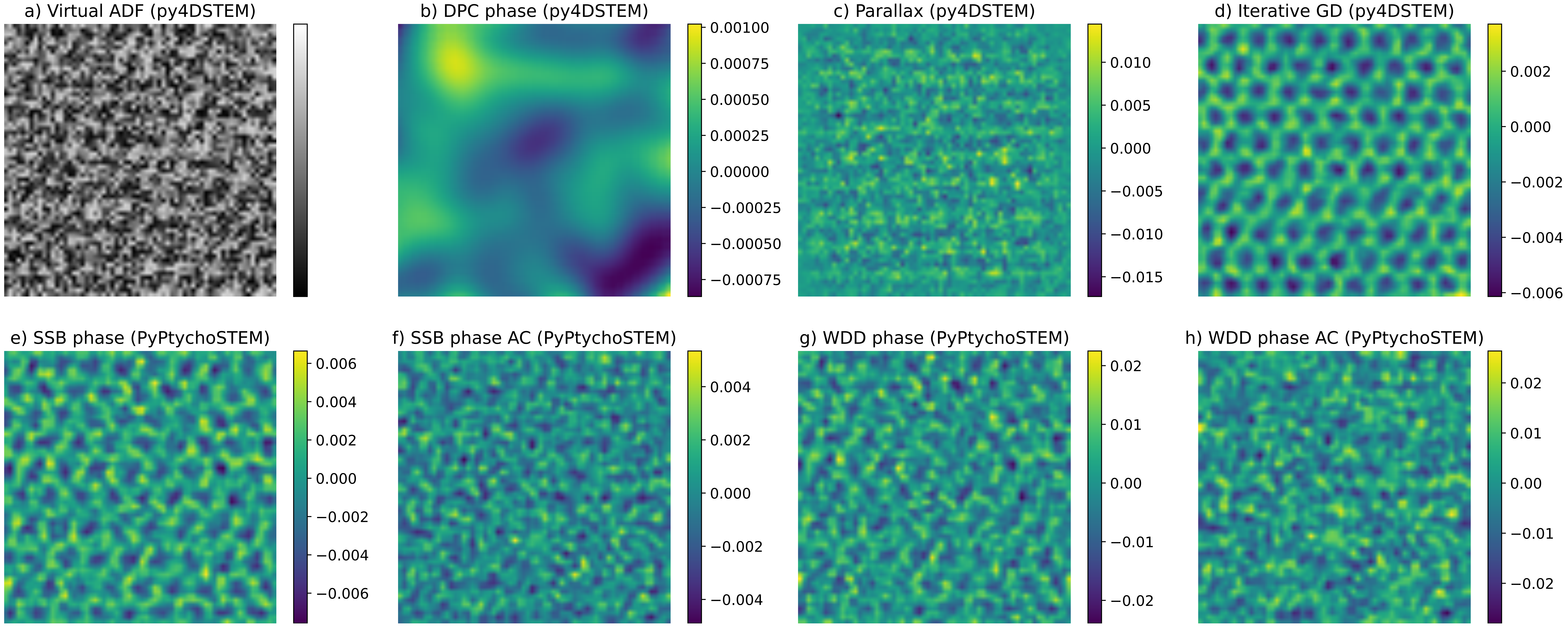
4× thinning in ***R***

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

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 thinning*)

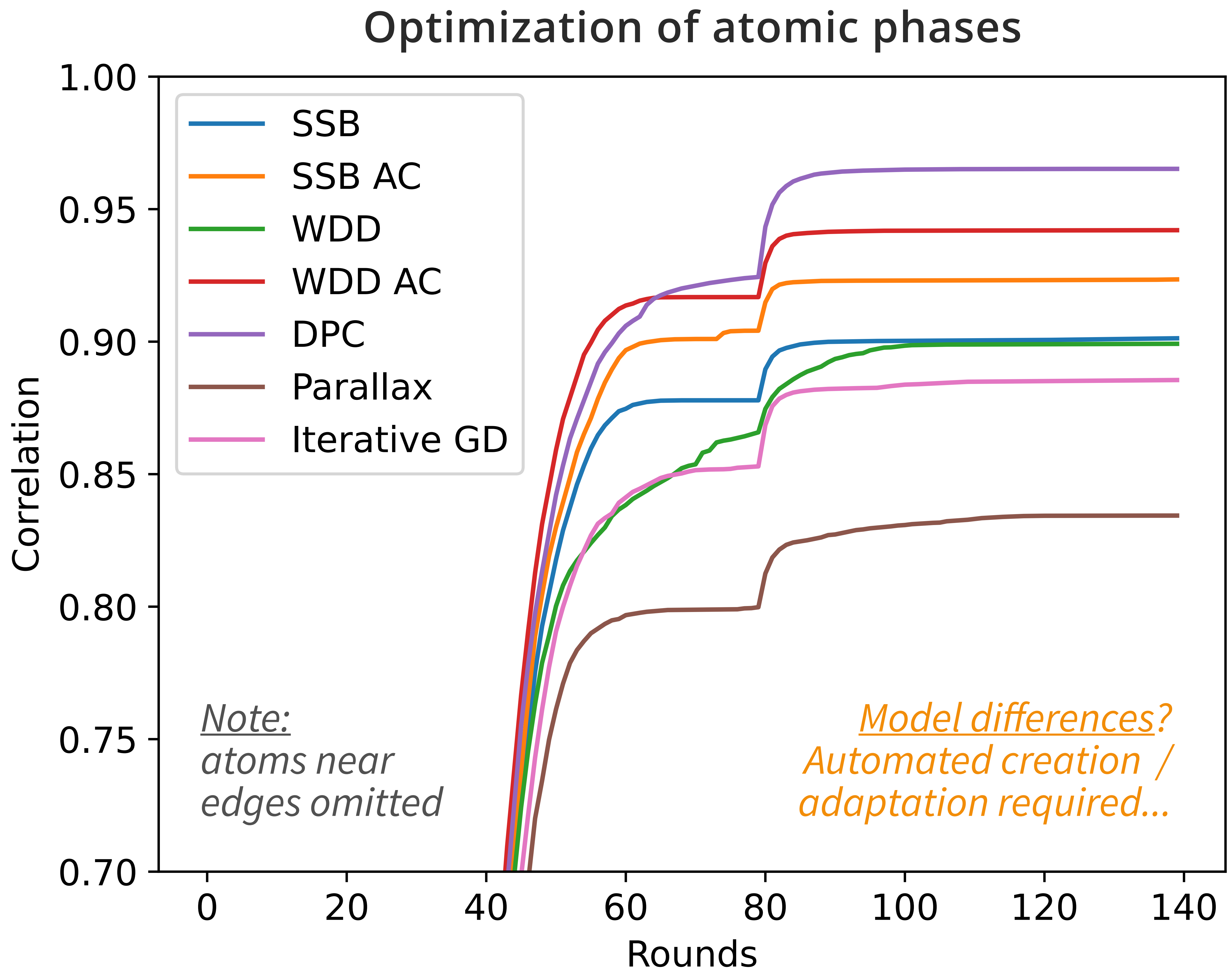


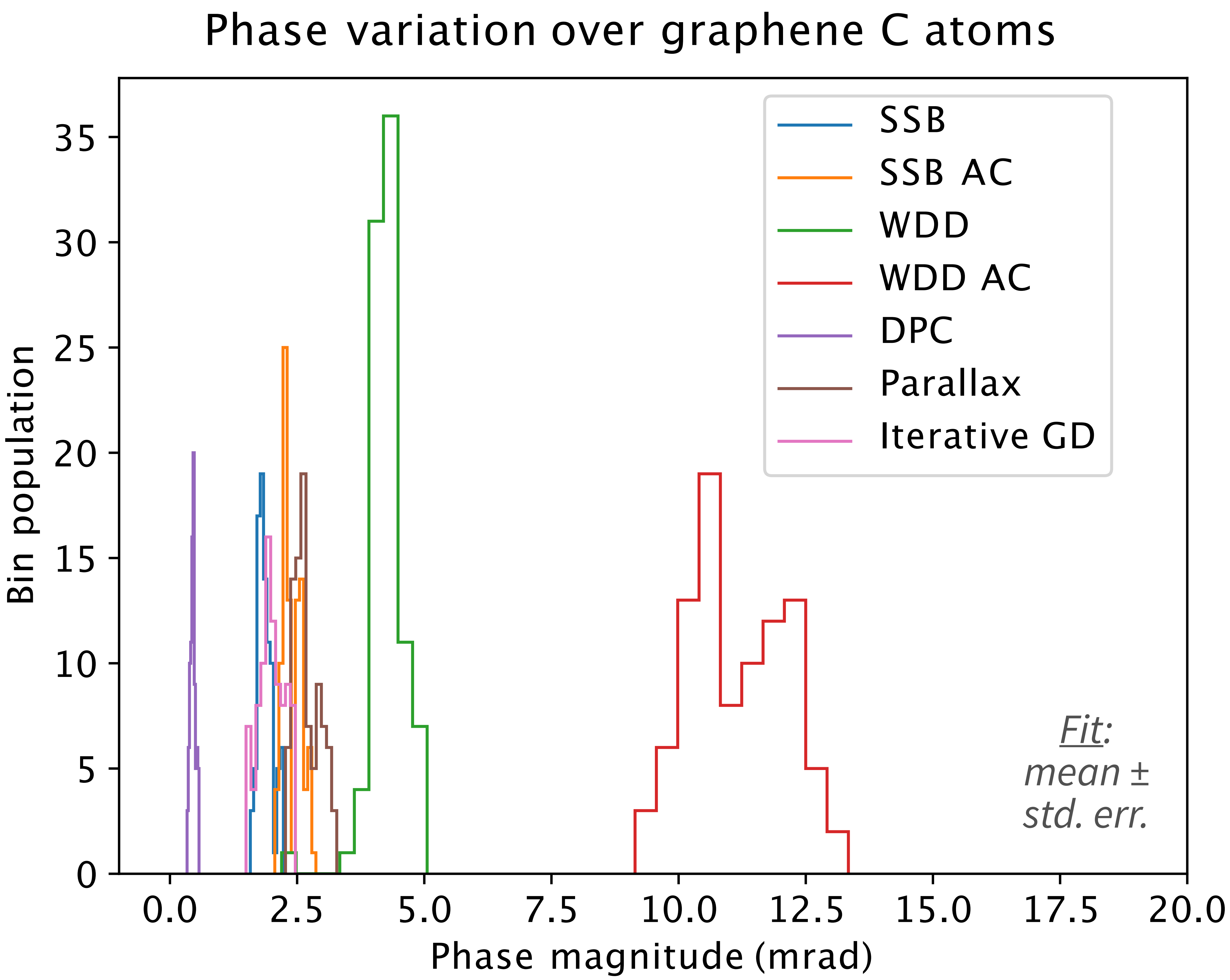
8× thinning in ***R***

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

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

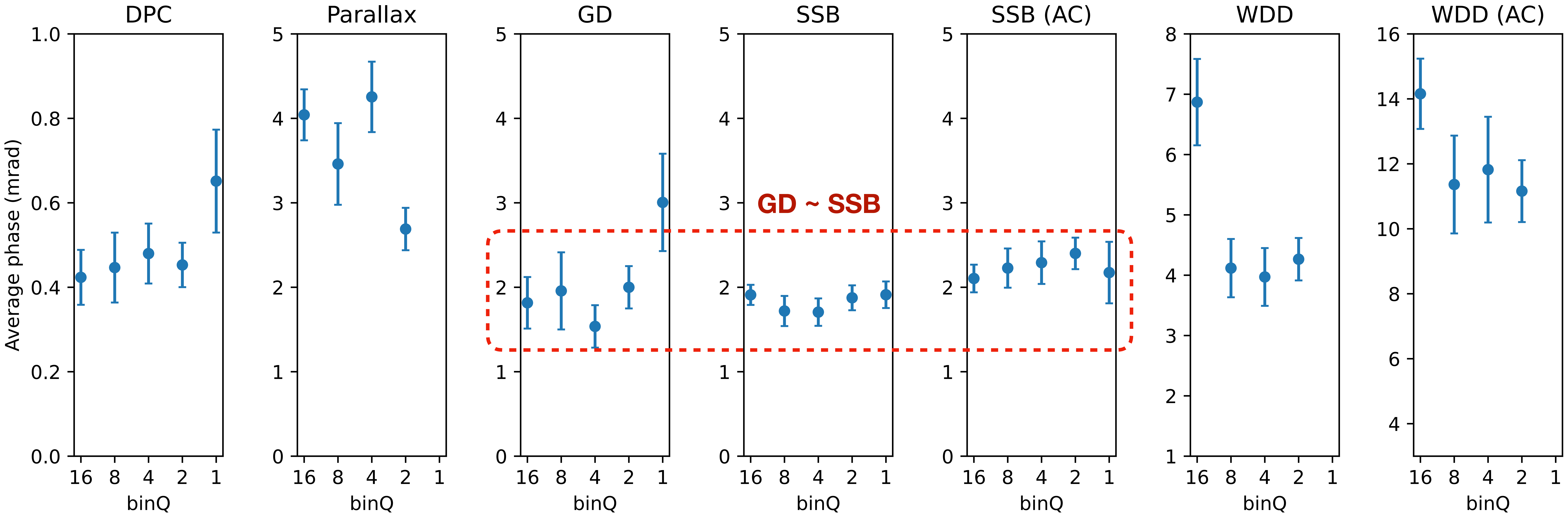




T. Susi, unpublished

Quantification of phase variation

Note: different *y*-scales!



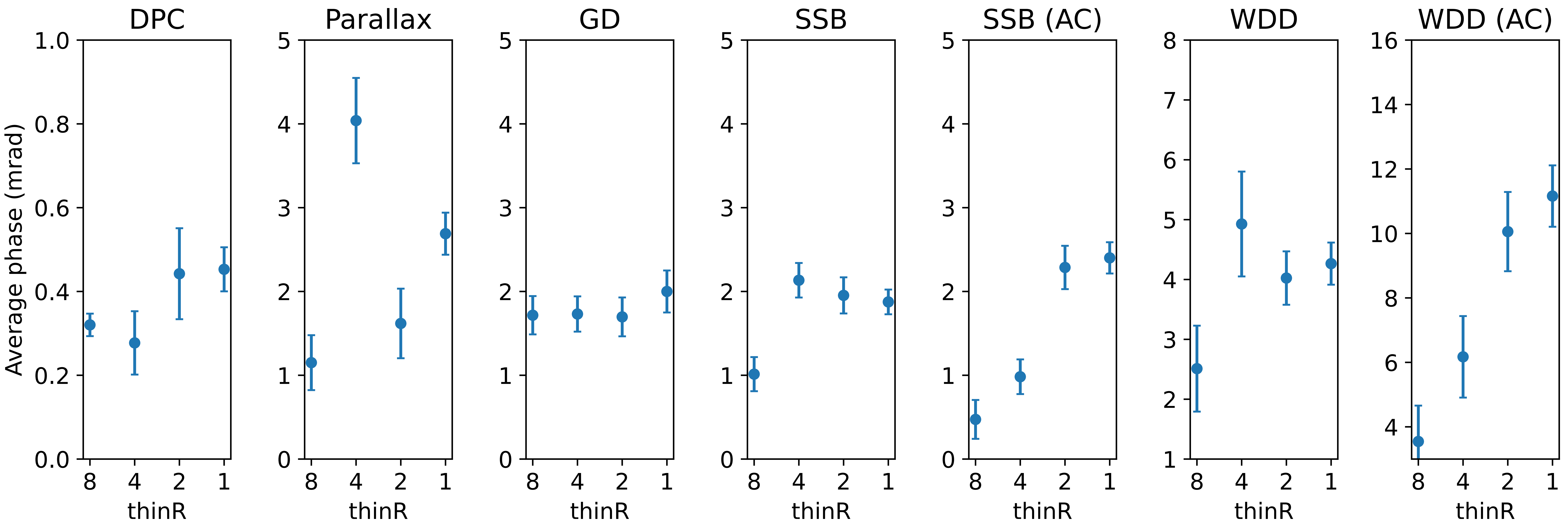
Binning in ***Q***: **36 mrad** max. scattering angle, 33.3 us dwell time, 512×512 ***R*** px, 192×192 ***Q*** pix

Plot: mean ± std. err.

T. Susi, unpublished

Quantification of phase variation

Note: different *y*-scales!

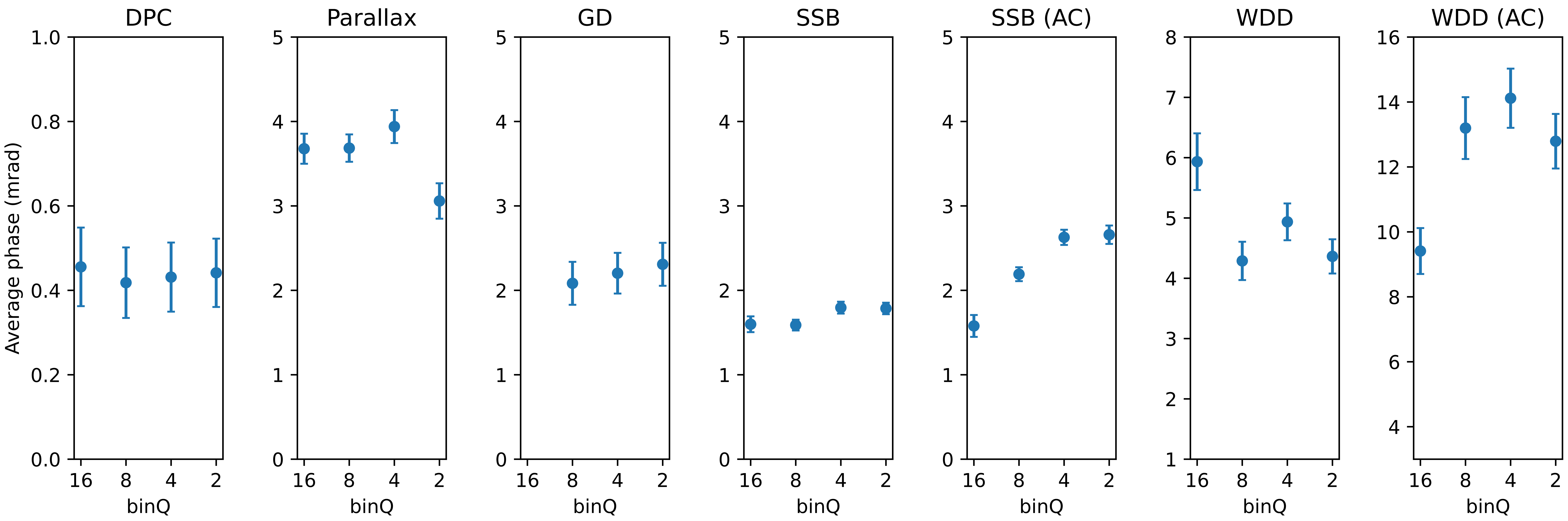


Thinning in ***R***: **36 mrad** max. scattering angle, 33.3 us dwell time, 512×512 ***R*** px, 96×96 ***Q*** pix

Plot: mean ± std. err.

Quantification of phase variation

Note: different *y*-scales!

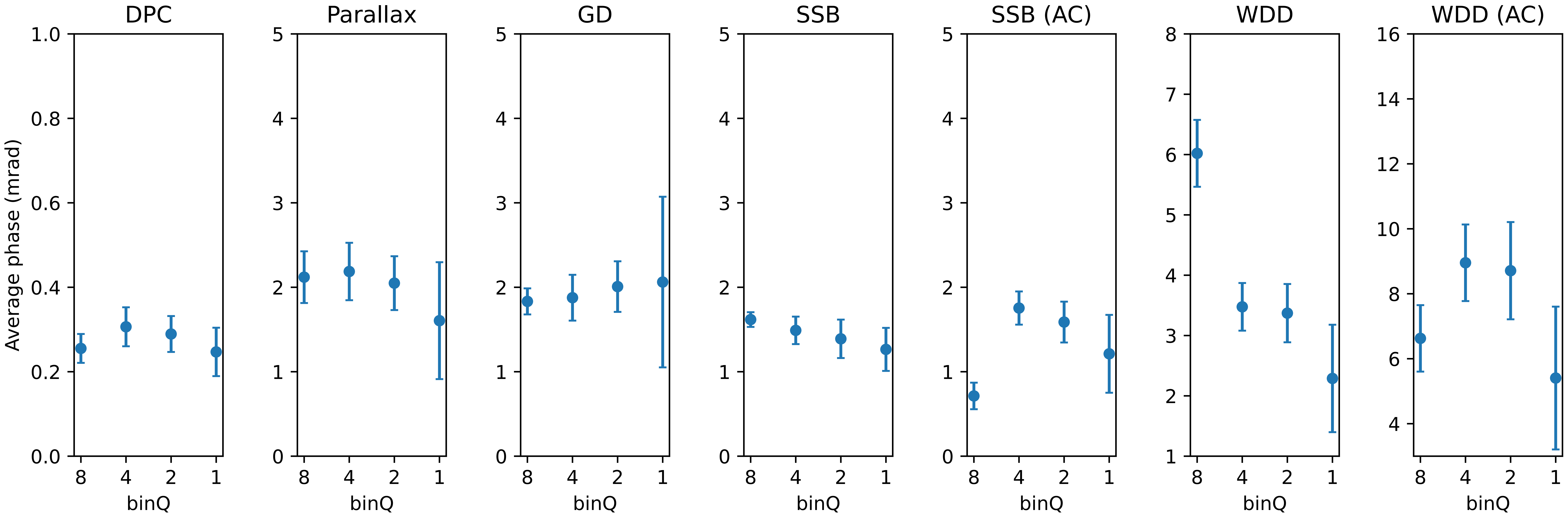


Plot: mean ± std. err.

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!



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

Plot: mean ± std. err.

T. Susi, unpublished

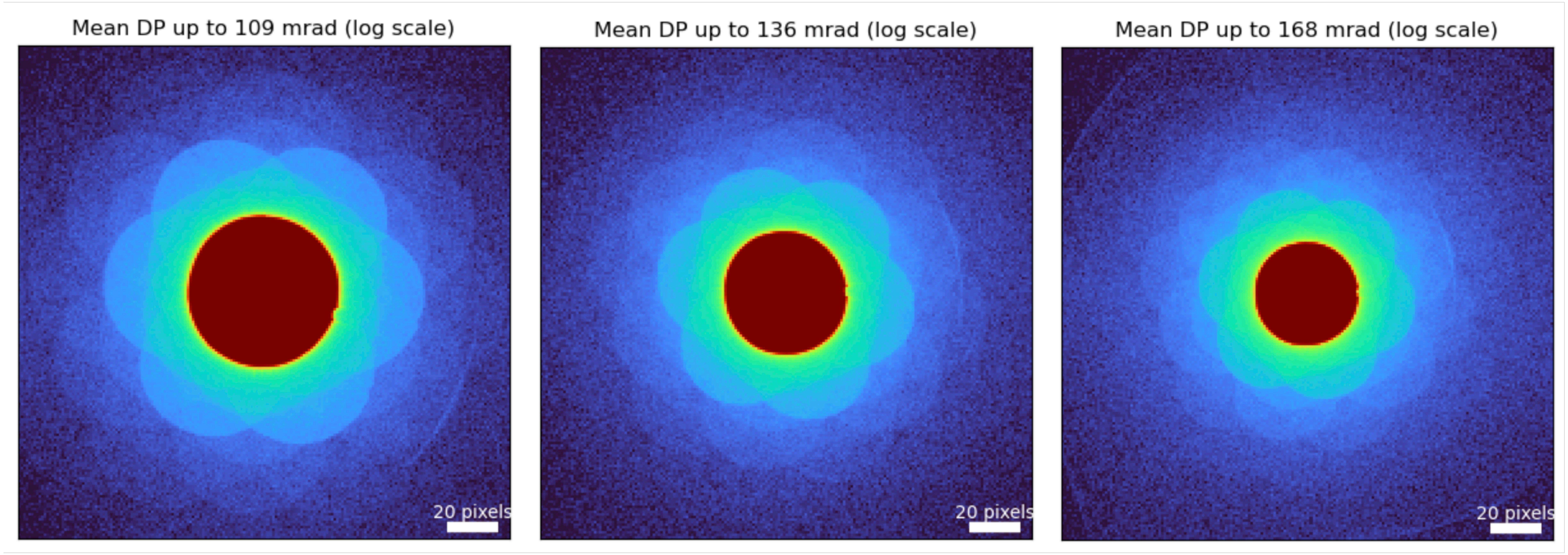
Computational cost (36 mrad datasets)

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

Hardware: 24-core Intel Xeon CPU + NVidia RTX4090 GPU

† Slightly different numbers of alignment iterations
‡ GPU-accelerated; varying Q-padding

Conclusions & future prospects



- Quantitative absolute phase values?
  - SSB and GD seem to ~agree (WDD?!)
- GD good even for focused (& fast on GPU)

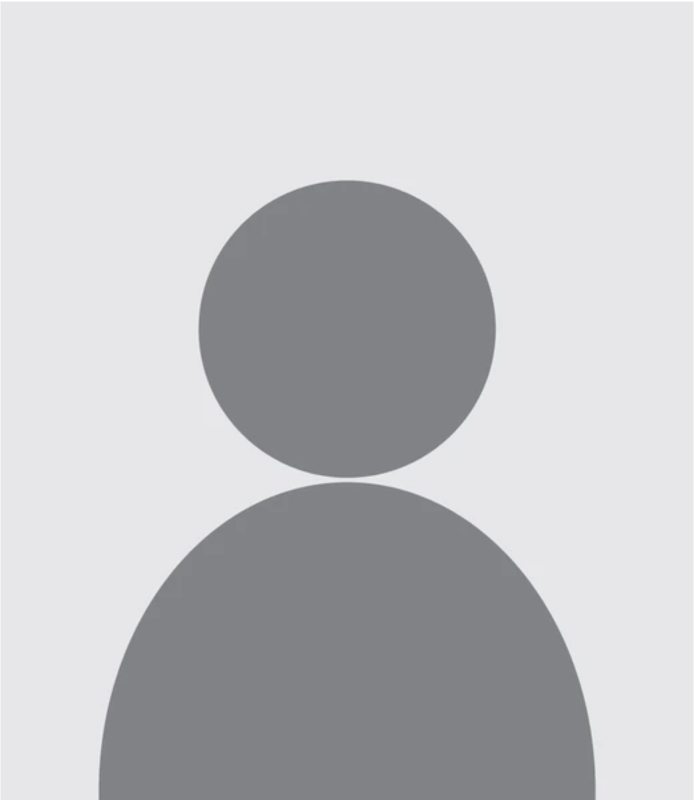
- Super-resolution iterative ptycho up to ~4 × alpha
- Live virtual detectors / (semi-)live SSB?
- Structure & charge transfer in various materials…

Upcoming Japan stay (Sep 2024 – Feb 2025)

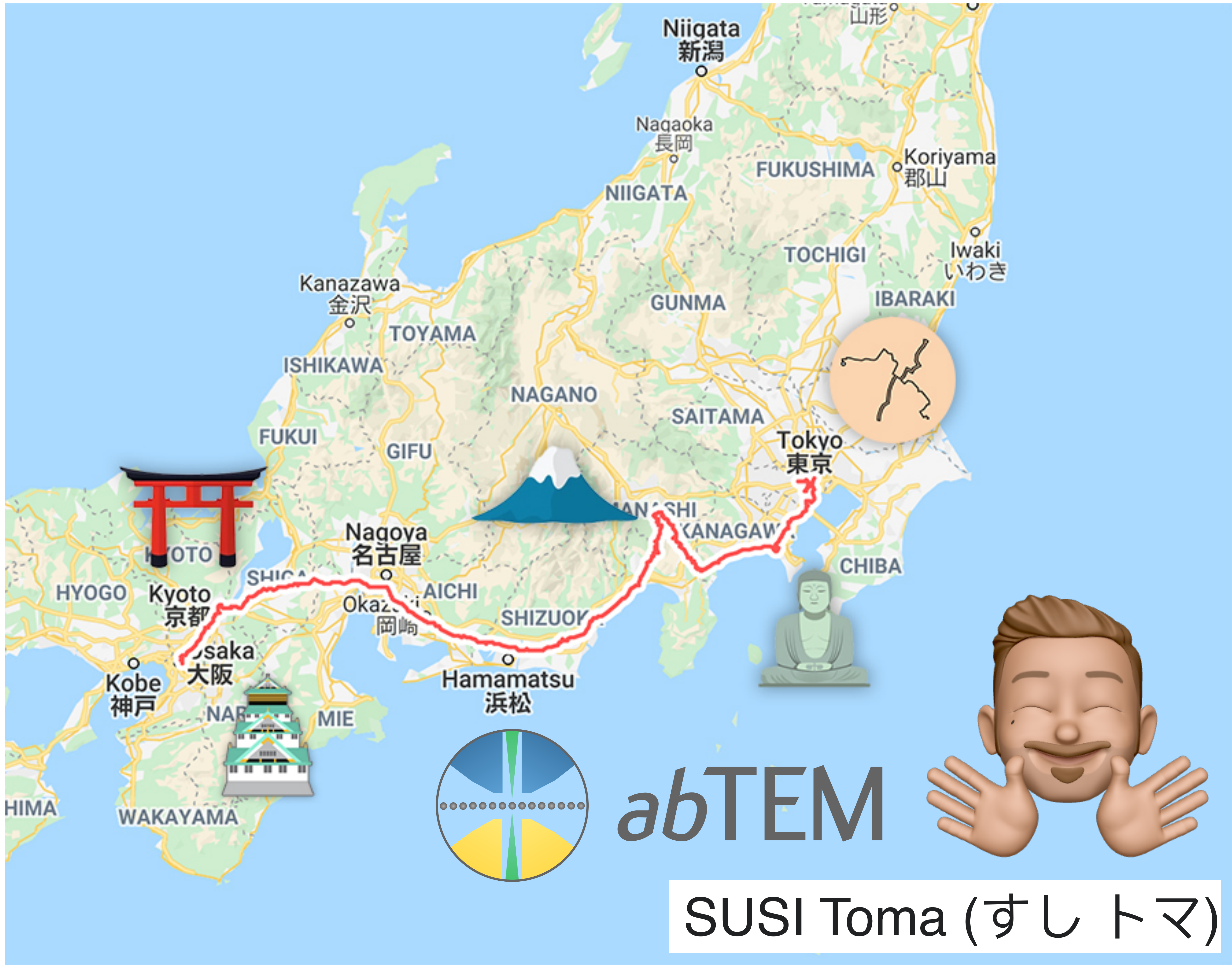
Nagashima (TUS, Sep)

Suenaga (Osaka, Oct)

Shibata (Tokyo, Nov–)



you?



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