Focused-Probe 4D-STEM 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)

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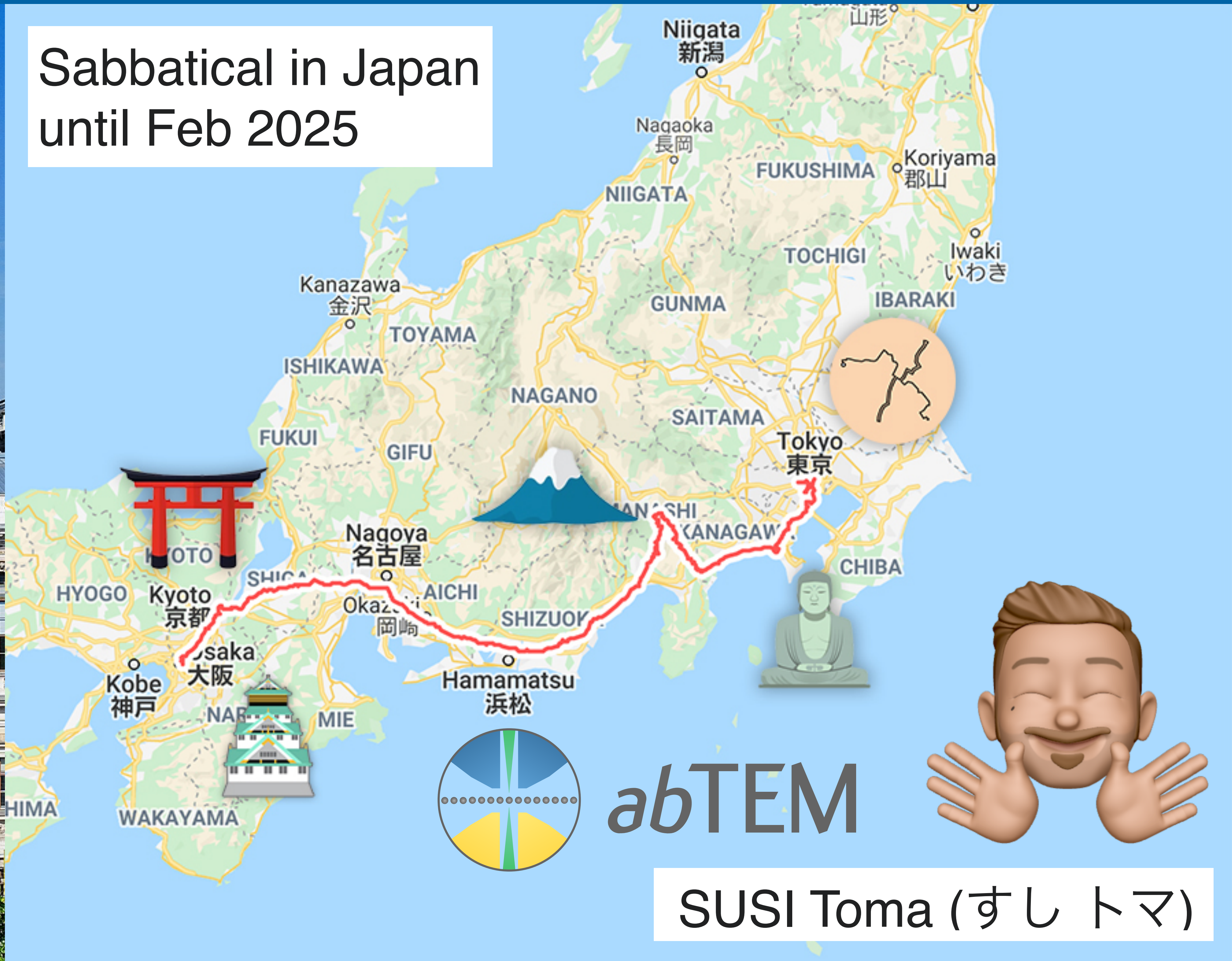


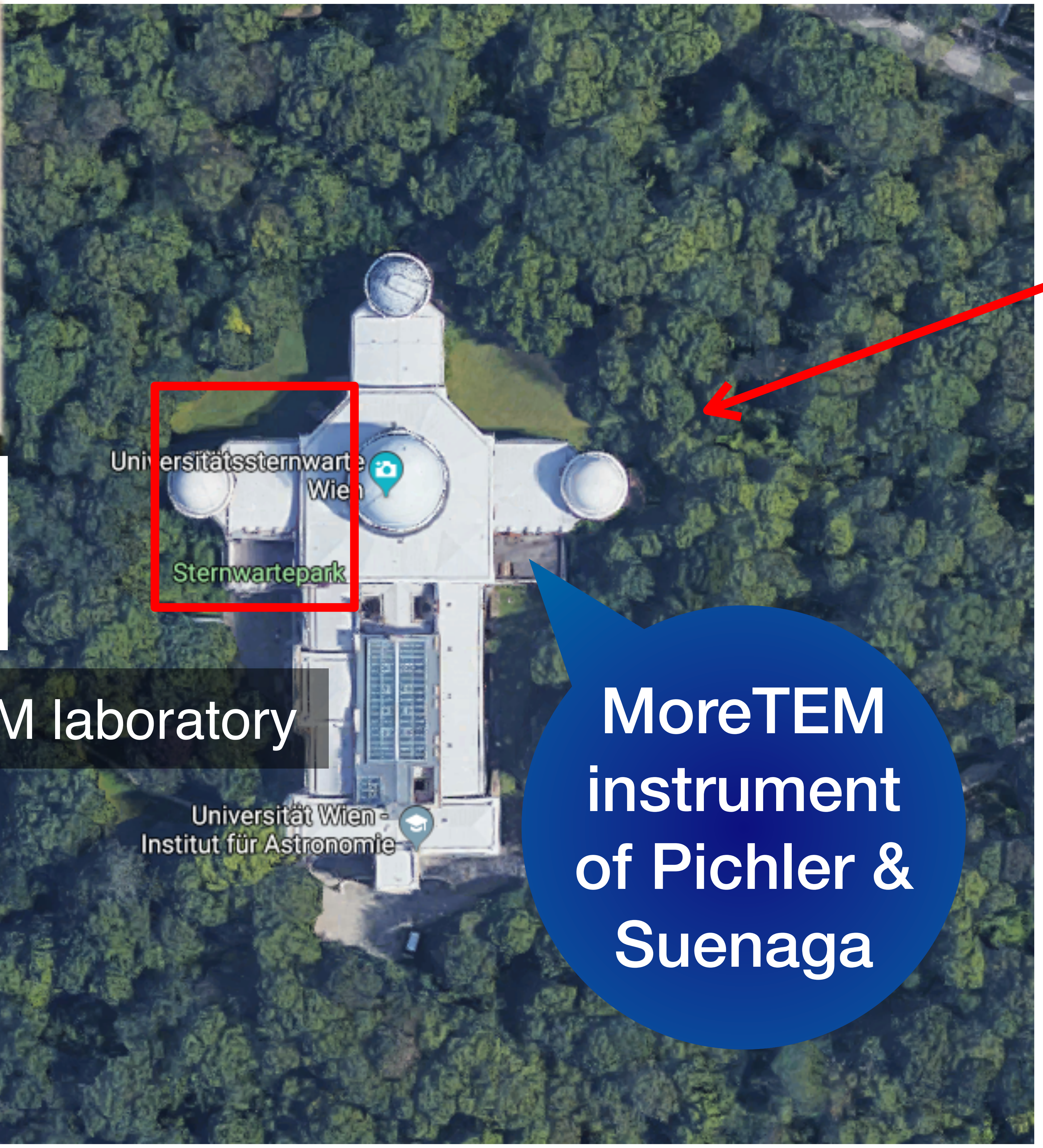
Physics of Nanostructured Materials (PNM)

Jannik Meyer
(U. Tübingen since 2018)

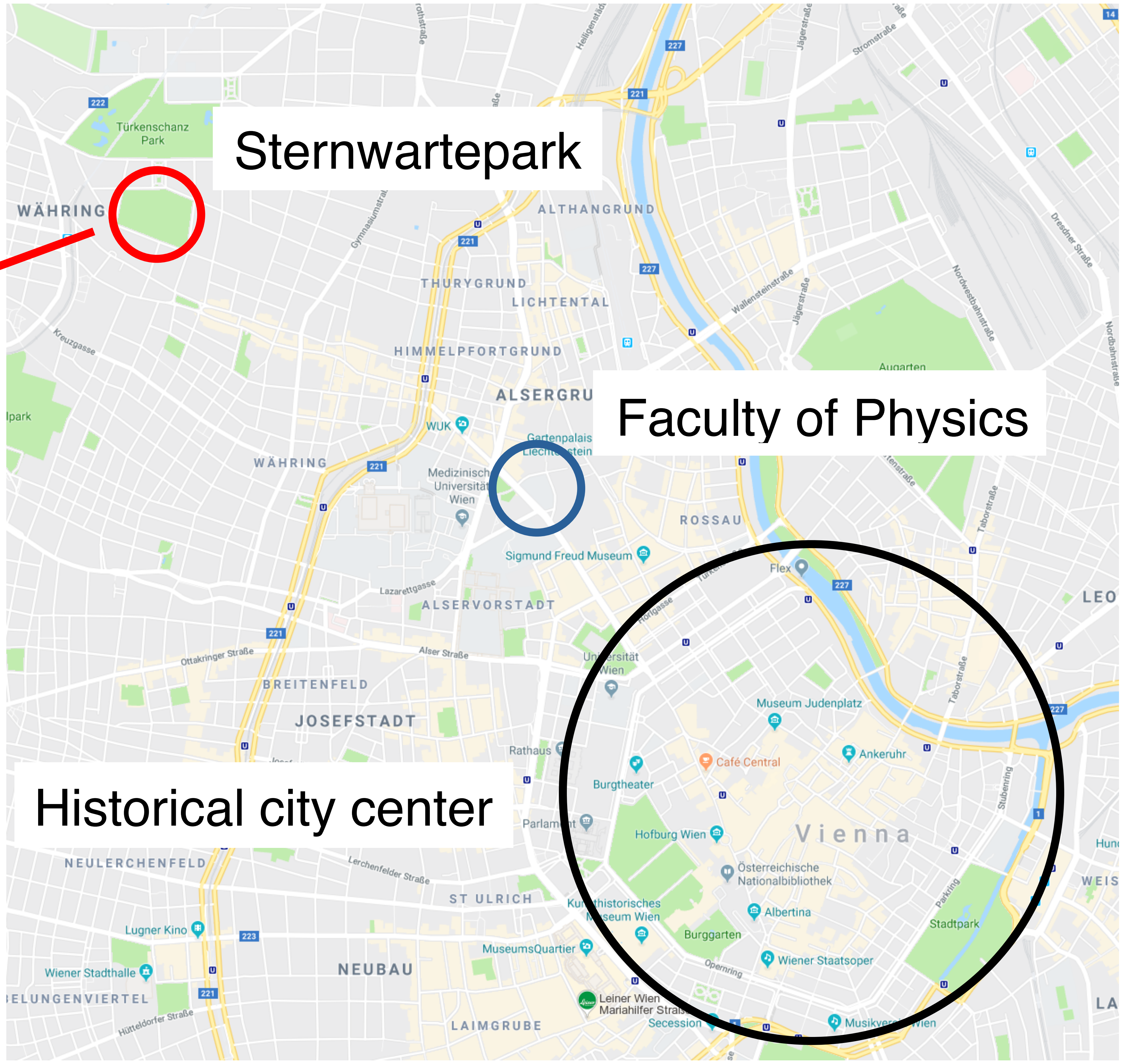
Jani Kotakoski
(group speaker)

Clemens Mangler
(staff scientist)

Sternwartepark



Vienna, Austria



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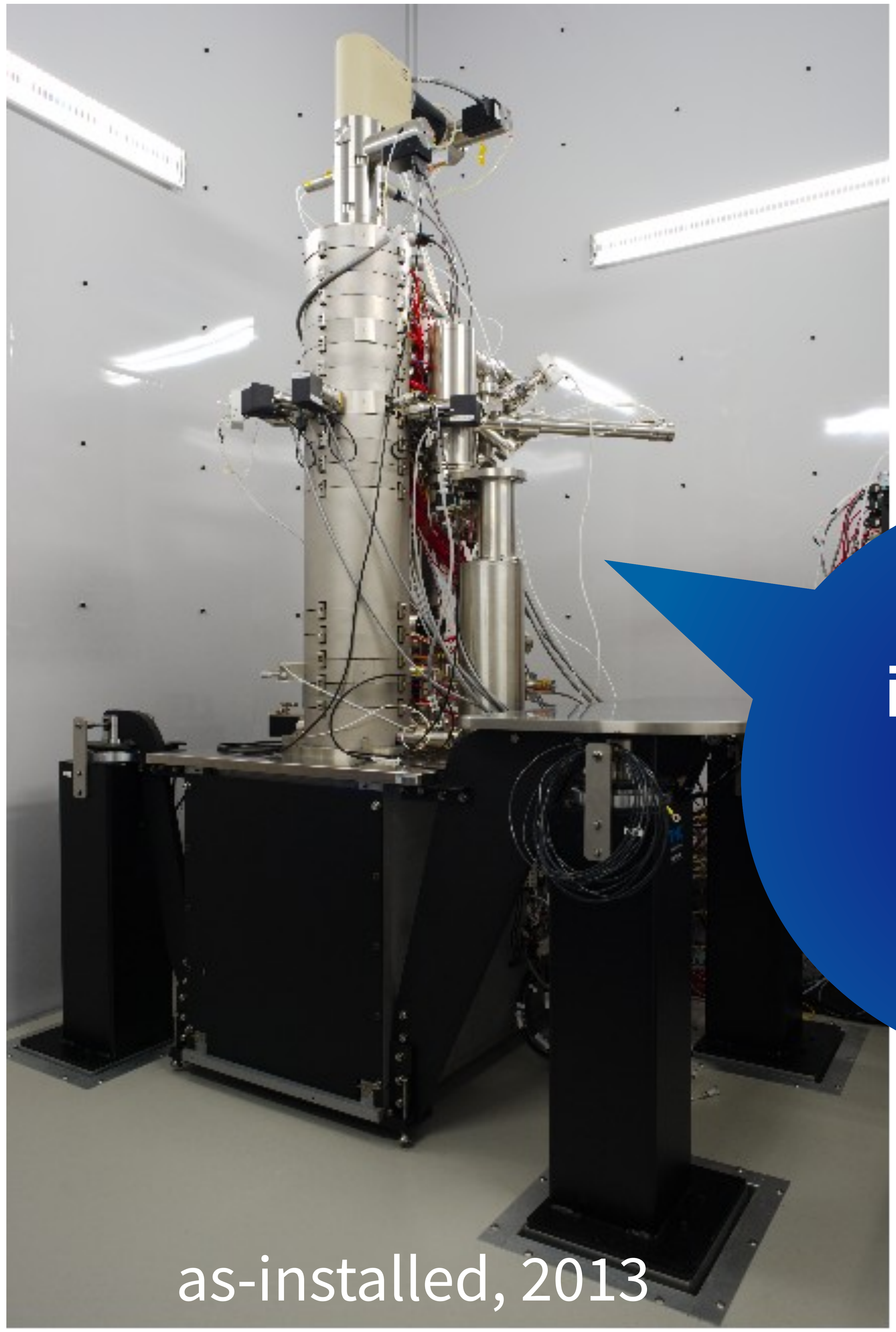
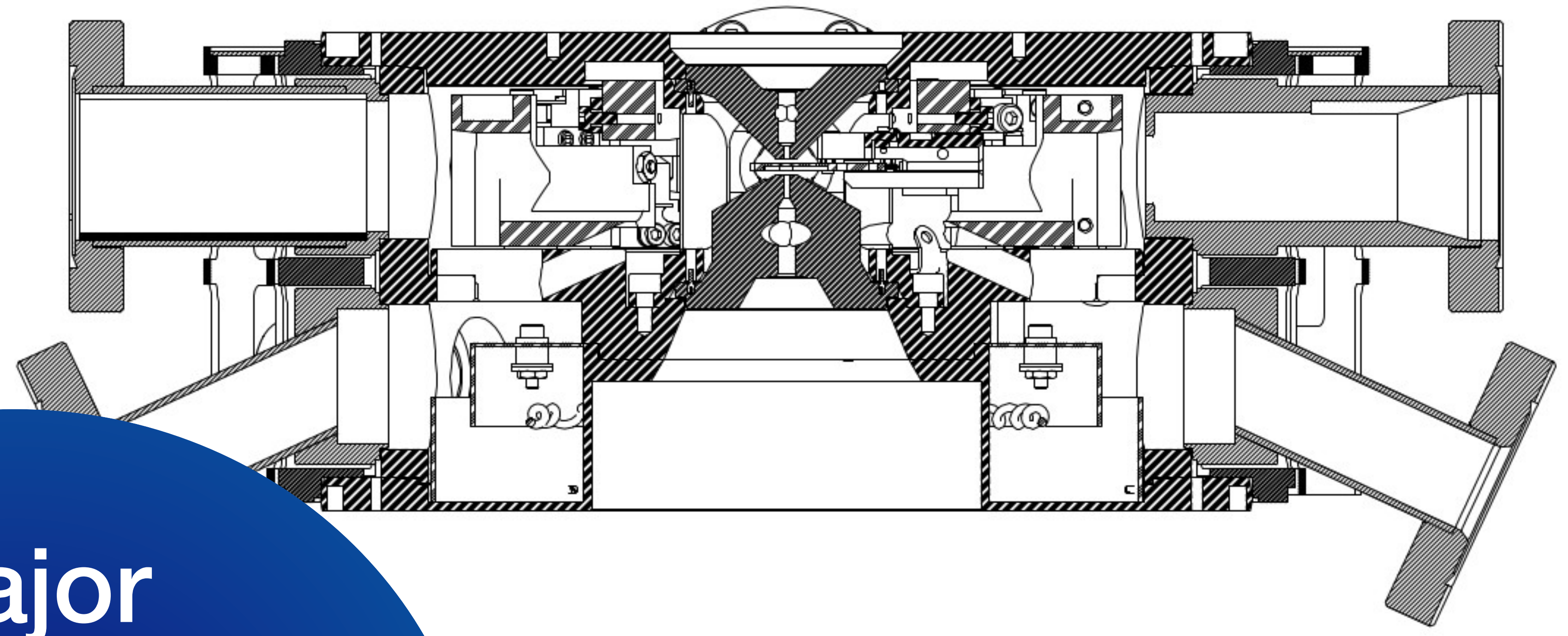
Vienna customized Nion UltraSTEM100



Major instrument upgrade coming by 2026!

Update, 2016



e UHV sample chamber, with an STM puck in the OL polepiece.

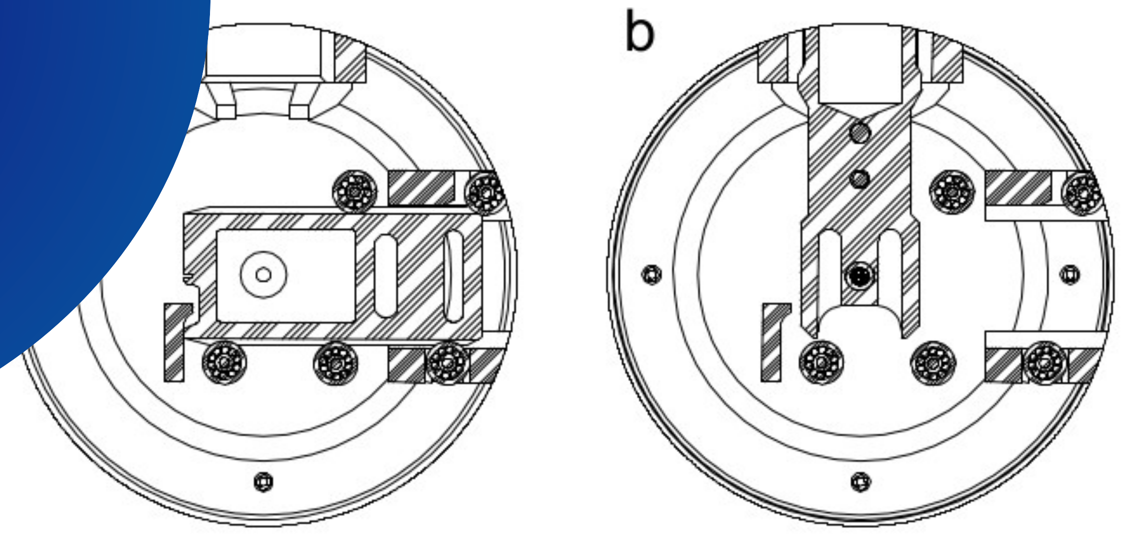


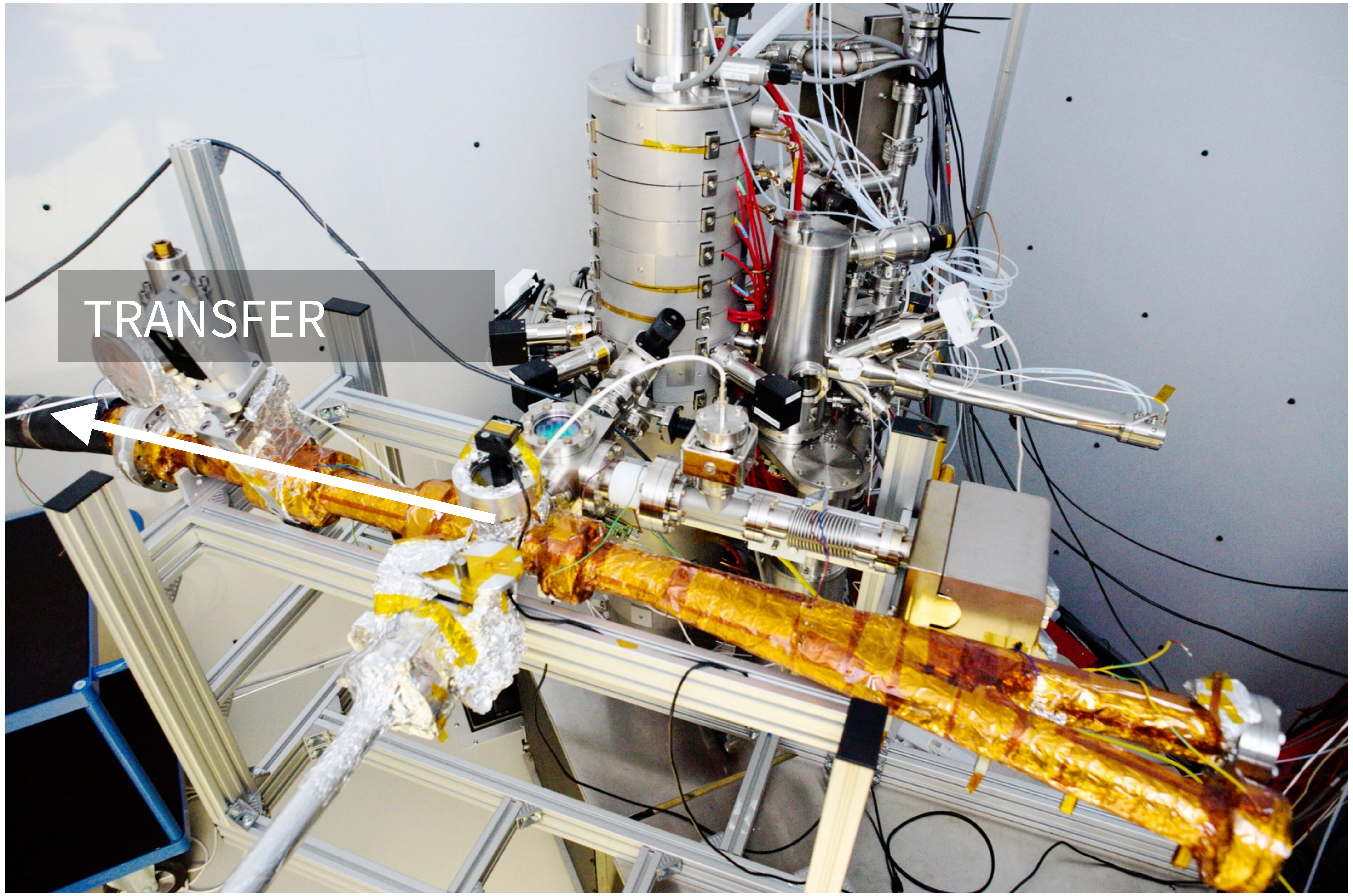
Figure 2. Horizontal cross-sections showing the OL region of the microscope operating with a) scanning tunneling microscope (STM) puck, b) regular Nion sample holder.

- 120 mm high
- Optimized for low Cc (Cc = 1.0 mm, Cs = 1.1 mm)
- Additional pumping
- New CF40 ports facing the sample
- New stage: two different holders

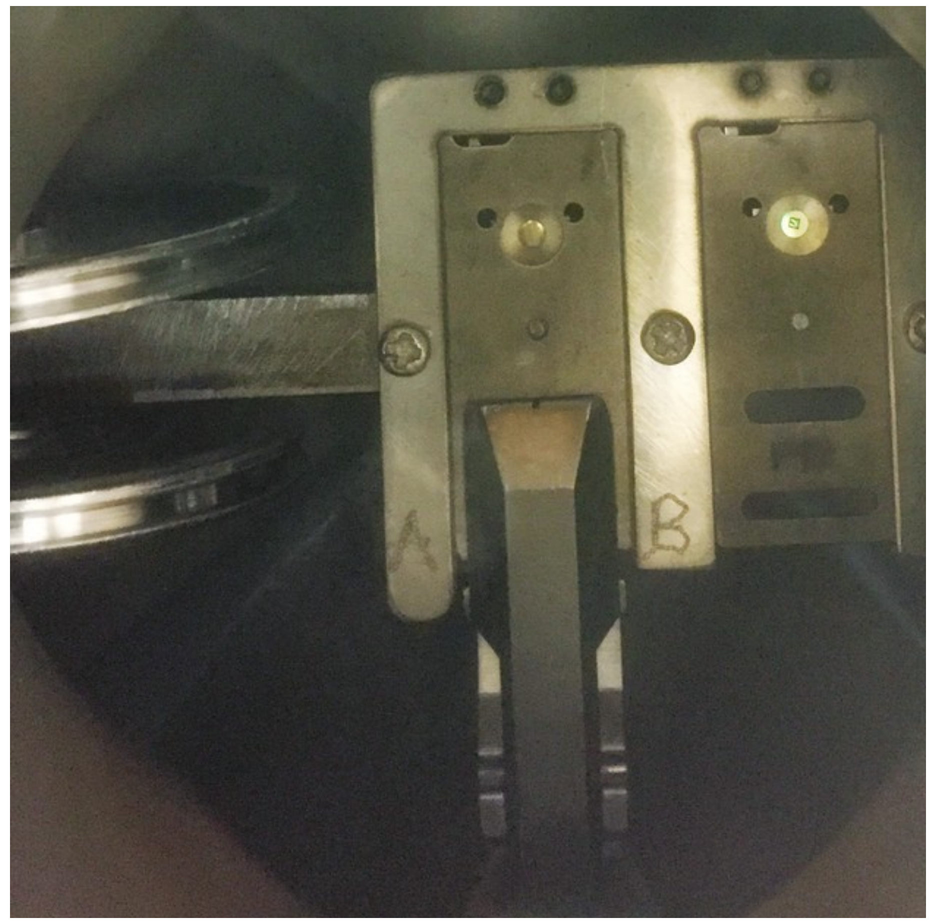
    1. Nion cartridge
    2. Titanium puck

M.T. Hotz *et al.*, Microscopy and Microanalysis 22(S3) (2016) 34

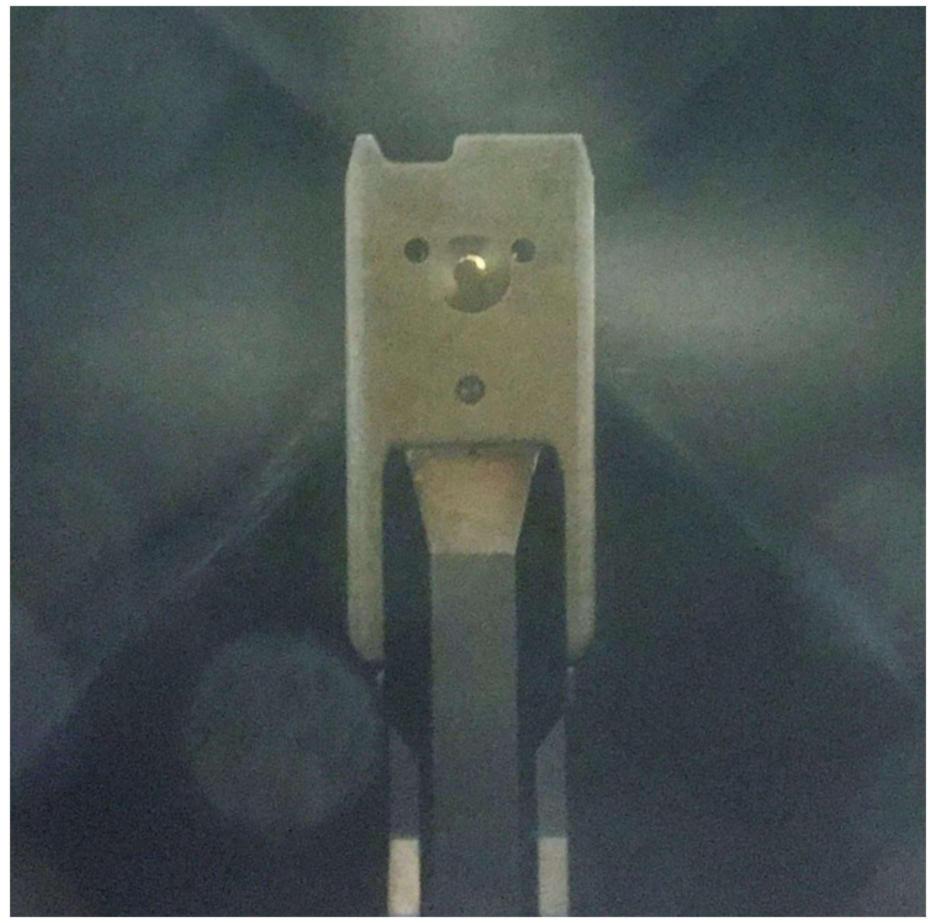
UHV sample transfer system: pucks & cars

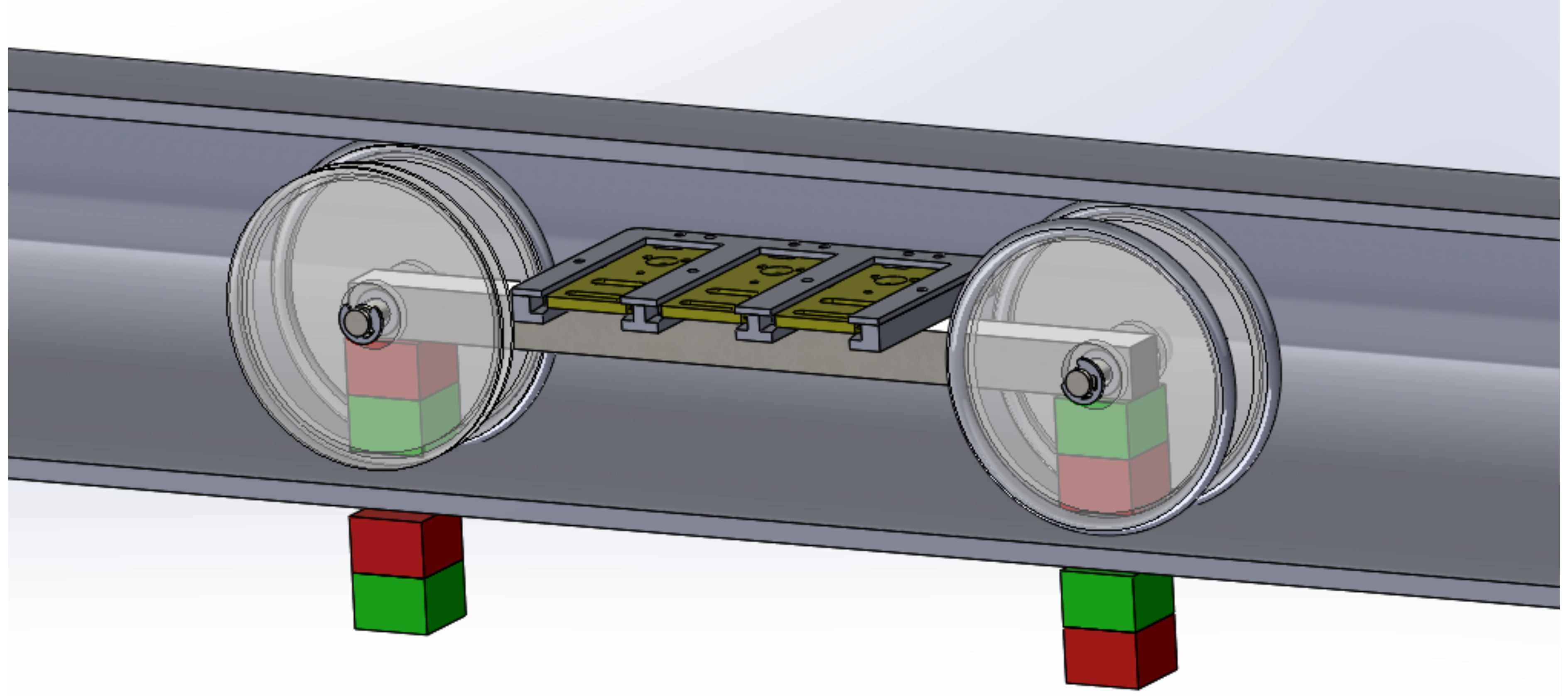


puck in car



puck on gripper

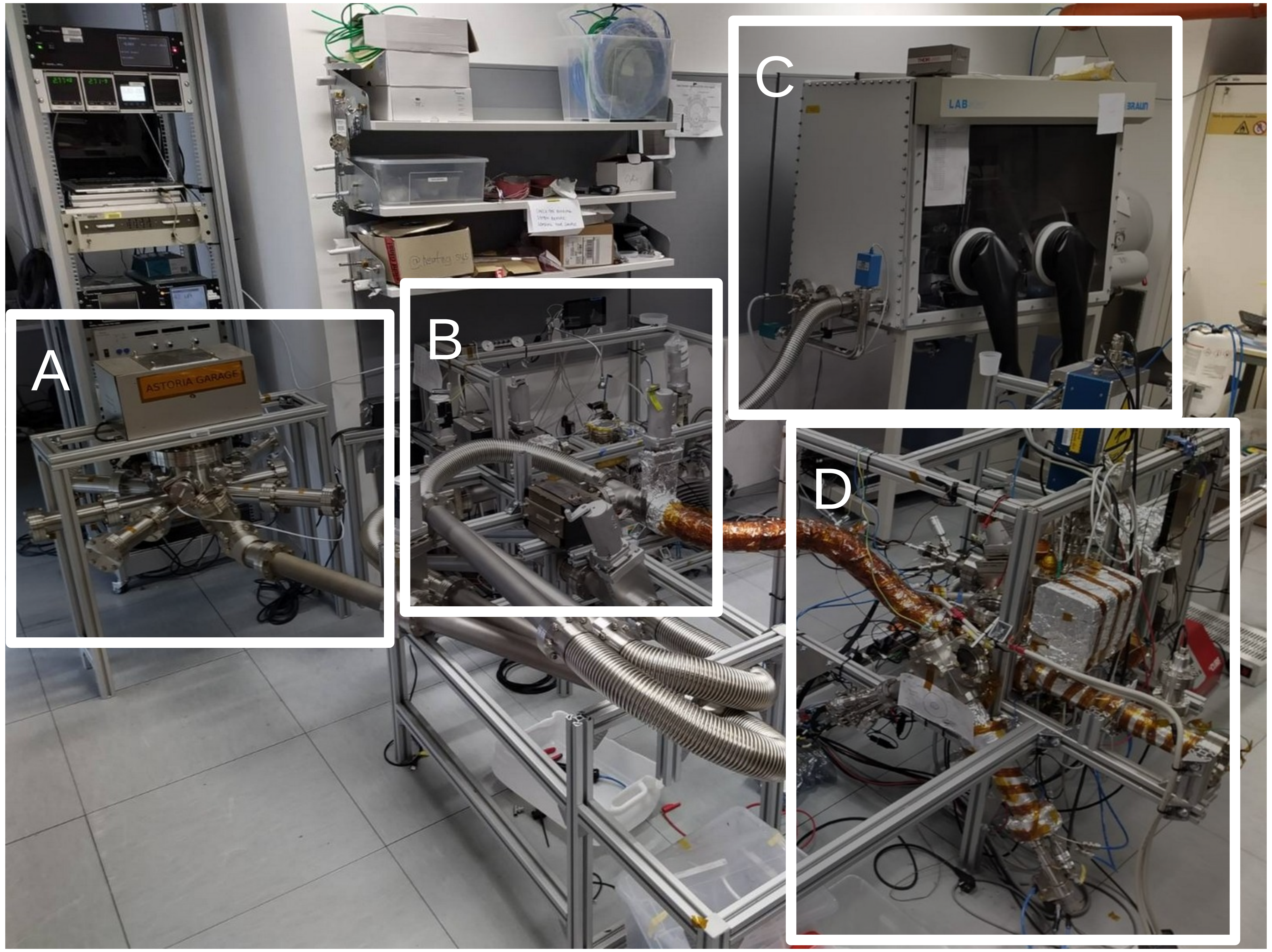




C. Mangler *et al.*, Microscopy and Microanalysis 28(S1) (2022) 2940

First floor: more storage, lots of prep, glove box

First floor



A HV STORAGE UNIT (ASTORIA)

3 x 13 samples, low 1e-9 mbar

B LOAD LOCK

Ar / vacuum load lock w/ 150°C bake

C GLOVE BOX

Ar atmosphere

Dry transfer setup

D TARGET CHAMBER

2 ebeam evaporators

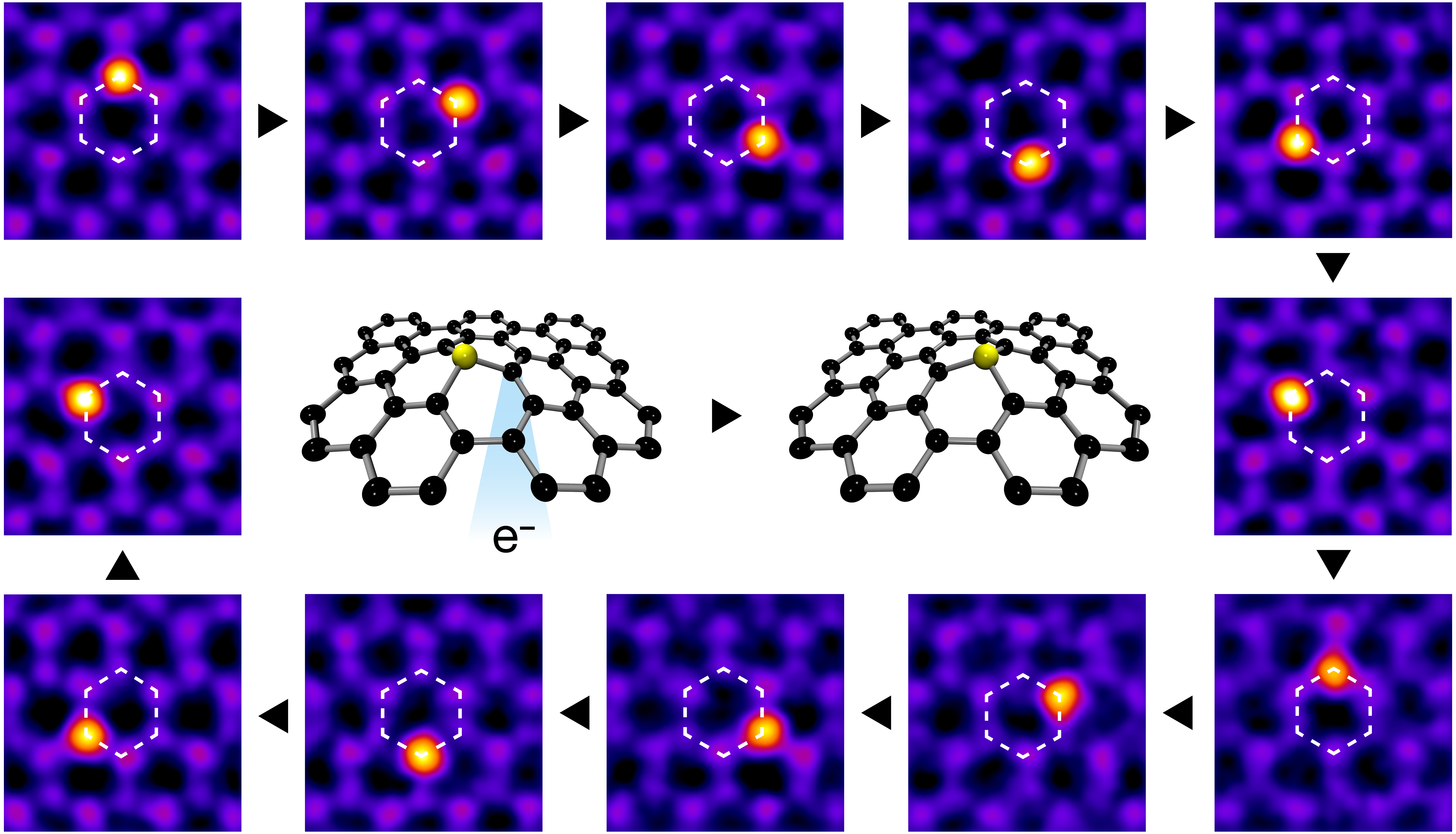
Knudsen cell evaporator

Magnetron plasma ion source

C. Mangler *et al.*, Microscopy and Microanalysis 28(S1) (2022) 2940

2017–2022: Manipulating covalently bound impurity atoms

Vienna
Nion UltraSTEM
60 keV, MAADF



M. Tripathi *et al.*, Nano Lett. 18 (2018) 5319
News Feature: Nature 563 (2018) 462–464

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Cite this: *Chem. Commun.*, 2022, **58**, 12274

Identifying and manipulating single atoms with scanning transmission electron microscopy

Toma Susi *

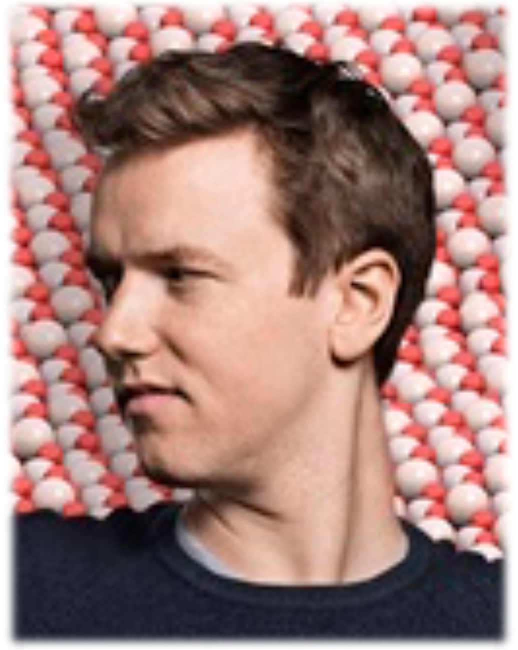
The manipulation of individual atoms has developed from visionary speculation into an established experimental science. Using focused electron irradiation in a scanning transmission electron microscope instead of a physical tip in a scanning probe microscope confers several benefits, including thermal stability of the manipulated structures, the ability to reach into bulk crystals, and the chemical identification of single atoms. However, energetic electron irradiation also presents unique challenges, with an inevitable possibility of irradiation damage. Understanding the underlying mechanisms will undoubtedly continue to play an important role to guide experiments. Great progress has been made in several materials including graphene, carbon nanotubes, and crystalline silicon in the eight years since the discovery of electron-beam manipulation, but the important challenges that remain will determine how far we can expect to progress in the near future.

Received 30th August 2022,
Accepted 28th September 2022

DOI: 10.1039/d2cc04807h

rsc.li/chemcomm

My favourite hobby project: abTEM



Jacob Madsen

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

Rafal Dunin-Borkowski @ Salve 2D22

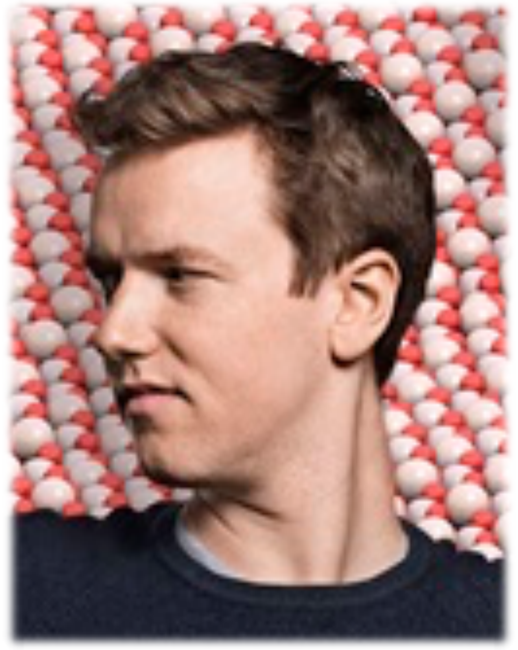
Our multislice solution

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

J. Madsen & **TS**, Open Research Europe 1:24 (2021)

Install: `pip install abtem`
https://abtem.github.io/doc/intro.html

My favourite hobby project: abTEM



Jacob Madsen

DIMENSIONS

165

CROSSREF

106

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

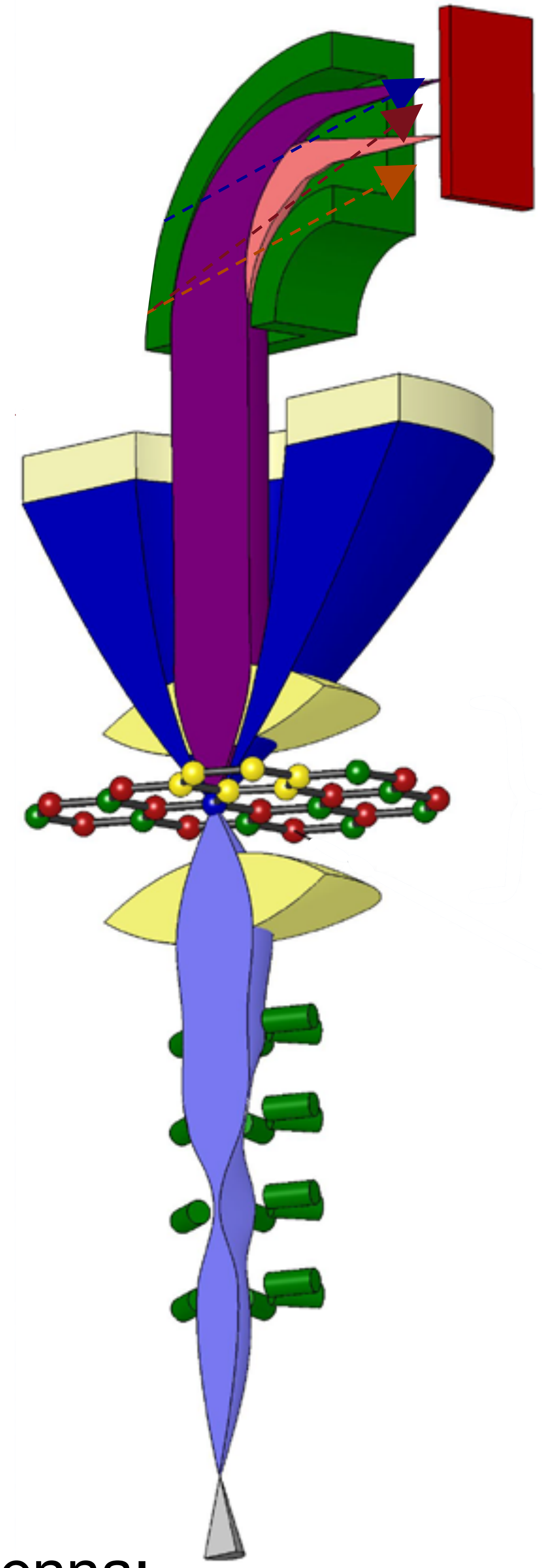
Major upcoming features: — Zarr
- Arbitrary tilts for 3DED / microED
- Plasmon loss for incoherence
- Pauli multislice for magnetic scattering
- True frozen phonon displacements (& vibrational EELS…)

J. Madsen & **TS**, Open Research Europe 1:24 (2021)

Install: `pip install abtem`
https://abtem.github.io/doc/intro.html

Ptycho powered by (Python) open source

MOLECULAR FOUNDRY



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**)

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

nion

Direct-electron detector for 4D-STEM

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!

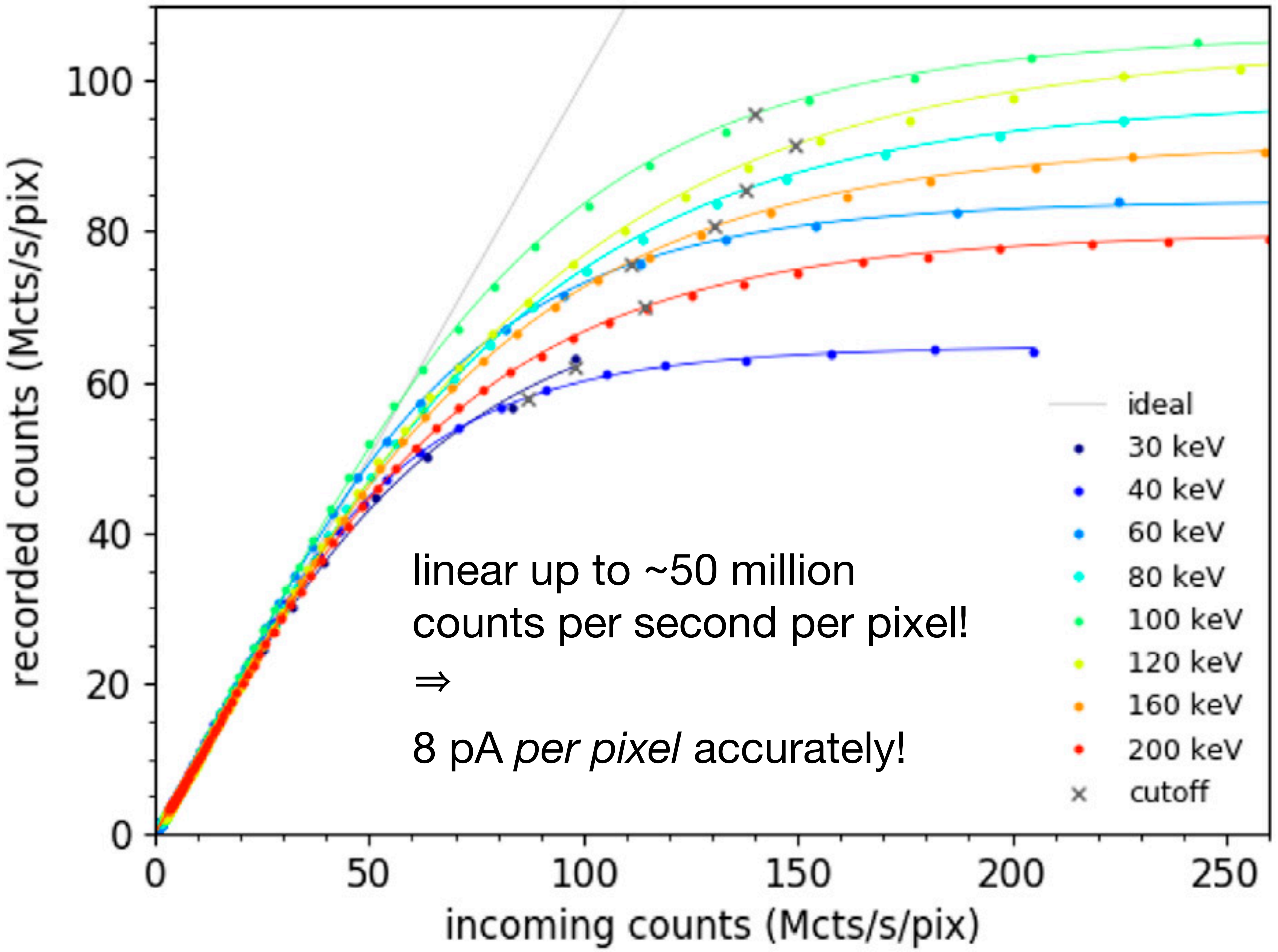
On-axis (btw. HAADF & MAADF)

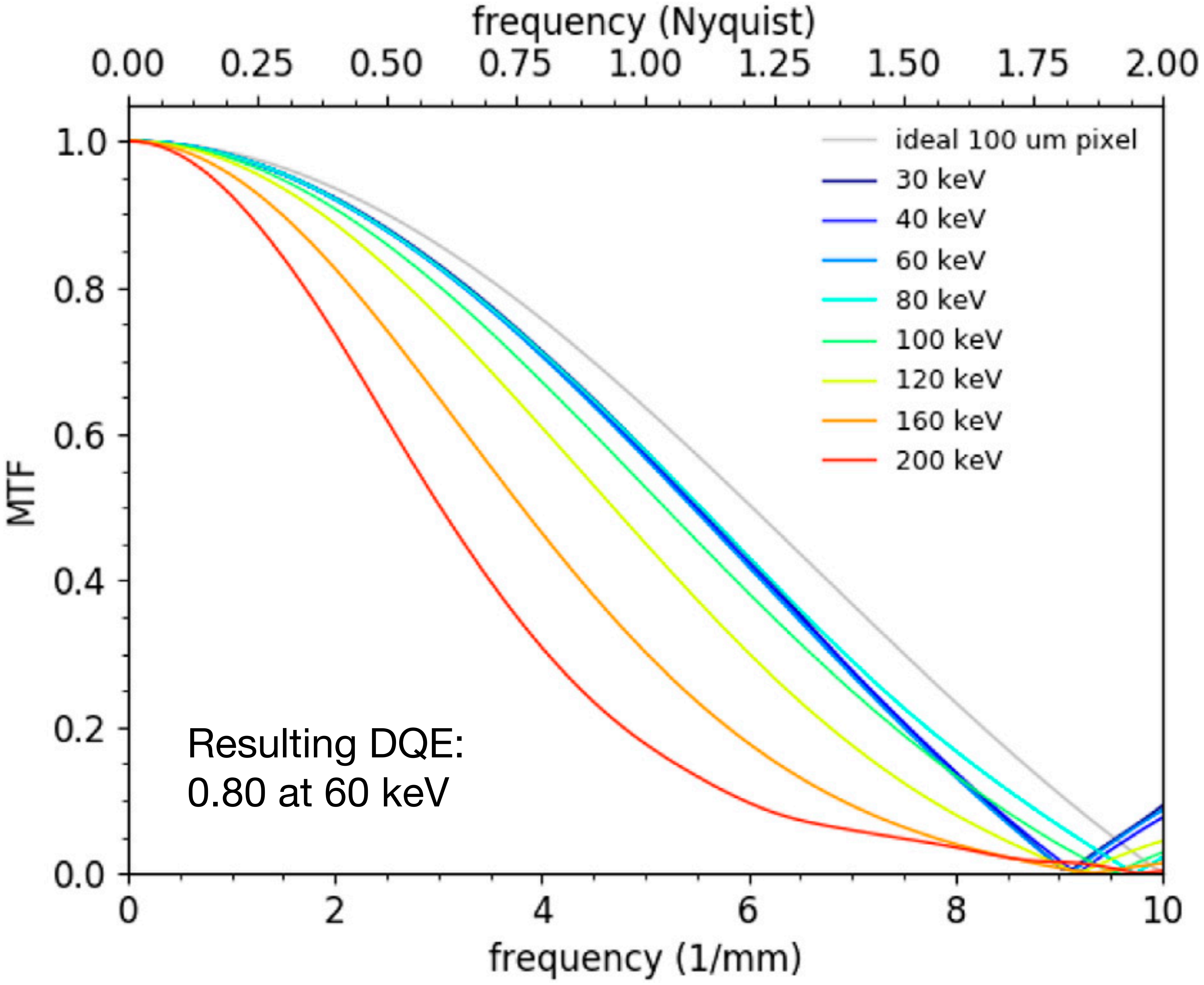
Near-ideal 4D-STEM performance at 60 keV

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

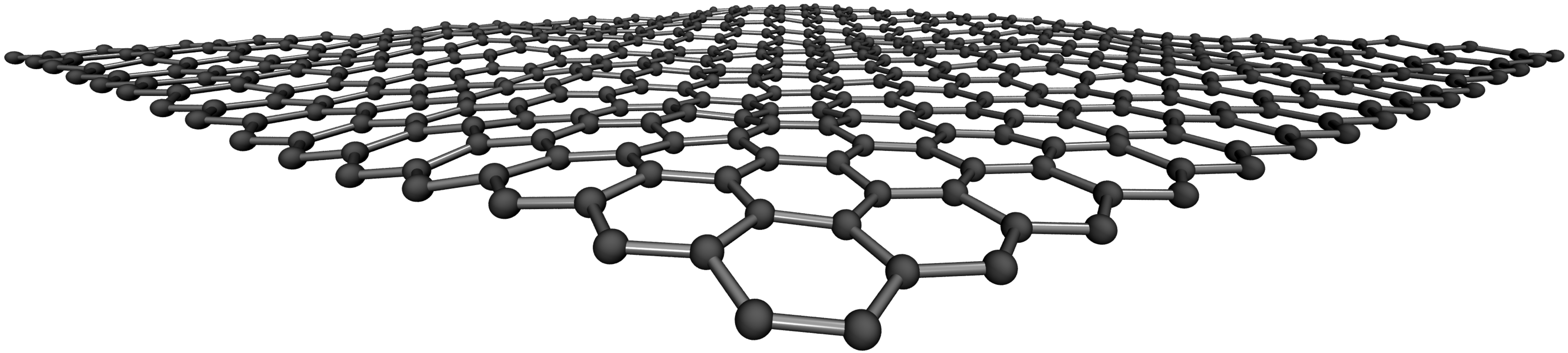




P. Zambon *et al.*, Front. Phys. 11:1308321 (2023)

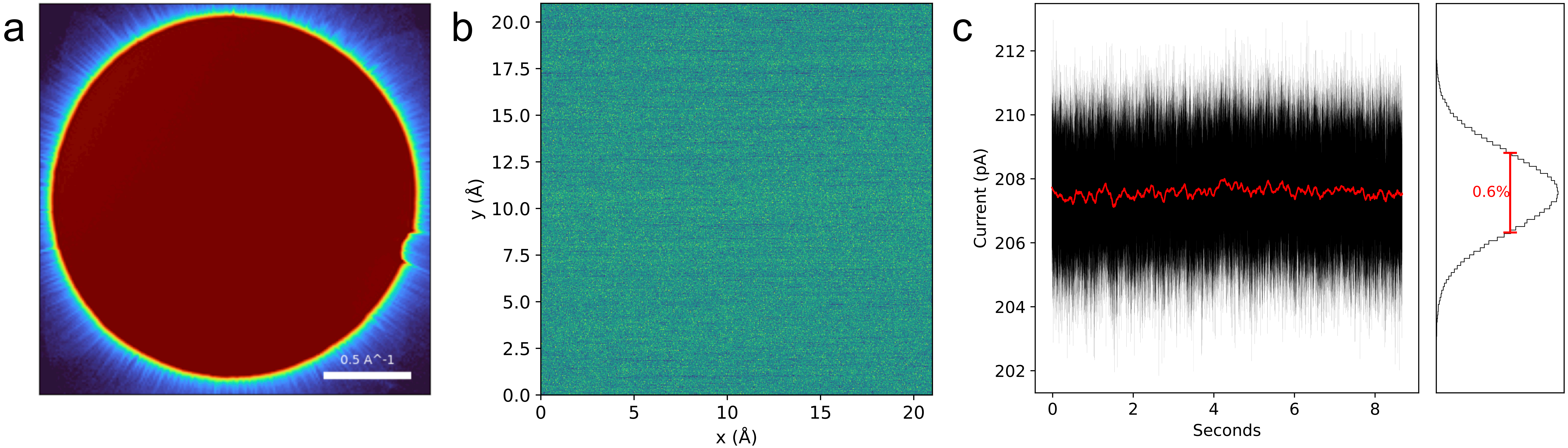
Graphene as ideal uniform phase object

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



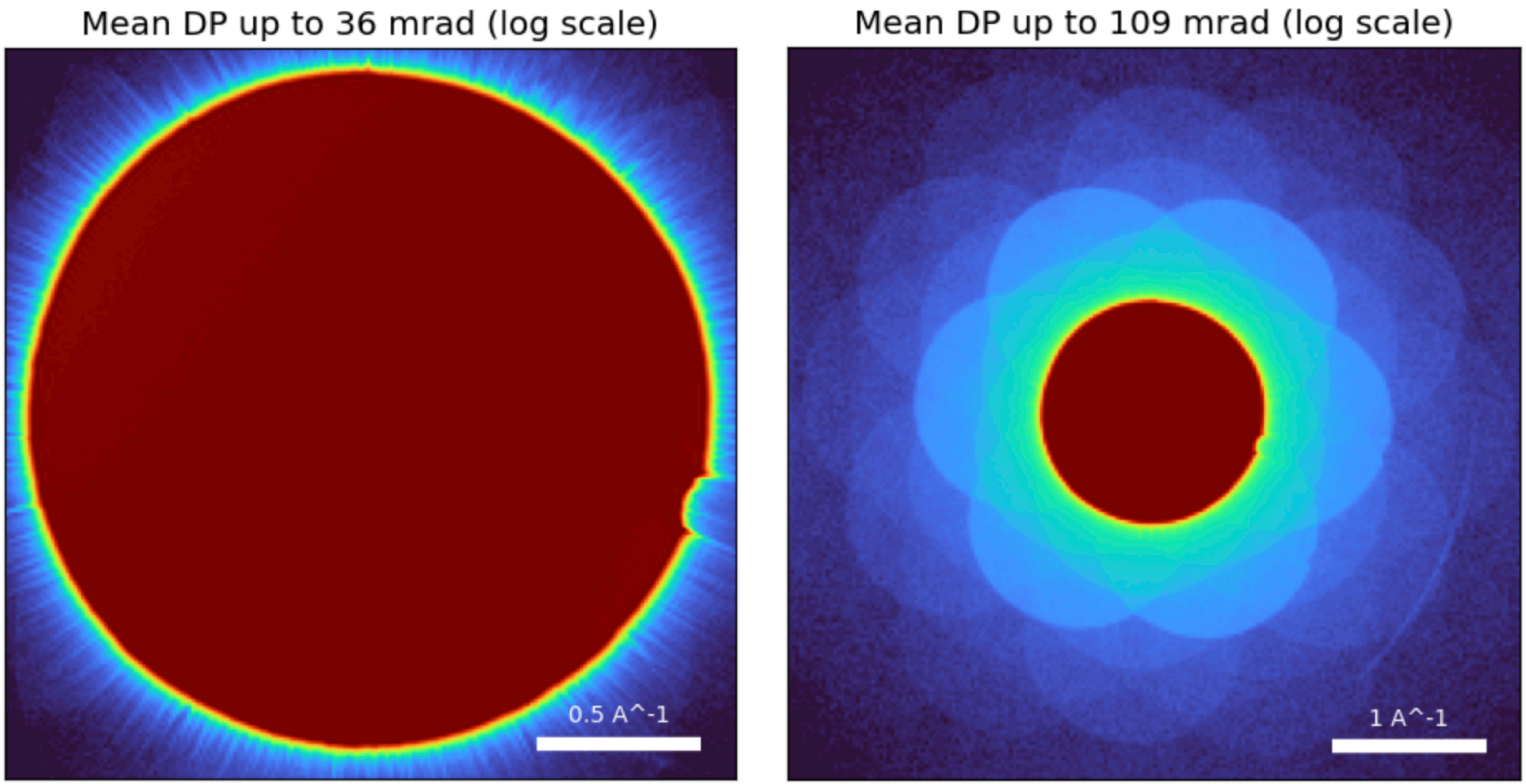
Nion + ARINA 4D-STEM data collection

Example: 512 × 512 px scan (***R*** **px**), 192 × 192 px CBED (***Q*** **px**), 33.3 us dwell time (207 pA!)



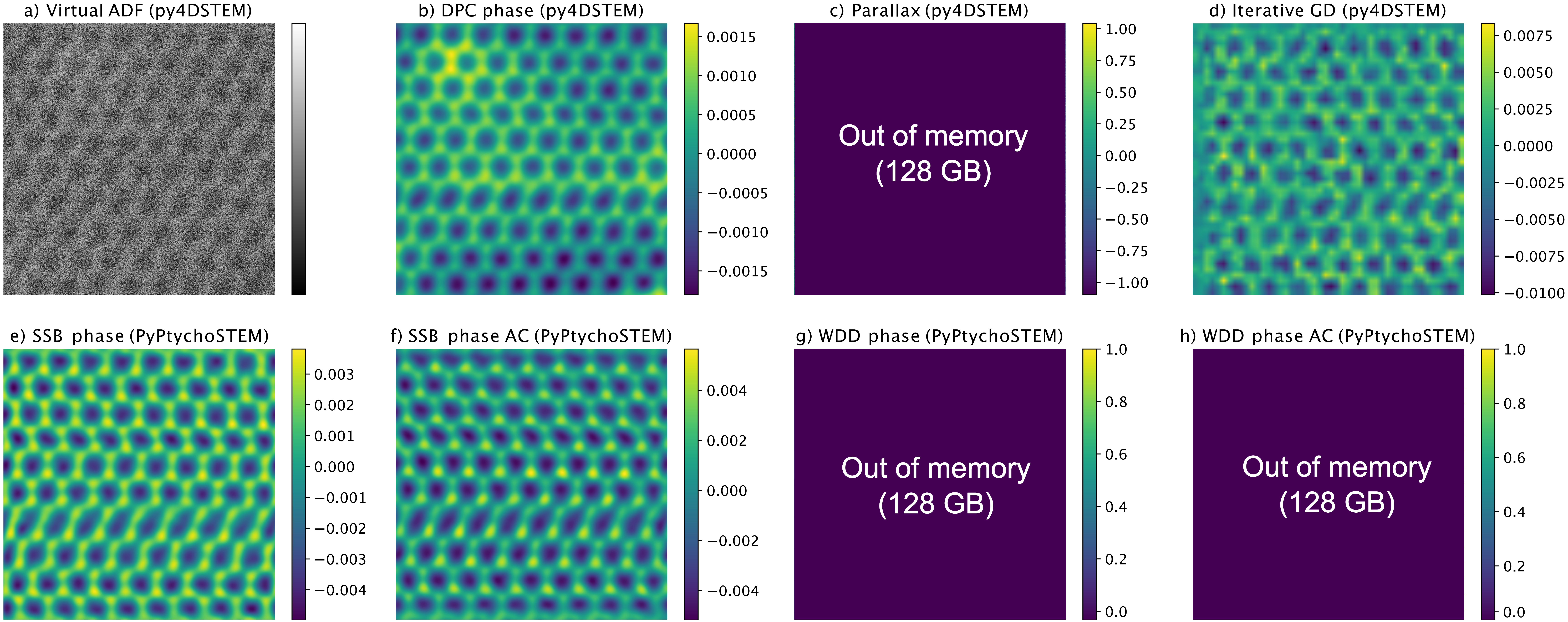
- 34 mrad semi-convergence angle, variable camera length (max collection angle)
- counting electrons: accurate dose (& dose per pixel can be normalized)

Nion + ARINA variable camera lengths



T. Susi, submitted (*Journal of Microscopy* ptychography special issue)

Comparing algorithms (short camera length $\boldsymbol{Q}$ *binning*)

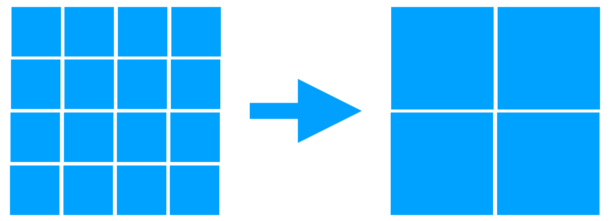


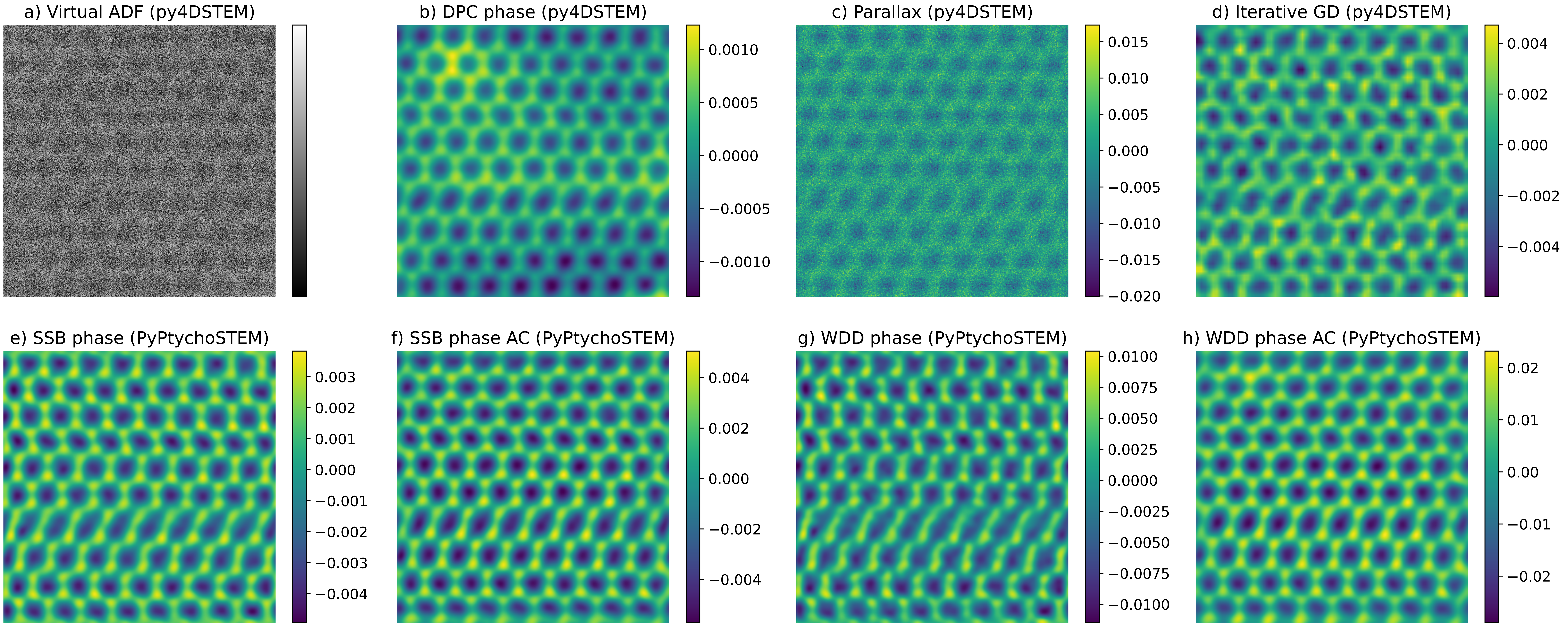
no binning in ***Q***

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 algorithms (short camera length ***Q*** *binning*)





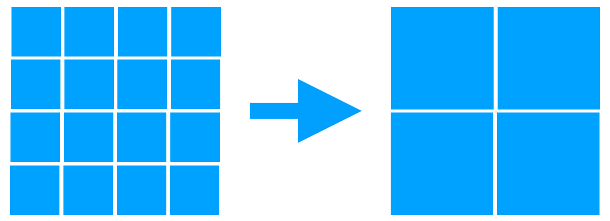
2× binning in ***Q***

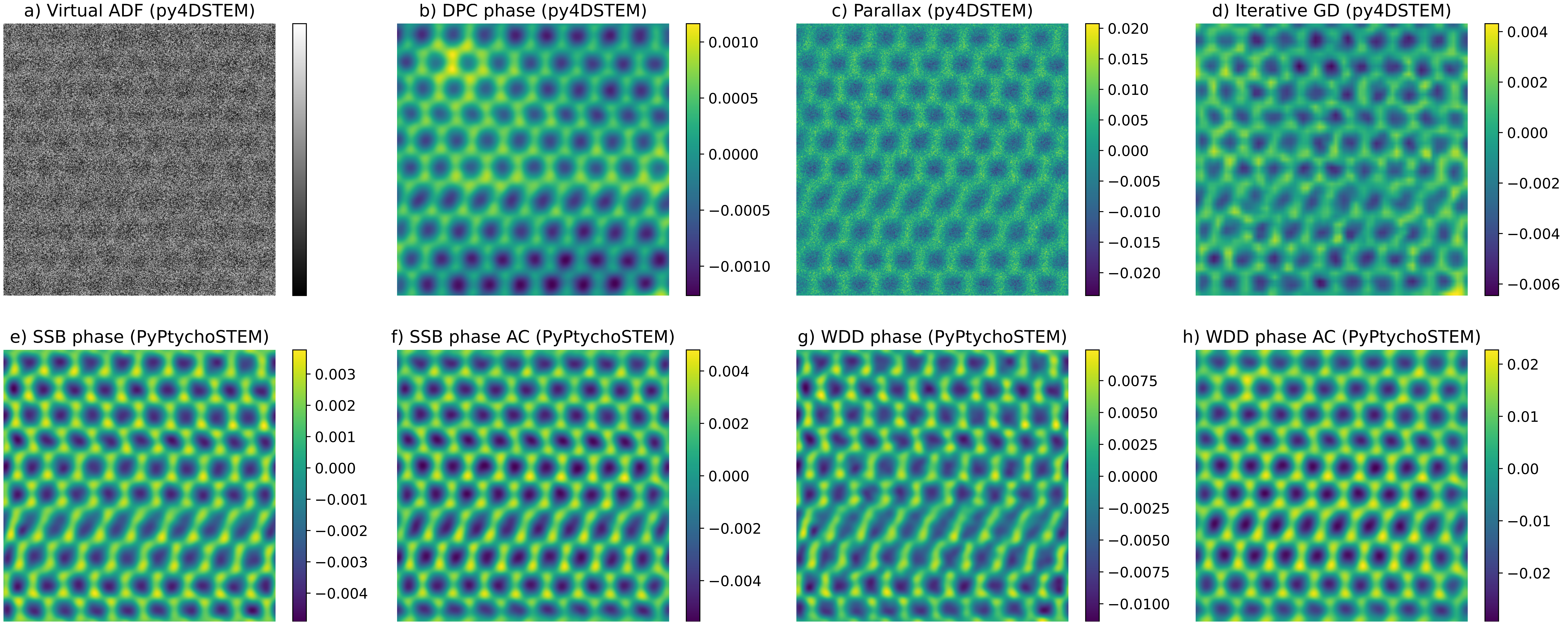
Binning in ***Q***: **36 mrad** max. scattering angle, 33.3 us dwell time, 512×512 ***R*** px, 192×192 ***Q*** px

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

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

Comparing algorithms (short camera length ***Q*** *binning*)





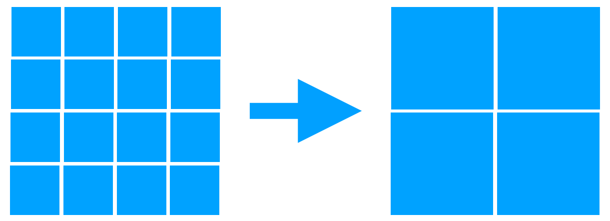
4× binning in ***Q***

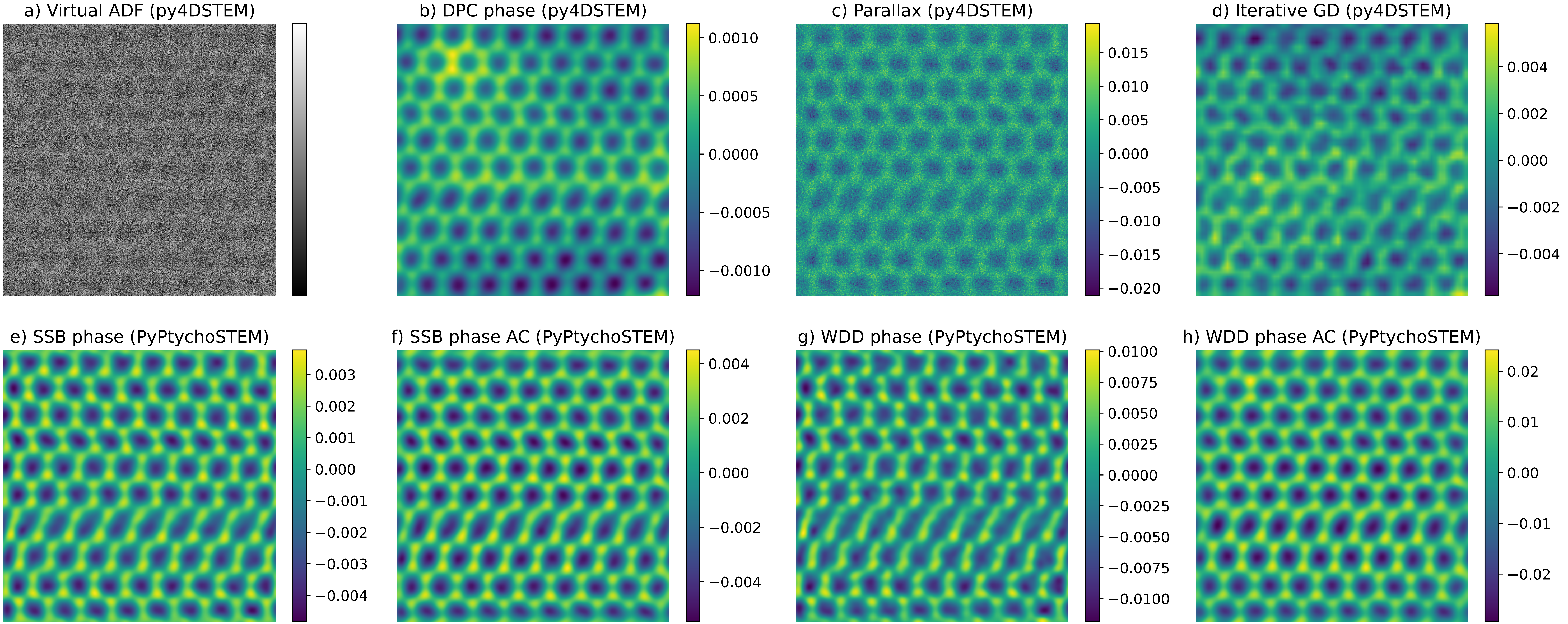
Binning in ***Q***: **36 mrad** max. scattering angle, 33.3 us dwell time, 512×512 ***R*** px, 192×192 ***Q*** px

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

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

Comparing algorithms (short camera length ***Q*** *binning*)





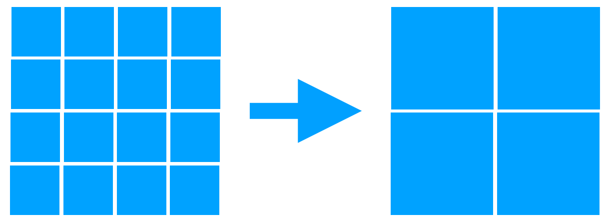
8× binning in ***Q***

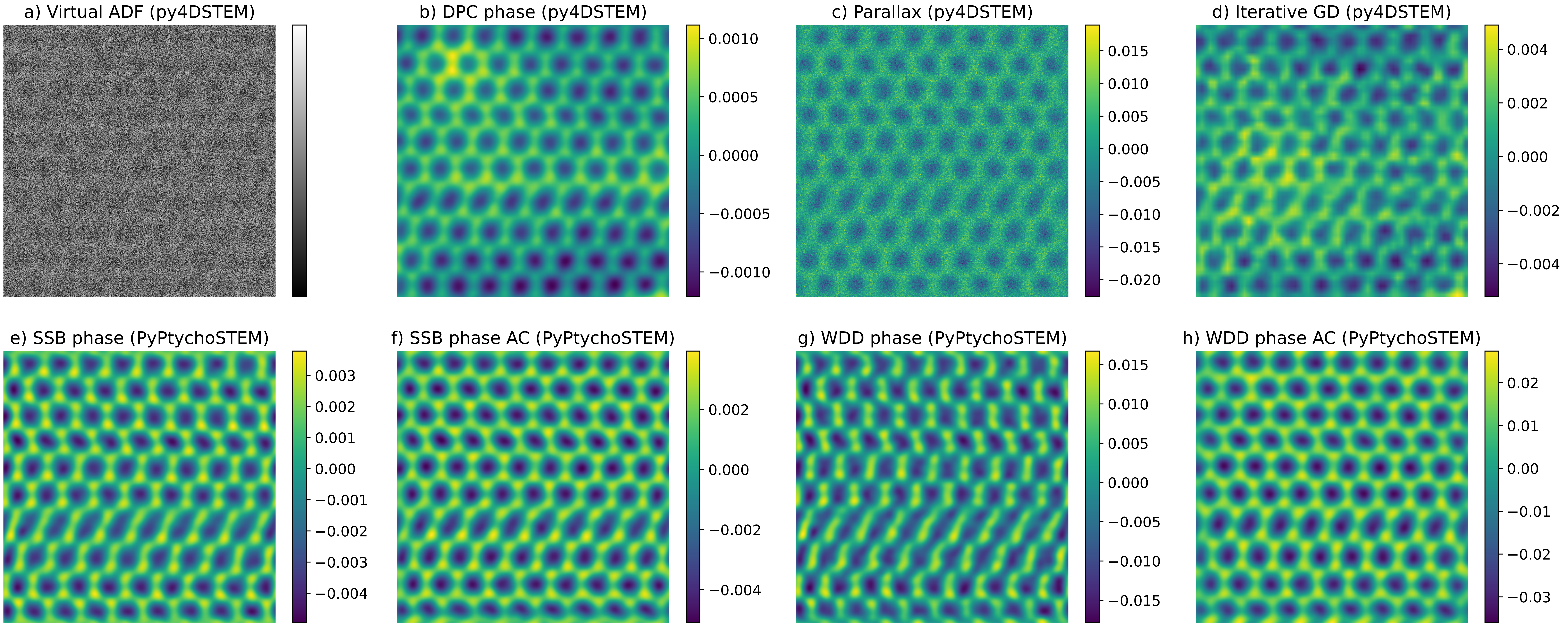
Binning in ***Q***: **36 mrad** max. scattering angle, 33.3 us dwell time, 512×512 ***R*** px, 192×192 ***Q*** px

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

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

Comparing algorithms (short camera length ***Q*** *binning*)



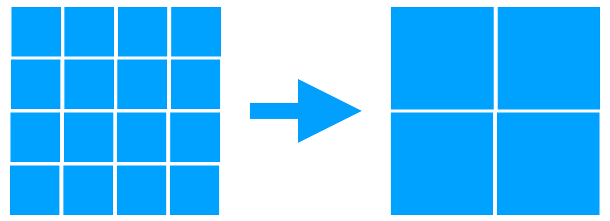


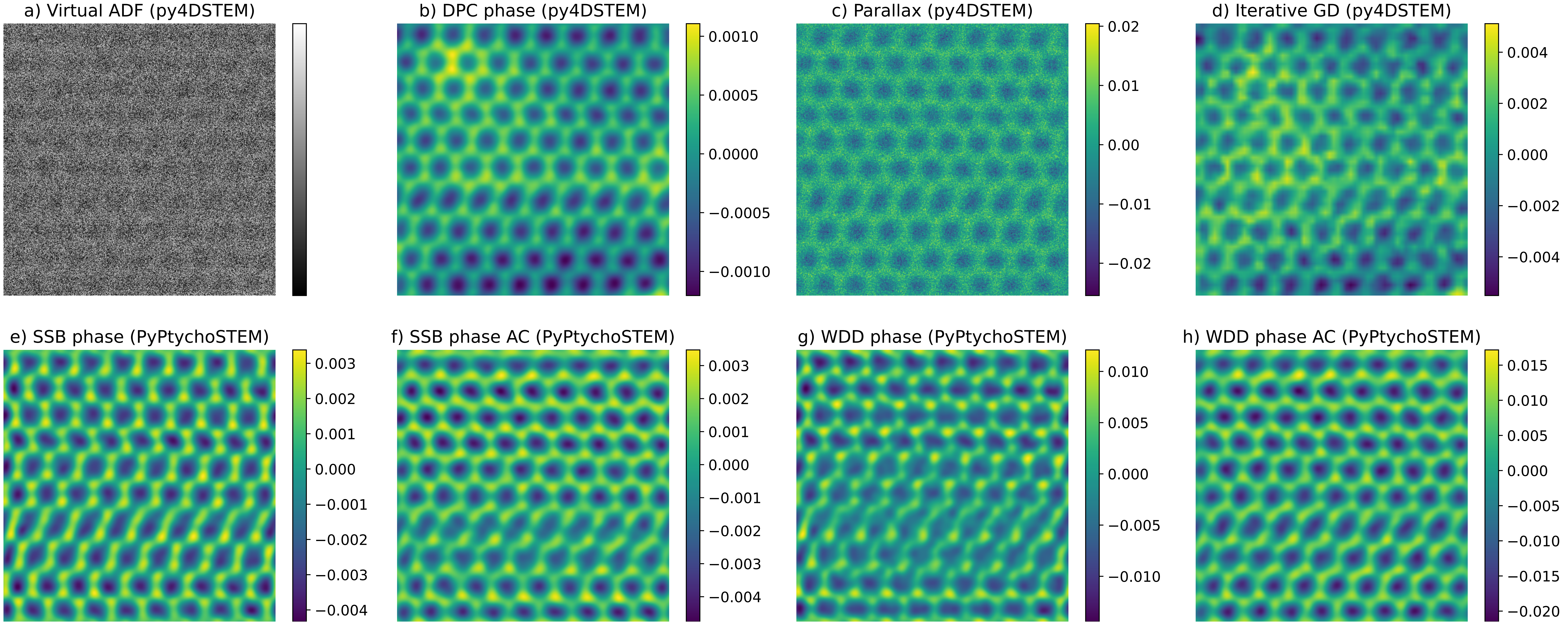
16× binning in ***Q***

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 algorithms (short camera length ***Q*** *binning*)





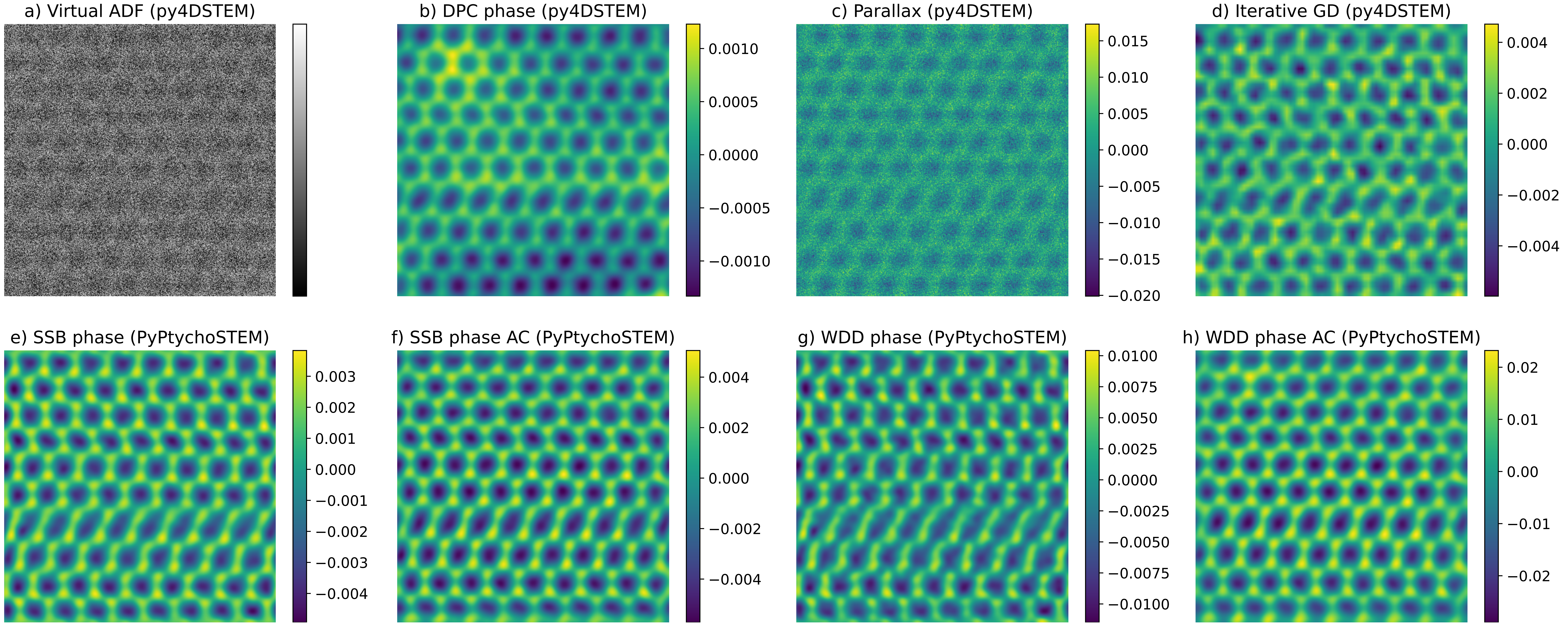
32× binning in ***Q***

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

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

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

Comparing algorithms (short camera length *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)

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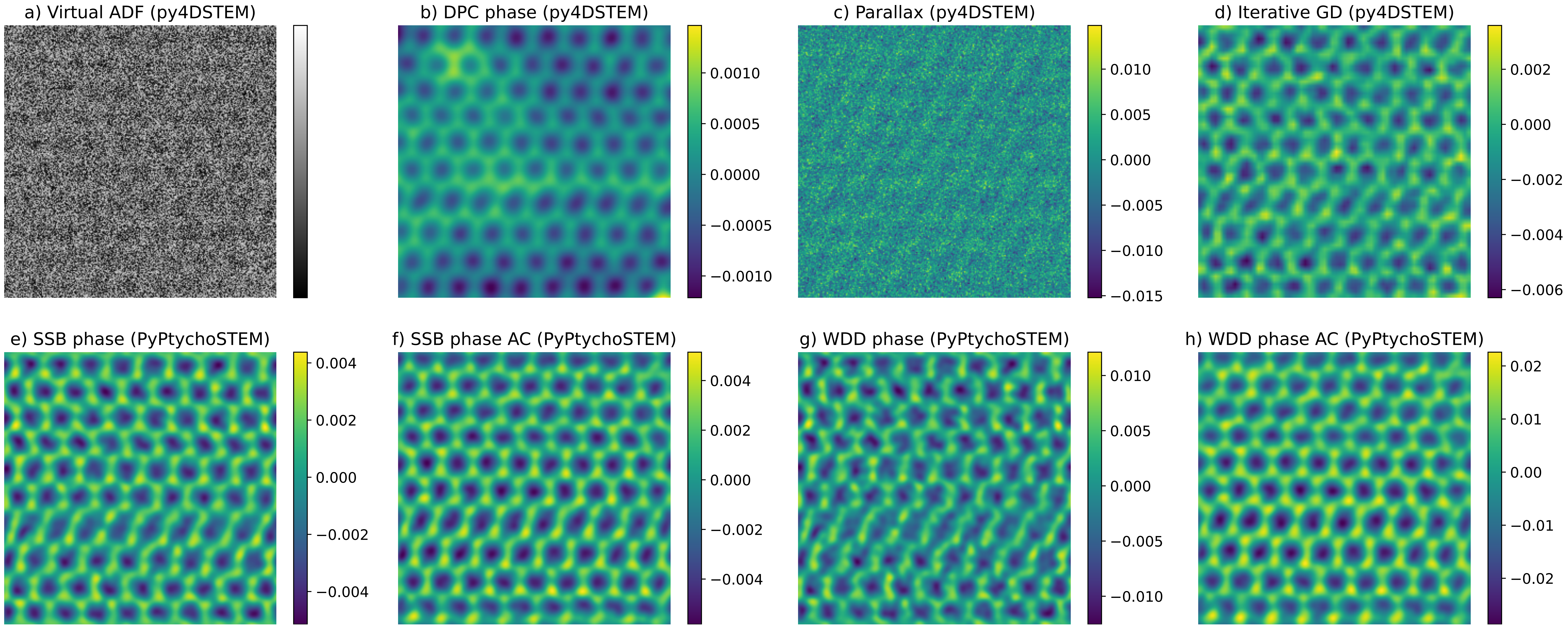
Comparing algorithms (short camera length ***R*** *thinning*)

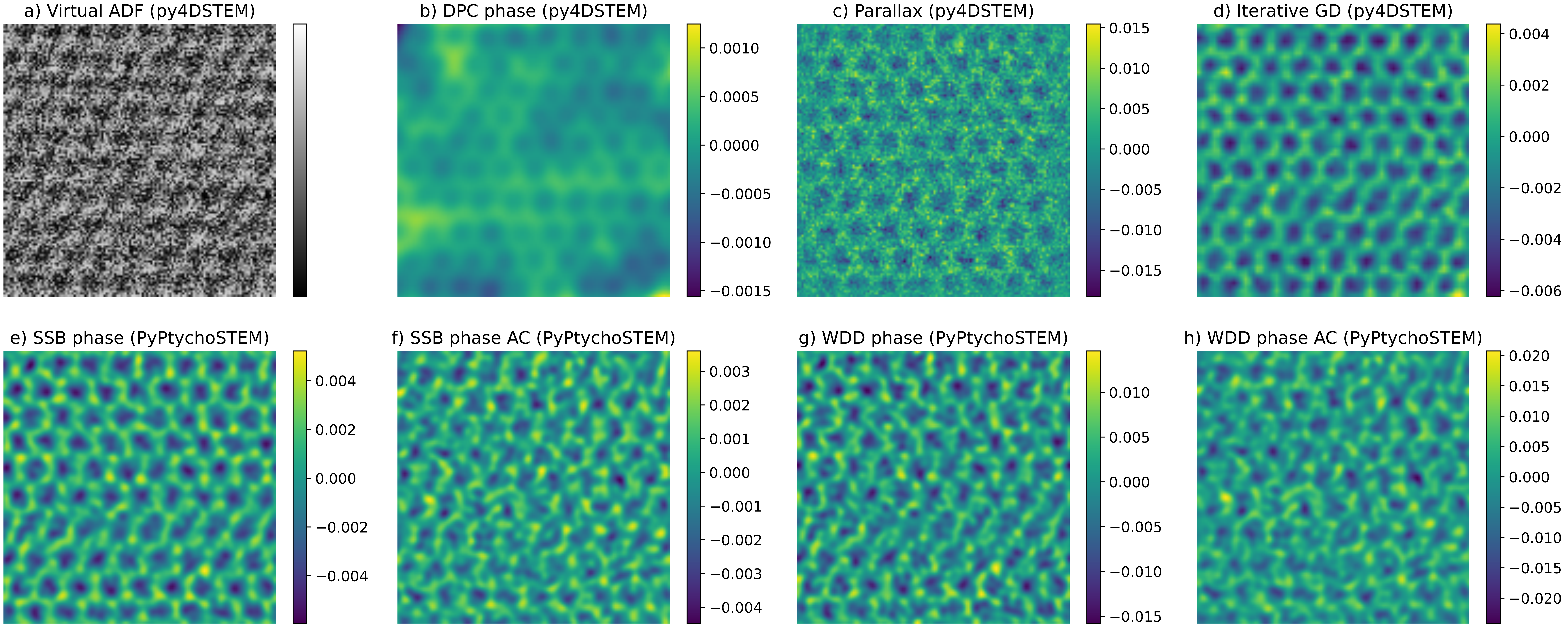


2× thinning in ***R***

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 algorithms (short camera length *R* *thinning*)

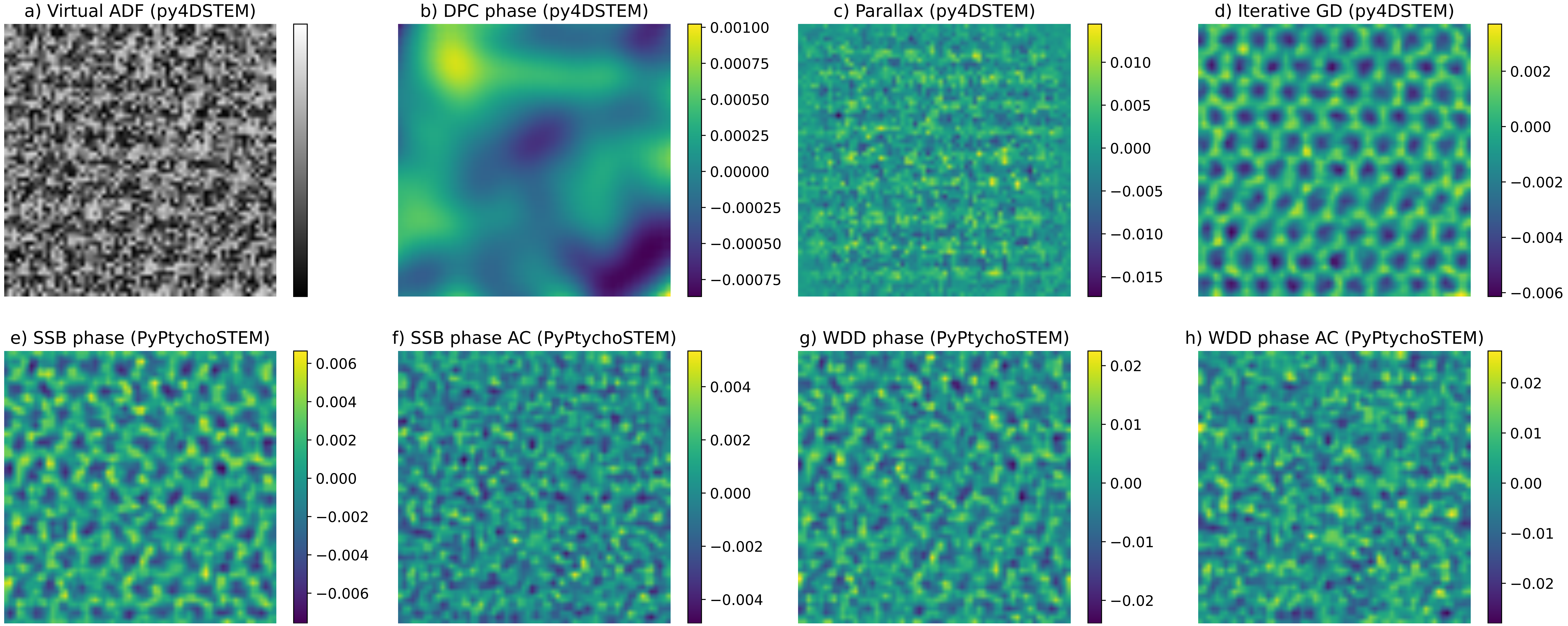


4× thinning in ***R***

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 algorithms (short camera length ***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)

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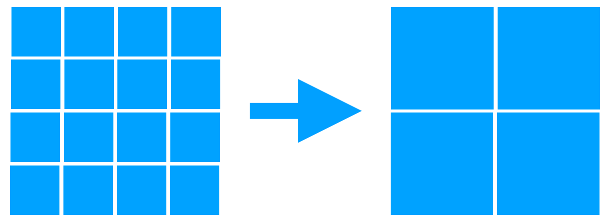
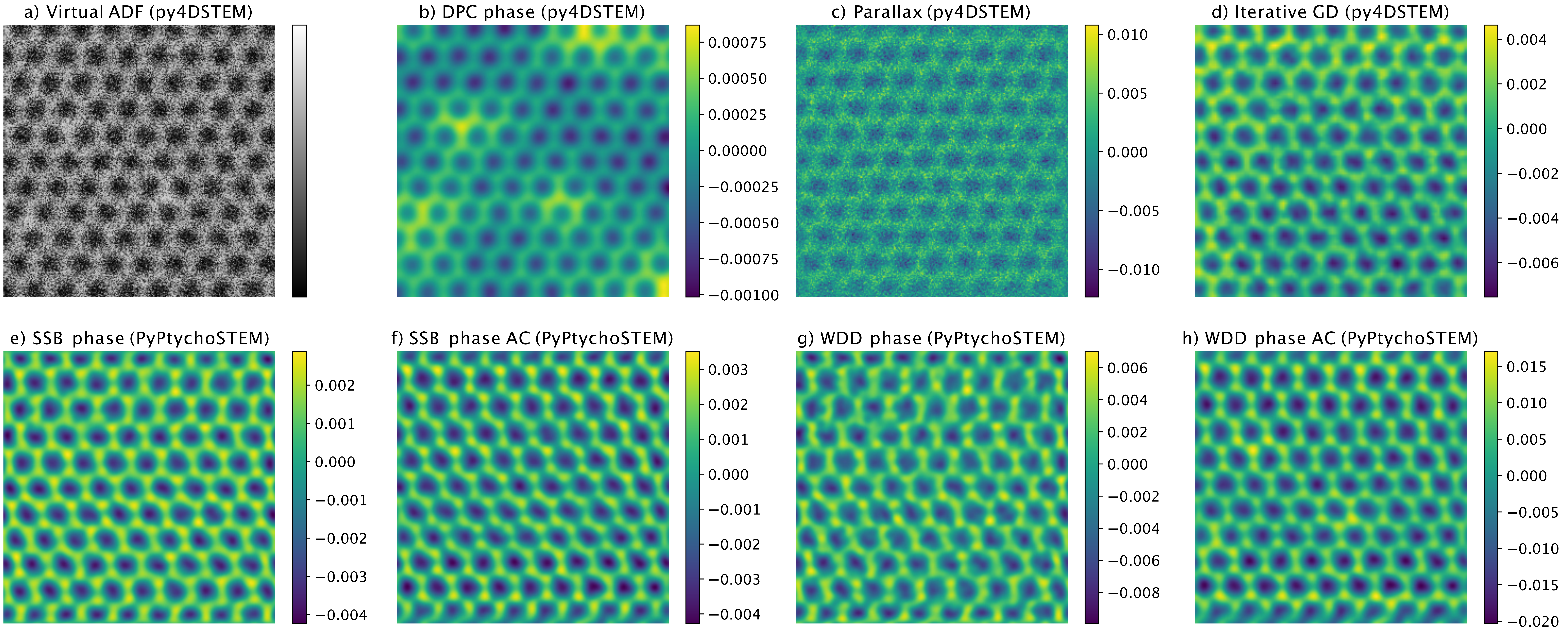
SSB & WDD: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167

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

Comparing algorithms (long camera length ***Q*** *binning*)





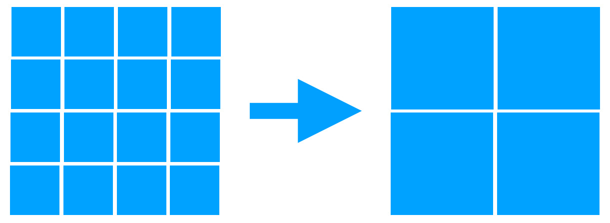
no binning in ***Q***

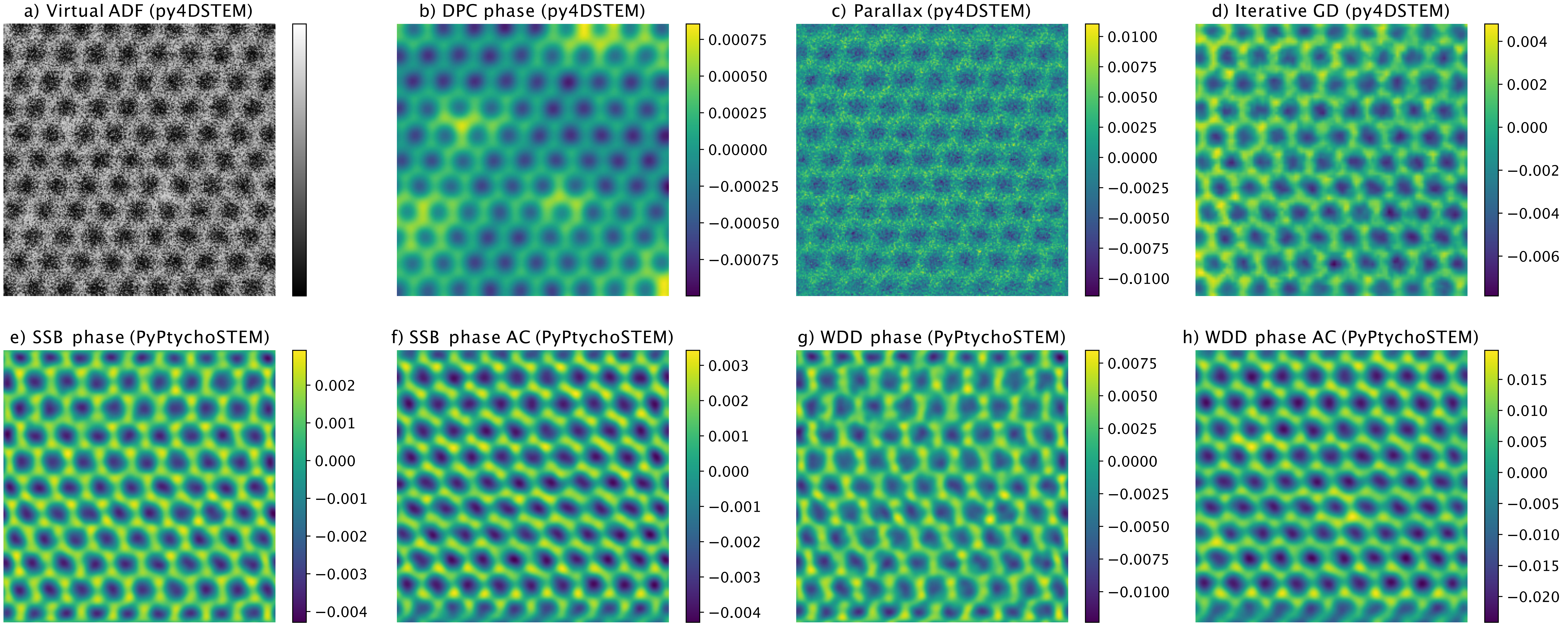
Binning in ***Q***: **109 mrad** max. scattering angle, 100 us dwell time, 256×256 ***R*** px, 192×192 ***Q*** px

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

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

Comparing algorithms (long camera length ***Q*** *binning*)





2× binning in ***Q***

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

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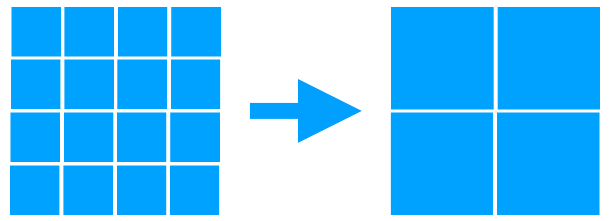
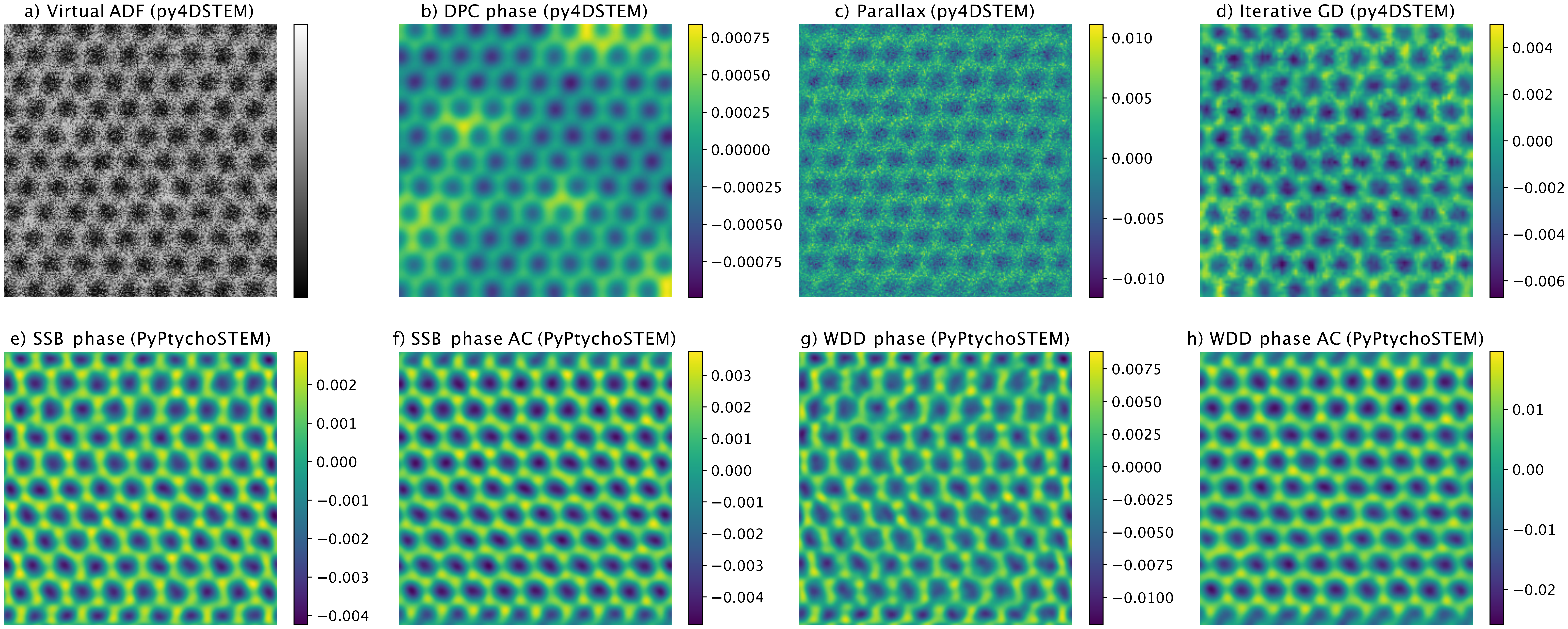
SSB & WDD: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167
DPC, Parallax, Iterative GD: G. Varnavides *et al.*, arXiv:2309.05250v2 (2024)

Comparing algorithms (long camera length ***Q*** *binning*)





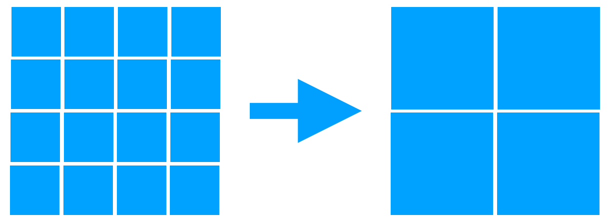
4× binning in ***Q***

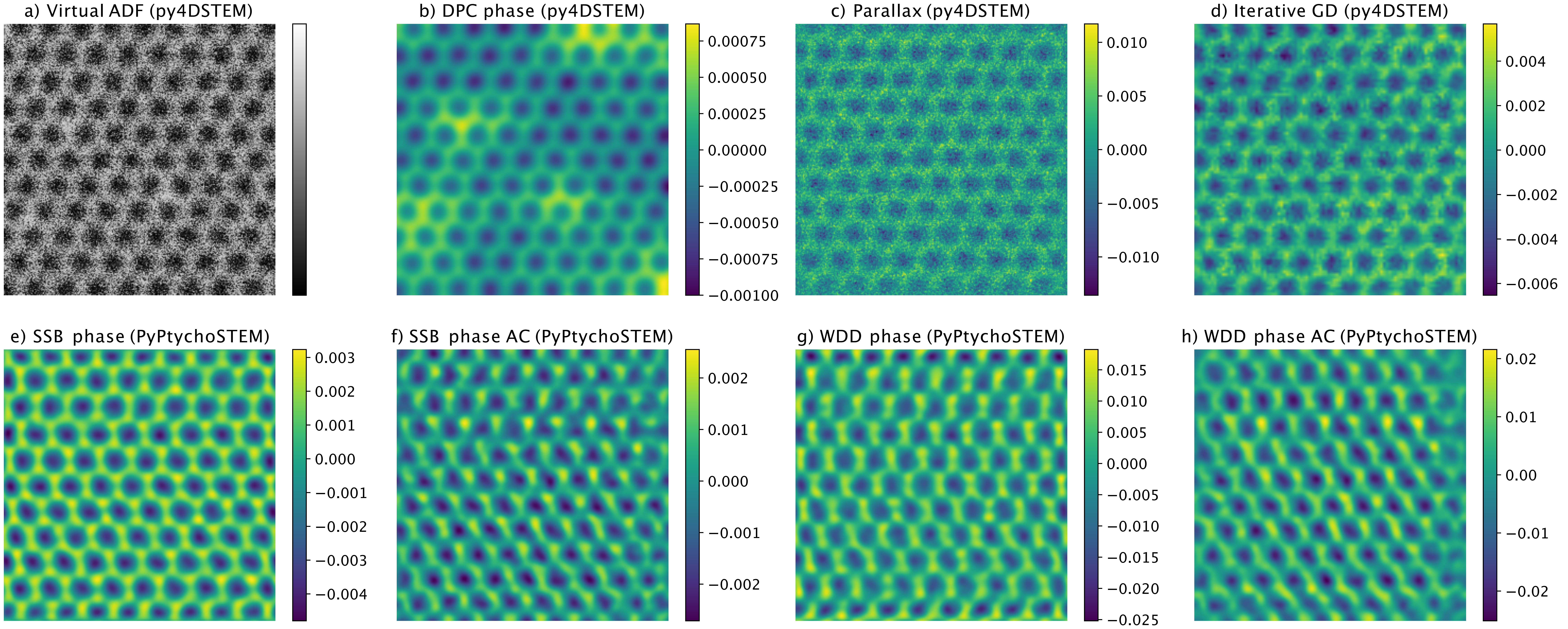
Binning in ***Q***: **109 mrad** max. scattering angle, 100 us dwell time, 256×256 ***R*** px, 192×192 ***Q*** px

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

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

Comparing algorithms (long camera length $\boldsymbol{Q}$ *binning*)





8× binning in $\boldsymbol{Q}$

Binning in $\boldsymbol{Q}$: **109 mrad** max. scattering angle, 100 us dwell time, 256×256 $\boldsymbol{R}$ px, 192×192 $\boldsymbol{Q}$ px

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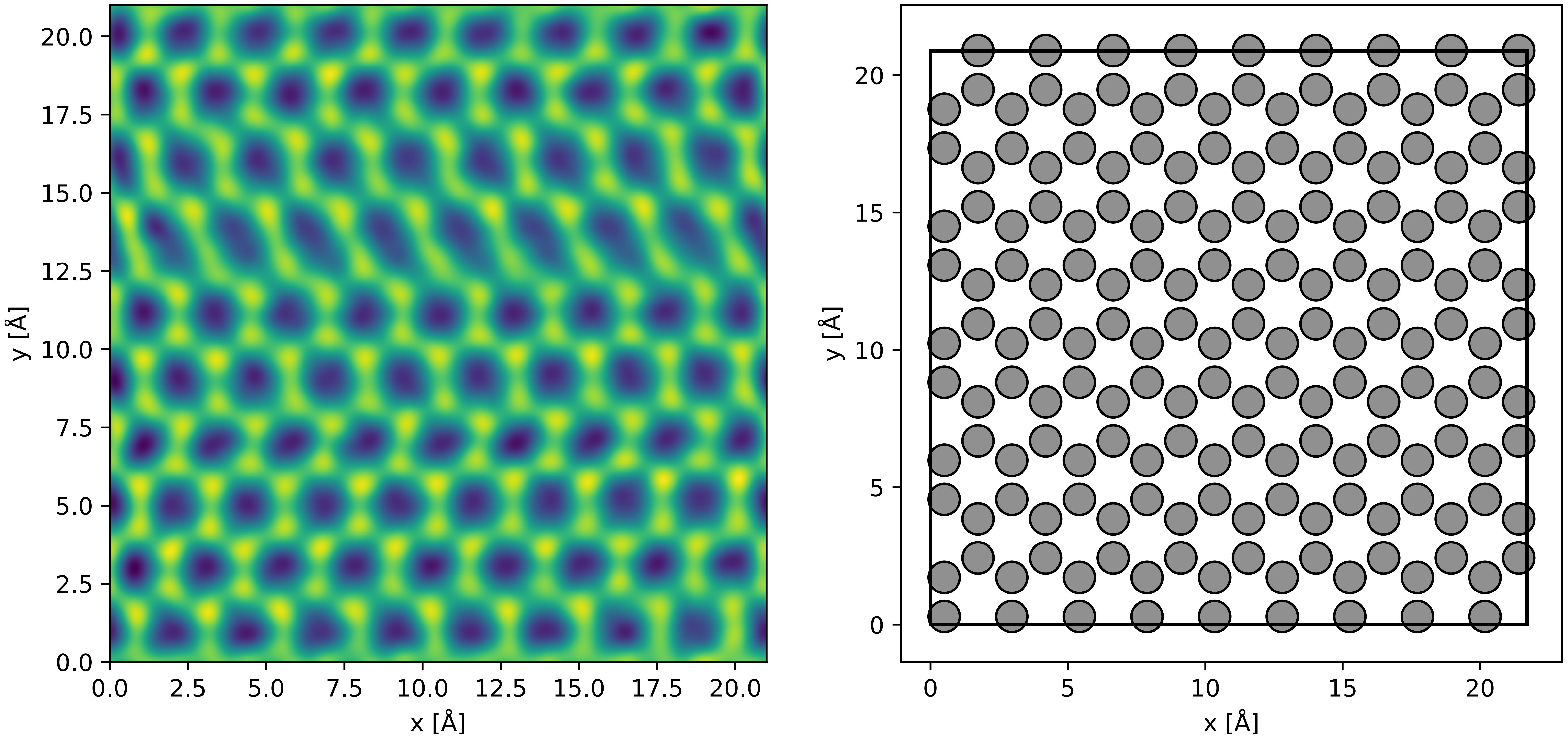
SSB & WDD: T. Pennycook *et al.*, Ultramicroscopy 151 (2015) 160–167
DPC, Parallax, Iterative GD: G. Varnavides *et al.*, arXiv:2309.05250v2 (2024)

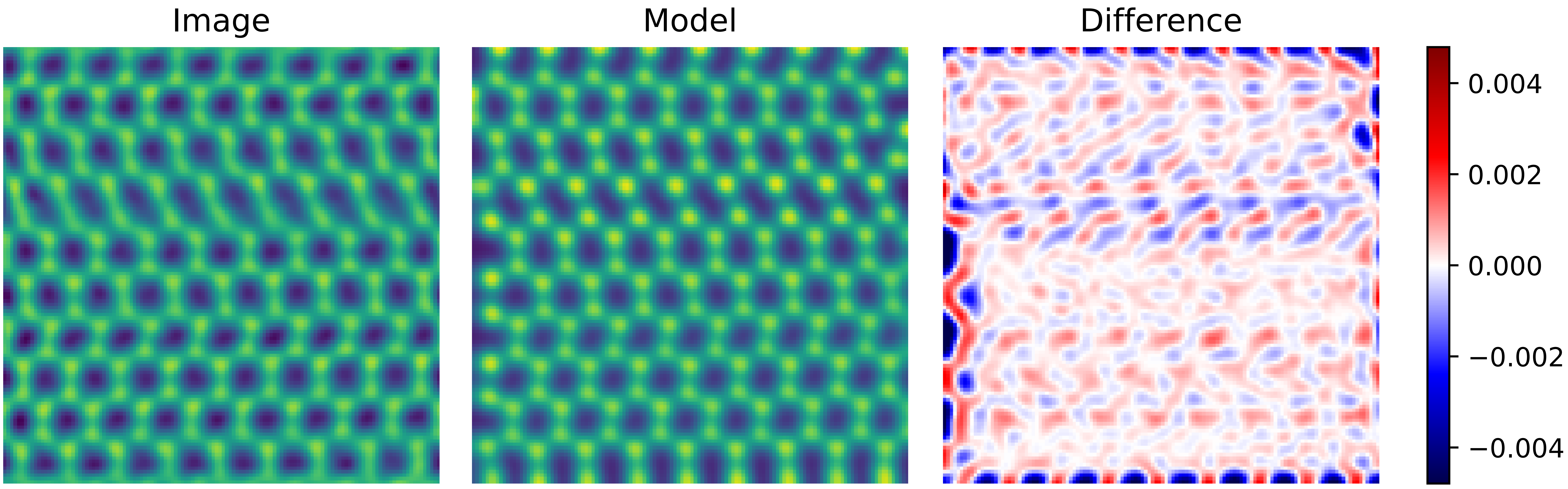
Iterative model-based phase quantification



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

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

Iterative model-based phase quantification



Can account for:

- ~~aberrations~~ *(not used here!)*
- incoherence
- scan distortions
- sample drift, tilt, etc...

Maximize correlation between experimental and simulated image

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

Note:
atoms near edges omitted

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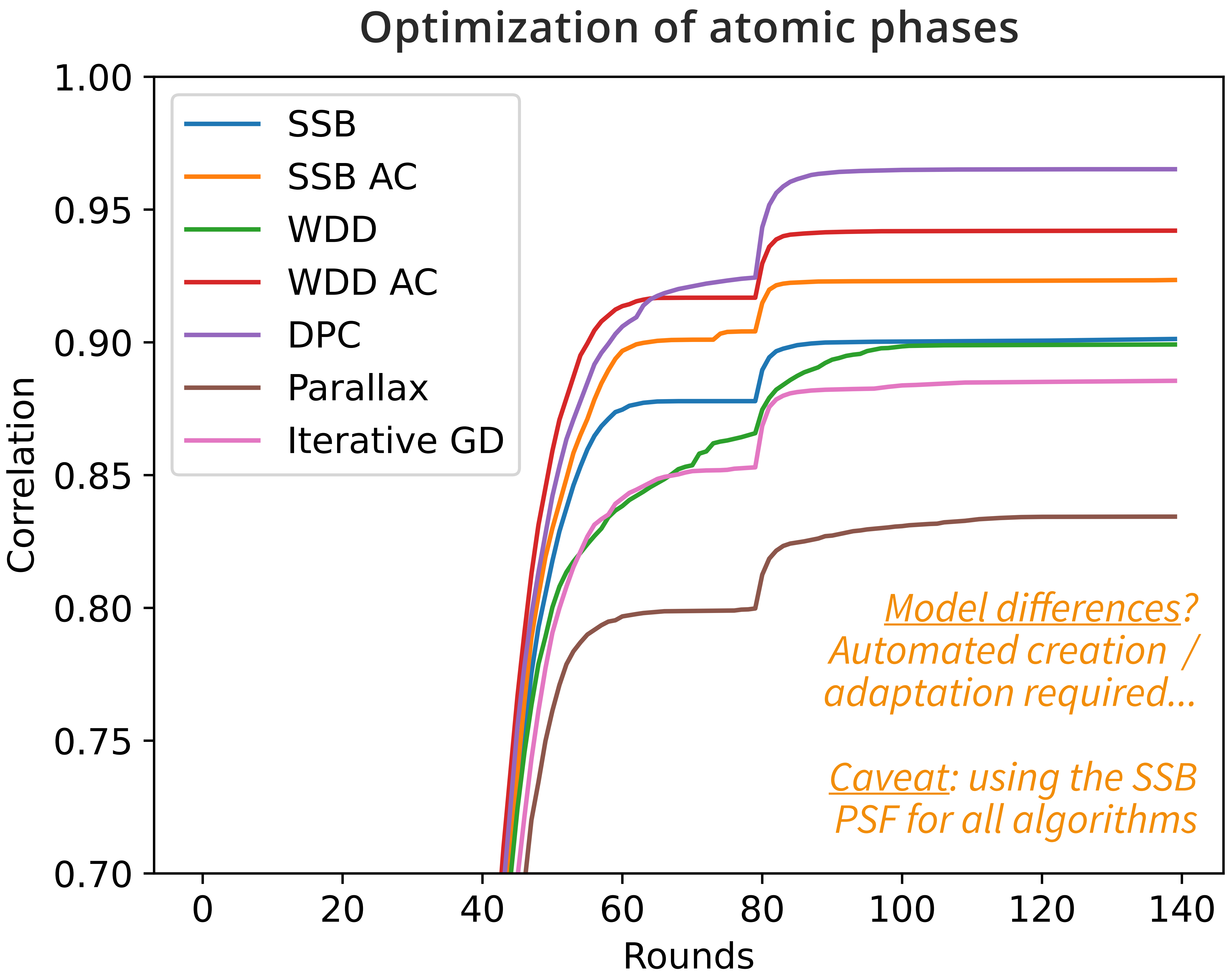
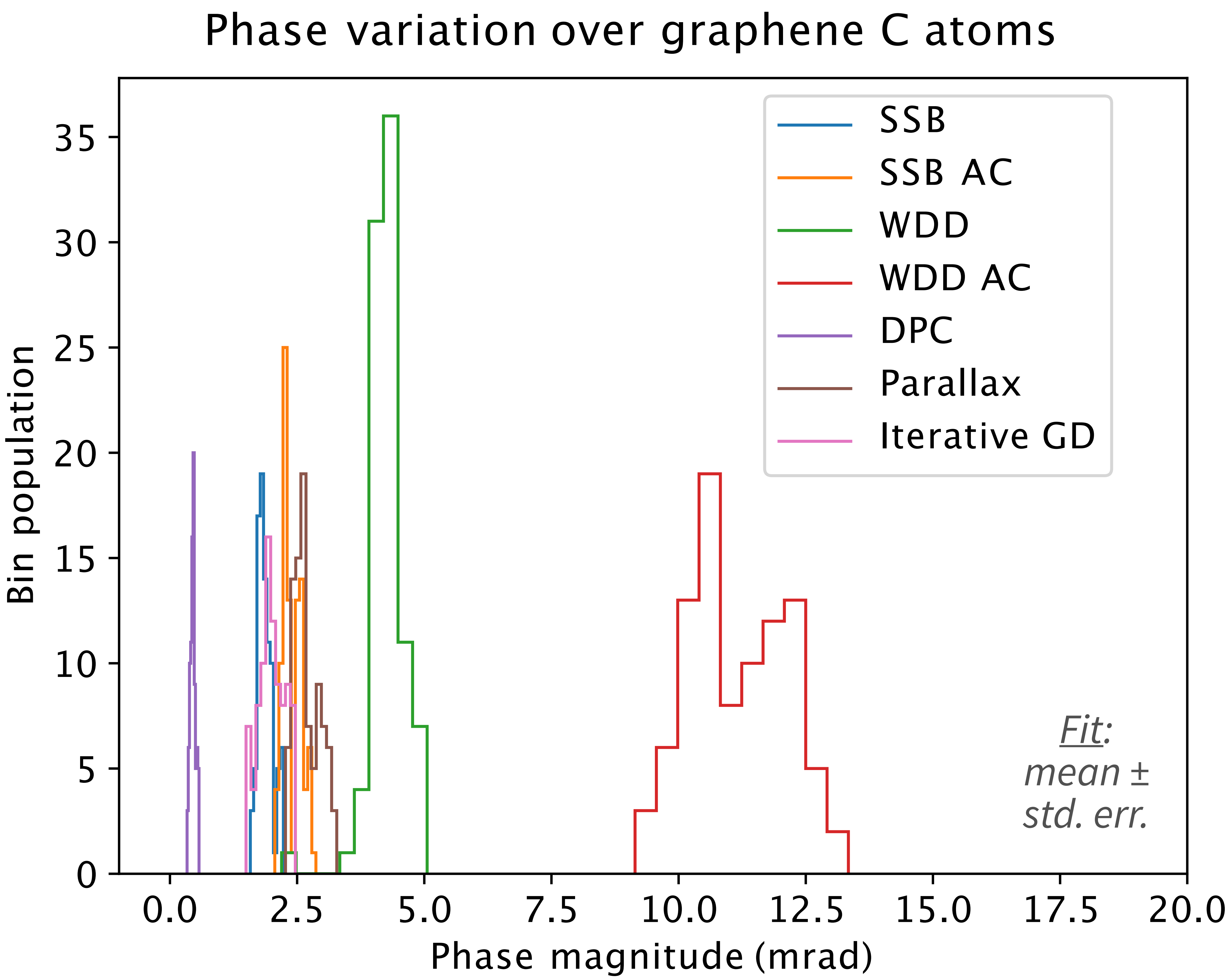
C. Hofer et al., Ultramicroscopy 227 (2021) 113292
* https://gitlab.com/christoph_hofer/stem_optimization

Atomic phase assignment

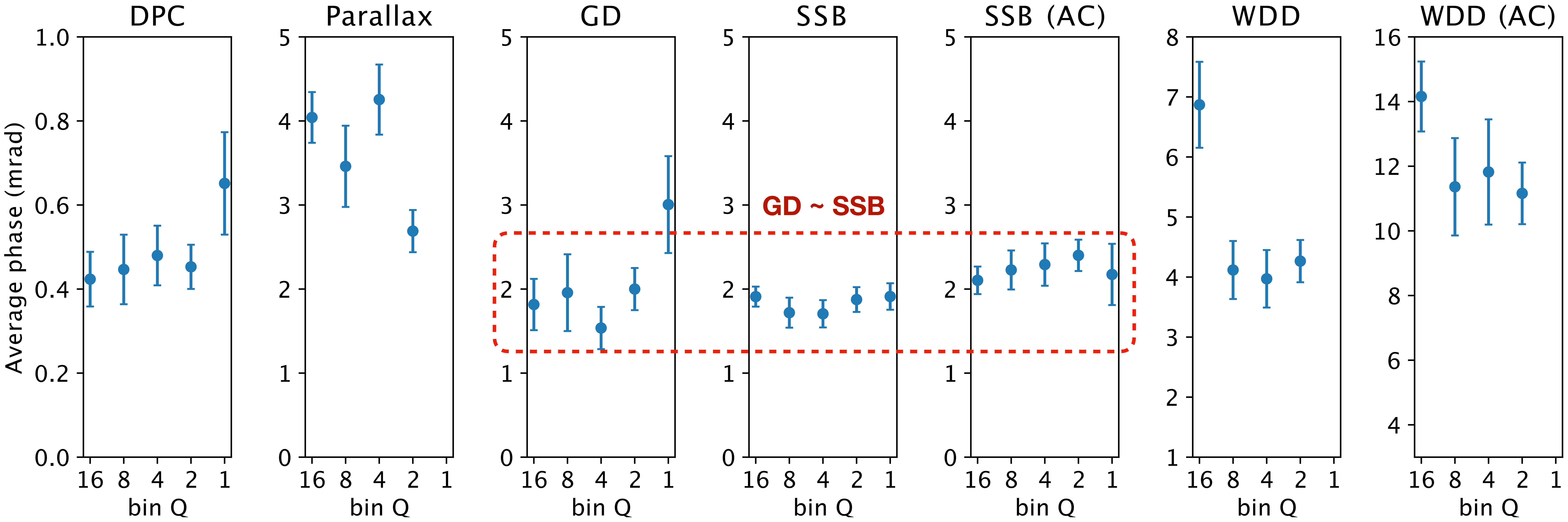




T. Susi, submitted (*Journal of Microscopy* ptychography special issue)

Quantification of phase variation

Note: different y-scales!

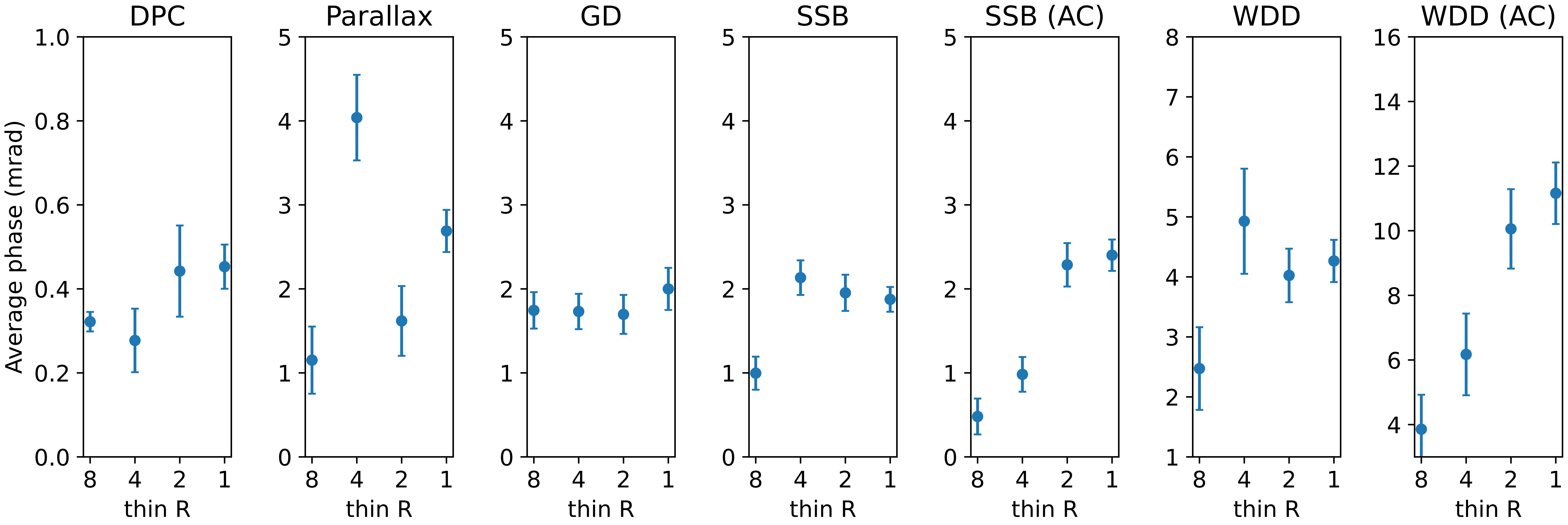


Plot: mean ± std. err.

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!

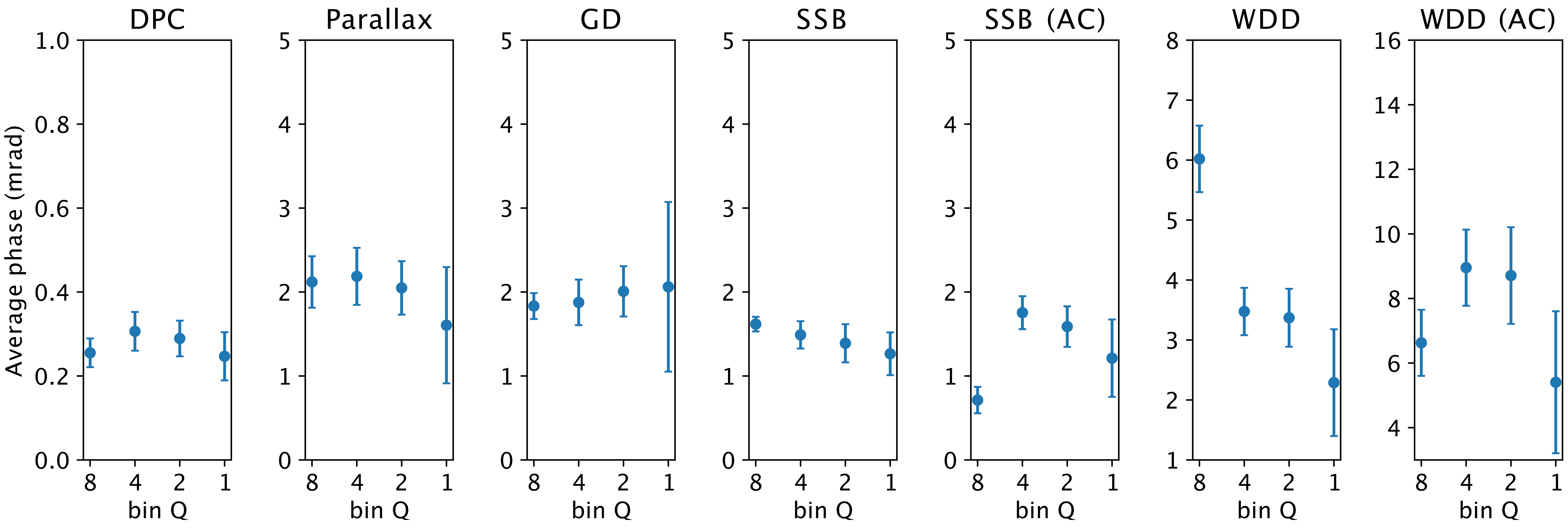


Plot: *mean ± std. err.*

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!



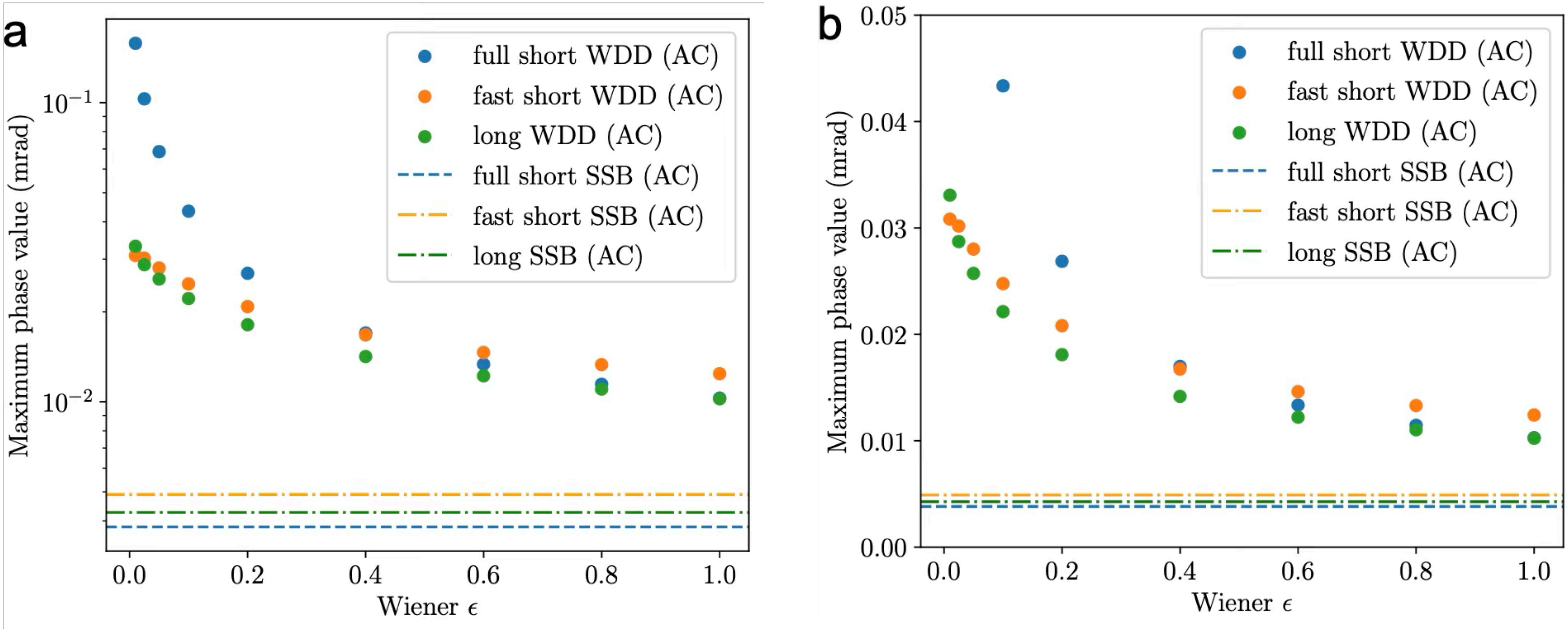
Binning in ***Q***: **109 mrad** max. scattering angle, 100 us dwell time, 256×256 ***R*** px, 192×192 ***Q*** pix

Plot: mean ± std. err.

T. Susi, submitted (*Journal of Microscopy* ptychography special issue)

Phase magnitude problem with WDD

ϵ is supposed to be a small parameter!*



* Rodenburg & Bates, Phil. Trans. R. Soc. Lond. A 339, 521-553 (1992)

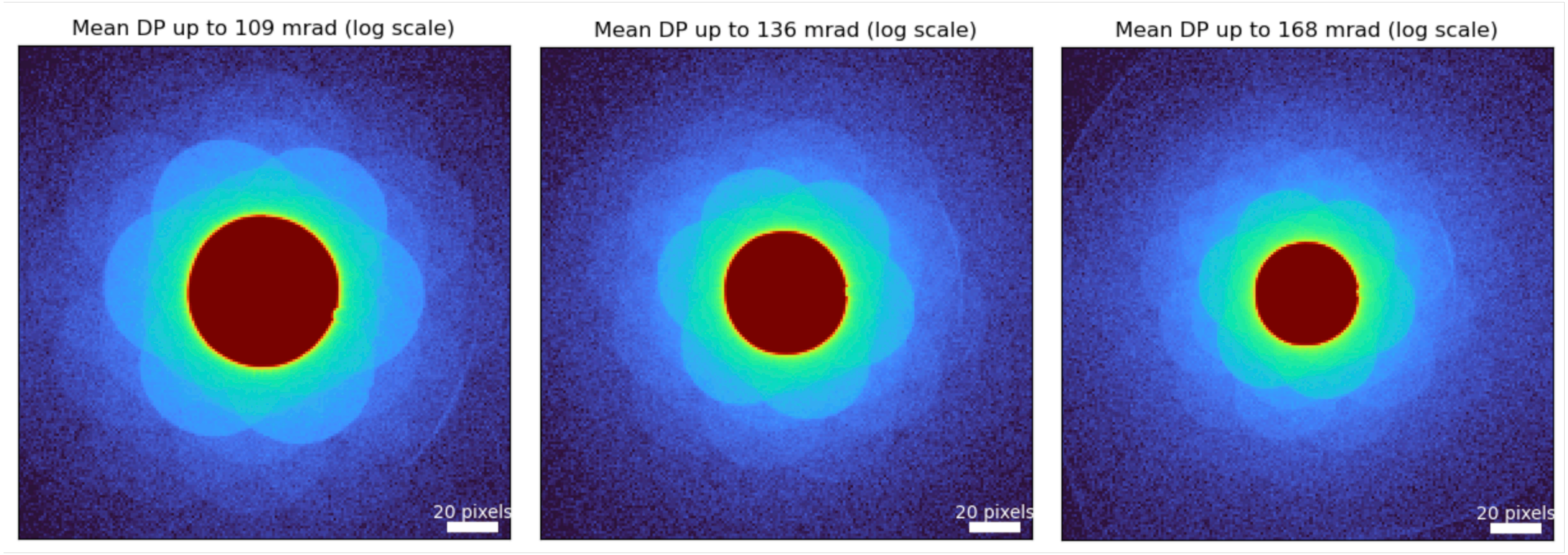
Computational cost (36 mrad datasets)

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
	32	6×6	38	4.4	22	122	7.1	35	144	147

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

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

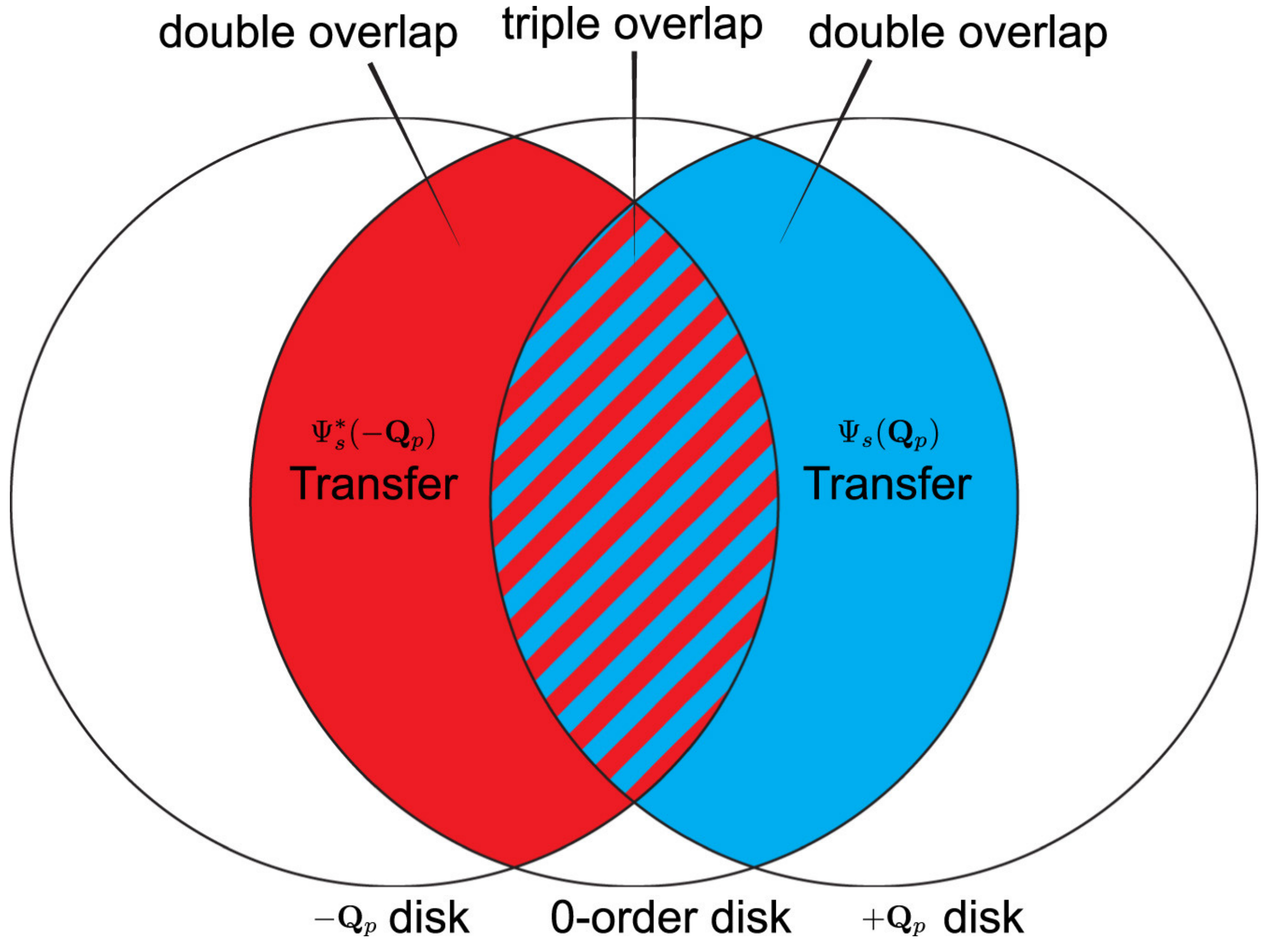
Conclusions & future prospects

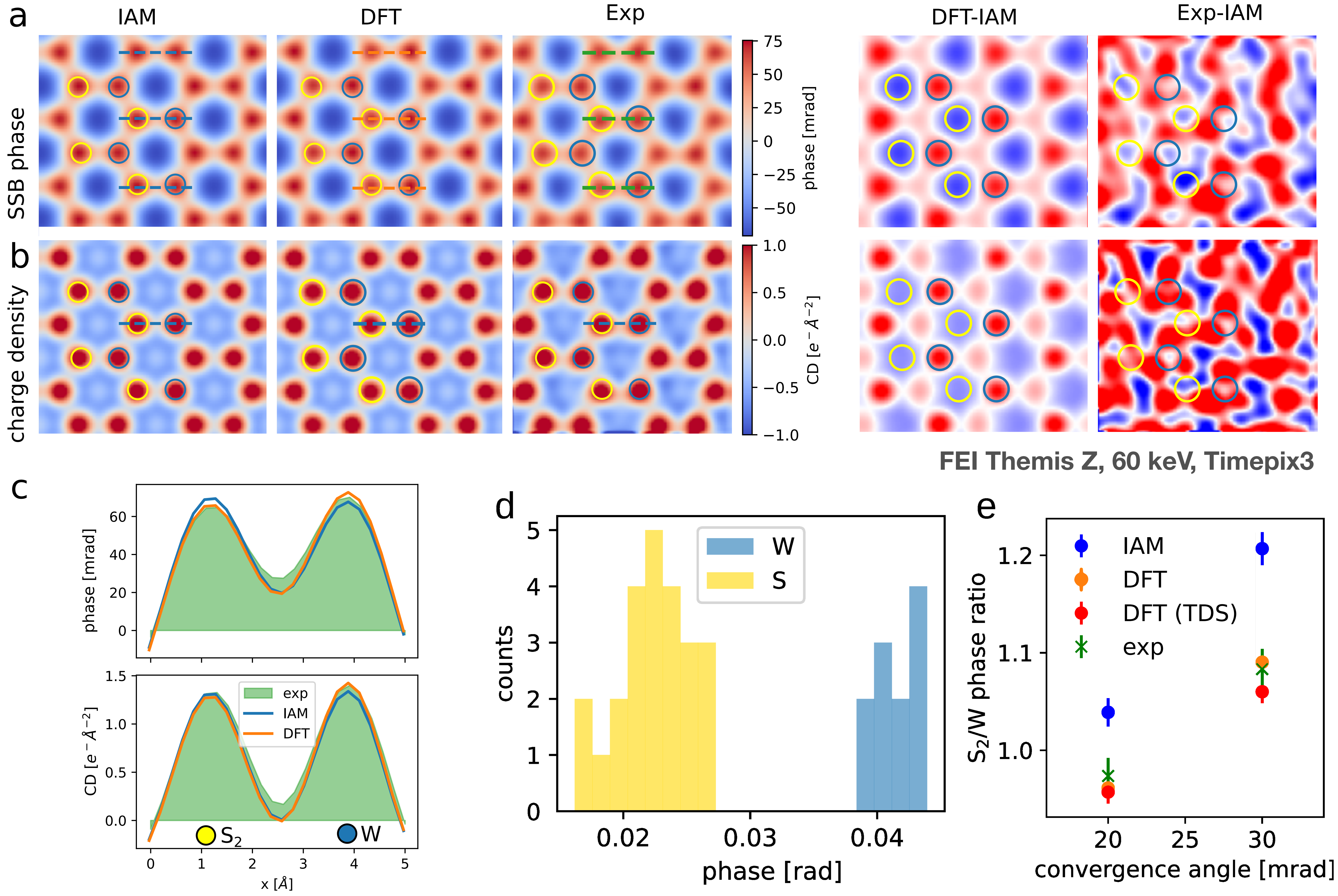


- Quantitative absolute phase values?
  - SSB and GD seem to ~agree (WDD?!)
- GD good even for focused (& fast on GPU)

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

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)

Thank you!

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

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

Open datasets
https://doi.org/10.25365/phaidra.564