Data Analysis and Machine Learning for 4DSTEM Characterization

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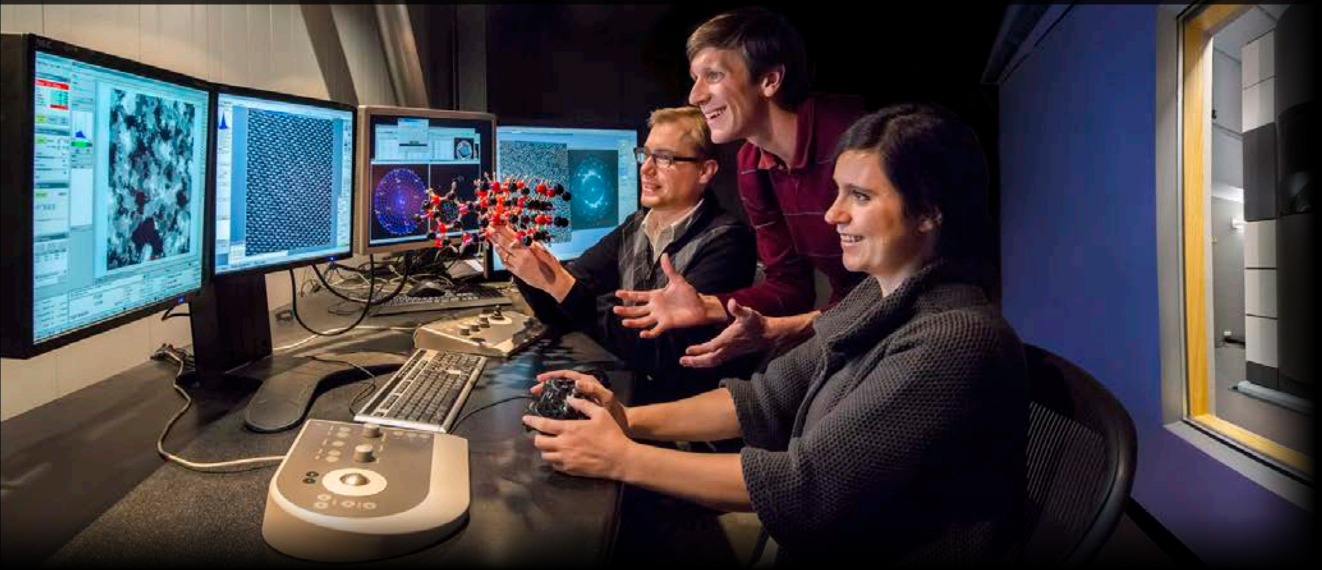
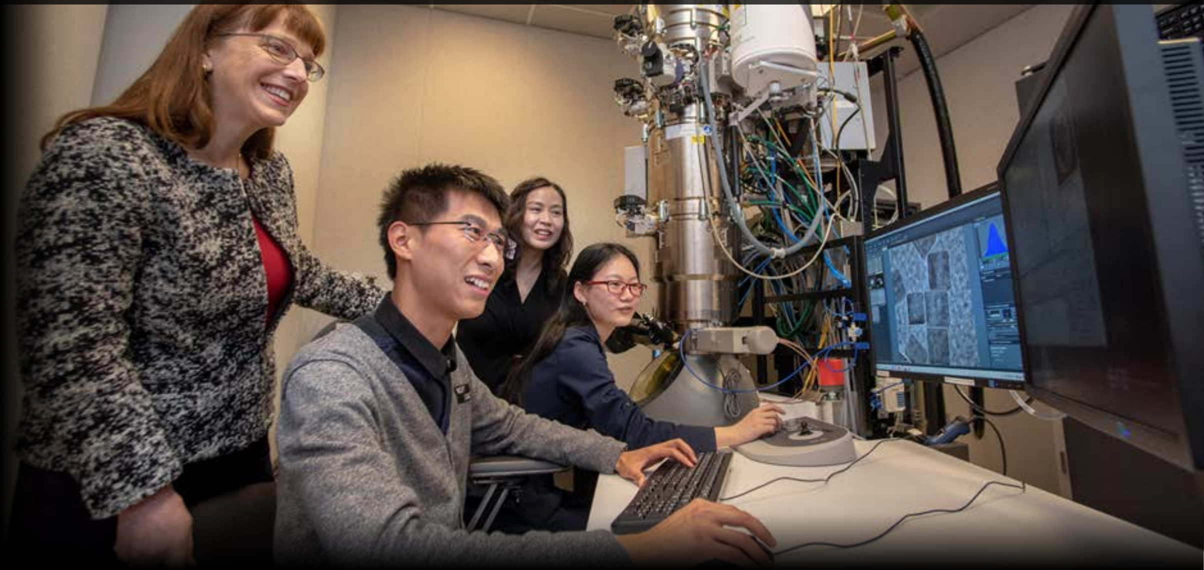
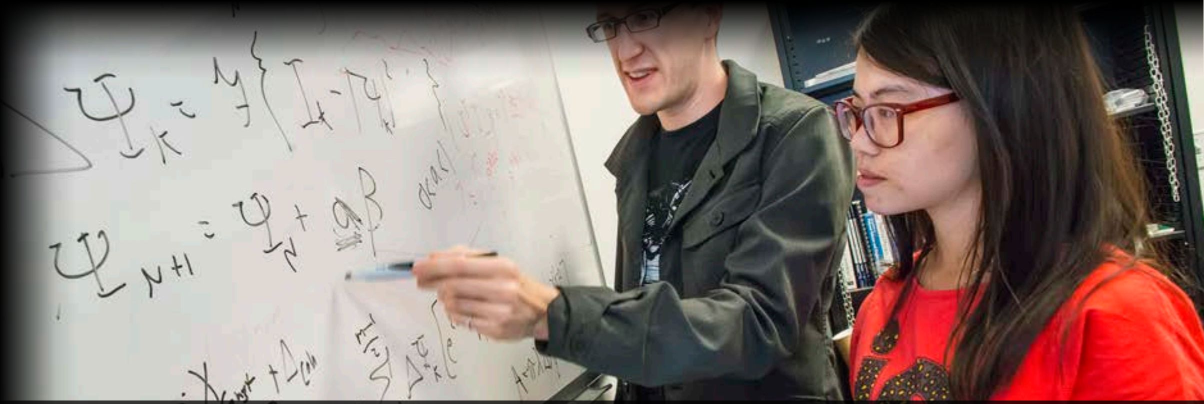
14:45 Tuesday, 18 April 2024 – Monterey Marriott in Monterey, CA
Frontiers of Characterization and Metrology for Nanoelectronics (FCMN)

Molecular Foundry User Facility – Sept 2024 – foundry.lbl.gov

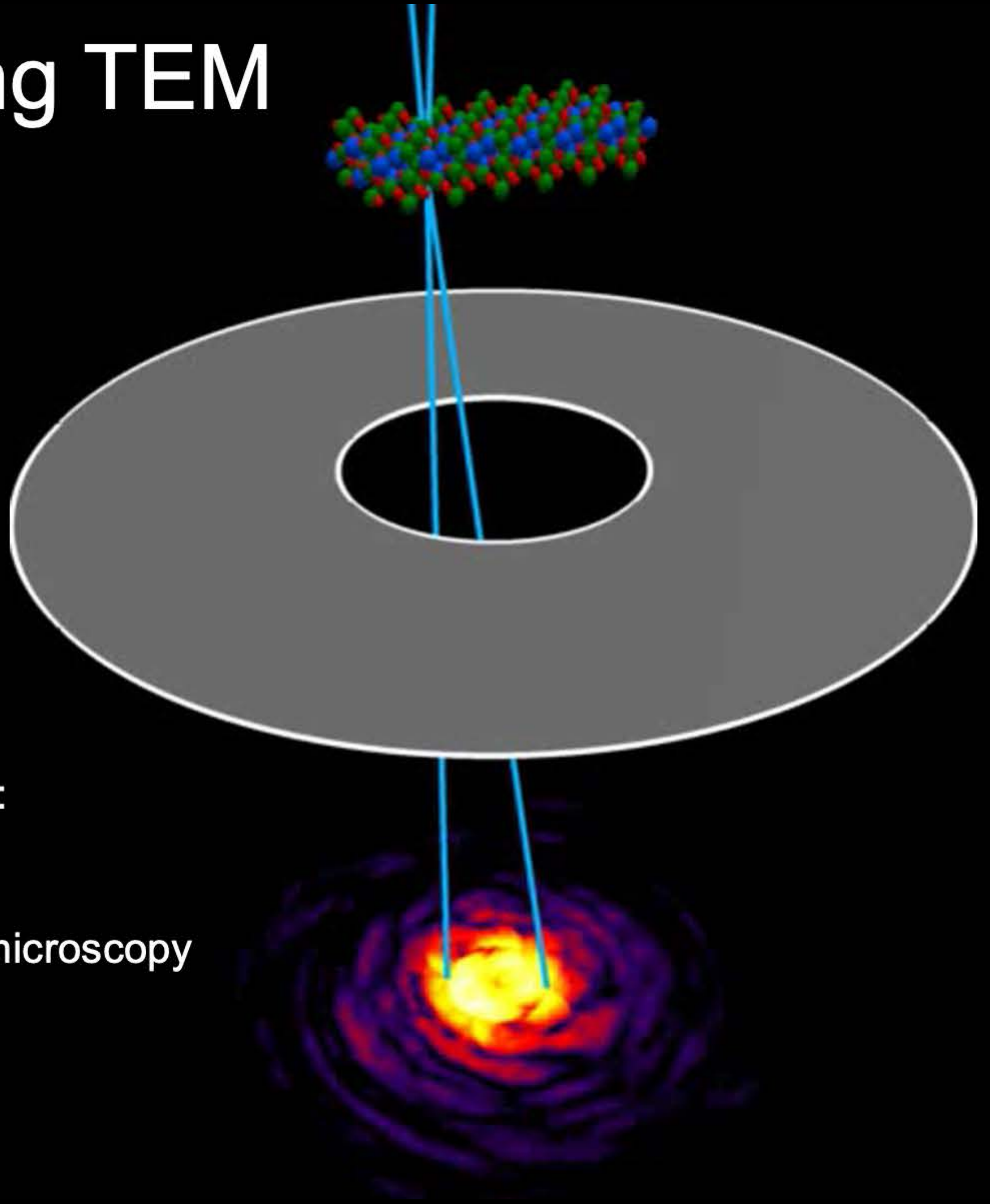






- We are a **user facility** at the Berkeley Lab, operated by the US Department of Energy.
- **Anyone** can submit a proposal (including for **computation**, **simulation** or **analysis**!).
- If accepted by independent review board, access to microscopes and staff is **free**.

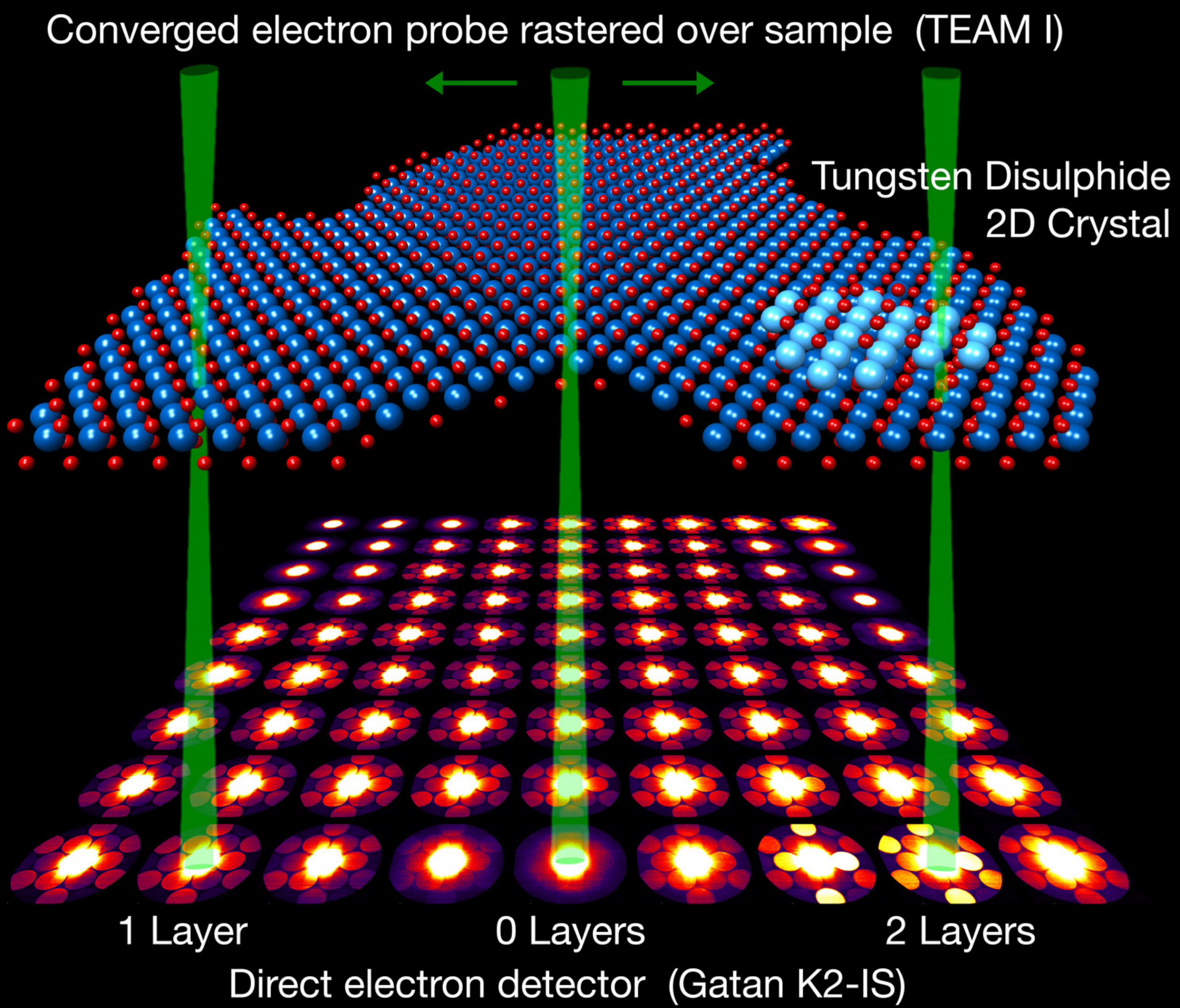
Intro to Scanning TEM



2D images recorded
over a 2D grid of probe positions:

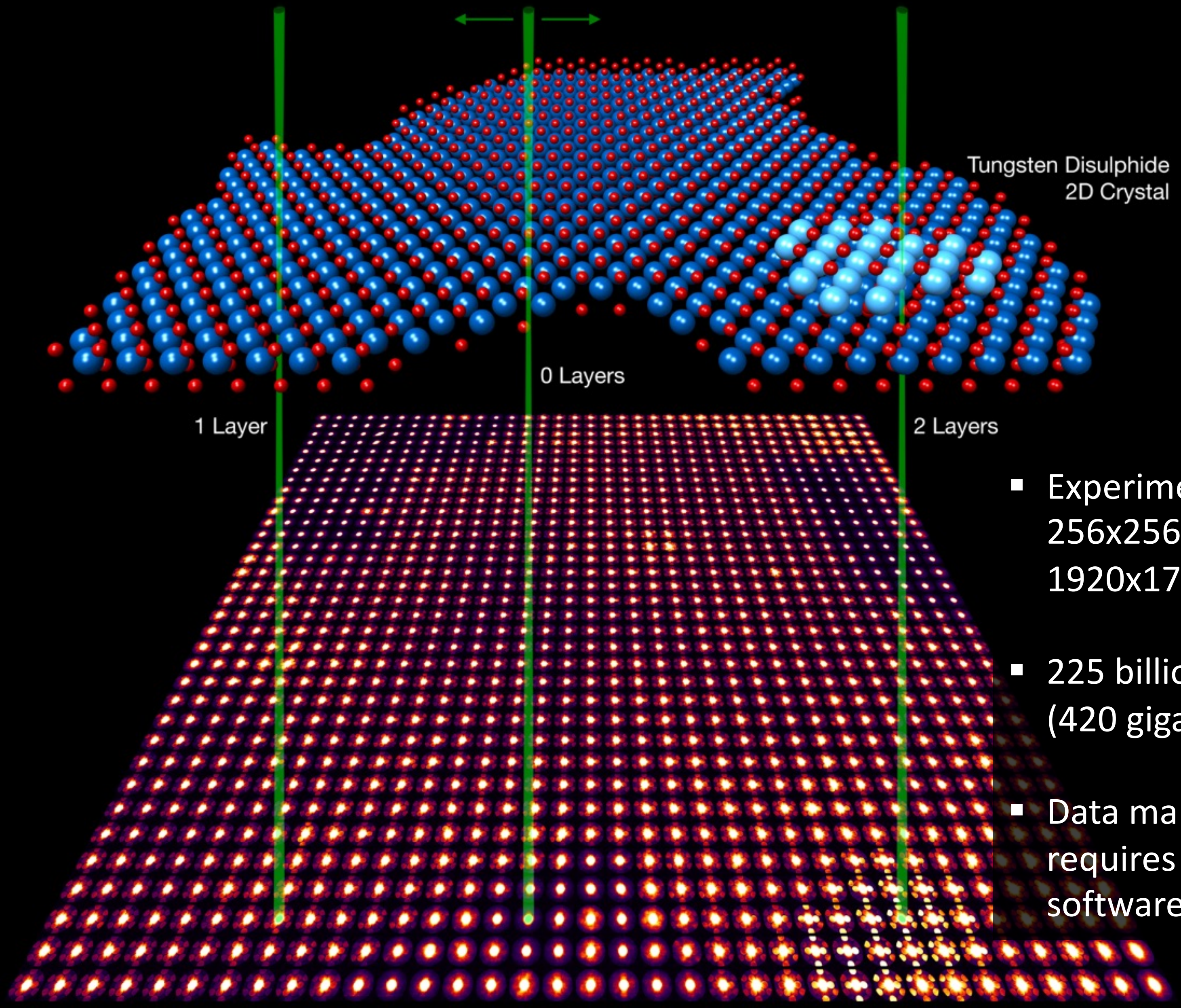
Four dimensional
scanning transmission electron microscopy
(4D-STEM)

4D STEM Experiments



Each image is
40 x 40 positions
averaged …

4D STEM Experiments

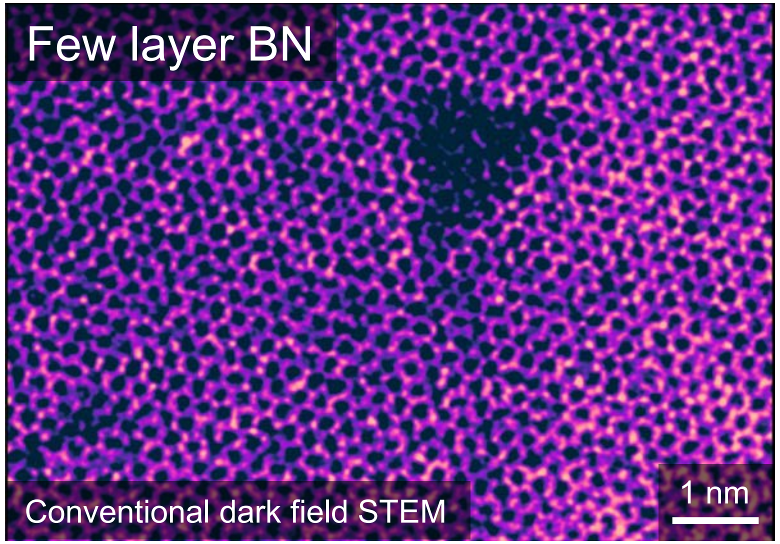


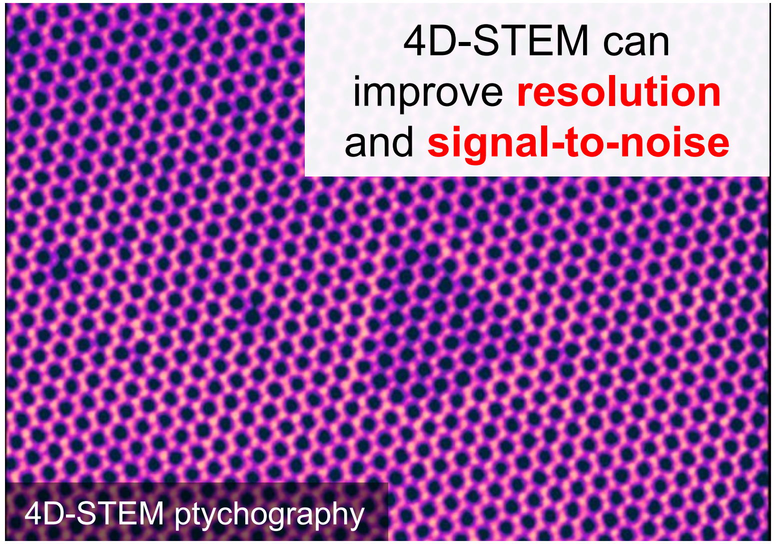
Each image is
7 x 7 positions
averaged …

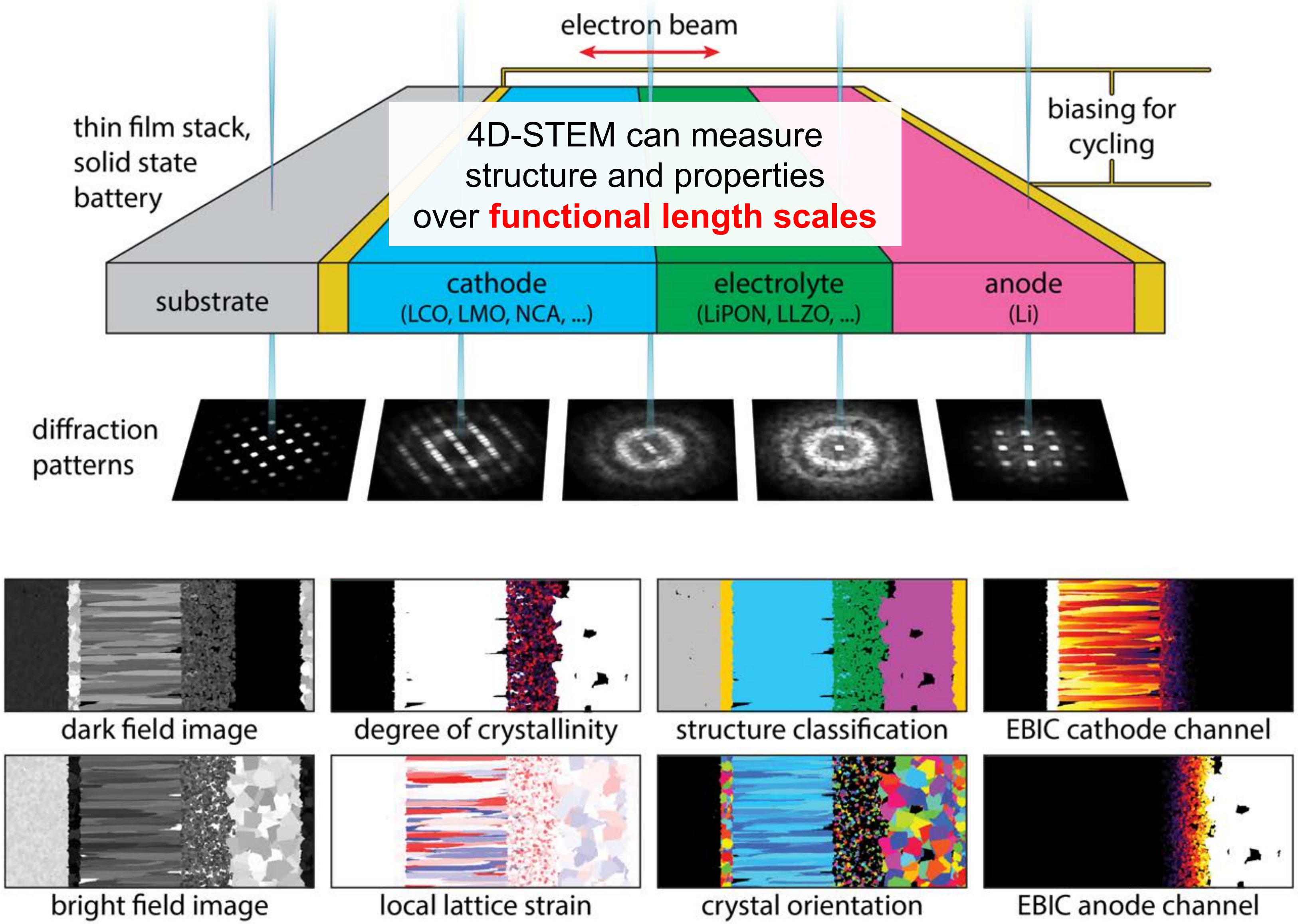
- Experimental K2 IS dataset, 256x256 probe positions, 1920x1792 CBED image sizes.
- 225 billion pixels (420 gigabytes) in ≈3 minutes.
- Data manipulation / analysis requires fast and robust software methods.

MOLECULAR FOUNDRY

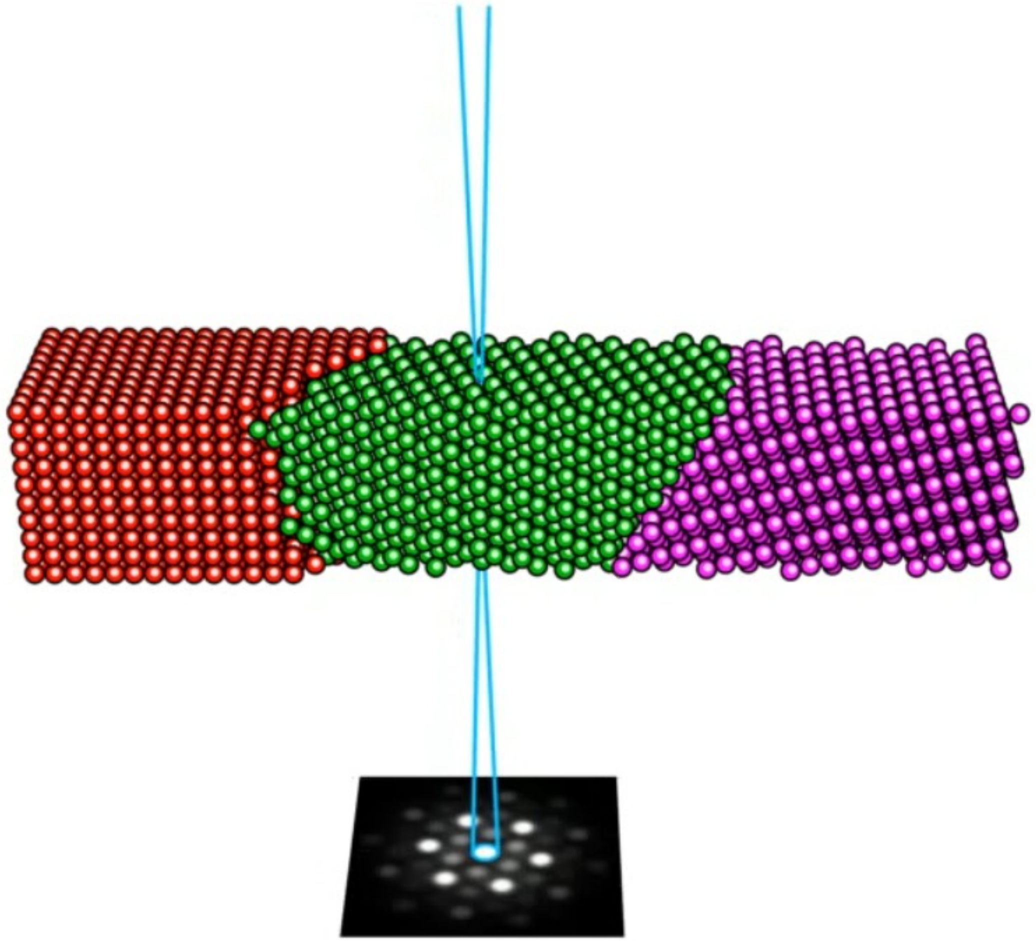
Motivation – Why do 4D-STEM?





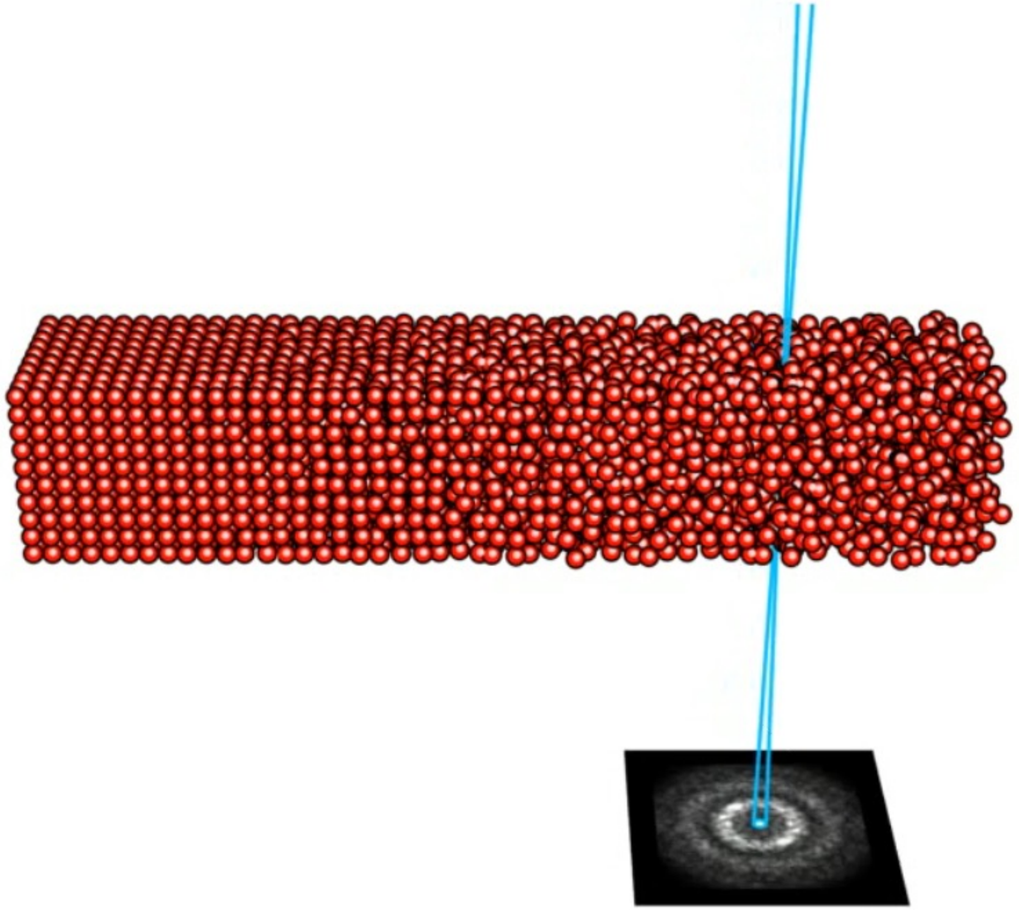


STEM Diffraction from Crystalline Samples



- Ideally, the diffracted signal is simply a 2D **Fourier transform** of the projected potential, multiplied by the probe intensity.
- Thus the position and intensity of **Bragg disks** of each diffraction pattern acts as a **fingerprint** for the local structure and orientation of the (crystal) sample.
- Interpretation is complicated by multiple / dynamical scattering (thickness effects), overlapping grains, background signals.

4D-STEM Diffraction from Amorphous Samples

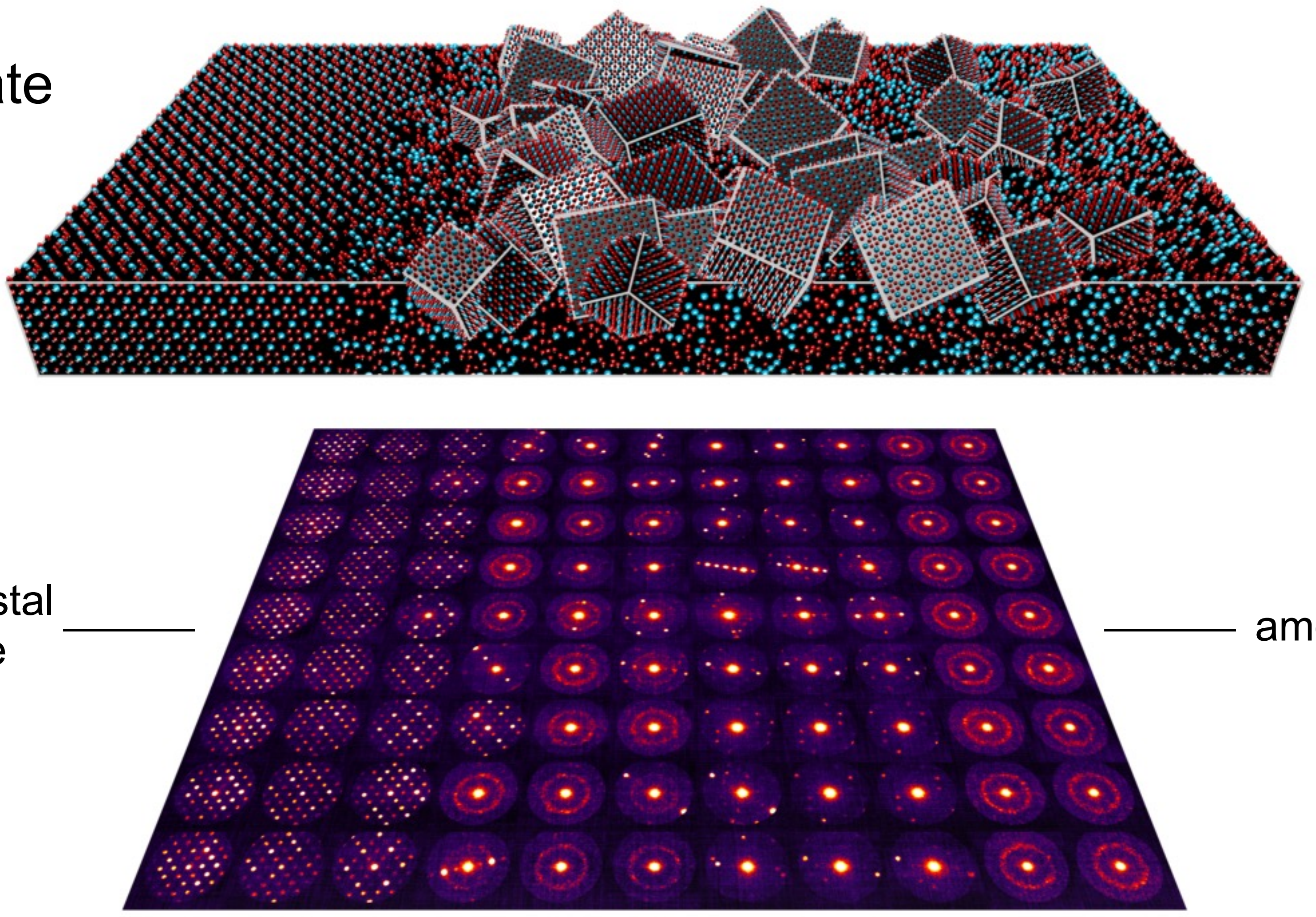


- Ideally, the diffracted signal is simply a 2D **Fourier transform** of the projected potential, multiplied by the probe intensity.

- The position and shape of **amorphous halos** of each diffraction pattern acts as a **fingerprint** for the local structure factor, given by the mean atomic arrangement.

- Interpretation is **complicated** by multiple / dynamical scattering (thickness effects), background, **more** than crystal diffraction!

Complex Sample Analysis with 4D-STEM



B Savitzky et al., Microscopy and Microanalysis (2021).

4D-STEM Analysis

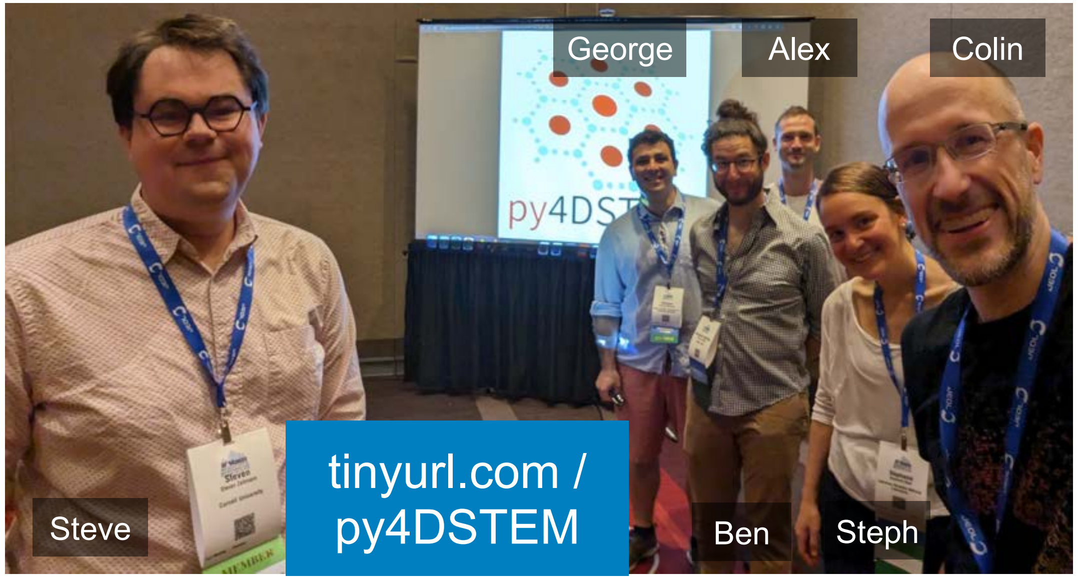
py4DSTEM

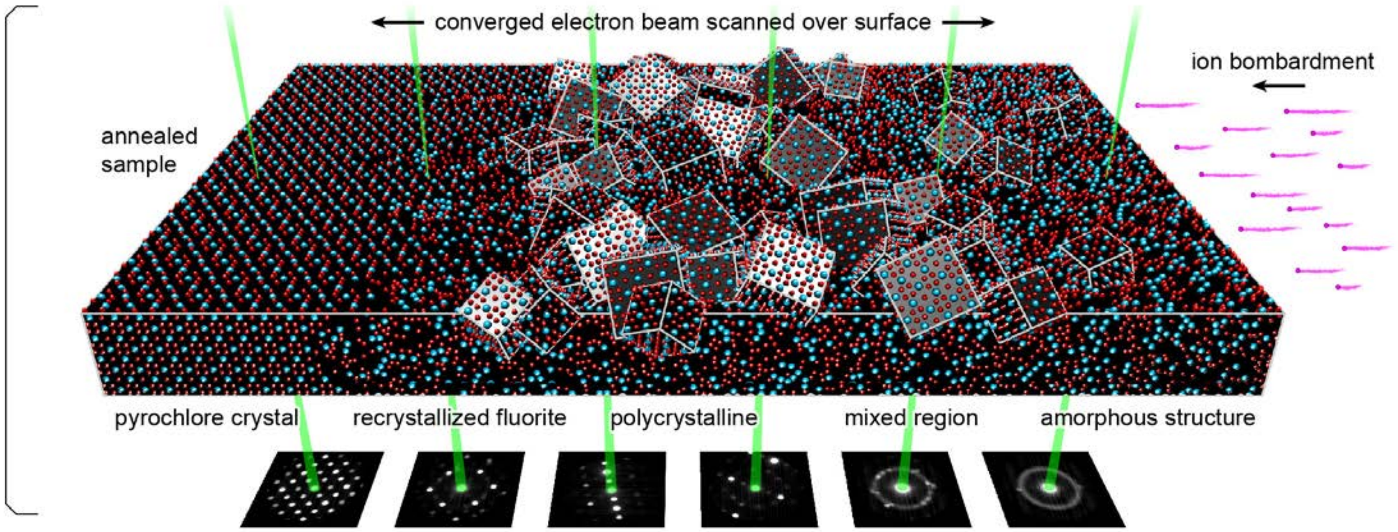
Created by Benjamin Savitzky & Colin Ophus.

Primarily funded by TOYOTA RESEARCH INSTITUTE

Core developer team also includes Steve Zeltmann, Steph Ribet, Alex Rakowski, and George Varnavides.

Teaching Workshop at M&M

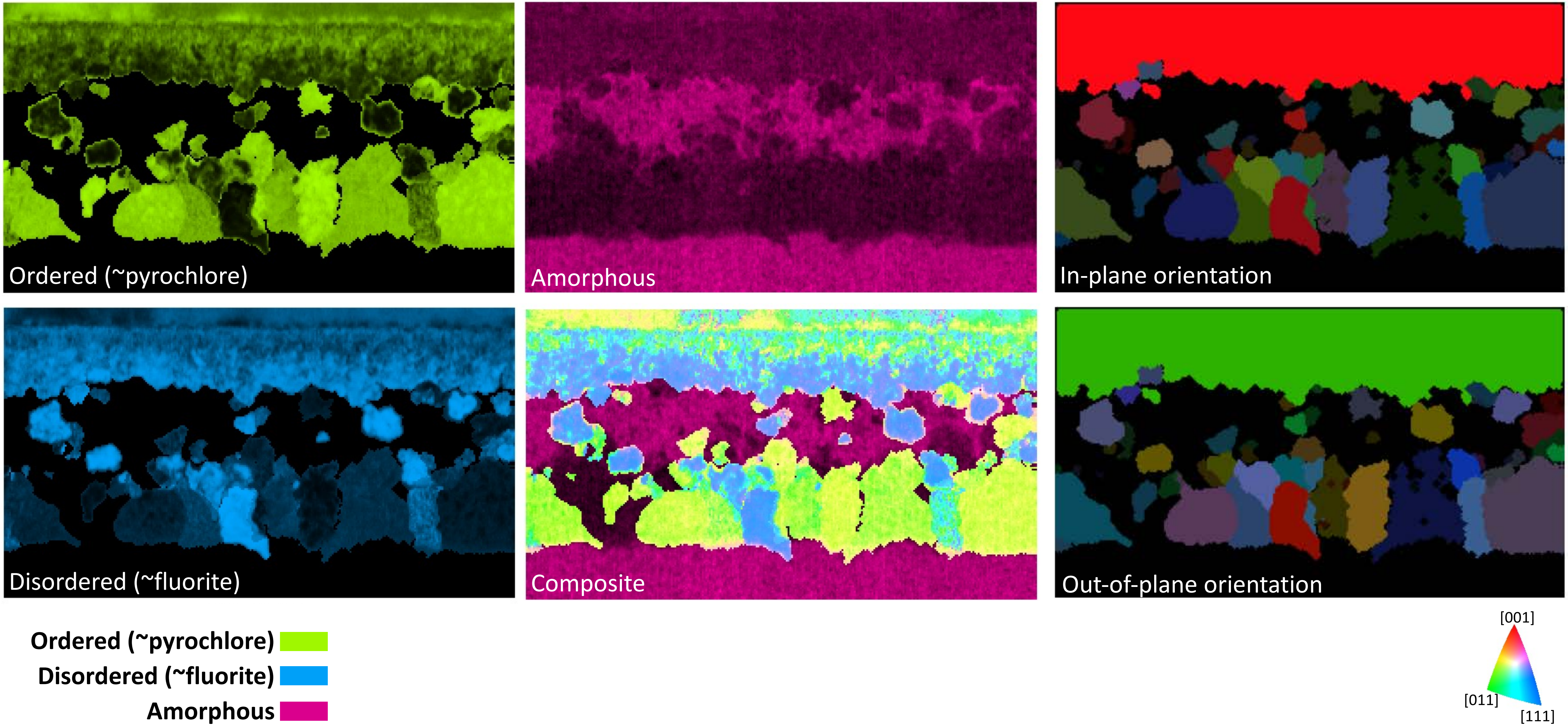




py4DSTEM can measure:

- Virtual imaging (bright field and dark field).
- Structure classification.
- Phase, orientation and strain mapping of crystal materials.
- Short range ordering (SRO) & FEM of amorphous materials.
- Phase contrast imaging methods:
  - Differential phase contrast
  - Parallax imaging
  - Ptychography
  - Ptychographic atomic tomography

4D-STEM – Irradiated & Annealed $Gd_2Ti_2O_7$

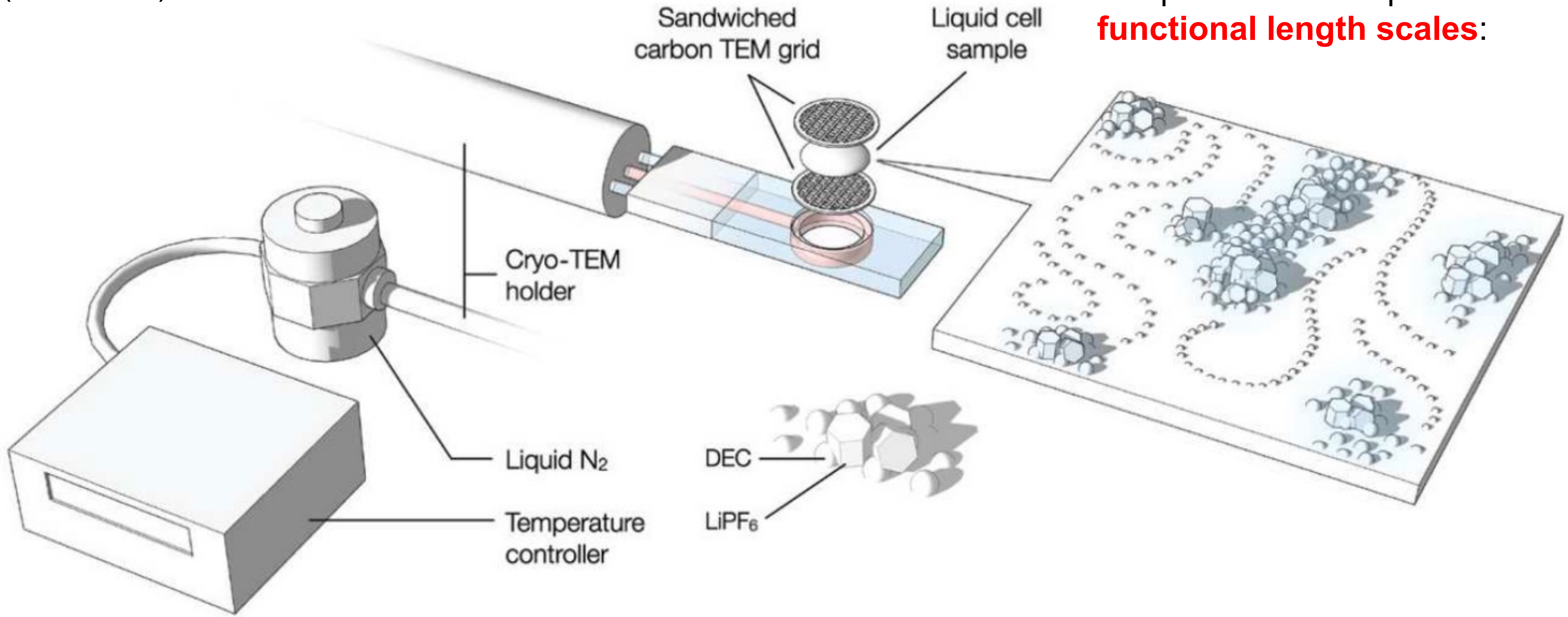


B Savitzky et al., *manuscript in preparation*

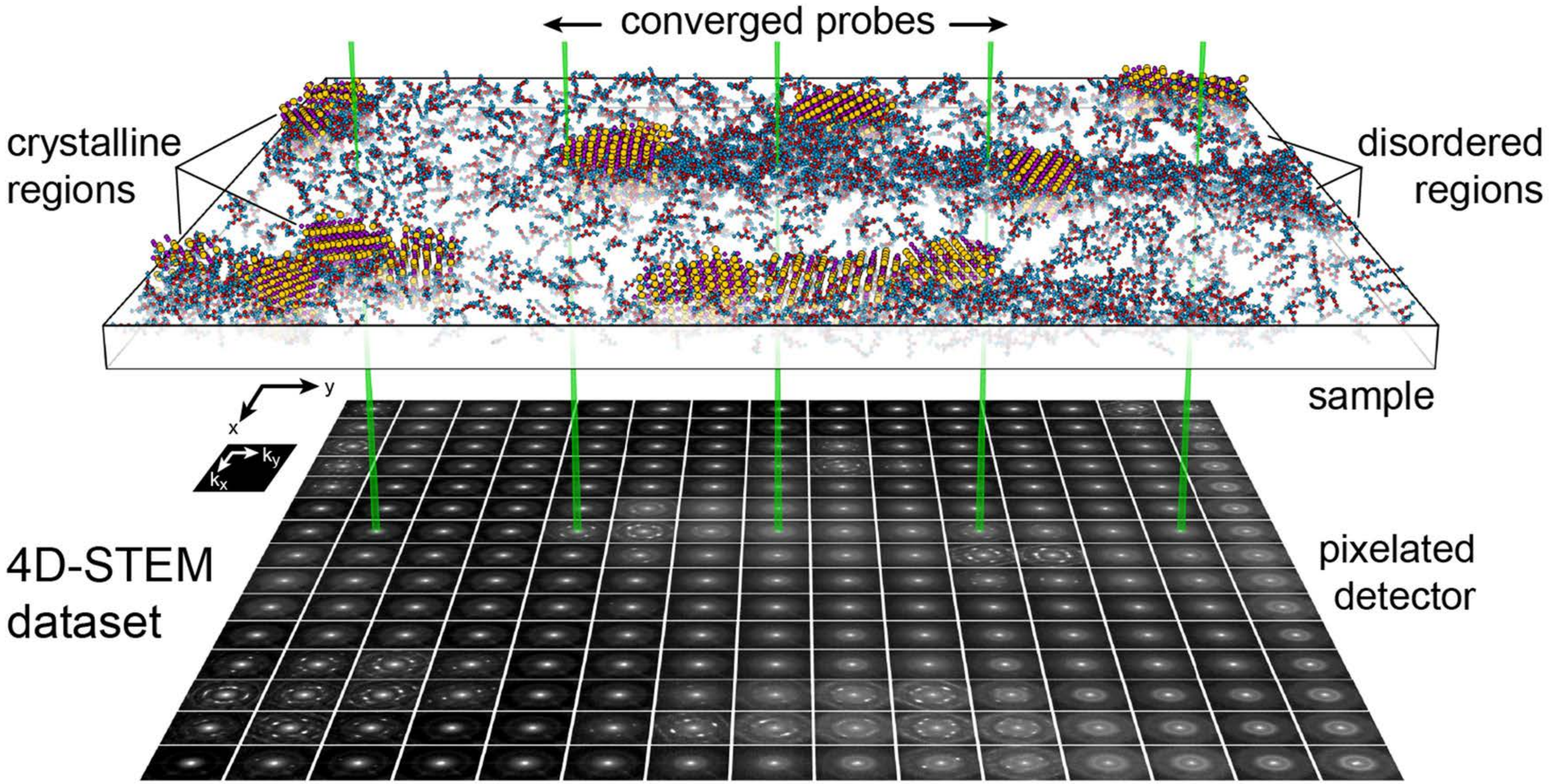
Characterization of Battery Electrolyte @ -30ºC

ethylene carbonate / diethyl carbonate
(**EC** / **DEC**)

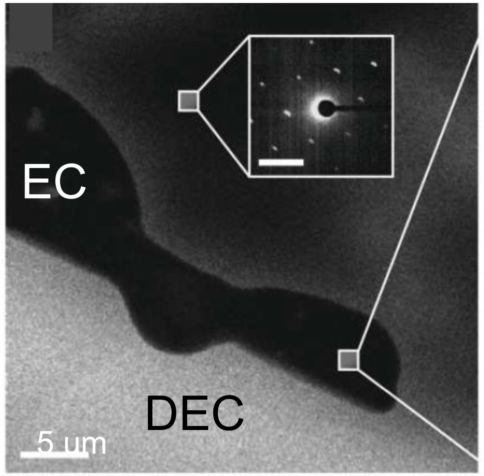
complex mixture of phases on **functional length scales**:

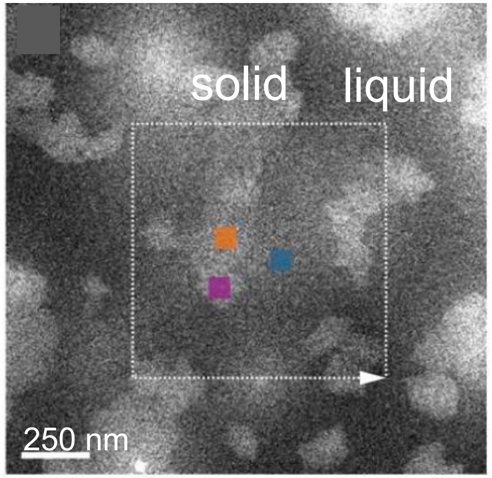


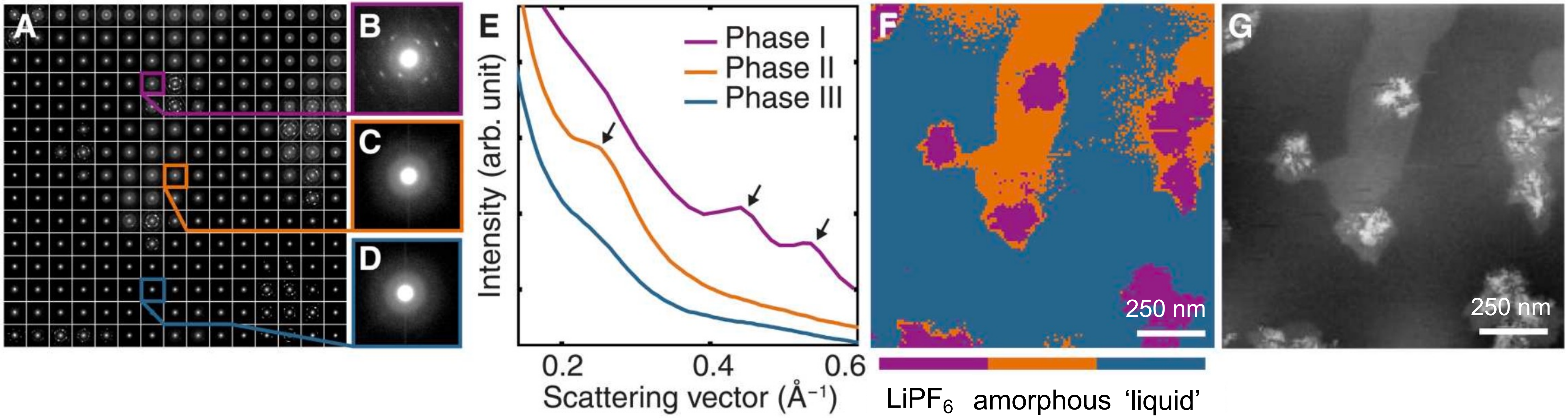
4D-STEM Characterization of Battery Electrolyte



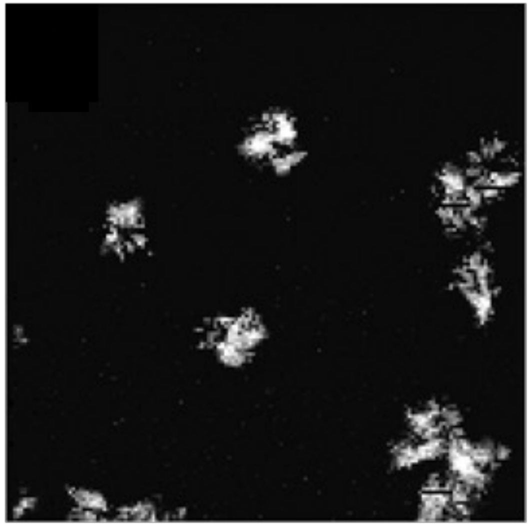
4D-STEM Characterization of Battery Electrolyte

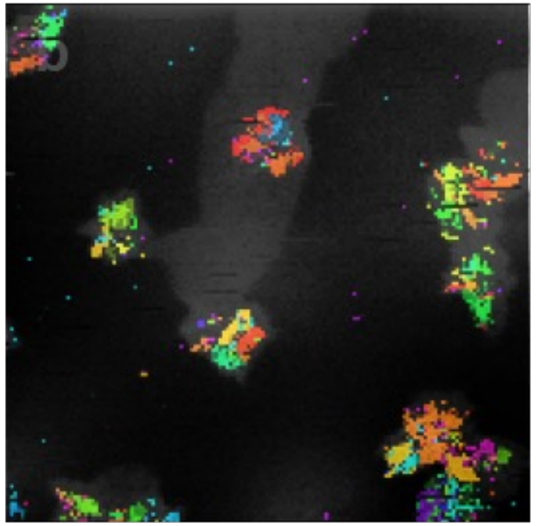


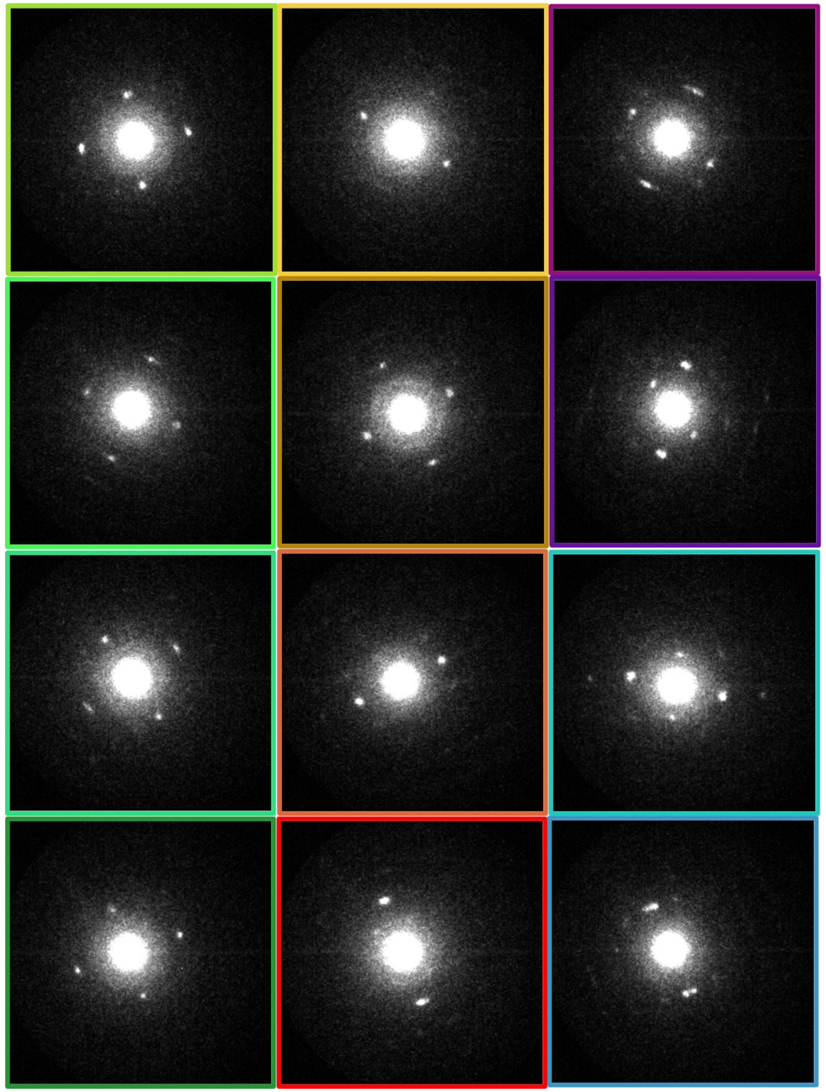


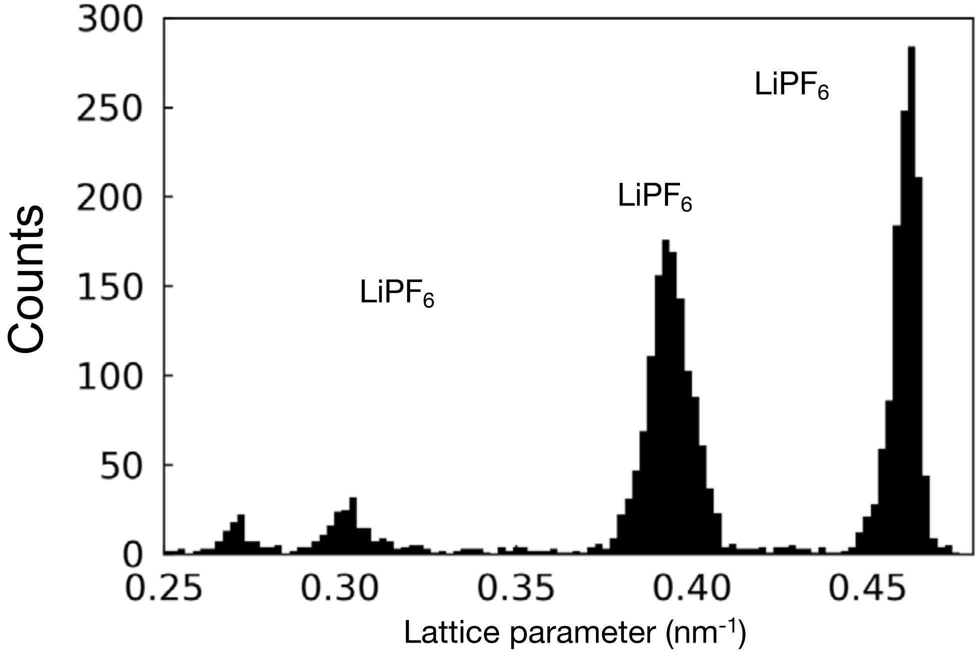


4D-STEM of Liquid Battery Electrolyte at -30ºC





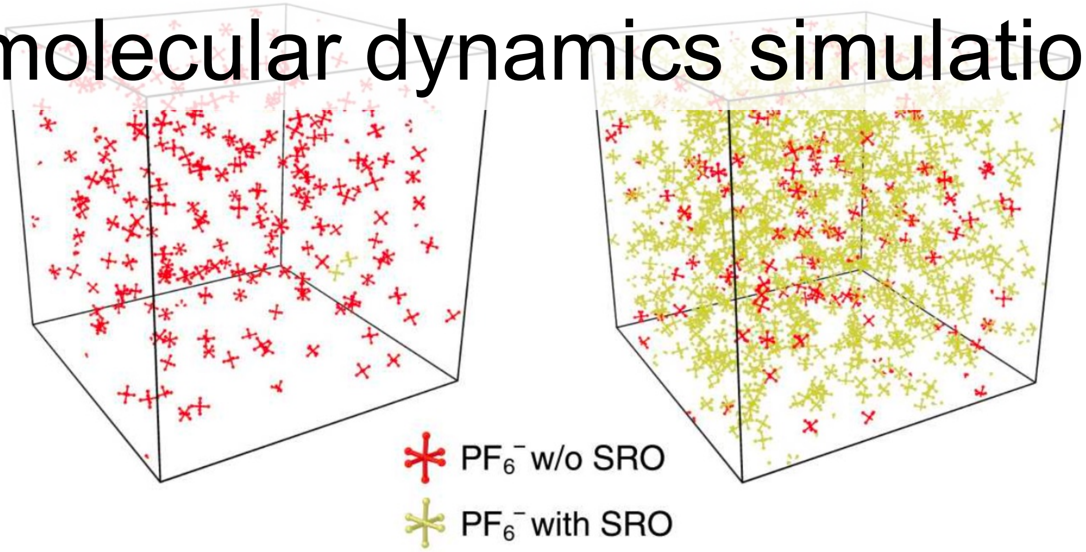


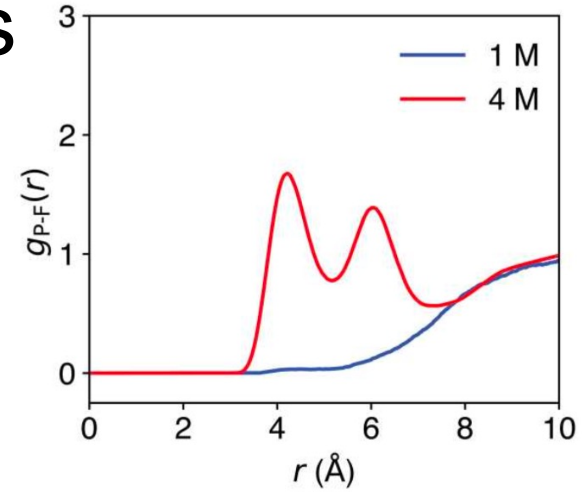


	[hkl]	d[Å]	d*[1/Å]
$LiPF_6$	[1 1 3]	2.167	0.462
LiF	[0 1 0]	2.556	0.391
$LiPF_6$	[0 -1 -2]	3.602	0.278

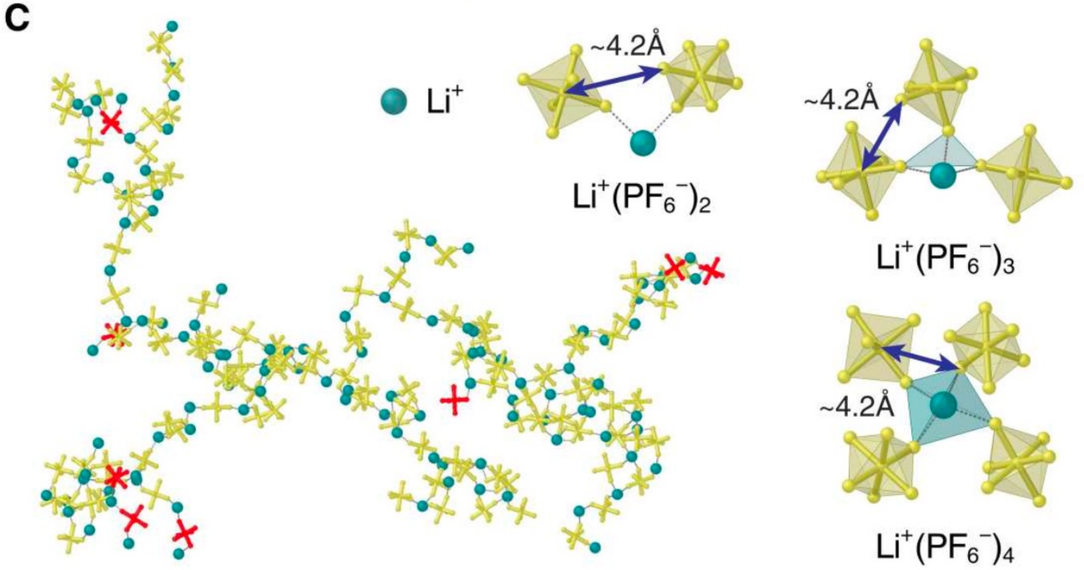
4D-STEM of Liquid Battery Electrolyte at -30ºC

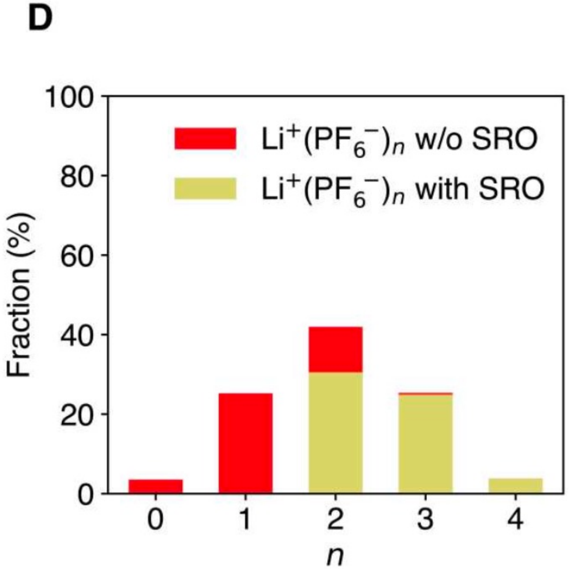
molecular dynamics simulations

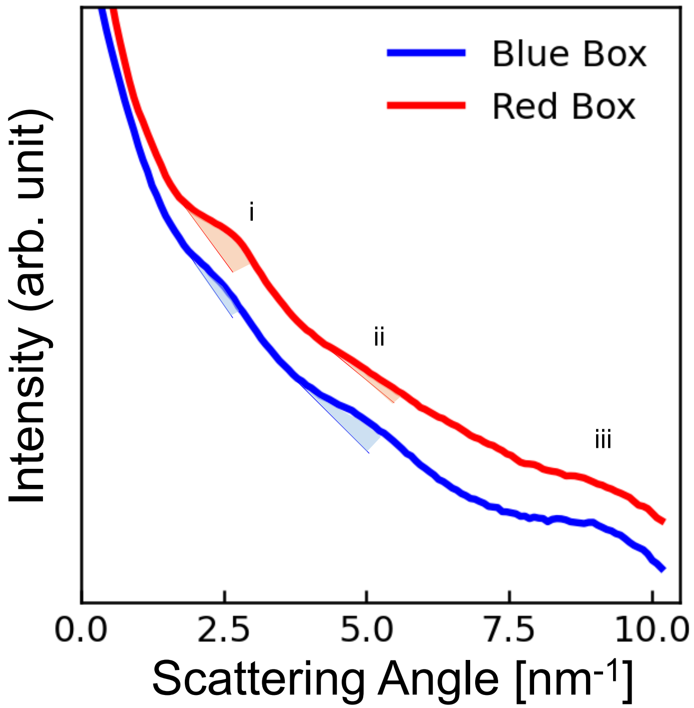




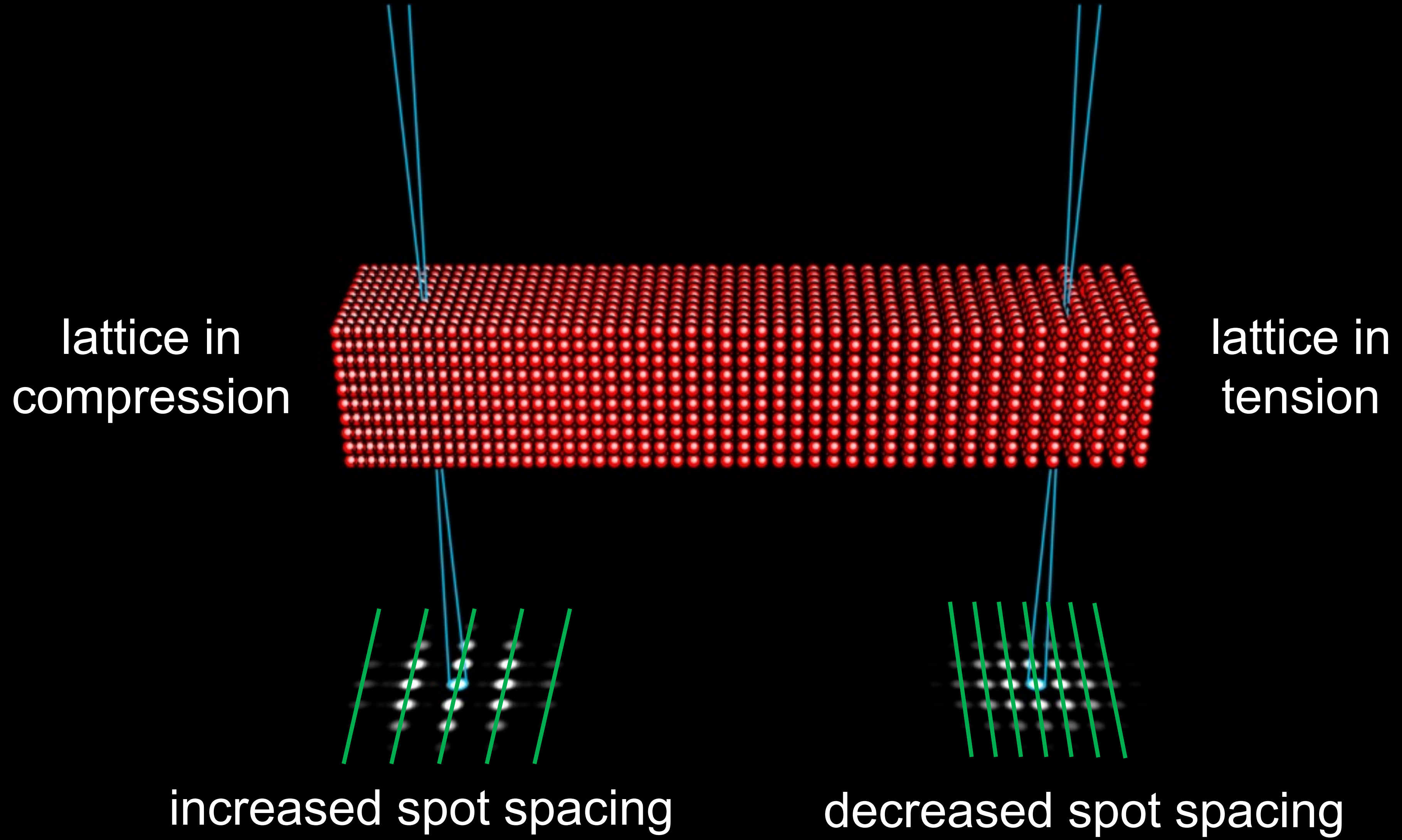
- Red region is more ordered, has higher density → **amorphous / viscous liquid phase**
- Blue region is less ordered, has lower density → **liquid**





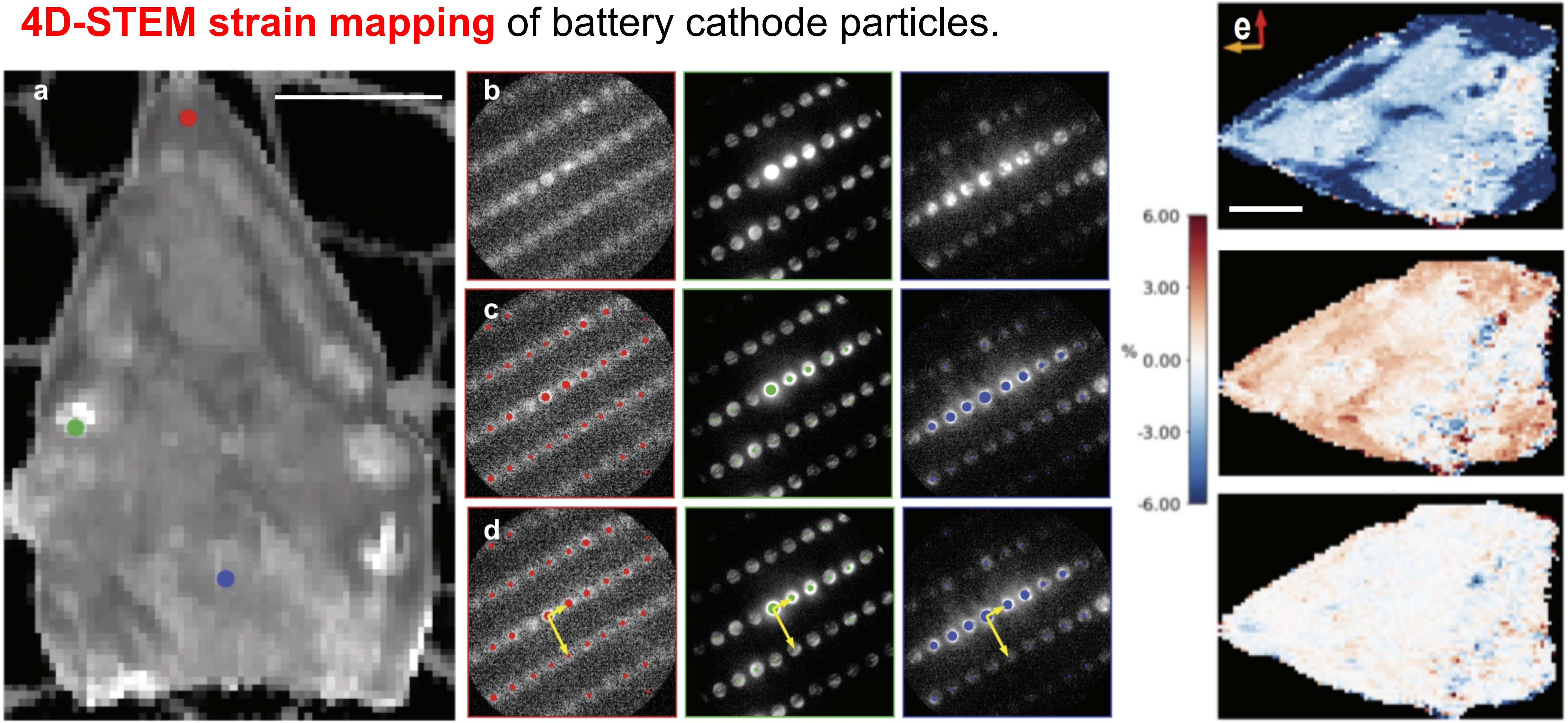


4D-STEM – Crystal Strain Mapping



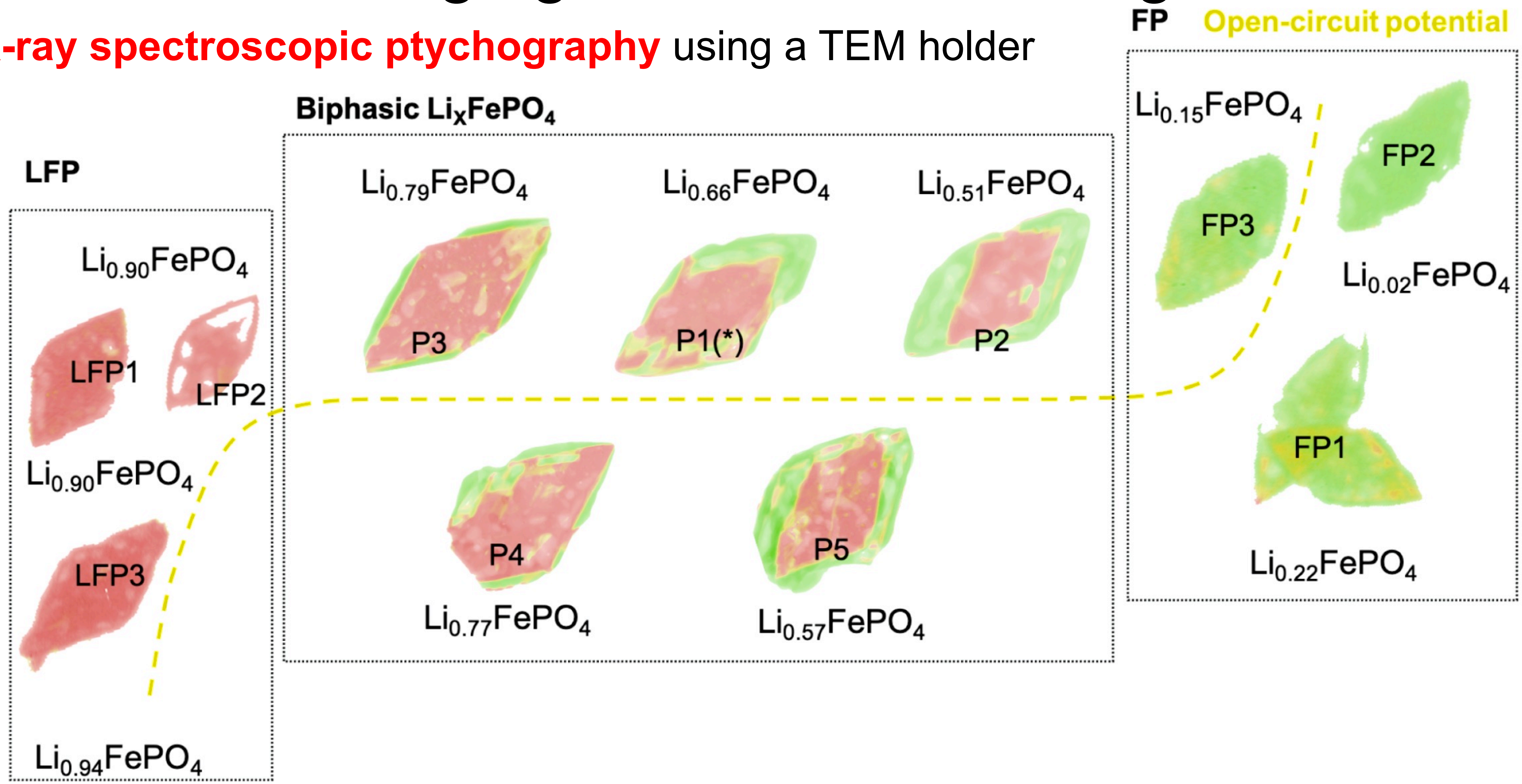
Correlative Imaging & Inverse Learning of $LiFePO_4$

4D-STEM strain mapping of battery cathode particles.



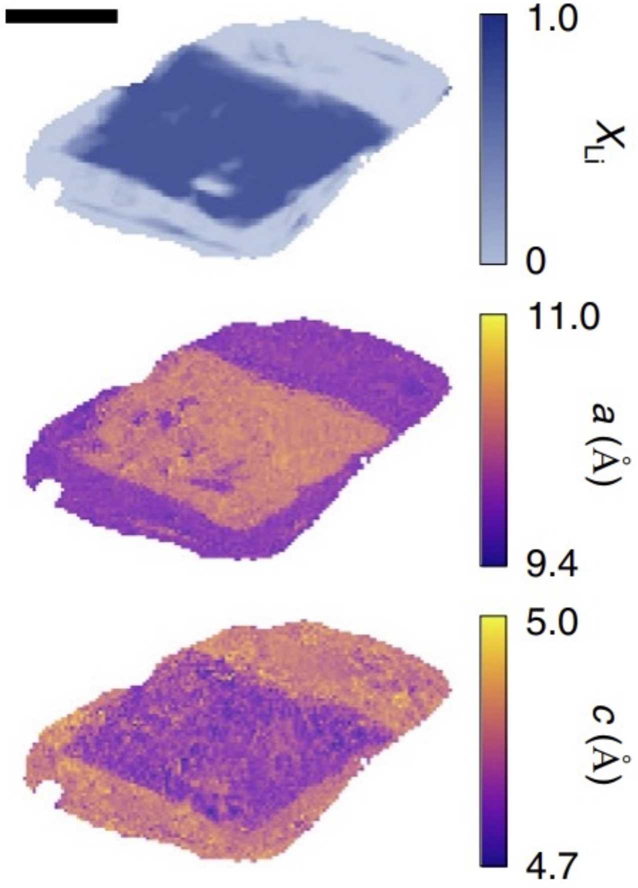
Correlative Imaging & Inverse Learning of $LiFePO_4$

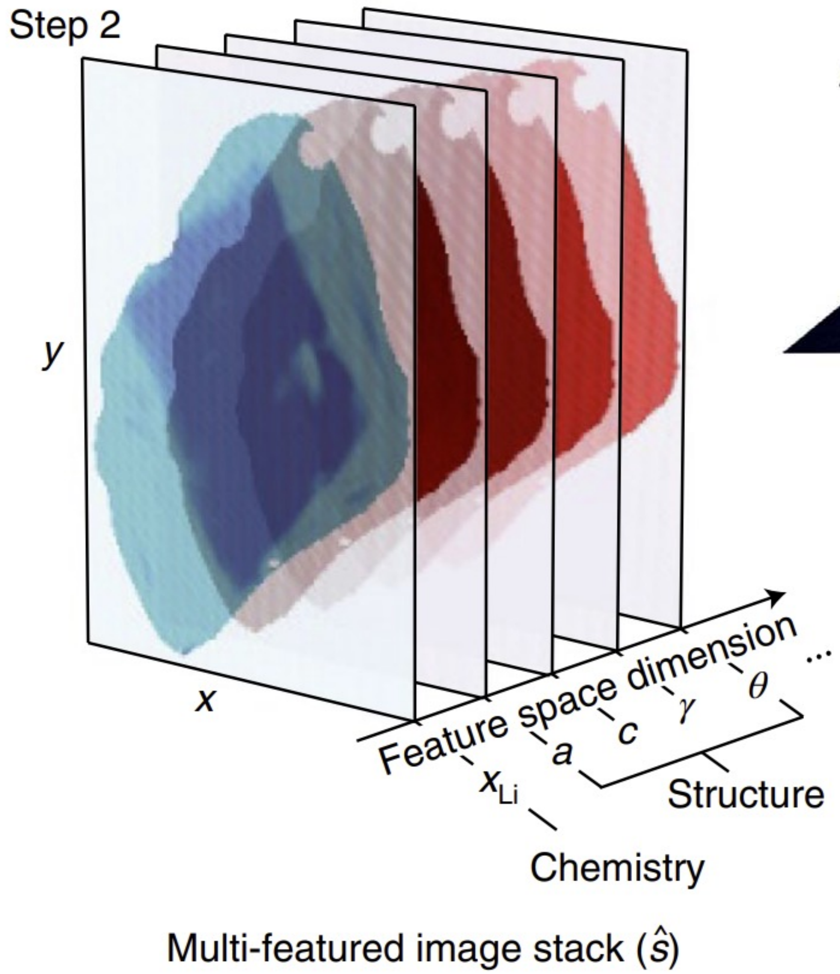
X-ray spectroscopic ptychography using a TEM holder

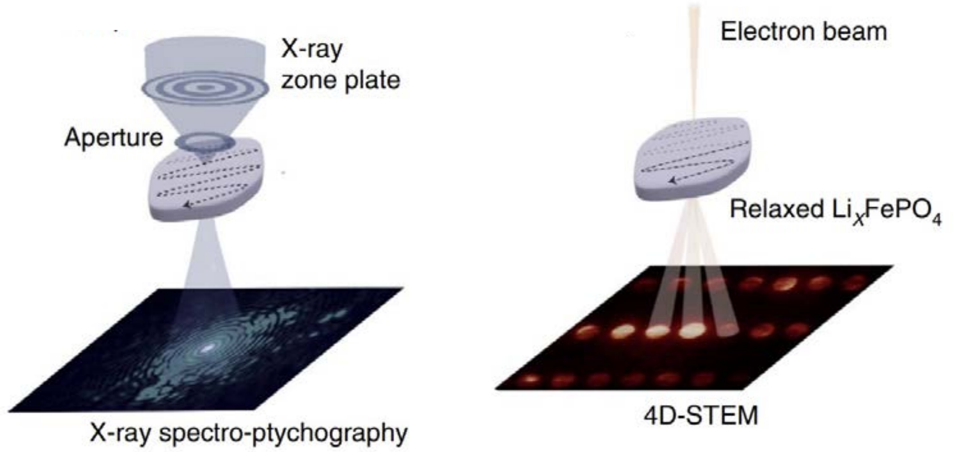


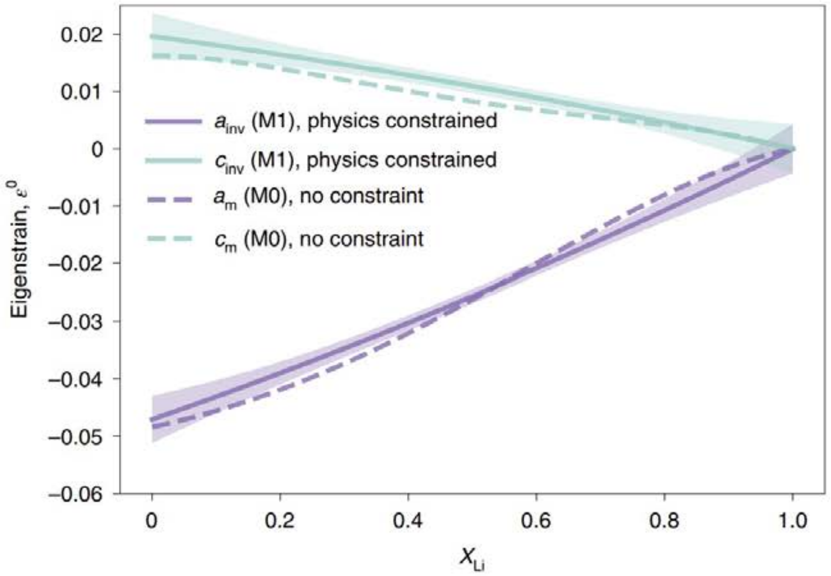
Correlative Imaging & Inverse Learning of $LiFePO_4$

Alignment and correlation of all channels → **inverse learning of constitutive law**



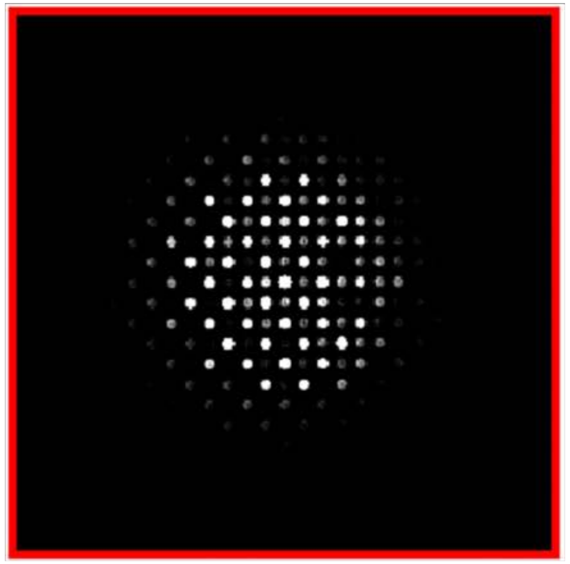


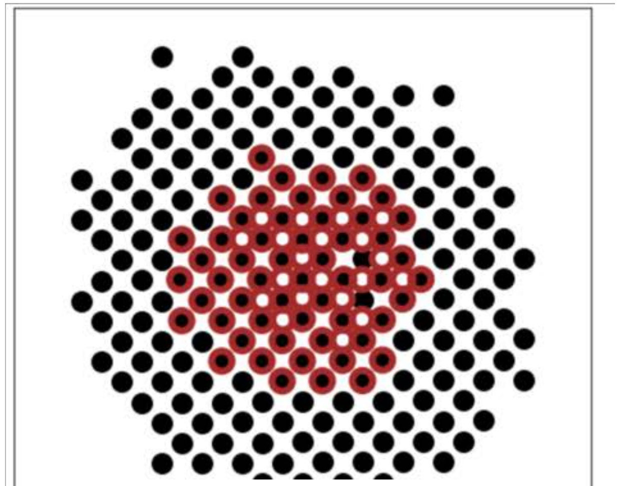




Dynamical Diffraction Complicates Disk Detection

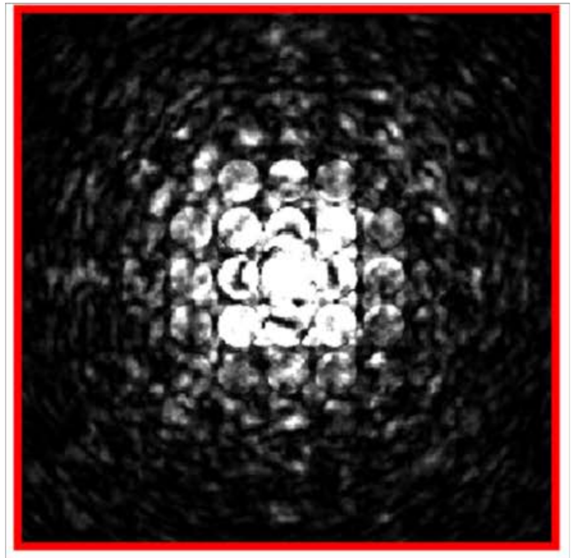
Diffraction pattern from using small convergence angle, thin sample

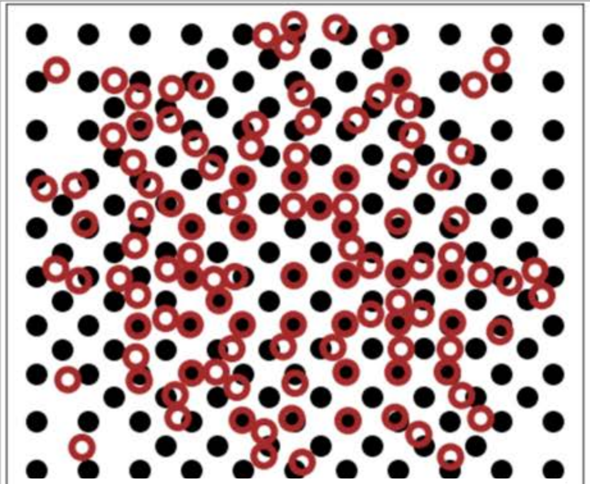




correlation detection

Diffraction pattern from using large convergence angle, thick sample





correlation detection

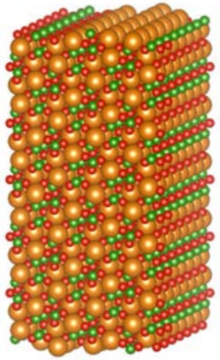
Can machine learning methods help us when our conventional image analysis pipelines fail?

Simulation Pipeline Infrastructure

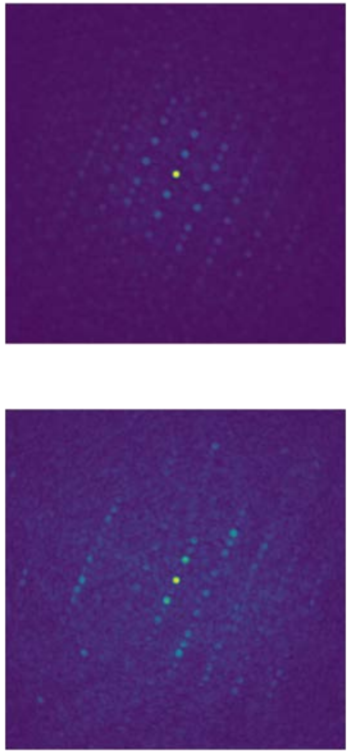
4D-SCRAPE & Manipulatt

The Materials Project
materialsproject.org

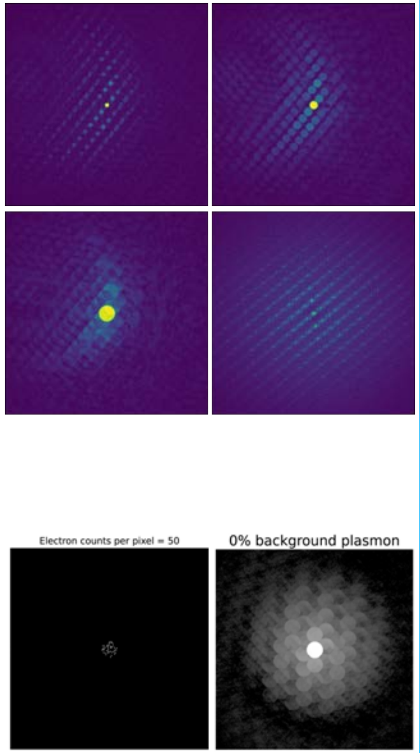
AFLOW
Automatic - FLOW for Materials Discovery



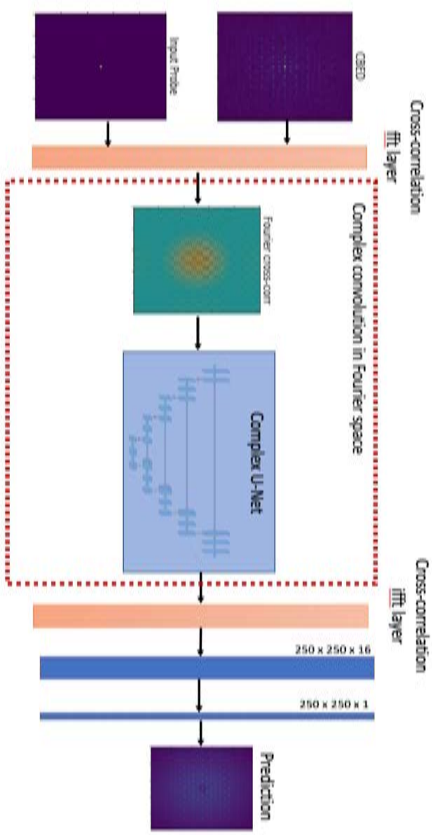
4D-MAKE



4D-PREP



4D-OPTIMIZE



Many open source software tools used:

py4DSTEM

RAPIDS

DASK

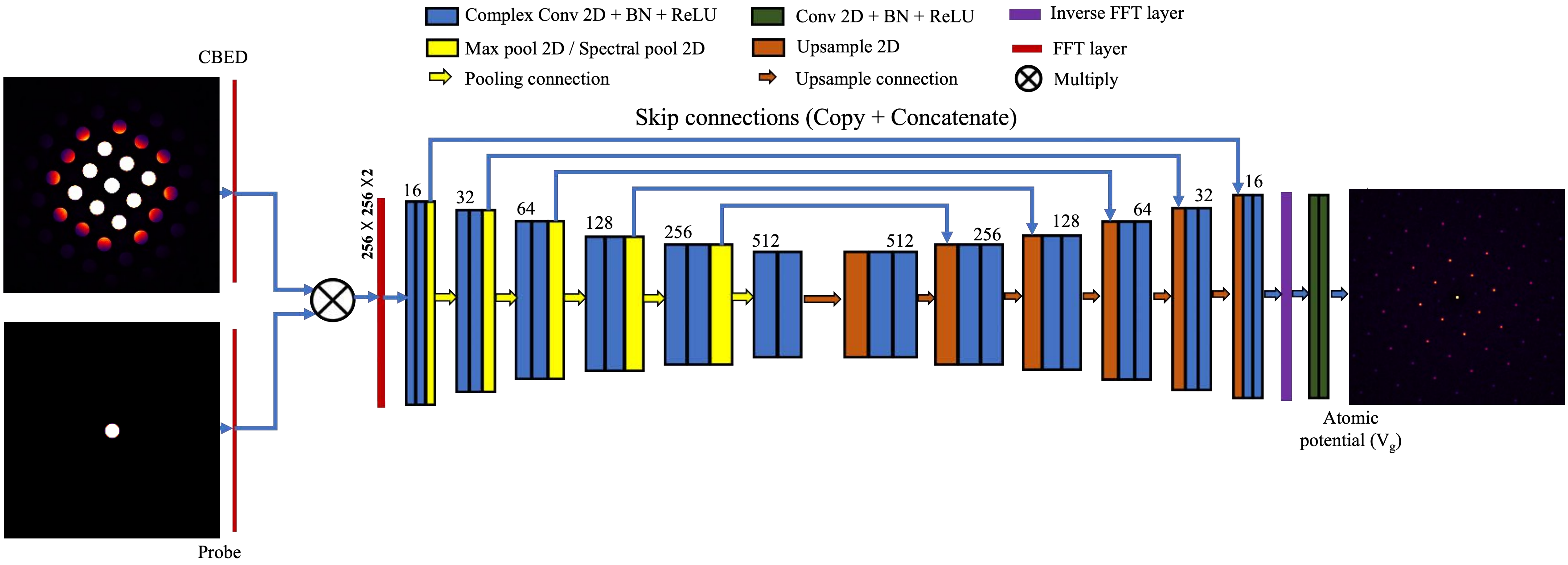
SHIFTER

HOROVOD

TensorFlow

tune

Solving Diffraction with Deep Learning – crystal4D



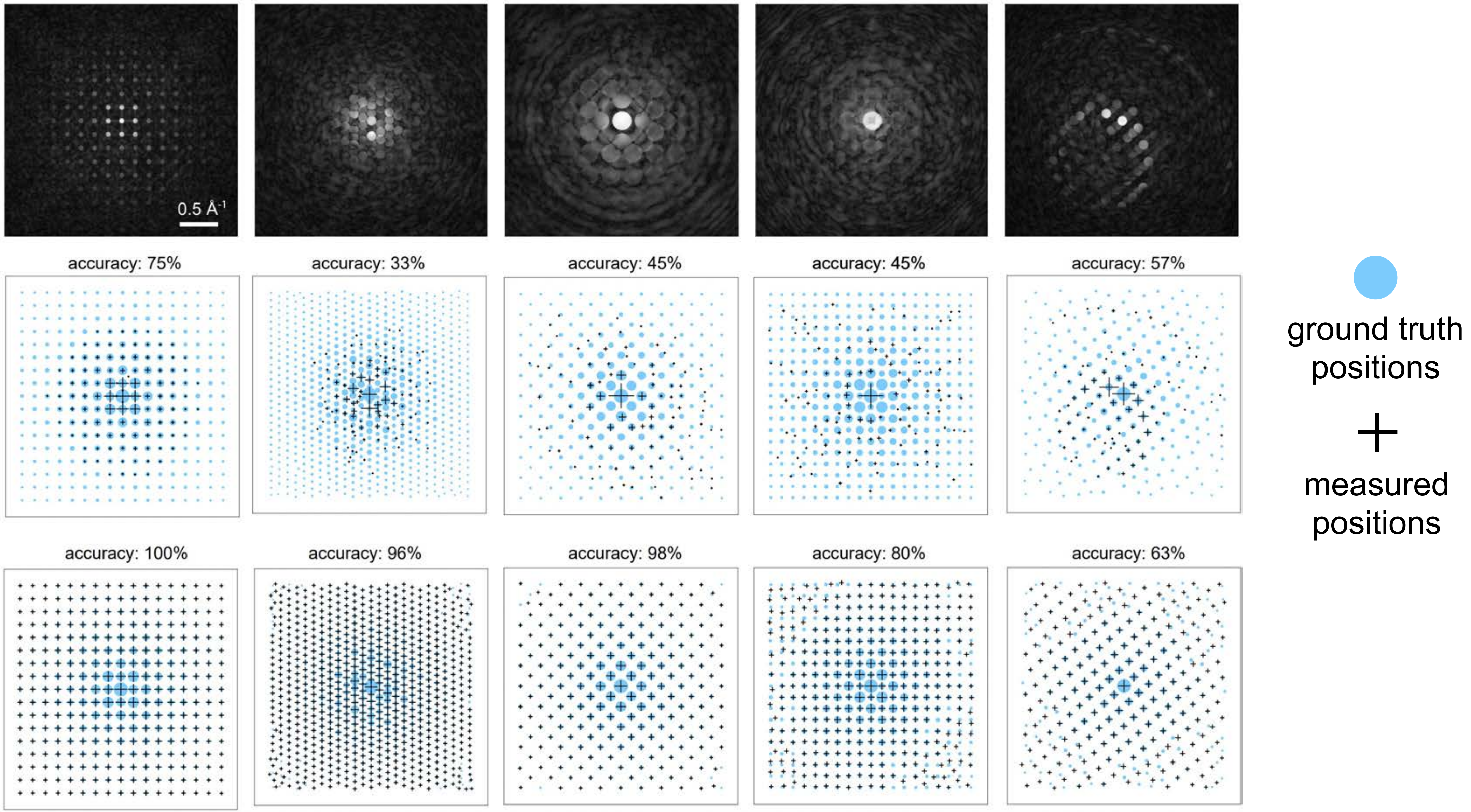
J Munshi*, A Rakowski*, et al., npj Computational Materials 8, 254 (2022)

Solving Diffraction with Deep Learning – FCU-Net

Simulated diffraction patterns

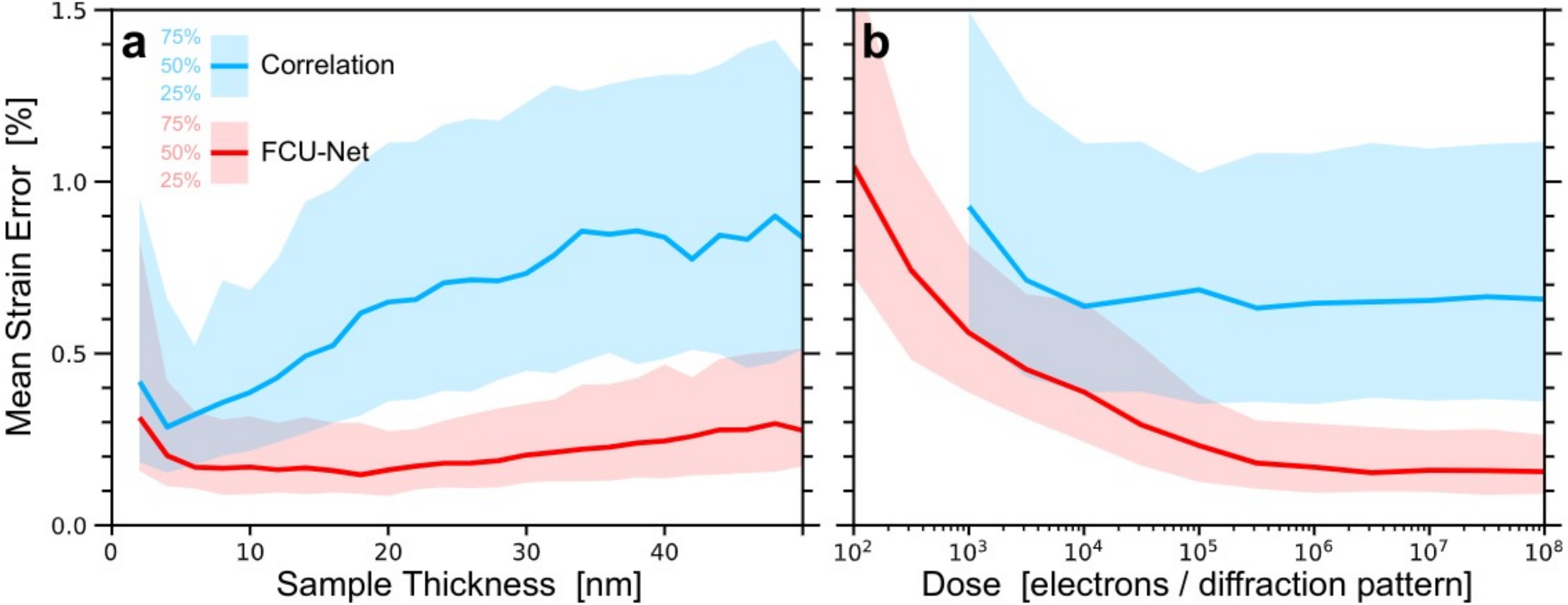
Correlation disk detection

Deep learning disk detection



J Munshi*, A Rakowski*, et al., npj Computational Materials 8, 254 (2022)

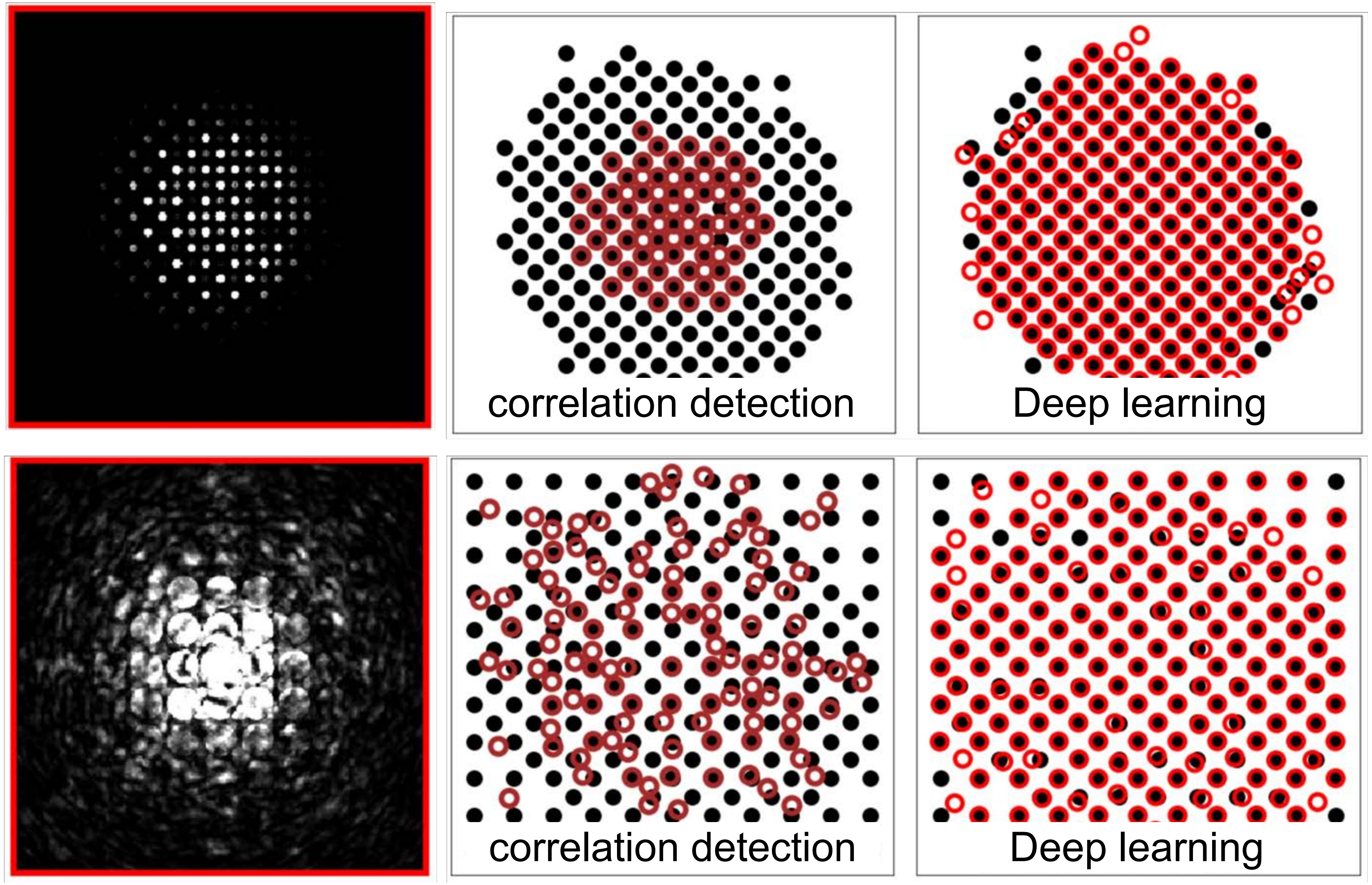
Solving Diffraction with Deep Learning – FCU-Net



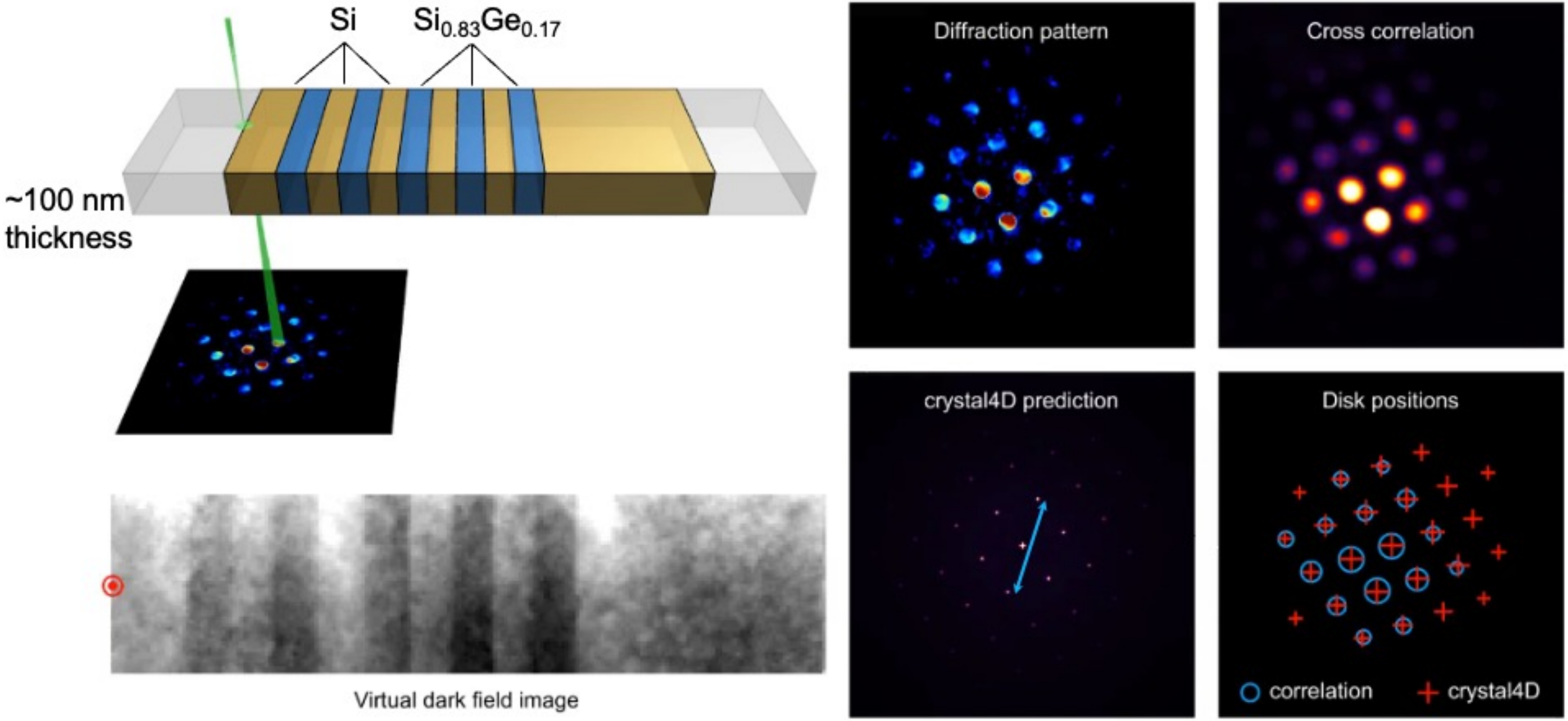
Dynamical Diffraction Defeated by Deep Learning

Diffraction pattern from using small convergence angle, thin sample

Diffraction pattern from using large convergence angle, thick sample

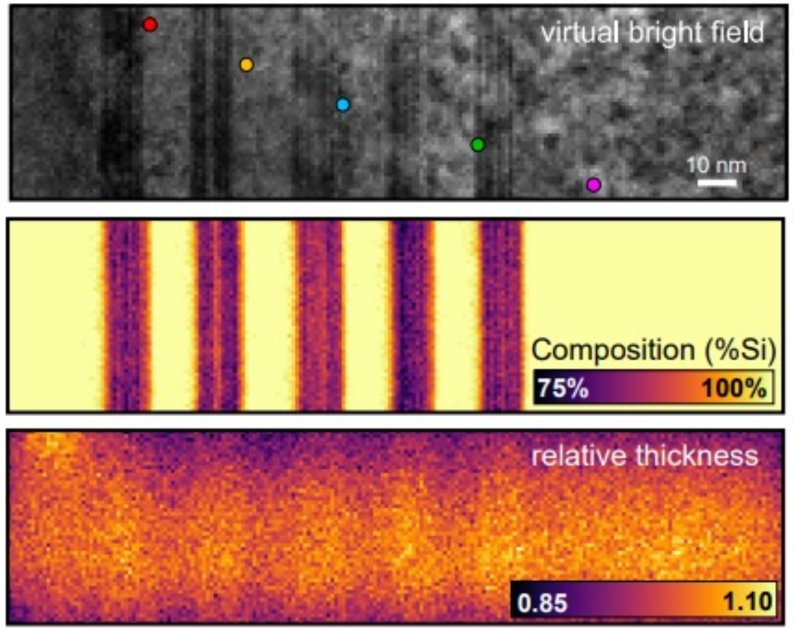


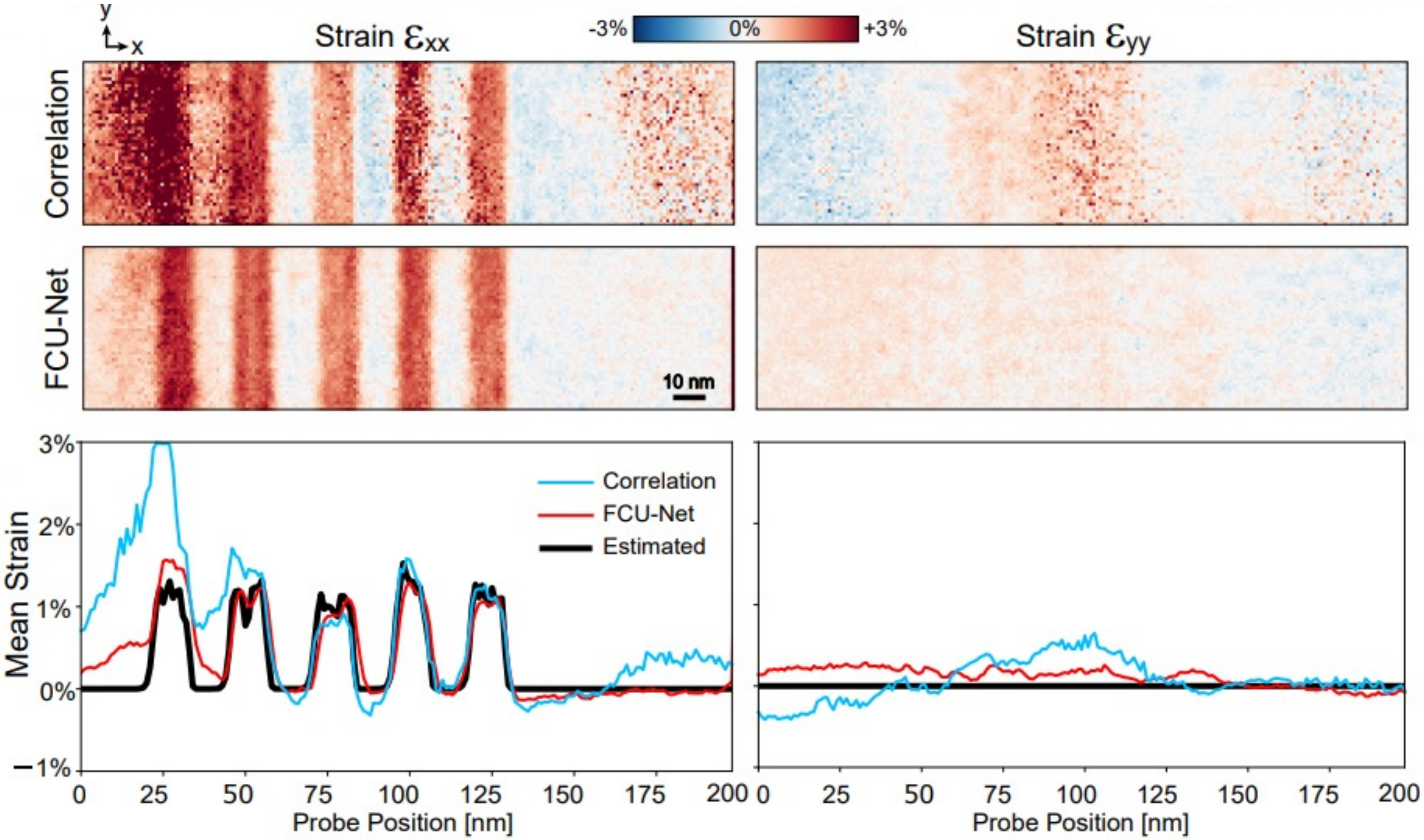
Strain Mapping of SiGe Multilayers w/ Deep Learning



J Munshi*, A Rakowski*, et al., npj Computational Materials 8, 254 (2022)

Strain Mapping of SiGe Multilayers w/ Deep Learning





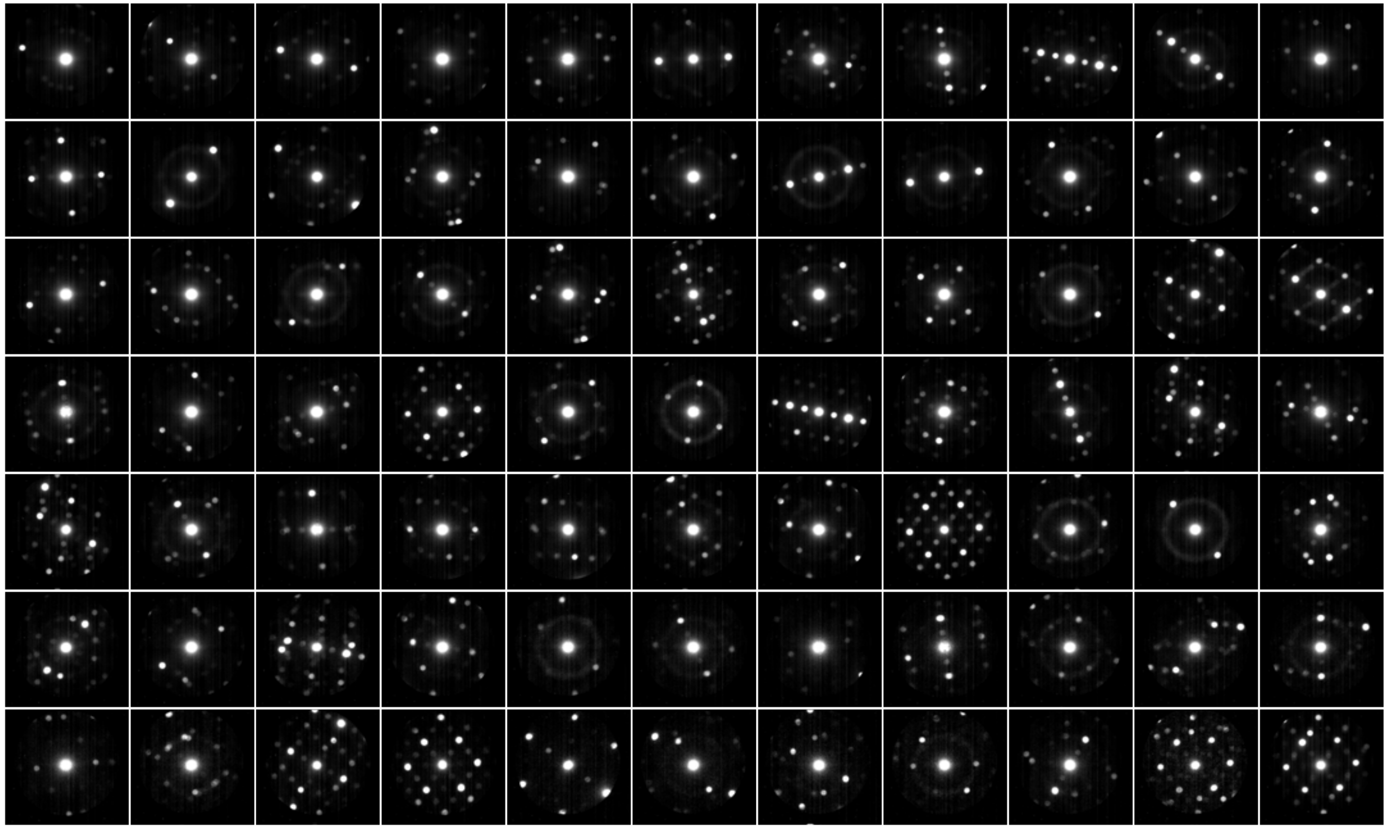
Strain mapping of alternating multilayers of Si and SiGe.

- Our deep learning approach significantly improves the **measurement accuracy** over conventional correlation, and **does not require** any **labeled training data**.

Blind Identification of Crystal Structures with ML

Our existing **phase mapping** methods work well for **known crystal structures**.

But what happens when we see an **unknown pattern** in an experiment?



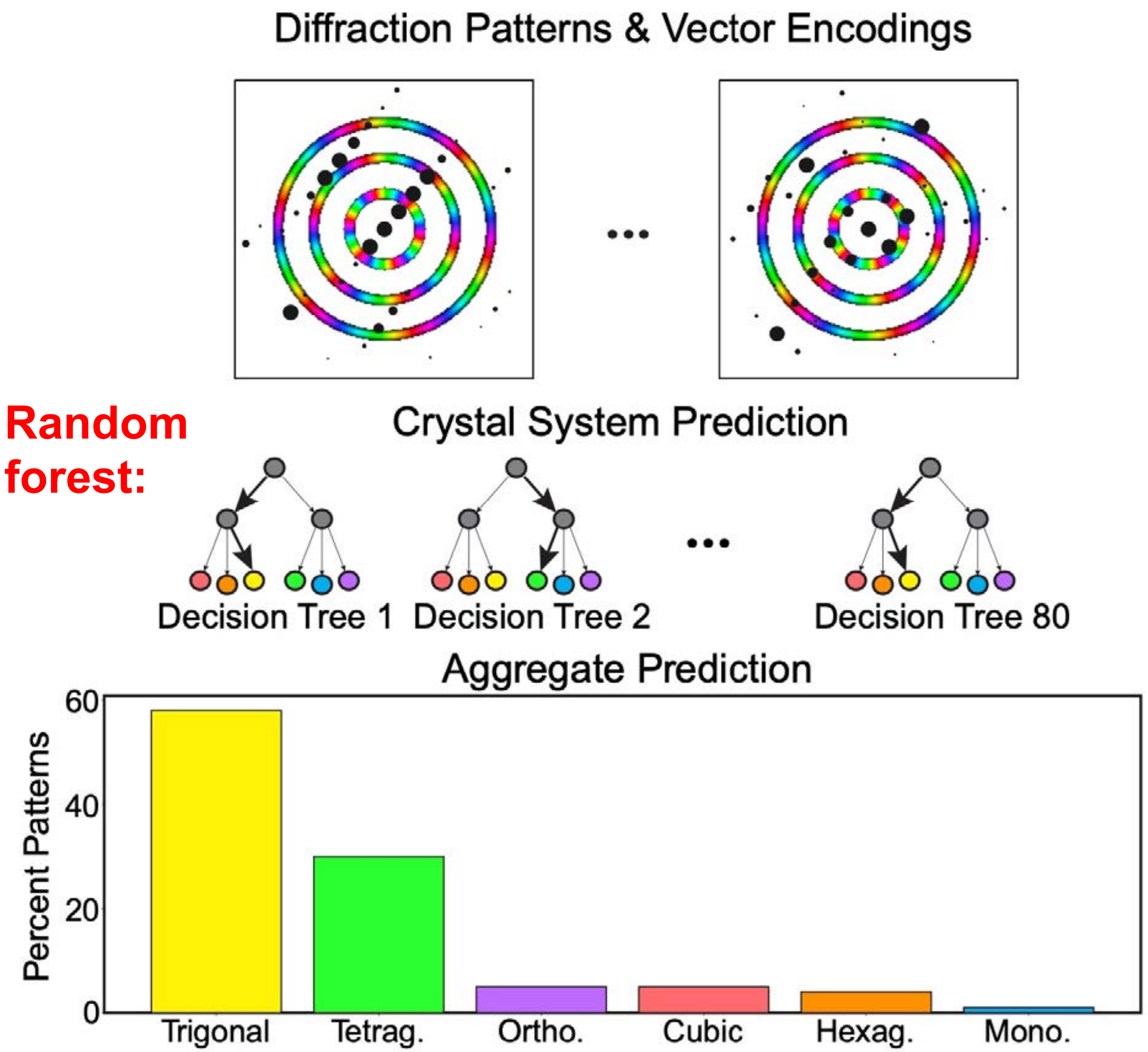
For example, **high-throughput** structure **discovery** experiments.

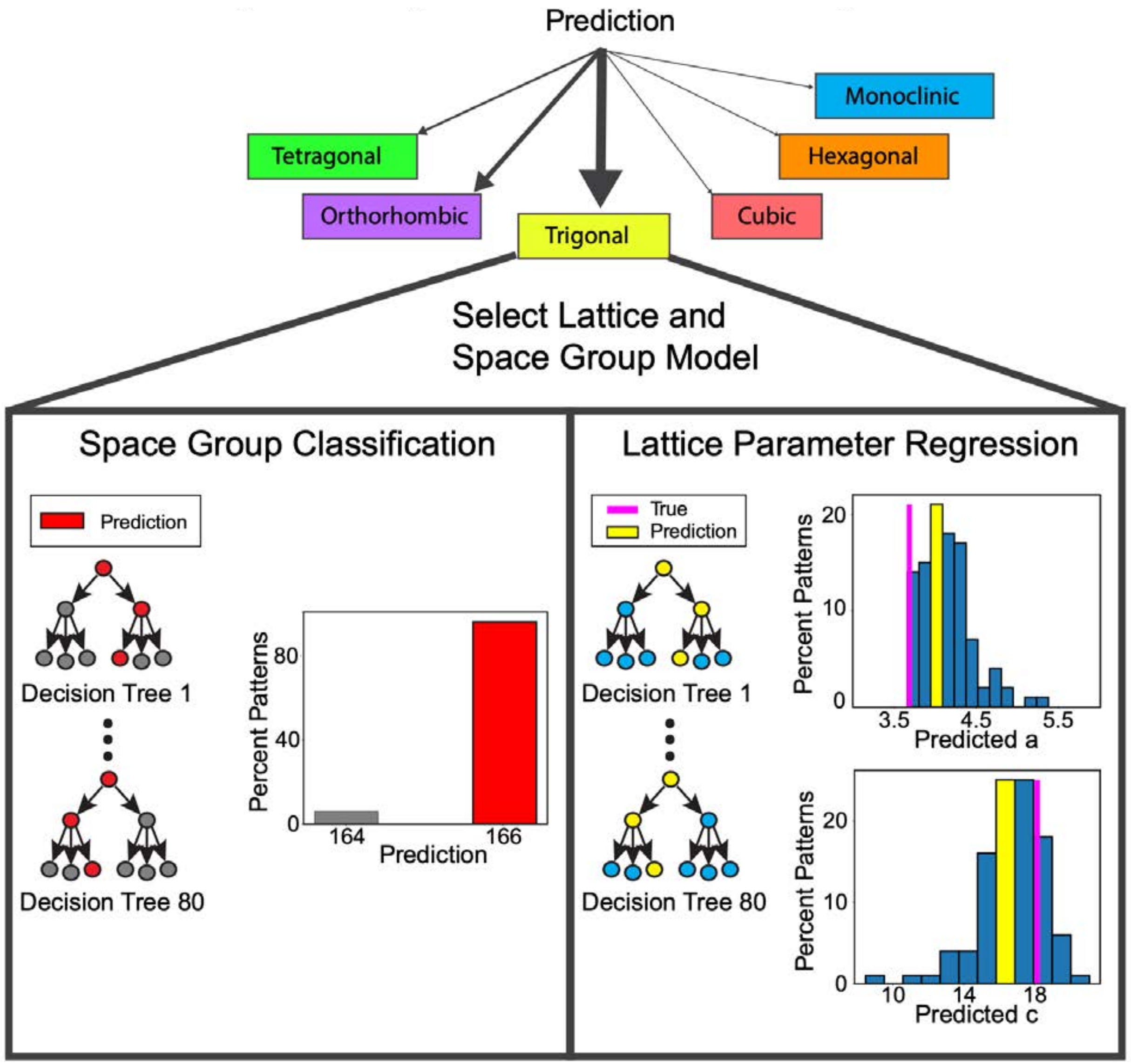
Could we predict **crystal system**, **space group**, or even the **lattice params** with ML?

A Gleason, A Rakowski, B Savitzky, M Henderson, J Ciston, and C Ophus, *manuscript in preparation*

Blind Identification of Crystal Structures with ML

Random forest:





Blind Identification of Crystal Structures with ML

Prediction accuracy (square brackets) and true positive rate (round brackets)

Fraction of Test Test

0% 5% 10% 15% 20%

Individual Pattern Confusion Matrix (Actual Values × Predicted Values)

	cubic	hex.	trig.	tetra.	mono.	ortho.
cubic	10	0.1	>0.1	0.2	0.5	0.8
hex.	0.2	7.4	0.2	0.9	1.2	2.8
trig.	0.1	0.7	2.9	1.1	1.3	1.9
tetra.	0.3	0.6	0.1	10	1.8	3.9
mono.	0.1	0.2	0.1	0.7	16	6
ortho.	0.2	0.5	0.1	1.3	5	20

Individual Pattern

Structure	Crystal System True Pos Rate (Sensitivity)	Space Group Accuracy	Median Lattice Contant Error (Å) a	b	c
Cubic	0.92 (0.86)	0.86	0.03	= a	= a
Hexagonal	0.78 (0.58)	0.65	0.12	= a	0.26
Trigonal	0.86 (0.36)	0.71	0.09	= a	0.60
Tetragonal	0.70 (0.61)	0.54	0.13	= a	0.54
Monoclinic	0.62 (0.69)	0.40	1.24	1.11	1.47
Orthorhombic	0.57 (0.74)	0.31	0.61	0.76	1.37

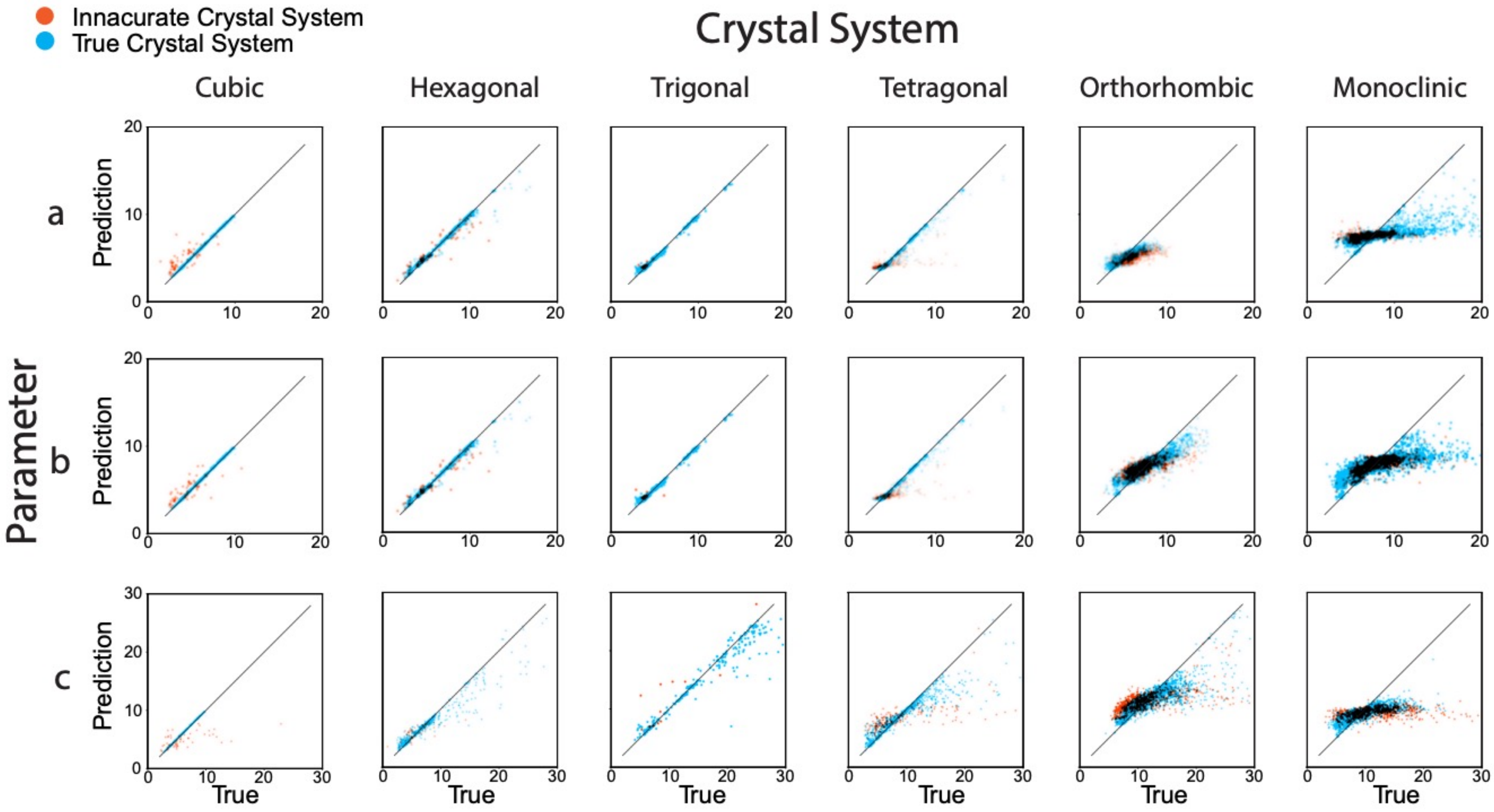
Aggregate Confusion Matrix (Actual Values × Predicted Values)

	cubic	hex.	trig.	tetra.	mono.	ortho.
cubic	11	0.1	0	0.1	0.5	0.7
hex.	0.1	9.2	0.1	0.3	1	1.9
trig.	0.1	0.5	3.8	0.7	1.4	1.5
tetra.	0.2	0.2	0	12	1.6	3.1
mono.	0	0	0	0.3	21	2
ortho.	0.2	0	0	0.7	3.5	23

10 Pattern Aggregate

Structure	Crystal System True Pos Rate (Sensitivity)	Space Group Accuracy	Median Lattice Contant Error (Å) a	b	c
Cubic	0.95 (0.89)	0.90	0.02	= a	= a
Hexagonal	0.92 (0.73)	0.82	0.09	= a	0.15
Trigonal	0.97 (0.48)	0.85	0.06	= a	0.41
Tetragonal	0.85 (0.70)	0.72	0.07	= a	0.28
Monoclinic	0.72 (0.90)	0.45	1.23	1.10	1.38
Orthorhombic	0.71 (0.84)	0.39	0.51	0.67	1.16

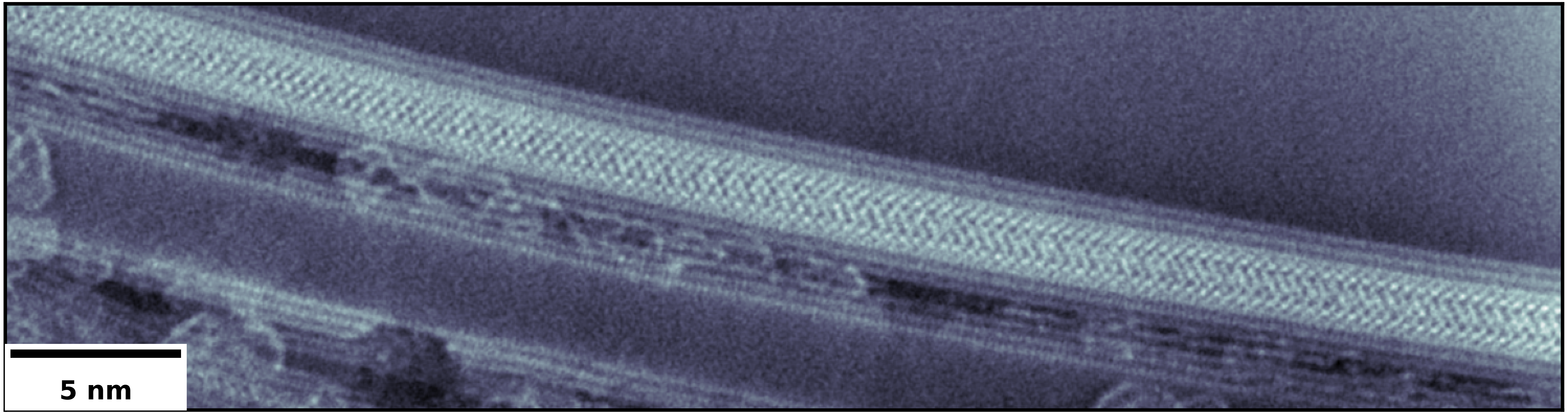
Blind Identification of Crystal Structures with ML



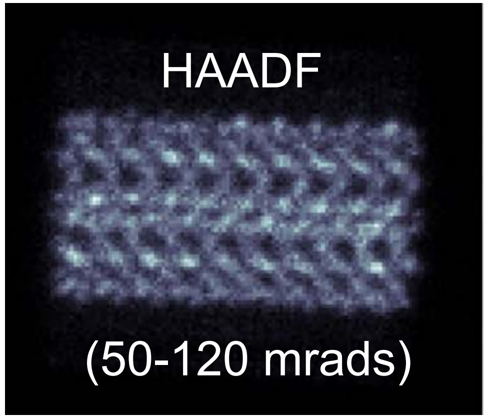
A Gleason, A Rakowski, B Savitzky, M Henderson, J Ciston, and C Ophus, *manuscript in preparation*

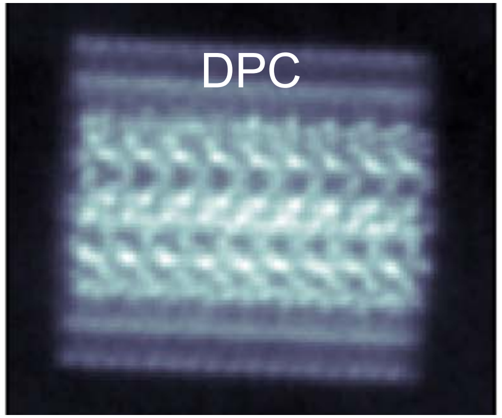
Ptychographic Atomic Electron Tomography

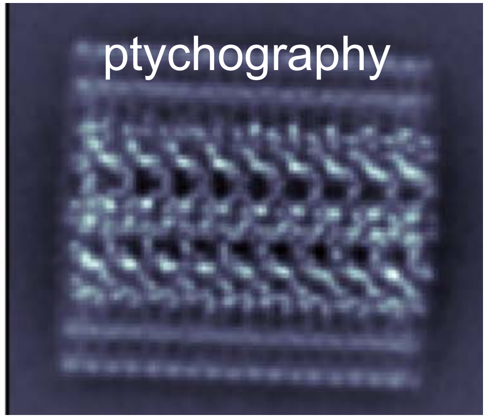
Sample – ZrTe nanowire, encapsulated in double-walled carbon nanotube



Different STEM imaging modes:

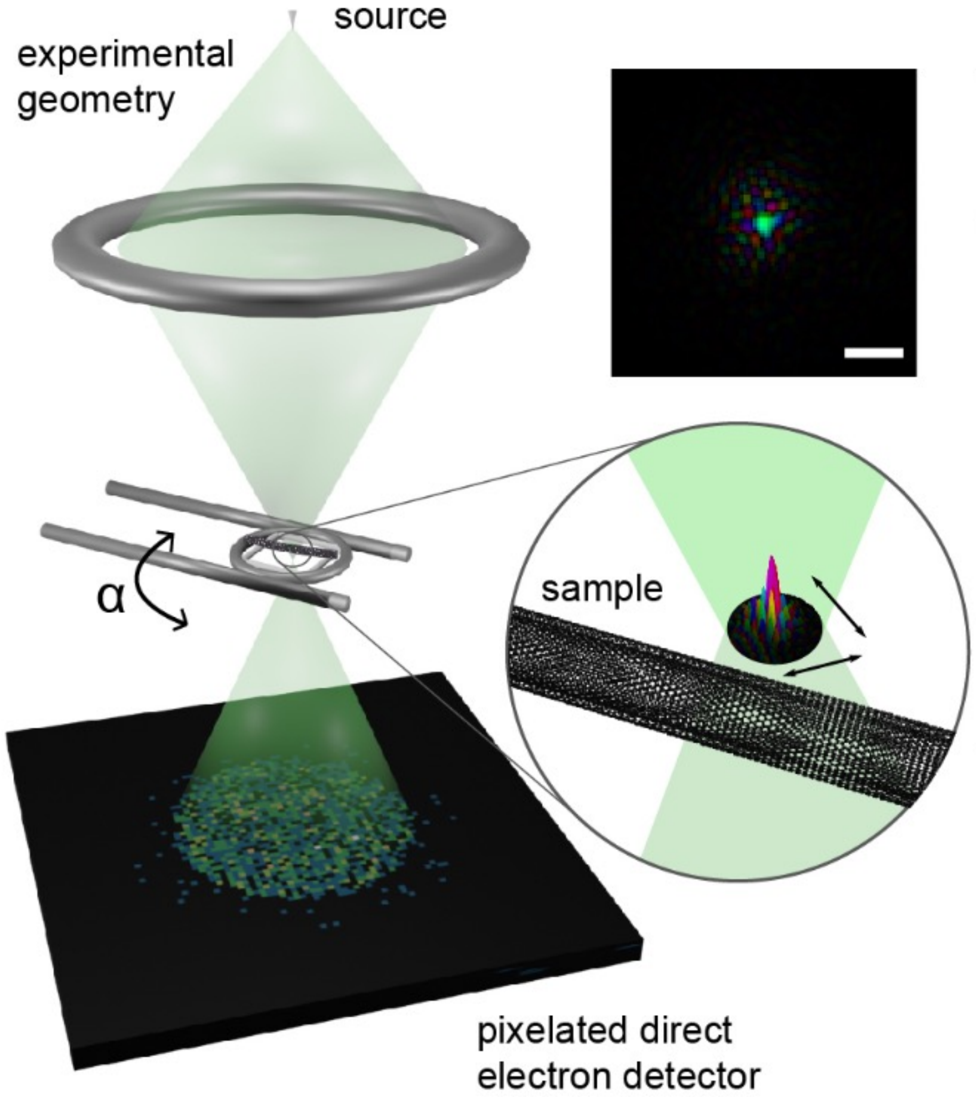


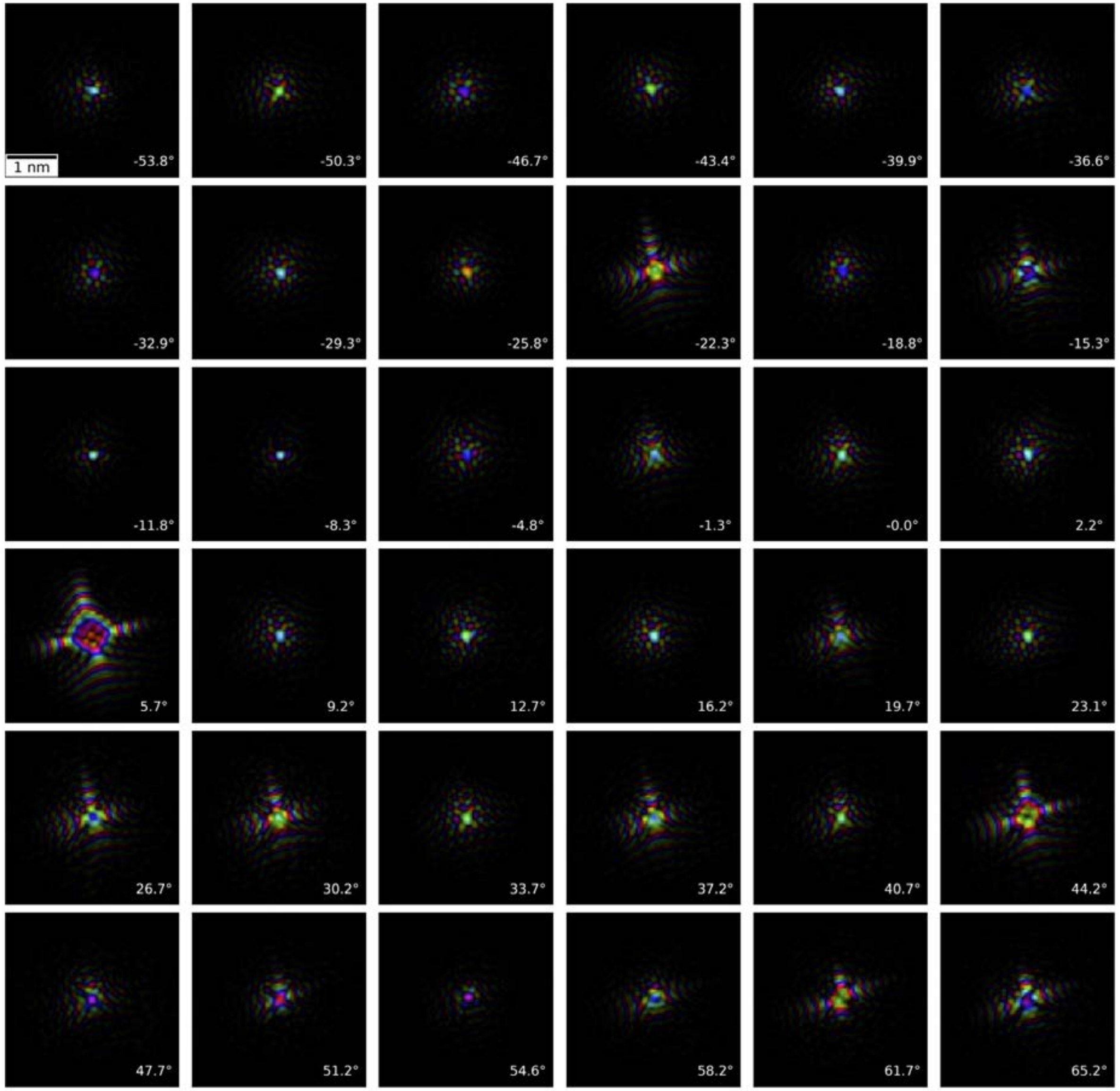




P Pelz et al., Nature Communications 14, 7906 (2023)

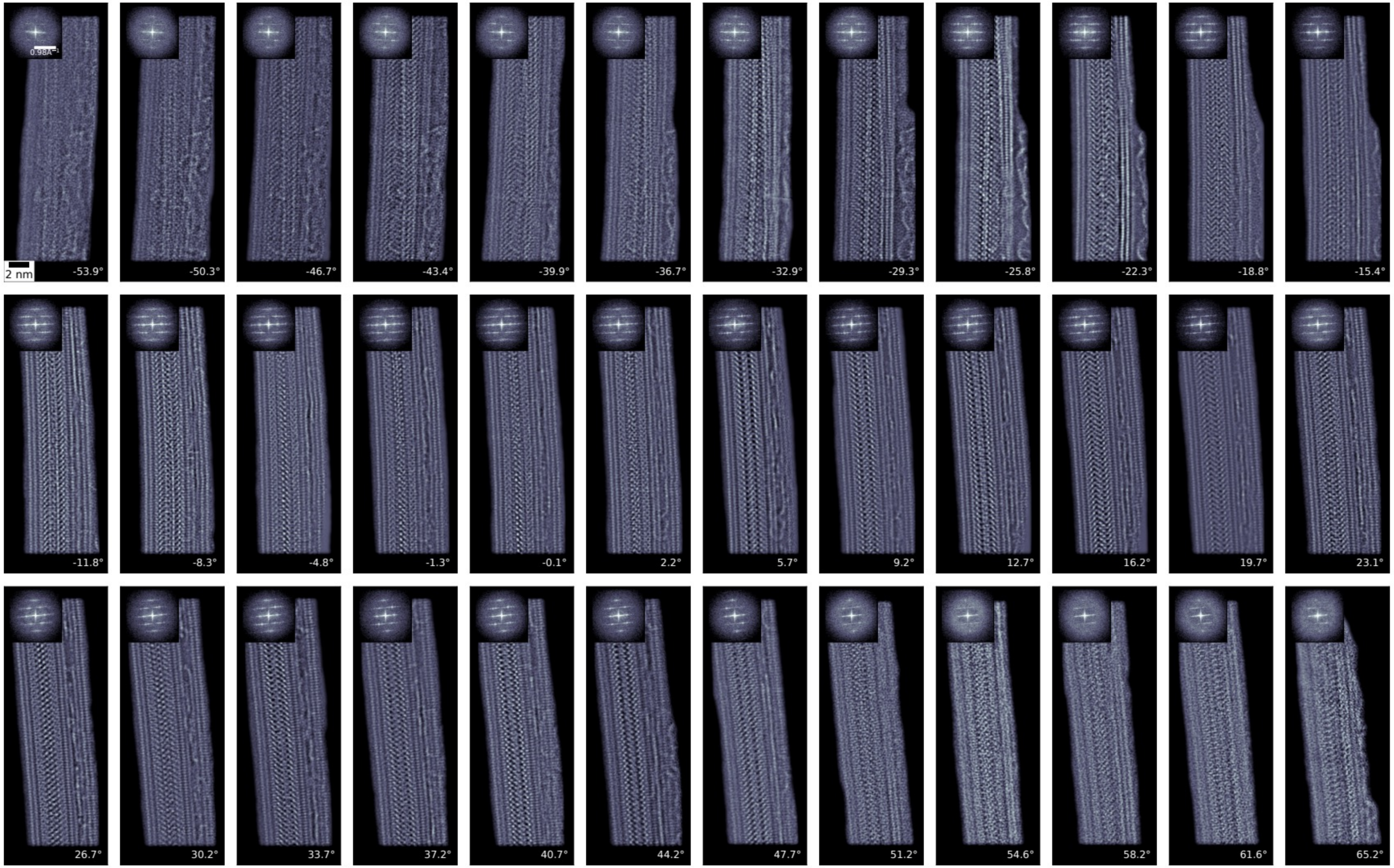
Ptychographic Atomic Electron Tomography



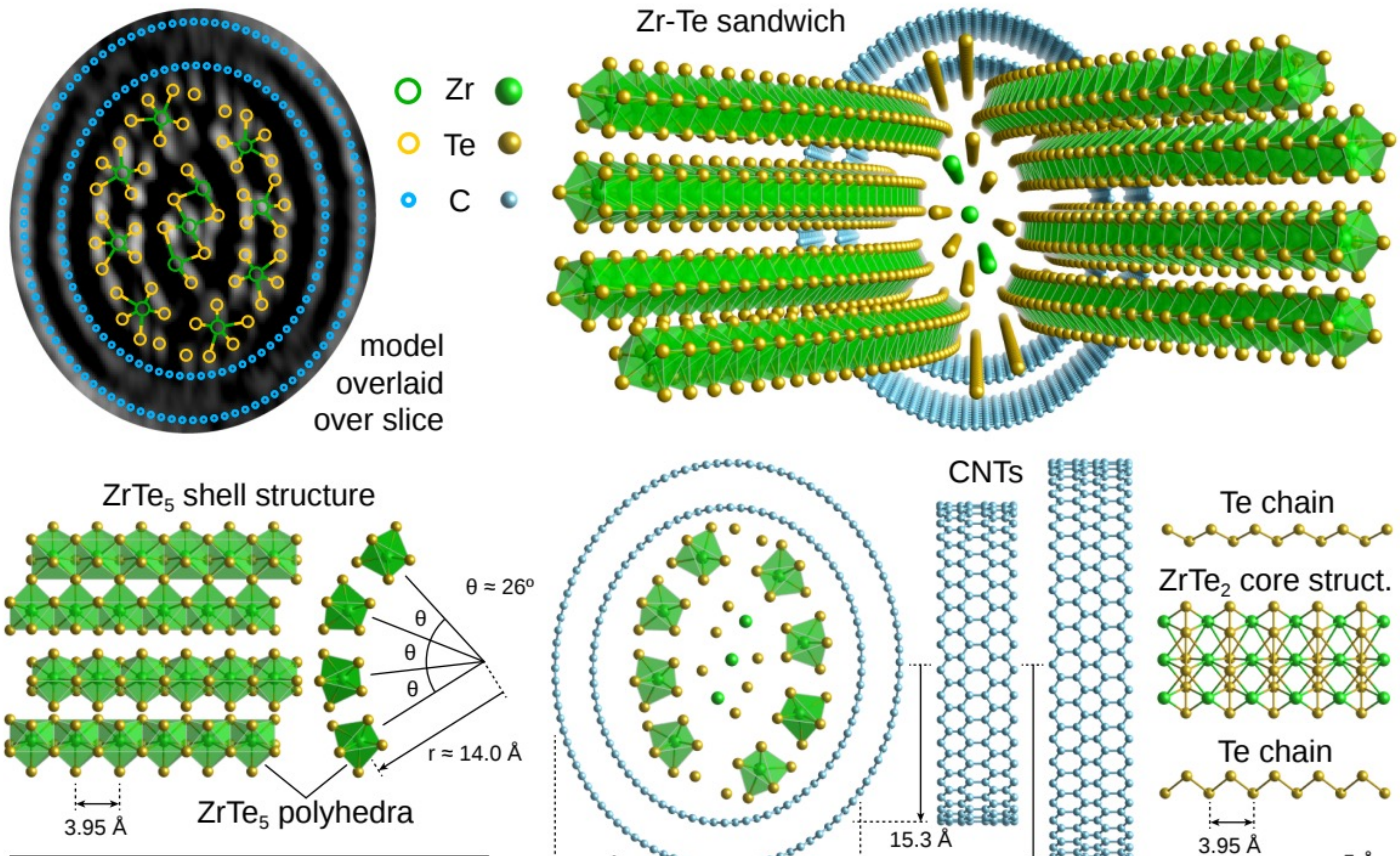


P Pelz et al., Nature Communications 14, 7906 (2023)

Ptychographic Atomic Electron Tomography

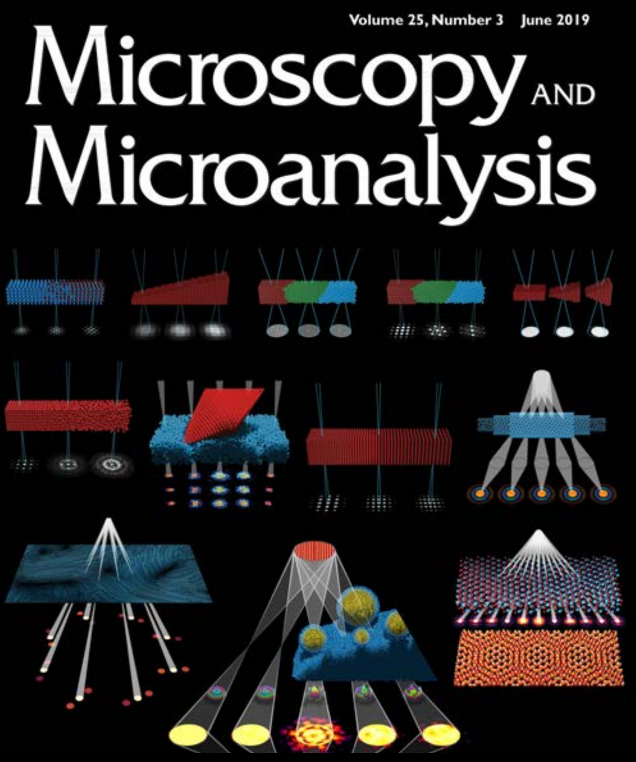


Ptychographic Atomic Electron Tomography



$8 \cdot ZrTe_5 + 3 \cdot ZrTe_2 + 4 \cdot Te = Zr_{11}Te_{50}$

More Info – contact me at **clophus@lbl.gov**

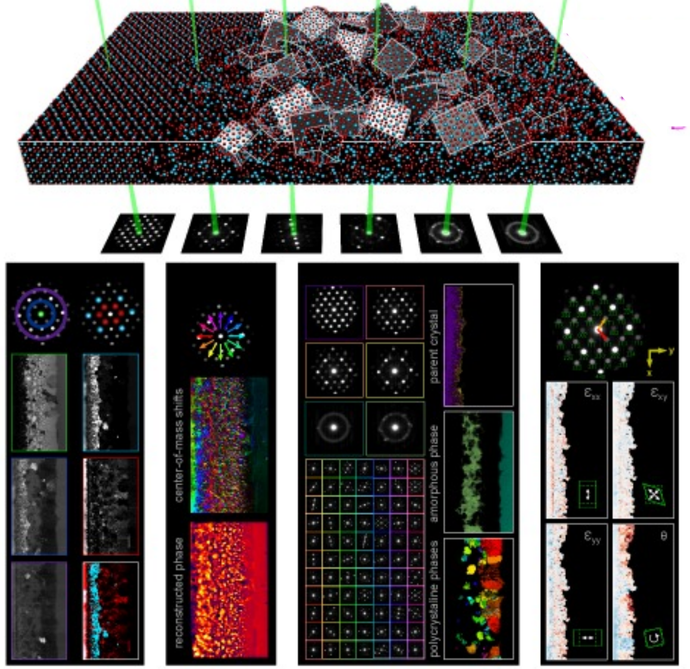


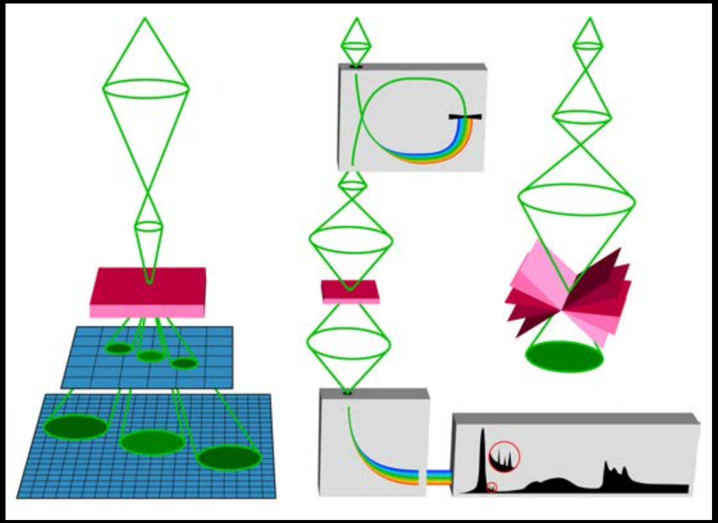
4D-STEM Review

C Ophus, Microscopy and Microanalysis 25, 563 (2019)

4D-STEM analysis: **py4DSTEM**

github.com / py4dstem
tinyurl.com / py4dstem





Quantitative STEM Imaging, Diffraction, Spectroscopy and Tomography

Colin Ophus, Annual Review of Materials Research 53, 105 (2023).

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