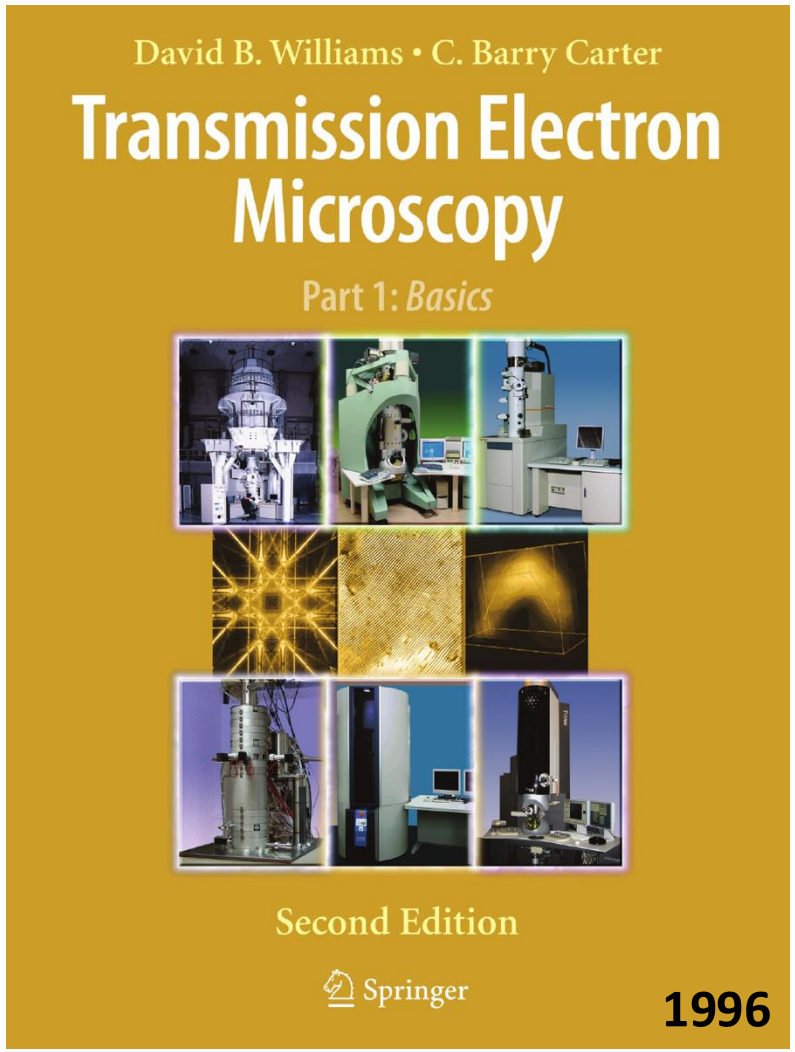
4D STEM

Austin Houston

THE UNIVERSITY OF TENNESSEE KNOXVILLE

Basics



21.8 OTHER METHODS

21.8.A Scanning Methods

Diffraction is as useful in scanning-beam instruments as in TEM. As we discussed back in Chapter 18, electron-backscatter diffraction (EBSD) studies are as ubiquitous today as SAD and CBED and very complementary. Many more crystals can be studied by EBSD and the specimen does not need to be thinned. EBSD patterns contain HOLZ lines just like CBED patterns.

Historically, electron channeling was another way to obtain crystallographic information in the SEM but the patterns are of such low contrast that EBSD has rendered the channeling technique obsolete (for now).

Scanning DPs can be obtained in STEMs using either one or two sets of coils both before and after the specimen. In both cases the beam is stationary at the plane of the specimen and rocks back and forth in a manner similar to the hollow-cone or precession methods we described in Chapters 18 and 20. Using only one scan coil below the specimen partially 'de-rocks' the beam but two coils fully 'de-rock' the beam such that, instead of the DP scanning across the BF STEM detector and being recorded sequentially, the 000 disk is always on the optic axis and thus on the BF detector. However, the HOLZ lines, etc., move continuously across the detector because, like Kikuchi lines, they are 'fixed' to the specimen. The resulting double-rocking patterns, which are often named after their inventor, Alwyn Eades, cover several degrees in comparison with the fractions of a degree visible in a normal CBED disk. These patterns are very striking as you can appreciate from Figure 21.16; such patterns can be viewed in BF or DF and, as with all CBED patterns, can be sharpened by energy filtering. These scanning methods can be used to study the occurrence of forbidden reflections which are important in crystal-symmetry determinations and more detail is given in the companion text.

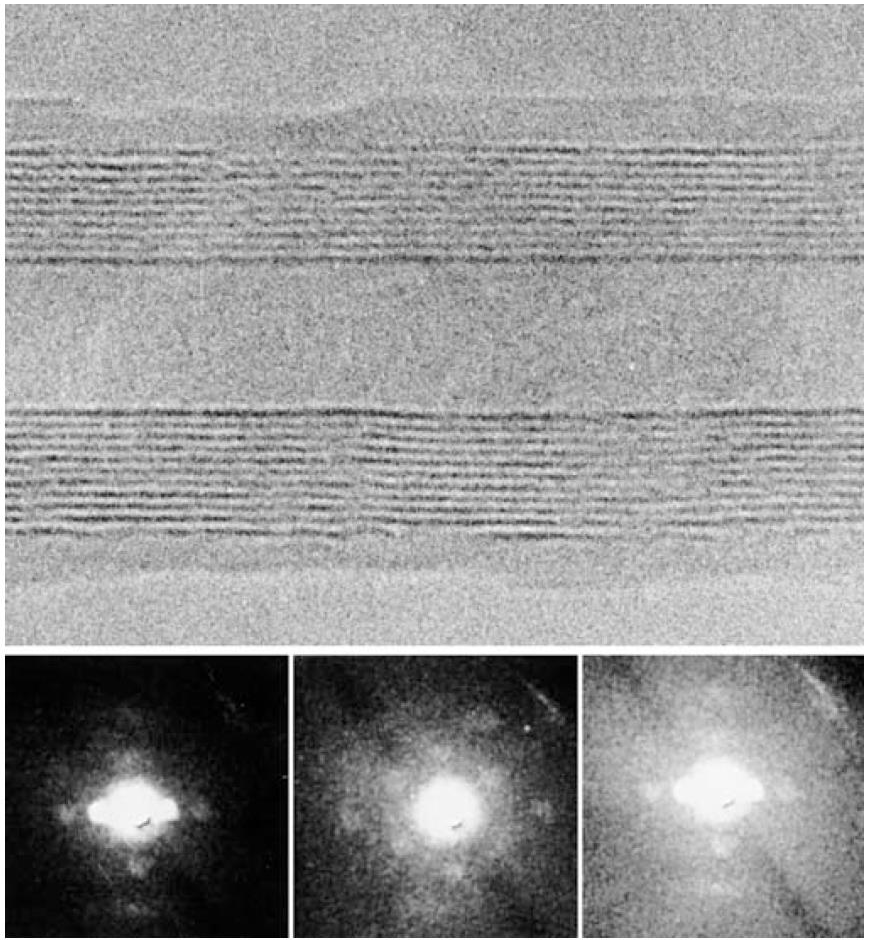
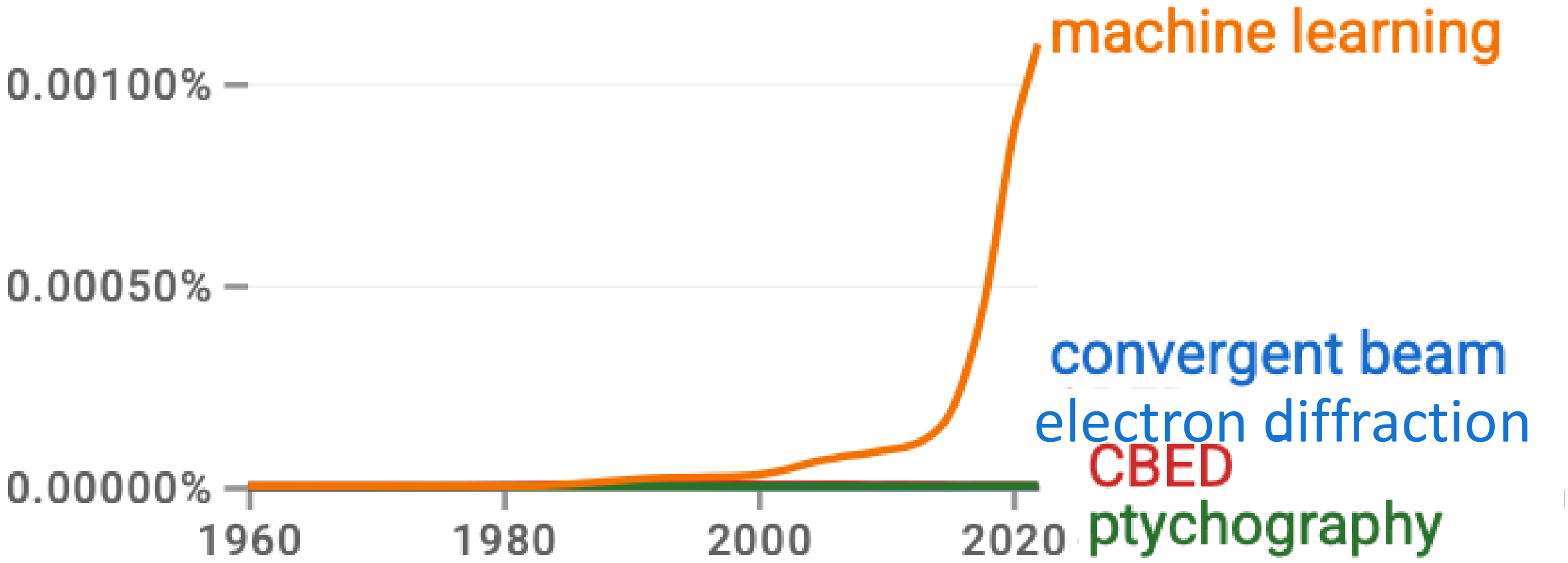


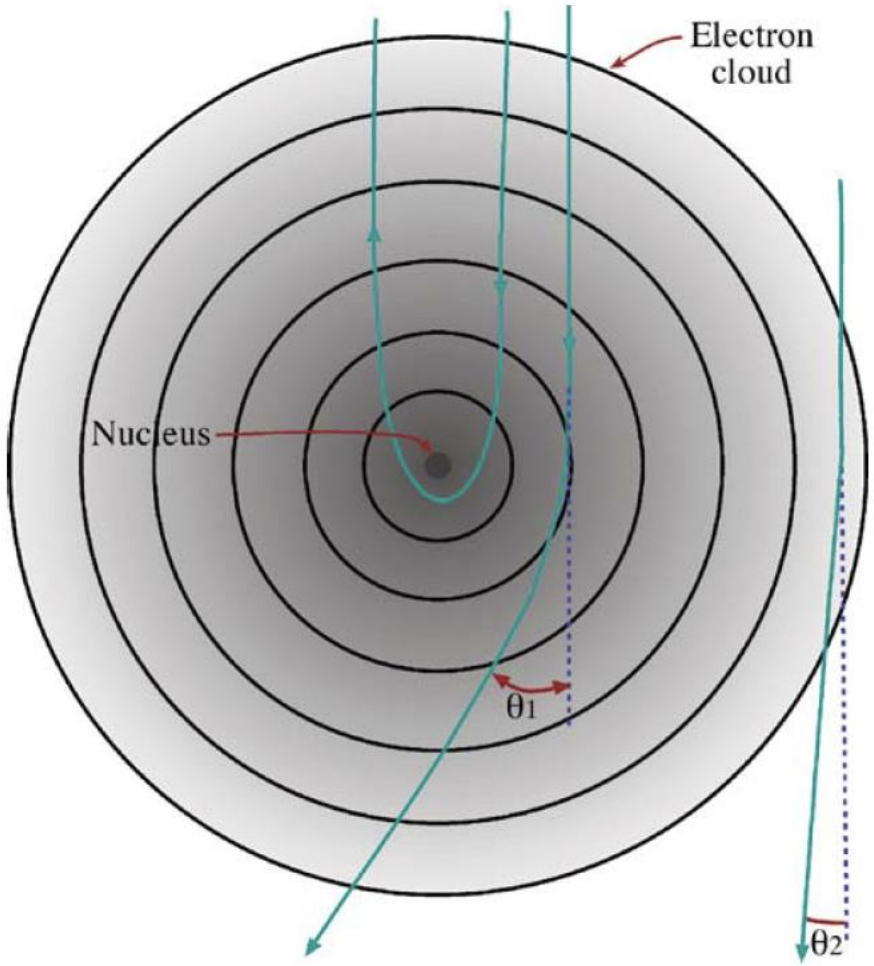
FIGURE 21.17 Nanodiffraction patterns from a multi-walled carbon nanotube (top).

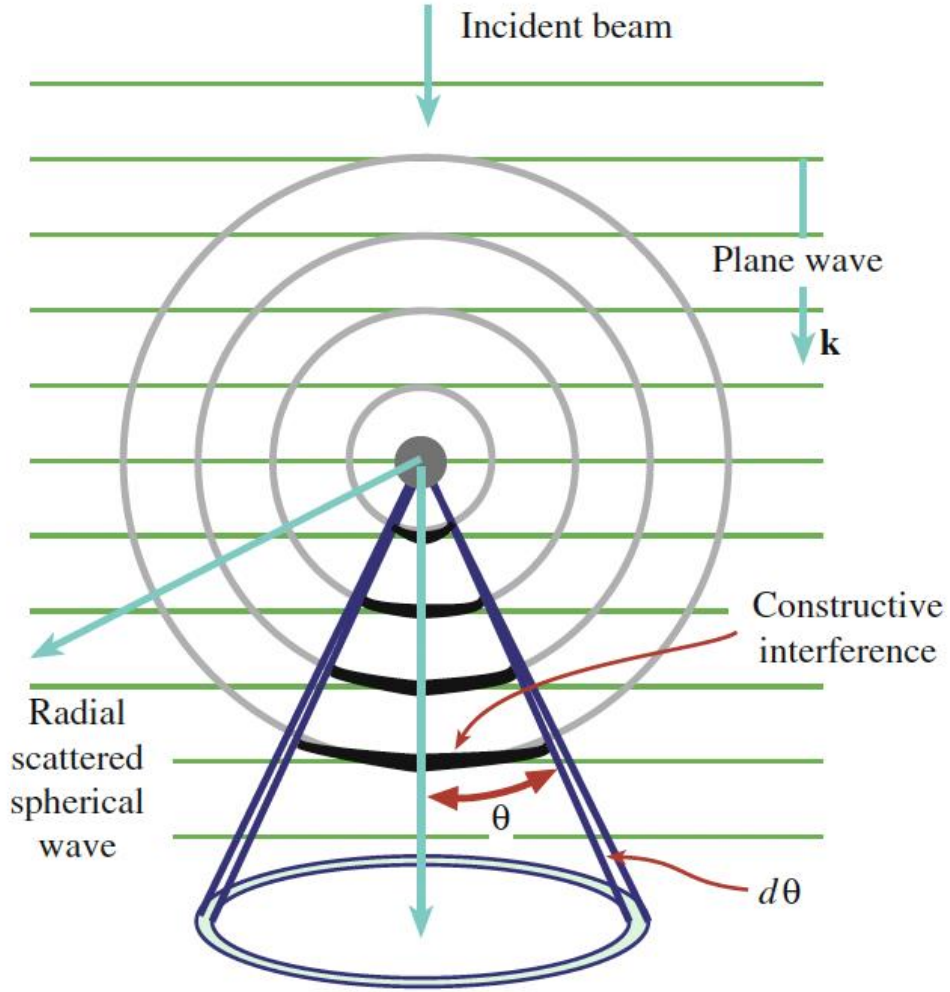
Things have changed…



Google Books Ngram Viewer

Basics



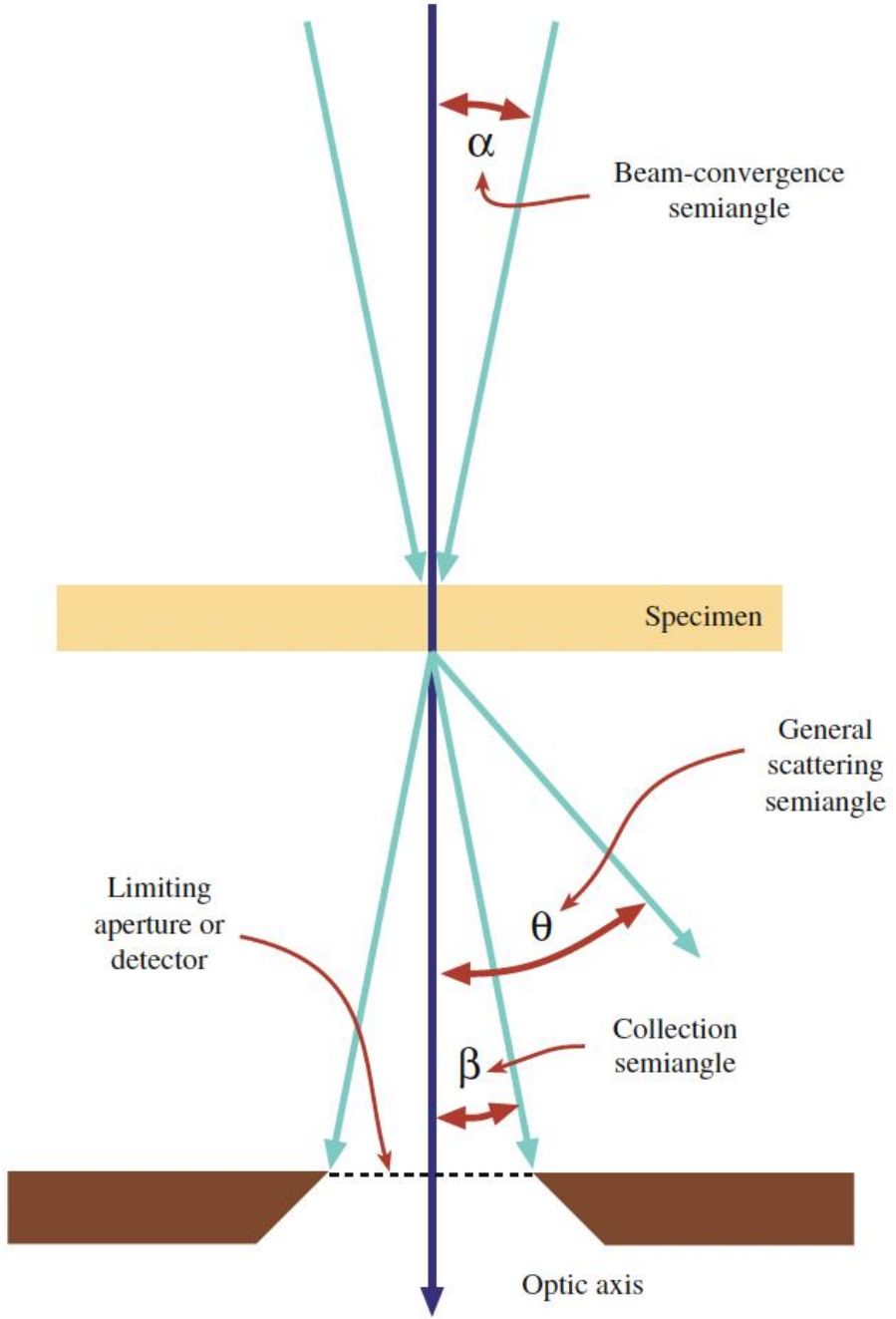


Variables

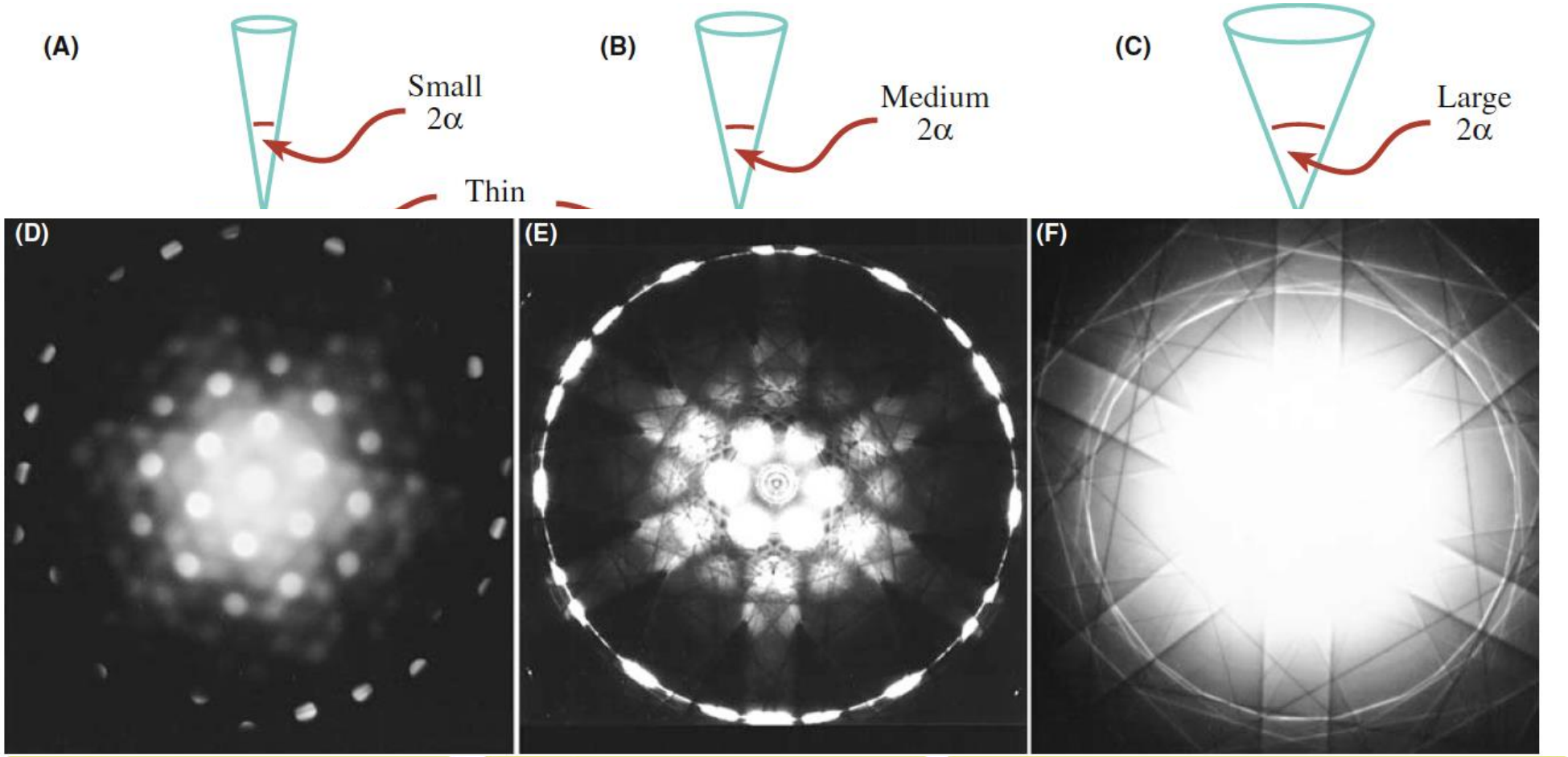
To form CBED patterns, you have to create a small (< 100 nm) beam with a convergence semi-angle (α) $\geq$10 mrads. There are at least five microscope variables you need to control when forming a CBED pattern

- The beam-convergence angle α (remember, we mean semi-angle)
- The camera length L (i.e., the magnification of the pattern)
- The size of the beam (the probe diameter)
- The thickness of the specimen
- The focus of the pattern (under/over or exact)

The last variable is the most complex because there are CBED techniques which require focused patterns and those where we deliberately defocus the beam out of the specimen plane to gain other advantages. So we'll deal with this last variable separately. You can also change the kV if you want and as we noted above, low kV gives better contrast, but this is more than offset by the drop in gun brightness and the increased beam spreading. So, for materials specimens, most CBED is done at the highest kV, as with most other TEM techniques.



Variable: Convergence angle



Variable: Camera Length

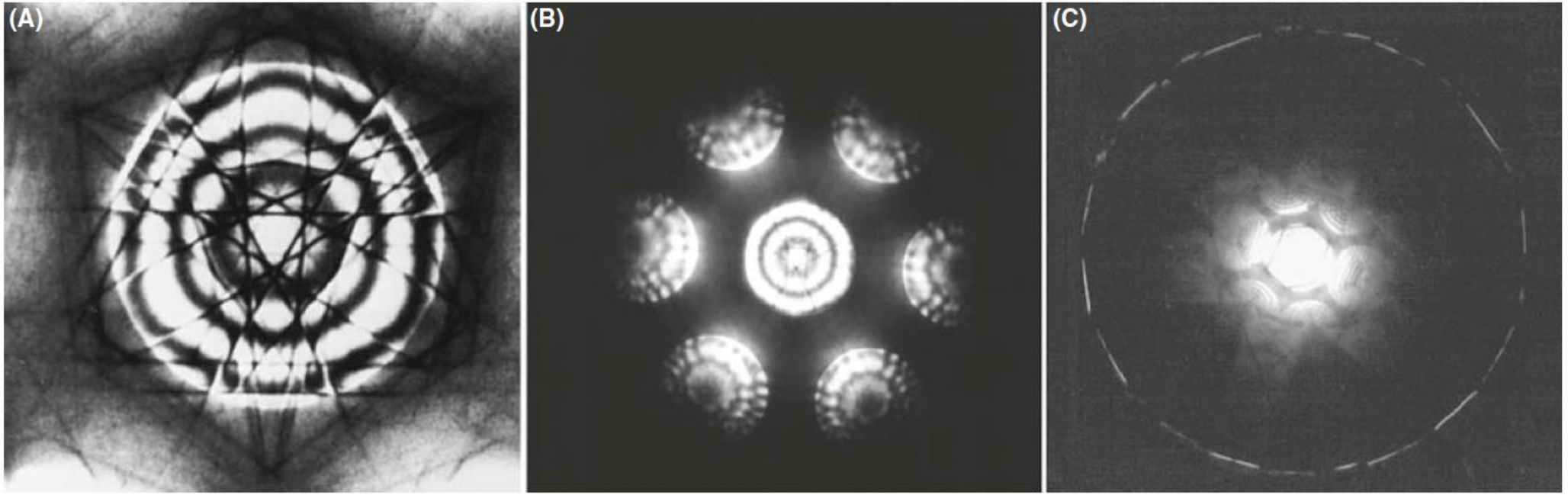


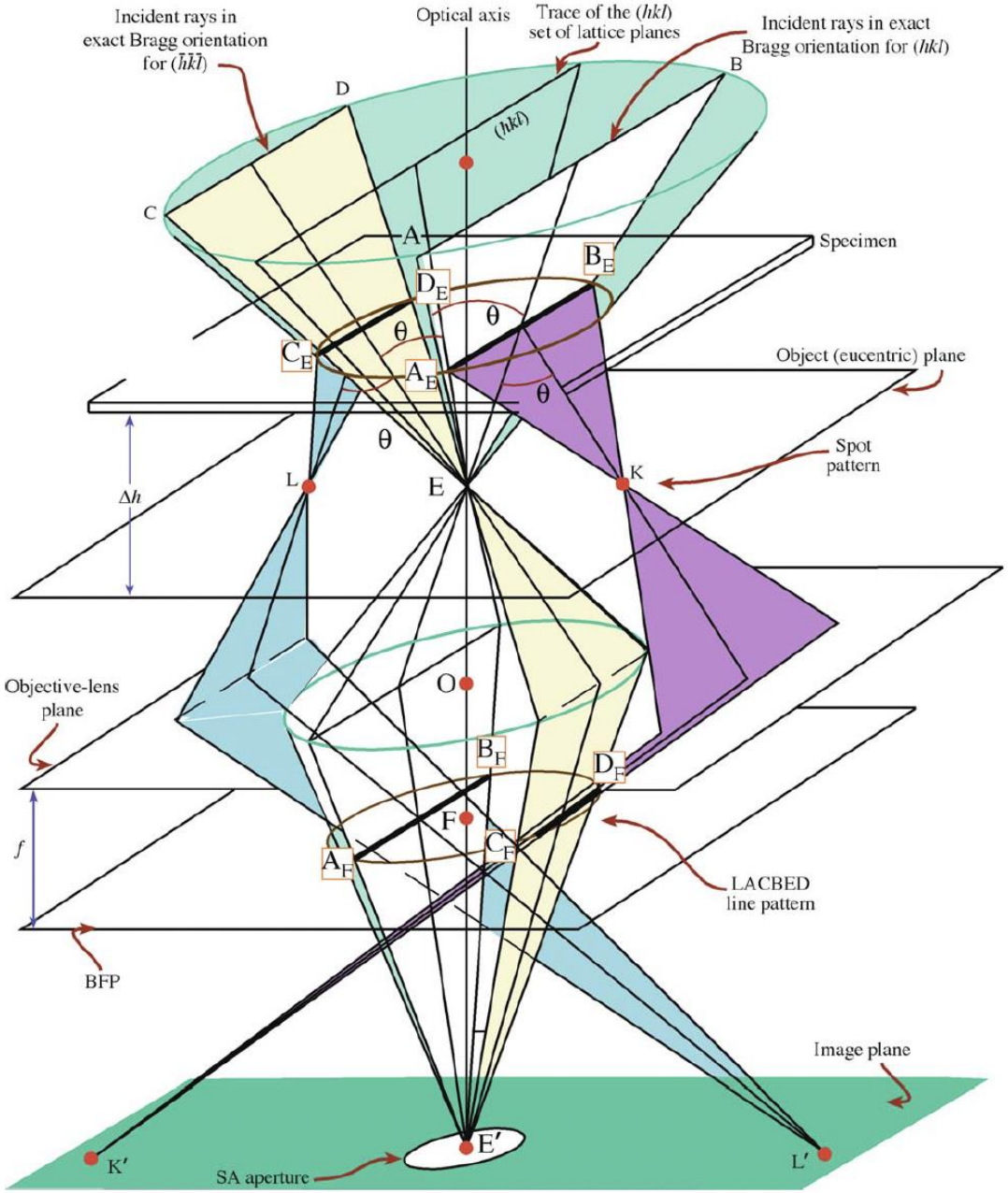
FIGURE 20.4. *Decreasing* the camera length, *L*, *increases* our view of reciprocal space. (A) Starting at high *L*, we see a CBED pattern containing only the 000 diffraction disk. As *L* decreases, we see in (B) the distribution of electrons in the ZOLZ, similar to a typical SADP. At the shortest camera length, (C) a ring of HOLZ intensity is faintly visible surrounding the bright ZOLZ disks. Typically, we can record electrons scattered over an angular range of $\pm 10^{\circ}$.

Simplify

Knowing everything we know about CBED patterns…

4D STEM can mean any of these techniques, as a function of (x,y) across the sample

And more!!

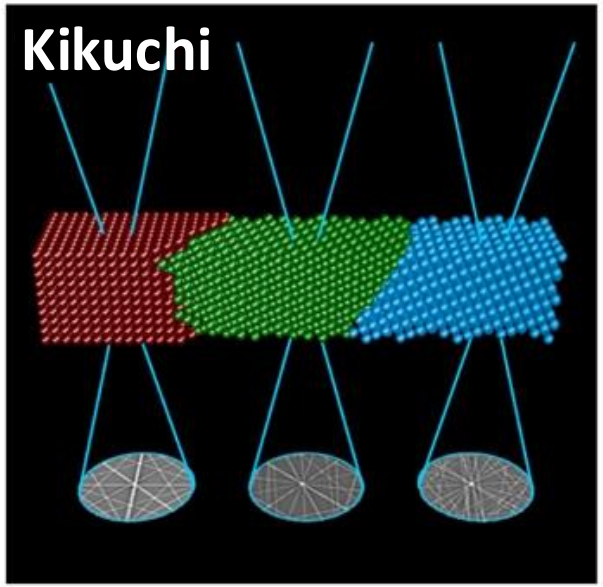


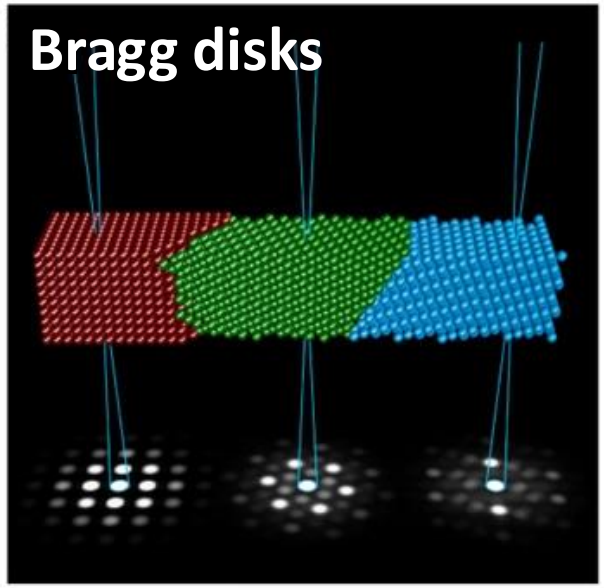
4D STEM - names

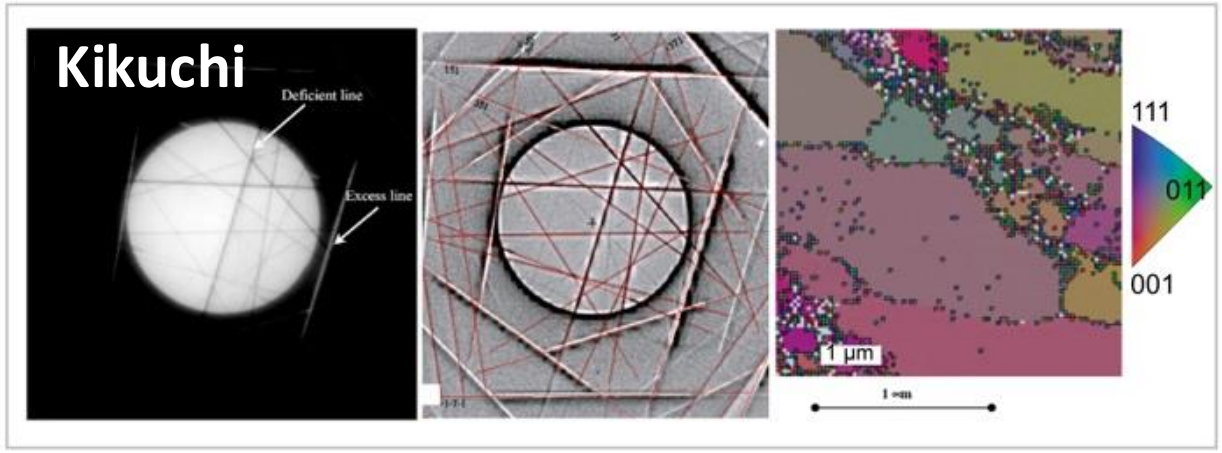
- position resolved diffraction (PRD), Zaluzec 2002
- spatially resolved diffractometry, Kimoto & Ishizuka 2011
- momentum-resolved STEM, Müller-Caspary 2018
- scanning electron nanodiffraction, Tao 2009
- nanobeam electron diffraction (NBED), Clément 2004
- pixelated STEM, MacArthur 2013
- universal detector, Hachtel 2018

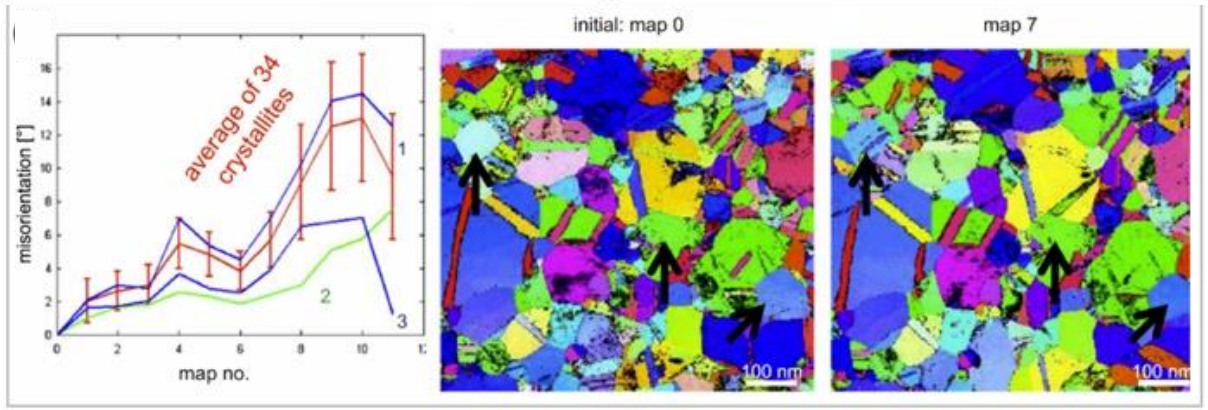
Colin Ophus, Four-Dimensional Scanning Transmission Electron Microscopy (4D-STEM): From Scanning Nanodiffraction to Ptychography and Beyond, *Microscopy and Microanalysis*, Volume 25, Issue 3, 1 June 2019

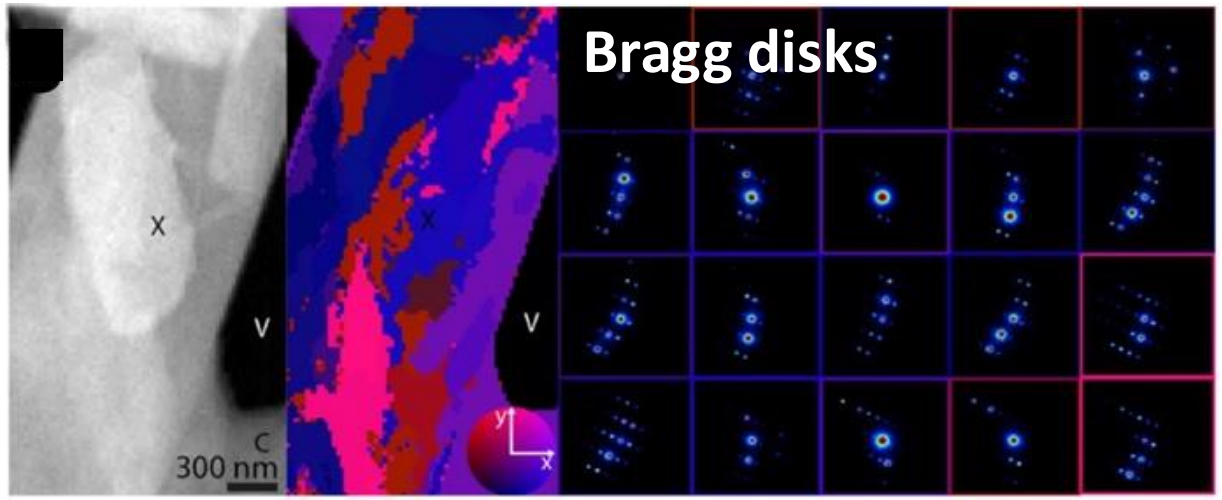
Orientation mapping in 4D-STEM





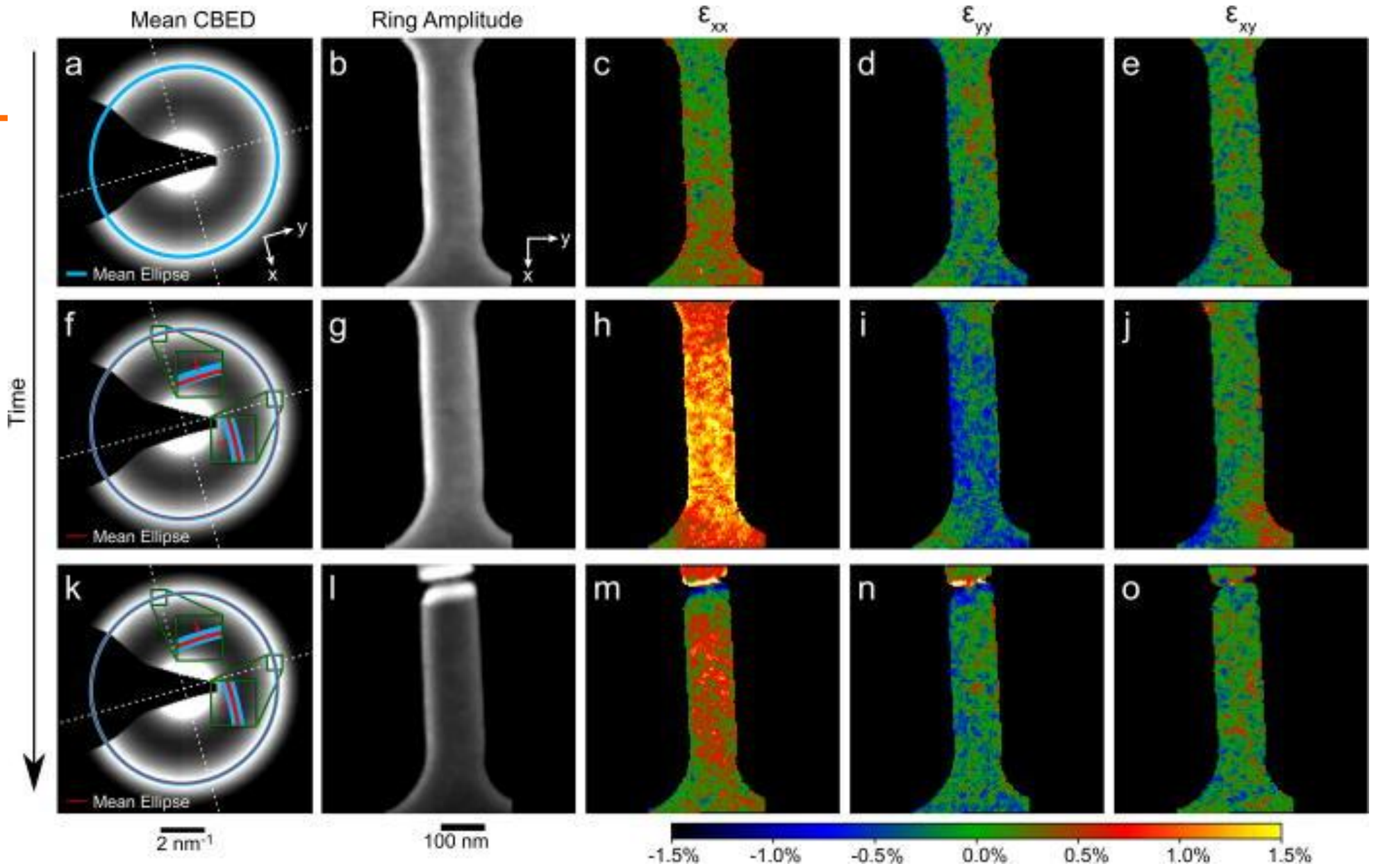






Colin Ophus, Four-Dimensional Scanning Transmission Electron Microscopy (4D-STEM): From Scanning Nanodiffraction to Ptychography and Beyond, *Microscopy and Microanalysis*, Volume 25, Issue 3, 1 June 2019

Strain

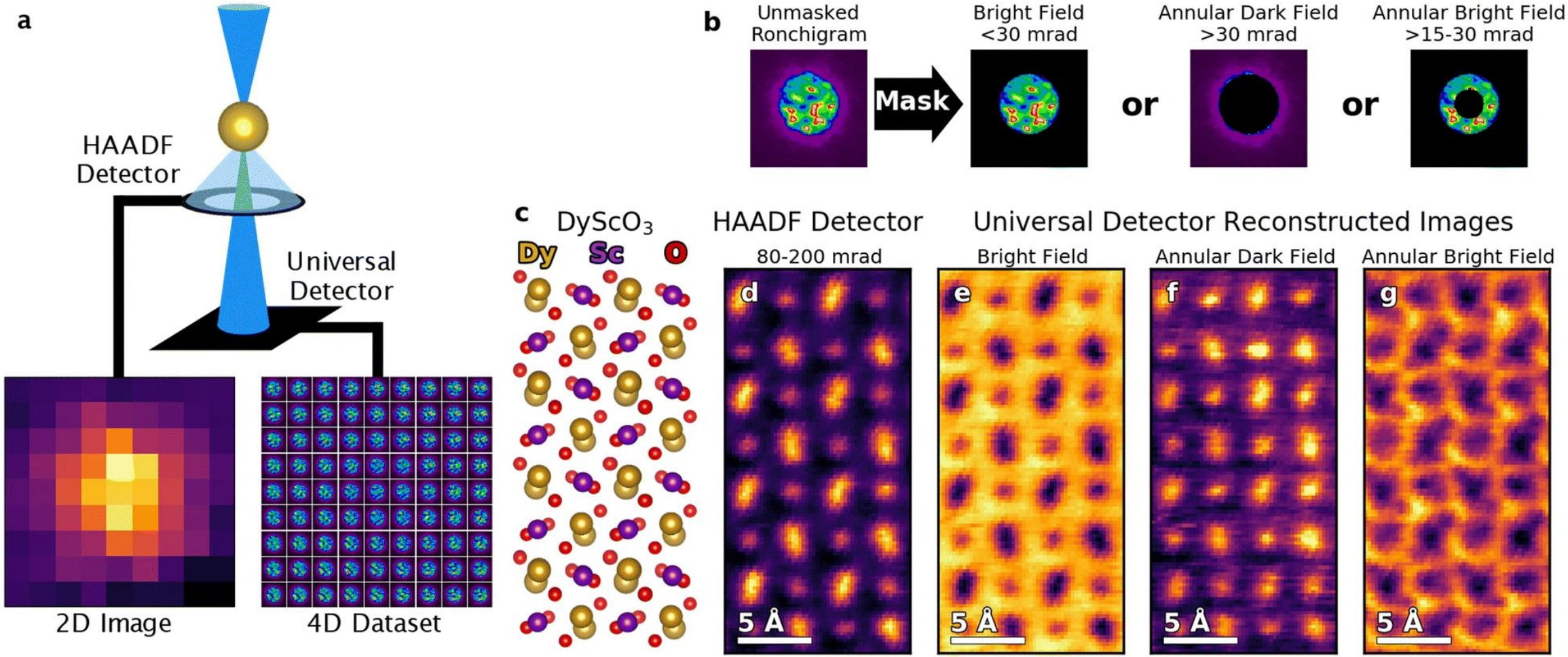


Christoph Gammer, Colin Ophus, Thomas C. Pekin, Jürgen Eckert, Andrew M. Minor; Local nanoscale strain mapping of a metallic glass during *in situ* testing. *Appl. Phys. Lett.* 23 April 2018

“more than the sum of the parts”

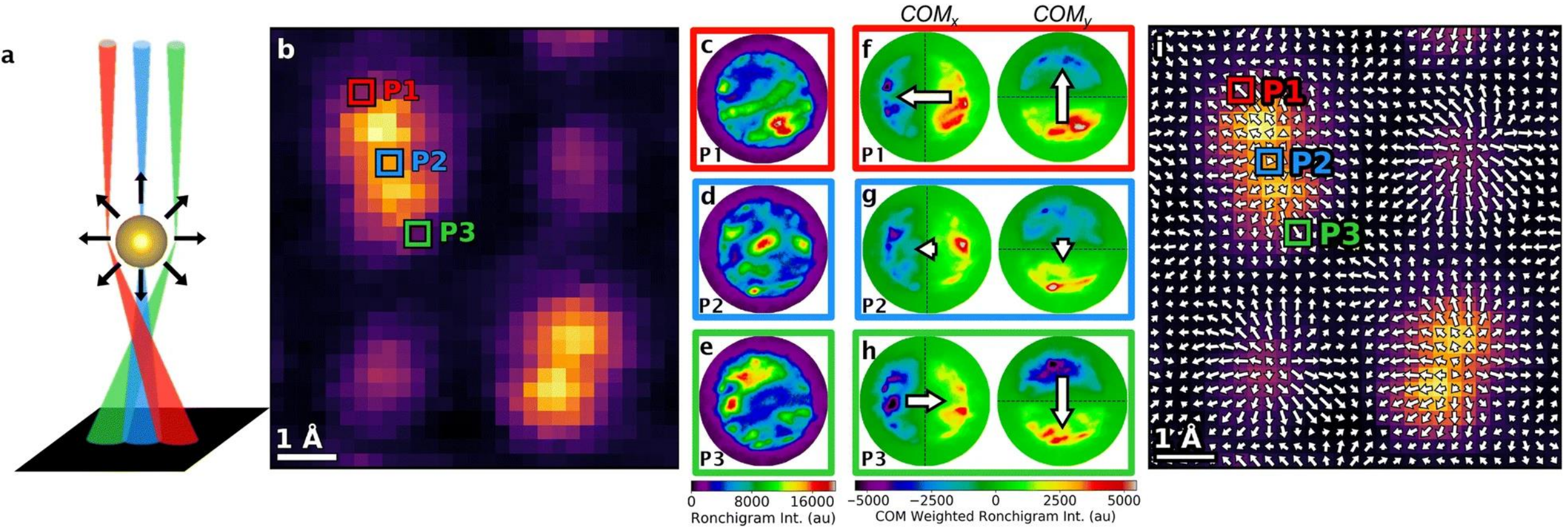
THINKING OUTSIDE THE HYPERCUBE

Virtual imaging



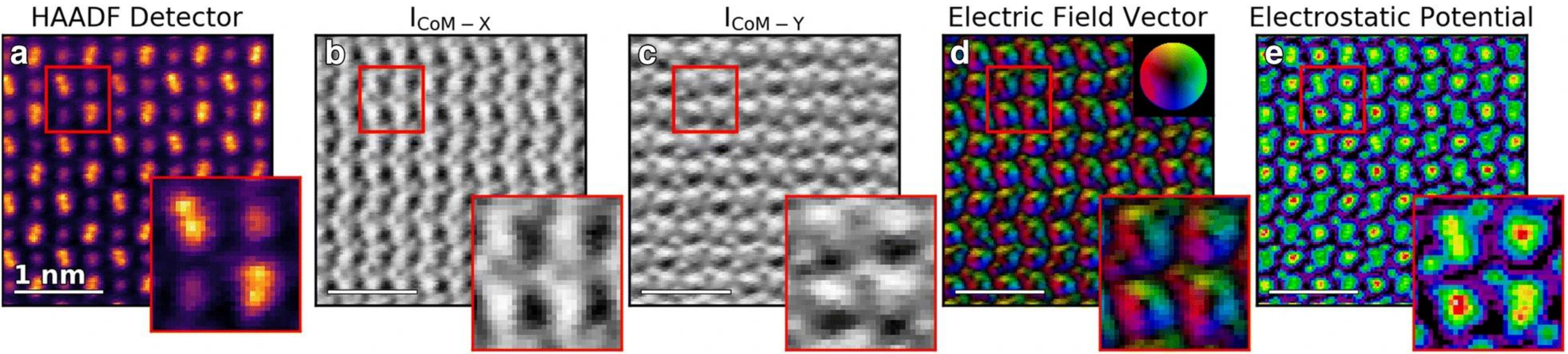
Hachtel, J.A., Idrobo, J.C. & Chi, M. Sub-Ångstrom electric field measurements on a universal detector in a scanning transmission electron microscope. *Adv Struct Chem Imag***4**, 10 (2018).

COM and I_{COM} Imaging



Hachtel, J.A., Idrobo, J.C. & Chi, M. Sub-Ångstrom electric field measurements on a universal detector in a scanning transmission electron microscope. *Adv Struct Chem Imag***4**, 10 (2018).

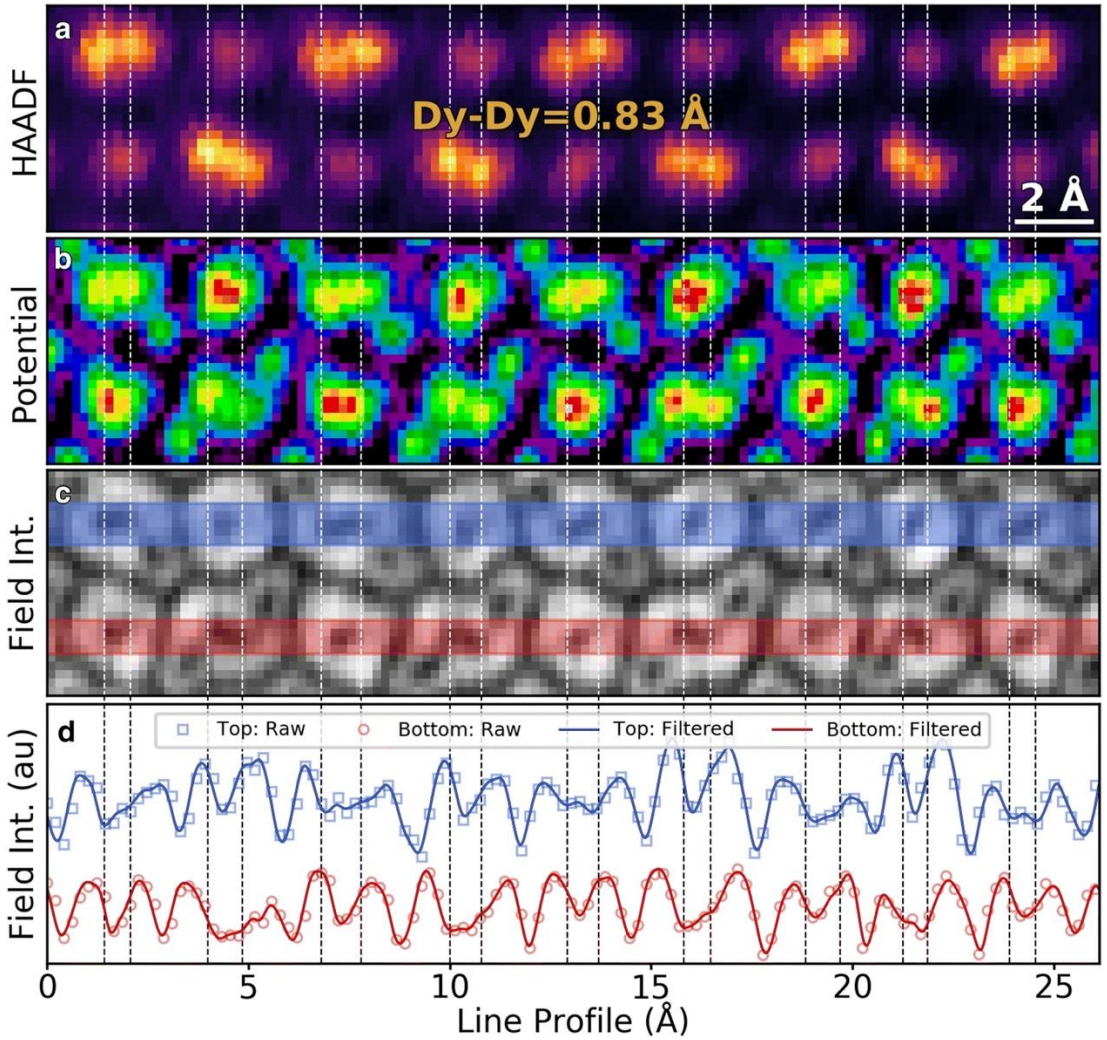
Imaging Electrostatic Potential



Look into this slide more

Hachtel, J.A., Idrobo, J.C. & Chi, M. Sub-Ångstrom electric field measurements on a universal detector in a scanning transmission electron microscope. *Adv Struct Chem Imag***4**, 10 (2018).

Sub-Å Electric Field Measurement

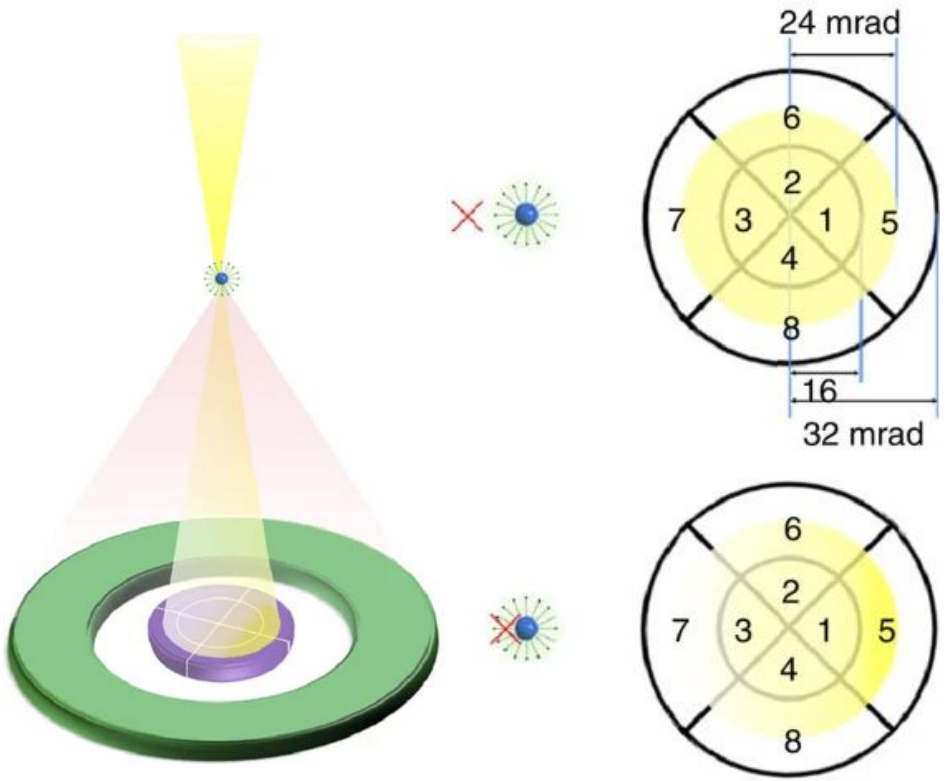


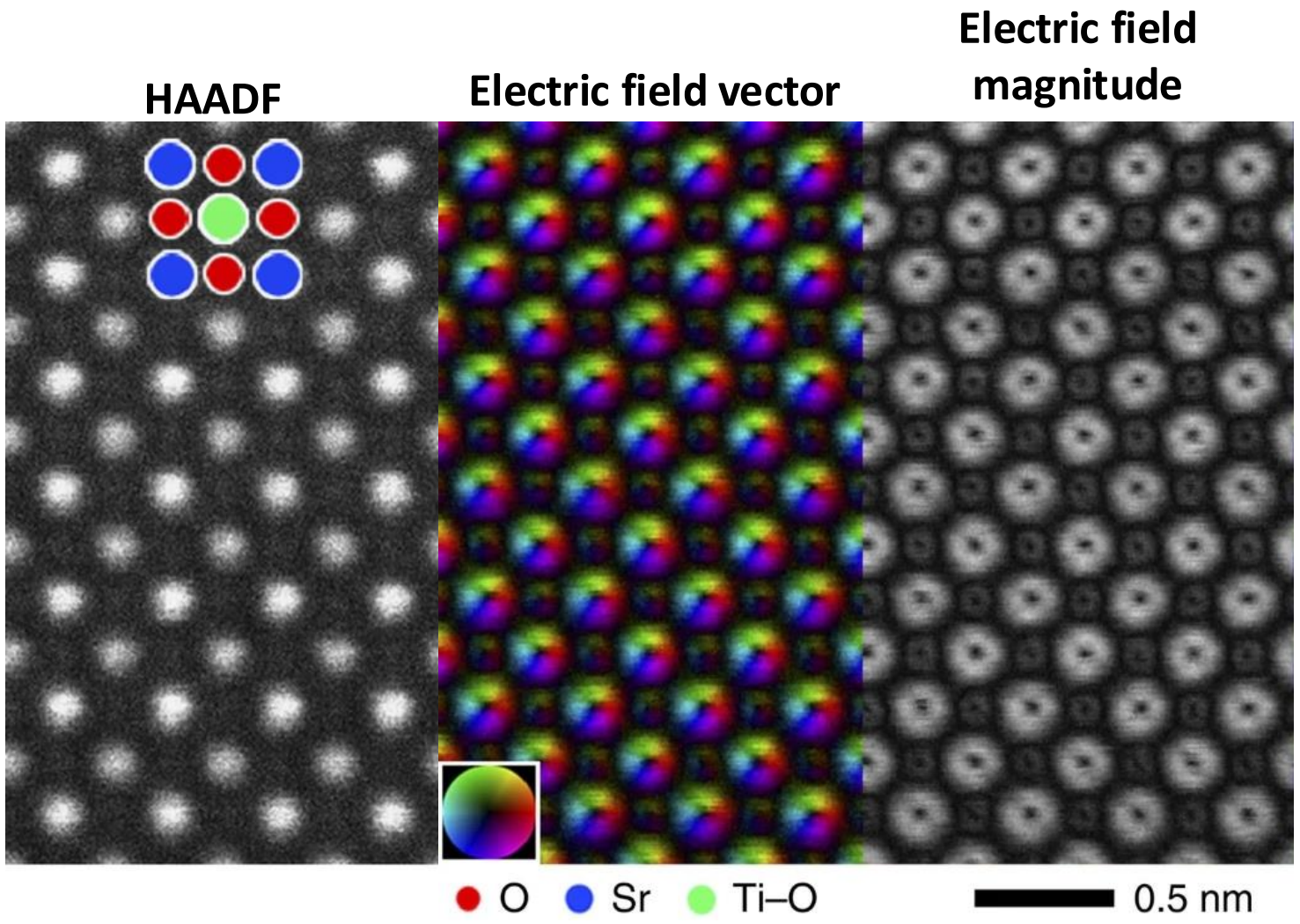
Hachtel, J.A., Idrobo, J.C. & Chi, M. Sub-Ångstrom electric field measurements on a universal detector in a scanning transmission electron microscope. *Adv Struct Chem Imag***4**, 10 (2018).

Integrated differential phase contrast

IDPC

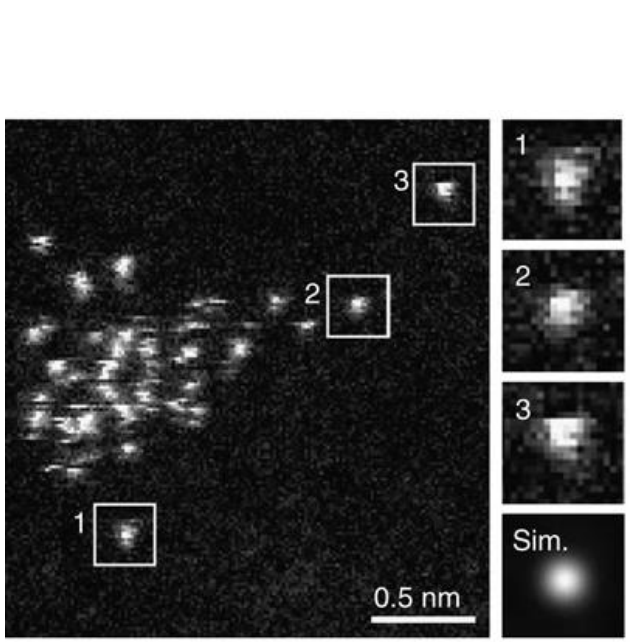
Single Atom Electric Fields

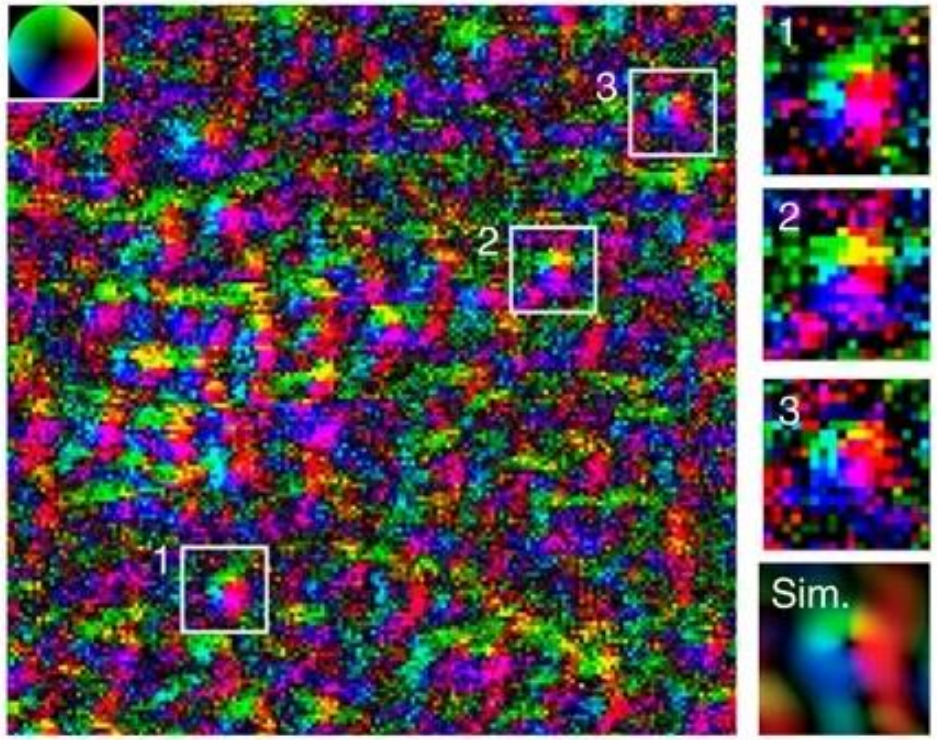


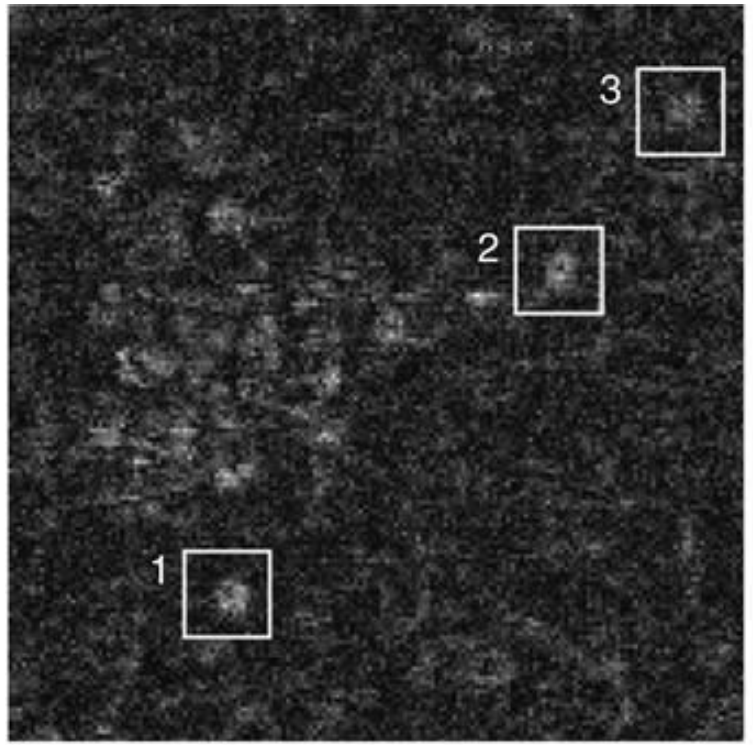


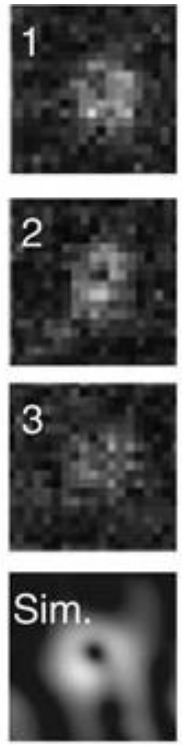
Shibata, N., Seki, T., Sánchez-Santolino, G. *et al.* Electric field imaging of single atoms. *Nat Commun* **8**, 15631 (2017)

Single Atom Electric Fields



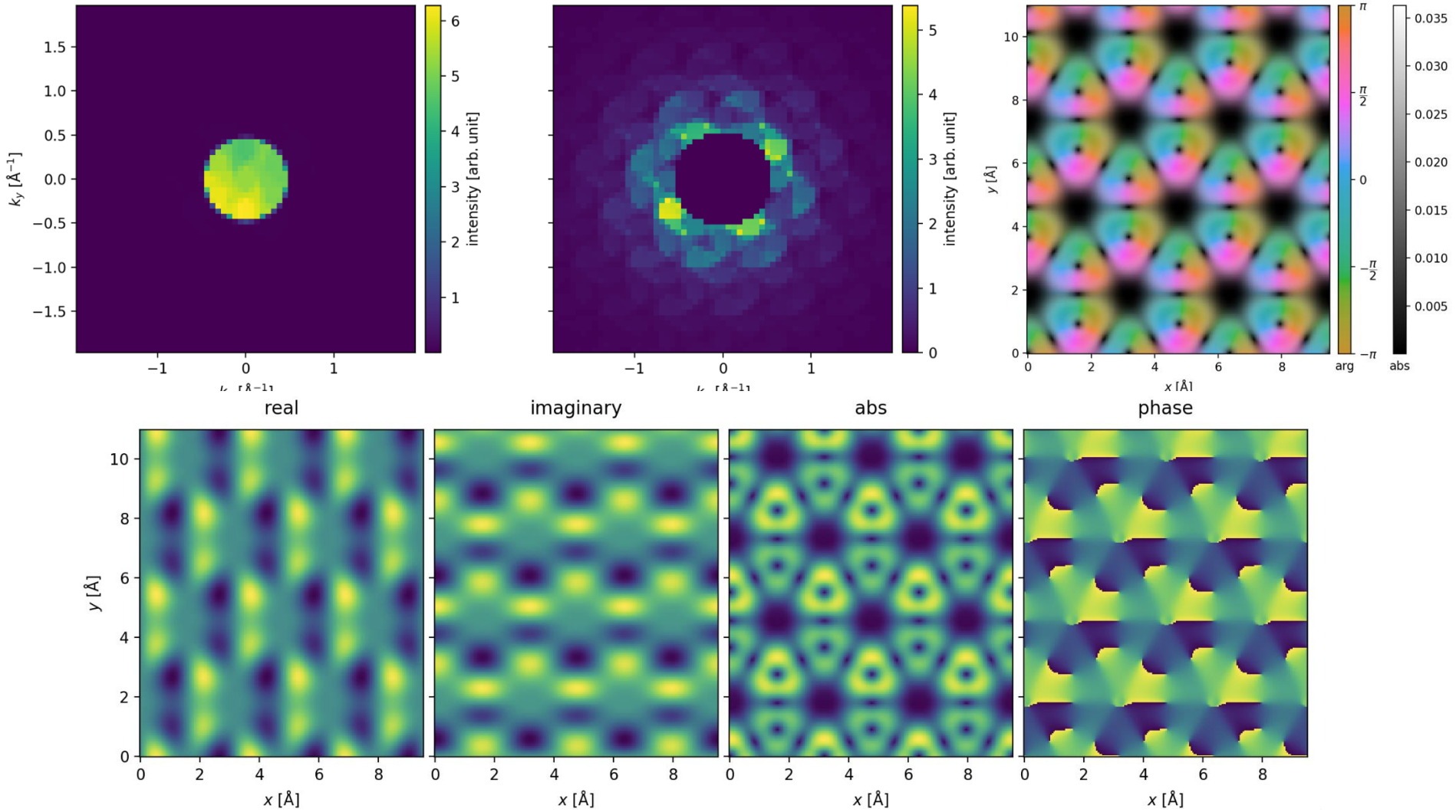






Shibata, N., Seki, T., Sánchez-Santolino, G. *et al.* Electric field imaging of single atoms. *Nat Commun* **8**, 15631 (2017)

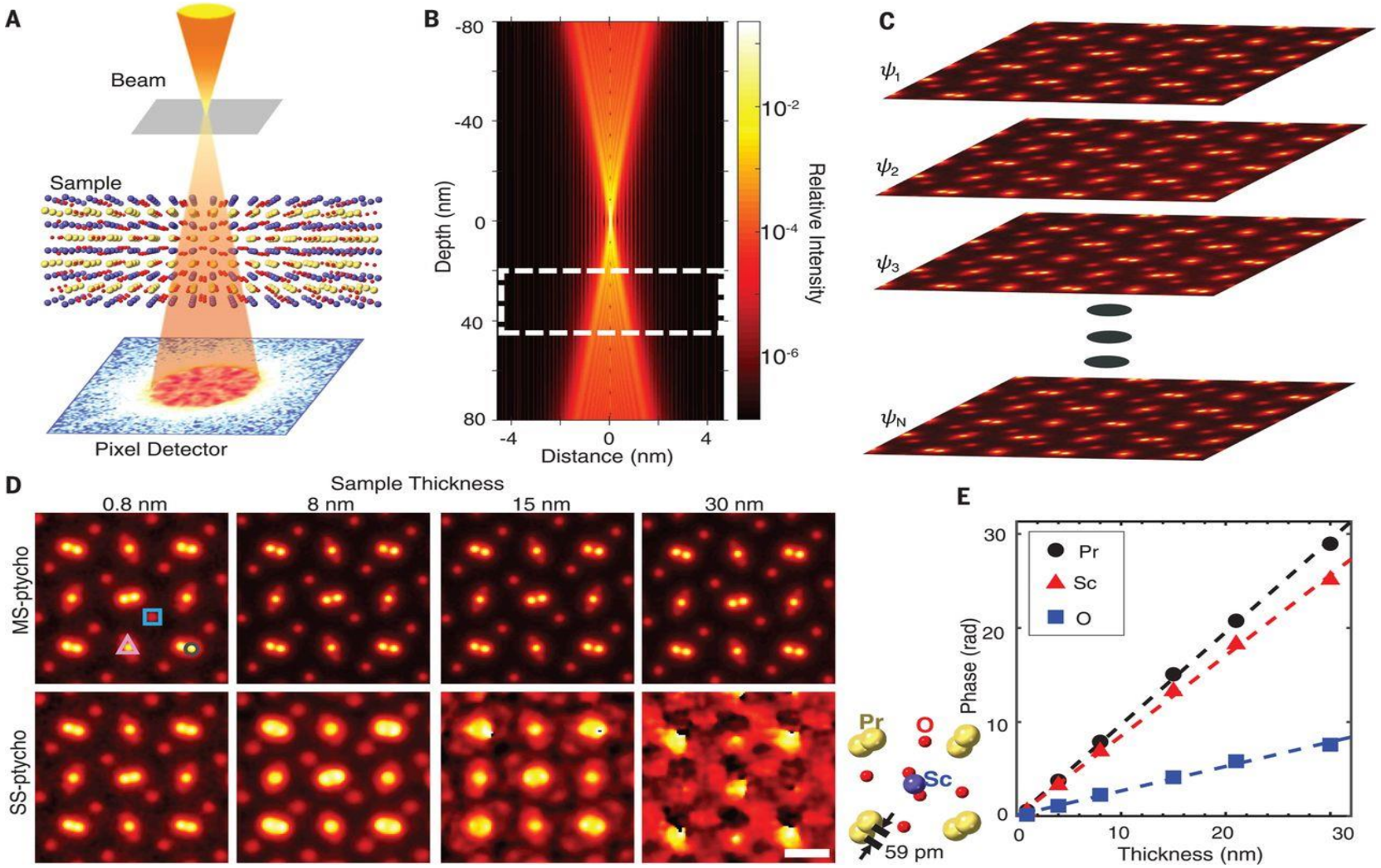
Notebook: iDPC simulation



Final topic

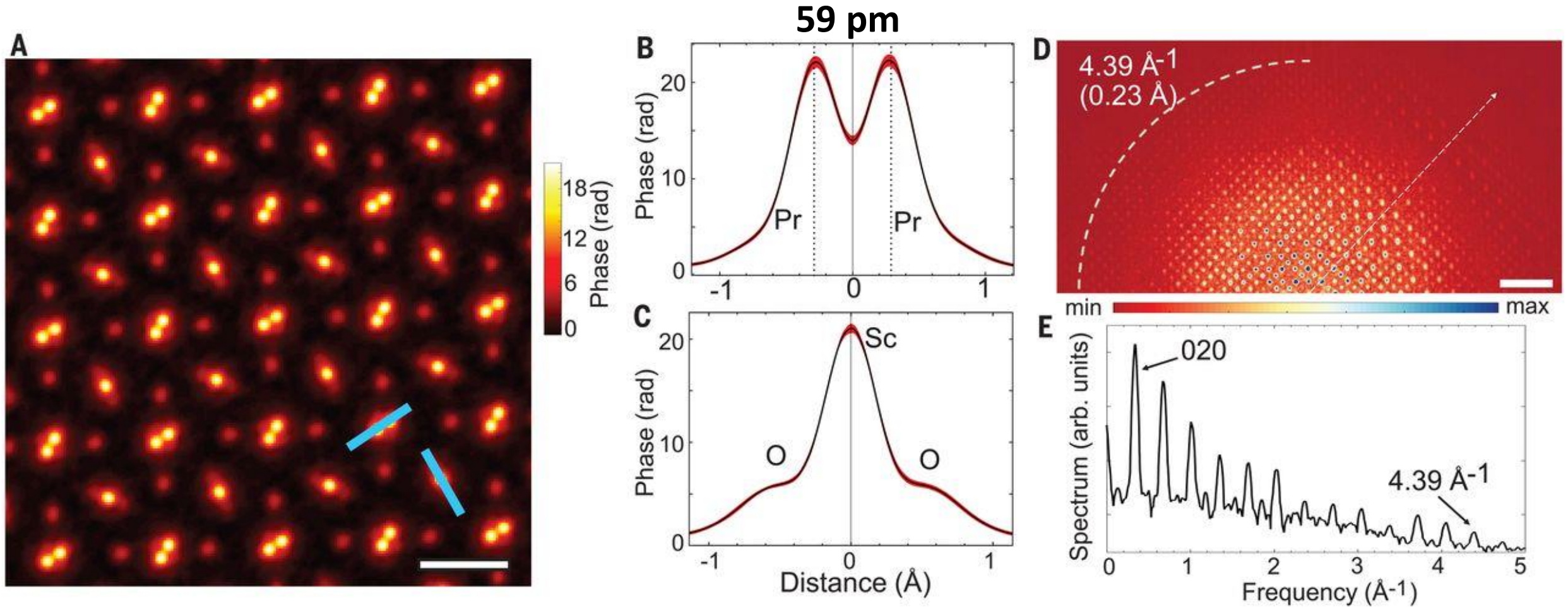
PTYCHOGRAPHY

Multislice Electron Ptychography



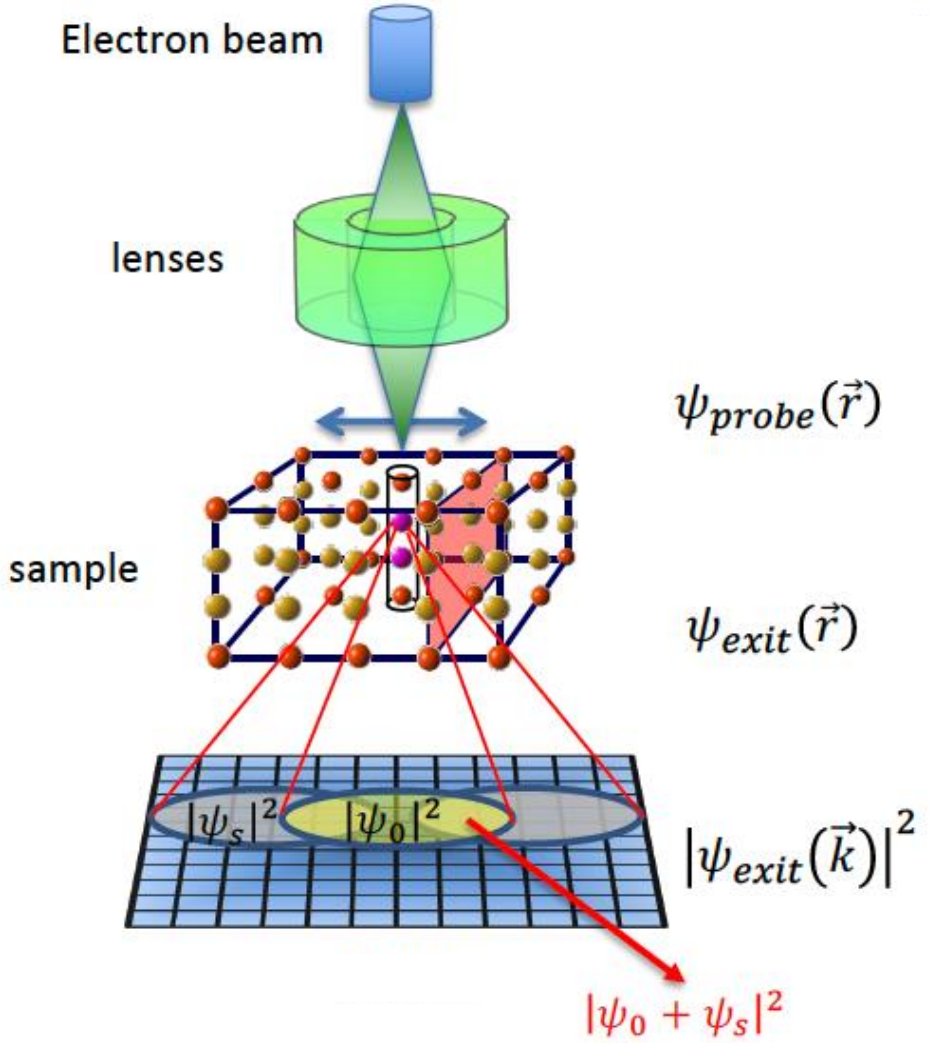
Zhen Chen *et al.,* Electron ptychography achieves atomic-resolution limits set by lattice vibrations.*Science***372**,826-831(2021)

Reconstruction of PrScO3



Zhen Chen *et al.,* Electron ptychography achieves atomic-resolution limits set by lattice vibrations.*Science***372**,826-831(2021)

Outline



Diffraction from an atomic-sized beam contains phase information in the overlapping beams.

The Strong Phase Approximation (SPA)

Model: $\psi(r) = \psi_0(r)\, e^{i\sigma V(r)}$

exit wave — Incident wave

$V(r)$ → specimen projected potential

σ → Interaction parameter σ(Beam Energy)

Probe shape does not change $|\psi(r)|^2 = |\psi_0(r)|^2$
Neglects beam spreading or propagation

Optimization Problem

- Step 1: Design a model + optimization problem
  - **Forward Model** (something that produces a diffraction pattern given a potential). e.g. : $I_{\vec{r}_p} = \left|\mathcal{F}\left[P\left(\vec{r}-\vec{r}_p\right)\cdot O\left(\vec{r}\right)\right]\right|^2$
  - **optimizer**: e.g. ePIE: A. M. Maiden et al. *Ultramicroscopy* **109,** 1256–1262 (2009)

$$\min_{P,O}\sum_{\vec{r}}\left|\sqrt{I_{\vec{r}_p}}-\left|\mathcal{F}\left[P\left(\vec{r}-\vec{r}_p\right)\cdot O\left(\vec{r}\right)\right]\right|\right|^2$$

probe object

measured diffraction pattern

- Step 2: find the designed solution w. iterative algorithms (solvers)

ePIE algorithm (extended Ptychographical Iterative Engine) is a widely used iterative solver that alternates updates to the probe and object using gradient-like rules

Intermission

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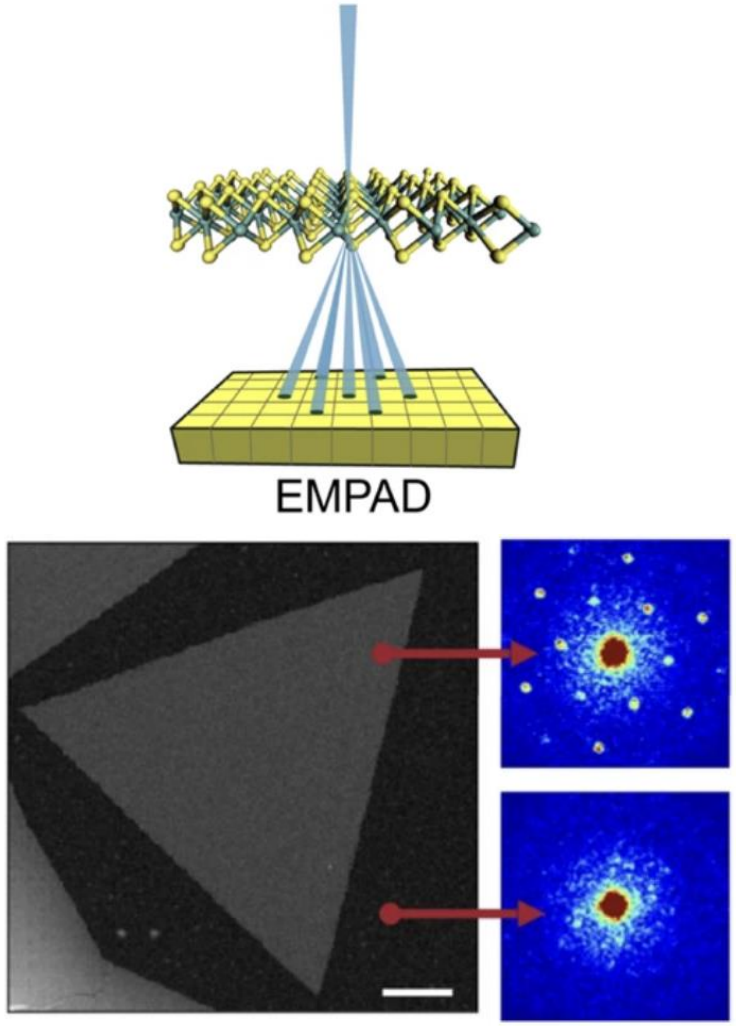
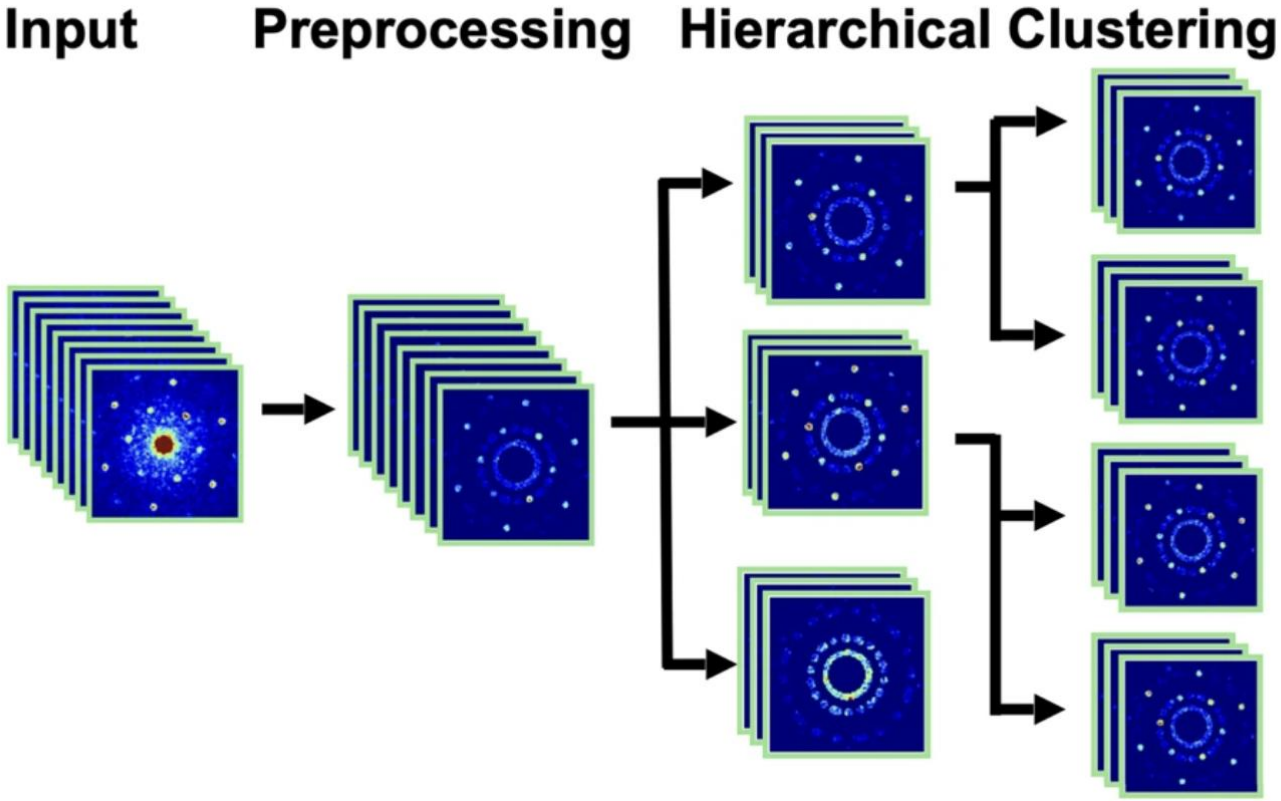
Let’s apply K-Means clustering

ML 4D STEM

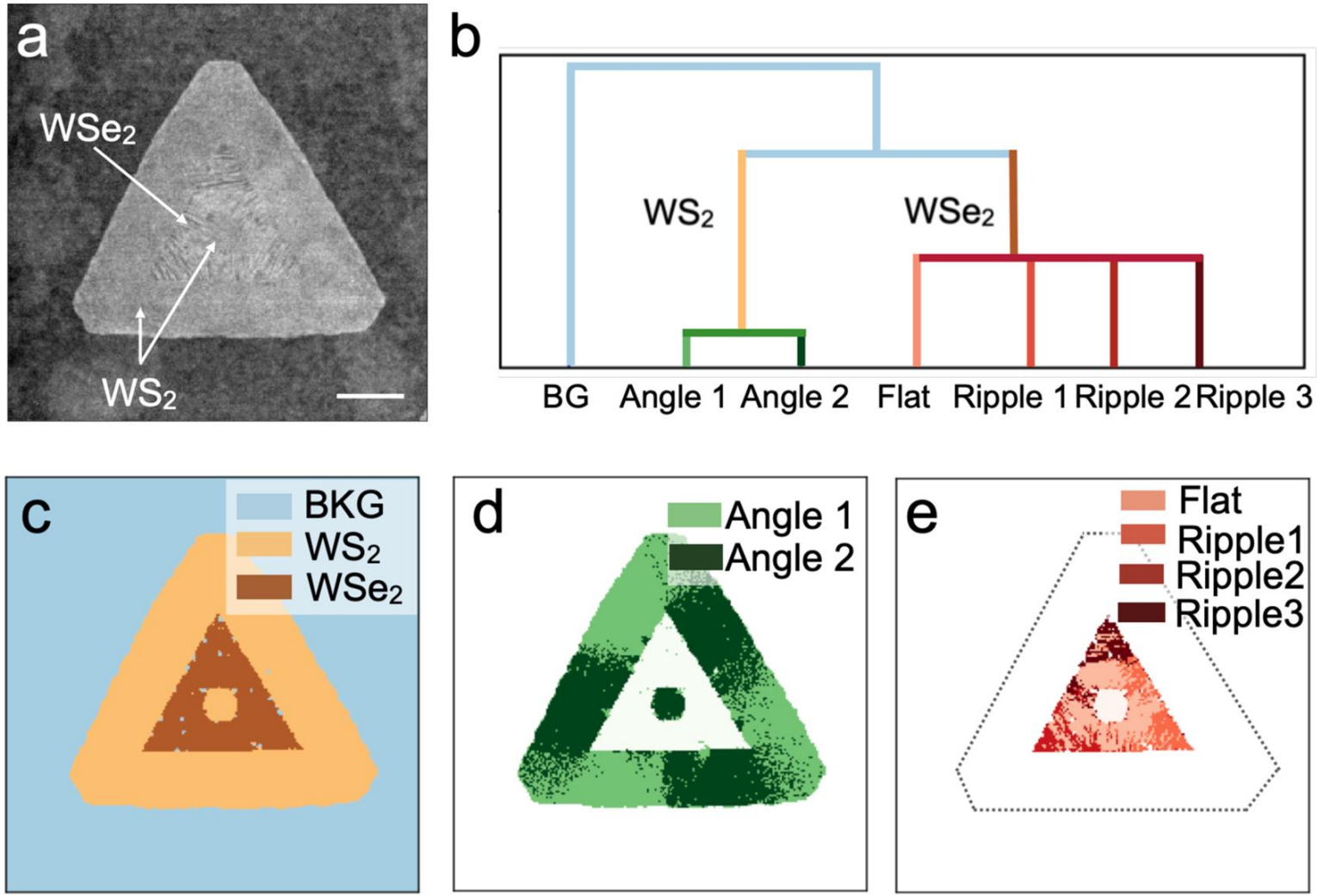
Hierarchical Clustering

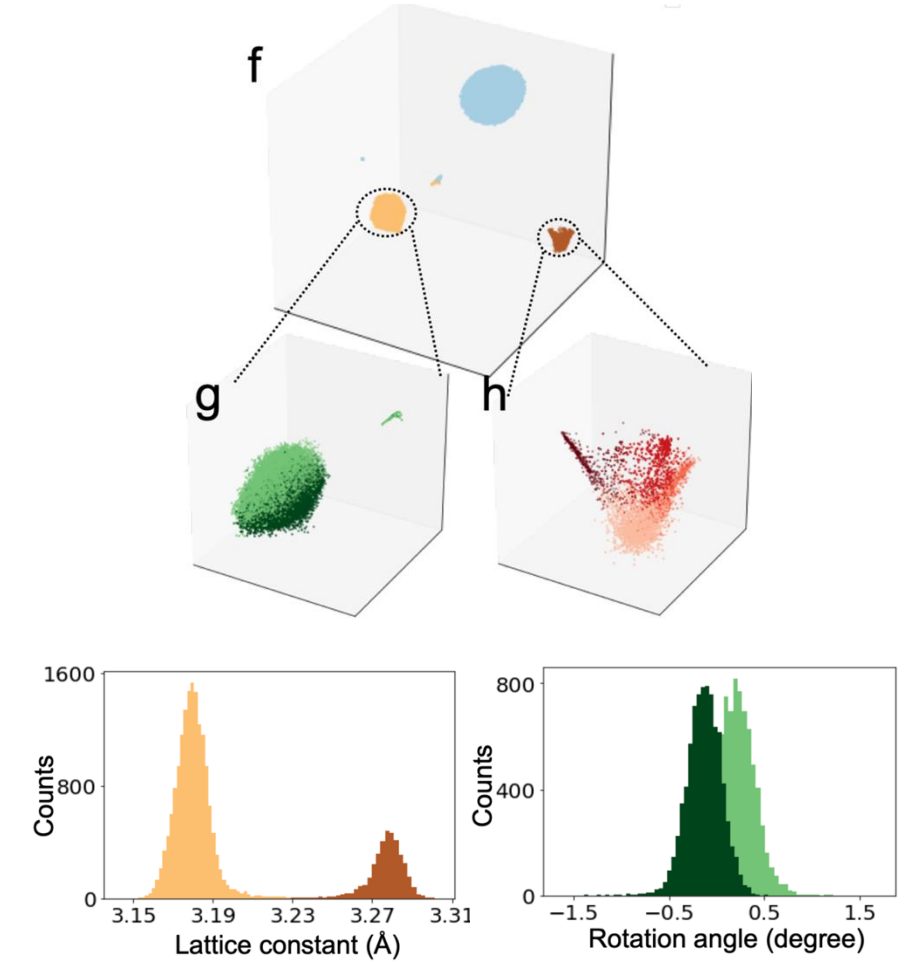




Shi, C., Cao, M.C., Rehn, S.M. *et al.* Uncovering material deformations via machine learning combined with four-dimensional scanning transmission electron microscopy. *npj Comput Mater* **8**, 114 (2022)

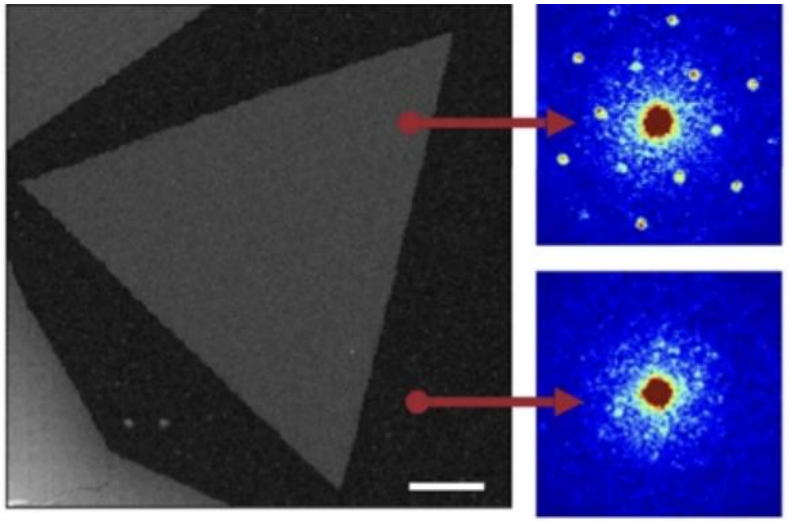
Hierarchical Clustering

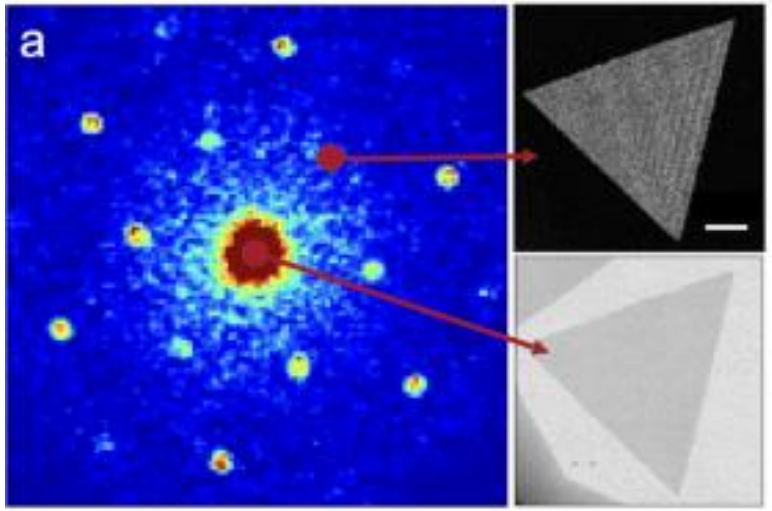


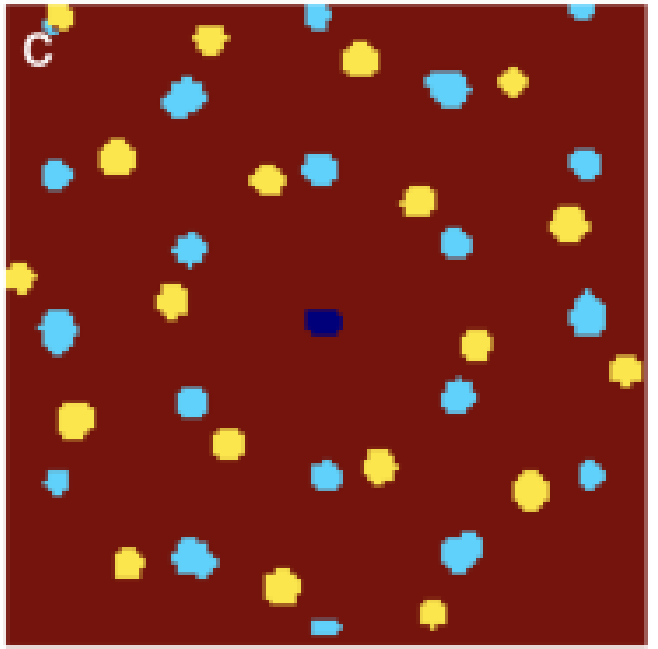


Shi, C., Cao, M.C., Rehn, S.M. *et al.* Uncovering material deformations via machine learning combined with four-dimensional scanning transmission electron microscopy. *npj Comput Mater* **8**, 114 (2022)

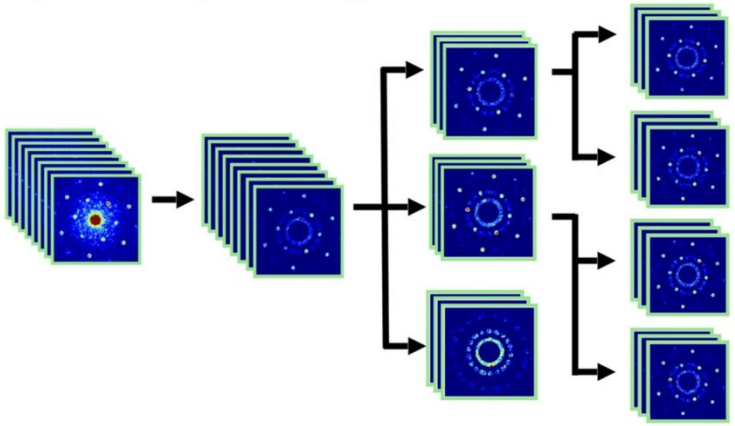
Creative Thinking



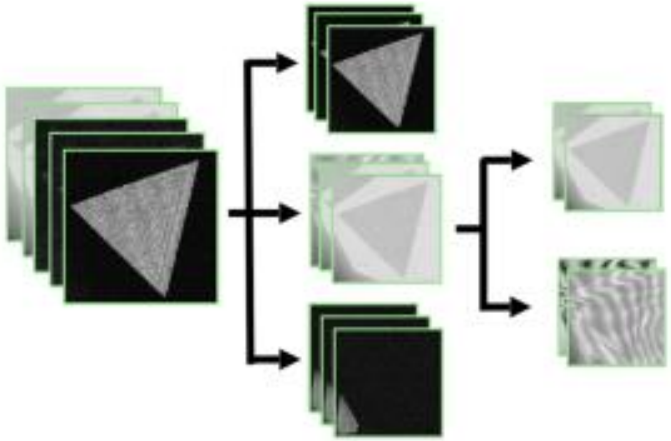


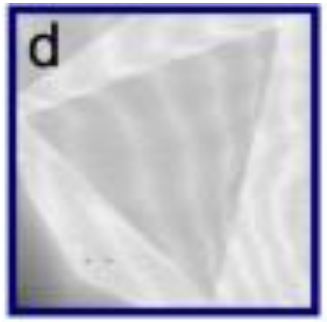


c Input Preprocessing Hierarchical Clustering



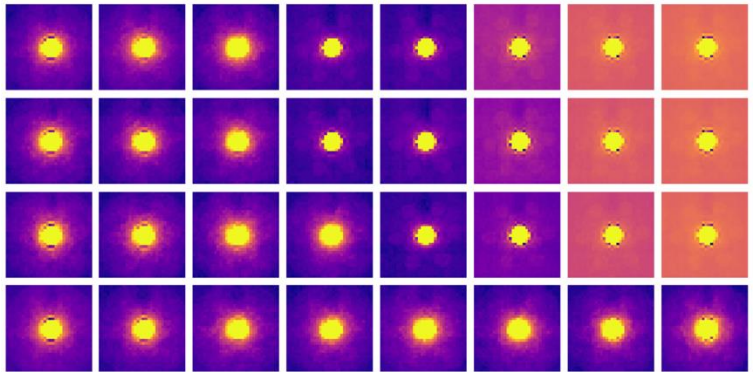
b Hierarchical Clustering in Real-Space Image

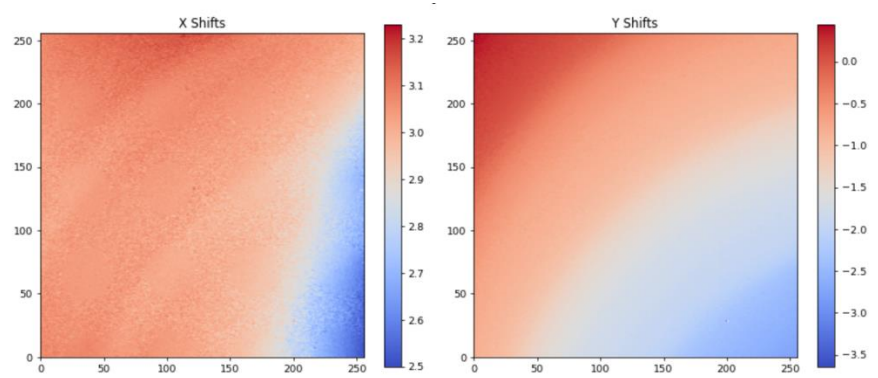


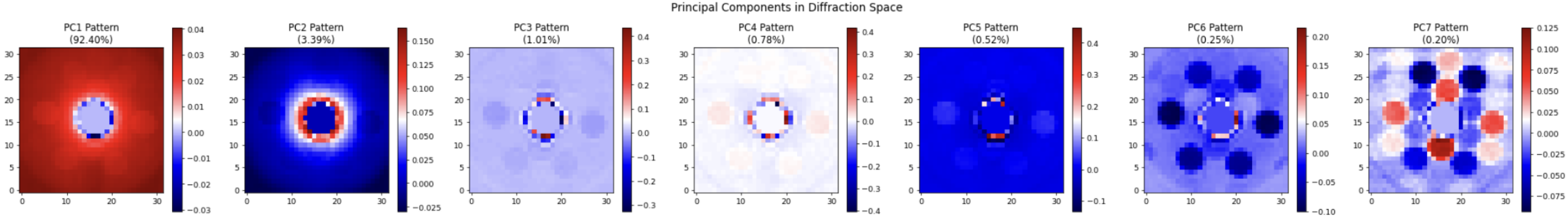


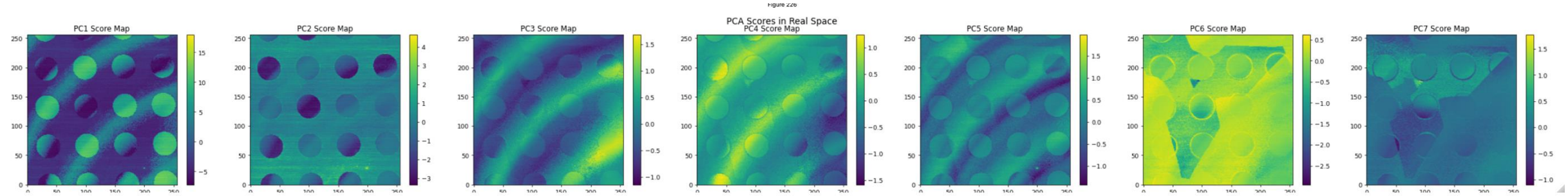
Shi, C., Cao, M.C., Rehn, S.M. *et al.* Uncovering material deformations via machine learning combined with four-dimensional scanning transmission electron microscopy. *npj Comput Mater* **8**, 114 (2022)

Notebook: negative example

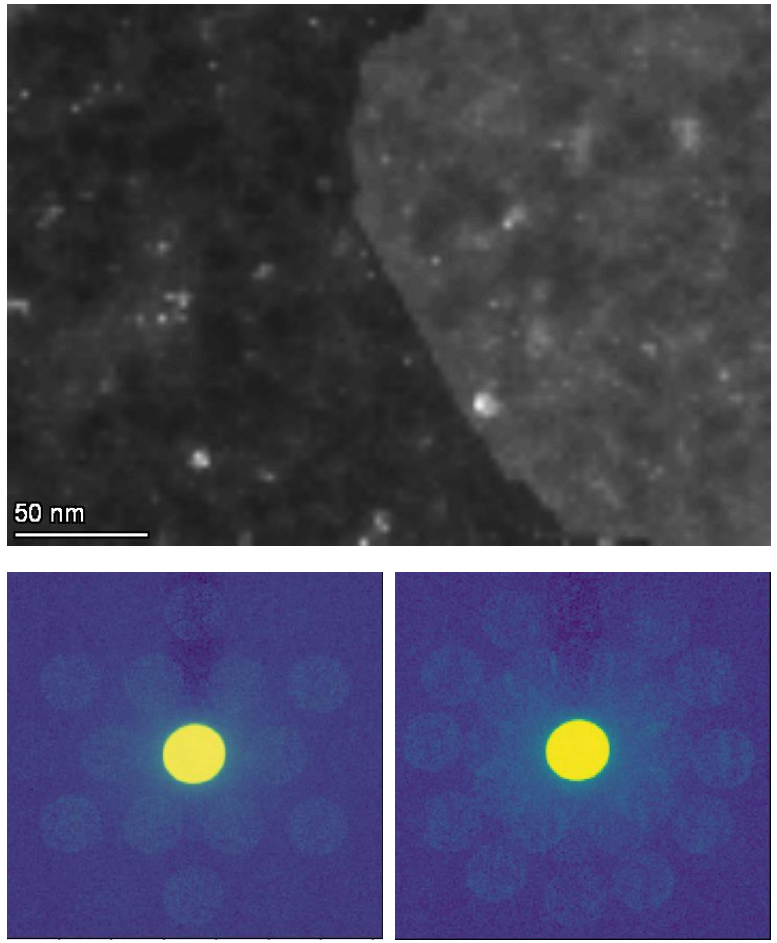


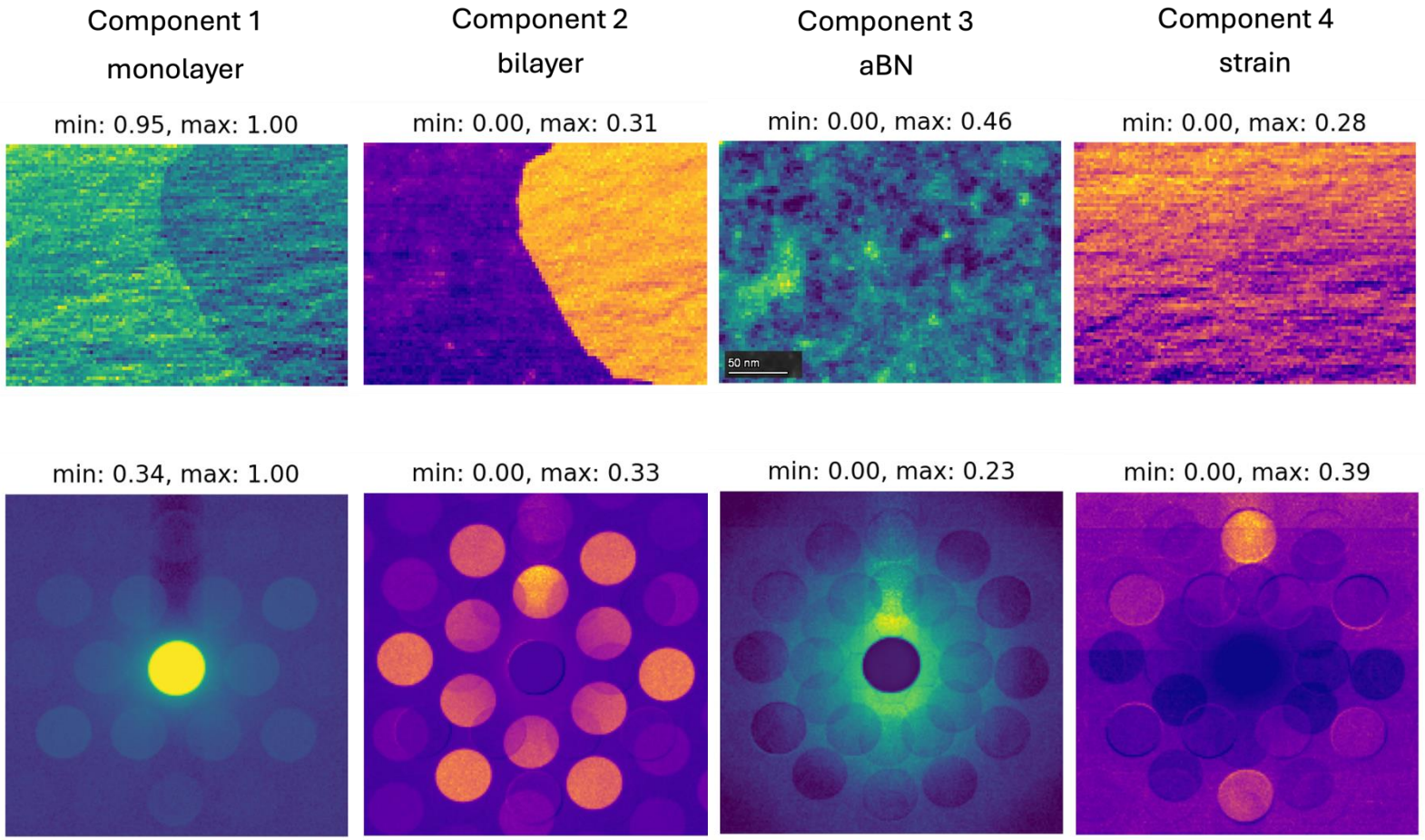






Notebook: positive example





Thank you!

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