Automated Experiments for STEM at CNMS

Andrew R Lupini

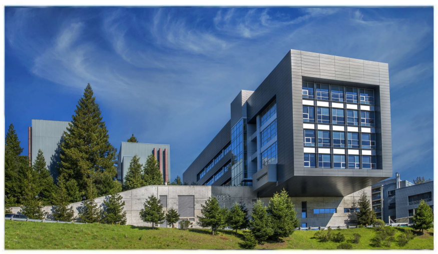
Center for Nanophase Materials Sciences

Oak Ridge National Laboratory

UTK 2025/05/23
Appalachian Regional Electron Microscopy Society Topical Conference
Third Summer School on ML for Electron Microscopy

CNMS is one of five Nanoscale Science Research Centers (NSRCs) located at Oak Ridge National Laboratory

Center for Nanoscale Materials
Argonne National Laboratory



Molecular Foundry
Lawrence Berkeley National Laboratory

Center for Functional Nanomaterials
Brookhaven National Laboratory

Center for Integrated Nanotechnologies
Sandia National Laboratories
Los Alamos National Laboratory

Center for Nanophase Materials Sciences
Oak Ridge National Laboratory

The eMMA Group at CNMS

- Mission "To advance atomic-scale electron-beam imaging and spectroscopy to enable new understanding of materials, quantum phenomena and energy technologies."

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Interim Group Leader

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Scanning Transmission Electron Microscopy

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Scanning Transmission Electron Microscopy

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Scanning Transmission Electron Microscopy

Andrew R Lupini
Scanning Transmission Electron Microscopy

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Scanning Transmission Electron Microscopy

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Magnetic imaging, TEM, STEM, STEM-holography

Haoran Yu
Analytical (S)TEM, Identical Location Electrochemistry, High-throughput Data Analysis

Motivation

"I would like to try and impress upon you the importance of improving the electron microscope by a hundred times. It is very easy to answer many of these fundamental (biological) questions; you just *look at the thing!*

I put this out as a challenge: **Is there no way to make the electron microscope more powerful?**

What would happen if we could arrange the atoms one by one the way we want them? (within reason, of course; you can't put them so that they are chemically unstable, for example)."

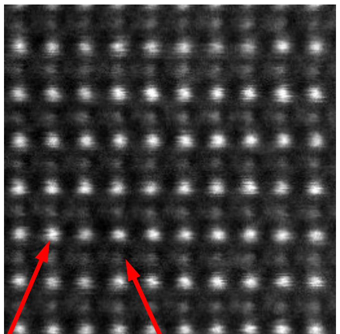
- R. Feynman, 1959
There's Plenty of Room at the Bottom

Build materials and devices atom by atom
Probe, understand and utilize novel properties

http://www.zyvex.com/nanotech/feynman.html

Scanning **T**ransmission **E**lectron **M**icroscope

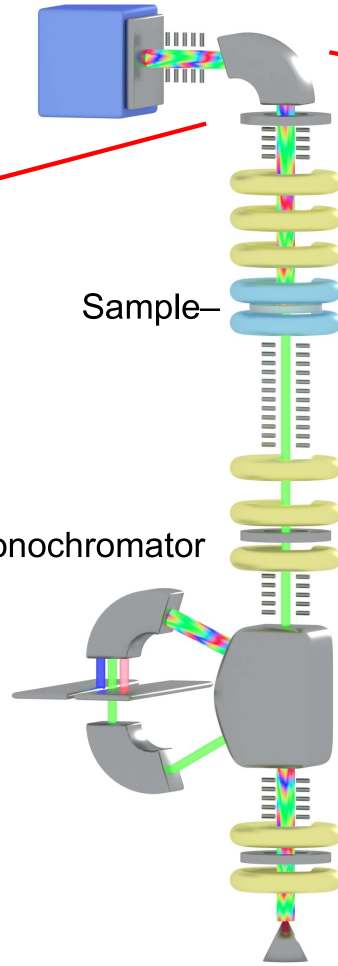
"Z-contrast"



Heavy Light

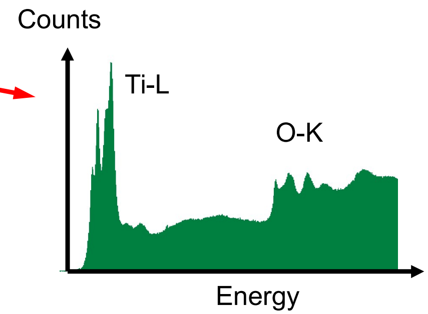
Scattering to high angles depends on atomic number Z

A probe is focused onto the sample. Images are formed by scanning the probe and detecting the scattering on various detectors. HAADF only uses a small fraction of the electrons.



Sample–

Monochromator



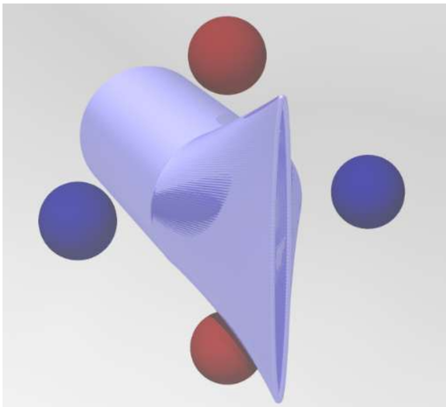
EELS: Chemical AND electronic information

Aberration corrector has hundreds of optical elements

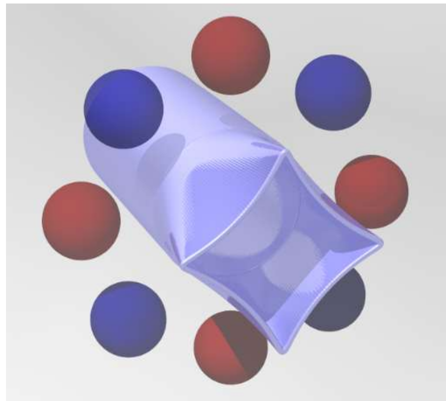
(P. Rez)

Aberration Correction

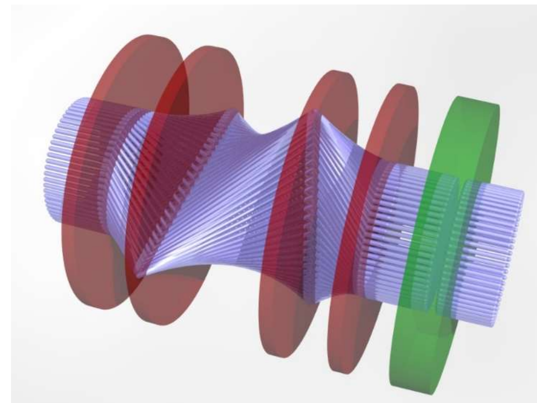
- Spherical aberration limits TEM resolution
- Scherzer (1936) – Spherical aberration unavoidable for static round lenses
- Scherzer (1947) – Combinations of **non-round** lenses can correct C_s



Quadrupole



Octupole

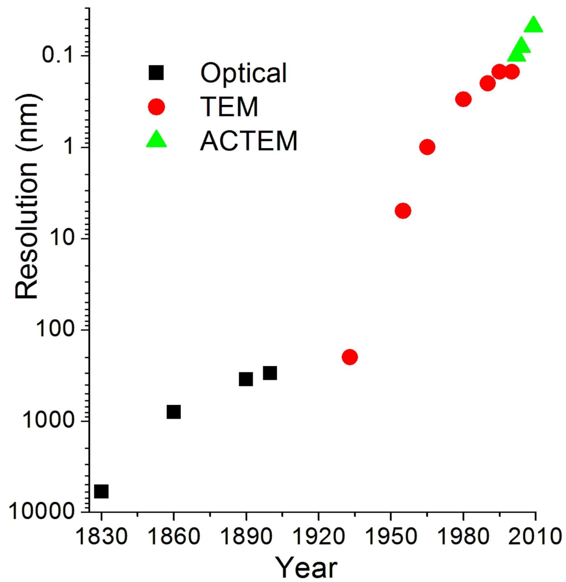


Corrector

- Octupoles have the right field to correct Cs but a 4-fold symmetry
- Aberrations will depend on the beam *shape* as well as the field.
- A corrector is a complicated system – only practical 50 years later.
- Krianek, et al, Haider et al. ~1997
- Lot of extra knobs – Computer control is essential!

Resolution Over Time – Aberration Correction!

~Atomic resolution



Pennycook, S.J.; Varela, M.; Hetherington, C.J.D.; Kirkland, A.I. "Materials Advances through Aberration-Corrected Electron Microscopy" (PDF). MRS Bulletin. 31: 36–43. doi:10.1557/mrs2006.4. S2CID 41889433.
After: H. Rose, Ultramicroscopy 56 (1994) p. 11

1977: First movies of single U atoms

Proc. Natl. Acad. Sci. USA
Vol. 74, No. 5, pp. 1802–1806, May 1977
Physics

Direct observations of atomic diffusion by scanning transmission electron microscopy

(atom motion/single atom visibility/time-lapse cinematography)

M. ISAACSON, D. KOPF, M. UTLAUT, N. W. PARKER, AND A. V. CREWE*
Department of Physics and the Enrico Fermi Institute, The University of Chicago, Chicago, Illinois 60637

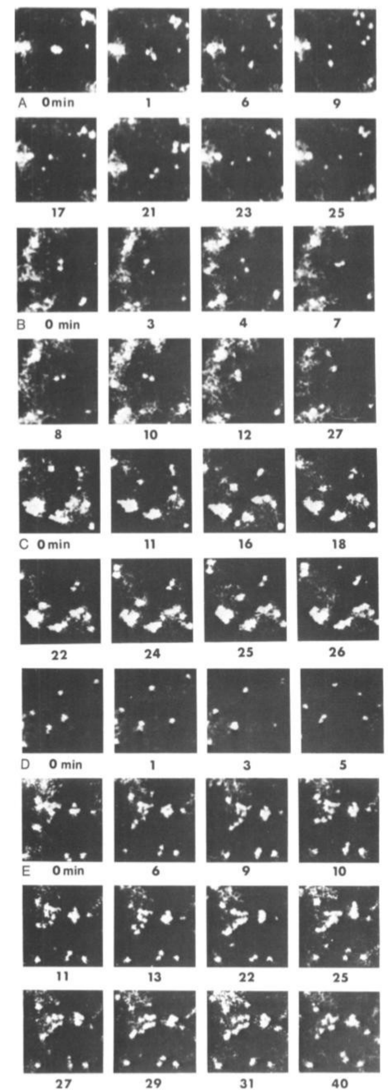
Contributed by Albert V. Crewe, January 21, 1977

"**I don't believe that atoms exist**." — Ernst Mach (1897) vs Boltzman

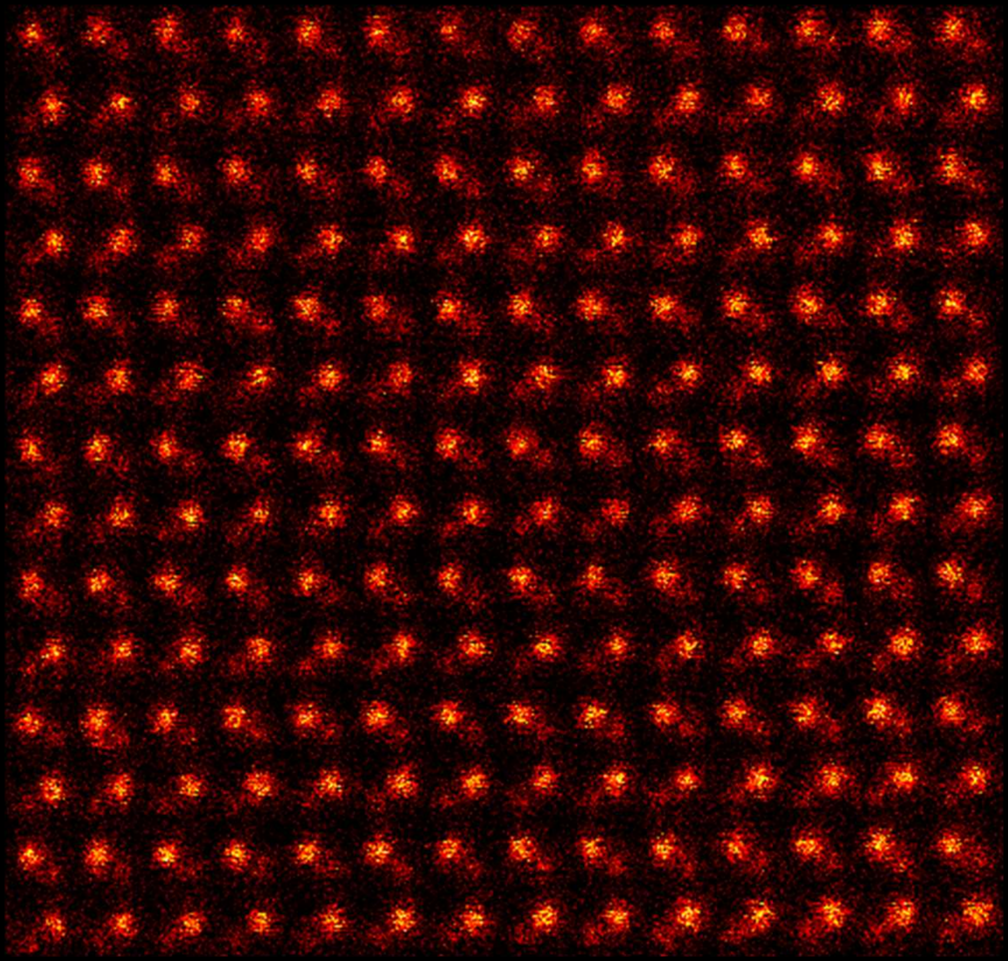
In 1871, Edmund Mills scathingly concluded that "**the atomic theory has no experimental basis, is untrue to nature generally, and consists in the main of a materialistic fallacy**."

Albert Crewe was at Argonne from 1958-1967 and director from 1961-67 (starting at age ~34)

Physics: Isaacson *et al.*



Single Ce-dopant diffusion in AlN

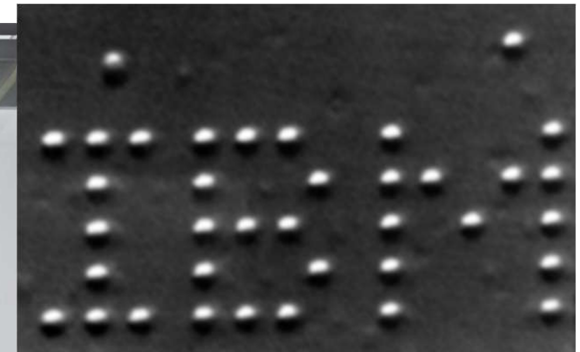


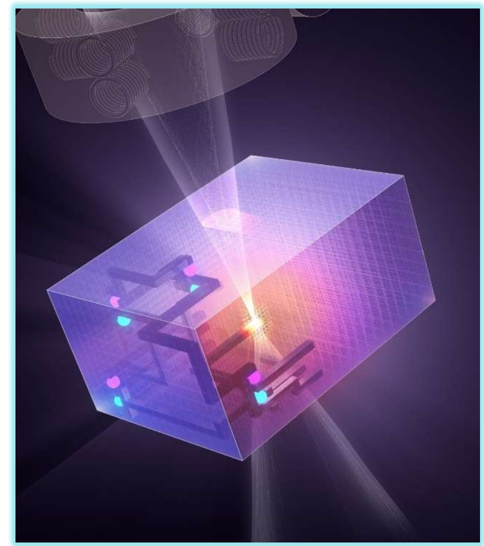
STEM Sequential imaging, 80 Frames

Ishikawa *et al*. PRL (2013), Sci Rep. (2014)

Direct atom placement

- Atom positioning state-of-the-art is AFM and STM
- STEM suggested as a tool to manipulate atoms
- Atomic devices require atoms below the surface
- *How do we control placement of dopants below a surface?*
- *“The Atomic Forge” – Kalinin*

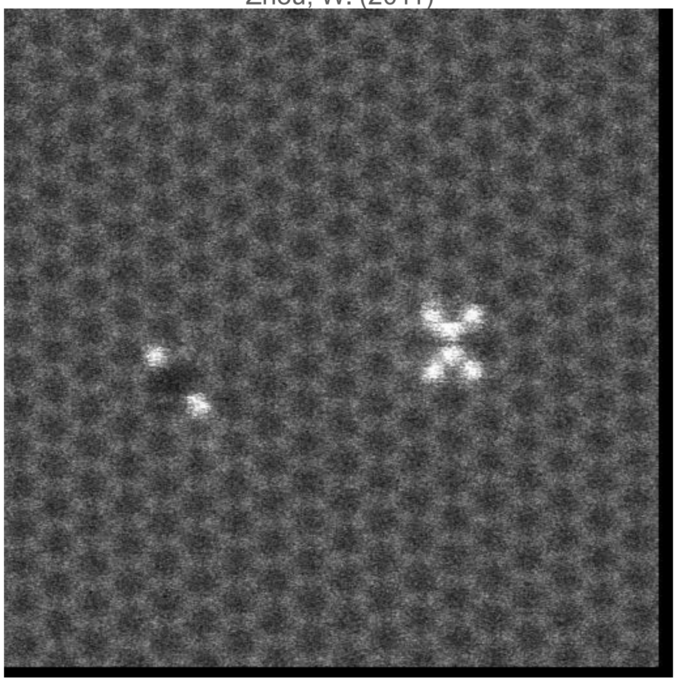


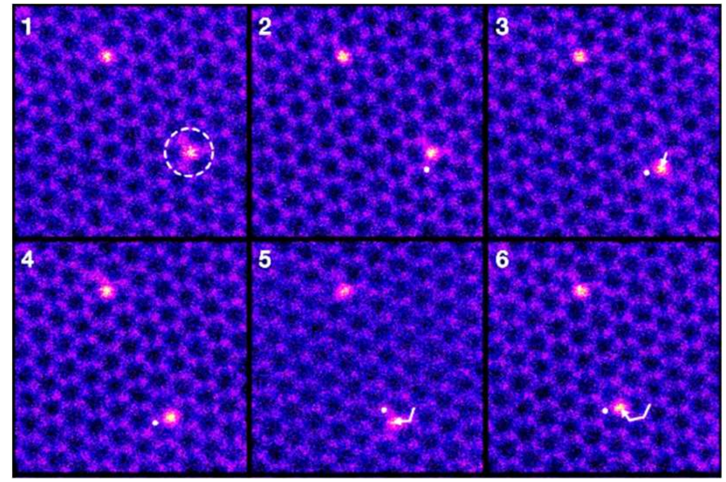


Eigler, D. M. & Schweizer, E. K. Positioning single atoms with a scanning tunnelling microscope. *Nature* **344**, 524–526 (1990).

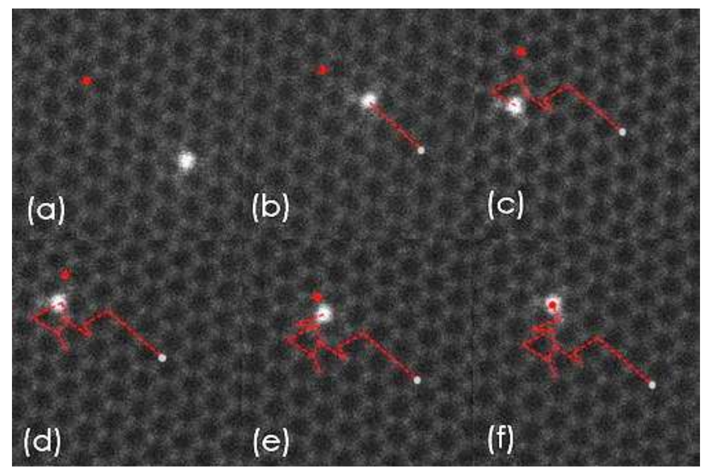
Moving Atoms in STEM
2D materials at 60 kV

Lee, J. *et. al. Nat Commun,* **4**, 1650 *(2013)*
Zhou, W. (2011)





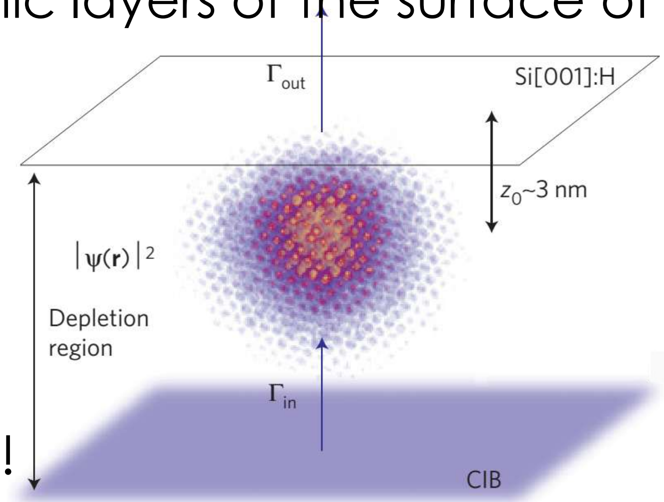
T. Susi *et al.*, Phys. Rev. Lett. **113**, 115501 (2014).
Susi, T. *et. al. Ultramicroscopy,* **180**, 163-172 (2017).



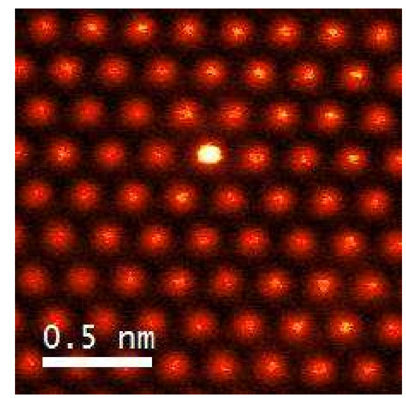
Dyck, O. *et. al. Appl. Phys. Lett.* **111**, 113104 (2017).

Single-atom quantum computing

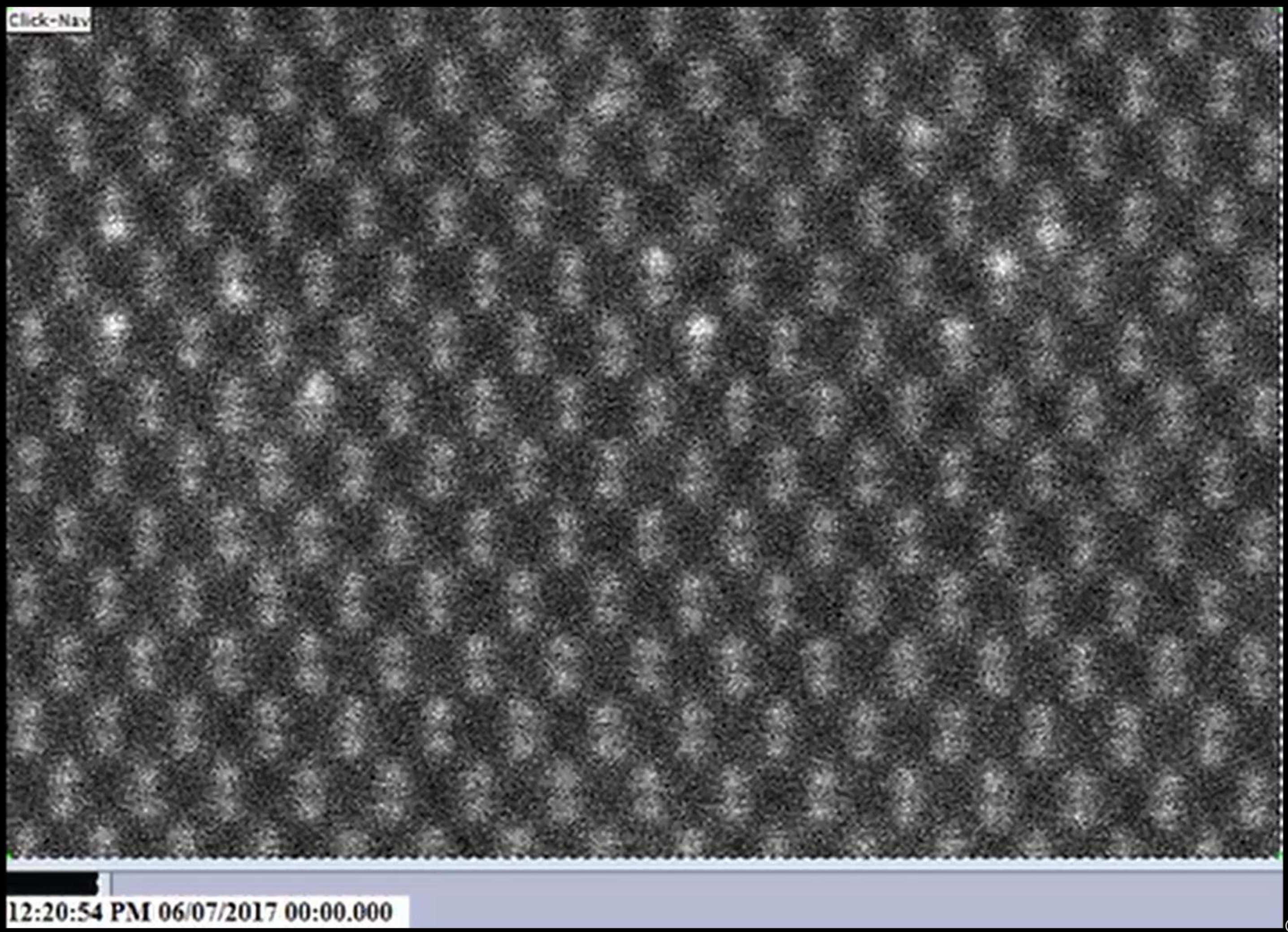
- Quantum computing could revolutionize cryptography, banking, scientific calculations, etc
- For functional single dopant qubits, one needs:
  - Accurate dopant positioning
  - Below but within a few atomic layers of the surface of a semiconductor
- Leverage existing technology:
  - Si based wafers
  - Single **P**, (As), Sb, <u>Bi</u>
  - Strong spin-orbit coupling
- Isotopically pure Si is expensive!
  - Use <u>*thin films*</u>
- Might only need a few dozen qubits to beat a supercomputer



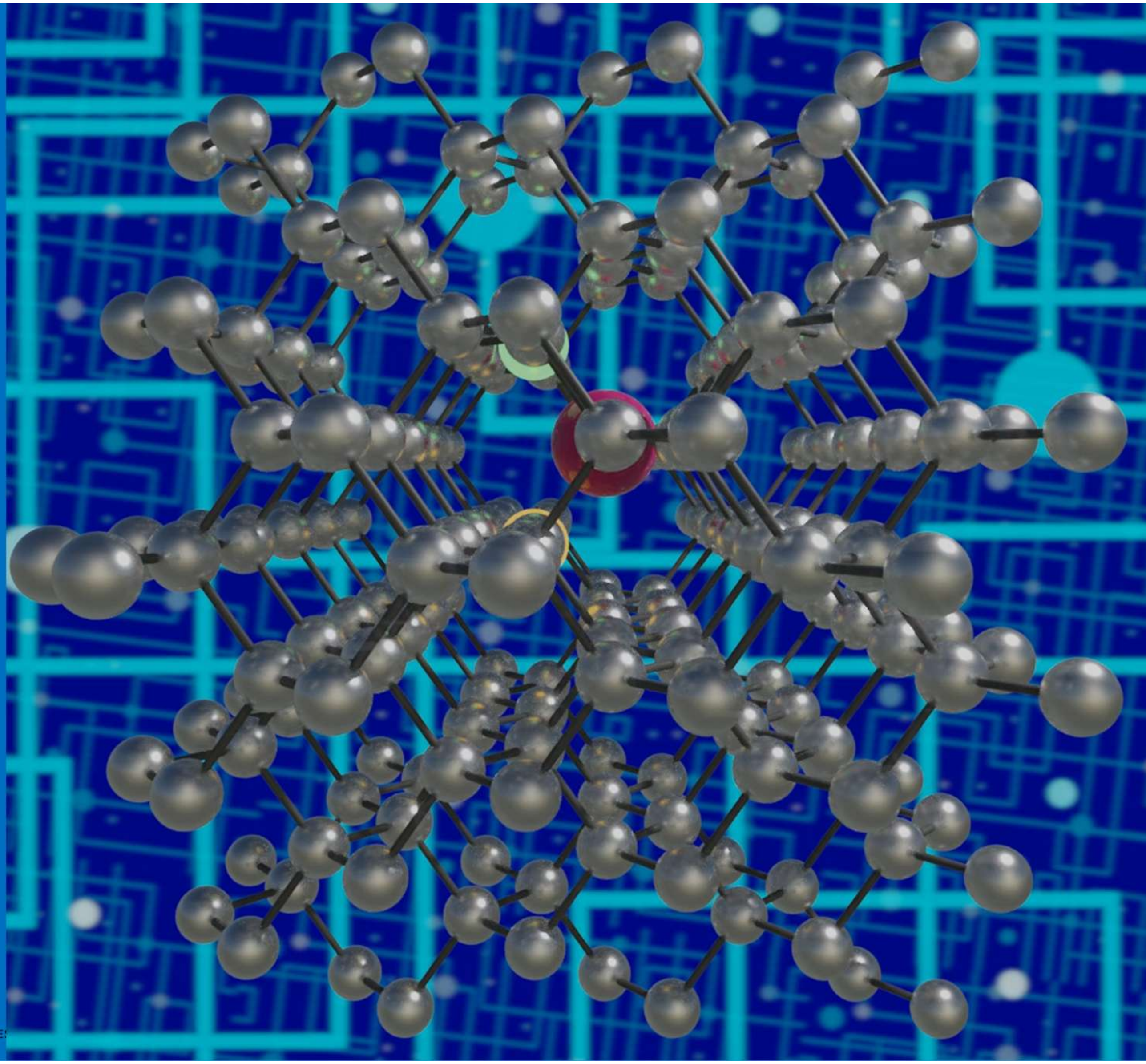
J. Salfi et al. Nature Materials '14



Assembling atomic structures @ 160kV



Hudak, B.M *et al.* Directed Atom-by-Atom Assembly of Dopants in Silicon. *ACS Nano (2018)*



M.D. simulations
T. Susi, &
A. Markevich

Open slide master to edit

Other improvements: Detectors

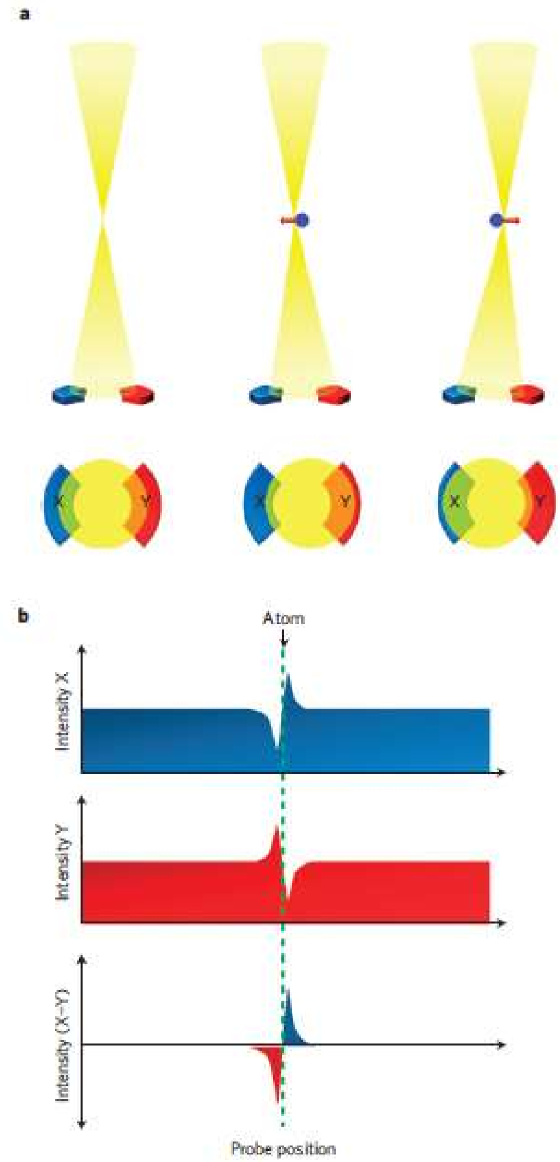
Differential phase contrast (DPC)

The in plane field deflects the beam causing a contrast by subtracting perpendicular segments of a detector below

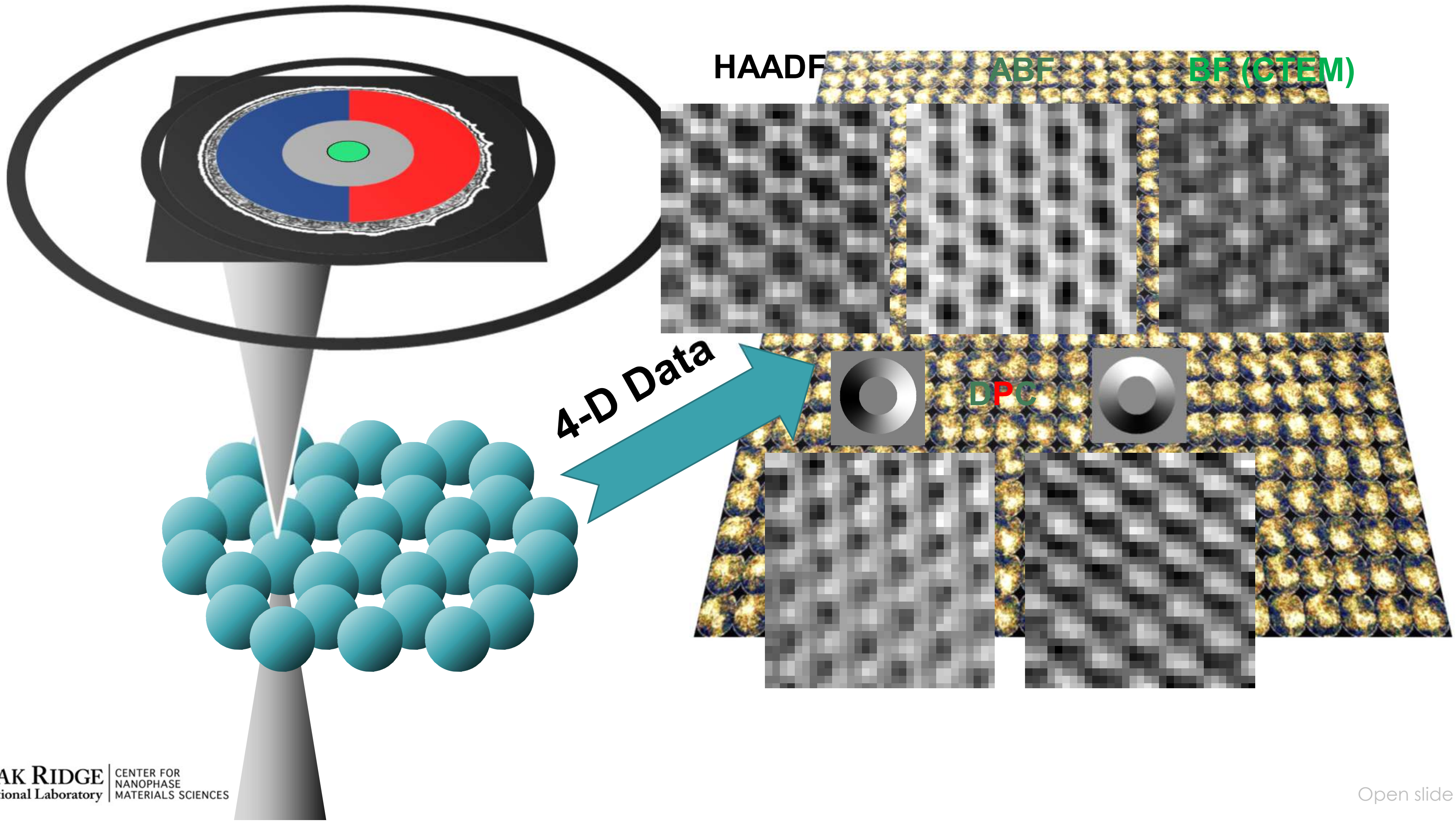
Dekkers & de Lang (1974) Optik
H.Rose *Optik*

Shibata & Ikuhara et. al. (2012) *Nature Physics.*

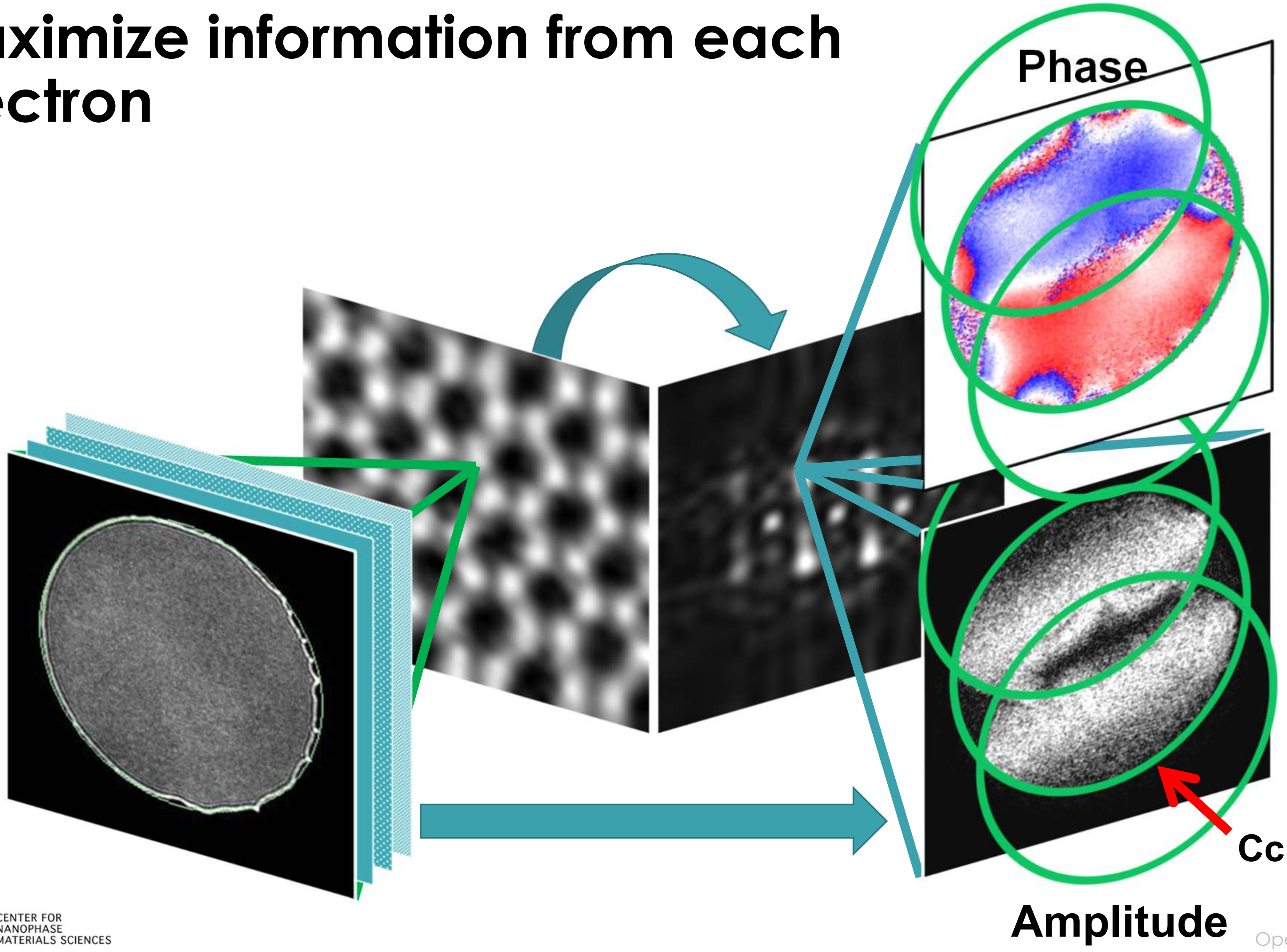
Integrated differential phase contrast
Bosch, Lazic & Lazar (2016)



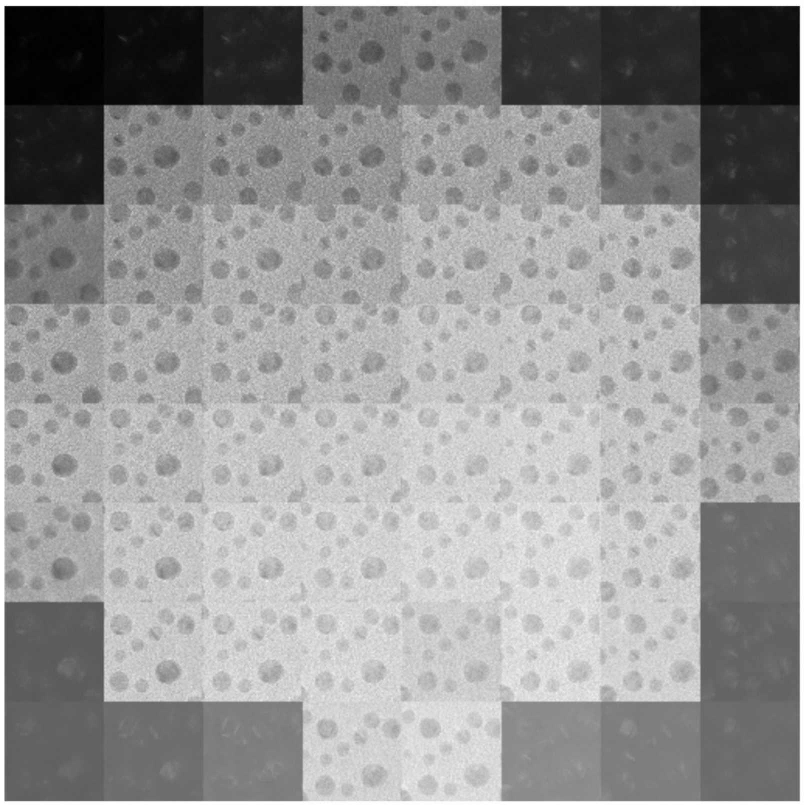
“Universal Detector”

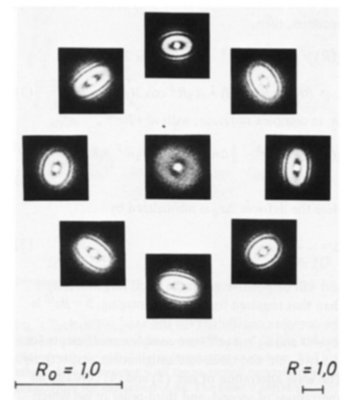


Maximize information from each electron

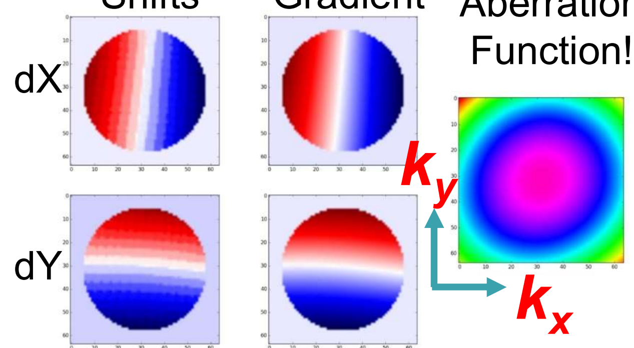


Form an image for each detector pixel



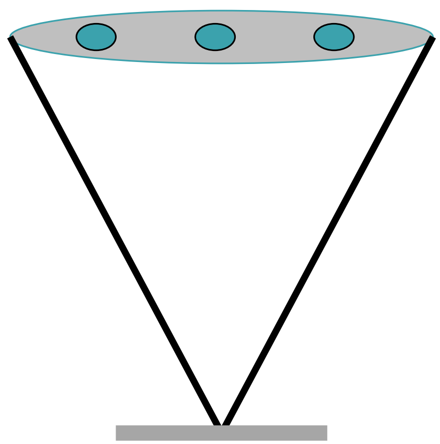


Zemlin Tableau



Principle of reciprocity: Equivalent to tilted TEM

Limitations of STEM Bright Field



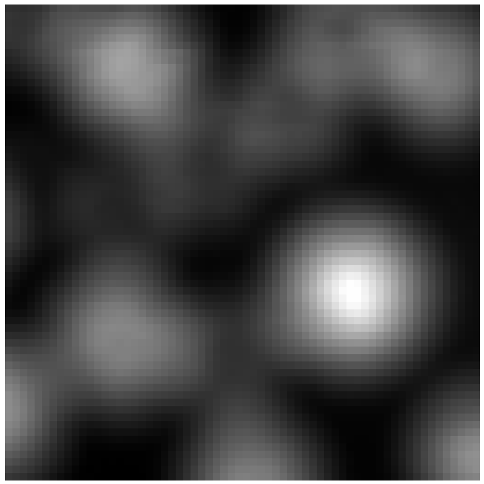
- **A large detector is the SUM of many smaller detectors at different angles**
- **Each would produce a different image**
- **The sum loses the smaller details…**
- **Damping envelope:**

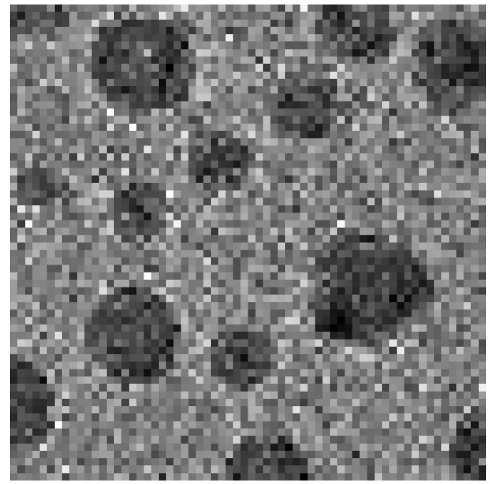
$$E(k) = \exp\left(-\left(\underbrace{\frac{\pi}{\lambda}\sigma}_{\text{Aperture}} \underbrace{\left| \nabla \chi(k) \right|}_{\text{Gradient of Aberration function}} \right)^2 \right)$$

Ref: J. Frank, Optik 1973

Simple Reconstruction

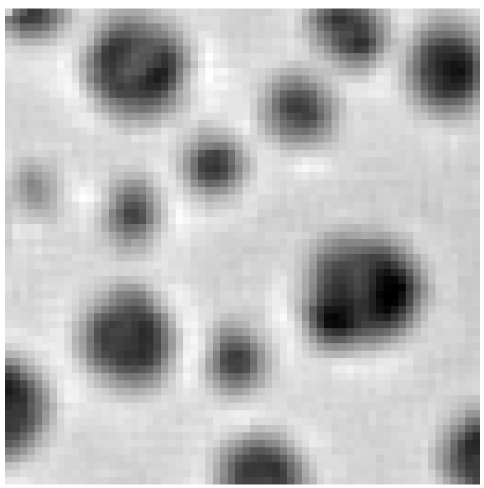
HAADF Image **64x64** probe positions, -250 nm defocus **Blurry!**

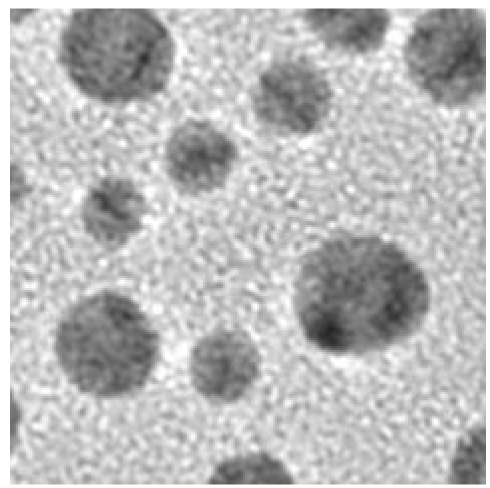




BF Image from single detector pixel 64x64 Probe positions Noisy!

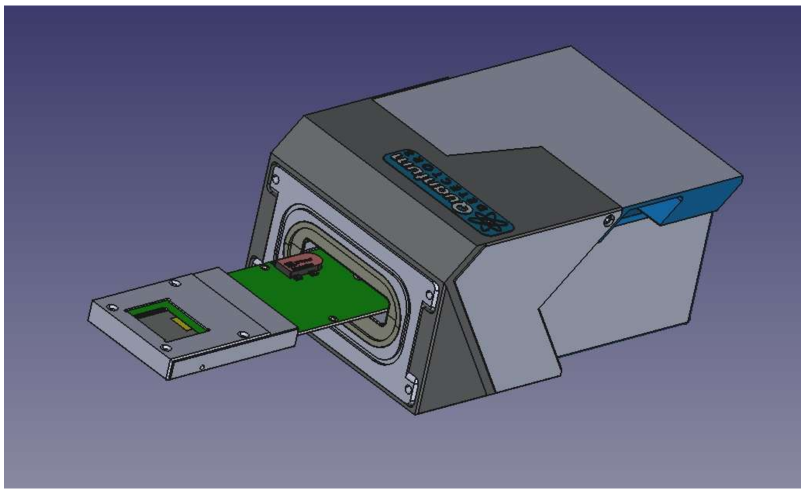
BF Image from large detector 64x64 Probe positions Blurry!





BF Image from shifting images by grad(chi) Reconstruct to **256x256** positions! (Lupini, Chi, Jesse, Journal Of Microscopy 2016)

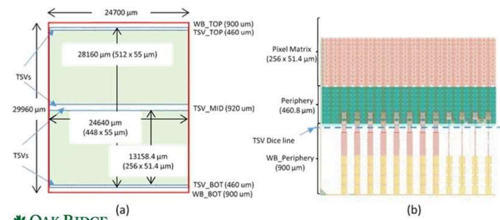
Better Detectors

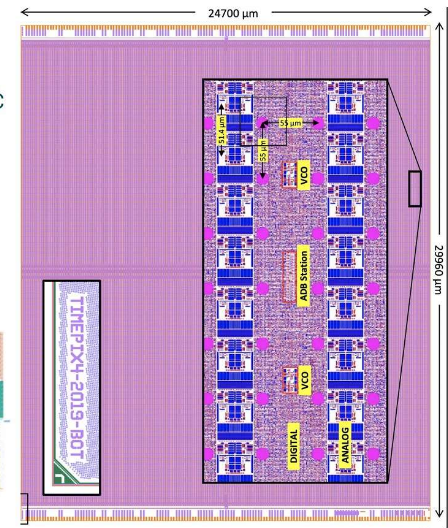


The Timepix4 ASIC (65 nm)

Timepix4 aims to be the highest bandwidth pixel ASIC on the market and **4-side buttable!**

- **Large size ASIC** : 25 x 30 mm
- 55x55μm^2 pixels, 800e threshold
- **Fully TSV compatible** with removable periphery
- 160Gbps, **3.6 Mhits/s/mm^2** capabilities
- **200 ps timing** capability on every pixel hit



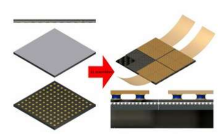


OAK RIDGE National Laboratory

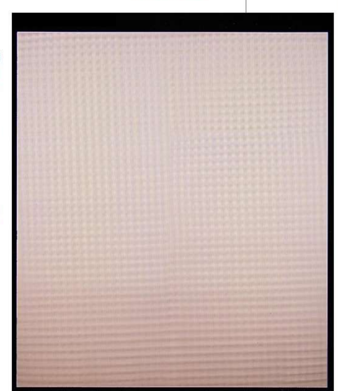
A Timepix4 with TSV

ORNL is collaborating with the CERN team in planning the processing of 12” wafers with IZM Berlin, securing 2-3 wafers (~70 chips)

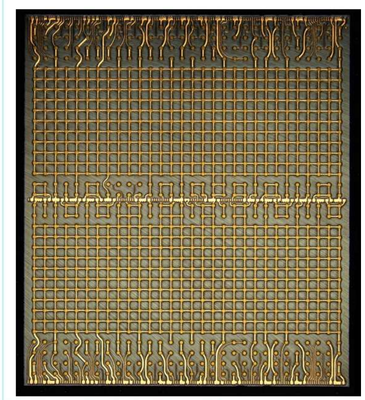
- TSV processing essential for large area tiling
- We plan on developing integration methodology using ACP/ACF or bumps that is ultimately wire-bond less



Pixel side



Back side



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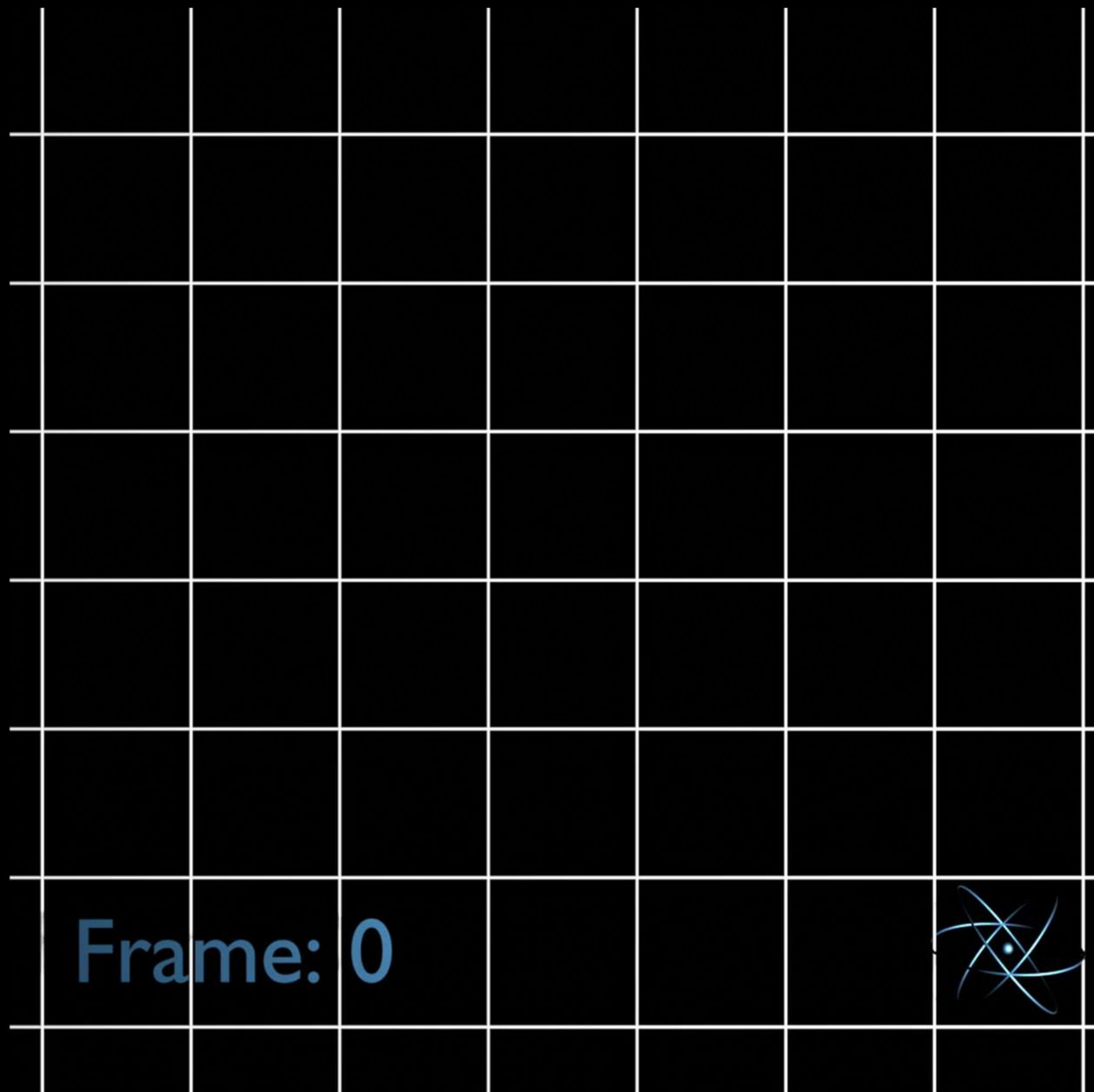
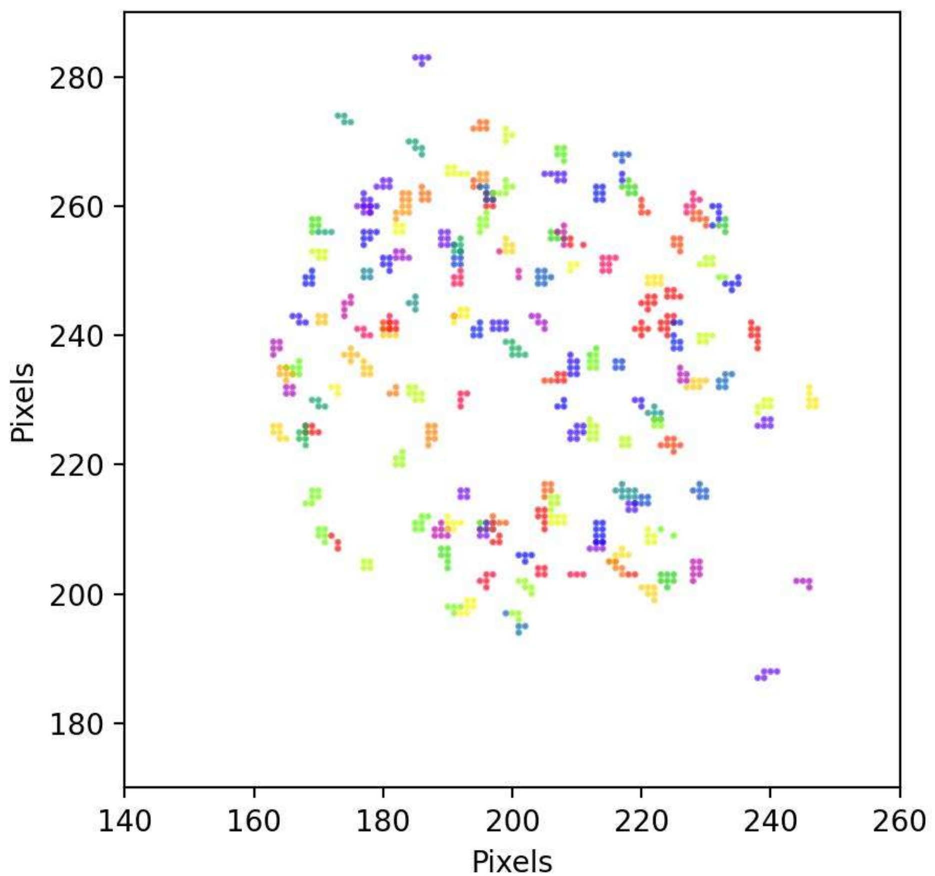
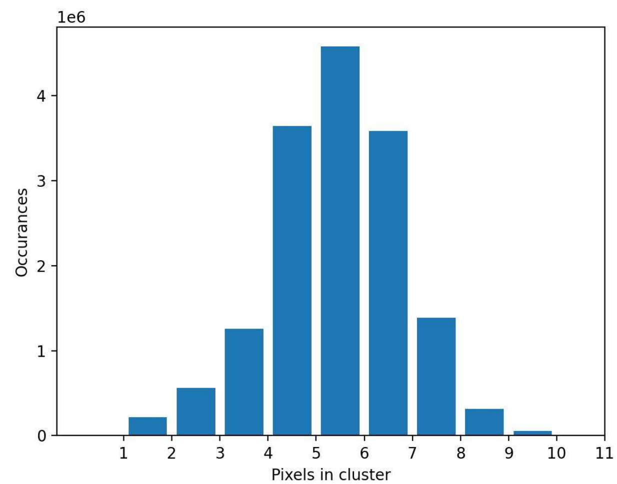
https://indico.cern.ch/event/1439336/contributions/6261676/attachments/2978870/5244628/drd3_extra_results.pdf


Frame: 0

Time (uS): 0.0

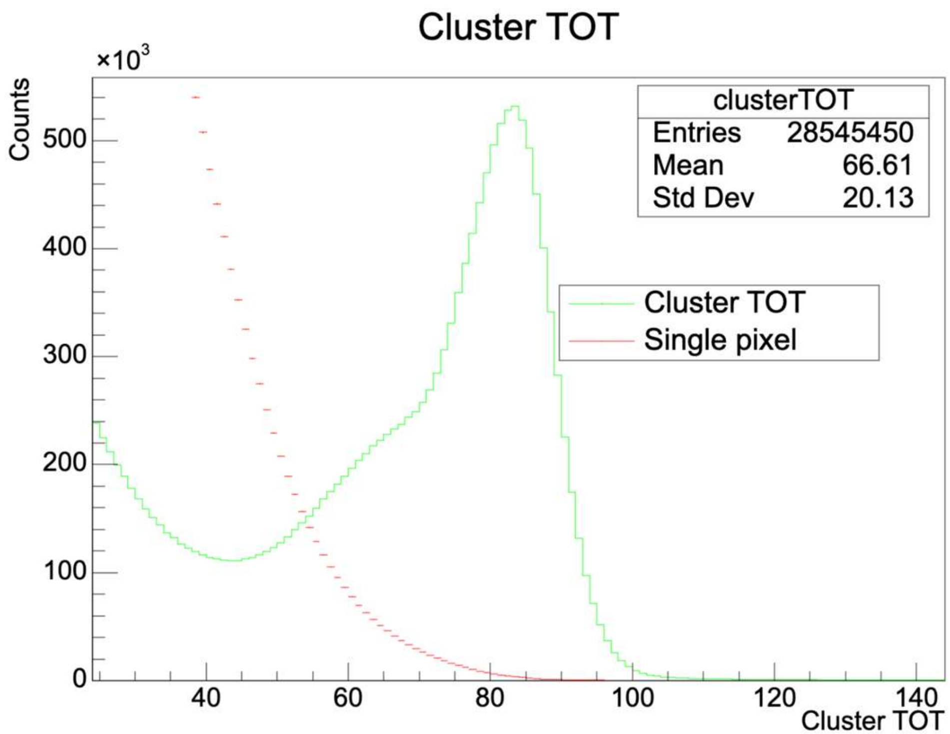
Electrons hit multiple pixels



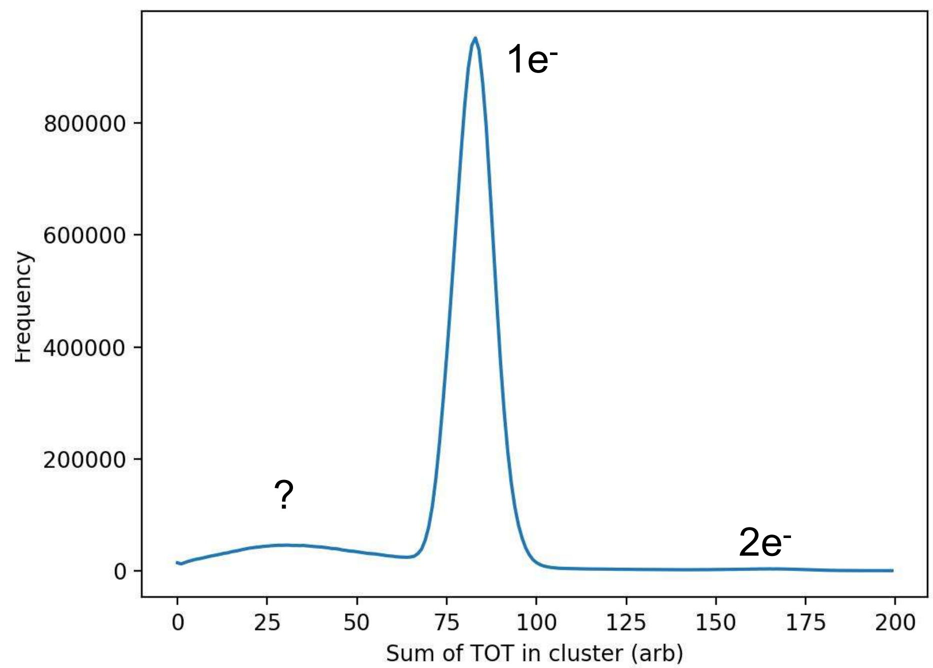


How to separate/cluster events?

Clustering algorithms

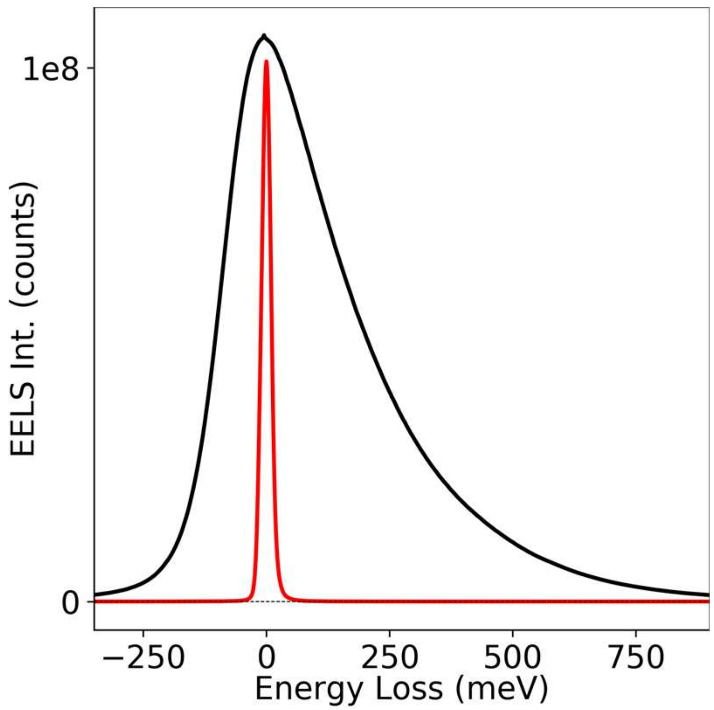


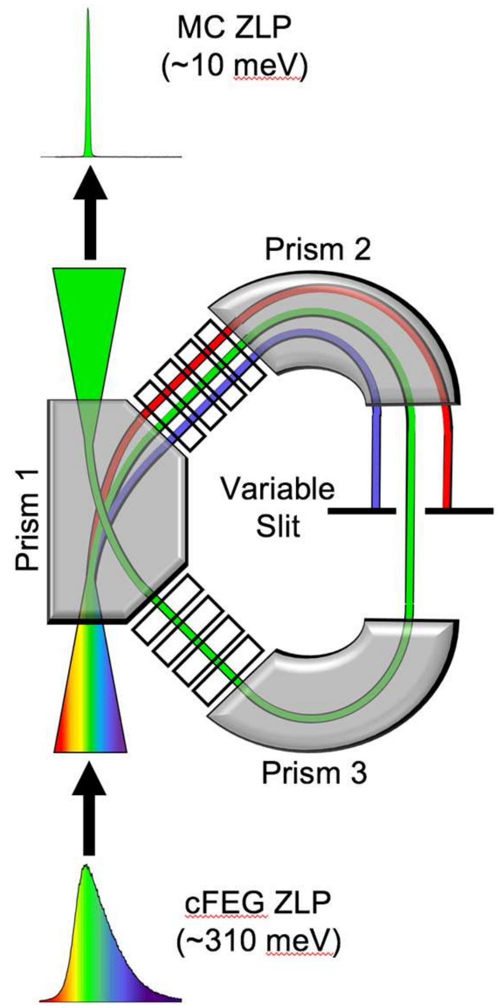
Separate clusters by time and position



Alternative handling for overlapping clusters

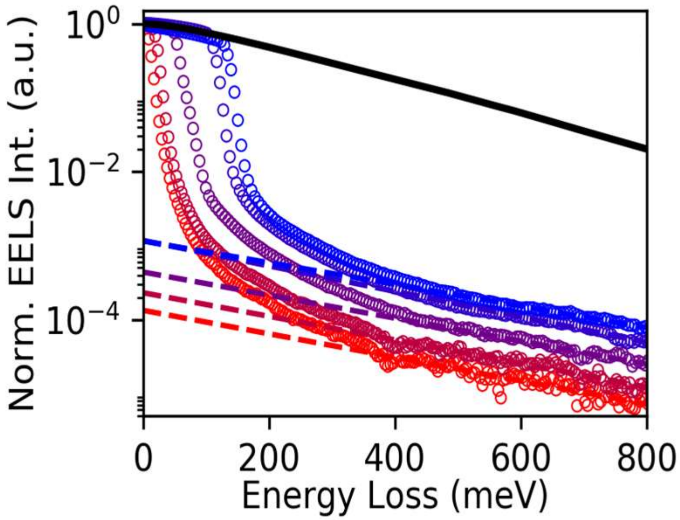
How else could the microscope be improved?



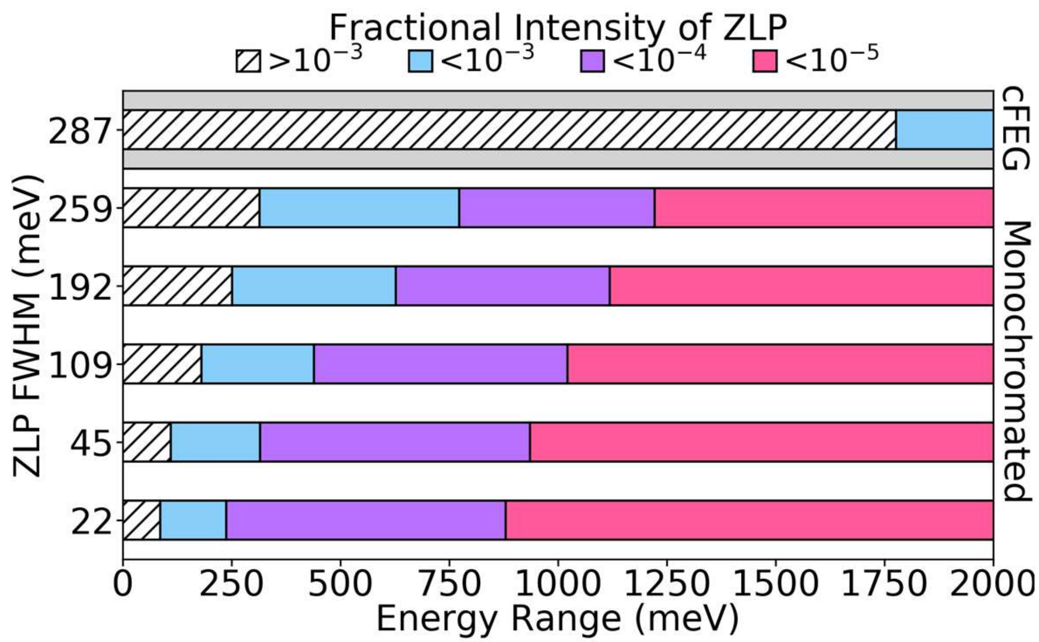


Krivanek et al (2014)
Hachtel et al (2018)

Reduced background in the infrared



3+ order of magnitude reduction in IR background



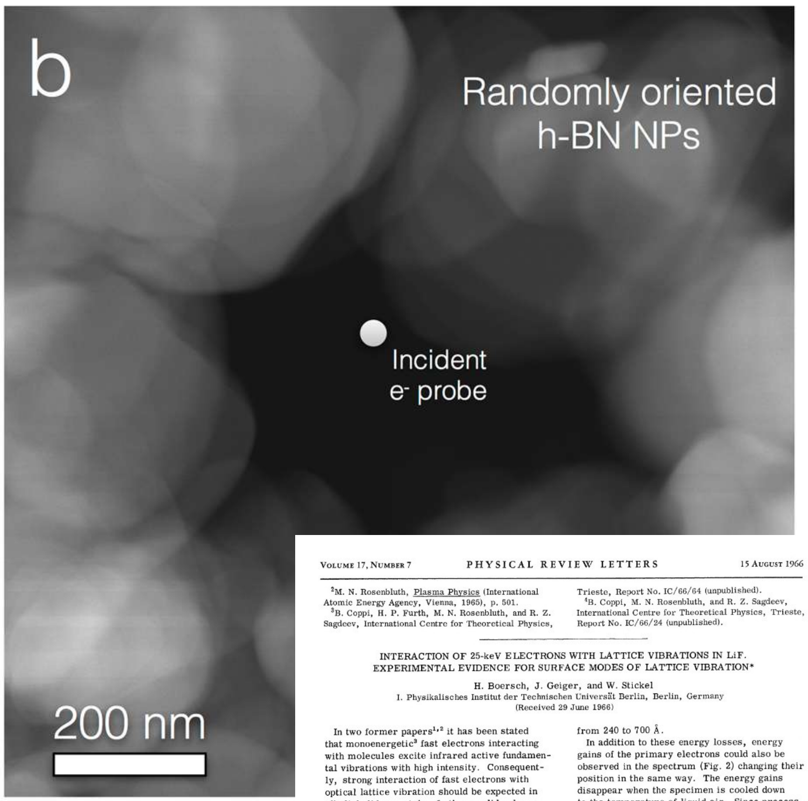
CFEG

Can't use cross-hatched region…

Zhou & Idrobo (2017), Hachtel *et al,*(2018)

Infra-red Phonon Spectroscopy in the Electron Microscope

Z-contrast Image



VOLUME 17, NUMBER 7 PHYSICAL REVIEW LETTERS 15 AUGUST 1966

[2]M. N. Rosenbluth, Plasma Physics (International Atomic Energy Agency, Vienna, 1965), p. 501.
[3]B. Coppi, H. P. Furth, M. N. Rosenbluth, and R. Z. Sagdeev, International Centre for Theoretical Physics, Trieste, Report No. IC/66/64 (unpublished).
[4]B. Coppi, M. N. Rosenbluth, and R. Z. Sagdeev, International Centre for Theoretical Physics, Trieste, Report No. IC/66/24 (unpublished).

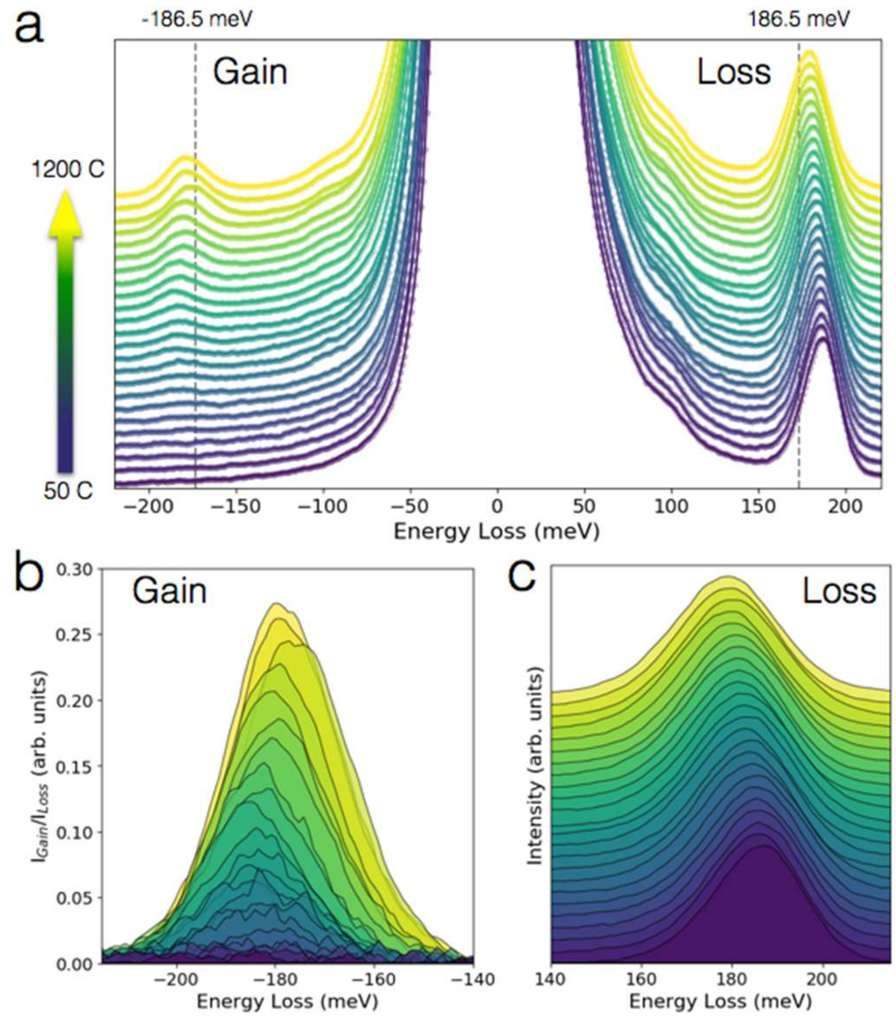
INTERACTION OF 25-keV ELECTRONS WITH LATTICE VIBRATIONS IN LiF. EXPERIMENTAL EVIDENCE FOR SURFACE MODES OF LATTICE VIBRATION*

H. Boersch, J. Geiger, and W. Stickel
I. Physikalisches Institut der Technischen Universät Berlin, Berlin, Germany
(Received 29 June 1966)

In two former papers[1,2] it has been stated that monoenergetic[3] fast electrons interacting with molecules excite infrared active fundamental vibrations with high intensity. Consequently, strong interaction of fast electrons with optical lattice vibration should be expected in alkali-halide crystals. In these solids, however, as is well known, electromagnetic waves excite transverse vibrations ω_T of the lattice, whereas a charged particle excites the longitudinal modes ω_L. The maximum of light absorption and the most probable energy loss appear at different energies.

from 240 to 700 Å.

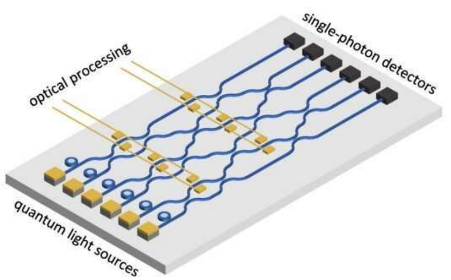
In addition to these energy losses, energy gains of the primary electrons could also be observed in the spectrum (Fig. 2) changing their position in the same way. The energy gains disappear when the specimen is cooled down to the temperature of liquid air. Since processes leading to energy loss and energy gain have equal probabilities, the intensity ratio of energy gain and energy loss is proportional to the occupation probability of the state, which is a function of the temperature of the specimen.



Principle of detailed balance
Unlike plasmons – no calibration needed

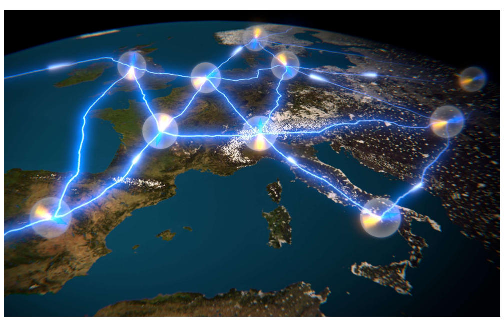
Single photons are ideal quanta for realizing quantum computing and networks

Quantum computing

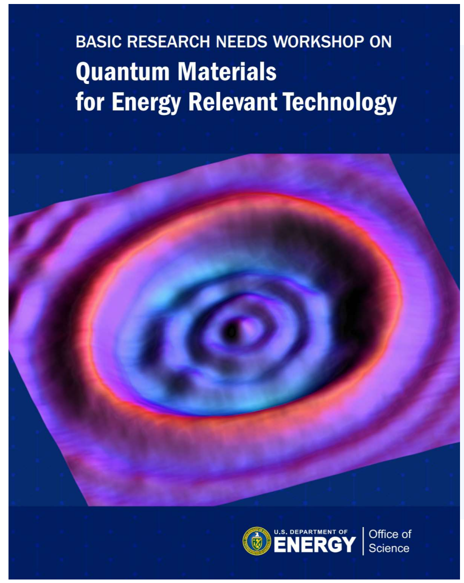


https://phys.org/news/2019-10-quantum-photonics.html

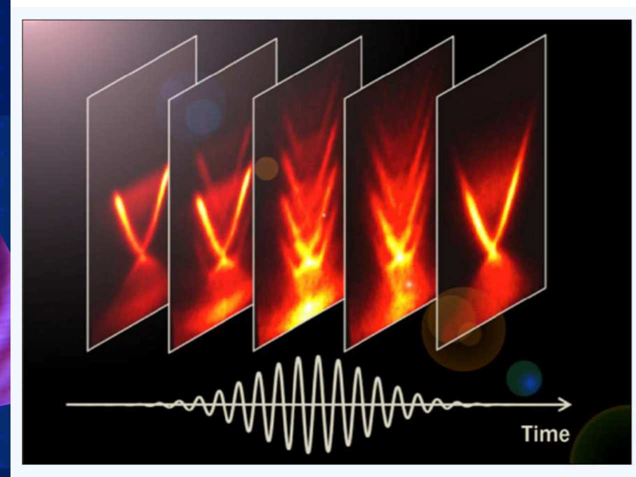
Quantum network

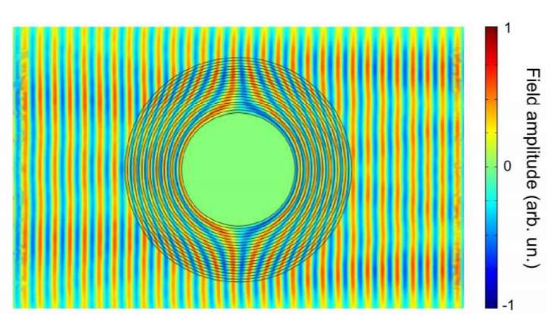


https://projectqsydney.com/quantum-teleportation-paving-the-way-for-a-quantum-internet/

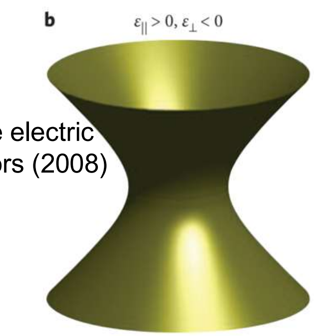


Light-Matter or electron-matter interactions:

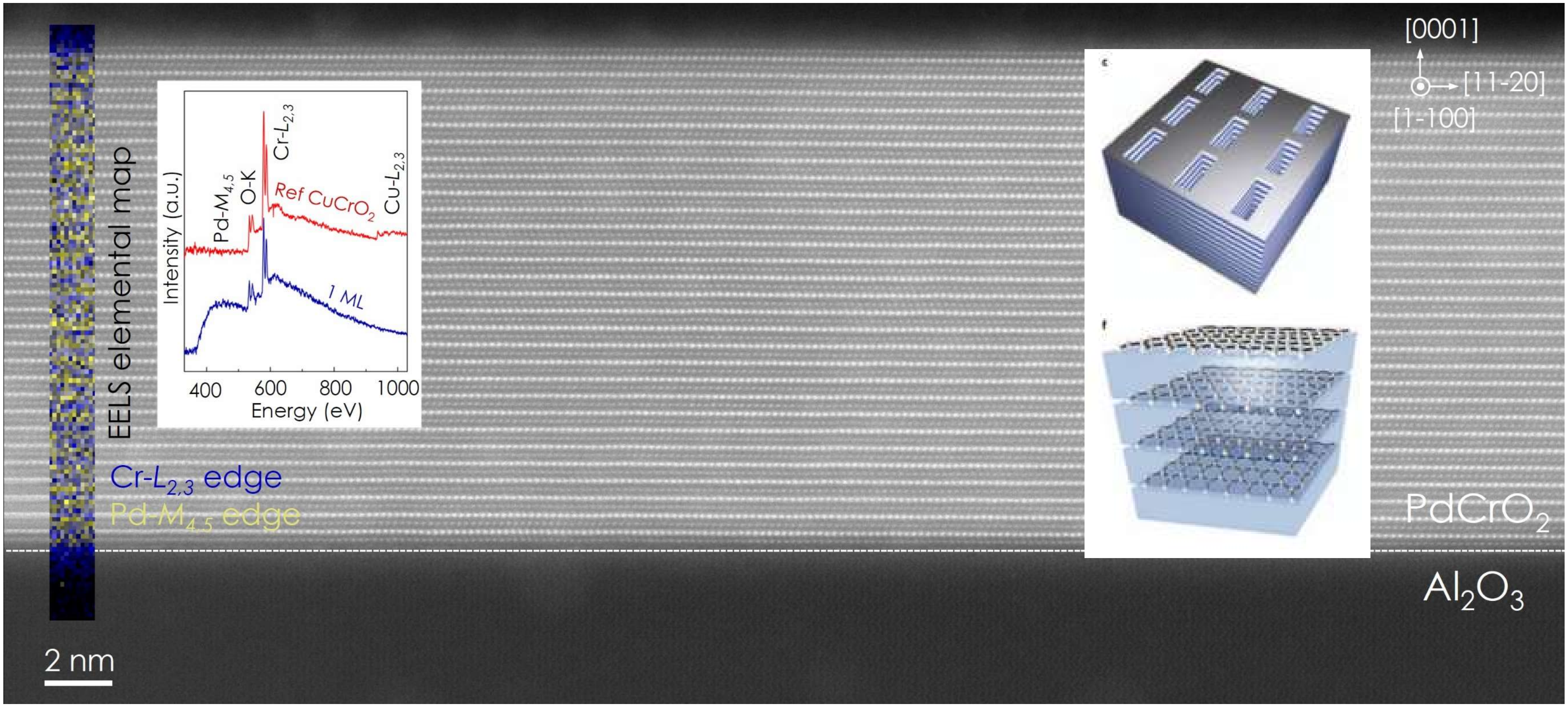




"Invisibility Cloak":
Infrared cloaking based on the electric response of split ring resonators (2008)
OPTICS EXPRESS 9191



Atomic Scale Hyperbolic Metamaterials
STEM EELS for composition and growth quality

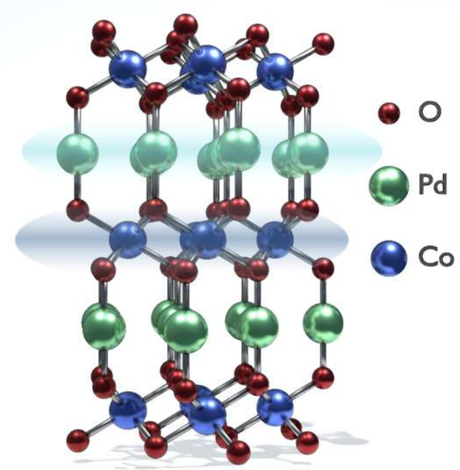


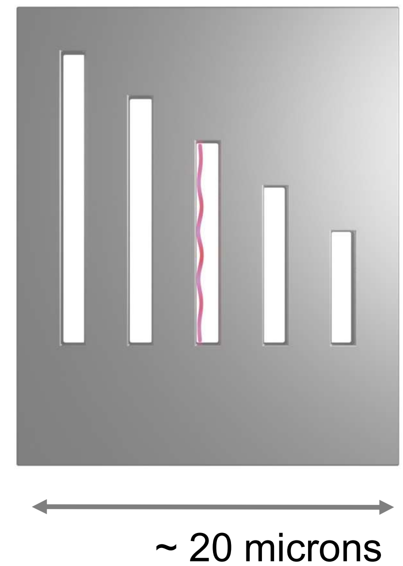
Buffer layer ($CuCrO_2$) essential for single phase growth

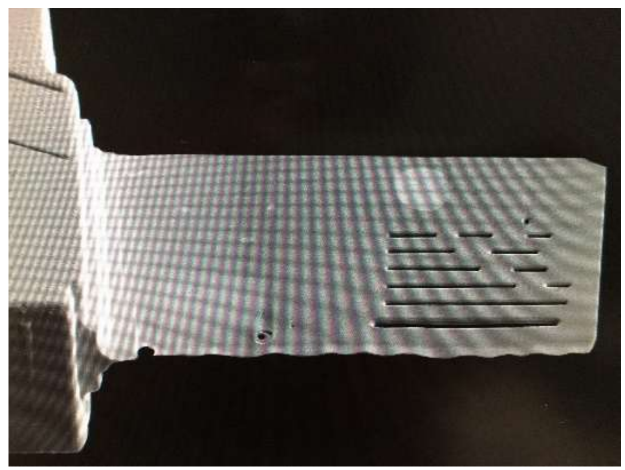
J.M. Ok *et al.* APL Materials 8 (5), 051104 (2020)

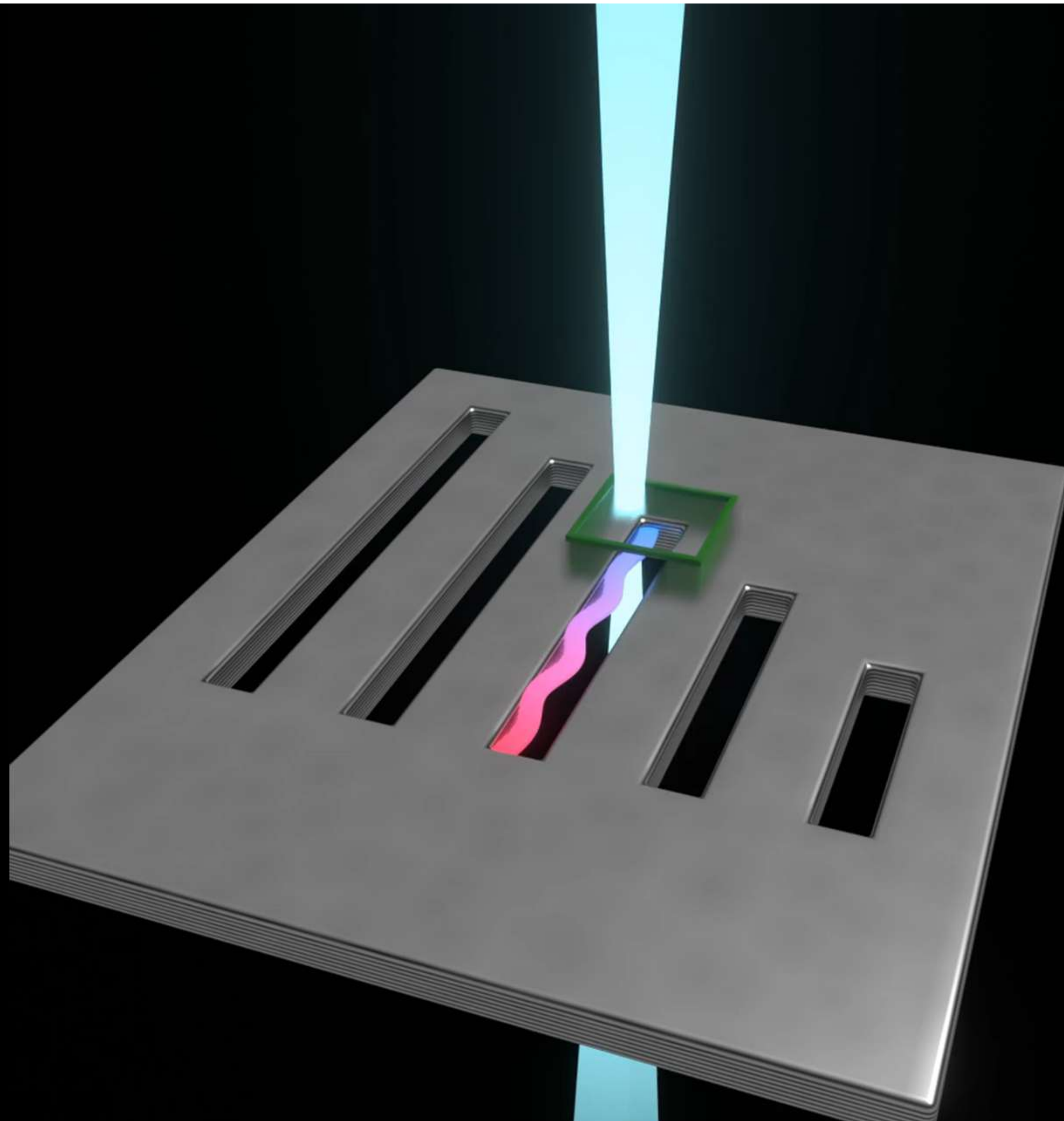
Experimental challenges:

- Want ~ 20 meV energy resolution
  - **MAC-STEM**
- Want ~ 10 nm spatial resolution
- Want ~ 20 micron Field of view
  - Limit ~ 0.5 micron (without distortion)
- Long and repetitive acquisition
  - Automated scripting and tiling
  - AI/ML for analysis?

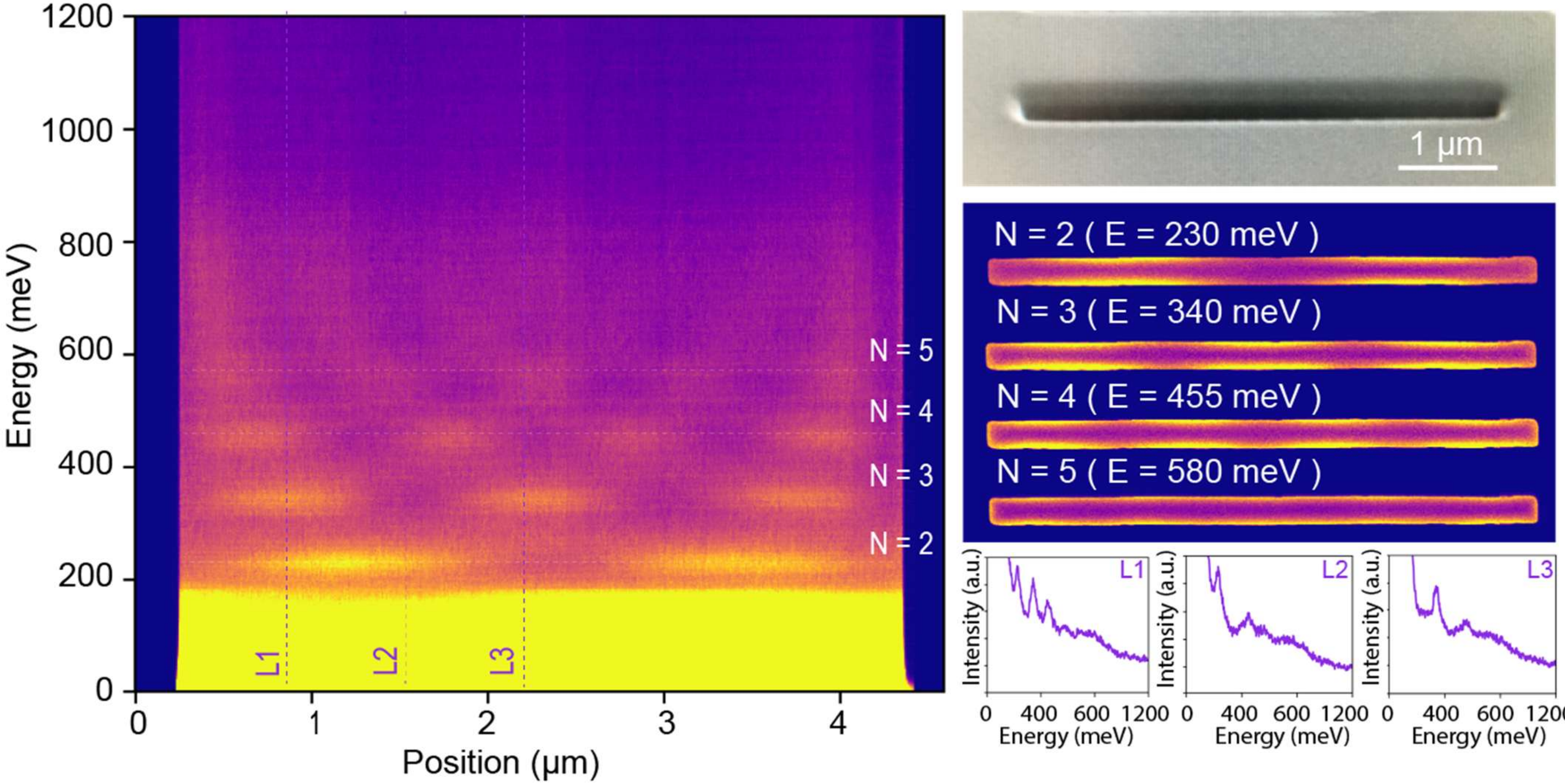




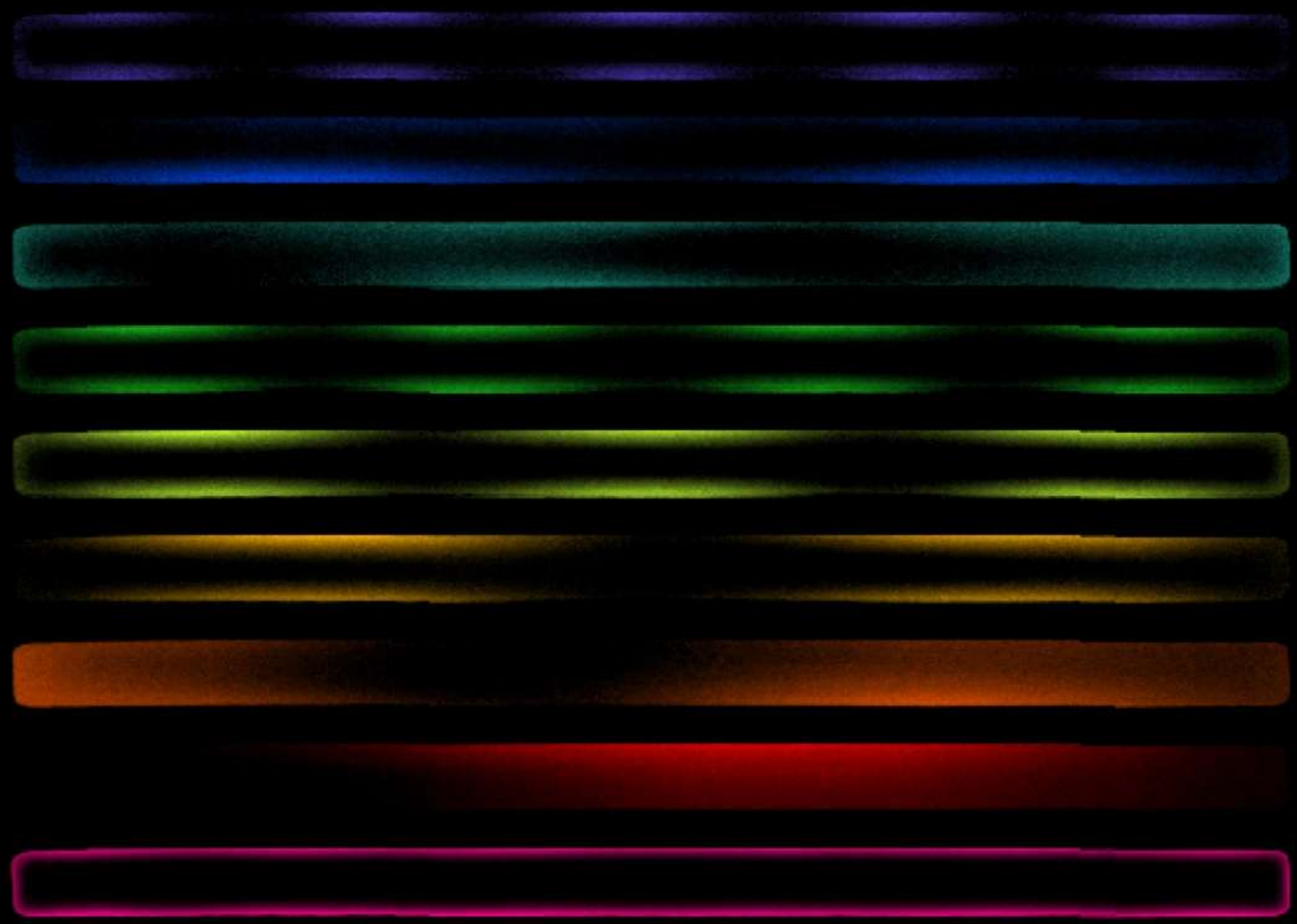




Plasmonic Response



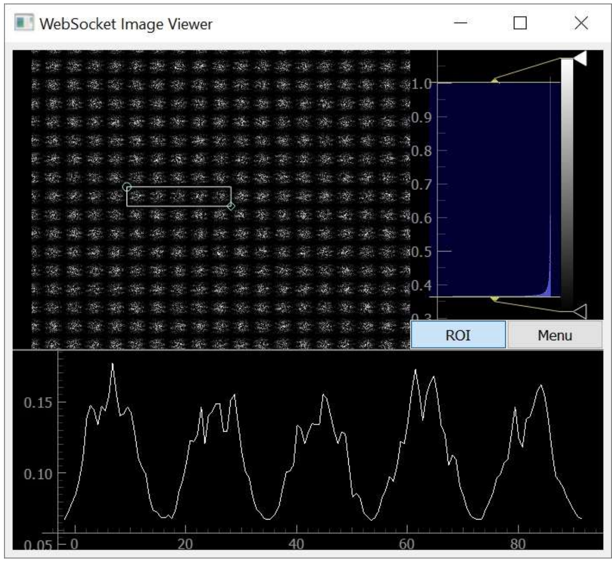
Non negative matrix factorization – Spatial map



4 micron slot

Obvious Areas for AI/ML/AE in (S)TEM

- Set up the microscope
  - Corrector, Monochromator tuning
  - Find your atom!
  - Keep the microscope focused at the same area
  - General Automation
- How representative is that little area?
  - Scan a whole sample / many samples
- Better data stream at high speeds
  - Tag each electron instead of keeping 4D stack
- Image / sample reconstructions and analysis
- Move many atoms
- Physics simulations – What are the properties?



Center for Nanophase Materials Sciences

A DOE User Facility for Creating, Characterizing, and Understanding Nanomaterials

Providing **free access to staff expertise and equipment** if intent is to publish results.

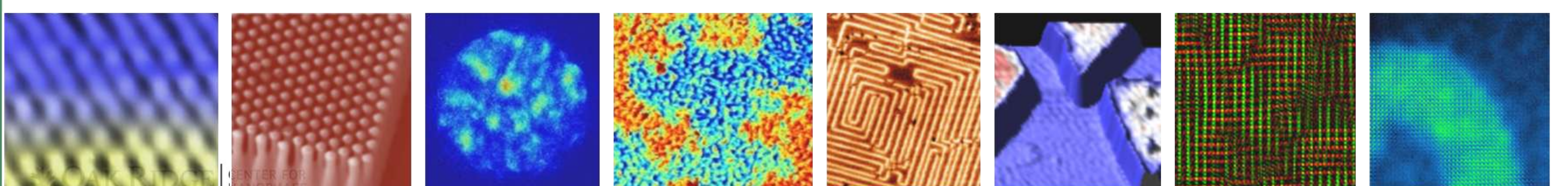
Proposals:

- Simple, two-page narrative
- Two general calls per year
- Short-term projects accepted continuously
- Joint proposals with neutron sources (SNS, HFIR)

cnms.ornl.gov

Research areas:

- **Synthesis** – Soft matter (precision synthesis, selective deuteration), 2D materials, hybrid structures, epitaxial oxides
- **Nanofabrication** – Direct-write (3D) fabrication, e-beam lithography, multiscale fluidics, 10,000 sq. ft. cleanroom
- **Advanced Microscopy** – AFM, STM, aberration-corrected and *in situ* TEM/STEM, He-ion microscopy, atom-probe tomography
- **Chemical Imaging** – Multiple approaches based on mass spectrometry or optical spectroscopies
- **Functional Characterization** – Laser spectroscopy, transport, magnetism, electromechanical phenomena
- **Theory/Modeling, Data Analytics** – Including interactions and co-development with leadership-class, high-performance computing
- **Gateway to Neutron Sciences** – deuterated materials, sample environments, multimodal measurements



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SHORT (5 min) paper summaries

Conference talks

Linked in: https://www.linkedin.com/in/ondrejdyck/

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