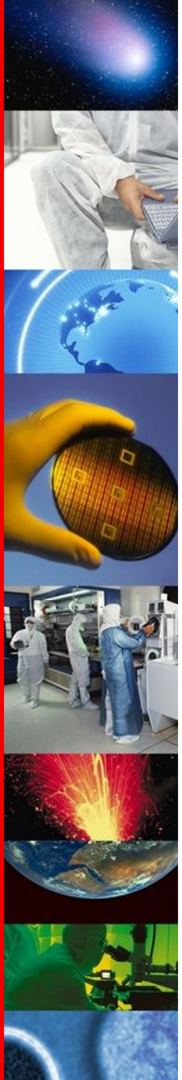

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In Pursuit of Atomic-Scale Tomography

Thomas F. Kelly

ASCI 2016

May 19, 2016

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Collaborators on Pending Programs

Project LaPlace

- Dierk Raabe, Baptiste Gault,
- Gerhard Dehm, Christina Scheu
  - Max Planck Institute Düsseldorf
- Integration of LEAP and STEM

Project Tomo

- Rafal Dunin-Borkowski
  - Forschungzentrum Jülich
- Joachim Mayer
  - RWTH Aachen
  - Forschungzentrum Jülich
- Dierk Raabe
  - Max Planck Institute Düsseldorf
- Max Haider
  - CEOS
- Integration of LEAP and TEM

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Other Active Collaborators

ATOM Project

- Simon P. Ringer
  - University of Sydney
- Michael K. Miller
  - Oak Ridge National Laboratory
- Krishna Rajan
  - Iowa State University
- Ondrej Krivanek, Niklas Dellby
  - Nion Instruments

LEAP-STEM Imaging

- Brian Gorman, David Dierks
  - Colorado School of Mines
- Christoph Koch,
  Wouter van den Broek
  - Humboldt Universität - Berlin

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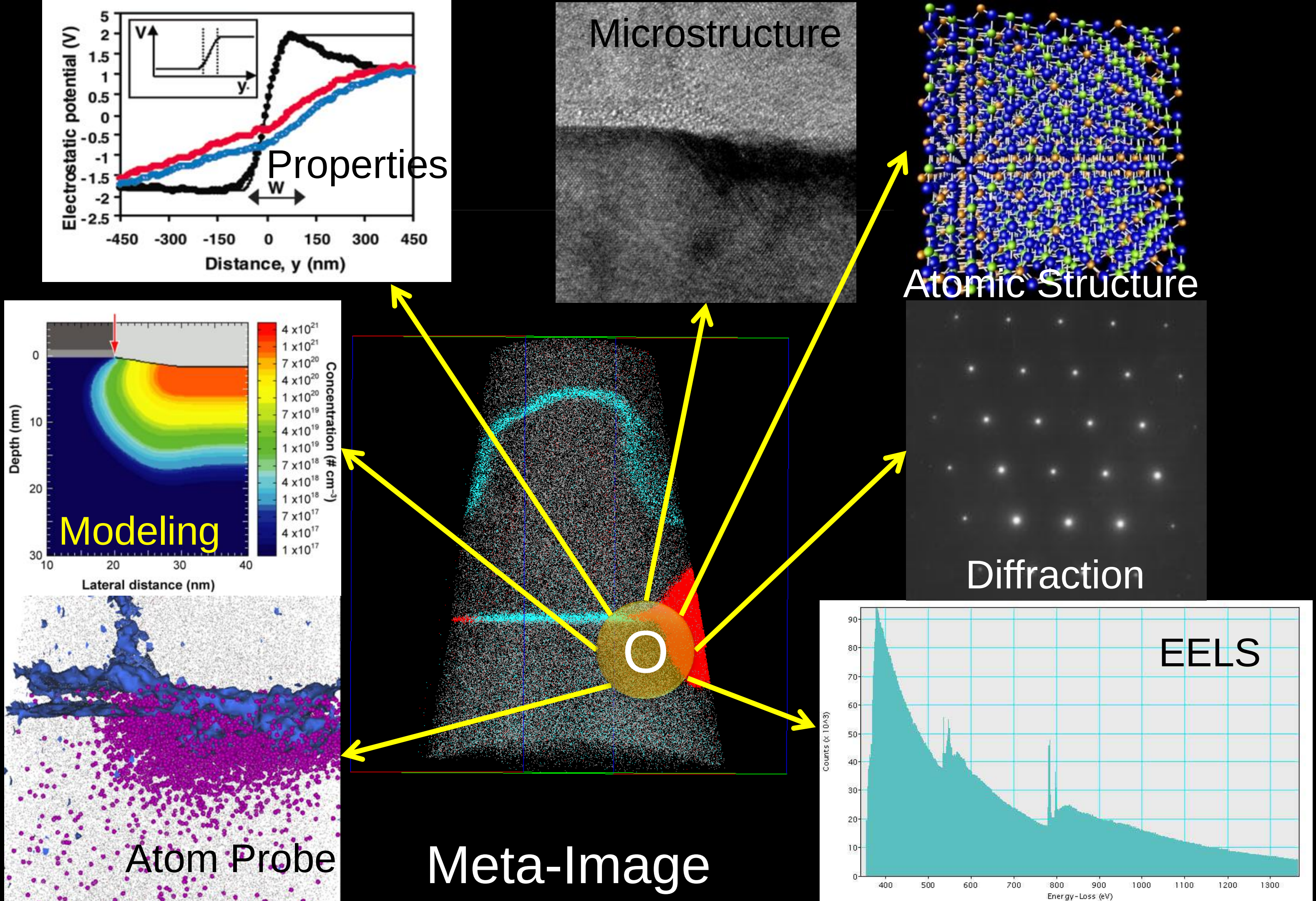
- Jeff Shepard
- David J. Larson
- Ty J. Prosa
- Brian P. Geiser
- Robert Ulfig
- Joseph H. Bunton

References

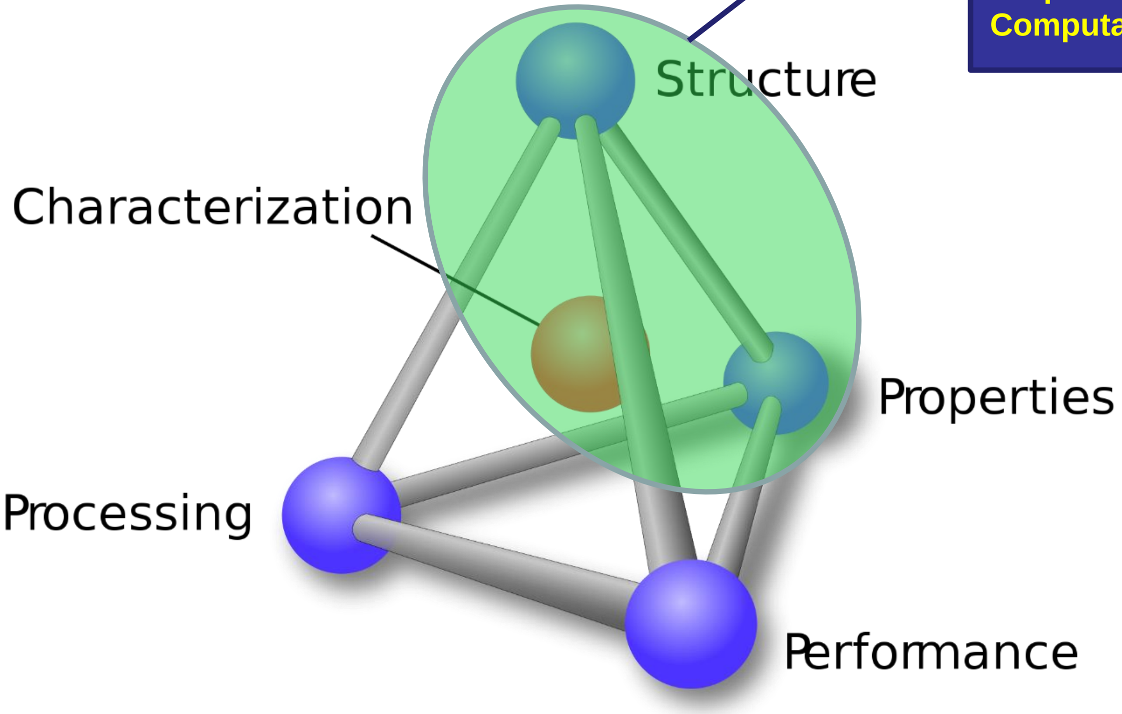
"Atomic-Scale Tomography: A 2020 Vision"
Thomas F. Kelly, Michael K. Miller, Krishna Rajan, and Simon P. Ringer, *Microscopy and Microanalysis*, Invited Review, vol. 19 (2013) pp. 652 – 664.

"Visions of Atomic-Scale Tomography"
Thomas F. Kelly, Michael K. Miller, Krishna Rajan, and Simon P. Ringer, *Microscopy Today*, May 2012, pp. 12-16.

"The Future of Atom Probe Tomography"
Michael K. Miller, Thomas F. Kelly, Krishna Rajan, and Simon P. Ringer, *Materials Today*, April 2012, vol. 15, no. 4, pp. 158-165.


Properties
Electrostatic potential (V)
Distance, y (nm)
W
Microstructure
Atomic Structure
Modeling
Depth (nm)
Lateral distance (nm)
Concentration (# cm⁻³)
Diffraction
EELS
Counts (x 10^3)
Energy-Loss (eV)
Atom Probe
O
Meta-Image

Structure-Properties Microscopy



Atomic-Scale Tomography gives **Structure** which can be connected to **Properties** through **Computational Materials Science**

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Analytical Tomography = LEAP + STEM

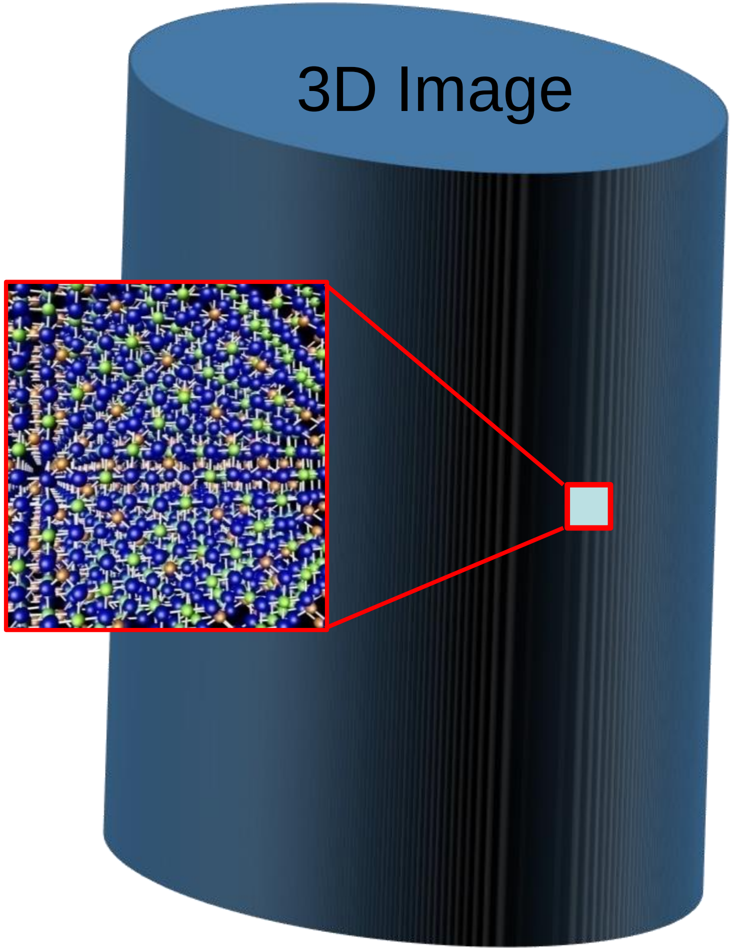
= Atomic-Scale Tomography?

- STEM tomography
  - Needle-shaped specimens
  - 360° rotation of needle
  - Full STEM image and analysis modes
- EELS adds chemical sensitivity
- Diffraction enhances structure certainty

+

- Atom probe tomography provides 3D atom positions
- Cryo specimen stage (20K)
- 1 ppm analytical sensitivity
- 0.2 nm spatial resolution in 3D

Atomic-Scale Tomography

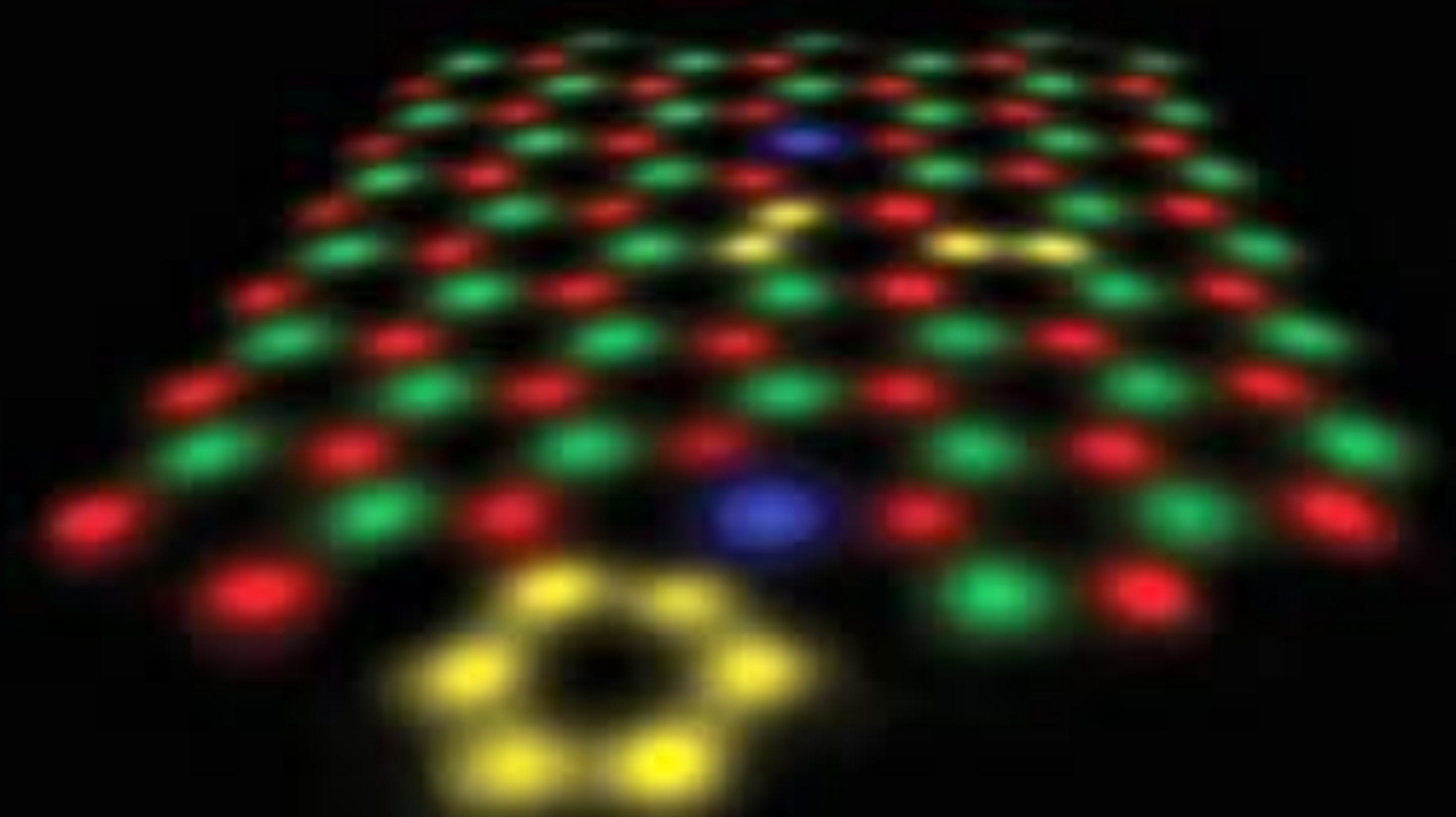


- Definition
  1. Atoms positioned with high precision
  2. 100% of atoms detected
     - Isotopic identity valuable at times
  3. Atoms identified with high precision
  4. Discrete 3-D image for a large volume ($500 \times 500 \times 500$ nm^3, i.e., billion atoms)

Kelly, Miller, Rajan and Ringer, *Microscopy and Microanalysis* (2013) **19(03)**, pp 652 – 664. DOI: http://dx.doi.org/10.1017/S1431927613000494.

Why Atomic-Scale Tomography?

- Characterization of small objects (<100 atoms) requires knowledge of every atom
  - Single atoms affect performance of atomic-scale devices
- Atomic structure (crystallography, radial distribution function) can be crucial for atomic-scale structures
- Atomic materials design requires characterization-based feedback
- Organic structures resolved better with 100% of atoms
- Maybe defect structures will be fully resolved
  - Grain boundaries
  - Dislocations
  - Vacancy distributions



Ondrej L. Krivanek, Matthew F. Chisholm, Valeria Nicolosi, Timothy J. Pennycook, George J. Corbin, Niklas Dellby, Matthew F. Murfitt, Christopher S. Own, Zoltan S. Szilagyi, Mark P. Oxley, Sokrates T. Pantelides & Stephen J. Pennycook
Atom-by-atom structural and chemical analysis by annular dark field electron microscopy. Nature 464, 571–574 (2010).

TEM and Atomic-Scale Tomography

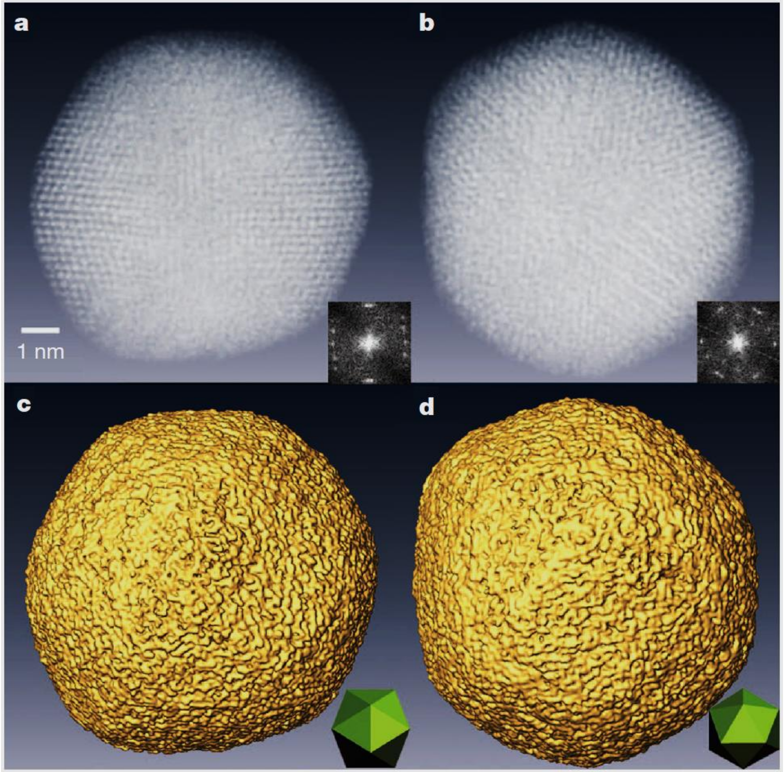


Figure 3 | 3D structure of the reconstructed gold nanoparticle. a, b, 3D volume renderings of the nanoparticle and their Fourier transforms (insets) at the two-fold (**a**) and three-fold (**b**) symmetry orientations. **c, d,** Iso-surface renderings of the nanoparticle at the two-fold (**c**) and three-fold (**d**) symmetry orientations. Insets show a model icosahedron at the same orientations.

doi:10.1038/nature10934

Electron tomography at 2.4-ångström resolution

M. C. Scott[1]*, Chien-Chun Chen[1]*, Matthew Mecklenburg[1]*, Chun Zhu[1], Rui Xu[1], Peter Ercius[2], Ulrich Dahmen[2], B. C. Regan[1] & Jianwei Miao[1]

444 | NATURE | VOL 483 | 22 MARCH 2012

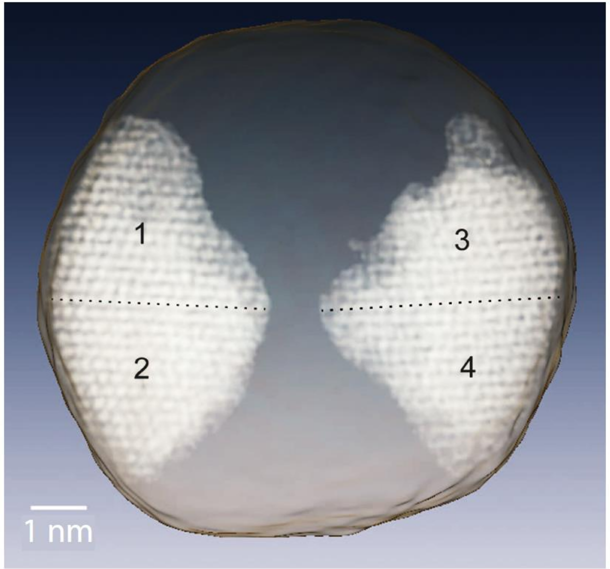
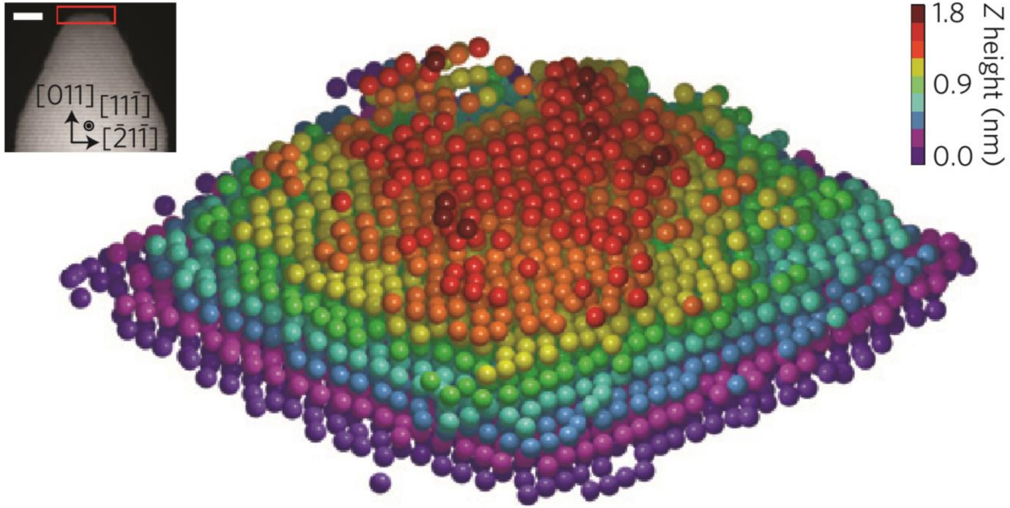


Figure 4 | Identification of four major grains inside the gold nanoparticle in three dimensions. Grains 1, 2 and grains 3, 4 are related by mirror-reflection across the horizontal interfaces marked by dotted lines. The angle enclosed by close-packed planes across these interfaces was measured to be 69.9° ± 0.8° between grains 1 and 2, and 71.3° ± 0.8° between grains 3 and 4, both of which are consistent with the angle for a face-centred cubic twin boundary (70.53°).

Electron Tomography & 3D Atom Positions



nature materials
LETTERS
PUBLISHED ONLINE: 21 SEPTEMBER 2015 | DOI: 10.1038/NMAT4426

Three-dimensional coordinates of individual atoms in materials revealed by electron tomography

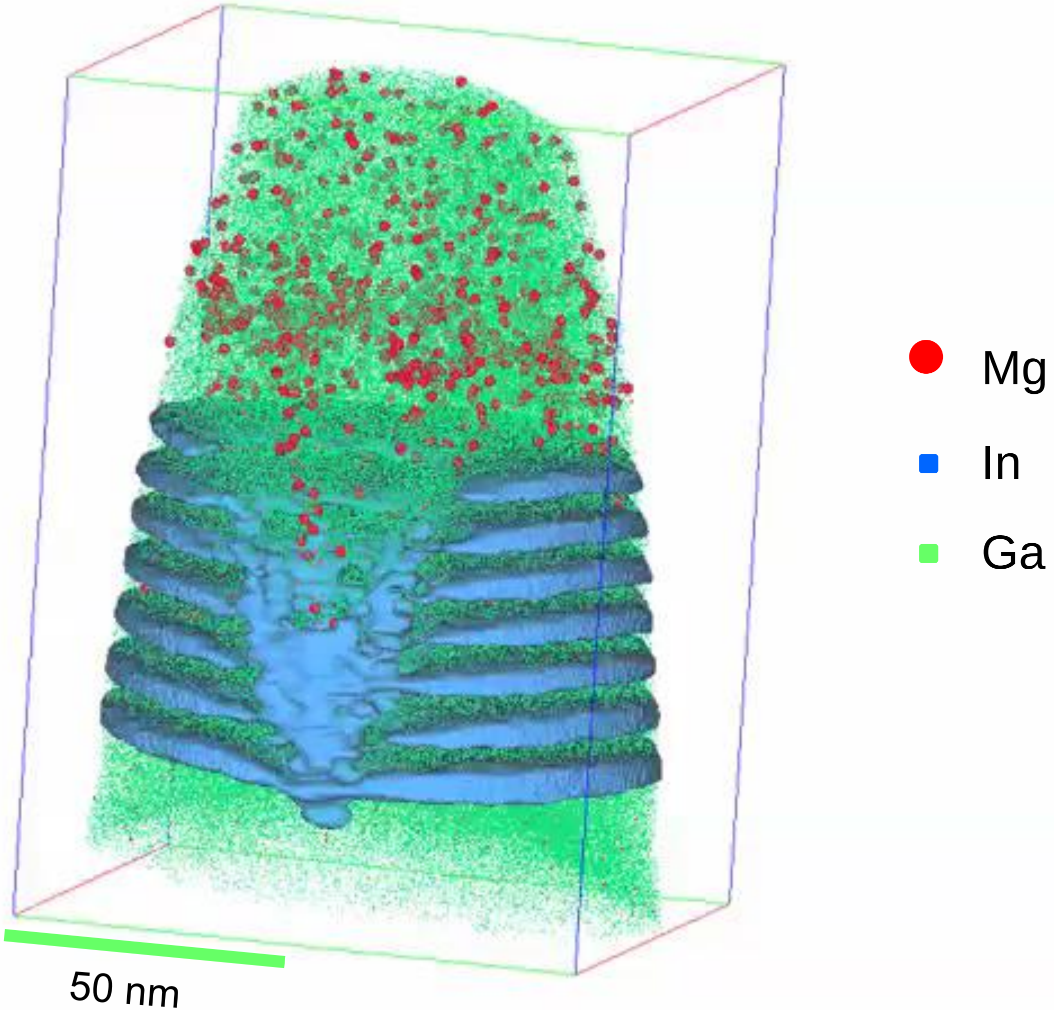
Rui Xu[1†], Chien-Chun Chen[1,2†], Li Wu[1†], M. C. Scott[1†], W. Theis[3†], Colin Ophus[4†], Matthias Bartels[1], Yongsoo Yang[1], Hadi Ramezani-Dakhel[5], Michael R. Sawaya[6], Hendrik Heinz[5], Laurence D. Marks[7], Peter Ercius[4] and Jianwei Miao[1†*]

- 3769 Tungsten atoms
  - 9 atomic layers
- Strain increases toward surface
  - May be presence of carbon
- Missing tungsten atom may be a carbon atom

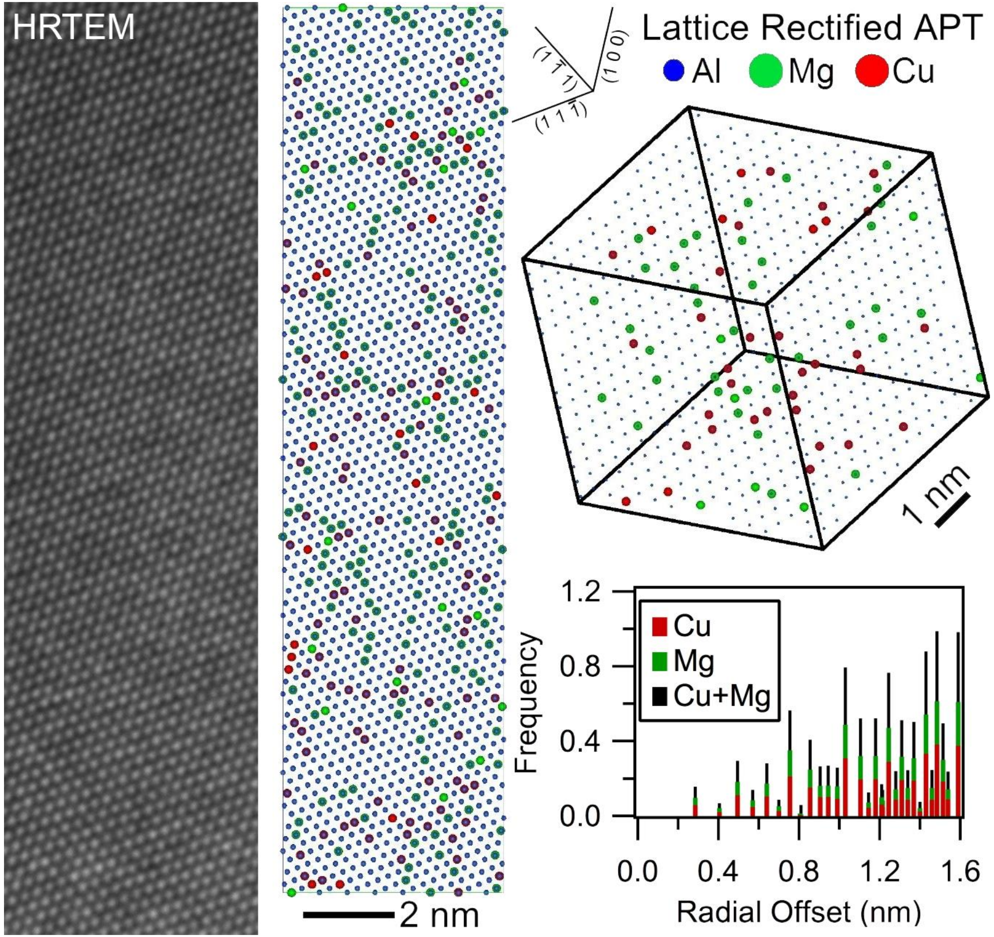
GaN LED for Lighting

- 3D image ☺
- 30 million atoms ☺

- 60% of the atoms ☺☹
- Spatial Resolution, δr
  - 0.1 nm < δr < 3 nm
    ☺ ☹



APT and Atomic-Scale Tomography



Lattice Rectification of Al-Zn-Cu-Mg alloy

M. P. Moody *et al.*, *Micros. Microanal.* **17**, 226 (2011).

- 40% of atoms are assigned by inference
- Limited to low-aberration regions
- Limited to simple crystal structures

Toward Atomic-Scale Tomography (AST)

What is needed for

APT → AST?

- Definition of AST
  1. Atoms positioned with high precision
     - Distortions corrected
  2. 100% of atoms detected
     - Isotopic identity valuable at times
  3. Atoms identified with high precision
  4. Discrete 3-D image for a large volume (500x500x500 nm^3, i.e., billion atoms)

To achieve AST with APT:

A. need complementary information

B. need new detector

B. need new detector

B. need new detector

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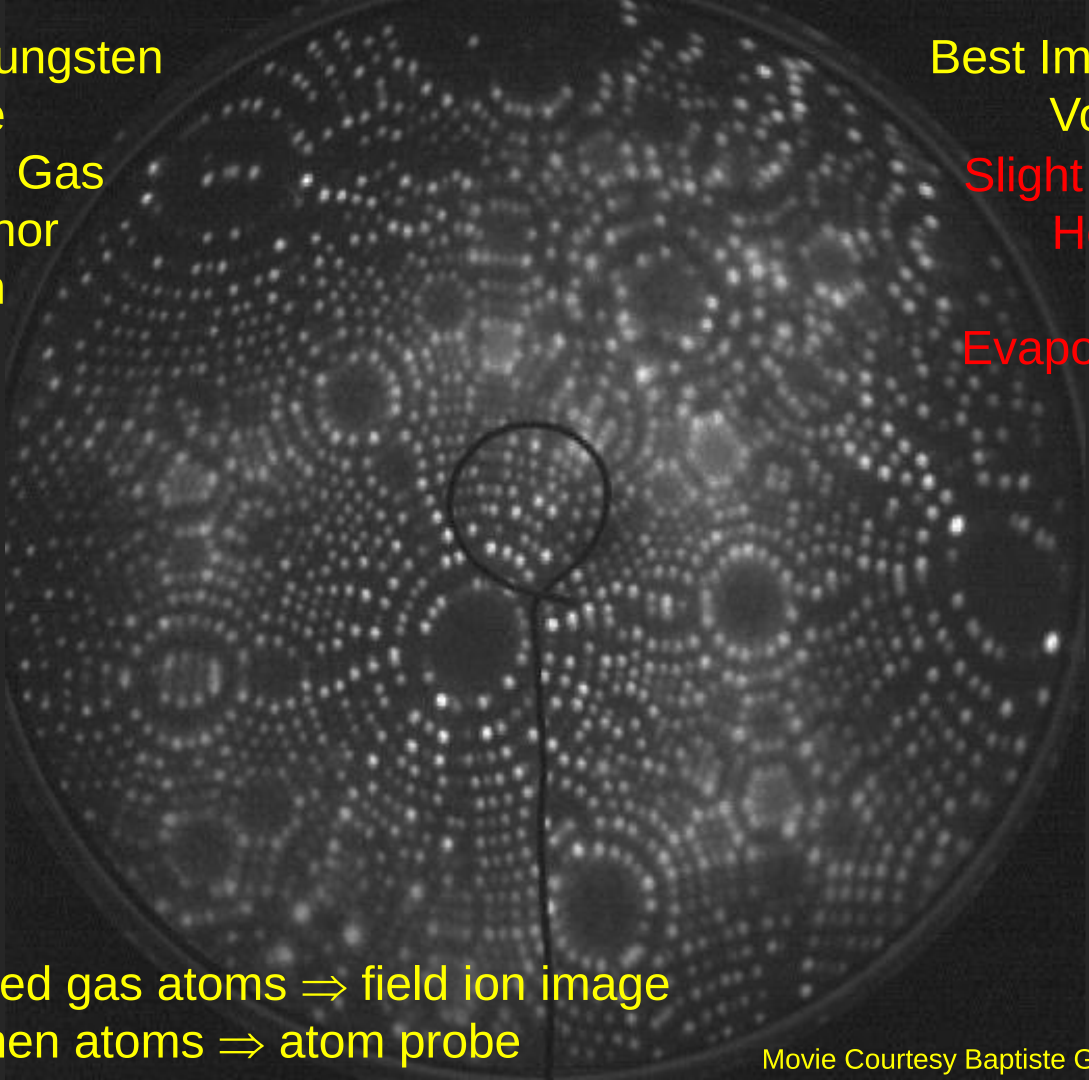
40 K Tungsten Needle
Helium Gas
Phosphor Screen

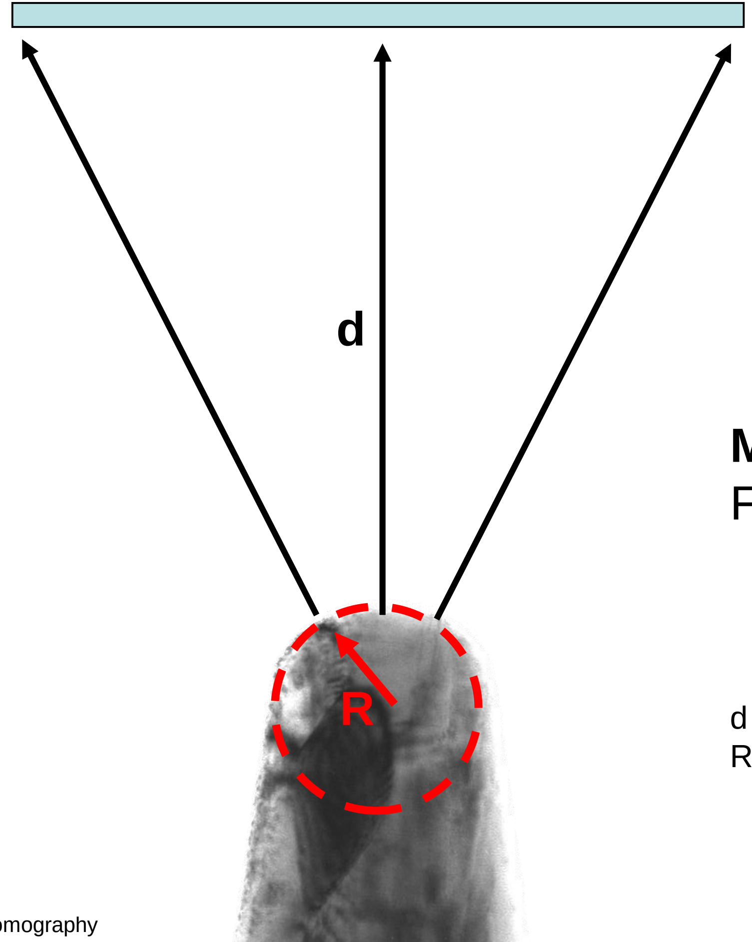
Best Imaging Voltage
Slight Laser Heating Field Evaporation



Adsorbed gas atoms ⇒ field ion image
Specimen atoms ⇒ atom probe

Movie Courtesy Baptiste Gault

Magnification of the Surface



Projection Microscope

Mag ~ d /R = 1 million X
For spherical endcap on conic frustum

d = constant distance from sample to detector (100 mm)
R = sample radius (e.g., 100 nm)

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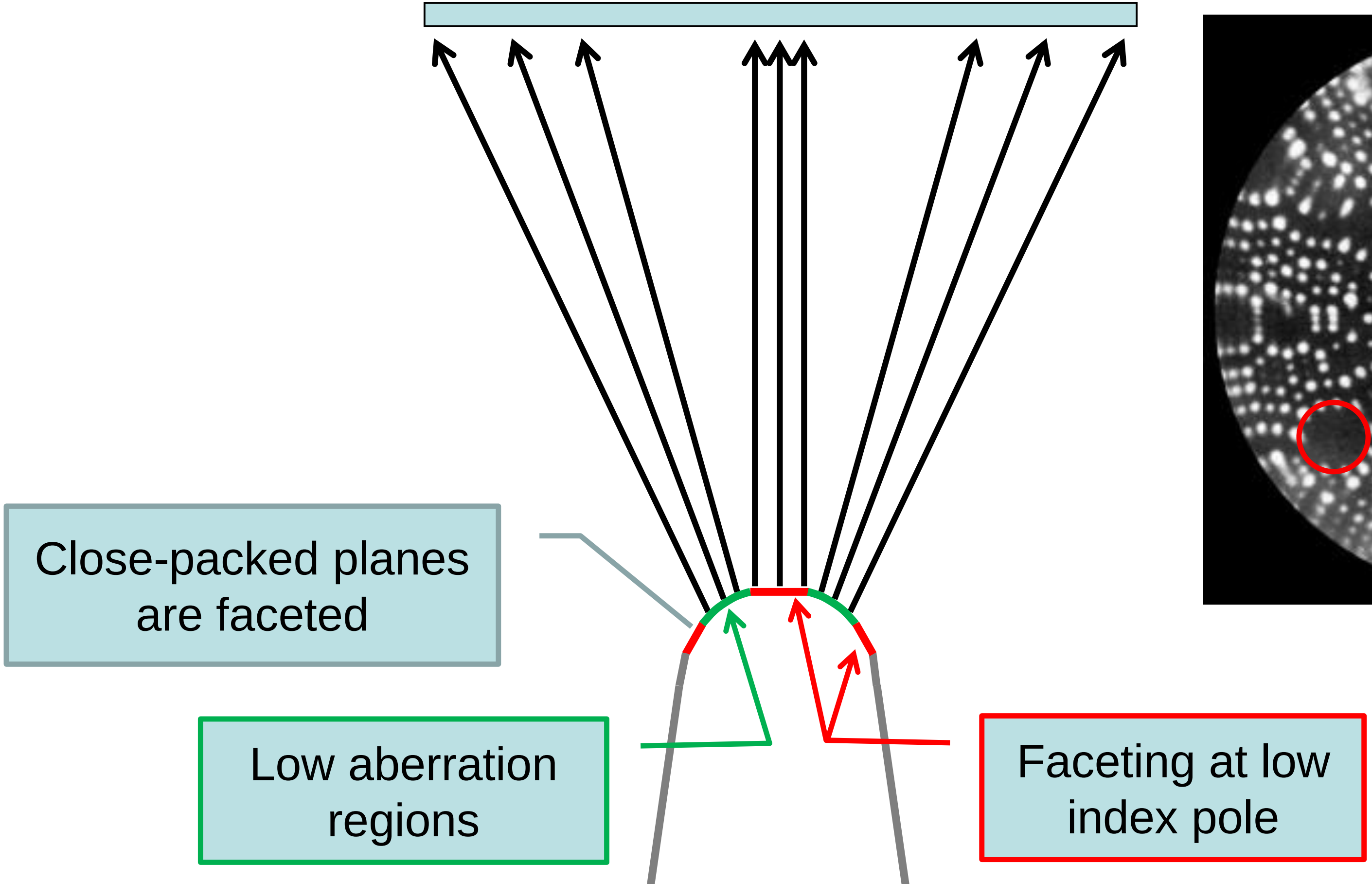
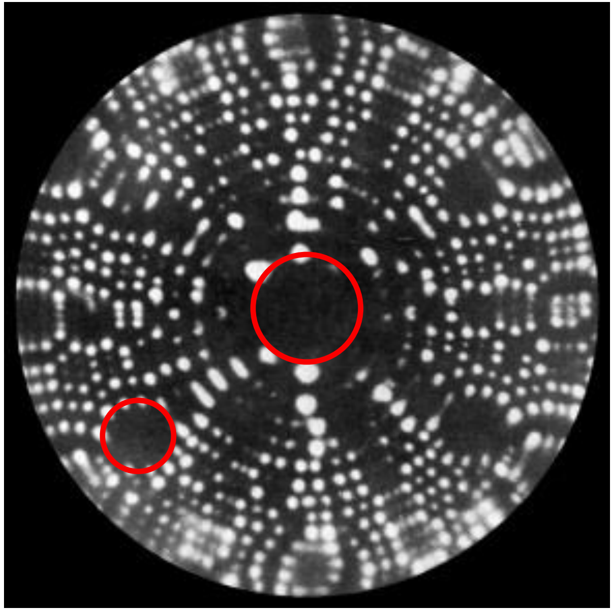
Monophase Distortions





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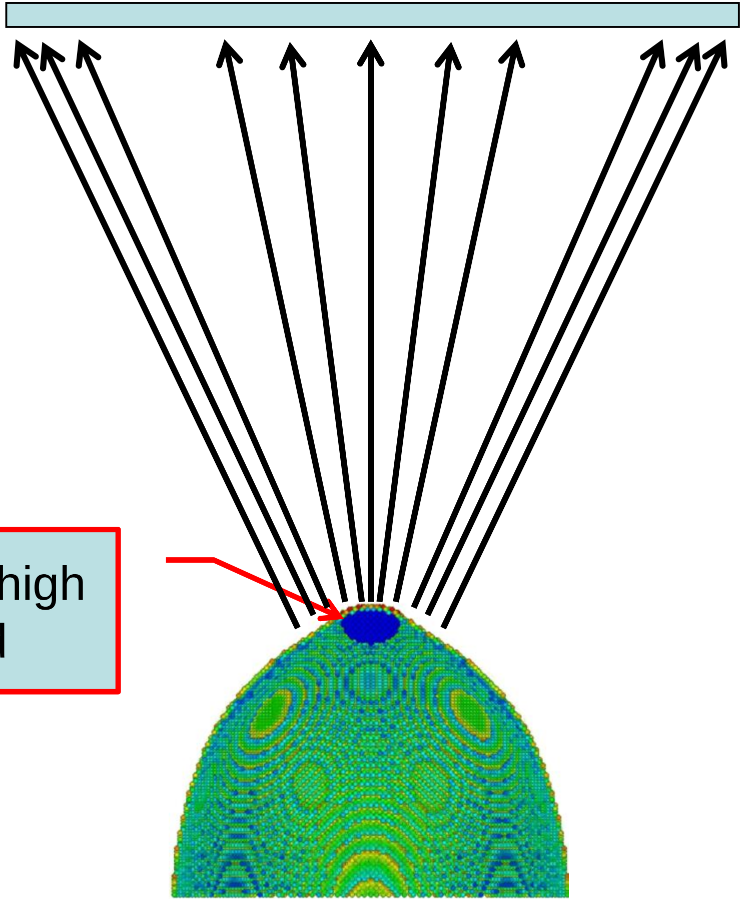
Polyphase Distortions



Second phase with high evaporation field

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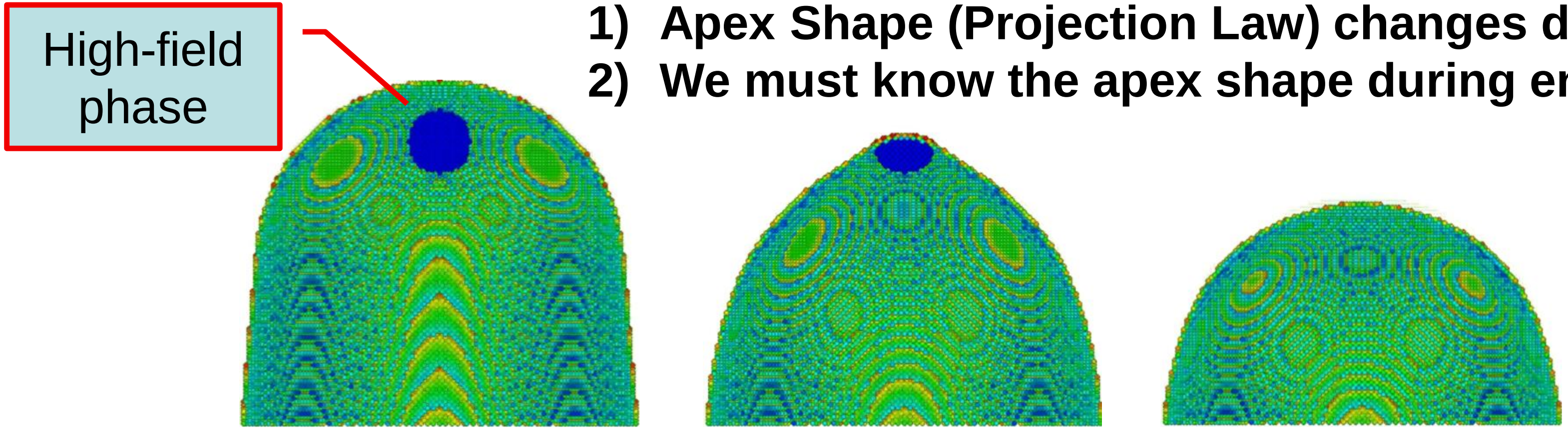
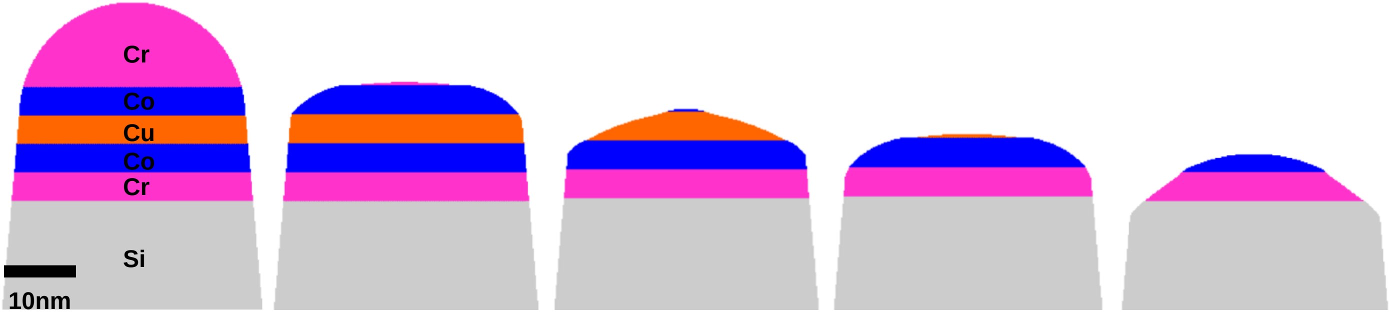
Second Phases Cause Non-Spherical Endforms

1) Apex Shape (Projection Law) changes during run
2) We must know the apex shape during entire run



O. Dimond, *Part II Thesis* (University of Oxford) (1999)

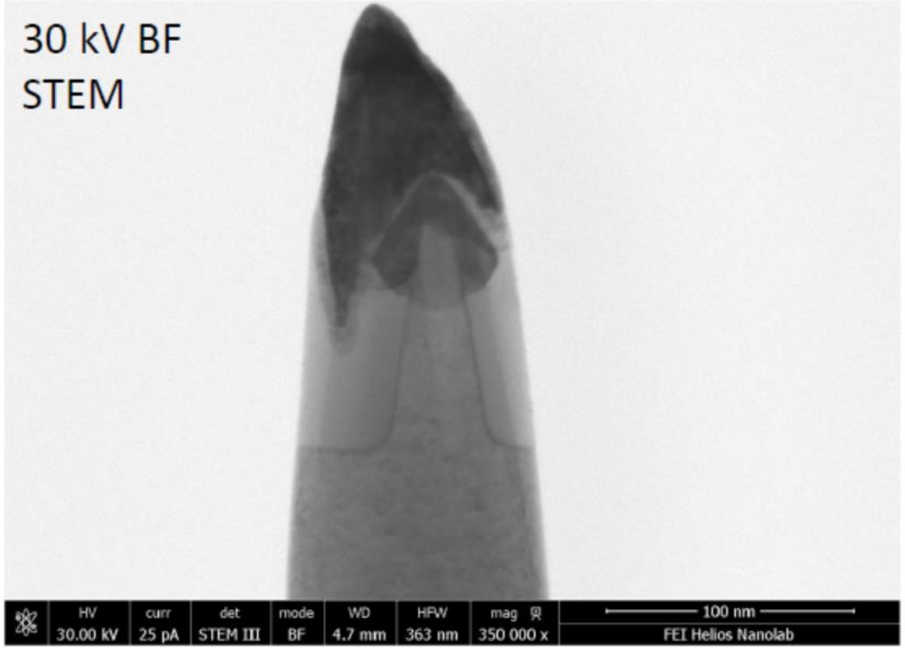


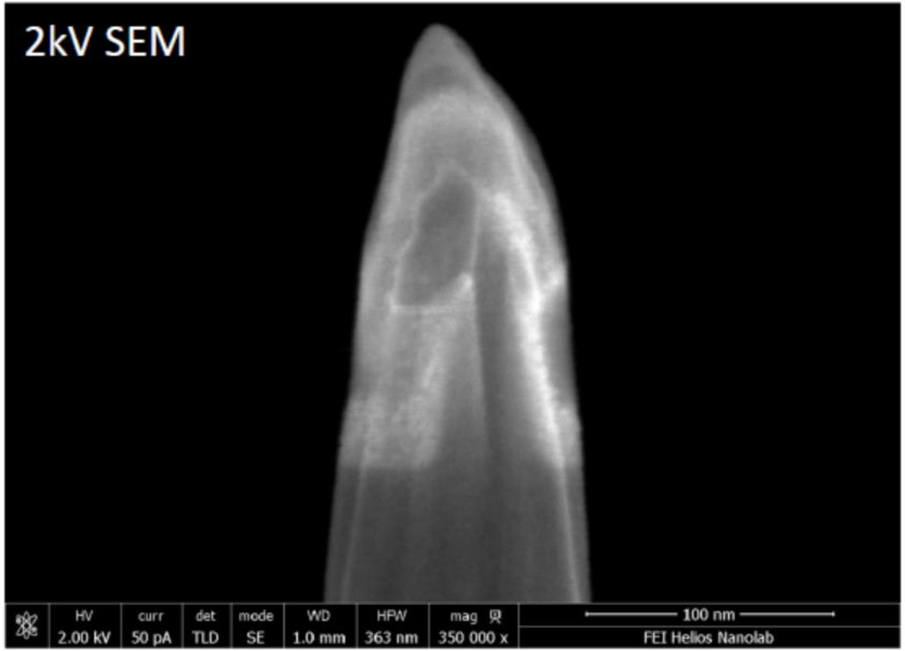
D. J. Larson et al., *Ultramicroscopy* 111 (2011) 506

Determination of Ion Trajectories (Specimen Apex Shape)

- Microscopy
  - Electron microscopy
    - TEM/STEM
    - SEM
    - …
  - Scanning probe microscopy
  - Field ion microscopy
- Simulation
  - Can simulation of field evaporation be good enough?
    - Iteration with actual data
    - Today’s algorithms are not sufficient
    - Need 100X increase in computing power or algorithm speed

STEM vs. SEM Comparison





Courtesy of FEI Nanoport

Outstanding Question

- What image resolution is sufficient? <1 nm
- It will depend on reconstruction and data processing algorithms
- Improved reconstruction will put atoms closer to correct position
- Lattice Rectification may be able to finish the job
  - Knowledge of lattice (a priori or derived from data) constrains atom positions

M. P. Moody *et al.*, *Micros. Microanal.* **17**, 226 (2011).
A. Breen, unpublished research (2014).

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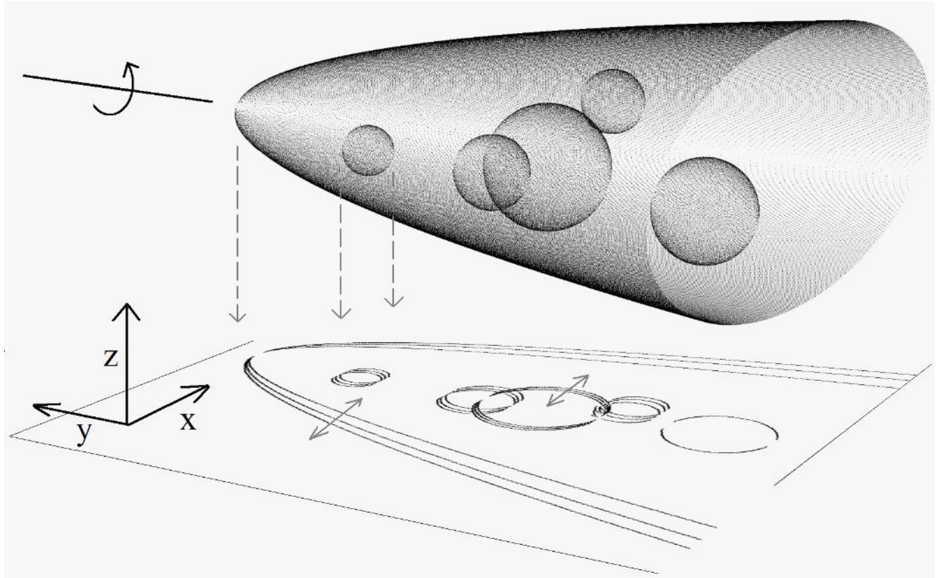
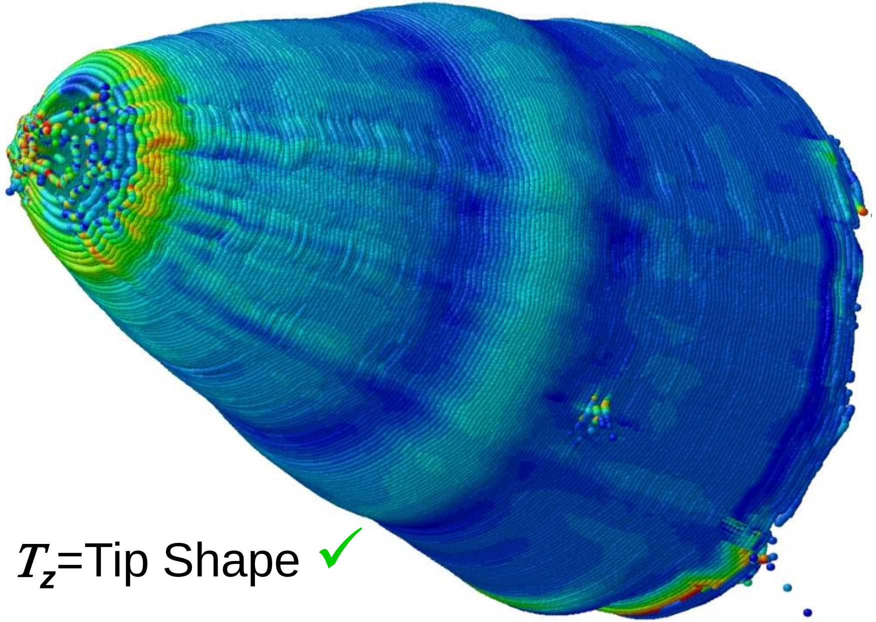
Tip Shape from Surface Tangent Algorithm





Petersen & Ringer, *Journal of Applied Physics*, **105**, 103518 (2009)
Petersen & Ringer, *Computer Physics Comms*, **181**, 676, (2010)

- Track the smooth movement of interest points in a tilt series.
- Need 8 images to determine surface shape.

- STA point cloud showing
  - Tip Shape
  - Mean Curvature (color scale)

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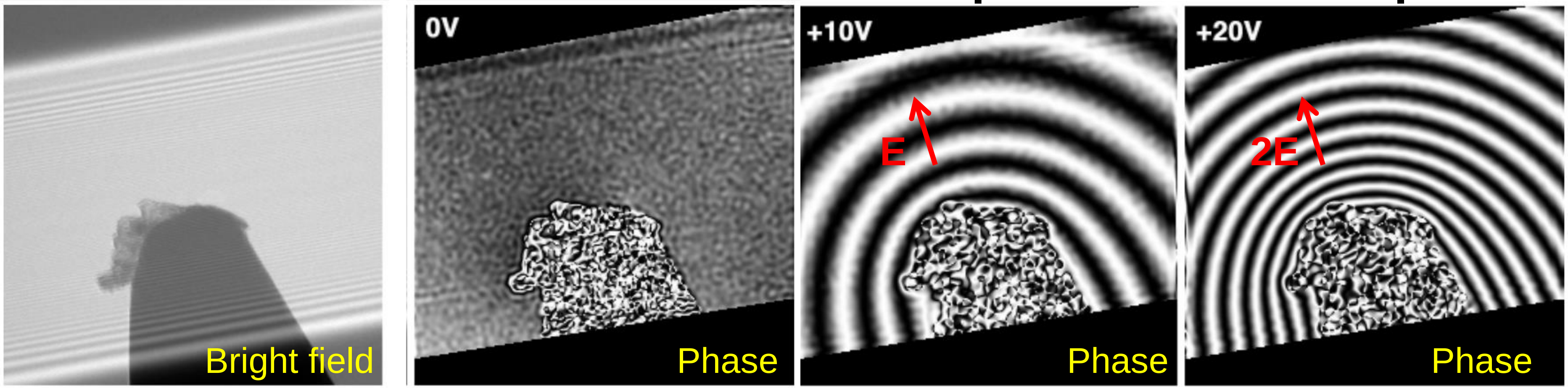
Holography of Tip at High Applied Field

Towards quantitative electron holographic mapping of the electric field around the tip of a sharp biased metallic needle

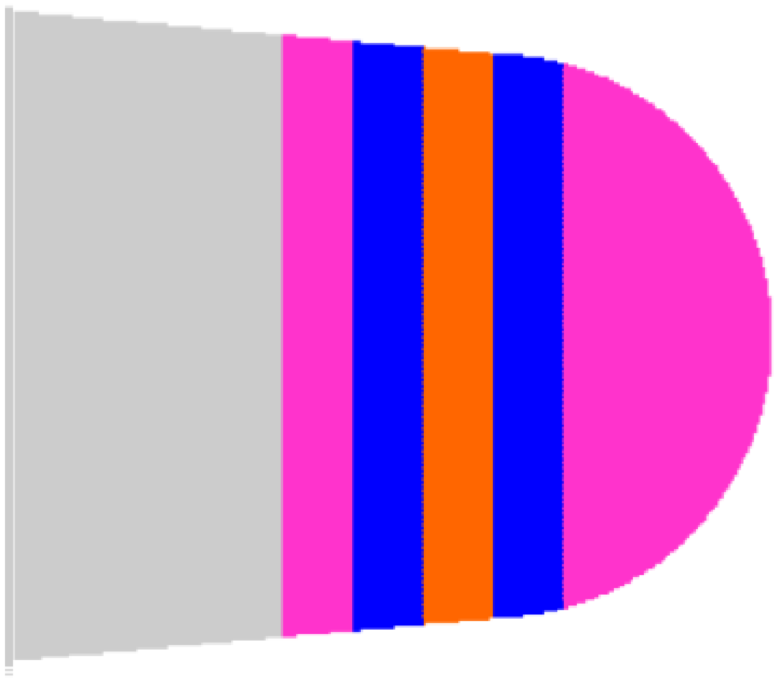
M. Beleggia,[1] T. Kasama,[2] D. J. Larson,[3] T. F. Kelly,[3] R. E. Dunin-Borkowski,[4] and G. Pozzi[5]
[1]) *Center for Electron Nanoscopy, Technical University of Denmark, Kongens Lyngby, Denmark*
[2]) *Center for Electron Nanoscopy, Technical University of Denmark, Kgs. Lyngby, Denmark*
[3]) *CAMECA Instruments, Inc., Madison, WI, USA*
[4]) *Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons and Peter Grünberg Institute, Forschungzentrum Jülich, Jülich, Germany*
[5]) *Department of Physics and Astronomy, University of Bologna, Bologna, Italy*

(Dated: 18 February 2014)

E_{ap} = on



Specimen Evolution Models



D. J. Larson et al., *Ultramicroscopy* 111 (2011) 506

- Experimental specimen apex shapes are expensive
- Simulated specimen apex shapes are expensive
- Interpolation of the apex shape between snapshots could solve this challenge
- Recent work is promising:
  - D. Haley, M.P. Moody, G.D.W. Smith, Microsc. Microanal., 19(06) (2013) 1709-1717. doi: 10.1017/S1431927613013299
  - D. Haley, T. Petersen, S.P. Ringer, G.D.W. Smith, J. Microscopy 244 (2011): 170–80. doi:10.1111/j.1365-2818.2011.03522.x.

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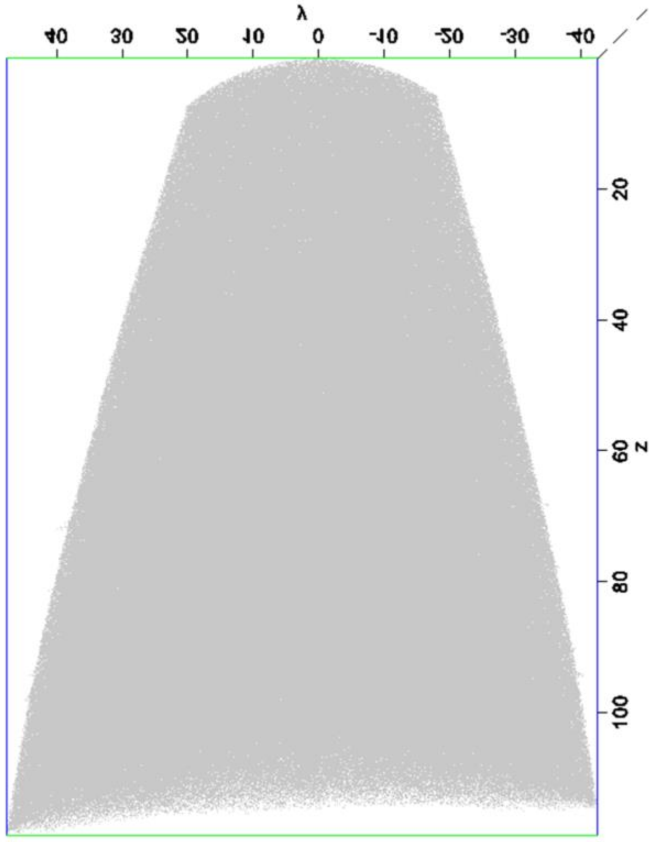
Interrupted Run



- Si Pre-sharpened Microtip
- Data were collected with field evaporation for 5 minutes, then field off for 5 minutes
- A new PSM was run in voltage mode in the reflectron tool at 15% PF, 1% DR, 50K, and 200 kHz pulse rate. The tip was run for 5 min, then the HV was disabled for 5 min.
- This cycle was repeated 20 times

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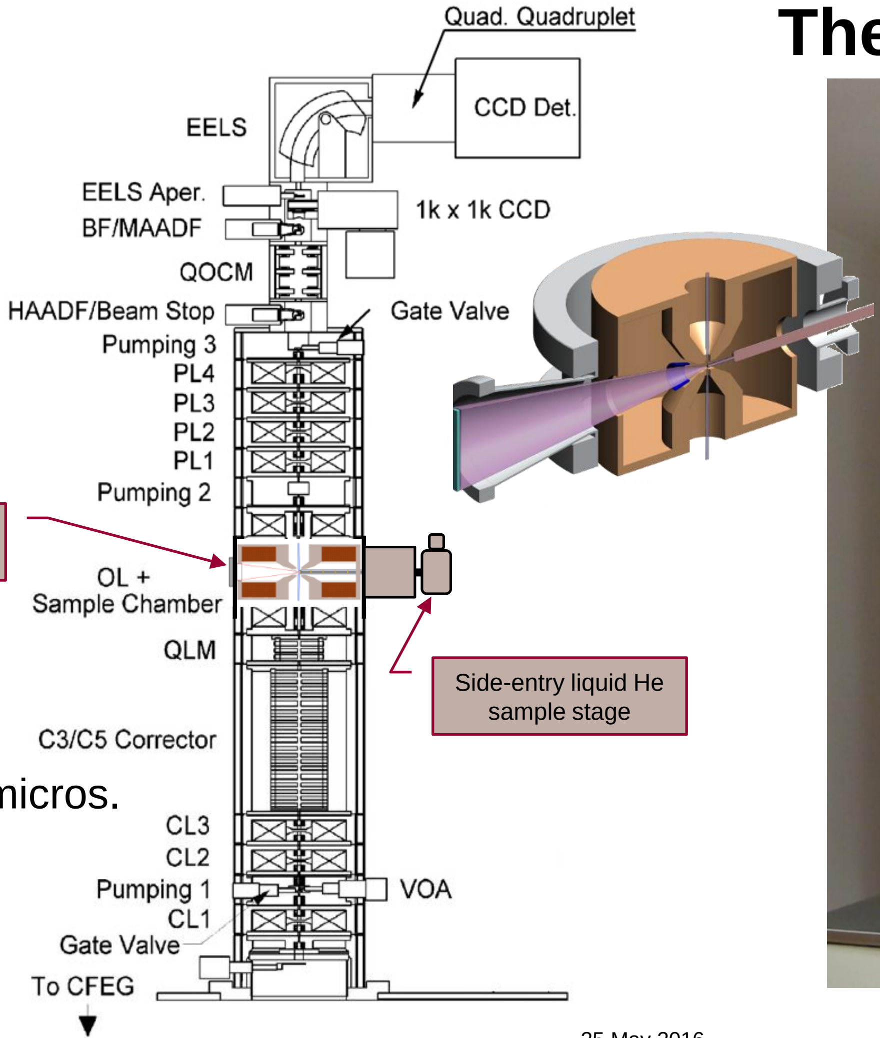
The ATOM Project

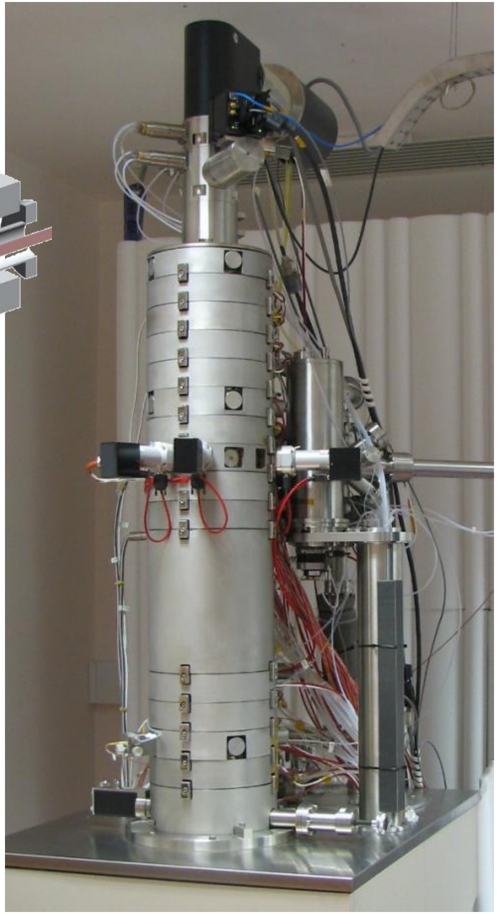
- Combine Nion STEM with LEAP
- Prepared proposal to US DOE in 2011
  - Kelly, Miller, Rajan, Ringer
- DOE budgets have not admitted to this level of project

The ATOM Project

STEM+LEAP

Build objective lens assembly with atom probe inside



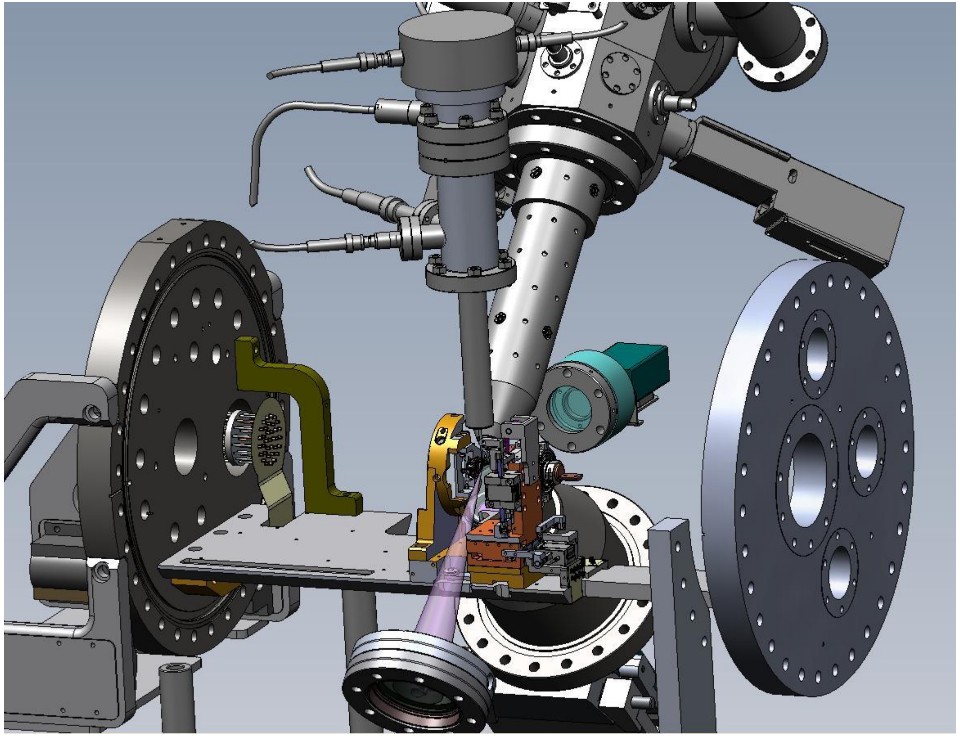


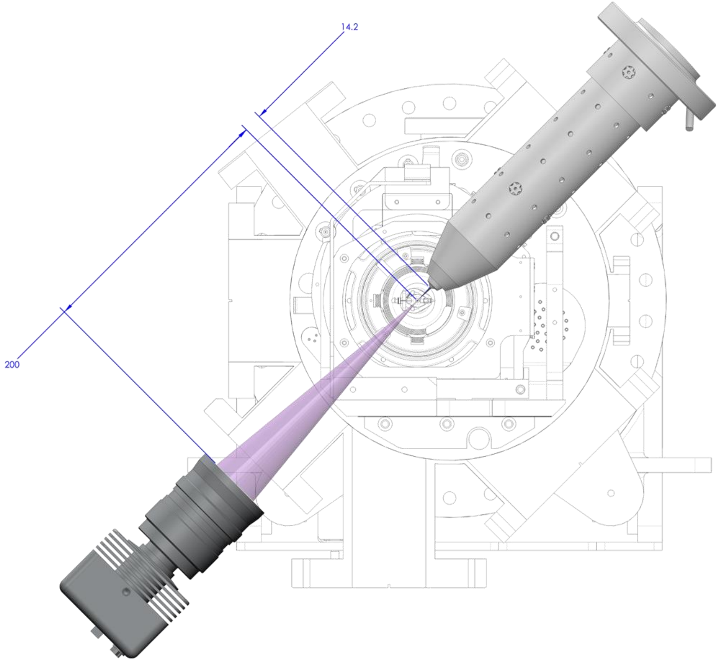
O. Krivanek et al., Ultramicros. 108 (2008) 179.

nion

Current Instrumentation Design

With Brian Gorman, David Dierks, Colorado School of Mines





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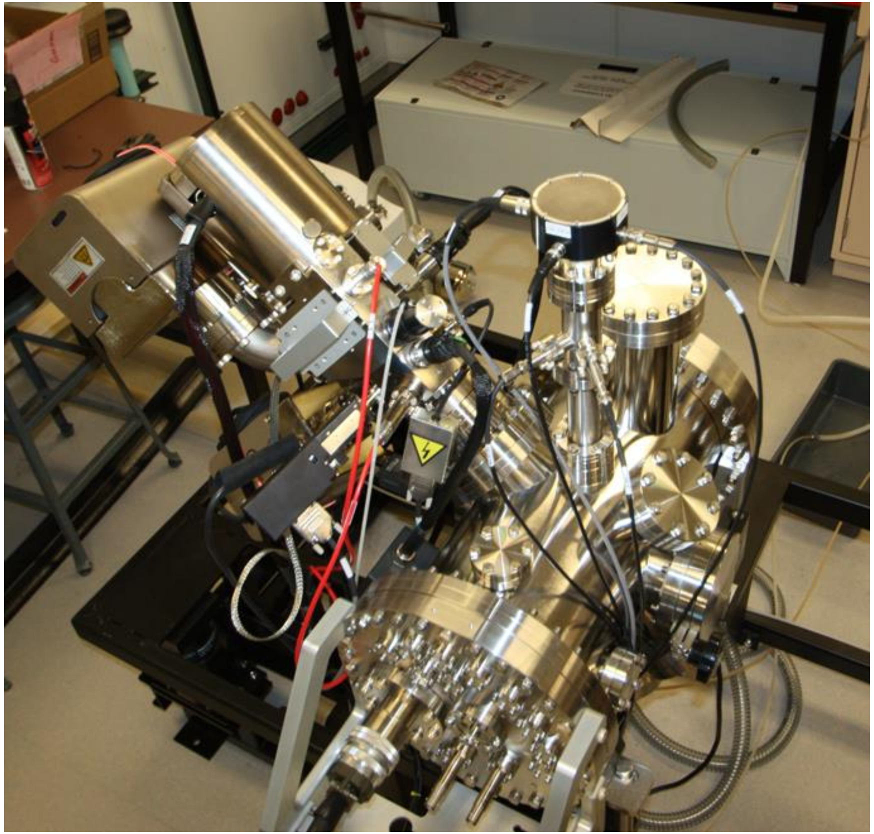
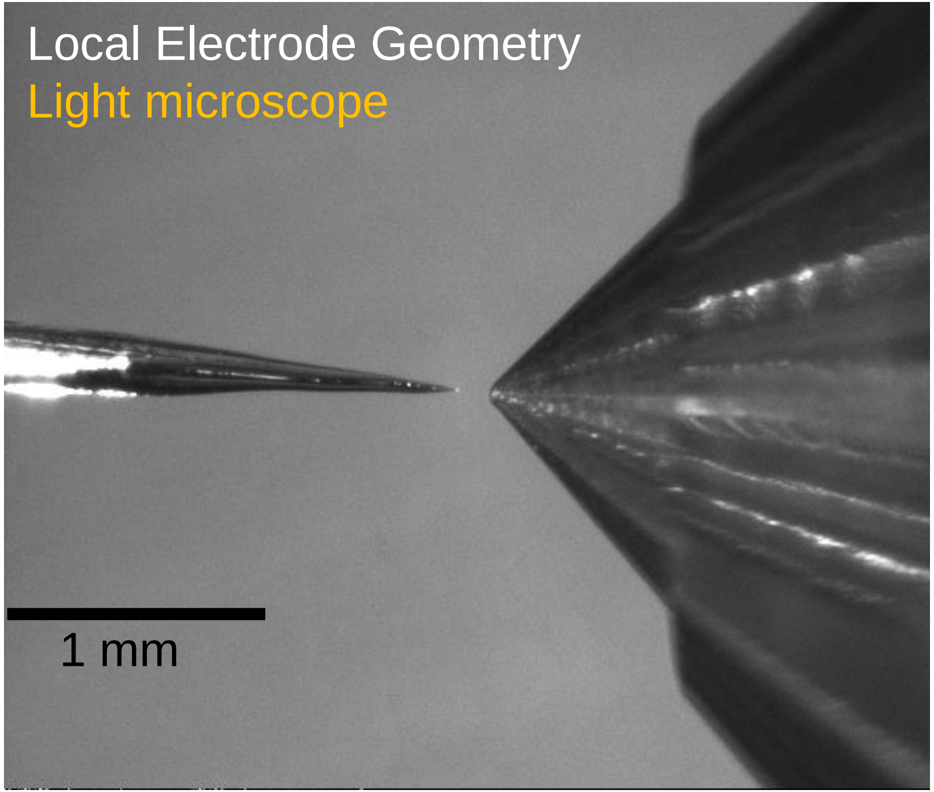
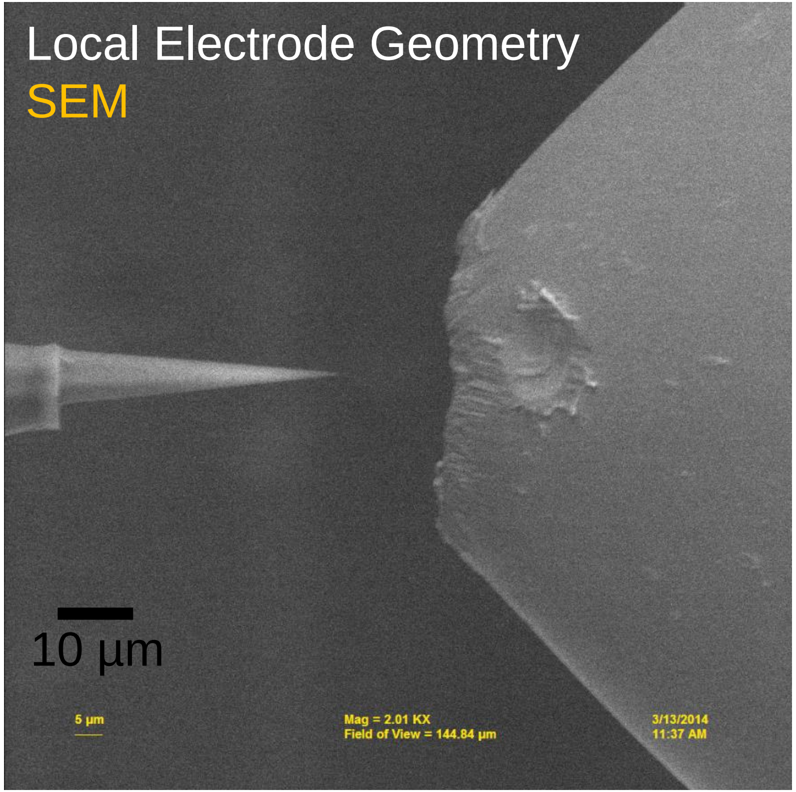
STEM+LEAP Experiment

With Brian Gorman, David Dierks, Colorado School of Mines



Colorado School of Mines Development System





The German Projects

- Project ULTRA (Ultimate Tomographic Reconstruction of Atoms)
  - This has split for now into two separate programs:
    - Project Tomo (LEAP-TEM) part of Project Ruska Center 2.0
      - Forschungszentrum Julich and Helmholtz Society/German Government
      - Proposal submitted in January 2016
    - Project LaPlace (LEAP-STEM)
      - Max Planck Dusseldorf and Max Planck Society
      - Two stage project:
        - » Stage I - Cryo-FIB-STEM-LEAP with cryo-UHV suitcase transport and Reaction Chamber
        - » Stage II – Build dedicated LEAP-STEM according to the concepts presented here

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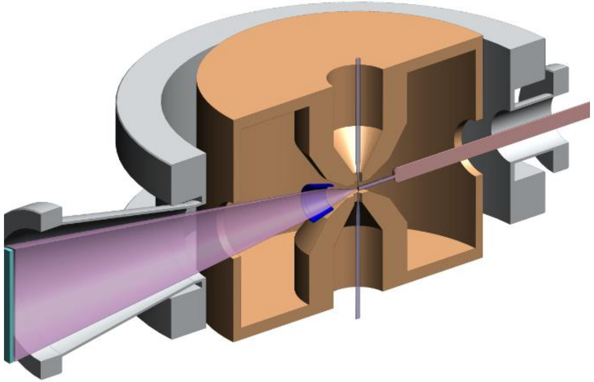
Project Tomo

TEM+LEAP



Build objective lens assembly with atom probe inside

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Summary of Imaging

- Record tomographic “snapshot” of specimen apex at regular intervals
- Use Specimen Evolution Models to interpolate surface between snapshots

- Result:
  Specimen apex shape during entire analysis
- Goal:
  Correct all atom positioning to <0.2 nm

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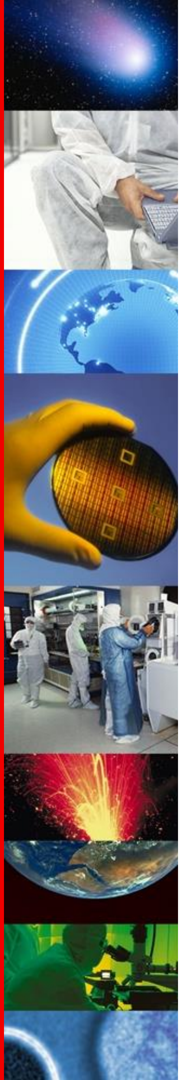

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Superconducting Detector Development Project

With Robert McDermott and Joseph Suttle, University of Wisconsin

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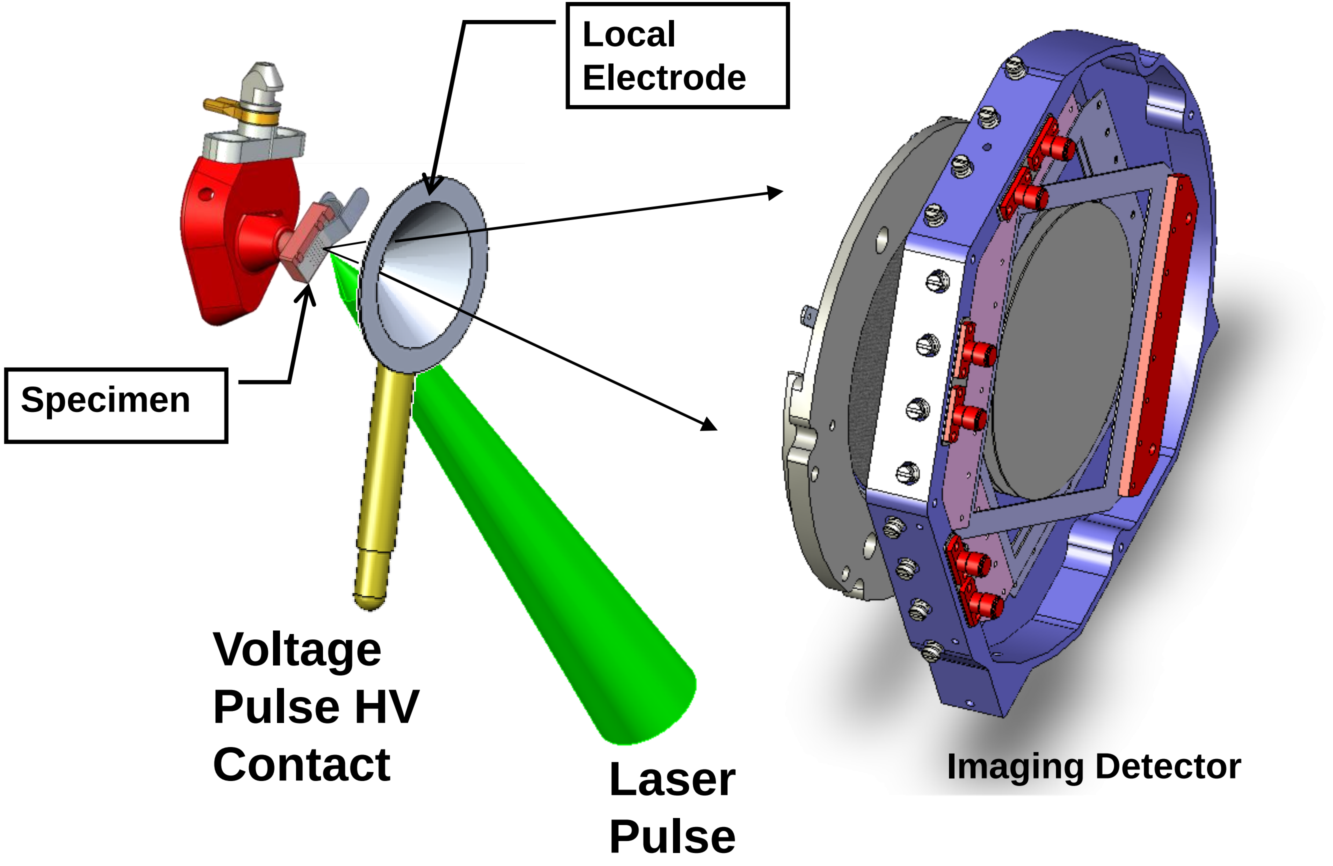
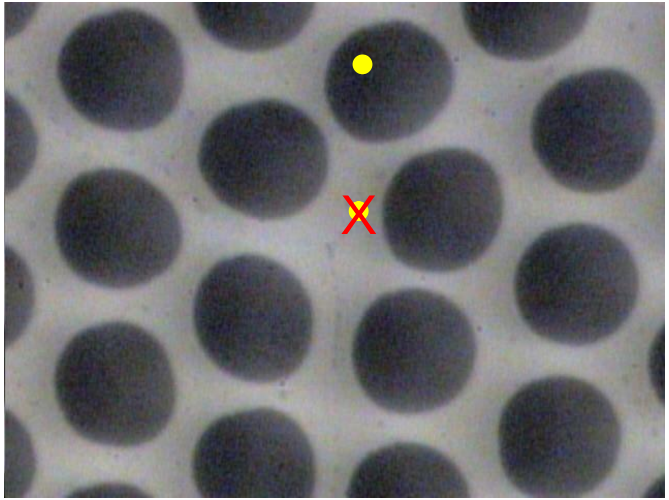
Atom Probe Microscope



MCP Construction and Performance

Conventional Pb-glass MCP



- Ions hitting in channel mostly get amplified
- Ions hitting on flat face mostly do not get amplified
- Sub 100 ps timing resolution possible
- High gain
- “No” variation in detection efficiency with atomic number

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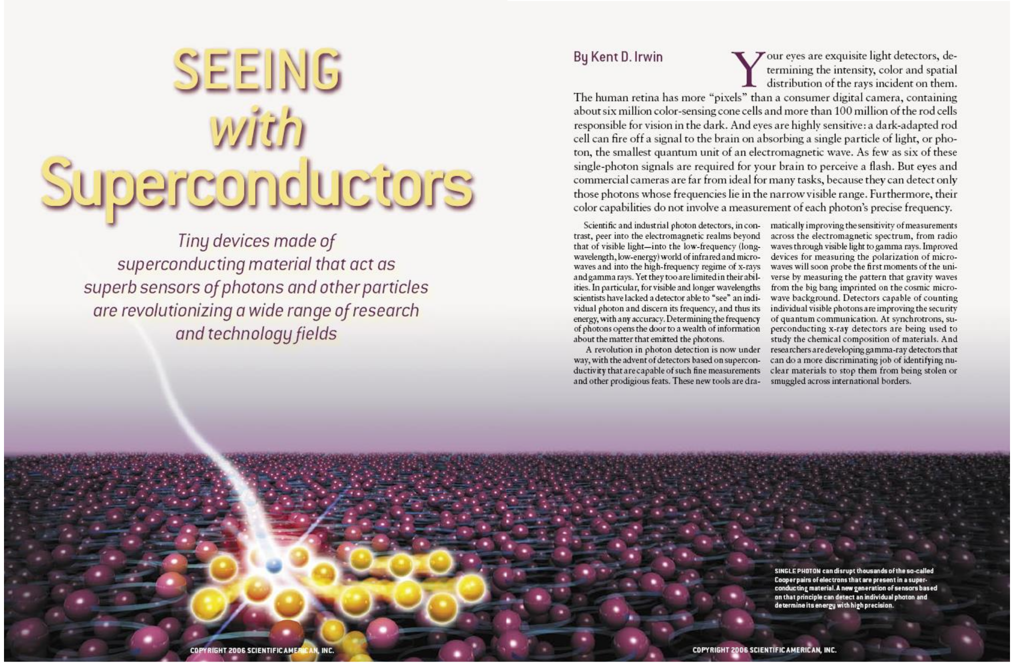
Why 100% Detection Efficiency?

- Improves sensitivity
- Needed for single atom sensitivity
- Needed for small cluster analysis
- Needed for improved crystallography
  - Needed if defect analysis is to be realized
- Needed for organic molecule identity
- Needed for atomic-scale tomography

Superconducting Detectors

Superconducting detector technology could enable 100% detection efficiency



SEEING with Superconductors

Tiny devices made of superconducting material that act as superb sensors of photons and other particles are revolutionizing a wide range of research and technology fields

By Kent D. Irwin

Your eyes are exquisite light detectors, determining the intensity, color and spatial distribution of the rays incident on them. The human retina has more "pixels" than a consumer digital camera, containing about six million color-sensing cone cells and more than 100 million of the rod cells responsible for vision in the dark. And eyes are highly sensitive: a dark-adapted rod cell can fire off a signal to the brain on absorbing a single particle of light, or photon, the smallest quantum unit of an electromagnetic wave. As few as six of these single-photon signals are required for your brain to perceive a flash. But eyes and commercial cameras are far from ideal for many tasks, because they can detect only those photons whose frequencies lie in the narrow visible range. Furthermore, their color capabilities do not involve a measurement of each photon's precise frequency.

Scientific and industrial photon detectors, in contrast, peer into the electromagnetic realms beyond that of visible light—into the low-frequency (long-wavelength, low-energy) world of infrared and microwaves and into the high-frequency regime of x-rays and gamma rays. Yet they too are limited in their abilities. In particular, for visible and longer wavelengths scientists have lacked a detector able to "see" an individual photon and discern its frequency, and thus its energy, with any accuracy. Determining the frequency of photons opens the door to a wealth of information about the matter that emitted the photons.

A revolution in photon detection is now under way, with the advent of detectors based on superconductivity that are capable of such fine measurements and other prodigious feats. These new tools are dramatically improving the sensitivity of measurements across the electromagnetic spectrum, from radio waves through visible light to gamma rays. Improved devices for measuring the polarization of microwaves will soon probe the first moments of the universe by measuring the pattern that gravity waves from the big bang imprinted on the cosmic microwave background. Detectors capable of counting individual visible photons are improving the security of quantum communication. At synchrotrons, superconducting x-ray detectors are being used to study the chemical composition of materials. And researchers are developing gamma-ray detectors that can do a more discriminating job of identifying nuclear materials to stop them from being stolen or smuggled across international borders.

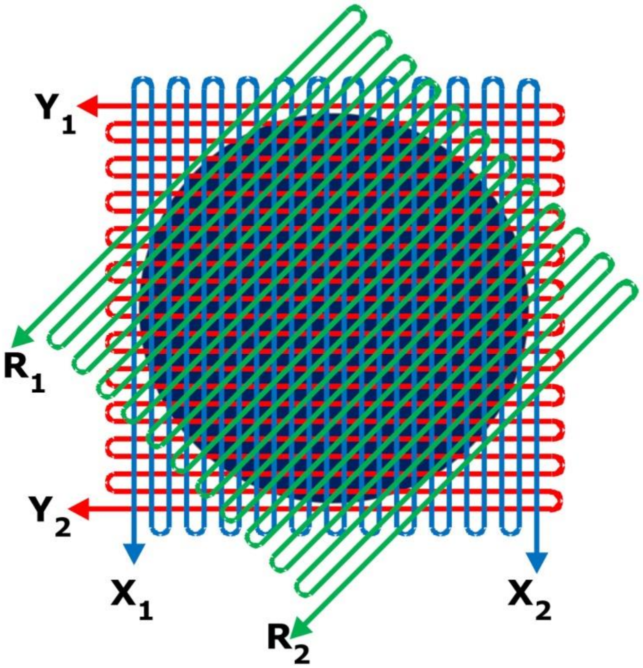
SINGLE PHOTON can disrupt thousands of the so-called Cooper pairs of electrons that are present in a superconducting material. A new generation of sensors based on that principle can detect an individual photon and determine its energy with high precision.

Kent D. Irwin, "Seeing with Superconductors," Scientific American, Nov. 2006, pp. 86-94.

- The fundamental properties of metallic superconductors are attractive and can do the job
- Superconductor band gap ~5meV
  - Large signal is generated
    - 5 keV ion generates 10^6 electrons
  - External amplifiers not needed
- The basic detection process takes ~ 30 ps
- The materials are well known, stable, and easy to deposit

Superconducting Detector Concepts

Double Meander Microwave Stripline Detector

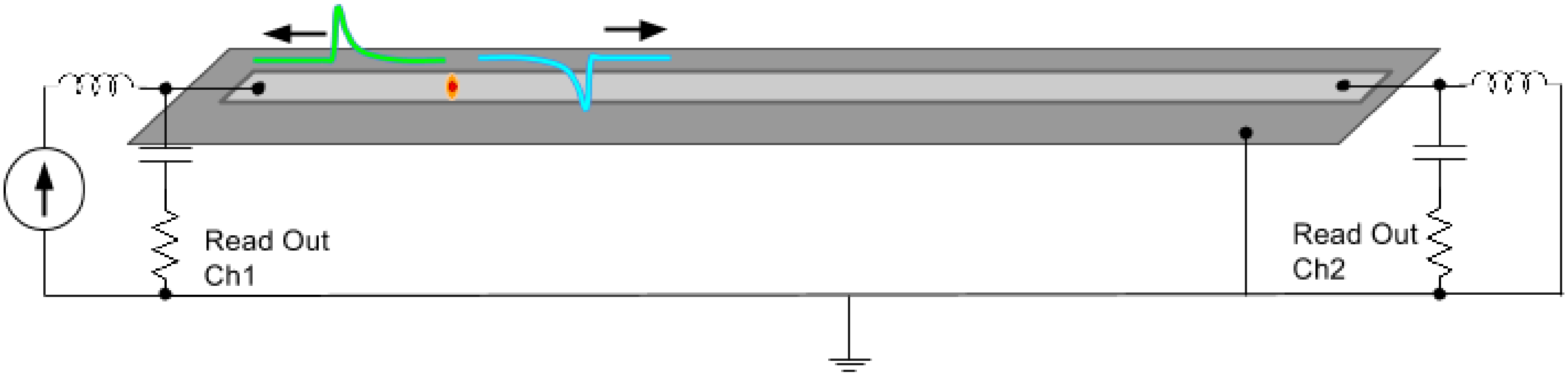


- The crossed delay line detector (XDL) has 2 or 3 electrodes
  - The third line is used to resolve multiple hits on a single evaporation pulse.
- Pulses from single hits are robust
- Timing resolution < 1ns expected
- 2D detectors currently being fabricated

Theory of Operation

New superconducting detector approach (McDermott and Suttle)

- Transmission lines can be expanded in size without loss of data collection rate
- By expanding to a single, continuous transmission line, we gain access to position information



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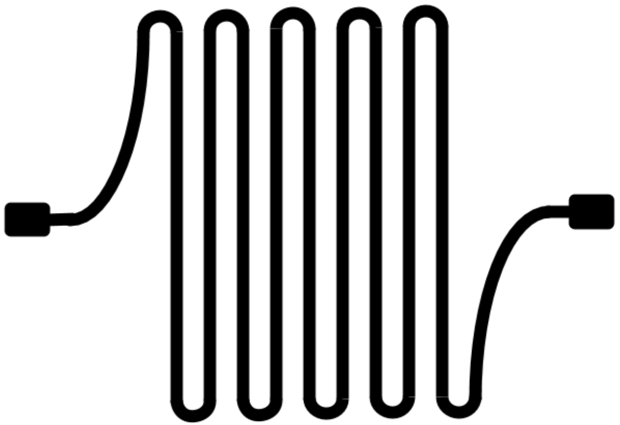
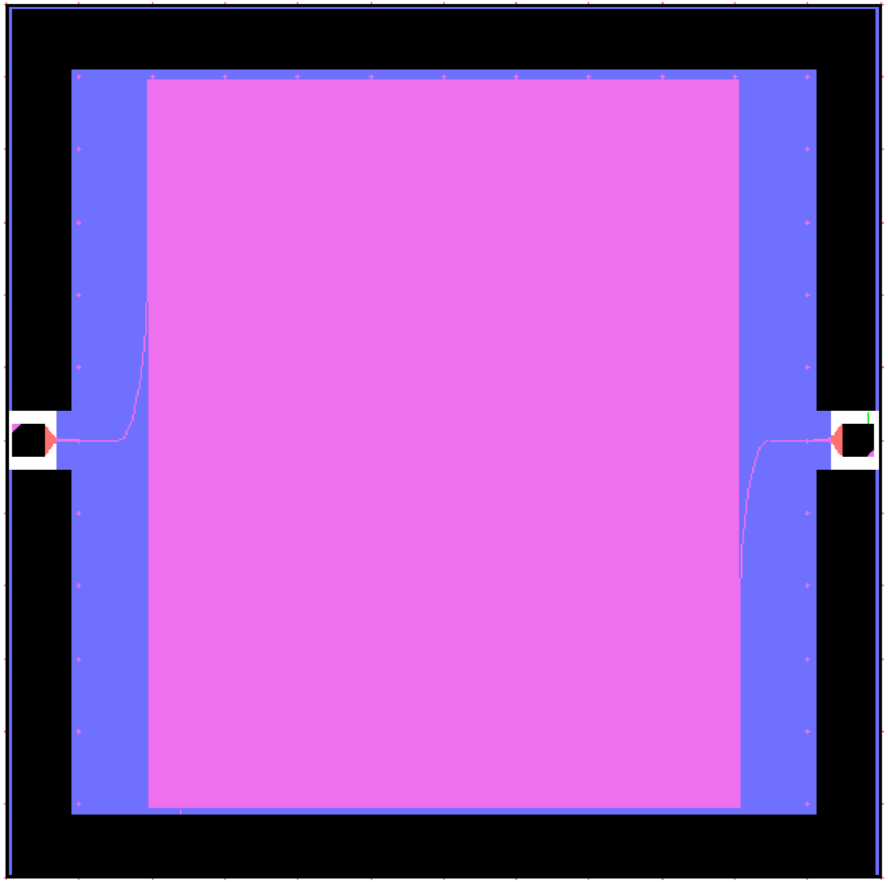
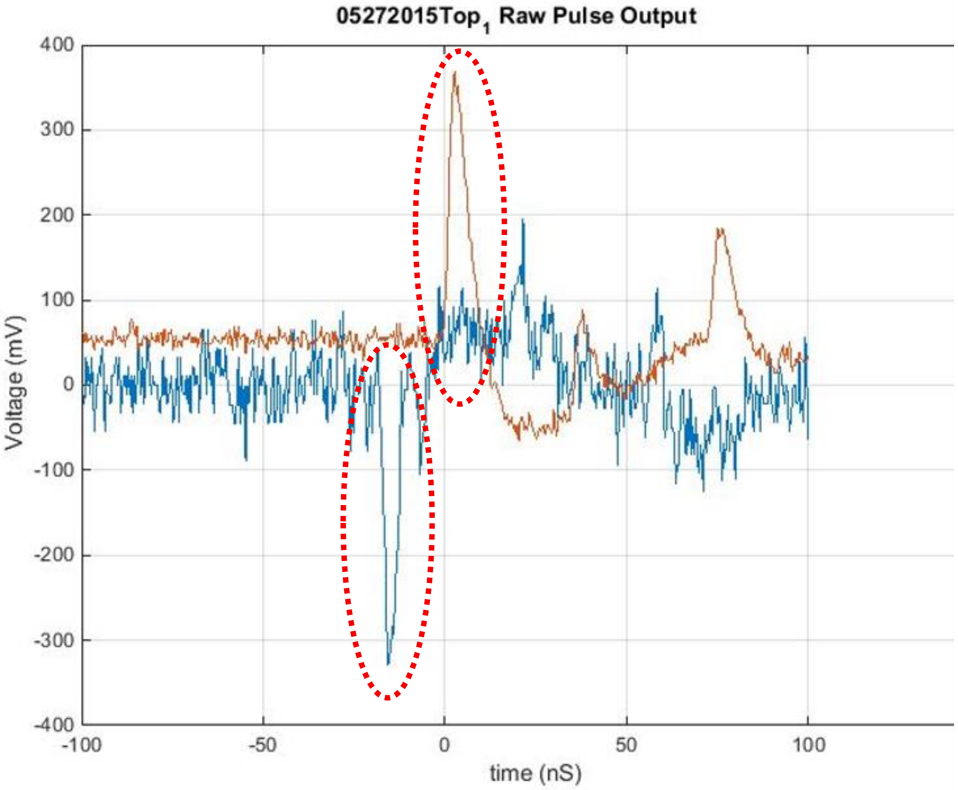
Lithographically Patterned Nb Meander





^{55}Fe Source for Energetic Particles

- Data from 1D transmission line
- Excellent pulse shape
- 300 mv amplitude
- No need for external amplification
- >1 ns rise



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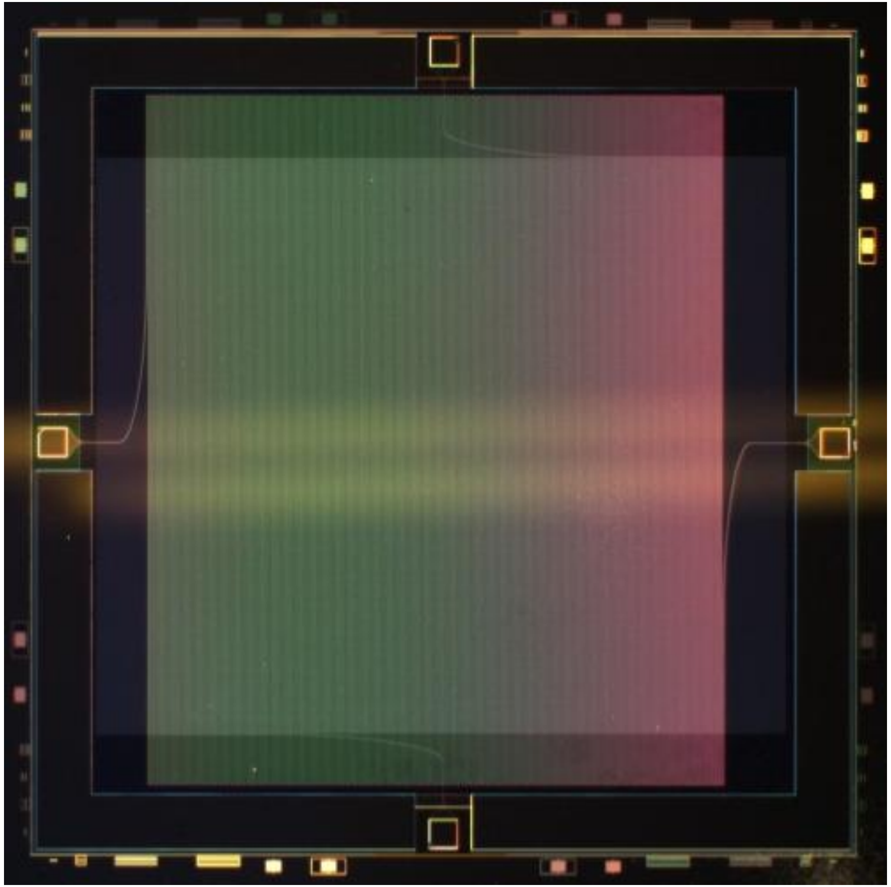
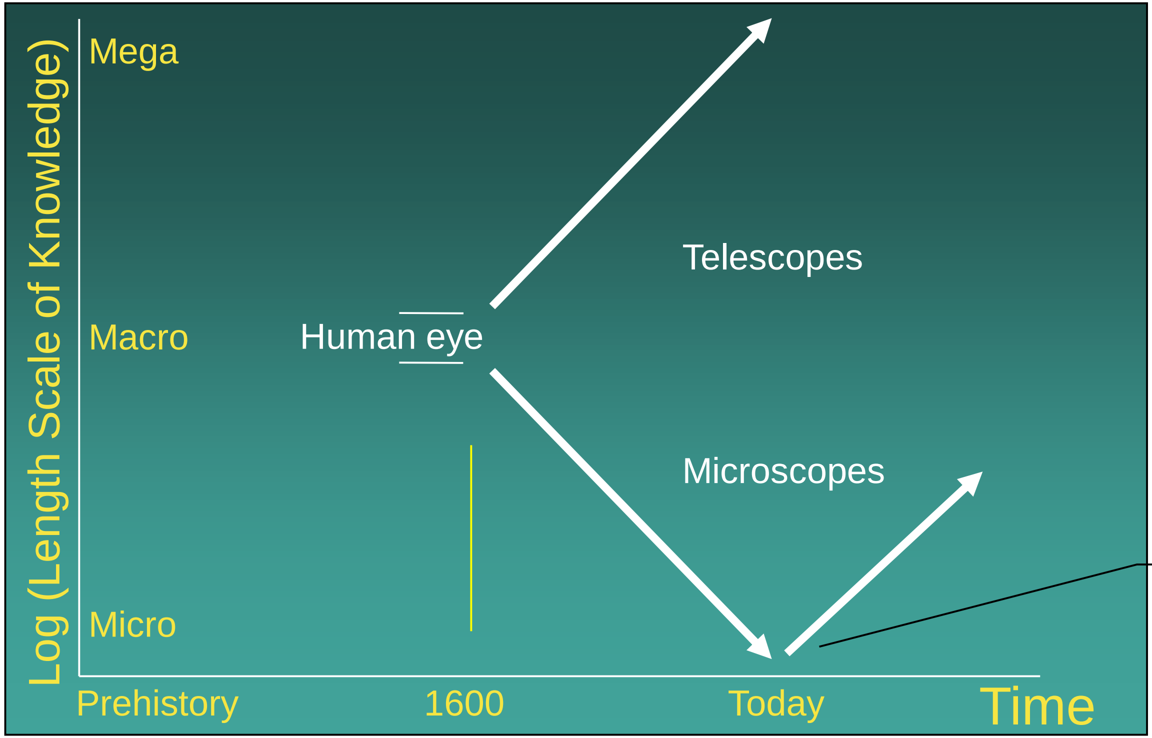
2D Detector

- Patterned detectors fabricated
- Test fixture for ions is being completed
- Testing will begin in next few months



In the History of Microscopy, We are at an Inflection Point



Inflection point

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The History of Microscopy

- The history of microscopy is the pursuit of learning "more and more about less and less"
- We have reached the atomic scale
  - This marks an inflection point
- The future of microscopy will be the pursuit of learning "more and more about more and more"

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