Methods for Achieving Atomic Scale Analytical Tomography

Brian P. Gorman, Megan Holtz

Department of Metallurgical and Materials Engineering, Colorado School of Mines, Golden, CO

Tom F. Kelly

Steam Instruments, Madison, WI

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Outline

How ASAT Might Be Achieved

4-D STEM

Example: Ge Hole Spin Qubits

CONCLUSIONS AND ACKNOWLEDGEMENTS

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How ASAT Might Be Achieved

Atomic Scale Analytical Tomography

- Individually analyze 100% of atoms within a volume

- Picometer spatial resolution in 3-D

- Single atom isotopic chemical information

- Electronic structure

- Correlate experiment with first principles theory

Kelly, Gorman, Ringer, Atomic Scale Analytical Tomography, Cambridge U.P. (2022)

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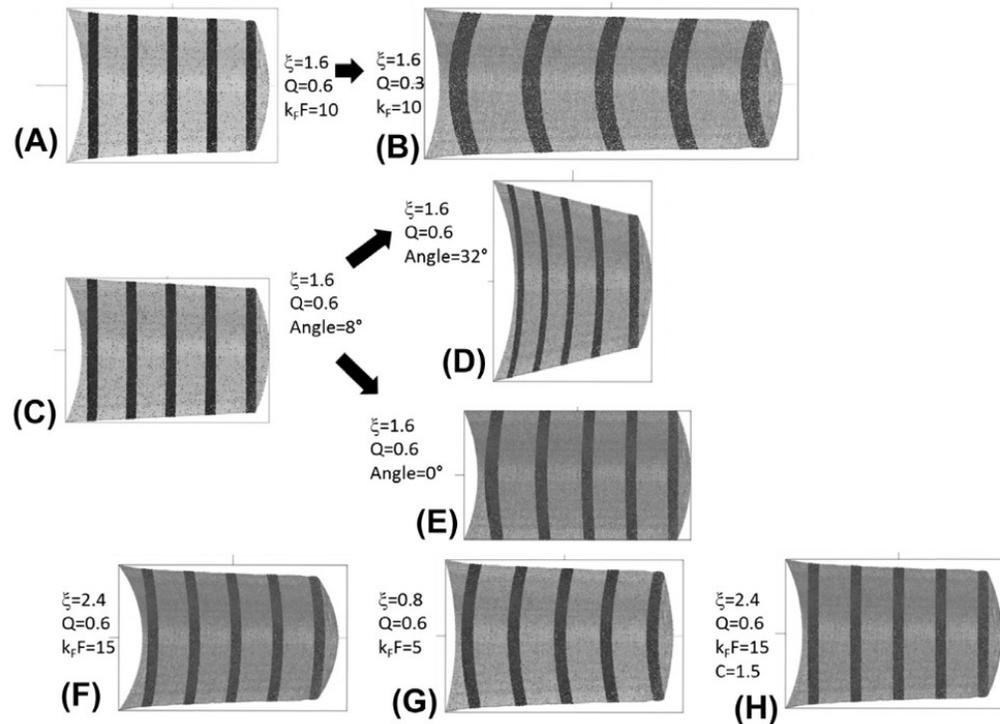
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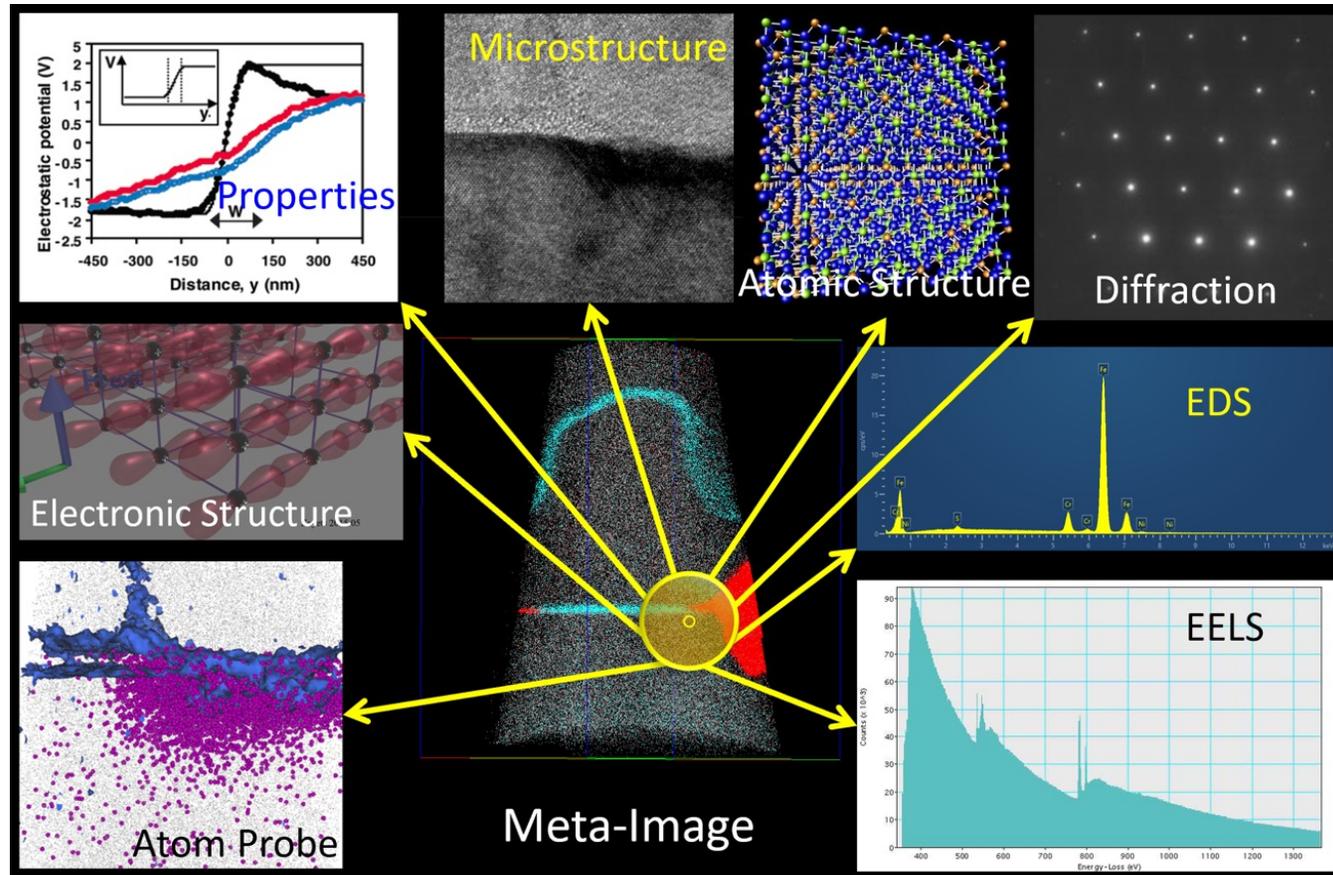
How Can We Achieve ASAT?
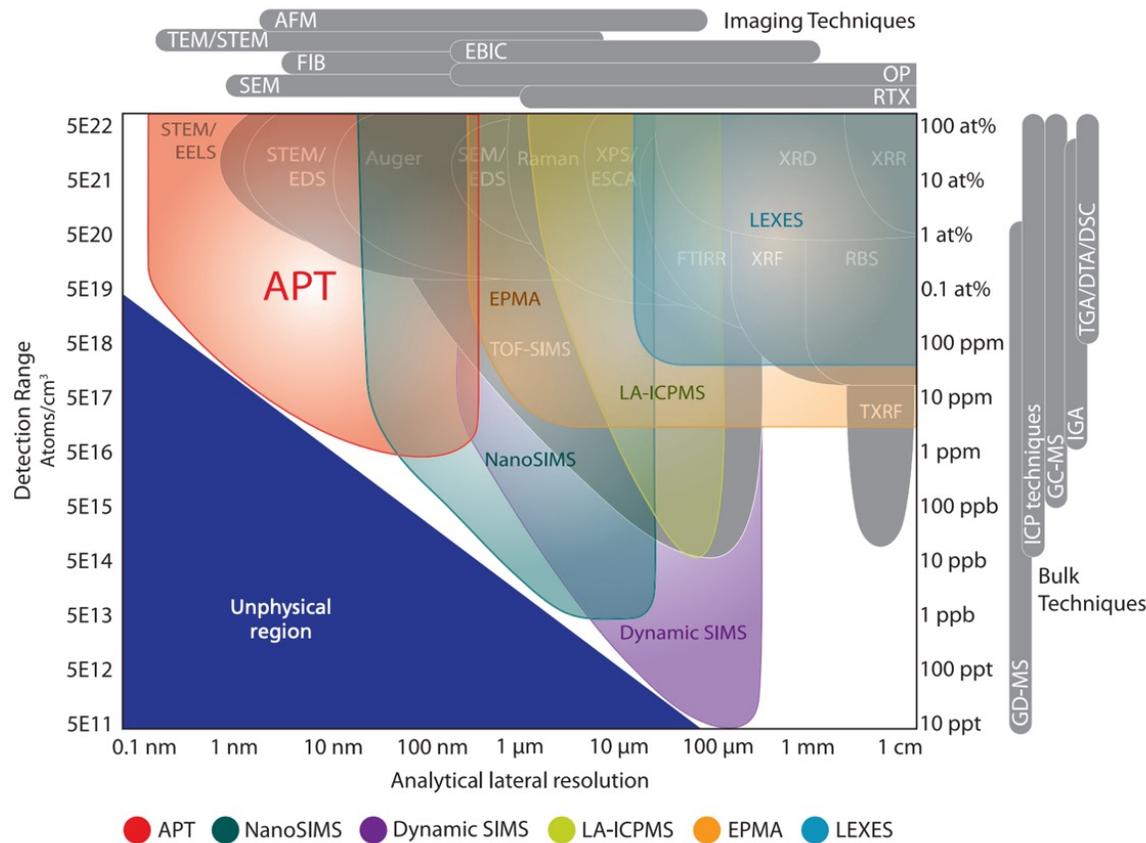
- (S)TEM
  - Approaching pm resolution in 2-D
  - Diffraction can achieve pm resolution on atomic spacings
  - Elemental information limited by detectability and spatial resolution

- APT
  - Single atom isotope information
  - 3-D spatial information
  - "Rubber Ruler" leads to inaccurate spatial information

- Combined APT + STEM?

APT "Rubber Ruler"

Geometric APT data reconstruction

Choice of data reconstruction variables can wildly alter the 3-D volume and representation of the structure
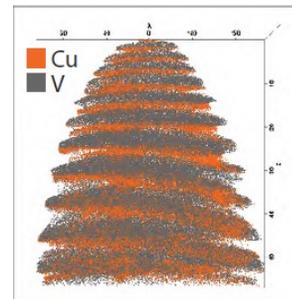
What is the correct answer? Extra information needed

F. Vurpillot, "Three Dimensional Reconstruction in APT" in Lefebvre-Ulrikson W, Vurpillot F, Sauvage X (eds). *Atom probe tomography: put theory into practice*. London: Academic Press, 2016.
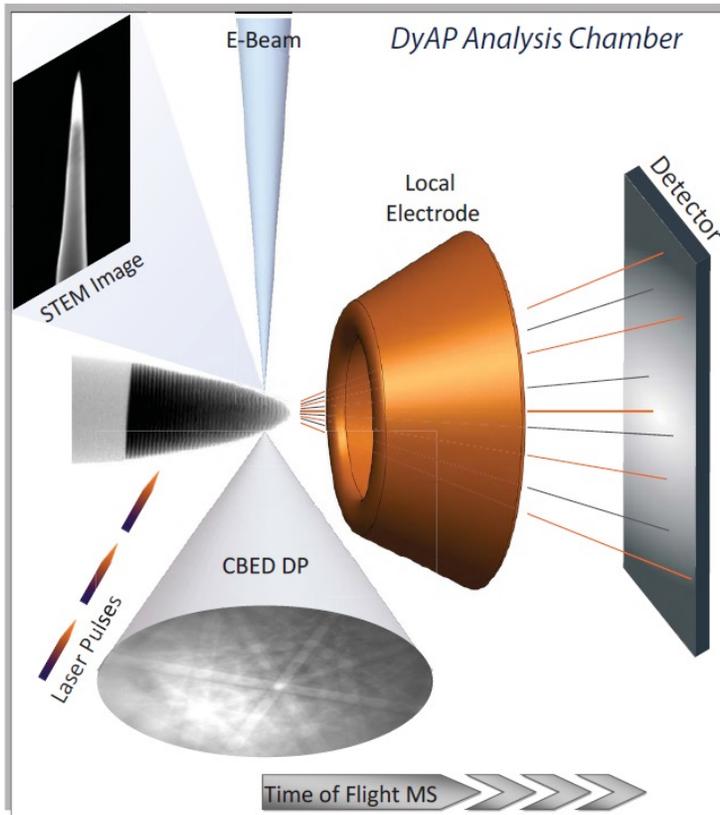
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STEM + APT Experiment

- Cylindrical ~100 nm diameter specimens prepared using FIB

- STEM imaging used to determine specimen shape

- 4-D STEM used to determine atom positions

- EELS used to determine electronic structure

- APT used to determine single atom isotopic information

TEM + APT Measurement

before APT

APT reconstruction

After APT

50 nm

Nd

O
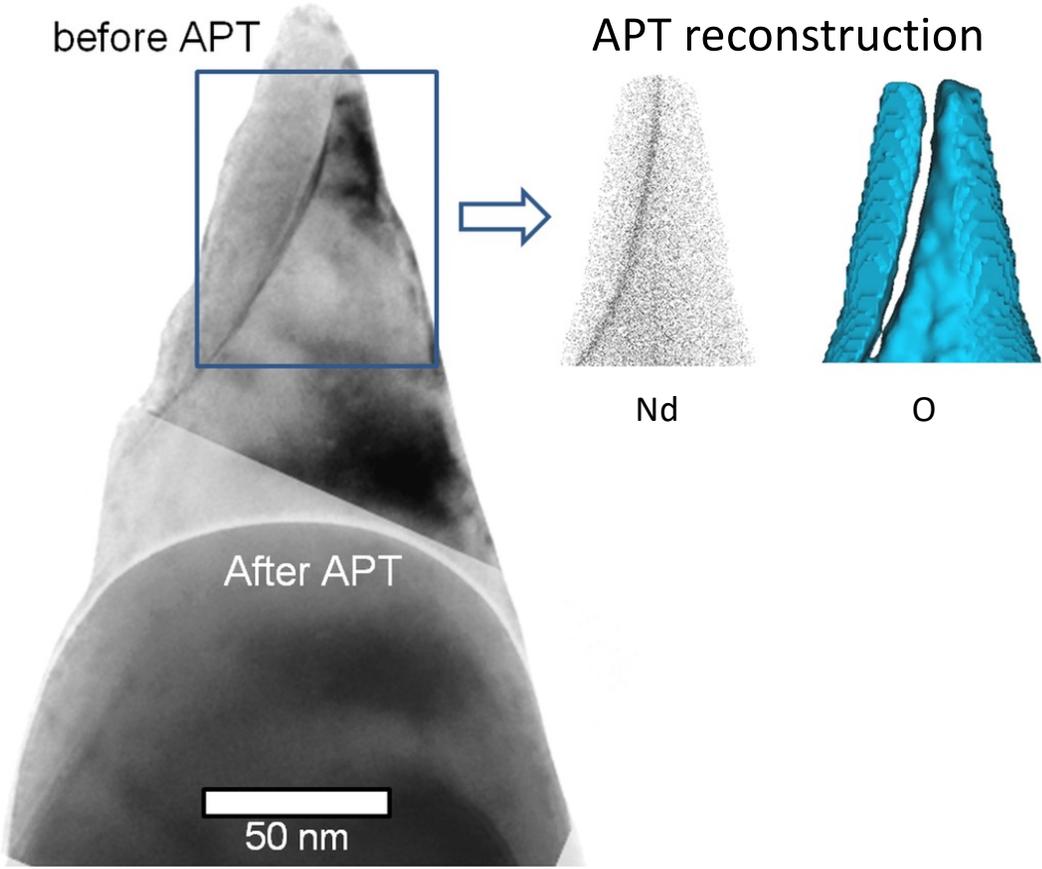
STEM + APT Programs

1. Forschungszentrum Julich + Cameca + ThermoFisher Scientific
   - Integrate APT into objective lens of ThermoFisher STEM
   - APT-centric using STEM and LTEM
2. Atomnaut
3. Univ. Rouen
   - Integrate APT into objective lens of JEOL 2000FX
4. Mines + Steam Instruments
   - Integrate UHV 4-D STEM + 100% efficient SCDL detector
   - STEM-centric ASAT

How Can We Achieve ASAT using TEM + APT?

- **APT-Centric Atom Positioning**

  - Use STEM information to determine the physics of the APT experiment
    - Electric field at the specimen apex
    - Ion trajectories
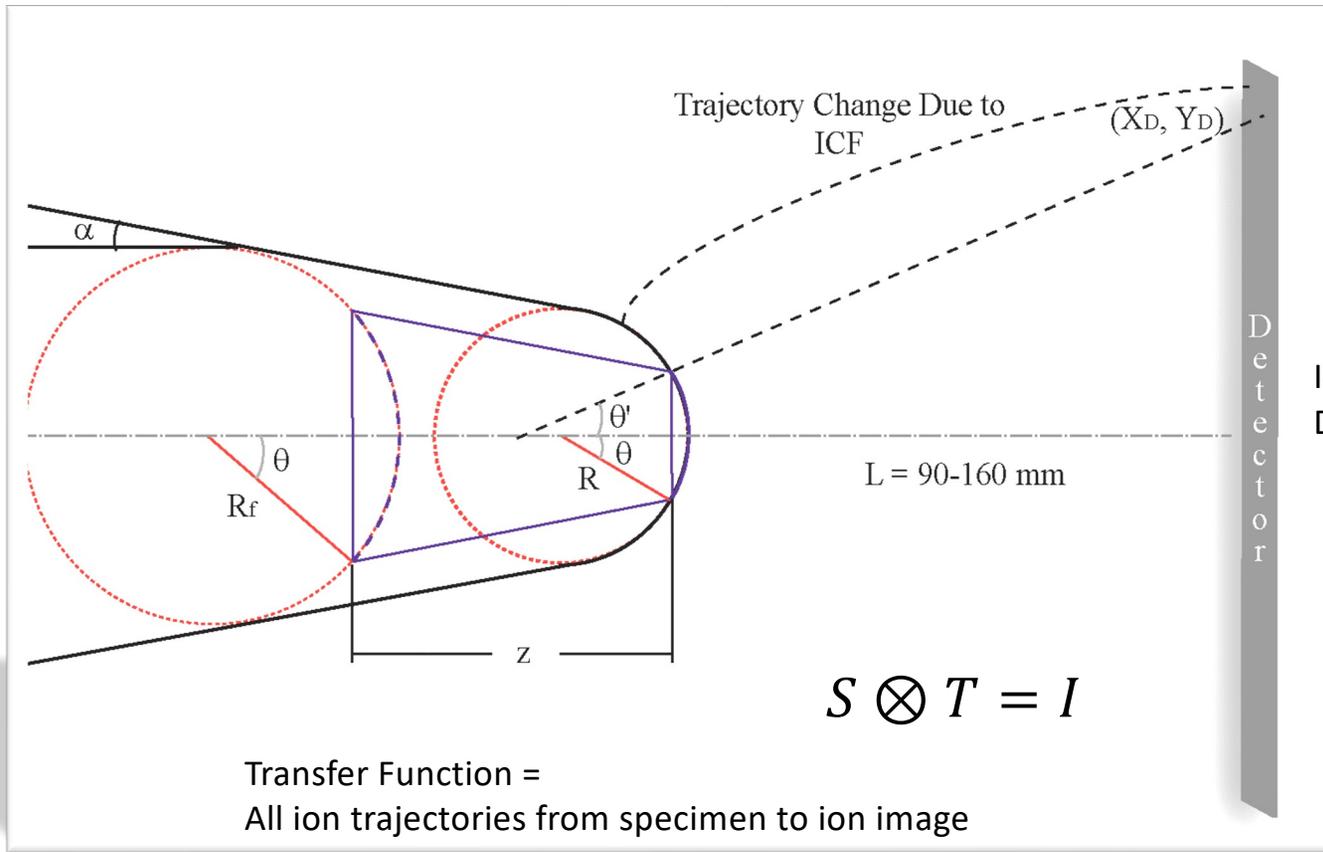 - Build physics models to correct ion trajectories

- **STEM-Centric Atom Positioning**

  - Use STEM information to determine atom positions before APT
    - Diffraction rather than HRSTEM
  - Use APT data to determine atomic isotopes

Kelly, Gorman, Ringer, <u>Atomic Scale Analytical Tomography,</u> Cambridge U.P. (2022)

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STEM-Centric APT Atom Positioning

Trajectory Change Due to ICF

(X_D, Y_D)

Specimen Function = Volume and atom positions before APT

Ion Image Function = Detector space image

θ'

θ

θ

R

R_f

$L = 90\text{-}160 \text{ mm}$

Detector

z

$$S \otimes T = I$$

Transfer Function = All ion trajectories from specimen to ion image

STEM-Centric Atom Positioning Example
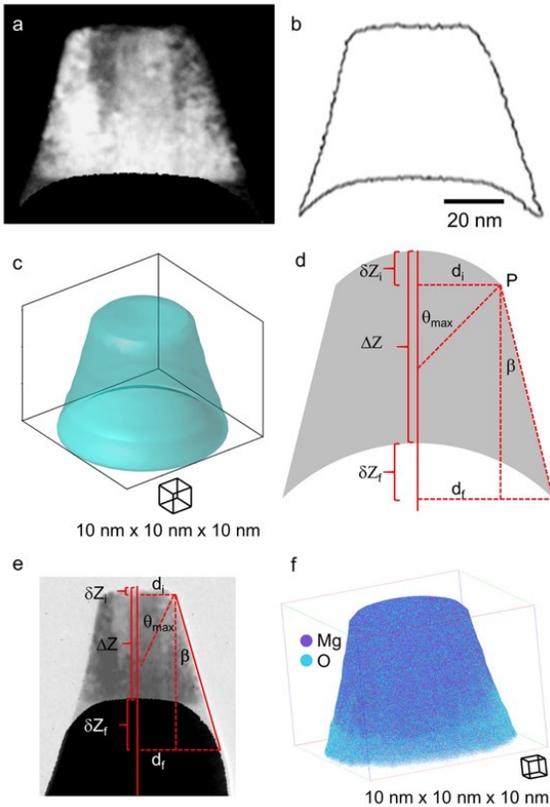
- ZnO // (0001) sapphire
  - Data collected by Rita Kirchhofer
- TEM imaging and diffraction used to define volume and atom positions
- APT ion data used to place the atoms back onto TEM defined final surface

- Additional structural information needed for ASAT
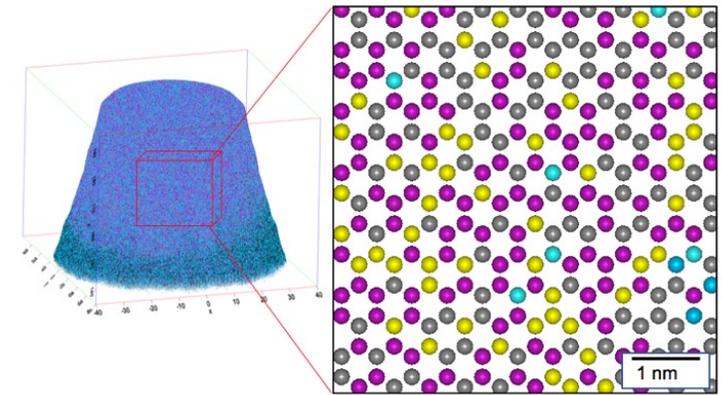
A. Ceguerra, A. Breen, J. Cairney, S. Ringer, B. P. Gorman, *M&M,* **27** (2021) 140-148

4-D STEM
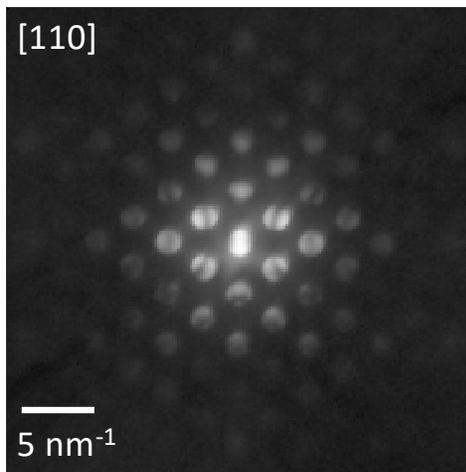
4-D STEM: Overview
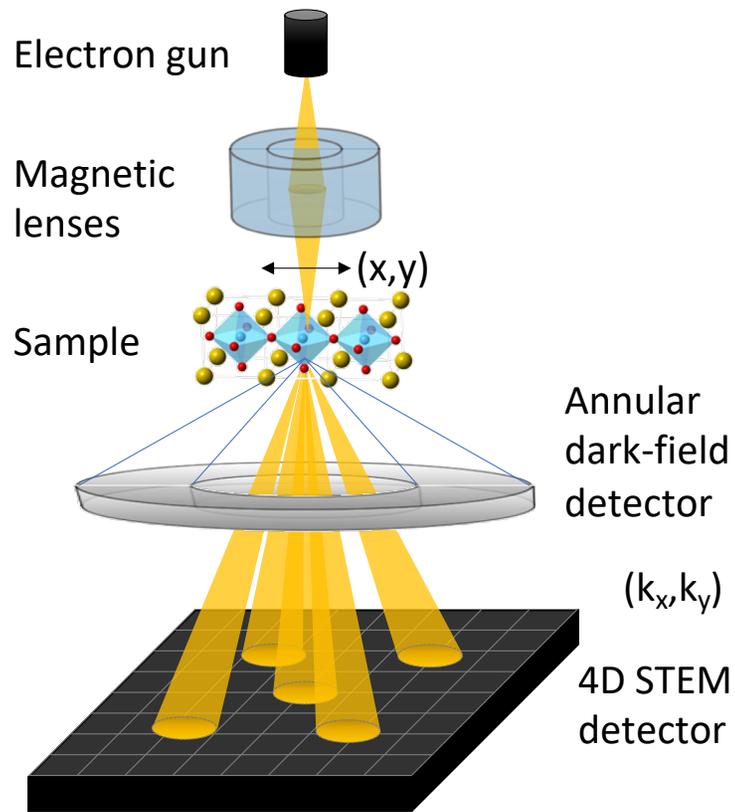
4D-STEM: Scanning electron diffraction

Collect the full electron diffraction pattern at every scan position in the sample.

- Crystallographic measurements with <nanometer real space resolution
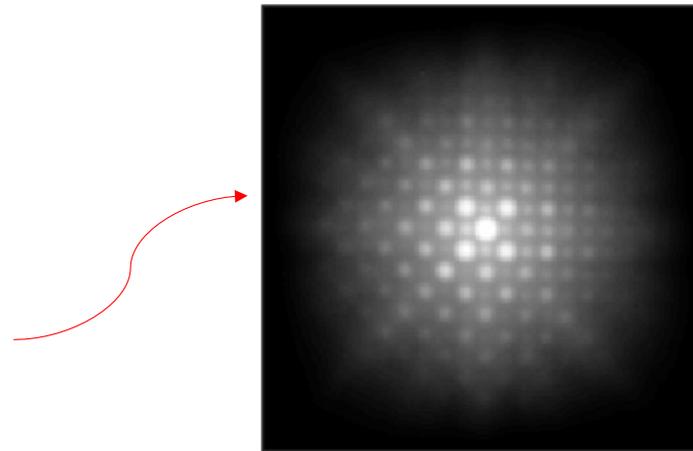
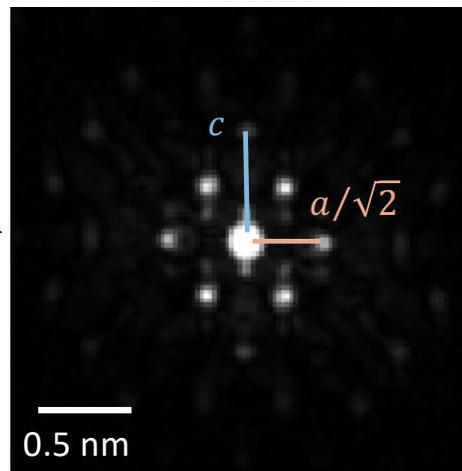# EWPC: Measuring Lattice Parameters with 4D-STEM

Log(NBED)

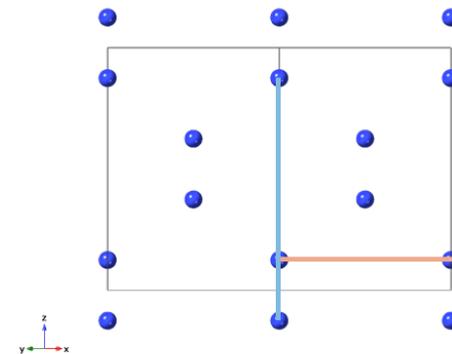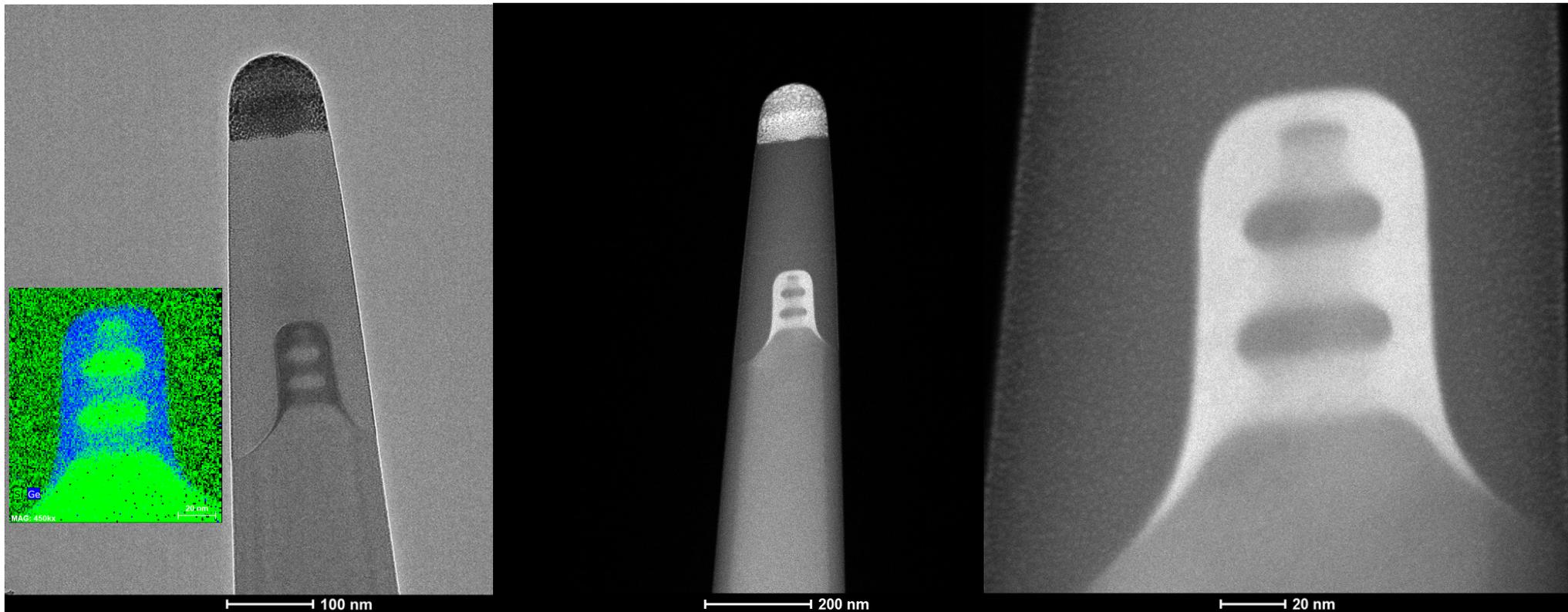The logarithm suppresses tilt and thickness effects

EWPC

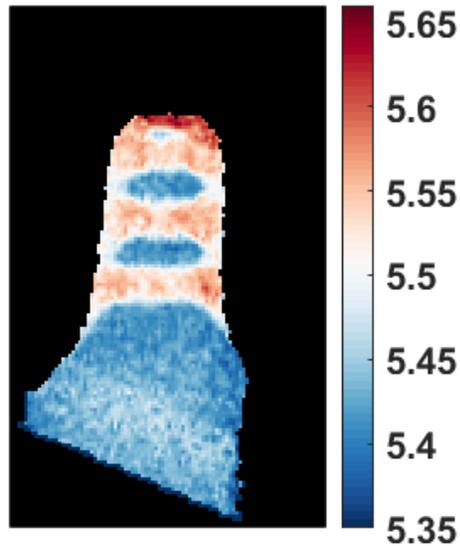Similar to a Patterson Function or Pair Distribution Function

Ge [110] zone

Directly measures interatomic distances!

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Example: BF and STEM-HAADF of SiGe / Si GAA Channel

Lattice Parameter Mapping with Scanning Nanobeam Electron Diffraction

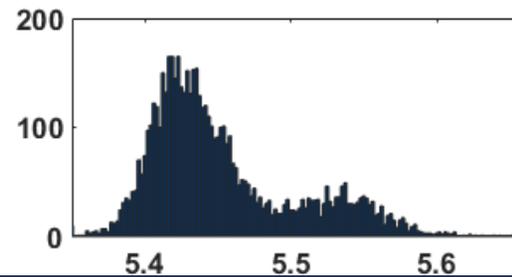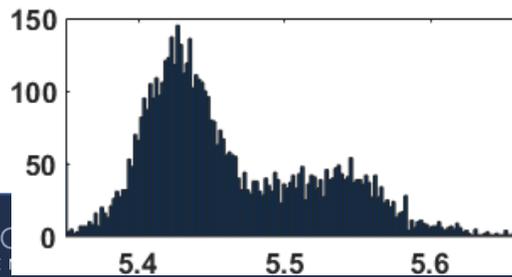

Vertical (out-of-plane)
Lattice paramter (Ang)
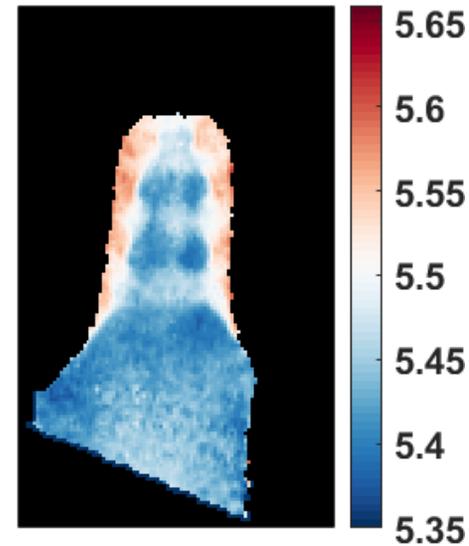
Horizontal (in-plane)
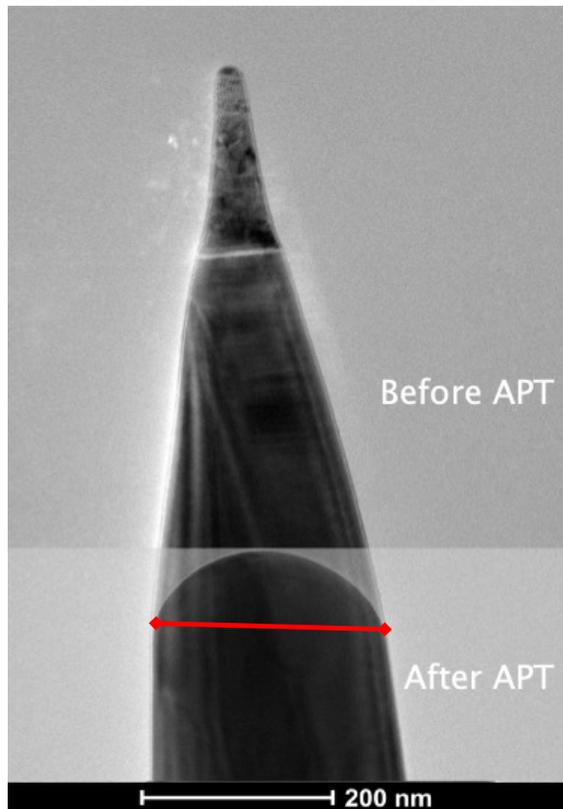Lattice paramter (Ang)

What ASAT Information Can We Get from 4-D STEM?

Before APT

After APT

|—— 200 nm

- Real space analyzed volume
  - Specimen thickness from HAADF intensity
- Crystallographic phase (space group)
  - Atomic ordering and defects
- Crystallographic orientation relative to detector axis
- Lattice parameters (0.05 Å precision)

- Combine real space and reciprocal space to give atom positions before APT reconstruction
  - Specimen function
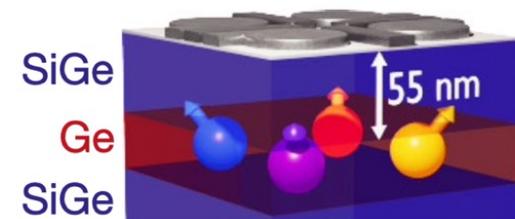
Example: Ge Hole Spin Qubits

Planar Ge QW Devices

- Ge gate defined devices outperform most other semiconductors due to valence band properties
  - Hole mobilities >10^6 cm^2/V*s
  - Strong spin-orbit coupling for holes
  - Low hole effective mass <0.05 when strained
  - T_1 >1 ms
- 4 qubit dot array recently demonstrated
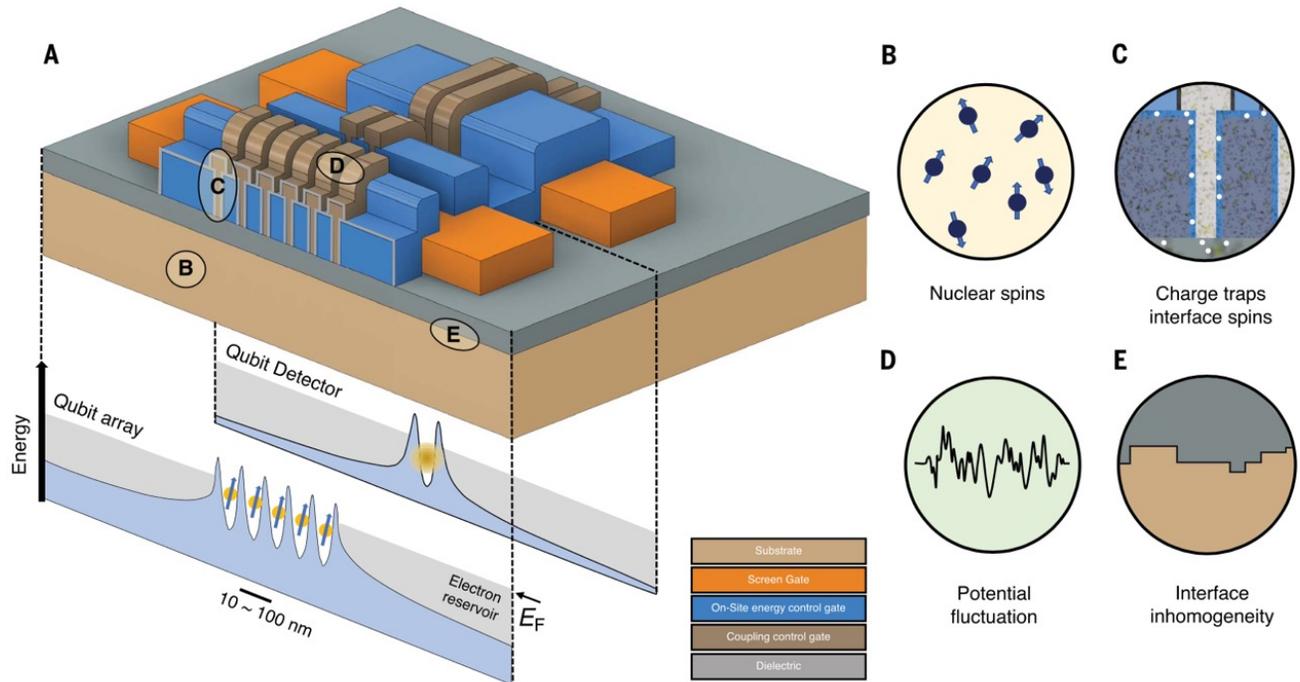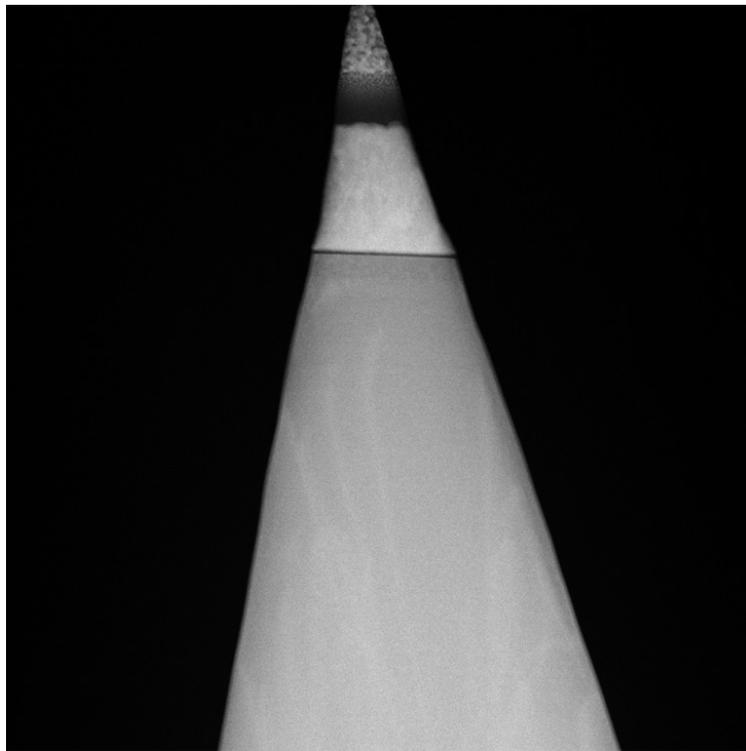 - Ge QW strained by SiGe epitaxy
  - Gate times ~2 μs
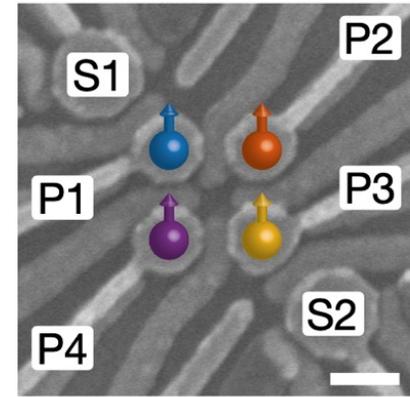
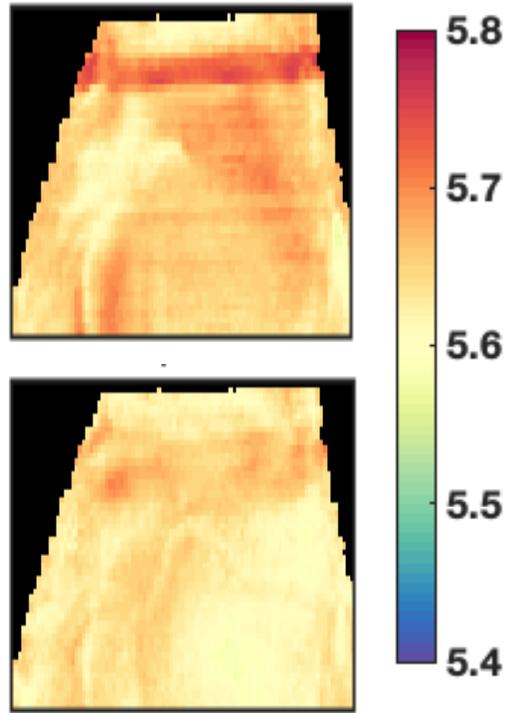# Decoherence Mechanisms in Planar Devices

1. Potential and Band Fluctuations
   - Variations in Strain
2. Interface Inhomogeneity
3. Nuclear Spins
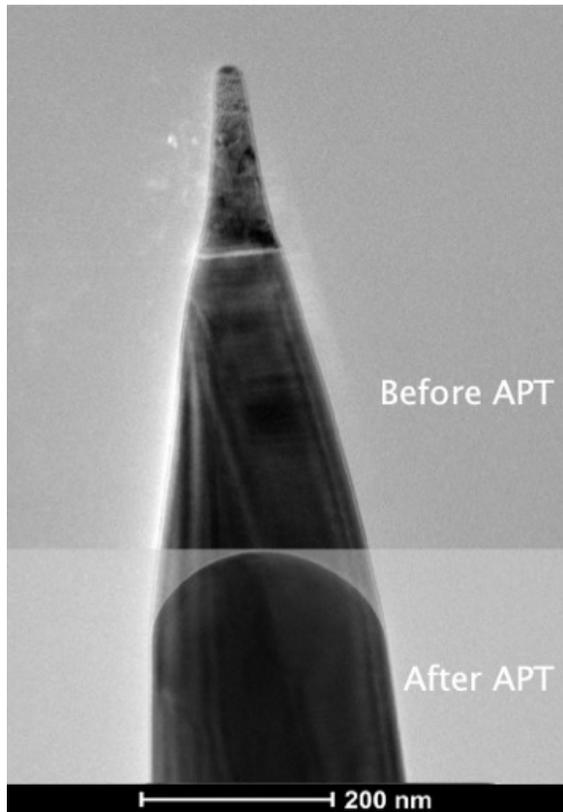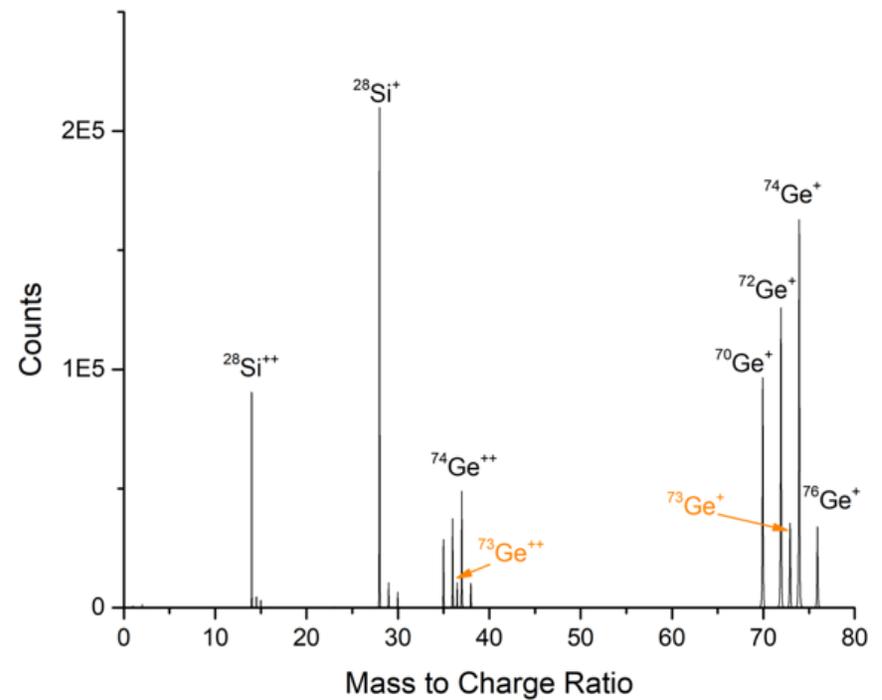4. Charge Traps and Interface Spins

4-D STEM

200 nm

100 nm

- 4-D STEM-EWPC applied again to a cylindrical specimen

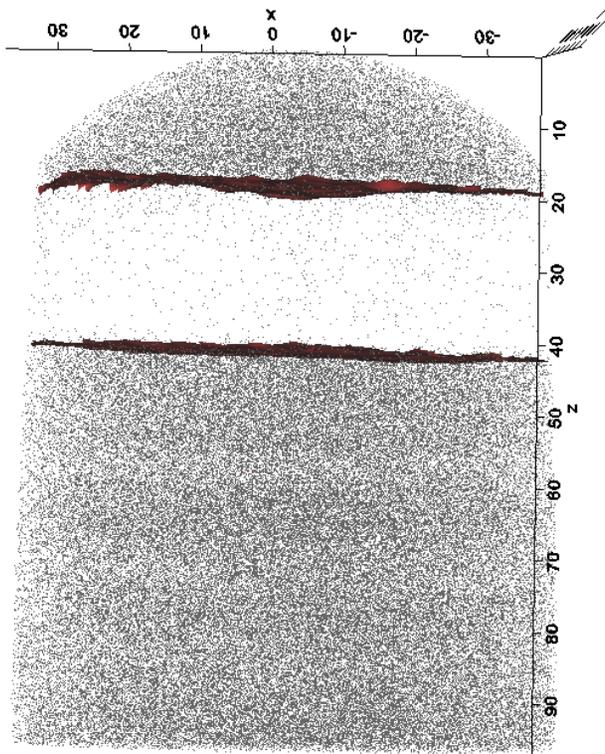- Information used to identify atom positions / specimen function before APT
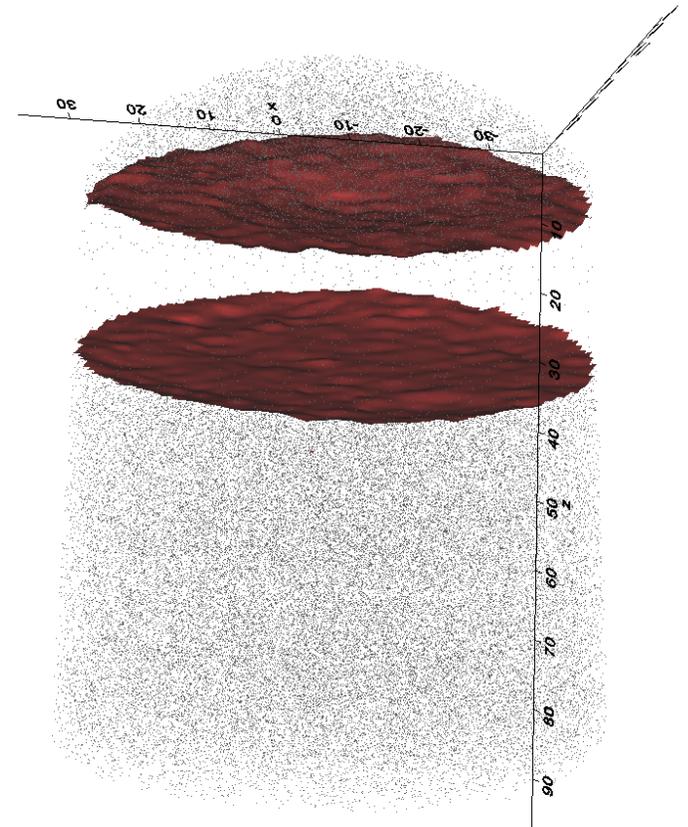
Atom Probe Tomography

- Pre- and Post-APT imaging of the specimen using TEM confines the 3-D data reconstruction

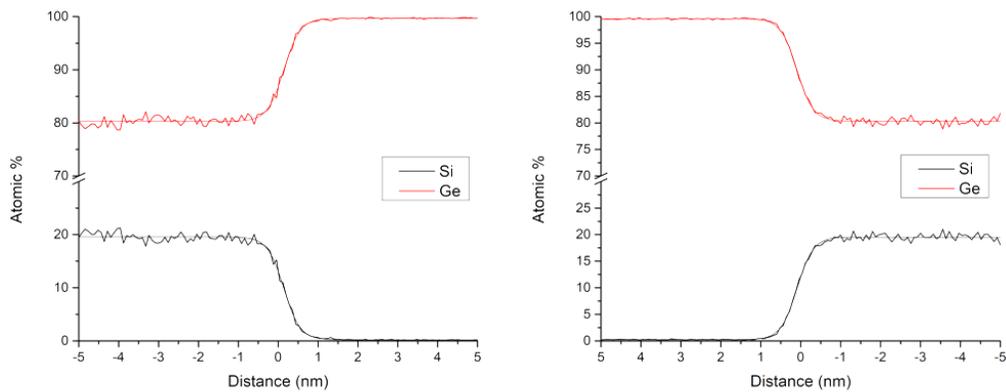- Individual ^{73}Ge isotopes easily identifiable
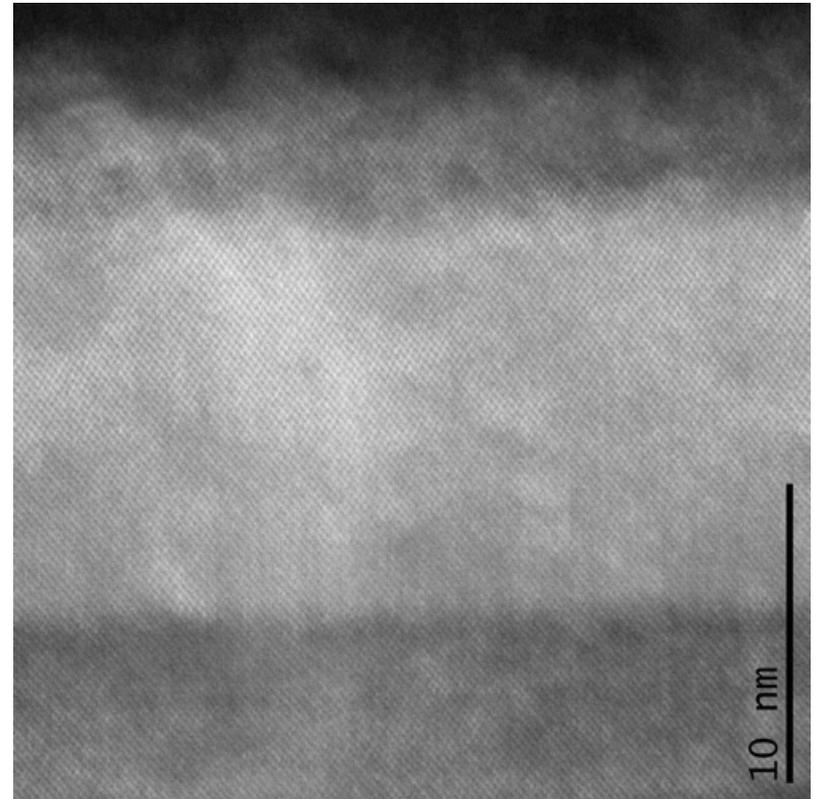
Interface Homogeneity

- Gray = Si
- Red = Ge

- Reconstruction confined by STEM information

- Determine internal structure of interfaces

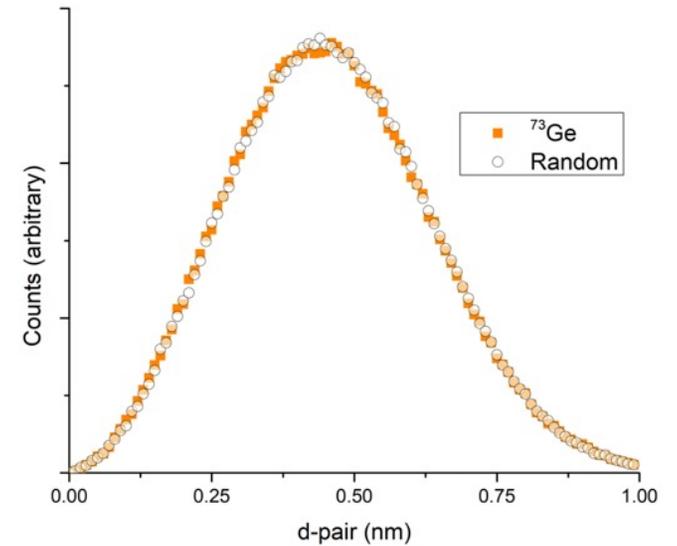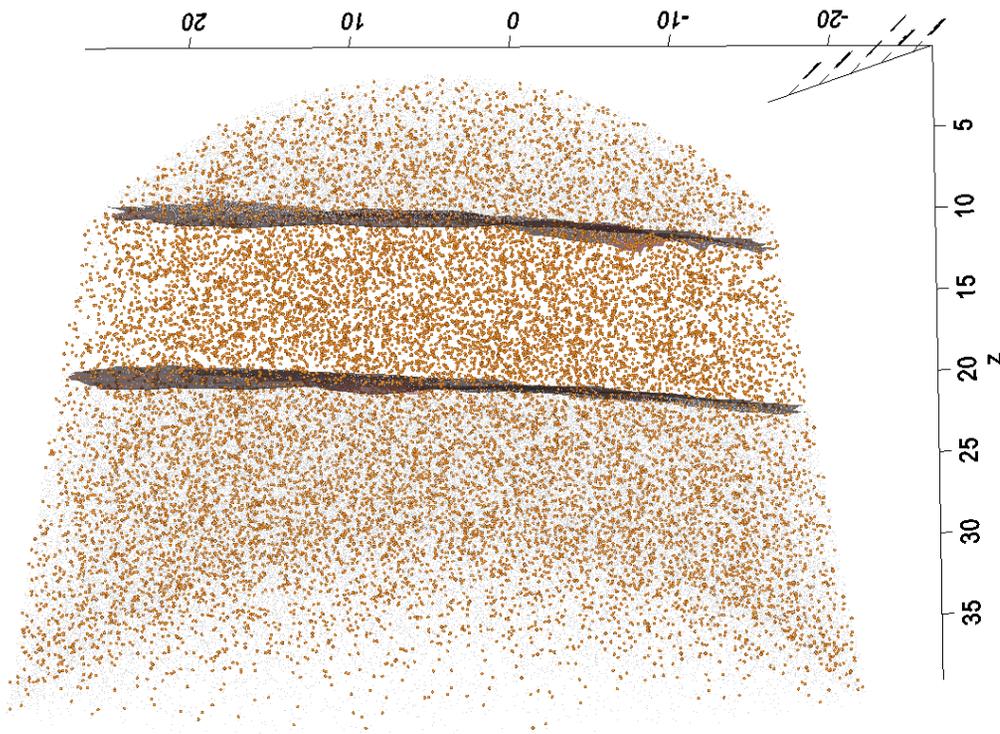# Interface Homogeneity

- Proximity histograms acquired at top and bottom QW interfaces

- Fit profiles to Boltzmann distribution

- Interface chemical abruptness is <1 nm

Nuclear Spins

- APT gives us the ability to locate nuclear spins in 3-D

- Orange spheres = ^{73}Ge (only 20% of atoms shown for clarity)

- Nearest neighbor distribution illustrates the ^{73}Ge are randomly distributed

Conclusions – Semiconductor Qubits

We can now experimentally access theories about decoherence mechanisms in planar Ge 2-D hole gasses

Structure-property relationships in quantum devices can be experimentally addressed

1. Potential and band fluctuations due to strain
    - 4-D STEM
2. Interface Heterogeneity
    - APT + 4-D STEM
3. Nuclear Spins
    - APT
4. Interface spins due to point defects
    - APT

CONCLUSIONS AND ACKNOWLEDGEMENTS

Conclusions

- ASAT may be possible using STEM + APT
  - TEM gives analyzed volume
  - 4-D STEM gives atom positions
  - APT gives single atom isotopic information

- Future
  - SCDL ion detector to capture ~100% of atoms
  - Single 4-D STEM + APT instrument
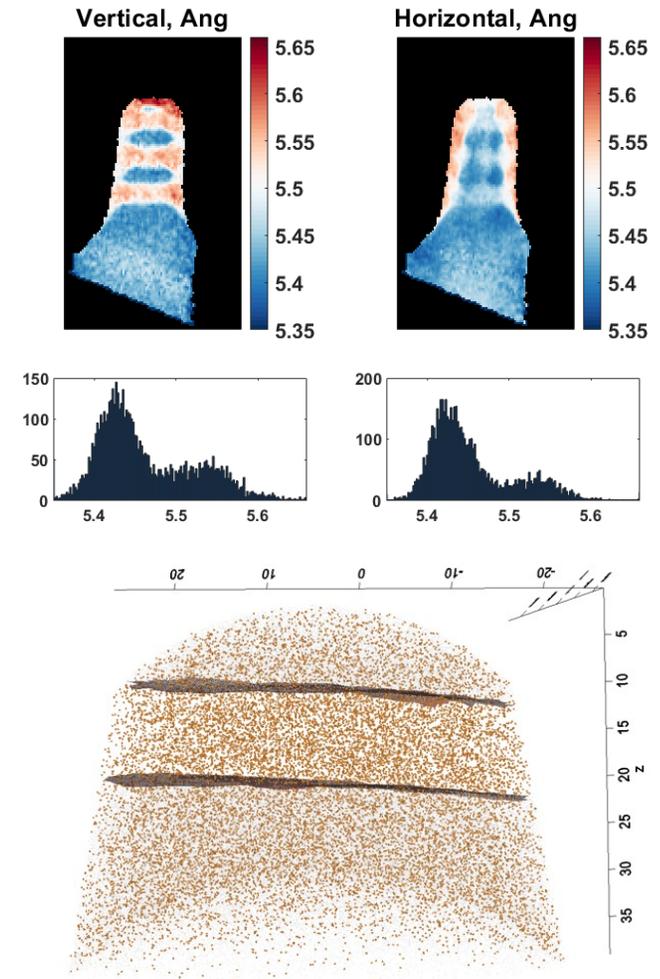
Collaborators

- Edwin Supple, *CSM*
- John Hunt, Igor Veryovkin, *Steam Instruments*
- Norman Salmon, Daan Hein Alsem, *Hummingbird Scientific*
- Andrew Breen, Simon Ringer, Julie Cairney, *Univ. Sydney*
- Brendan Foran, *The Aerospace Corp.*
- Kevin Jones, *Univ. Florida*
- Chris Richardson, Chomani Gaspe, *Univ. Maryland and LPS*

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We Need Your Input

- Materials Engineering Department at Mines is establishing a new Ceramic Engineering undergraduate degree
  - Strong focus in electronic materials
  - Laboratory courses in fab processing and metrology

- We need industrial input to give students the skills they need

- bgorman@mines.edu