



High Resolution 3-D Characterization of Nanomaterials using Tilt Tomography in the Scanning Transmission Electron Microscope

Ilke Arslan

Sandia National Laboratories, CA



Outline and Motivation: 3D Analysis

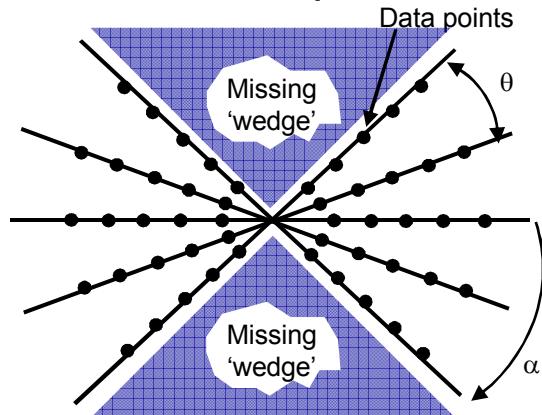
- Why STEM tomography?
 - Instrumentation
 - Reconstruction methods
- High Resolution Single Axis : Sn QDs in Si
- Quantitative Single Axis : Fischer Tropsch catalysts
- The Need for Dual Axis: CdTe tetrapods
- On-axis geometry for comparison to APT : Ag in Al
- Conclusions



Anisotropic Spatial Resolution

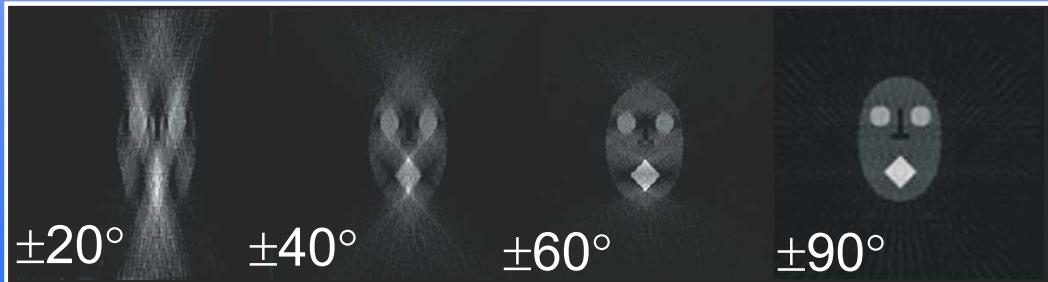


Representation of sampling in Fourier space



θ is the angular sampling
 α is the maximum tilt angle

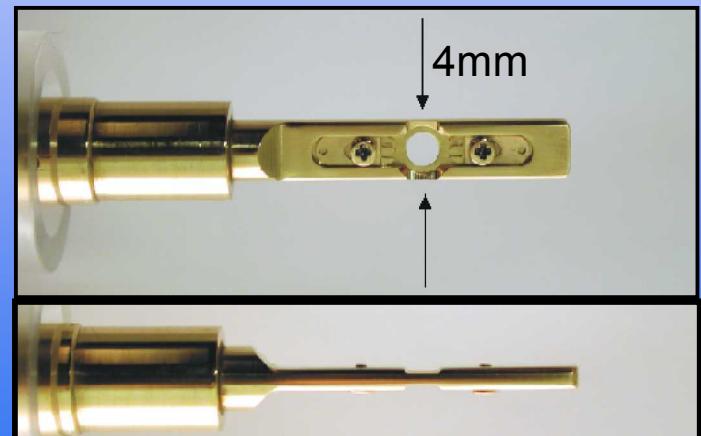
Elongation if limited tilt angle - *missing wedge*



FEI Tecnai F20

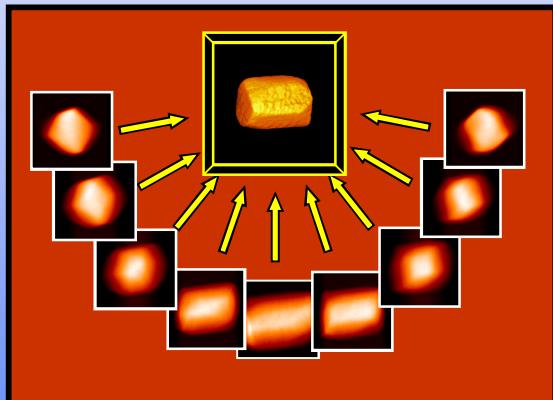
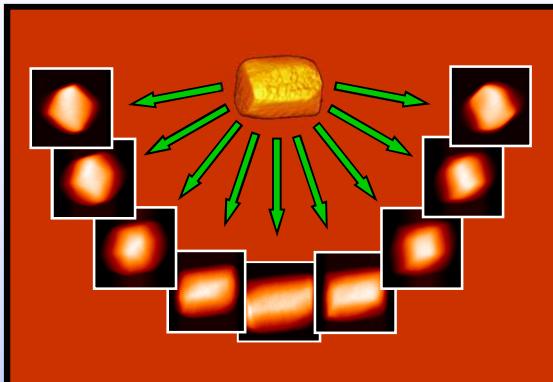
Super Twin Objective Lens
Tilt limited to +/- 40° with conventional holders

Fischione Ultra High Tilt Holder
Full 360° tilt – limited to $\pm 80^\circ$ only by microscope goniometer

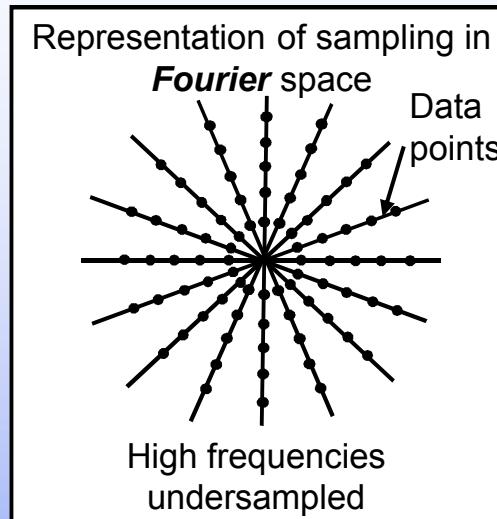




Tomographic reconstruction



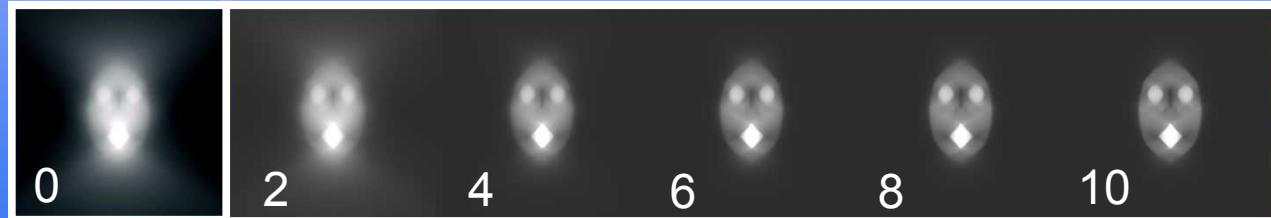
The Principle of
(a) Backprojection



(b) Weighted backprojection
– to account for undersampling of high frequencies

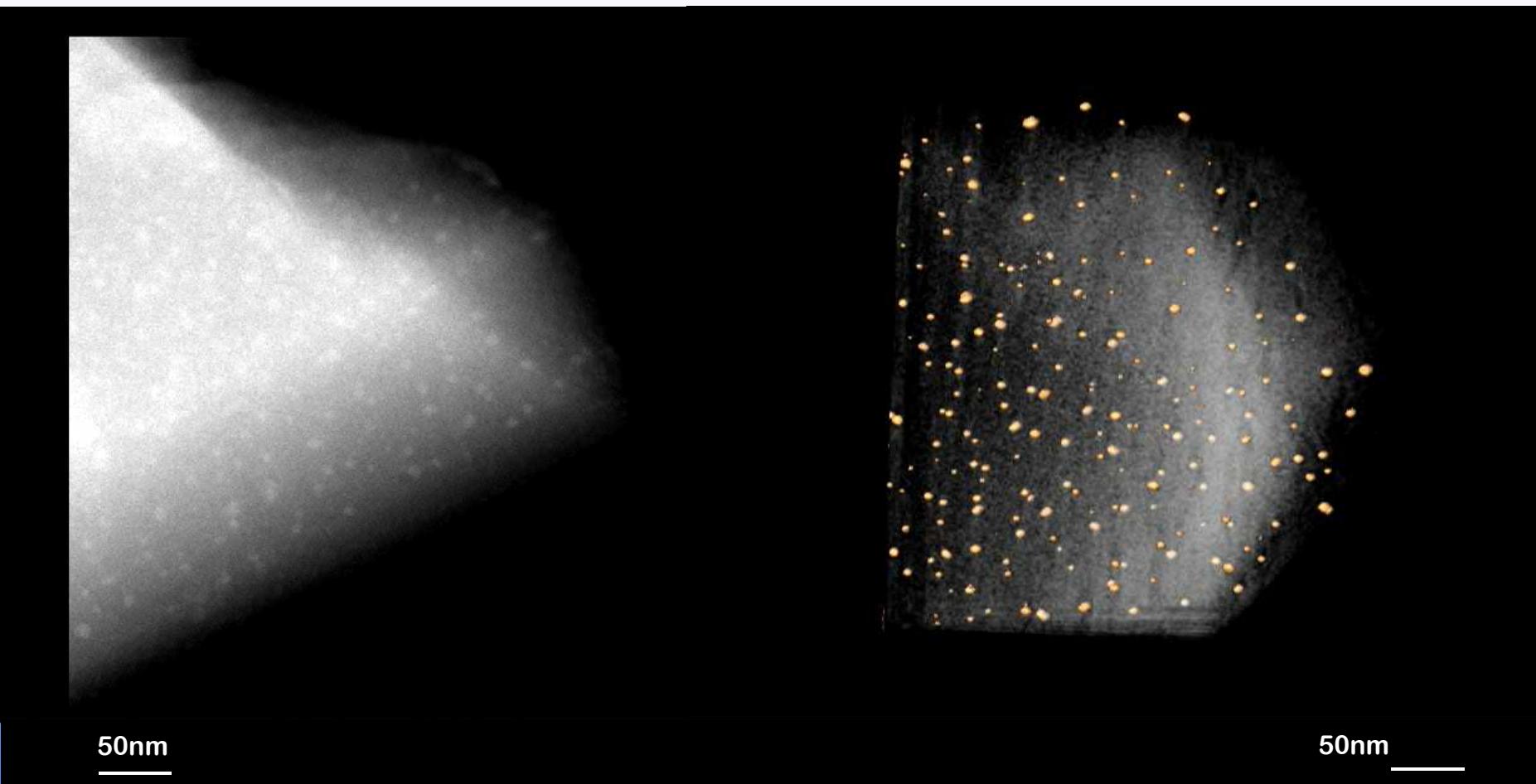


(c) Iterative backprojection – iteratively constrain the reconstruction to give re-projected images that are identical to the original projections.





3-D Analysis-Tomography



Tilt Range: -74° to $+74^\circ$ using 1° increments: 149 images



Distribution in Layers





New and Improved Reconstruction





In-Layer Quantum Dot



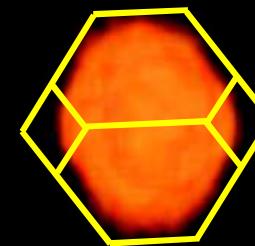
001 projection



5nm

10nm

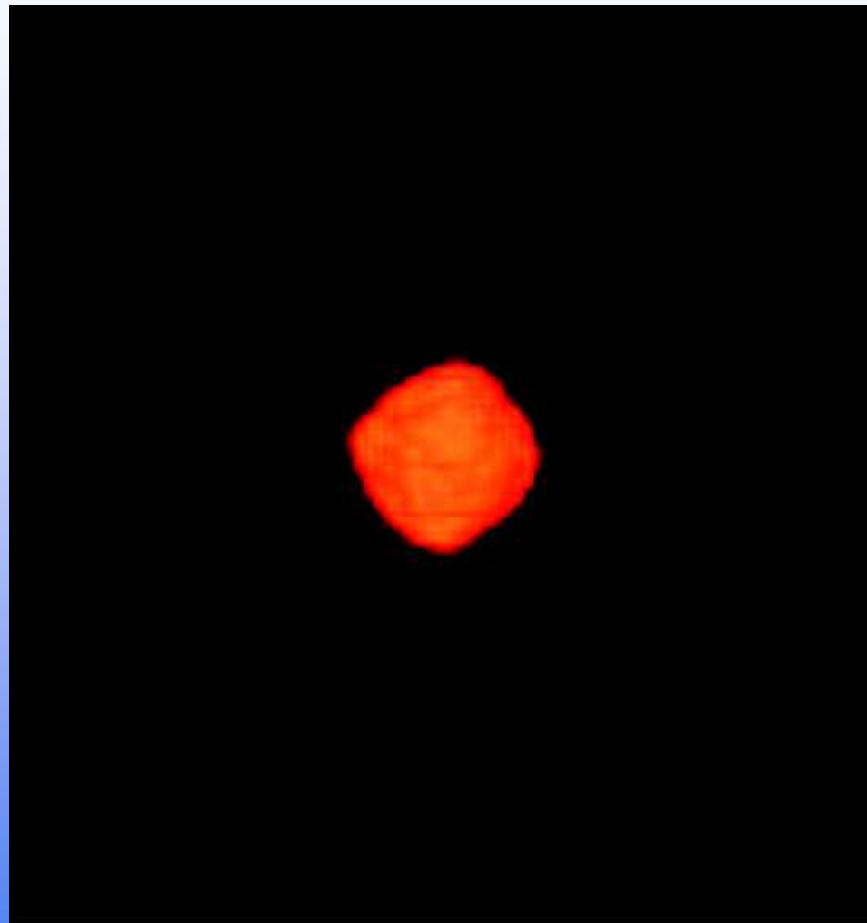
110 projection



5nm



In-Layer Quantum Dot



7nm quantum dot shows 3-D uniformity



Out-of-Layer Quantum Dot



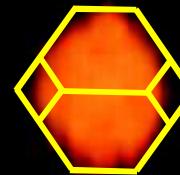
001 projection



5nm

10nm

110 projection

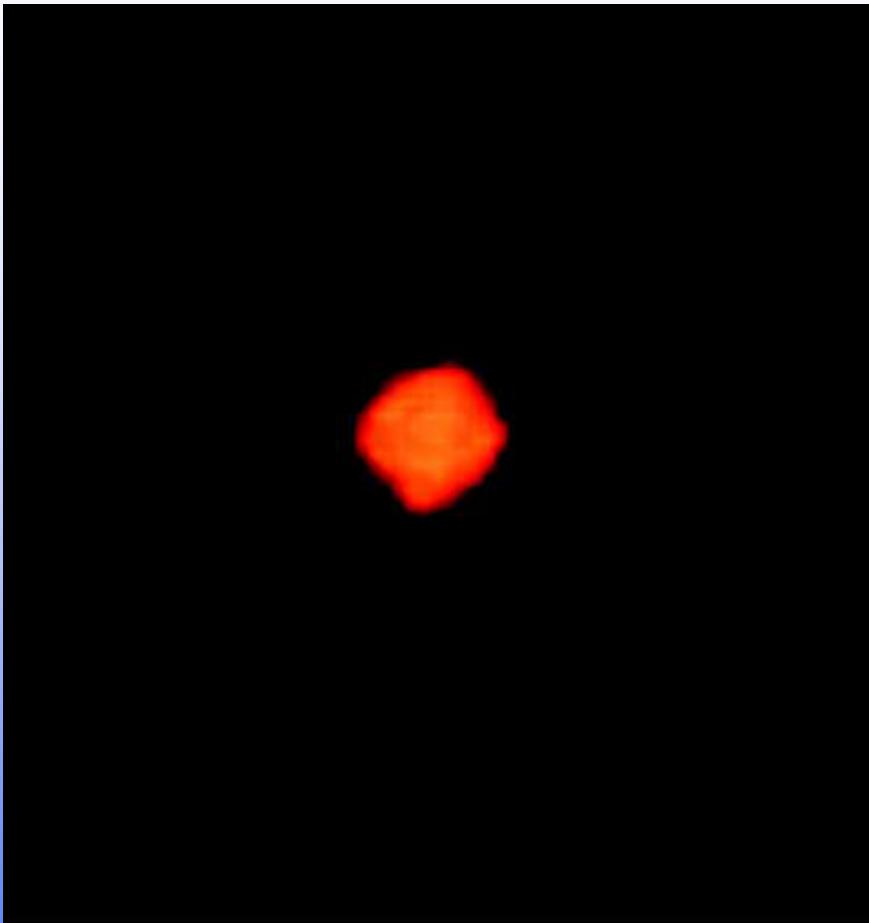


5nm

3.5nm quantum dot reveals filling of void from one side



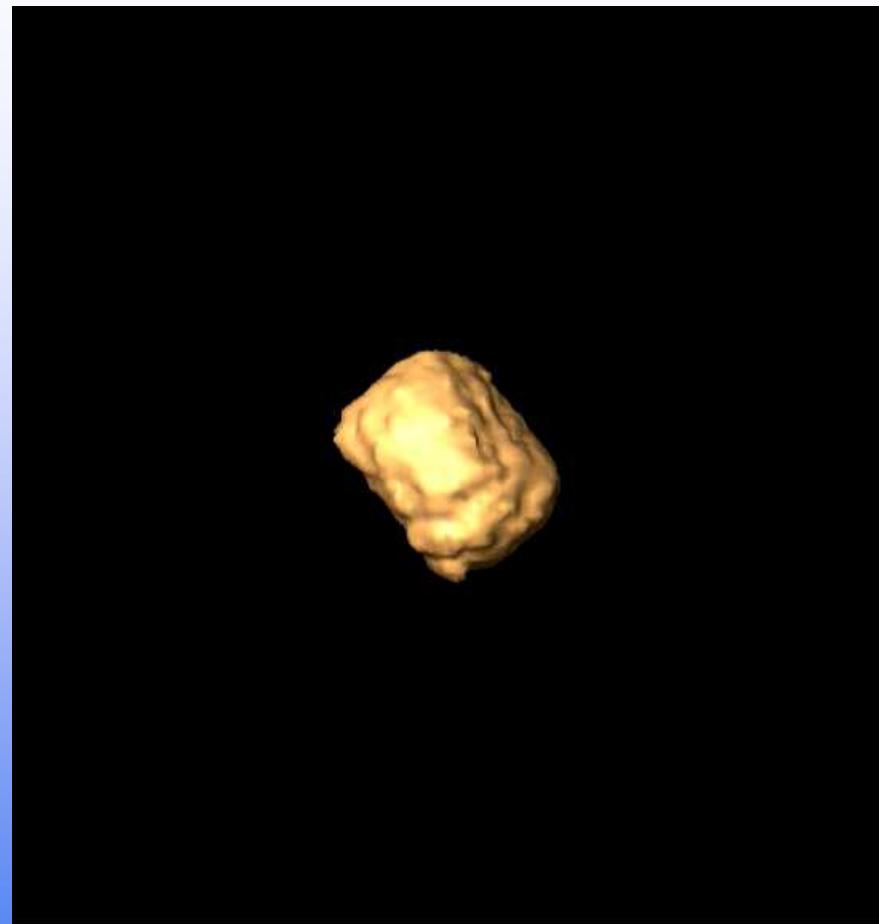
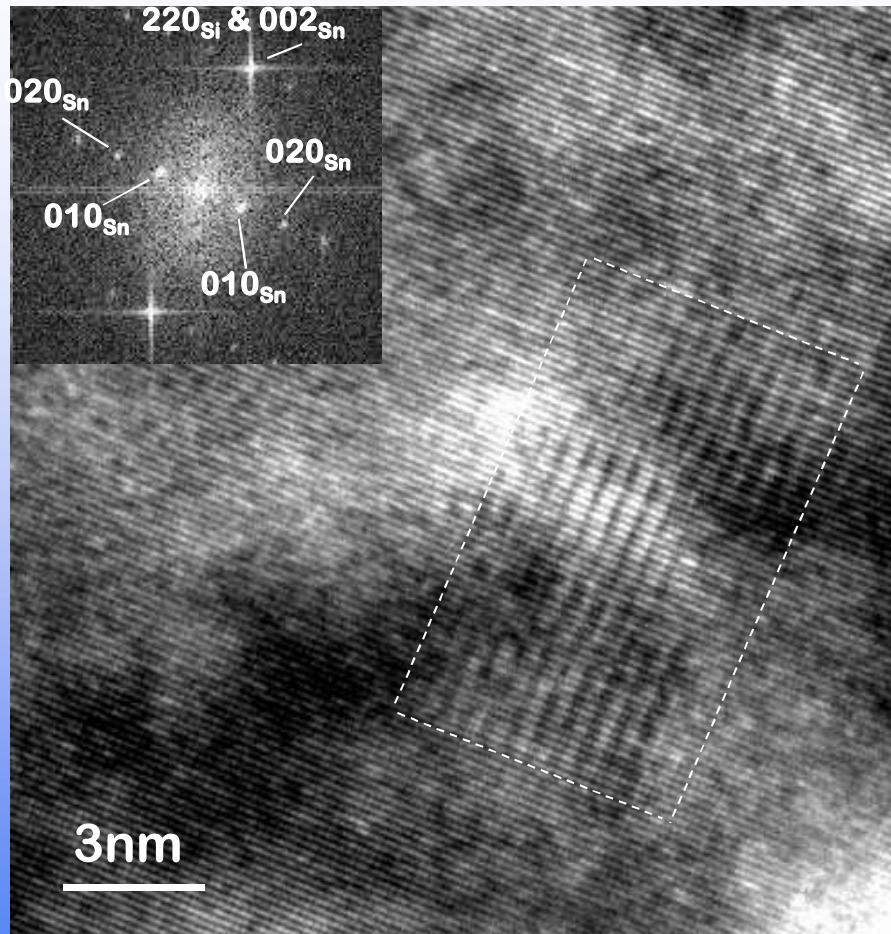
Out-of-Layer Quantum Dot



3.5nm dot shows filling from one side-not uniform in 3D



Larger Dots- A Different Phase?



Elongated dots have transformed into beta-tin

I. Arslan et. al., *Science* 309, 2195-2198 (2005).

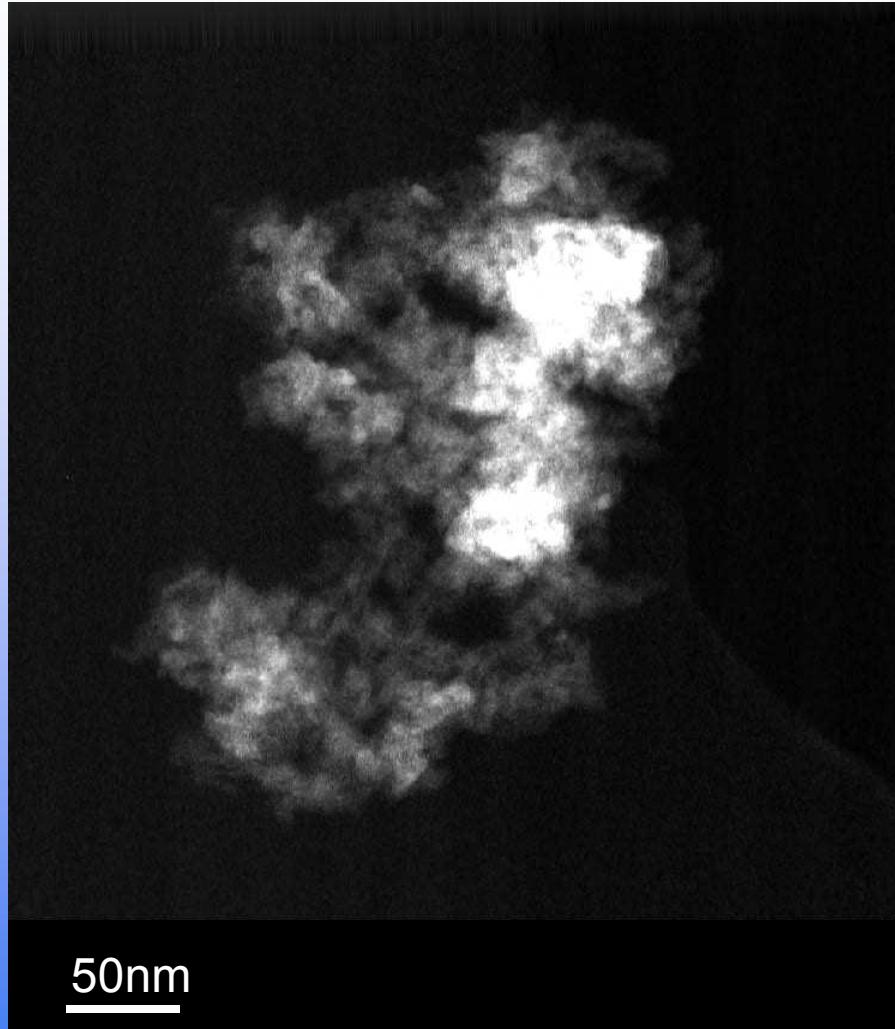


Co₃O₄-alumina Fischer-Tropsch Catalysts

- FT process converts coal and natural gas to useful, clean hydrocarbon fuel.
- Need to diversify energy sources with minimum environmental impact due to increasing crude oil prices.
- Surprisingly little is known about the morphology of the cobalt particles on the nanoscale, the detailed interaction with their support, the pore structure of the support and how these affect the catalytic activity and selectivity.



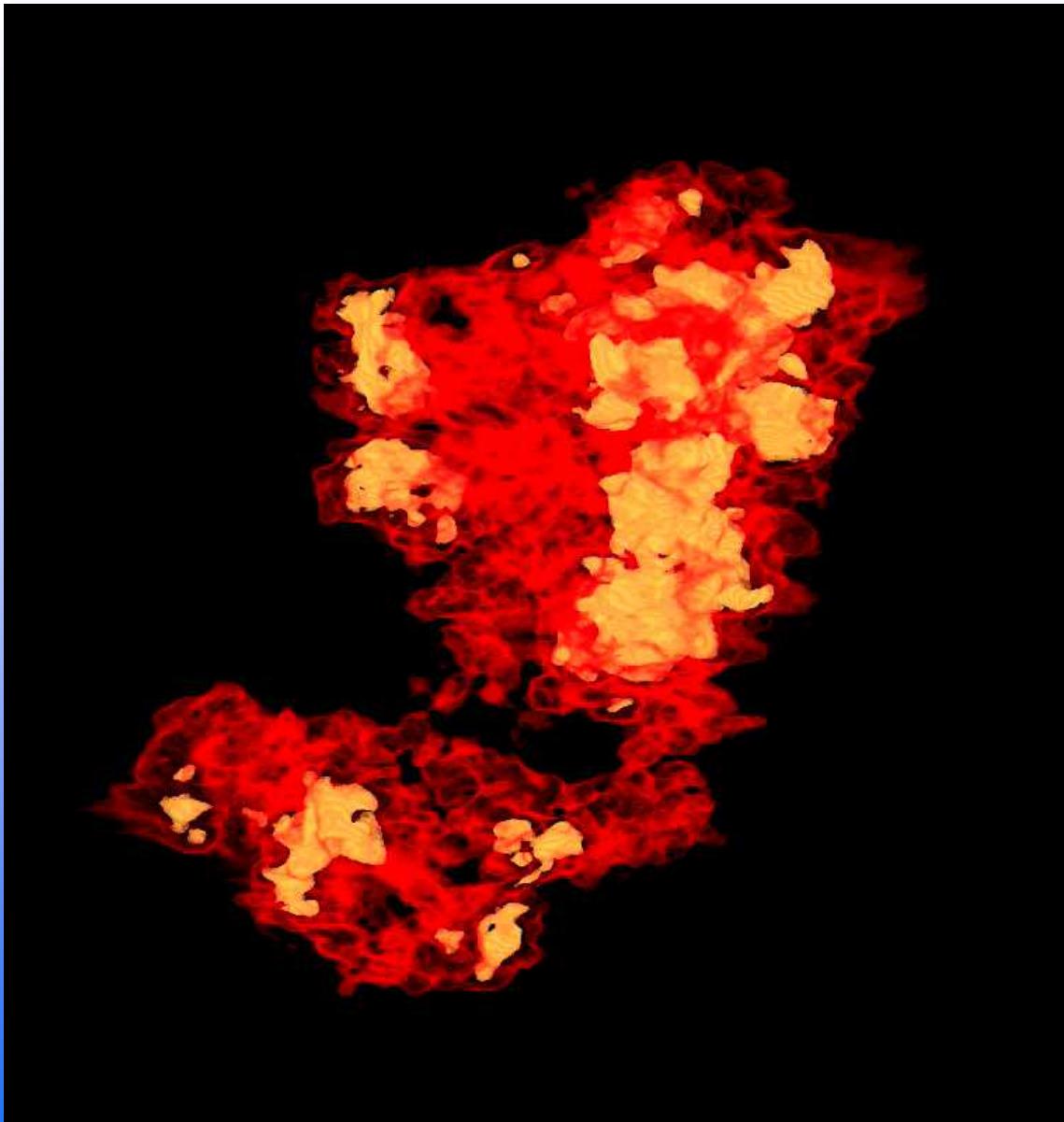
Catalysis: Co_3O_4 on γ -alumina



Tilt Range: -70° to $+70^\circ$ using 1° increments: 141 images

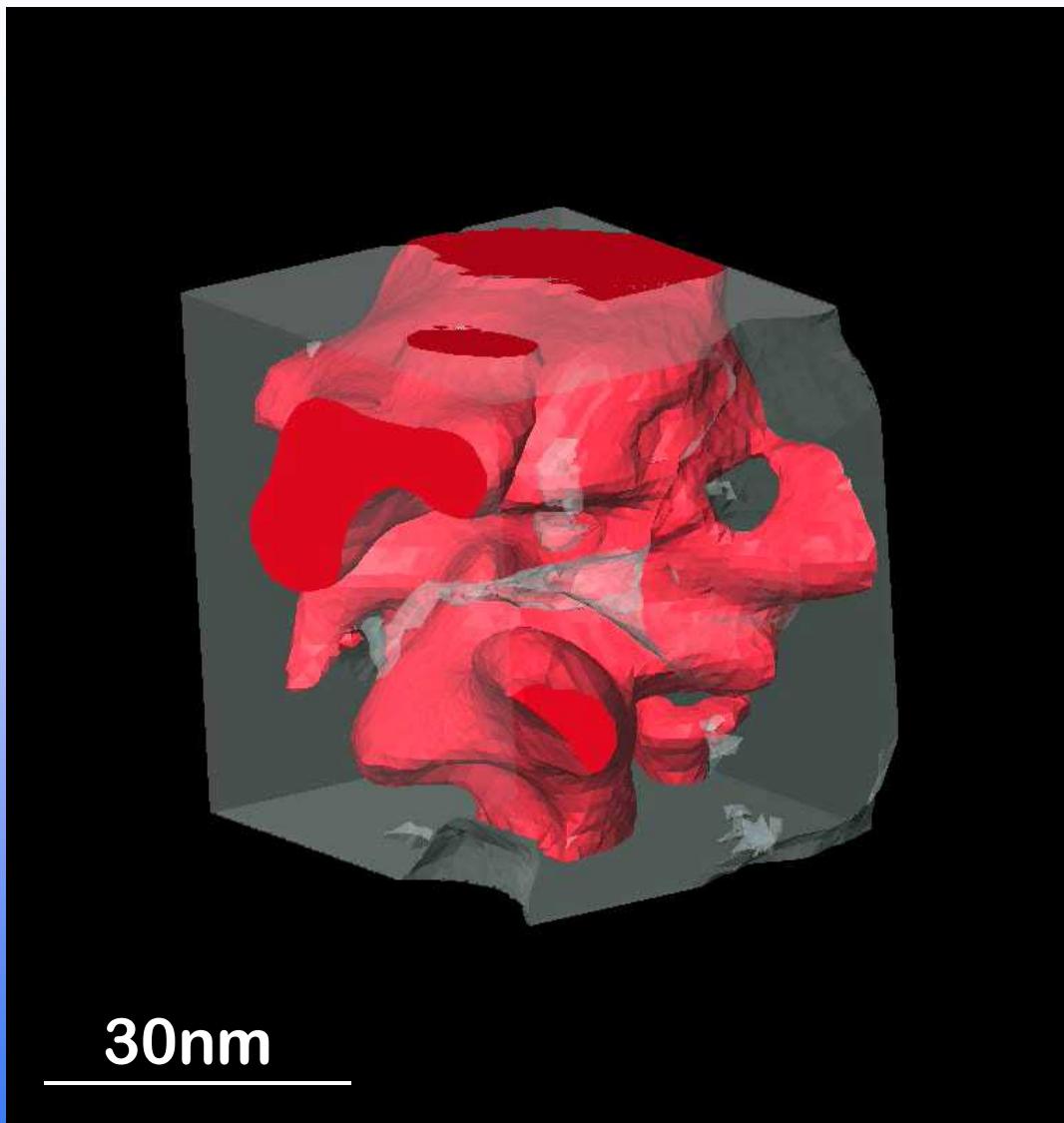


Catalysis: Co_3O_4 on γ -alumina





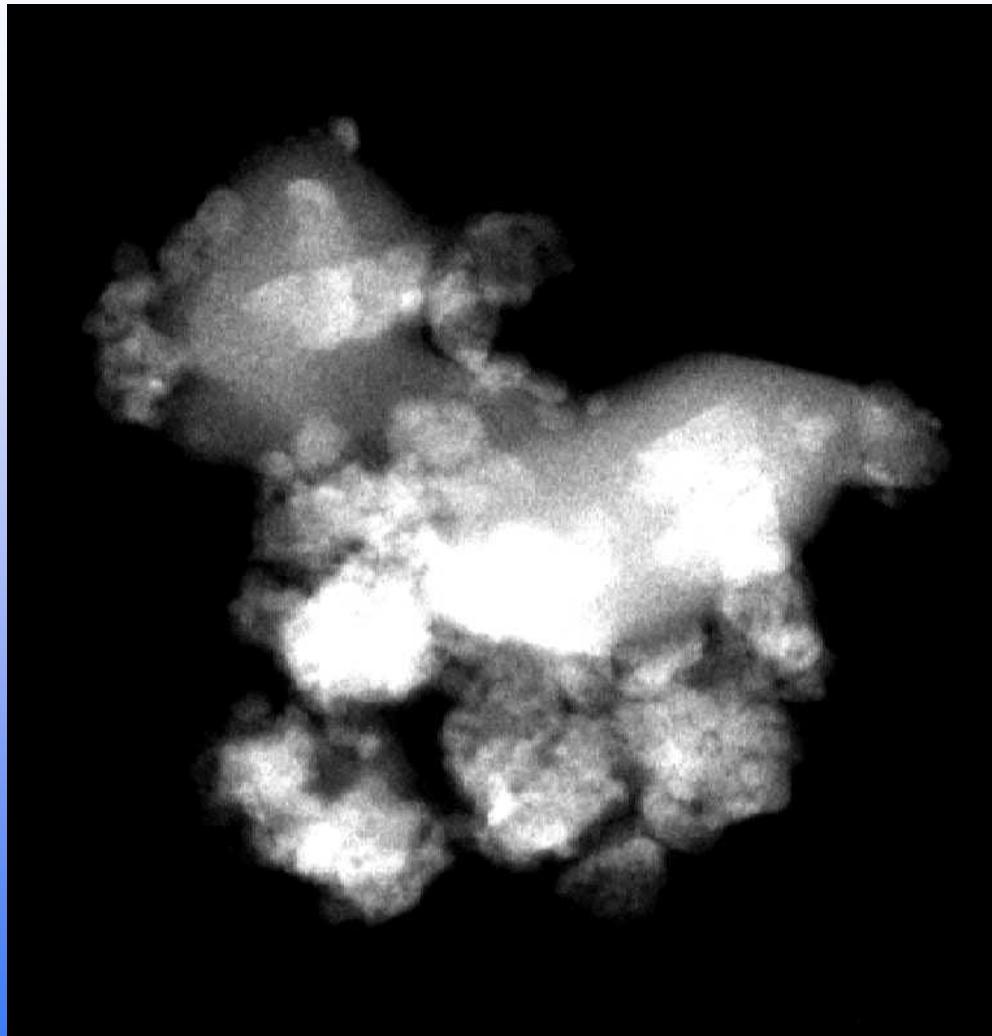
Catalysis: Co_3O_4 on γ -alumina



Porosity: Catalyst: 73% Support: 77% Bulk: 67%



Catalysis: Co_3O_4 on α -alumina/Ni-aluminate

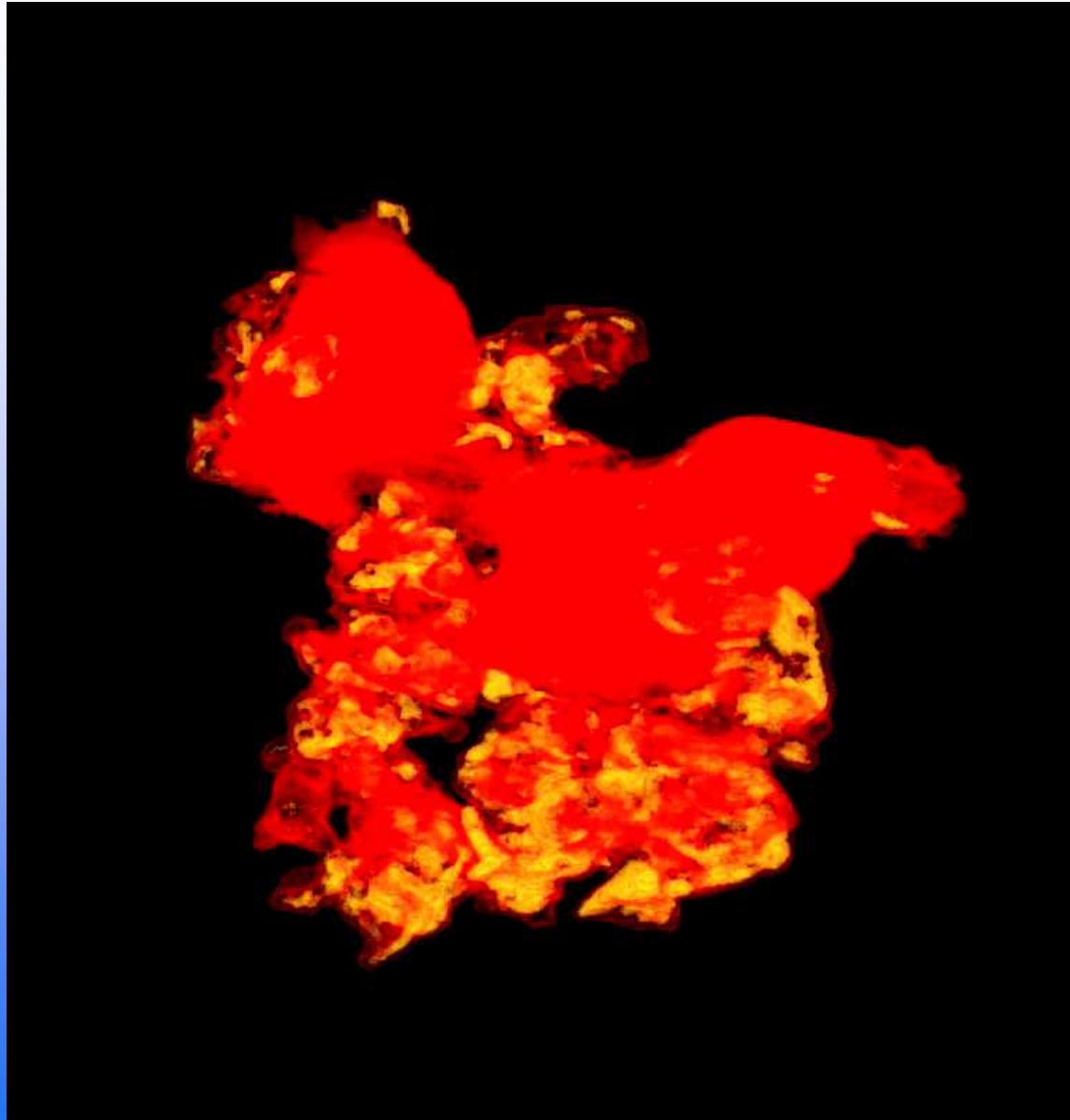


50nm

Tilt Range: -70° to $+70^\circ$ using 1° increments: 141 images

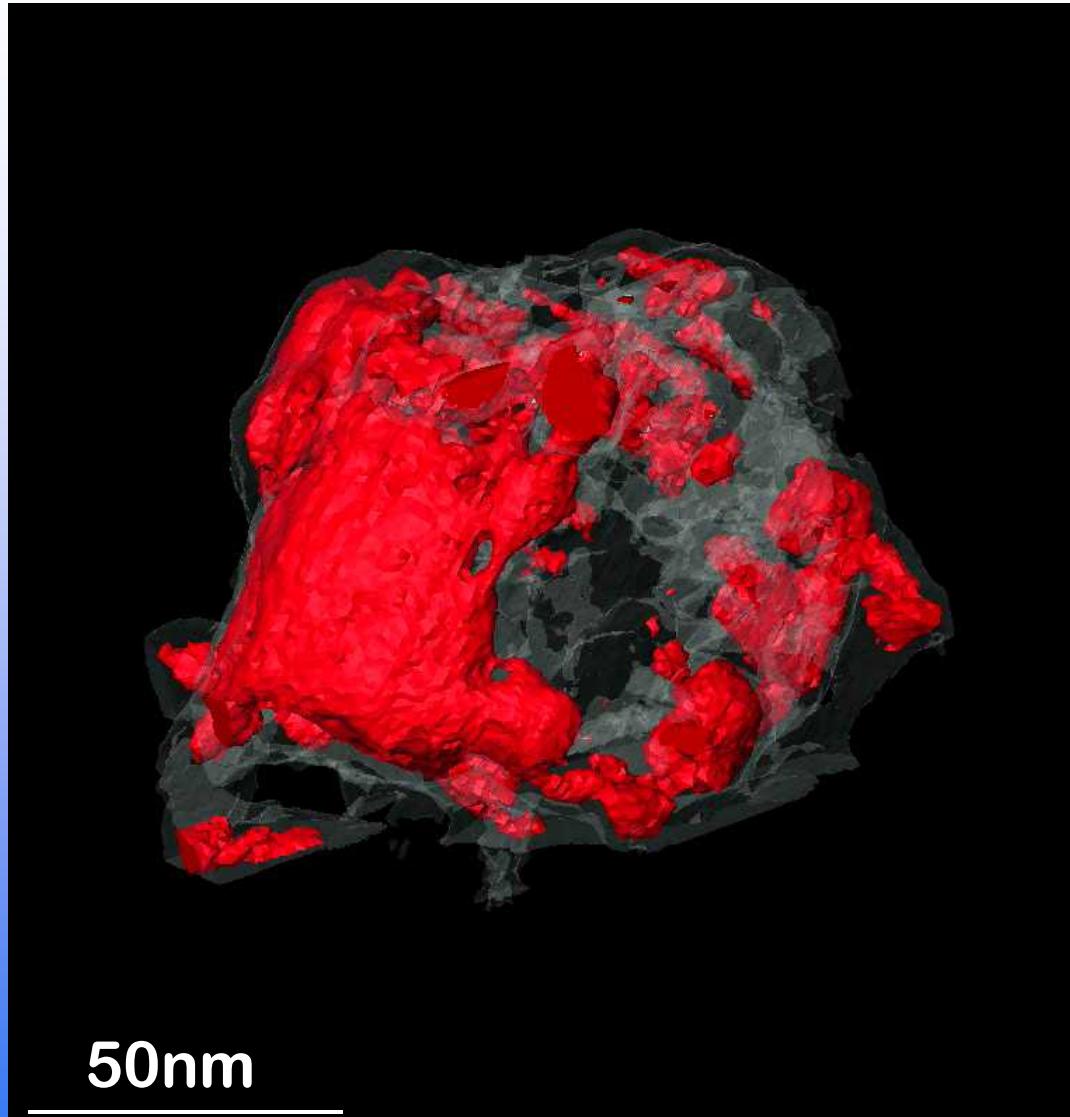


Co_3O_4 on α -alumina/Ni-aluminate





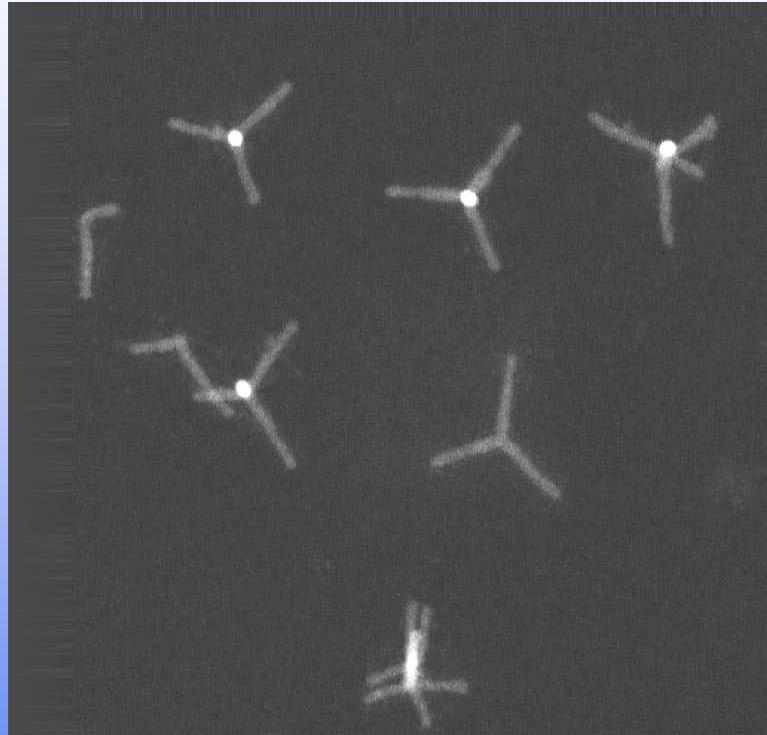
Co₃O₄ on α -alumina/Ni-aluminate



Porosity: Support: 57% Bulk: 42%



Tomography of CdTe Tetrapods



50nm

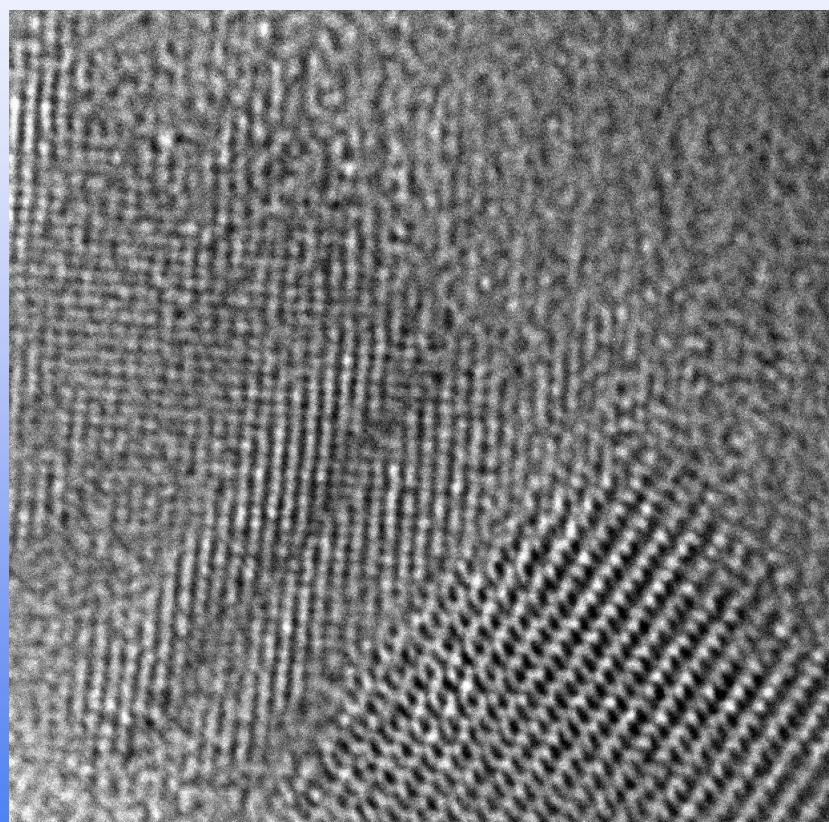
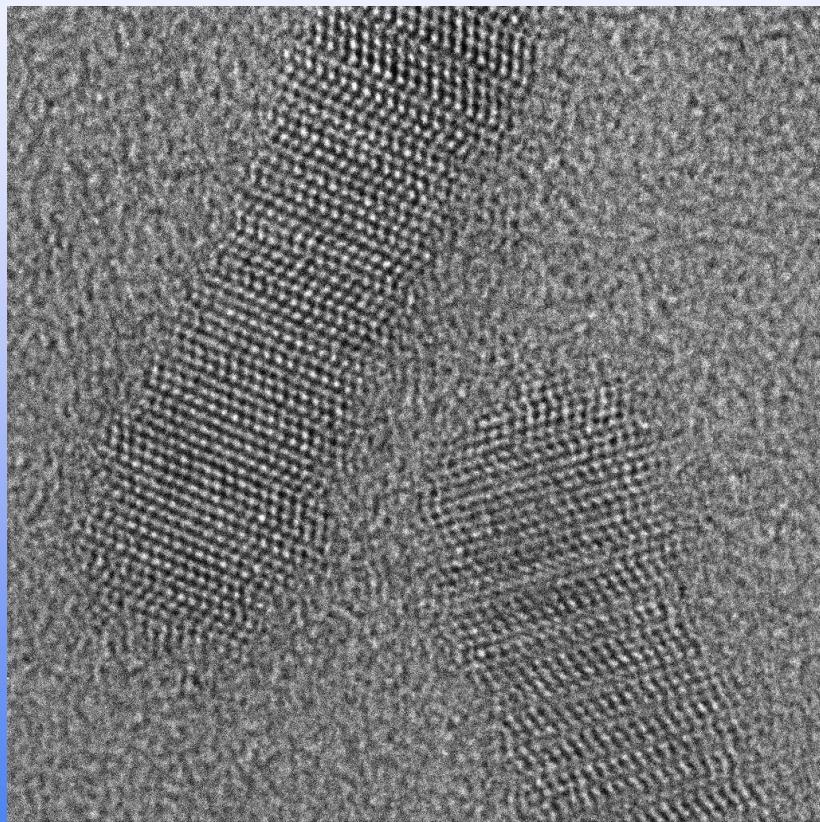


50nm

Tilt Range: -75° to +75° using 1° increments: 151 images

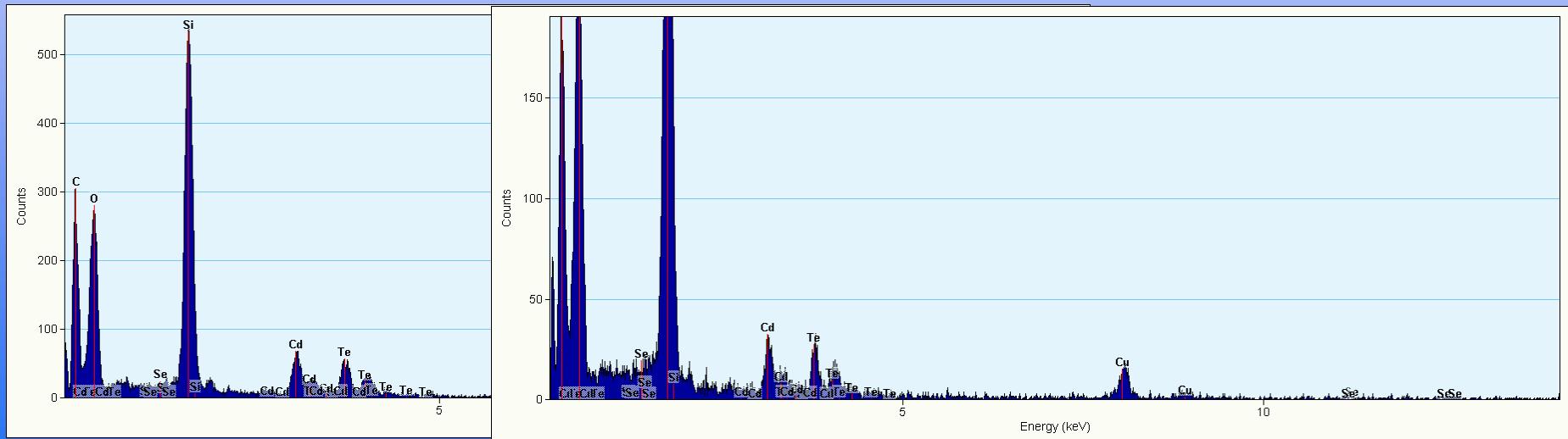
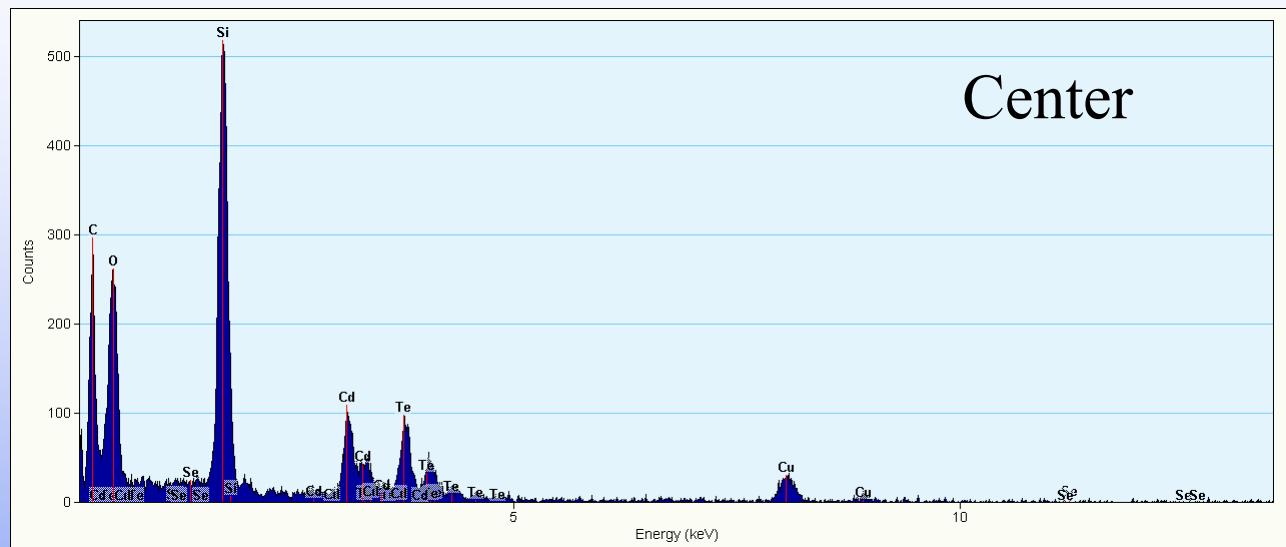
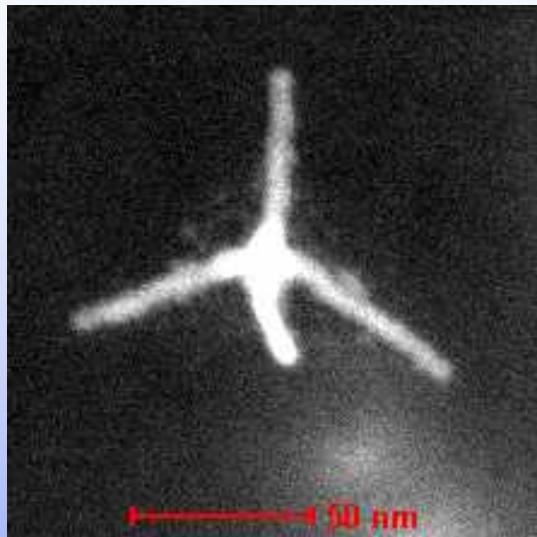


HR-TEM Images of CdTe Tetrapods



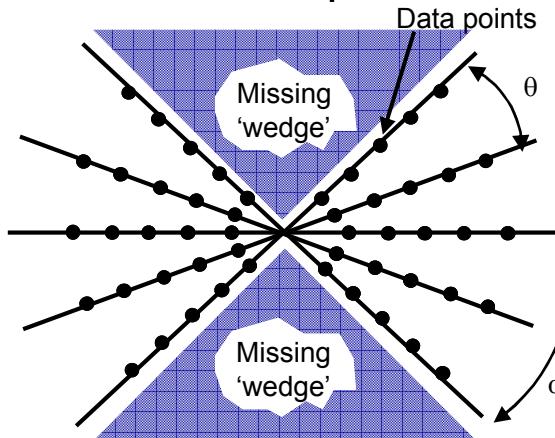


EDX Analysis of CdTe Tetrapods

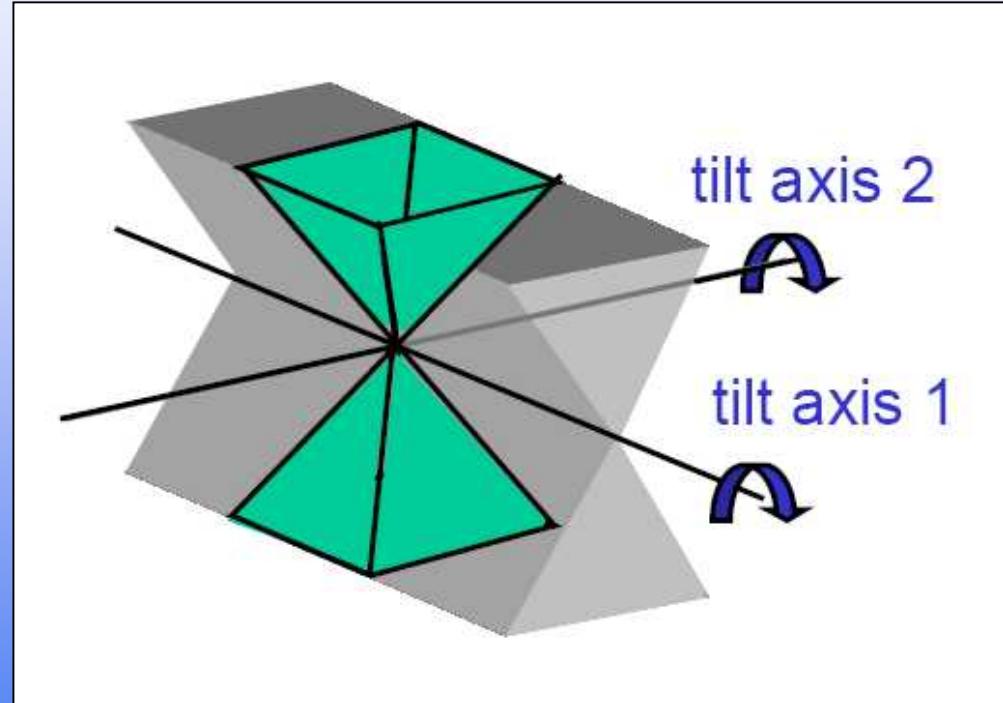


Dual axis tomography: Reducing the missing wedge

Representation of sampling in Fourier space



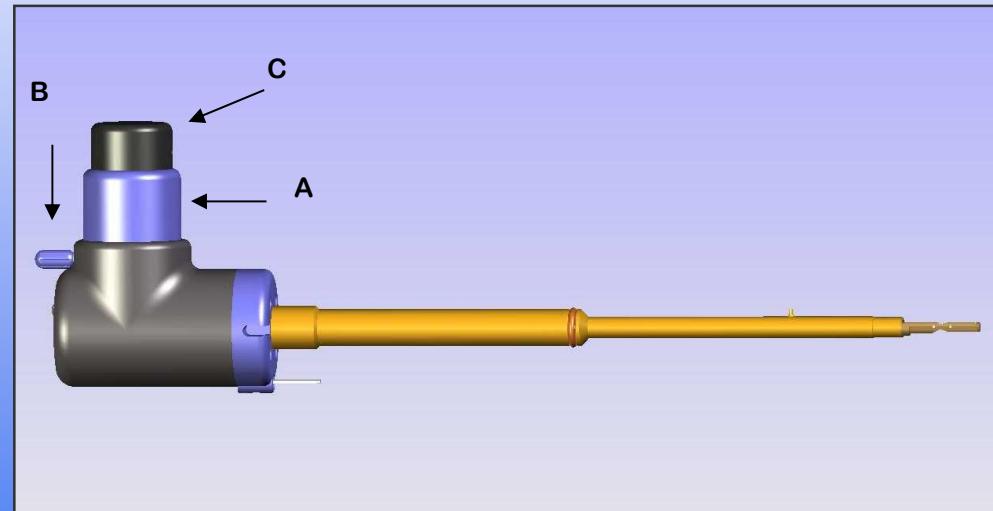
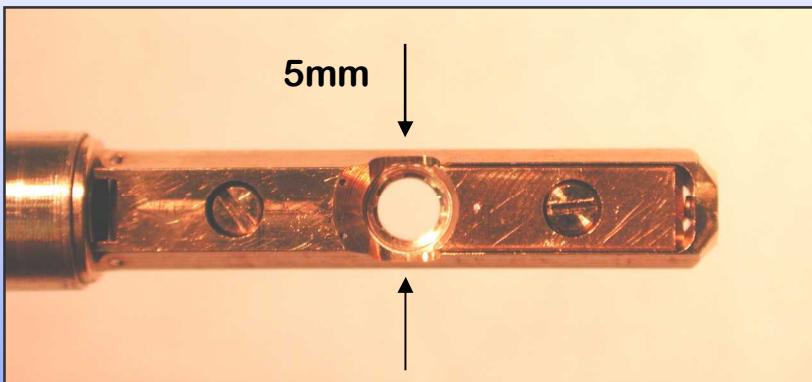
θ is the angular sampling
 α is the maximum tilt angle



Dual axis tomography reduces the amount of missing information in reconstructions from $\sim 20\%$ to just $\sim 2\%!!$



Dual axis tomography: Fischione Advanced Tomography Holder





Dual Axis Reconstructions



Two Perpendicular Tilt Series:

Reconstruct each using WBP
Reconstruct each using SIRT



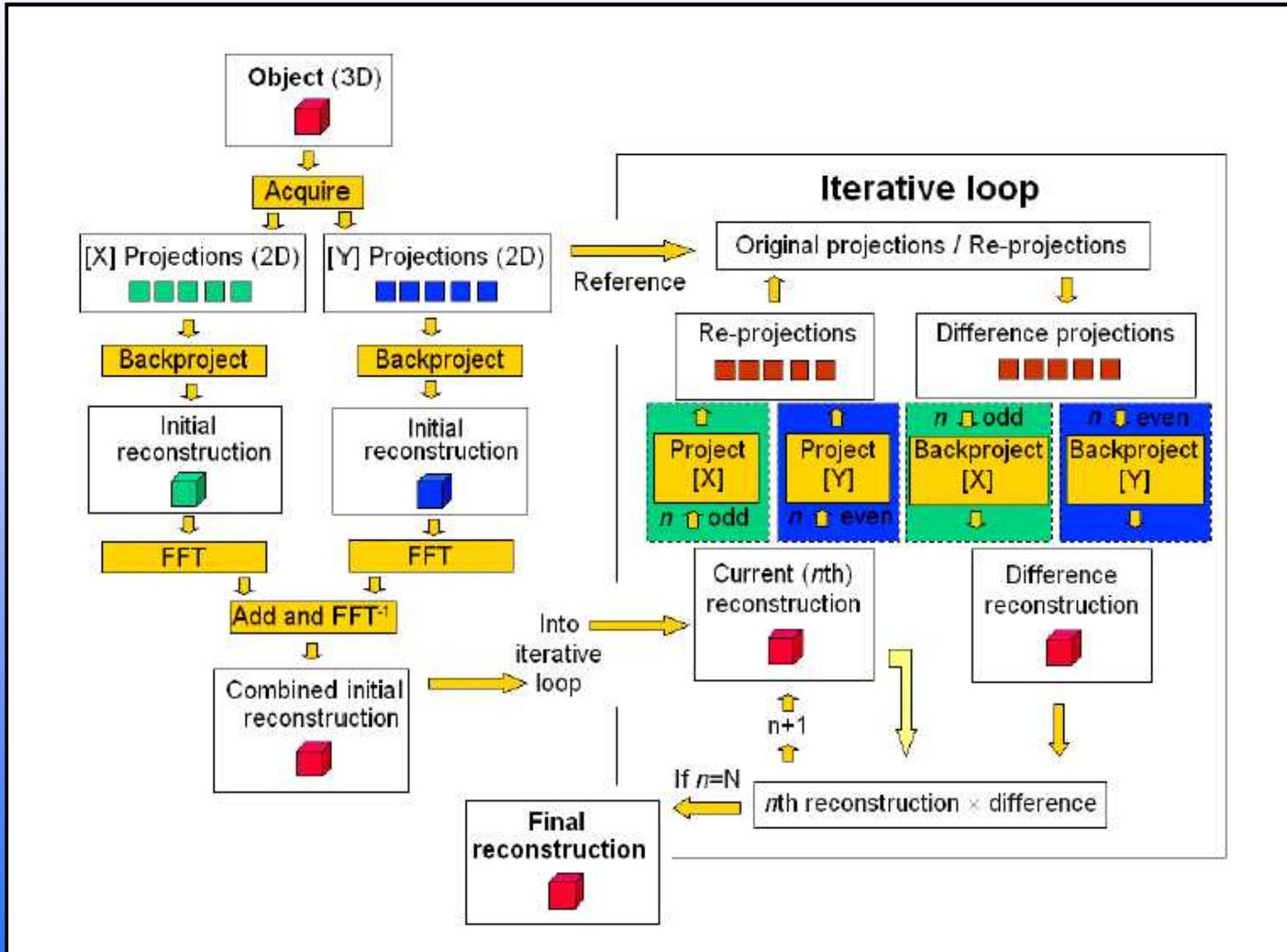
Sum two reconstructions in Fourier space

OR

Alternating Dual Axis SIRT (ADA-SIRT)

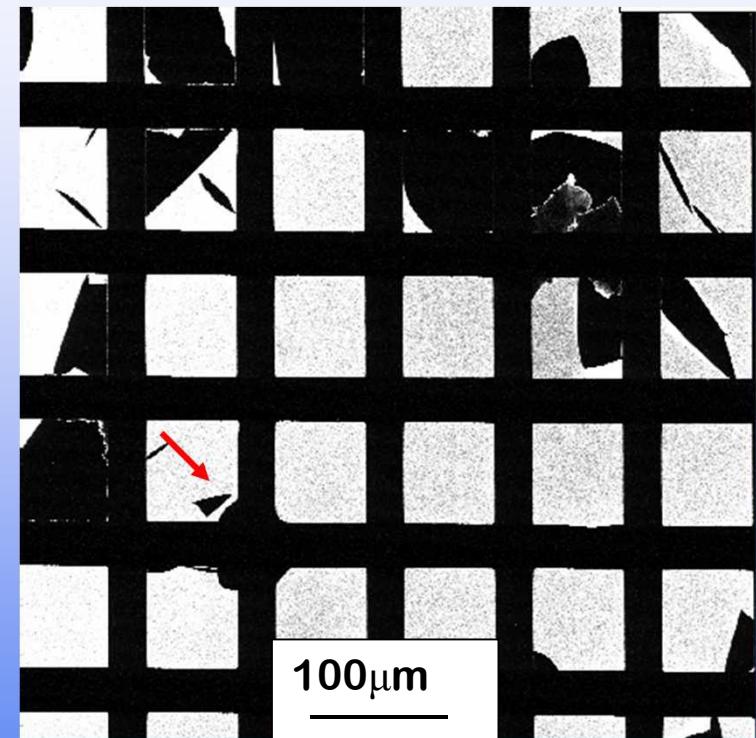
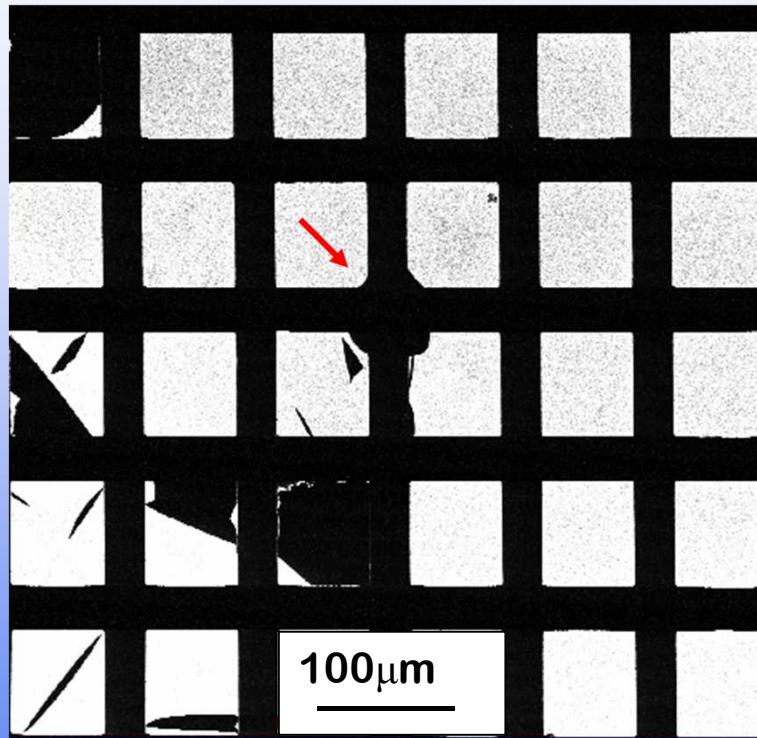


Reconstruction Algorithm: ADA-SIRT





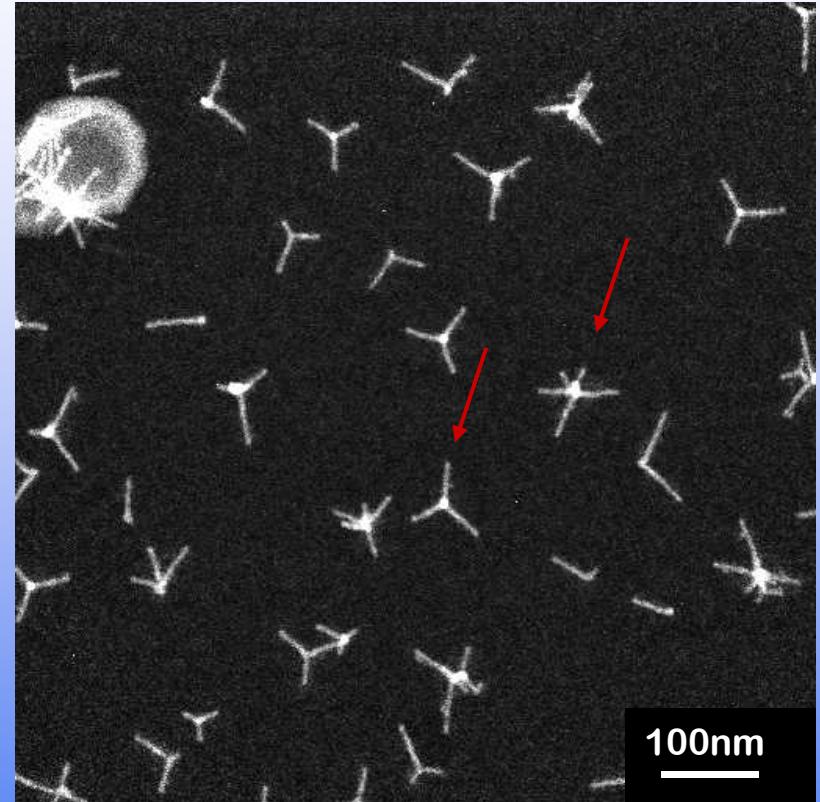
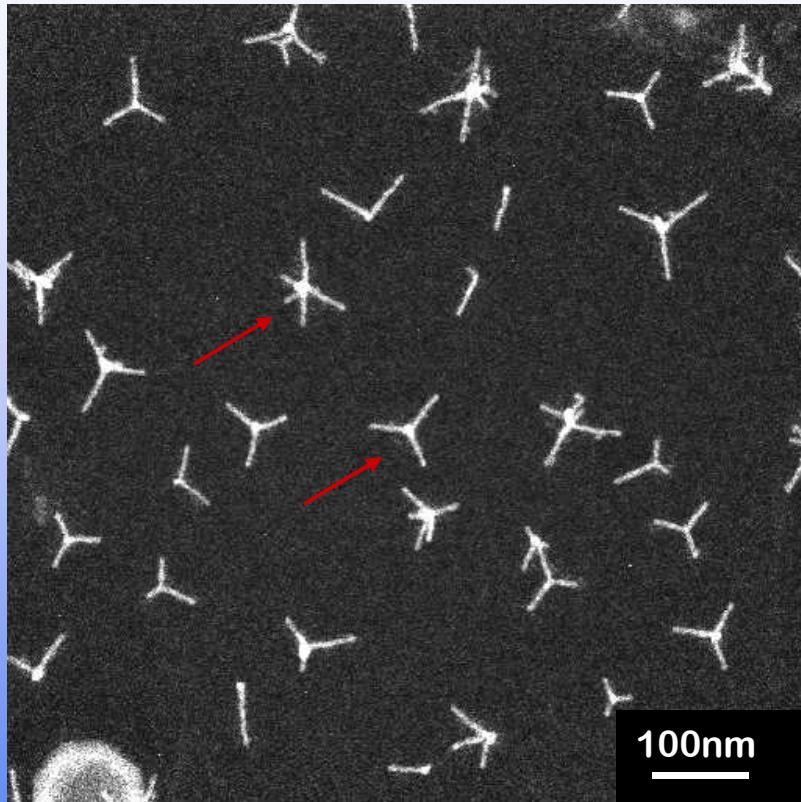
Dual Axis Tomography



Sample is rotated 90° with an accuracy better than 1°.



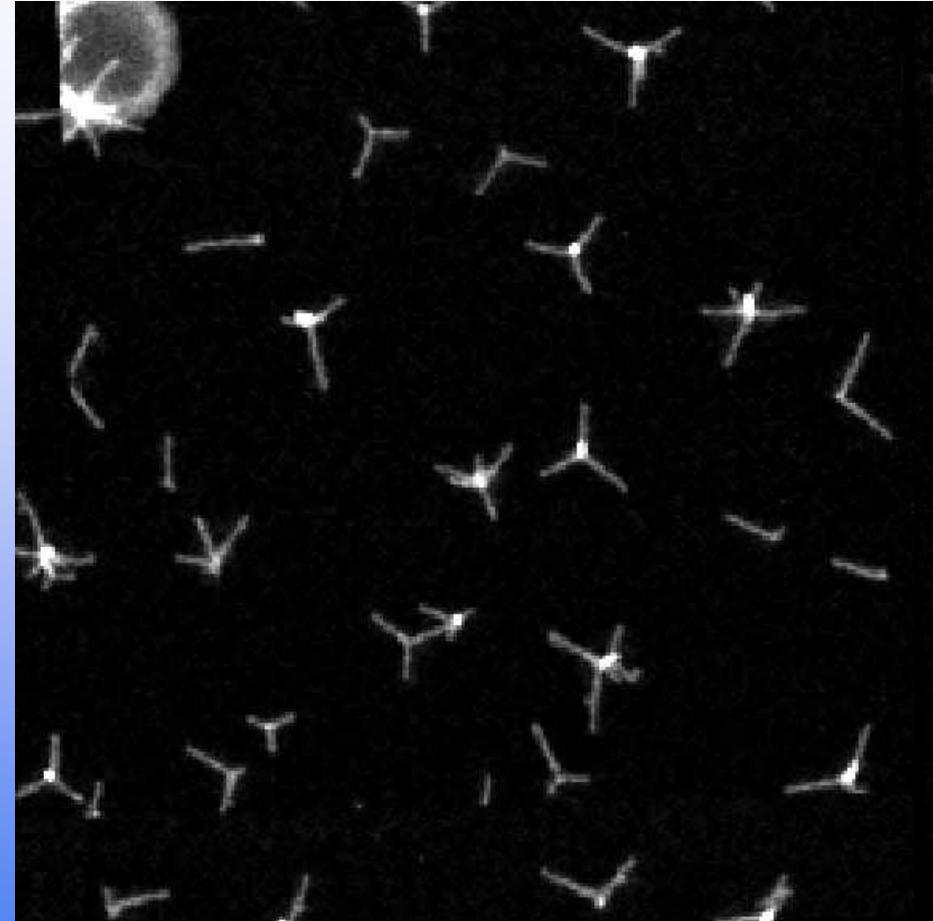
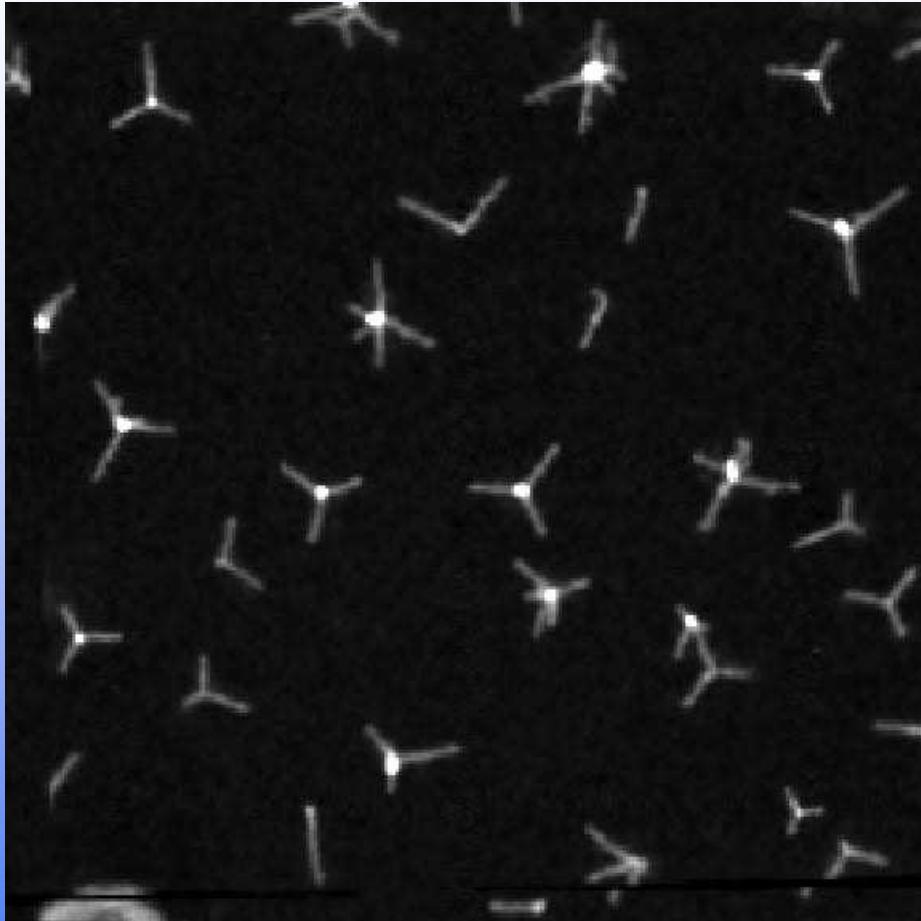
Dual Axis Tomography



0° projection of two perpendicular tilt series



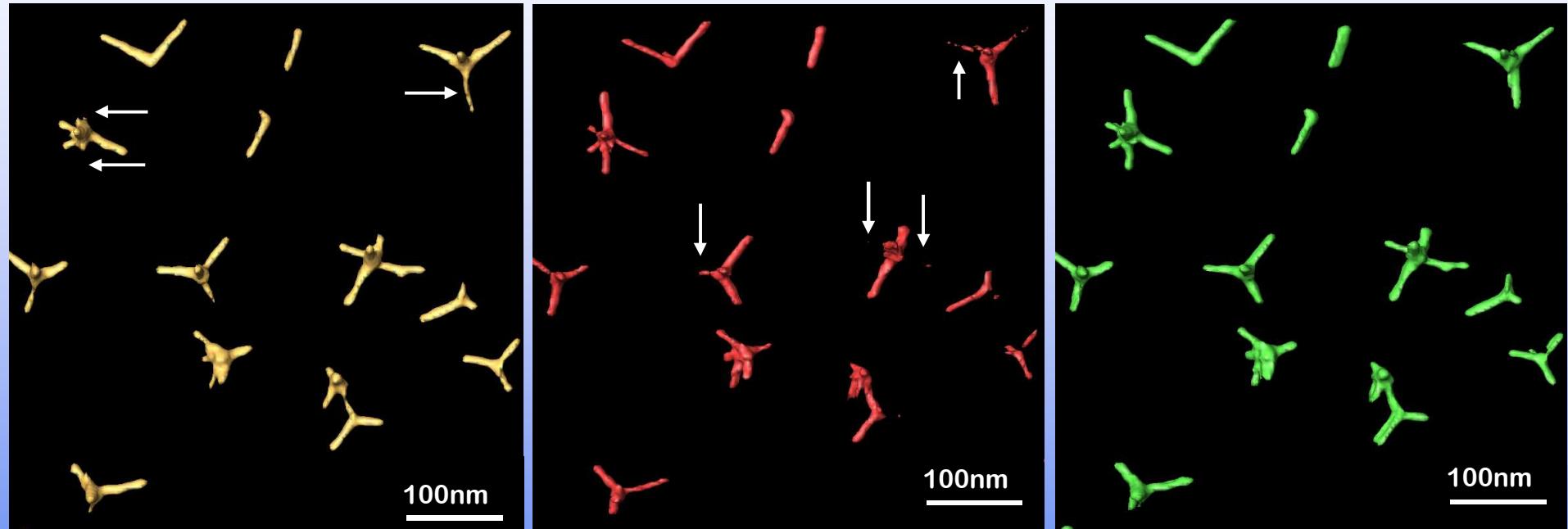
Dual Axis Tomography



69 images each, $\pm 70^\circ$, -65° to $+70^\circ$ for second



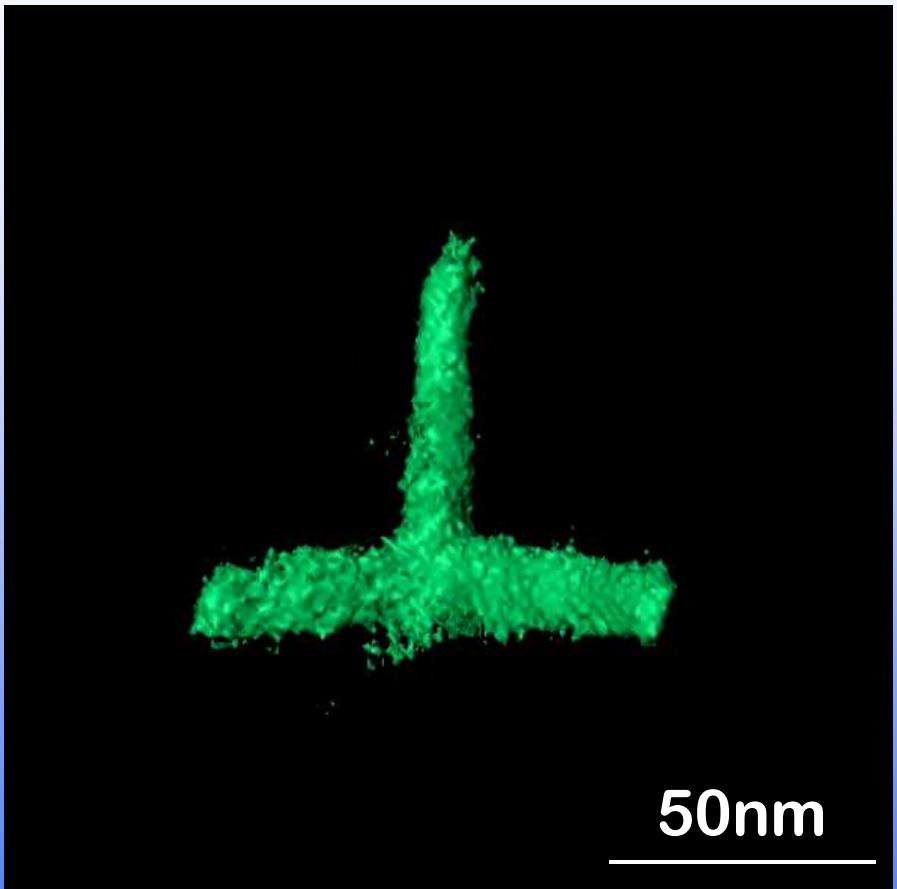
Dual Axis Reconstructions



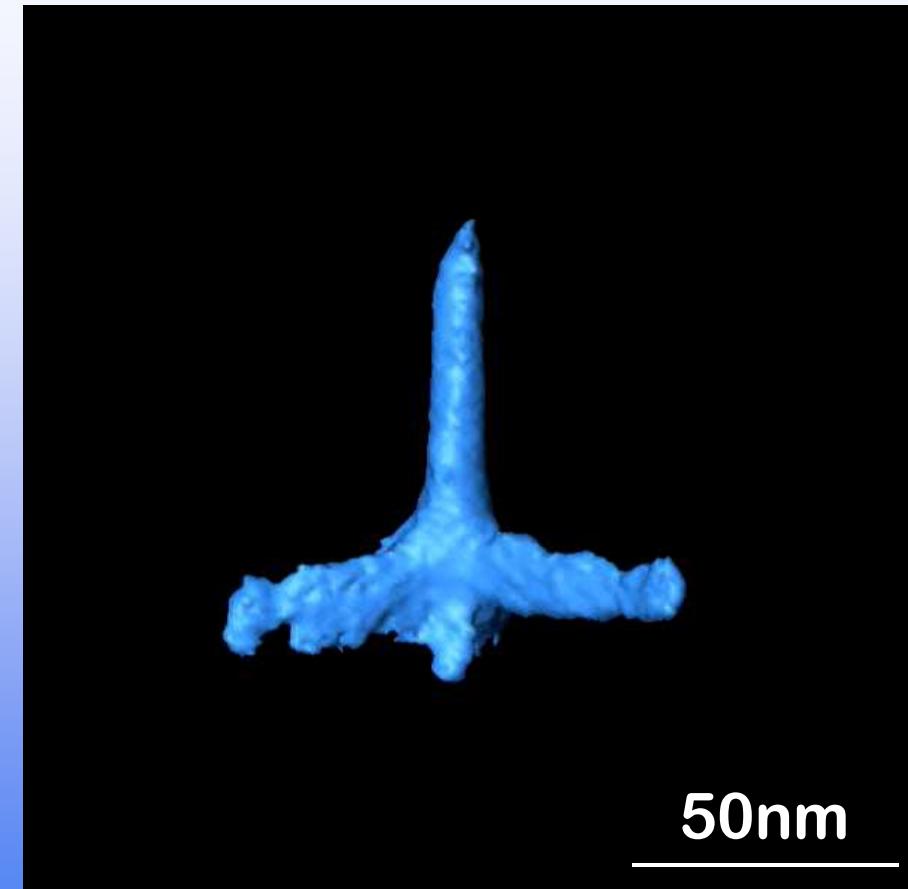
Dual axis reconstructs all legs



Dual Axis Tomography



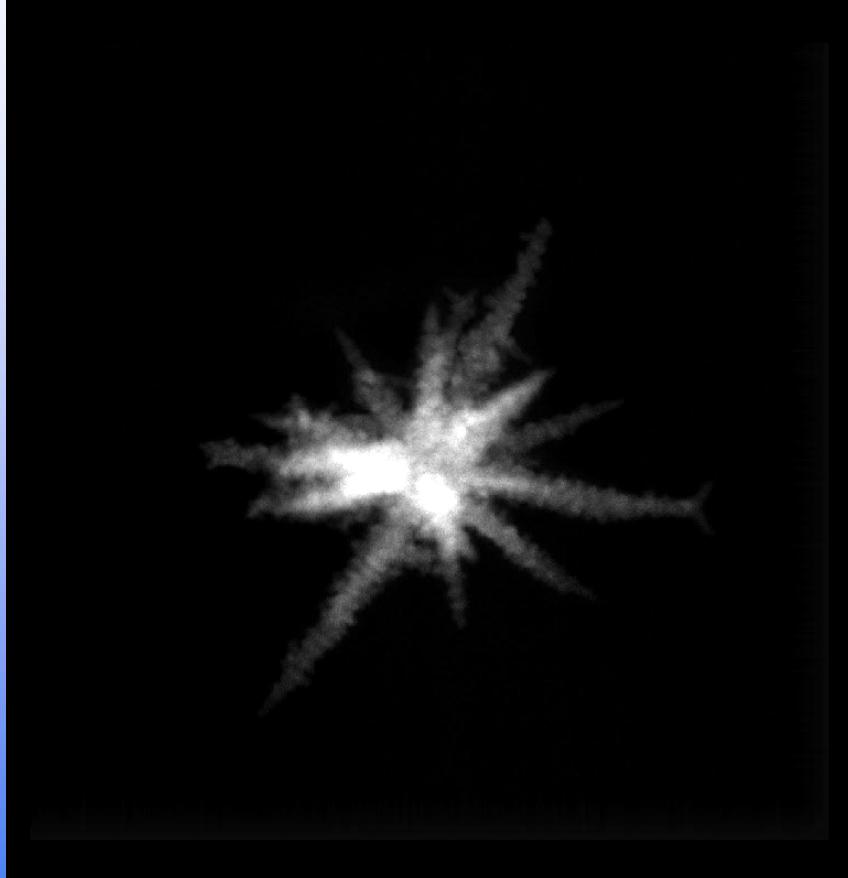
Fourier Sum



ADA-SIRT

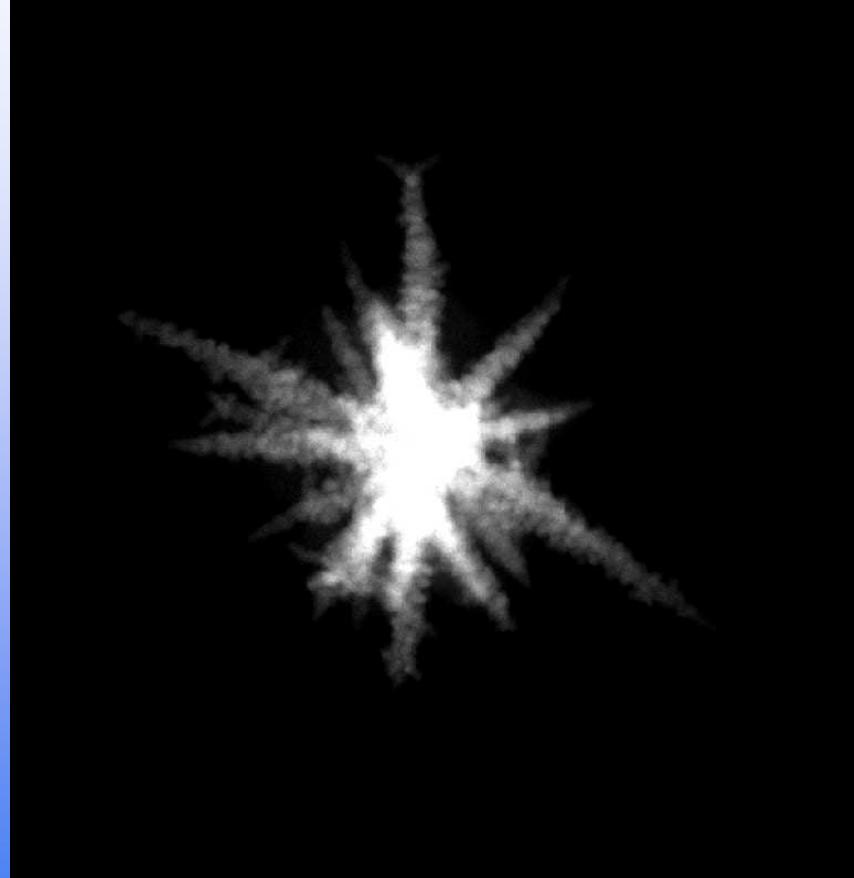


Dual Axis Tomography of Multi-pod



50nm

149 images, -74° to $+74^\circ$

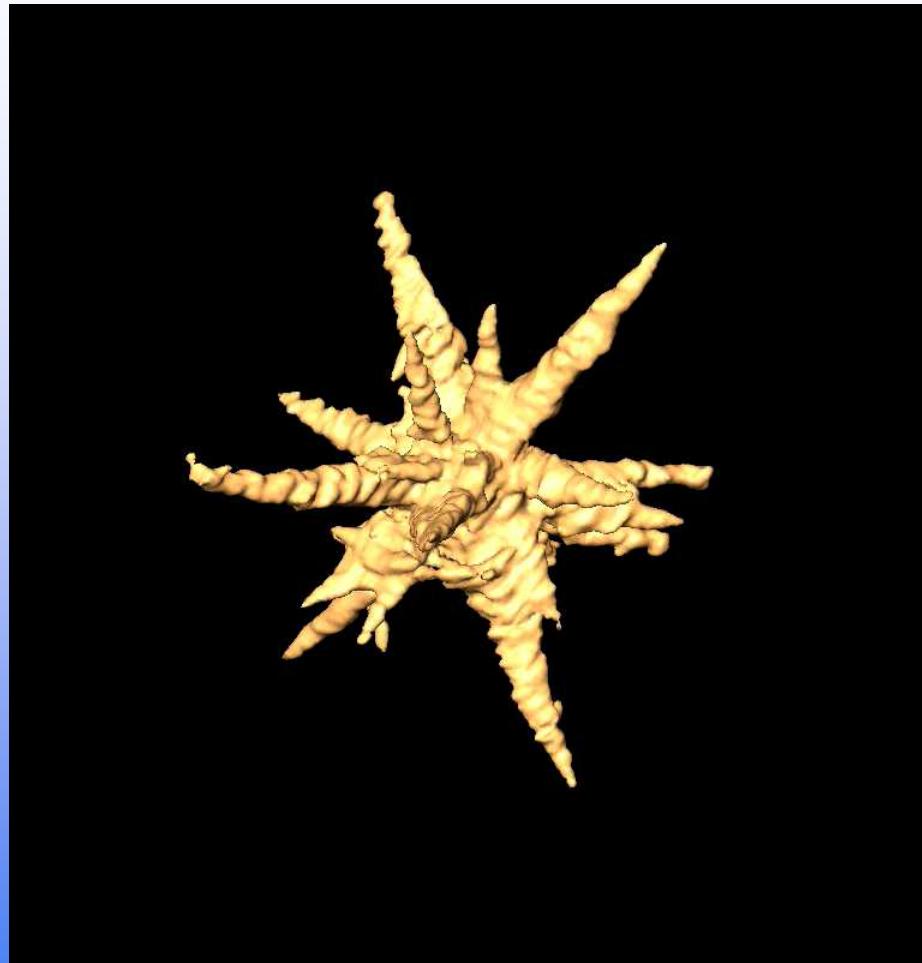


74 images, -74° to $+62^\circ$

50nm



Multi-pod Reconstruction

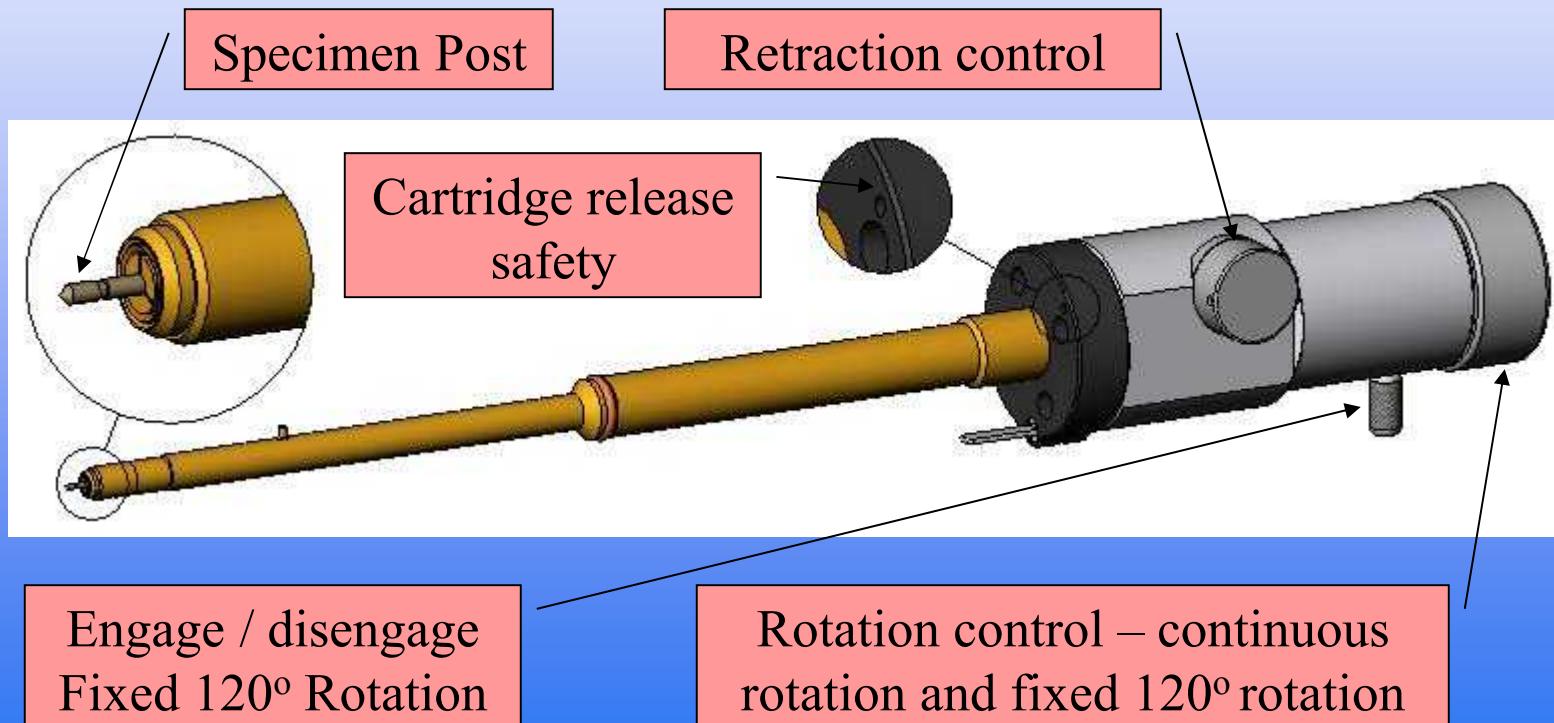


50nm



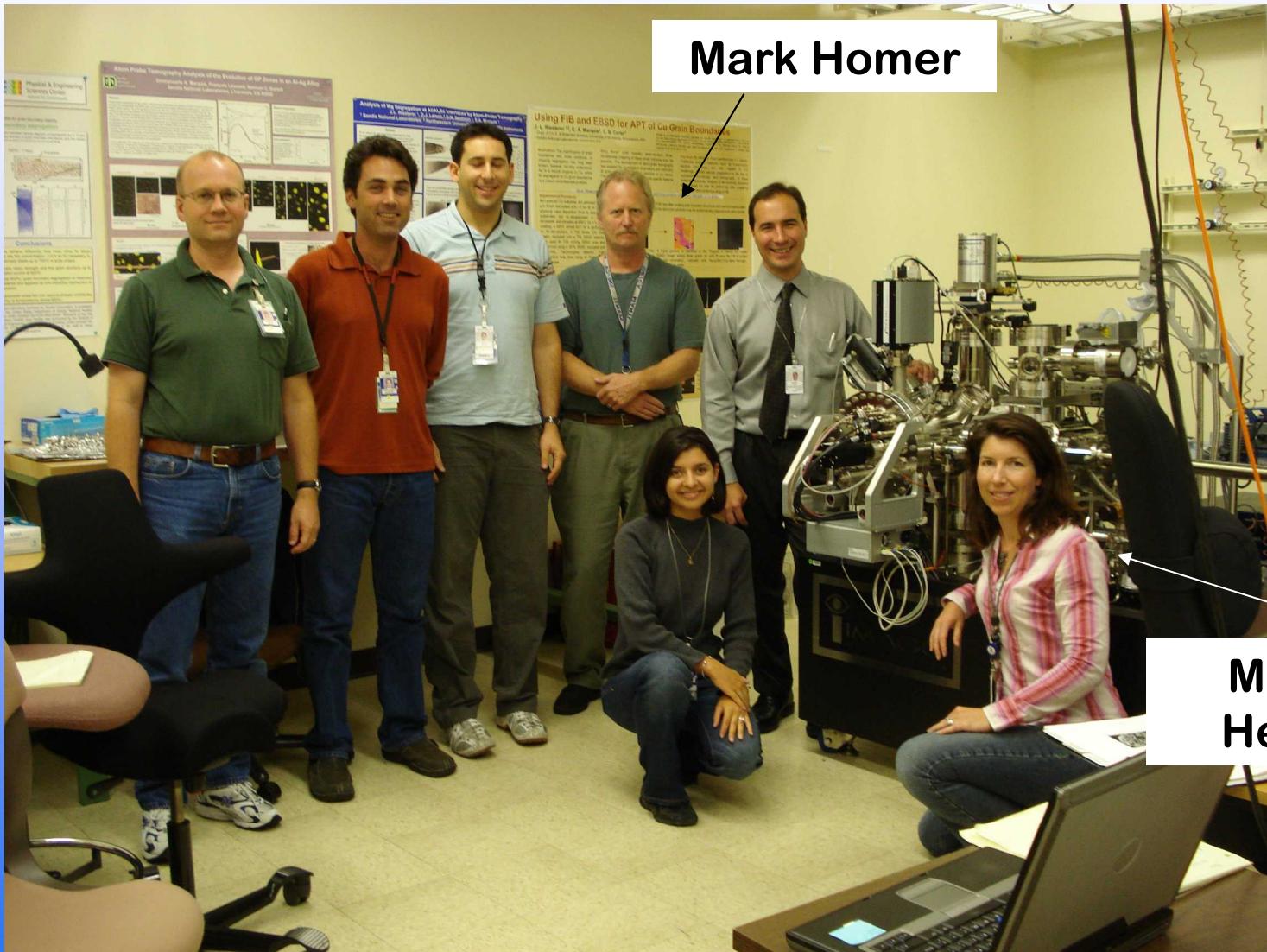
Correlative Techniques-Electron Tomography and Atom Probe Tomography

Model 2050 On-Axis Rotation Tomography Holder



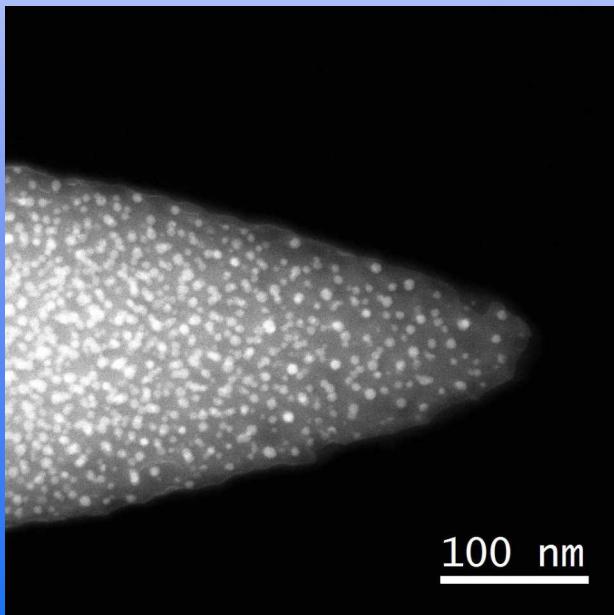
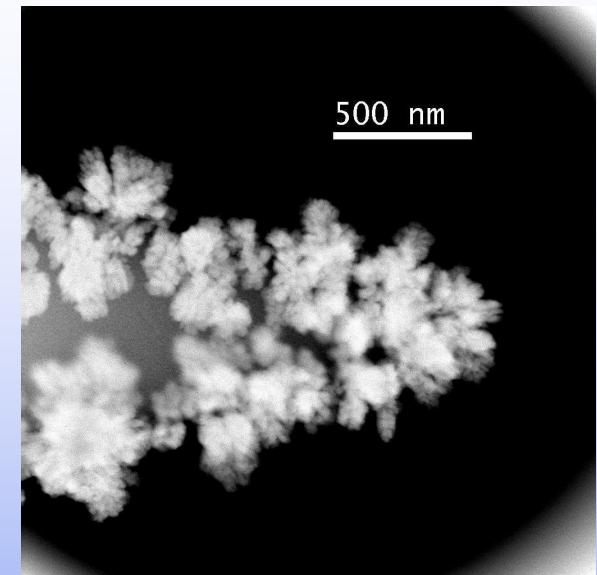
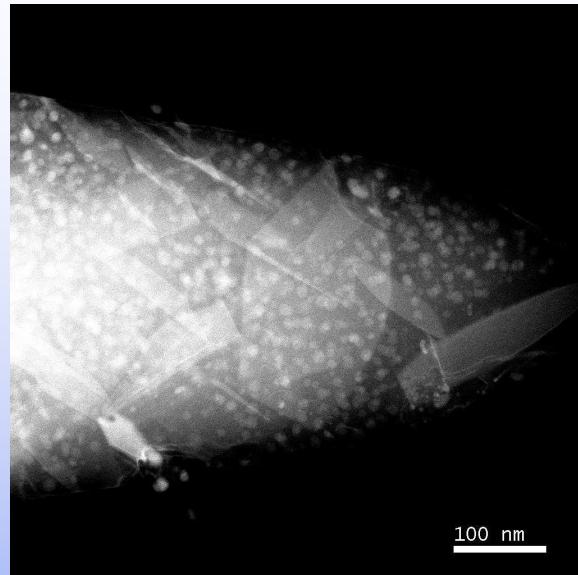
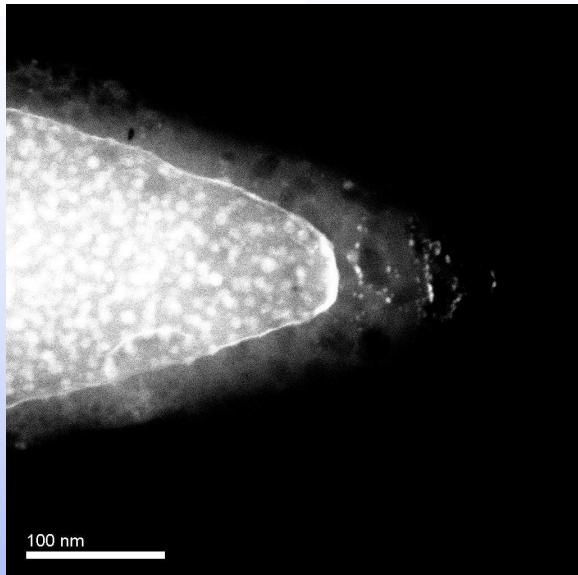


Atom Probe at Sandia/CA





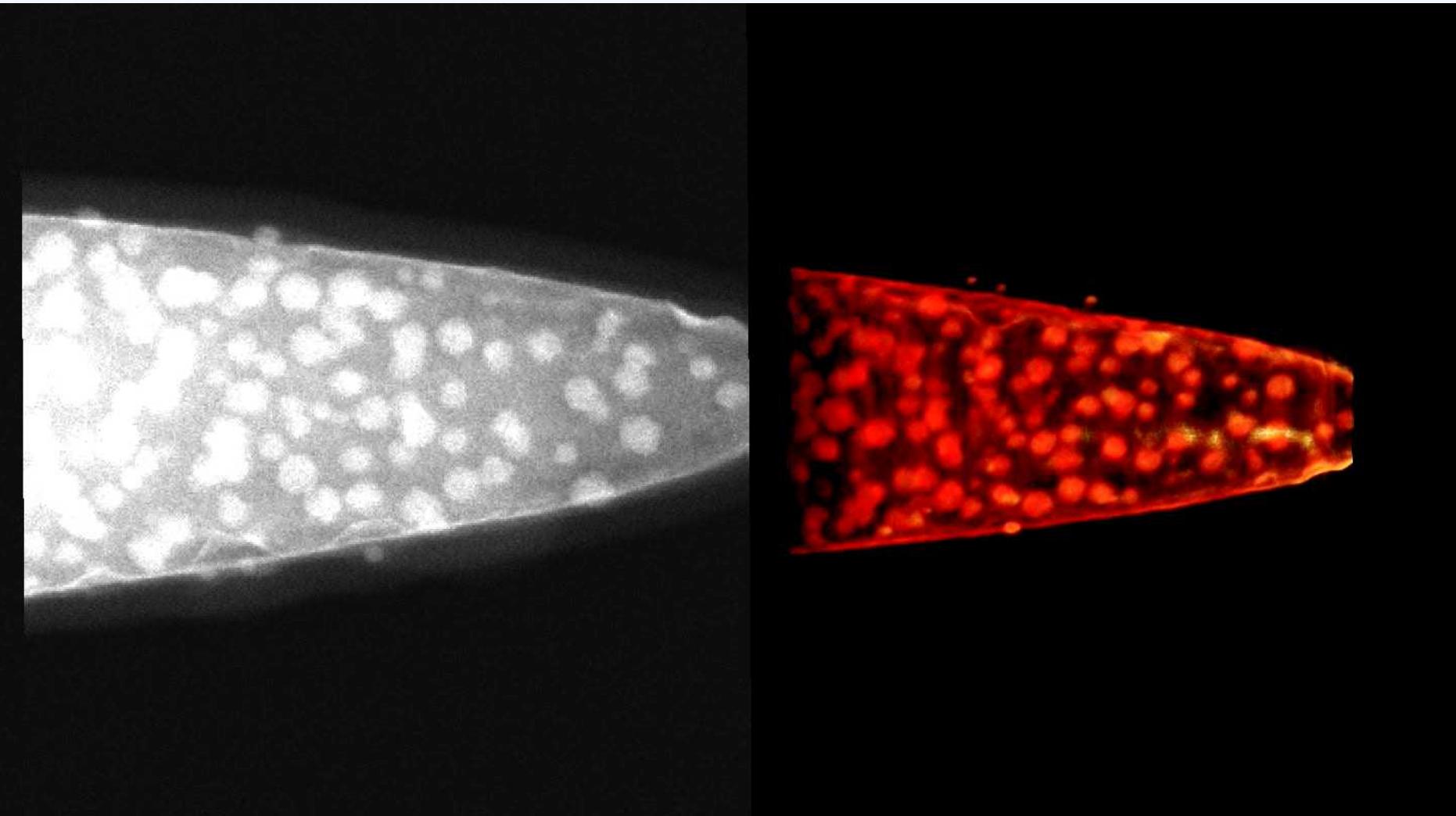
Correlative Techniques-Electron Tomography and Atom Probe Tomography



On average, 1 out
of 5 tips run in the
atom probe

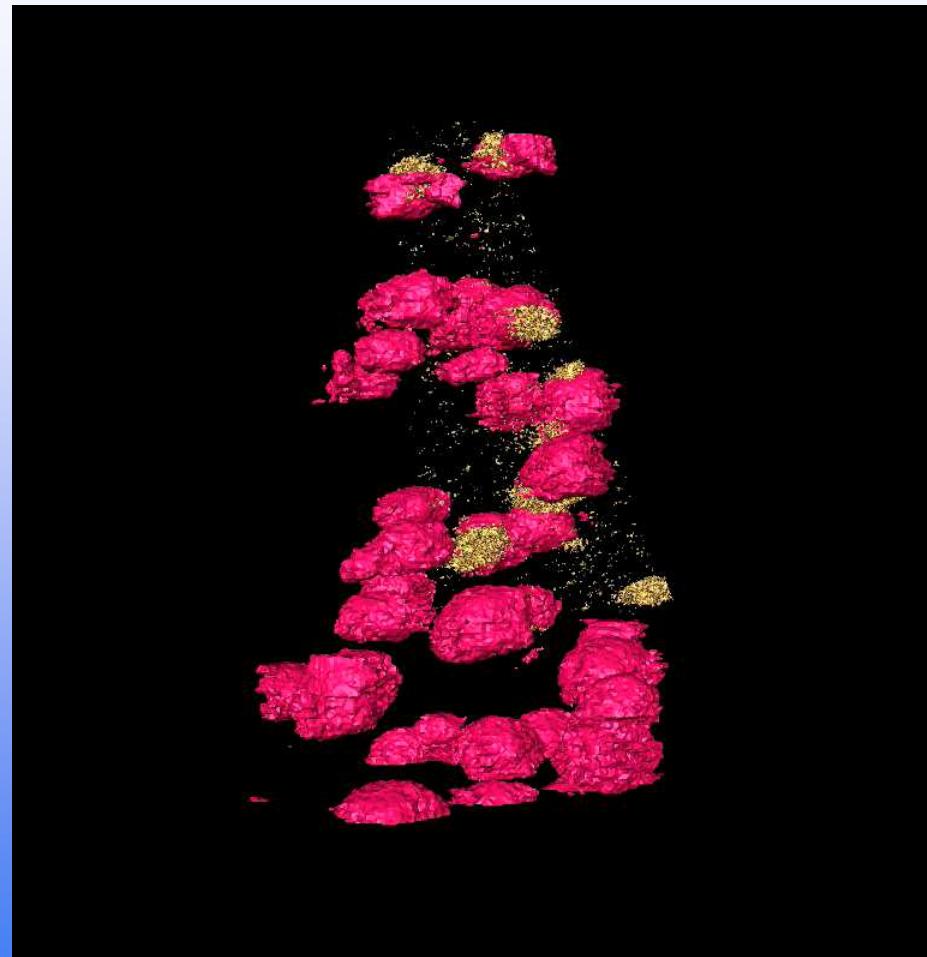
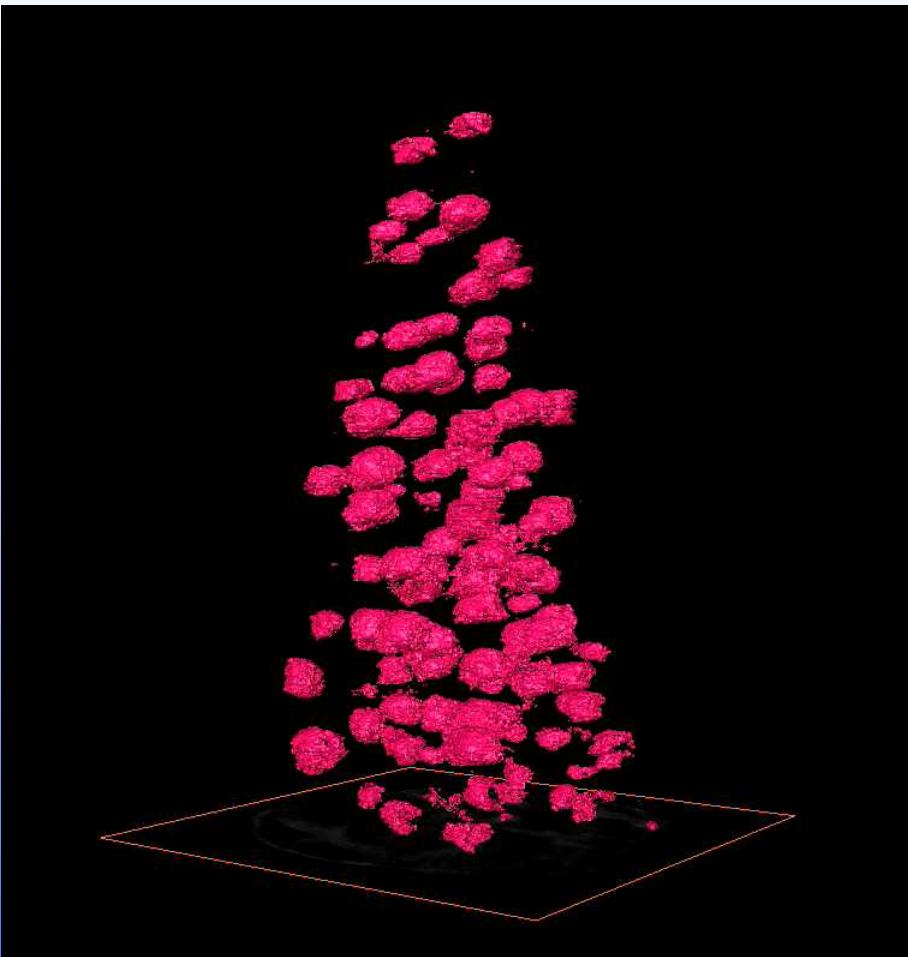


On-Axis Tomography





On-Axis Tomography





Acknowledgements



- Paul Midgley
- Timothy Yates
- Jenna Tong
- Nigel Browning
- John Walmsley
- Erling Rytter
- Edvard Bergene
- Chris Booth et. al.
- Norman Bartelt
- Mark Homer
- Michelle Hekmaty
- Emmanuelle Marquis
- Alivisatos Group

University of Cambridge
University of Cambridge
University of Cambridge
UC Davis/LLNL
SINTEF
Statoil Research Centre
SINTEF/Statoil
Gatan, Inc.
Sandia/CA
Sandia/CA
Sandia/CA
Oxford University
UC Berkeley

Funding

Sandia's President
Harry S. Truman
Fellowship in National
Security Science and
Engineering, a
Laboratory Directed
Research and
Development Program
(LDRD)