X-ray and Neutron Microdiffraction

Gene E. Ice

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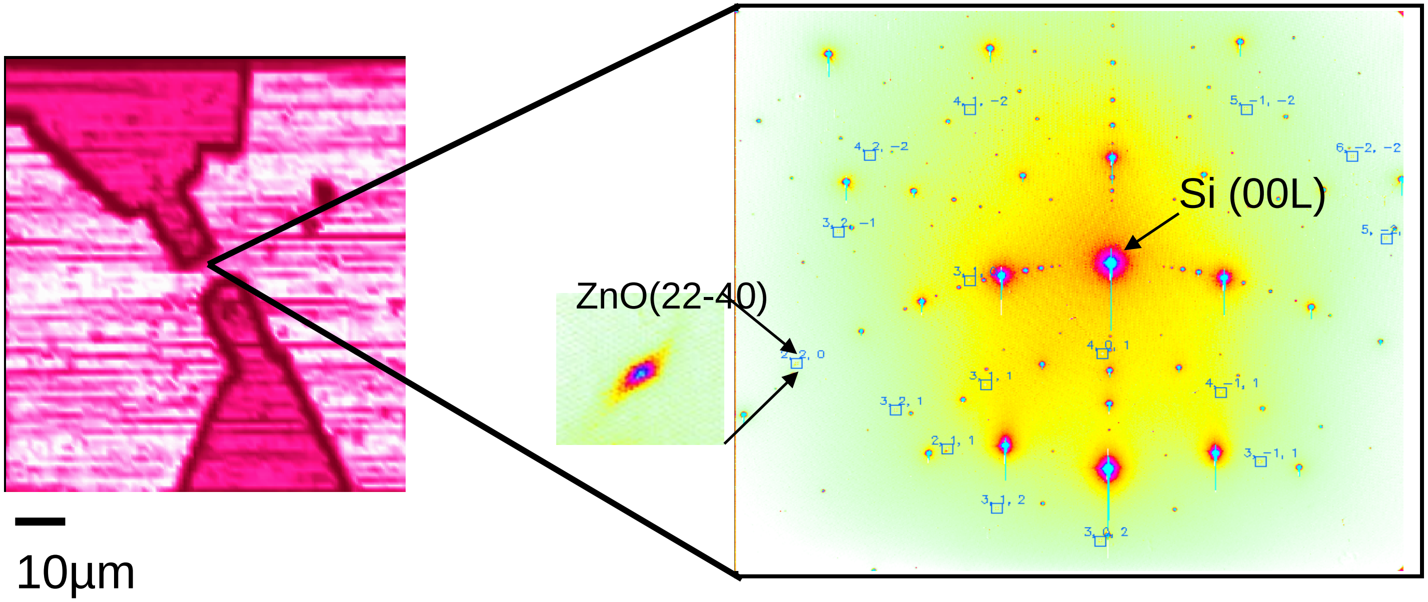


2009 Neutron X-ray Summer School

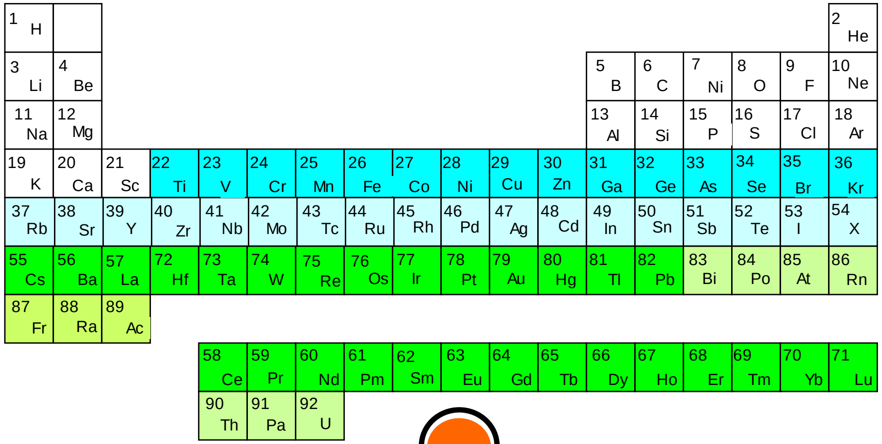
QuickTime™ and a
decompressor
are needed to see this picture.

Two words

Spatial Resolution

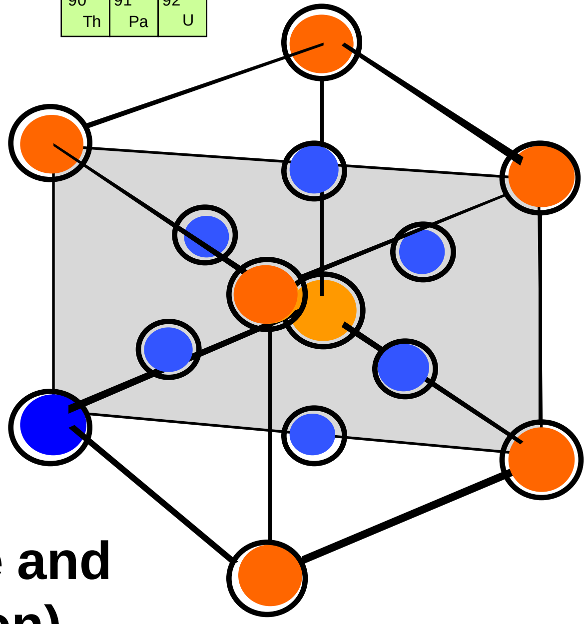
Materials characterization begins 3 questions

- **What is the elemental composition?**



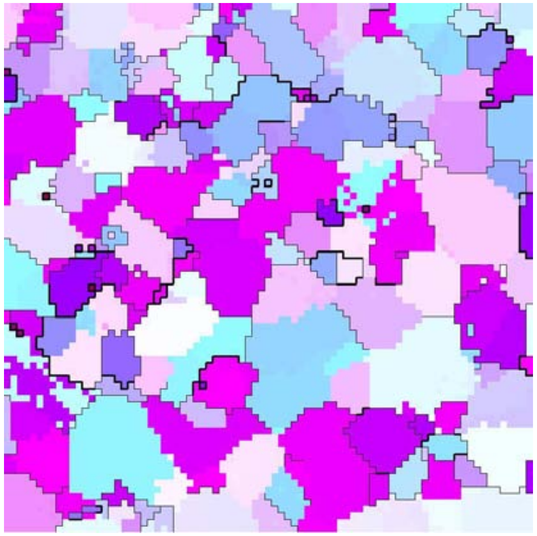
- **What is the crystal/local structure?**

- **What are the defects?**

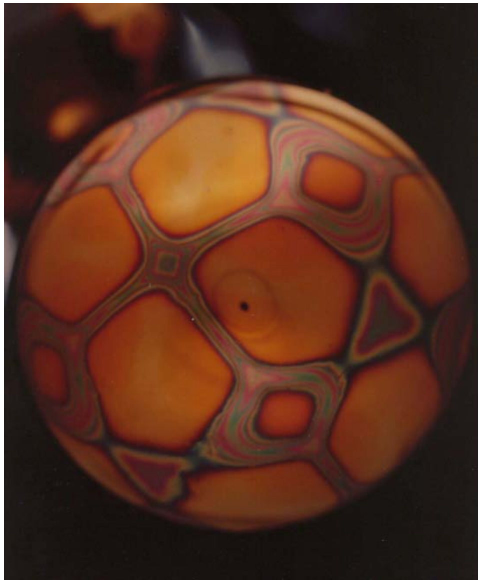


X-rays and neutrons probe structure and defects through scattering (diffraction)

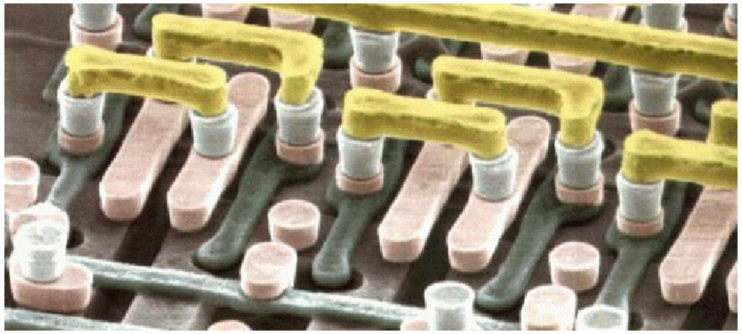
Spatial resolution essential!



- **Most materials** *polycrystalline***(0.1-50 μm)**
  - **Anisotropic**
  - **Heterogeneous**
  - **Plastic/elastic deformation/ diffusion/ oxidation/**



- **Even** *within* **single and "perfect" crystal:**
  - **Strain**
  - **Defects**
  - **Spontaneously organize to reduce energy**



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Spatial resolution essential for most advanced energy systems

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TIFF (Uncompressed) decompressor
are needed to see this picture.

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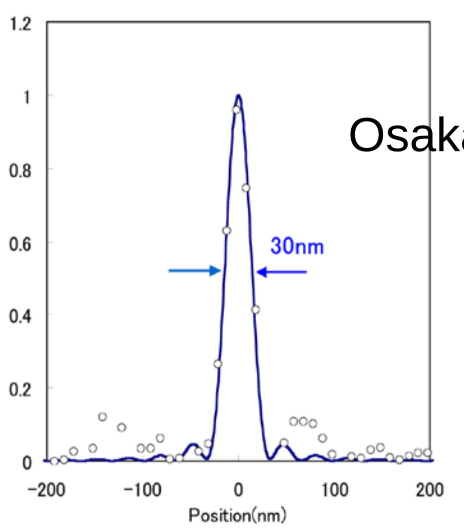
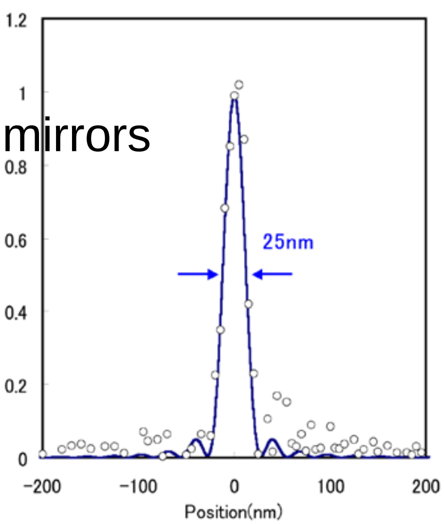
are needed to see this picture.

Optics improving rapidly

- **Both chromatic and achromatic <20 nm**
- **New choices**
  - **Mirrors**
  - **Zone plates**
  - **Laue zone plates**
  - **Compound refractive optics**

QuickTime™ and a
TIFF (Uncompressed) decompressor
are needed to see this picture.

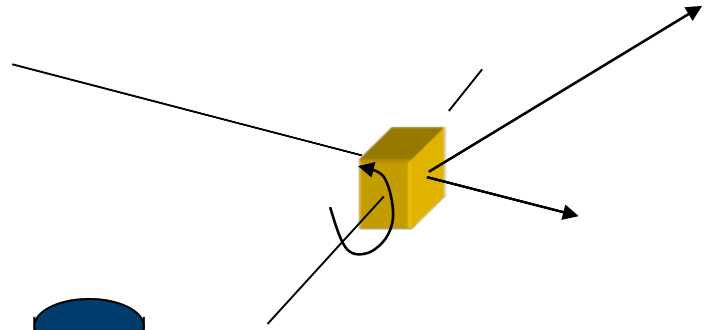




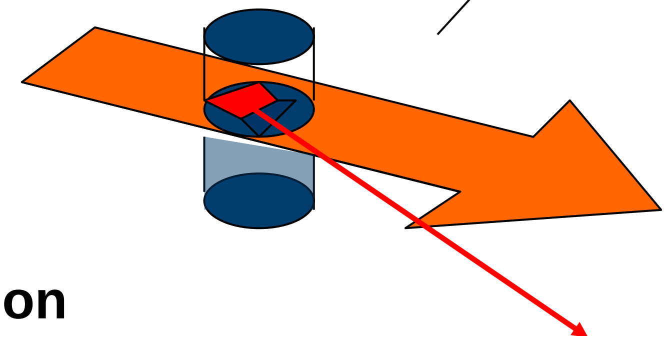
Osaka mirrors

Three directions in Microdiffraction

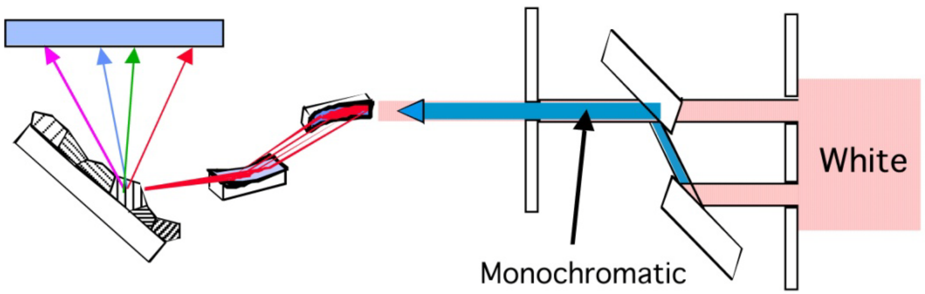
- **Monochromatic crystallography**
  - **best S/N for simple systems**



- **RISØ/ESRF 4DXRD**
  - **best depth probe, fast**



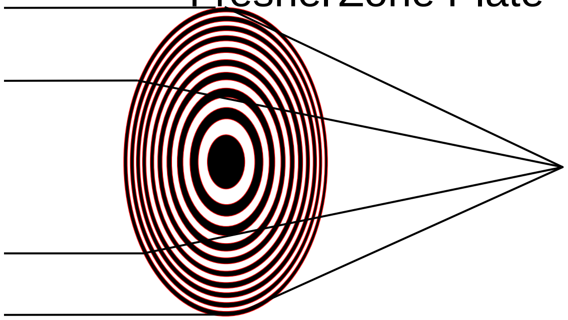
- **Polychromatic microdiffraction**
  - **Best spatial resolution**
  - **Deviatoric/absolute elastic+plastic strains**



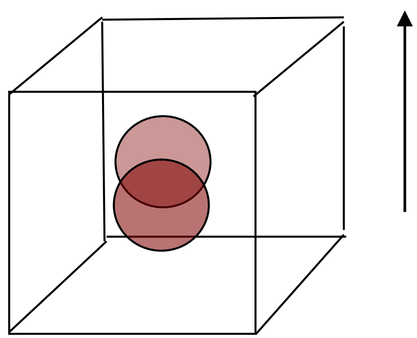
Coherent diffraction provides additional capabilities

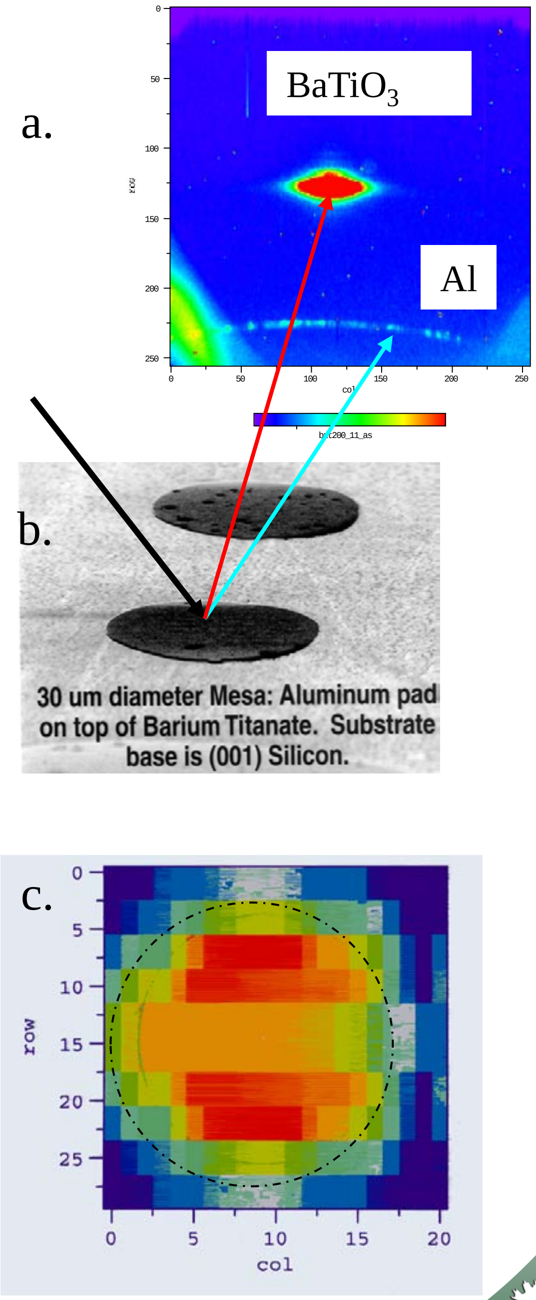
Monochromatic micro crystallography probes simple crystal systems

Fresnel Zone Plate



Wide-range of focusing choices

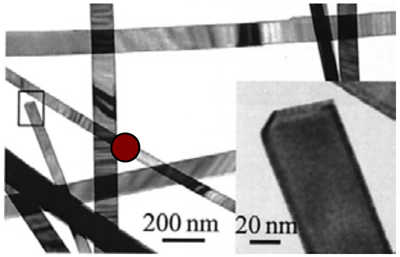




Ferroelectrics ideal samples

Thompson et al. Study domain growth/switching etc.

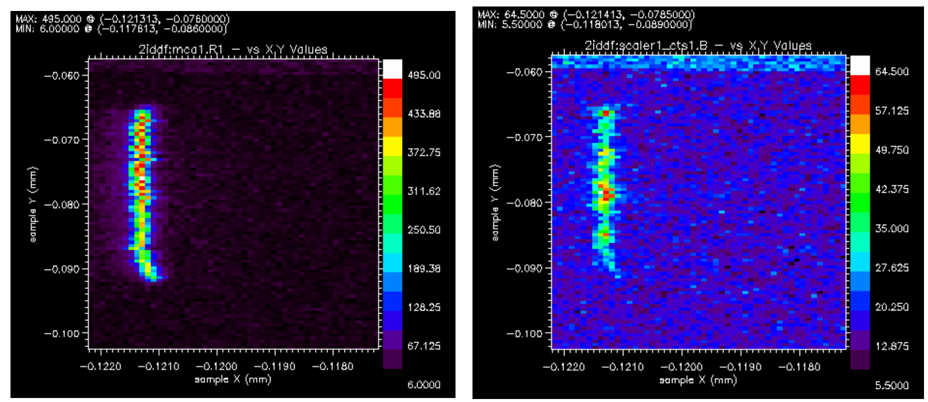
Cai et al. and others study ultra-small nanocrystalline volumes with existing microbeams

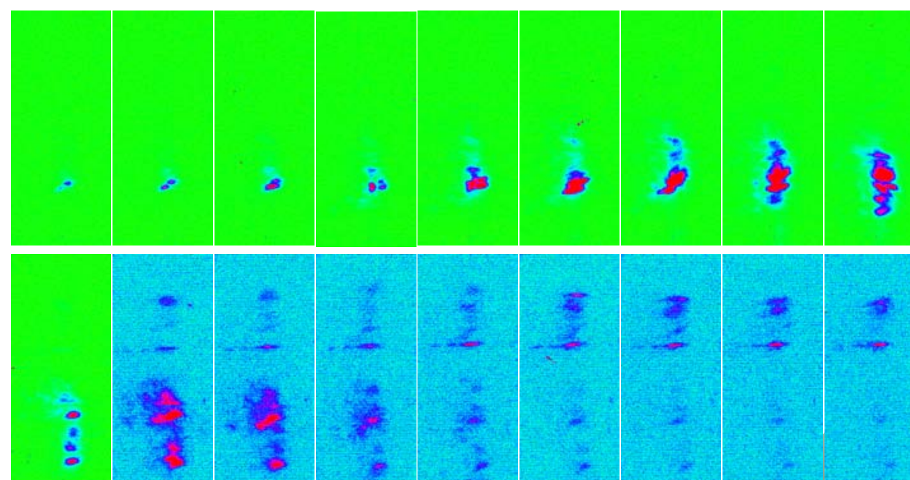


- 150 nm beam resolves crystalline substructure in individual Sn_2O_3 nanobelts

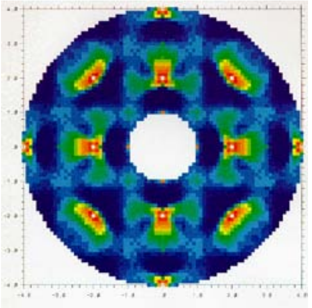
Fluorescence map

Diffraction map

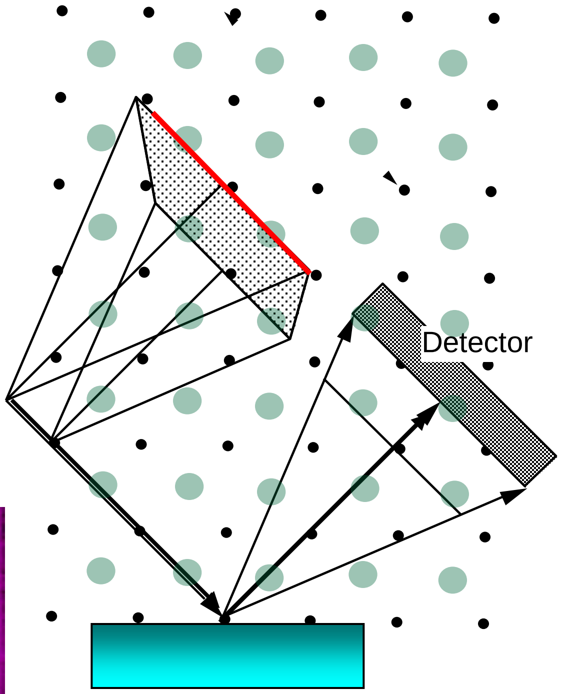


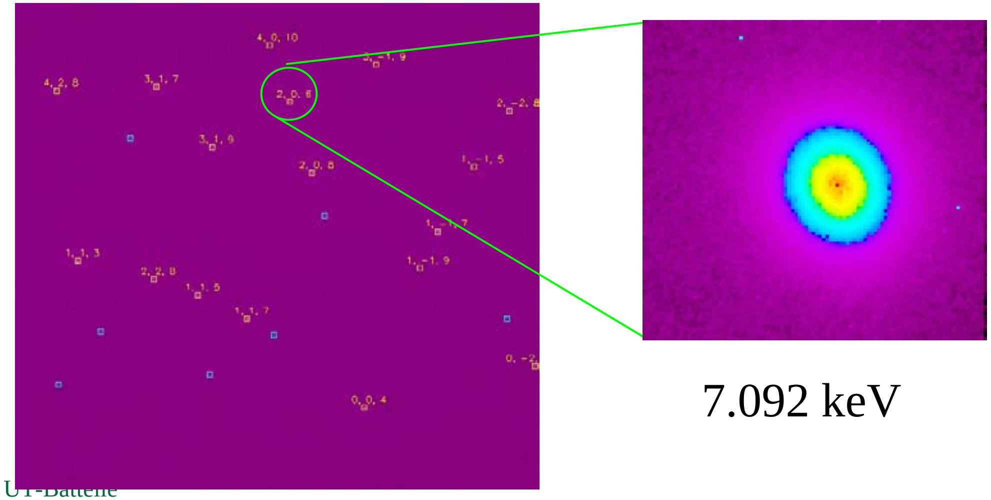


Short-range chemical order heterogeneity magnetically annealed crystal now accessible



- **6 orders of magnitude weaker than Bragg scattering**
- **Area detector integrates over plane in reciprocal space**





7.092 keV

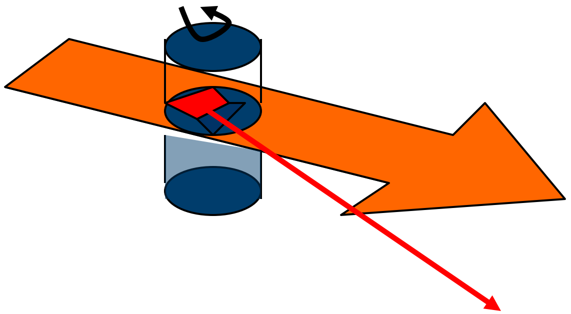
APS *Nanoprobe*- opens new opportunities for spatially resolved

- **Diffraction proposals compelling**
- **Physics of small**
- **Integrated circuit materials**

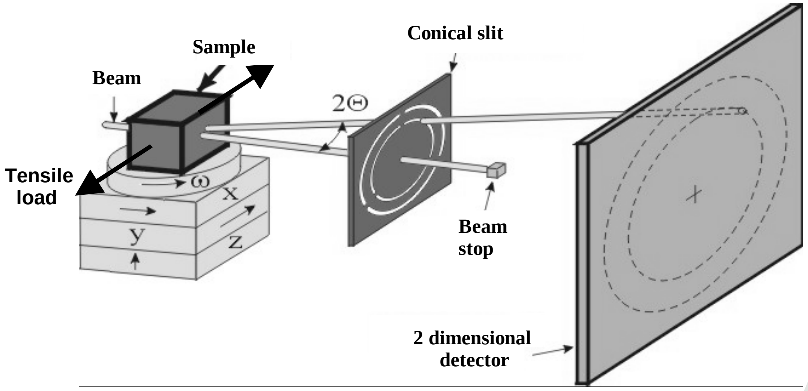
~30 nm target/ <10 nm possible in near future/ NSLSII ~1 nm!

4DXRD Microscope emerging tool for studying mesoscale dynamics-single rotations

- **Singly focused monobeam illuminates numerous grains**
  - **Bragg condition satisfied by single rotation**
  - ***Time resolution! (4D)***



- **Grain outline determined**
  - **Ray tracing**
  - **conical slit**
  - **Back-projection tomography**



- **E>50 keV allows deep measurements**

Best with high-energy beams/Beamline 1 at APS

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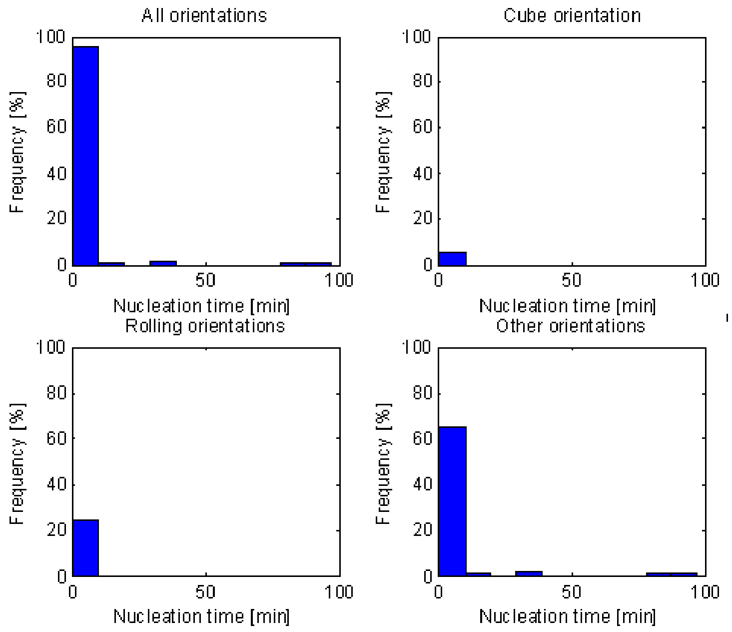
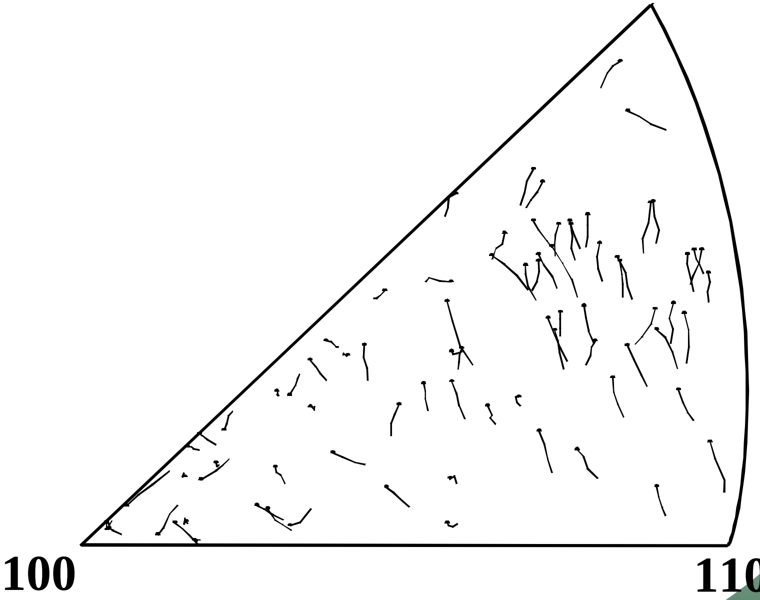
4DXRD Microscope powerful dynamics probe



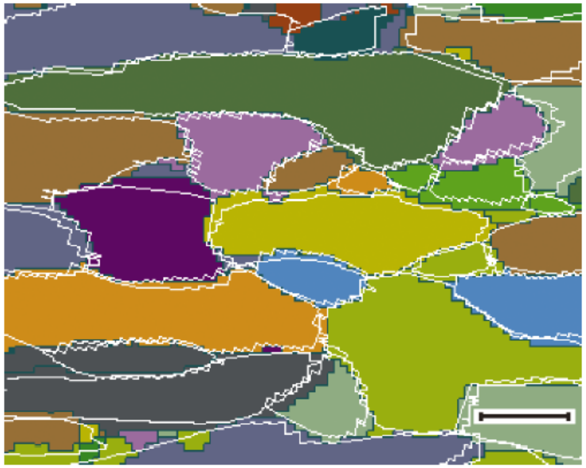
- **Recrystallization growth individual grains-deep**
  - **E. M. Lauridsen, D. Juul Jensen, U. Lienert and H.F. Poulsen (2000). *Scripta Mater., 43, 561-566***

- **Rotations/texture evolution individual grains during deformation**
  - **Tests deformation models**
  - **L. Margulies, G. Winther and H.F. Poulsen, Science** 291**, 2392-2394 (2001).**



4DXRD Microscope provides additional powerful capabilities

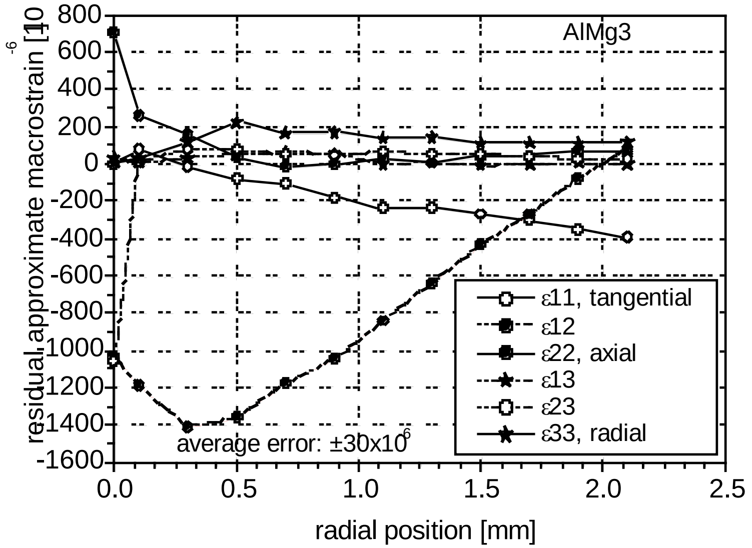
- **Grain boundary mapping in coarse grained materials-5μm**
  - **Poulsen et al. J. Appl. Cryst. 34 751-756 (2001)**



- **Single crystal refinement for polycrystals**

- **Macro/microstrain**

Ideal for neutrons! But needs high-resolution detectors!



Strain tensor elements in torsion sample

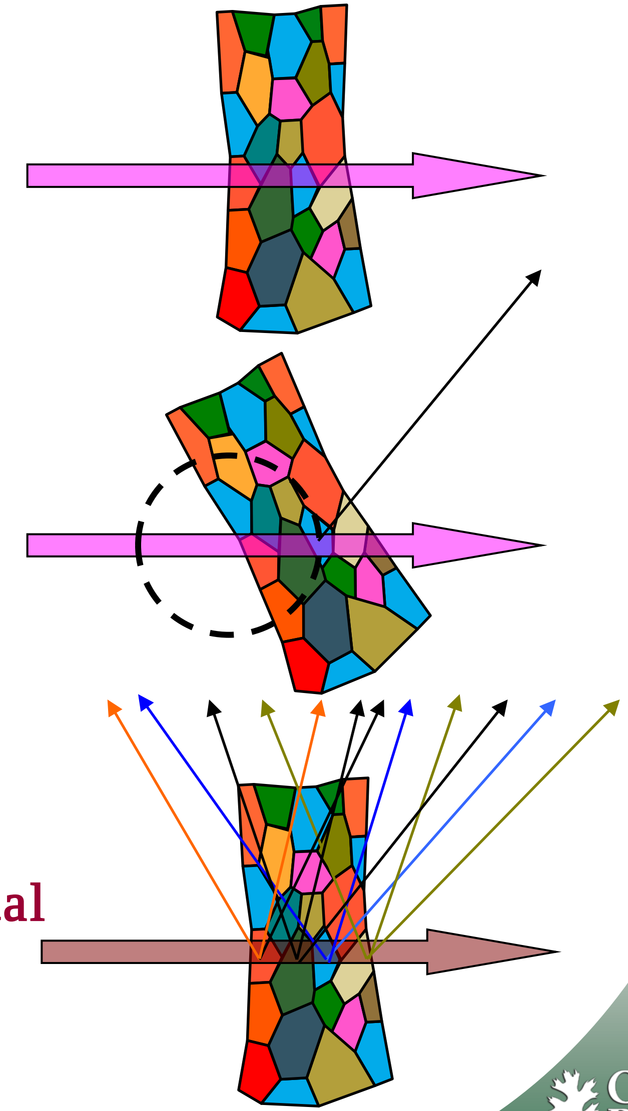
Polychromatic simplifies microdiffraction

Solves intrinsic problem with conventional microdiffraction-

-Sample does not need to be rotated!

Special software required- Can index polycrystalline samples

3D nondestructive probe of stress/strain/crystal structure!



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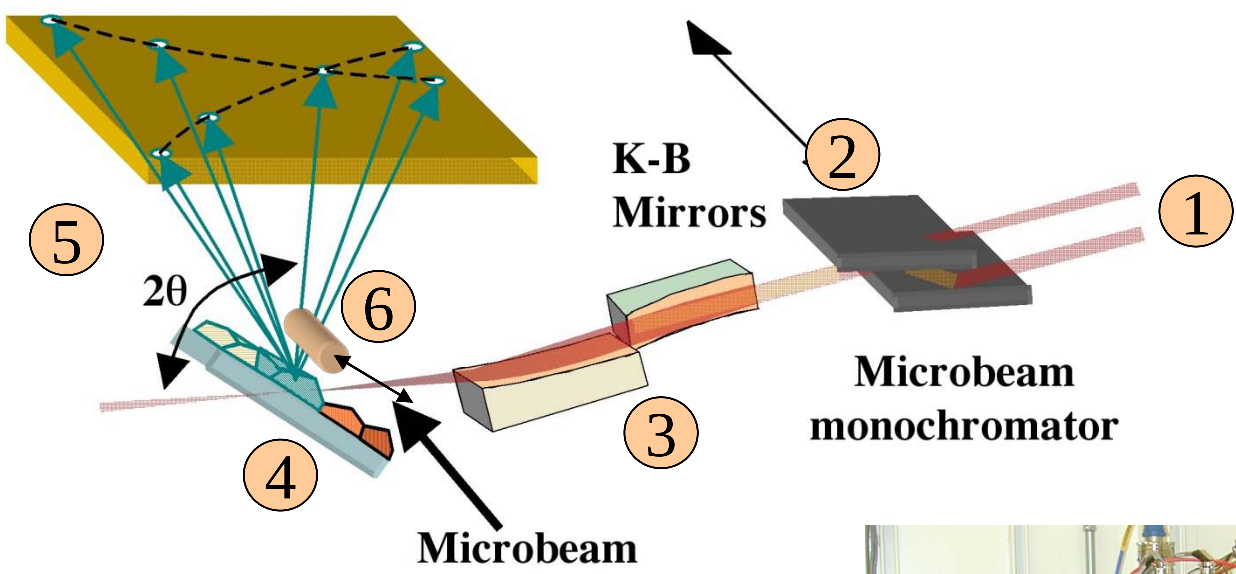
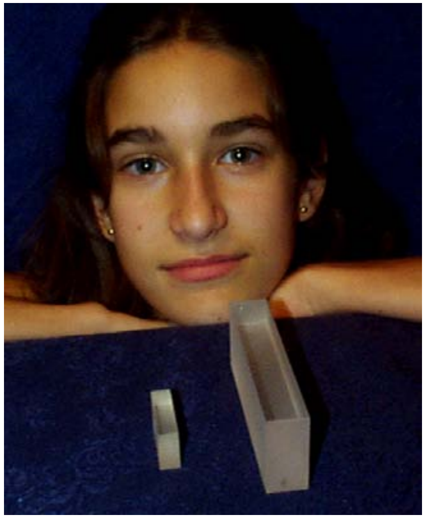
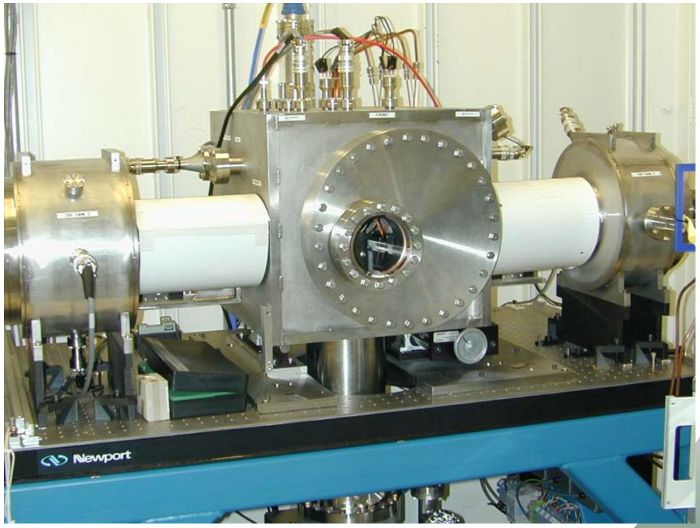
3-D X-ray Crystal Microscope has 6 key Elements

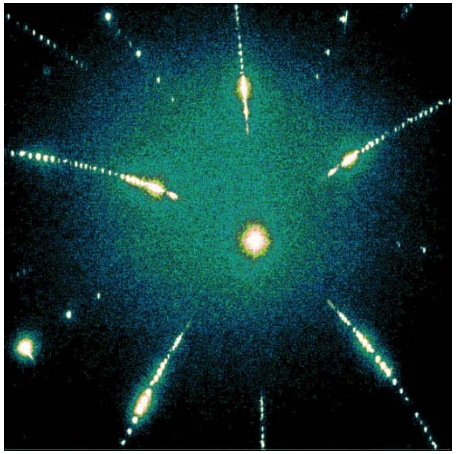




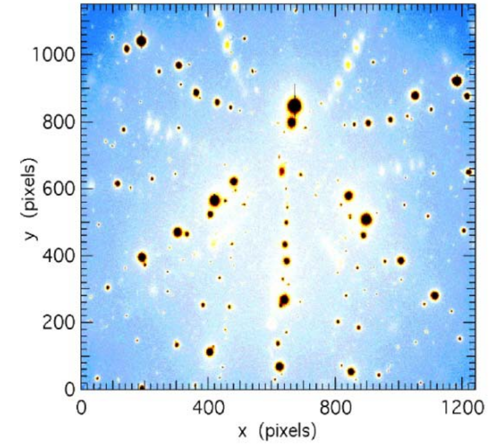


<0.3 x 0.3 x 0.5 μm^3
strain~10^{-4}-10^{-5}

MicroLaue patterns depend on internal structure/orientation

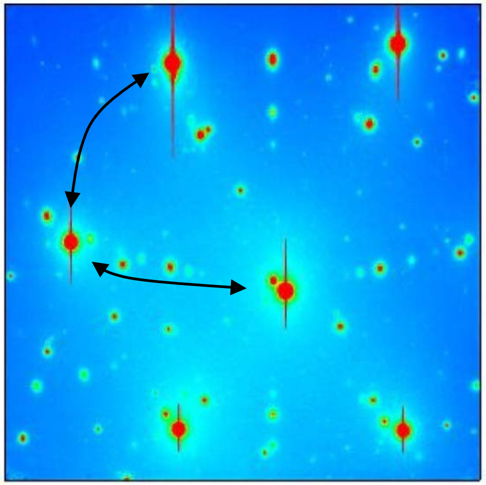


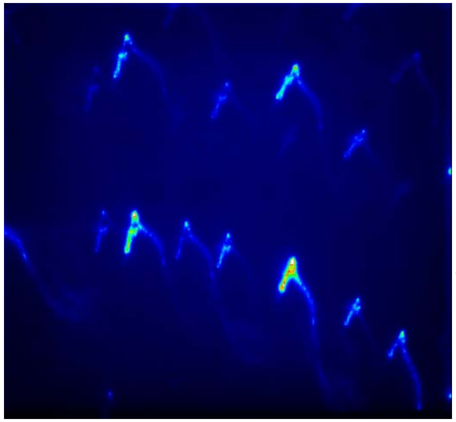
Phase/phase boundaries



Texture (0.01º)/grain boundaries

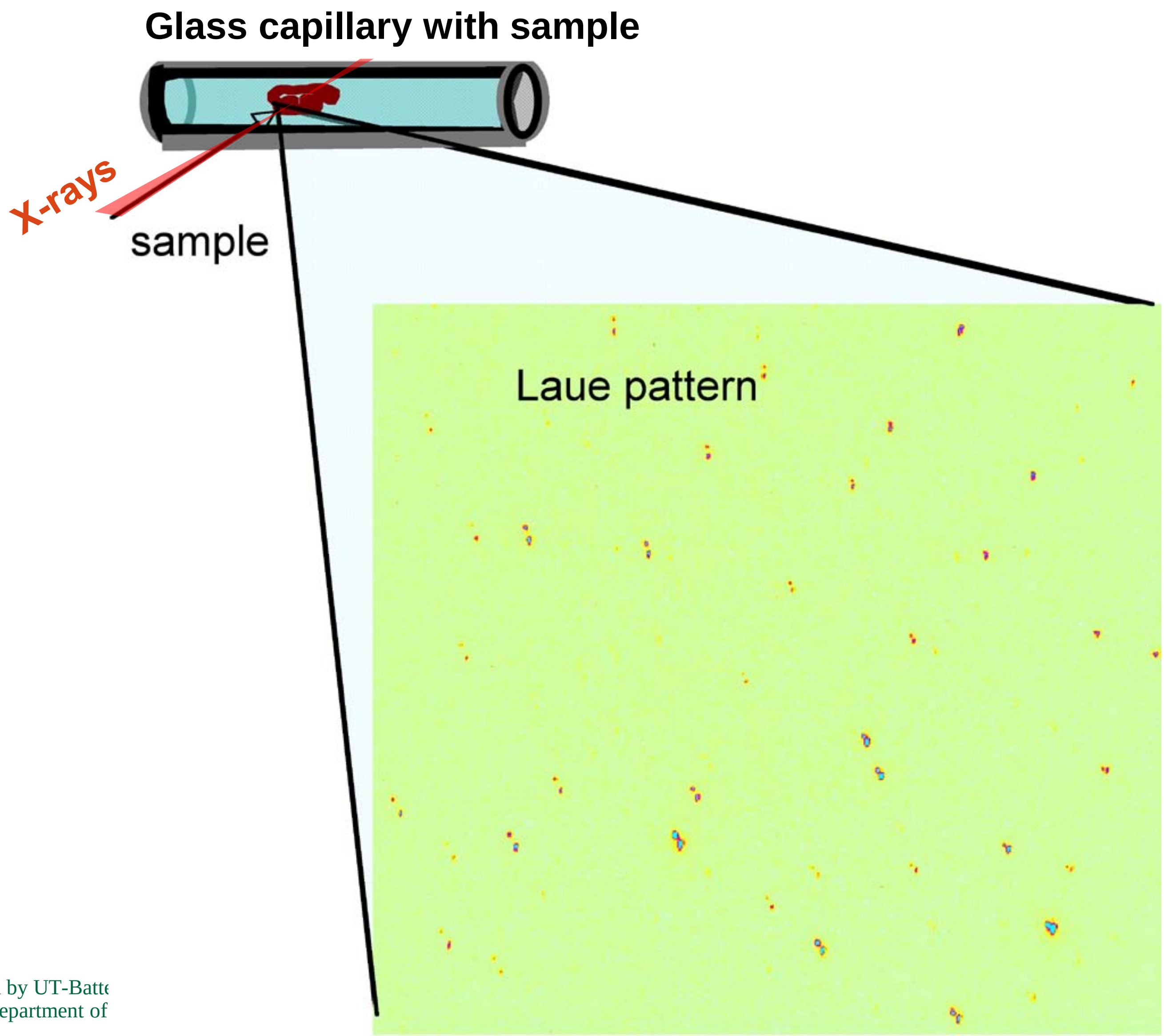
Elastic strain tensor(1 x 10^{-4})





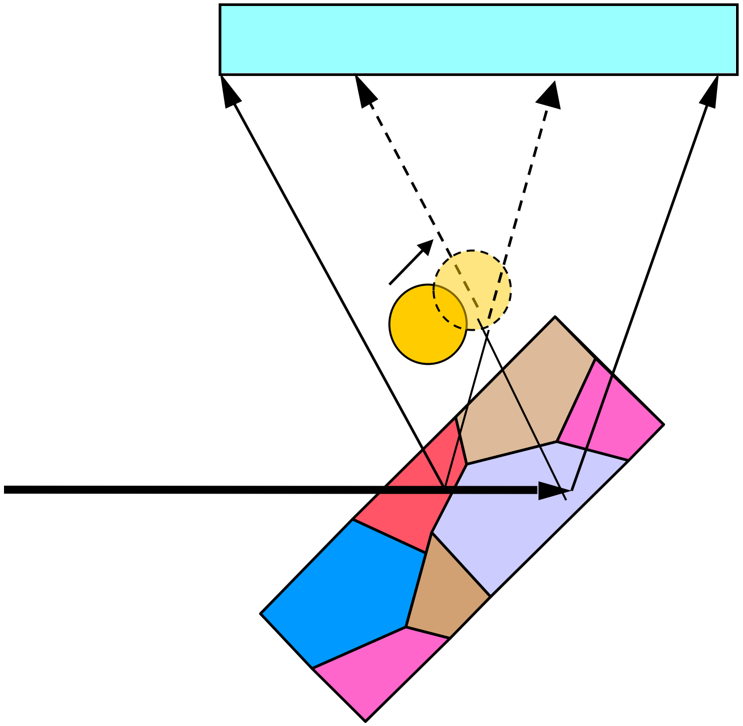
Dislocation Tensor

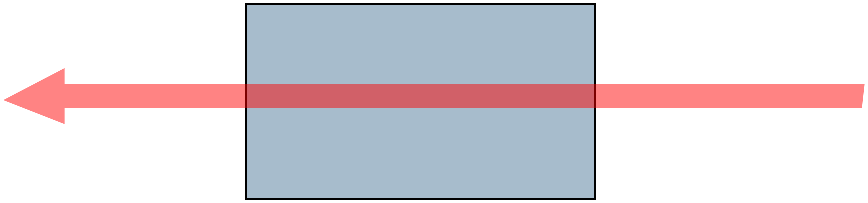
Laue methods essential for some samples



Differential aperture microscopy resolves submicron along incident beam!

- Simplifies data interpretation
- Submicron Z resolution
- Isolates weak diffraction from strong
- First demonstration by Larson et al. on deformed Cu -





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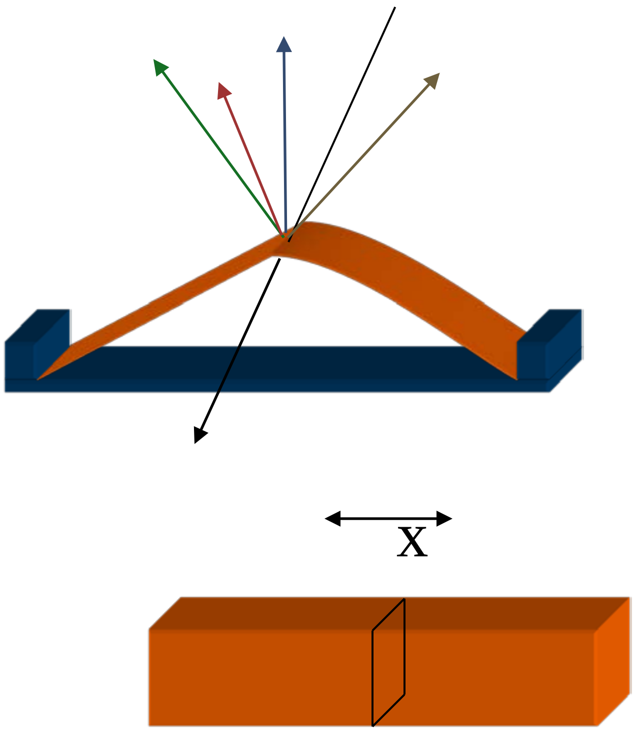
As wire moves its edge cuts through Laue spots

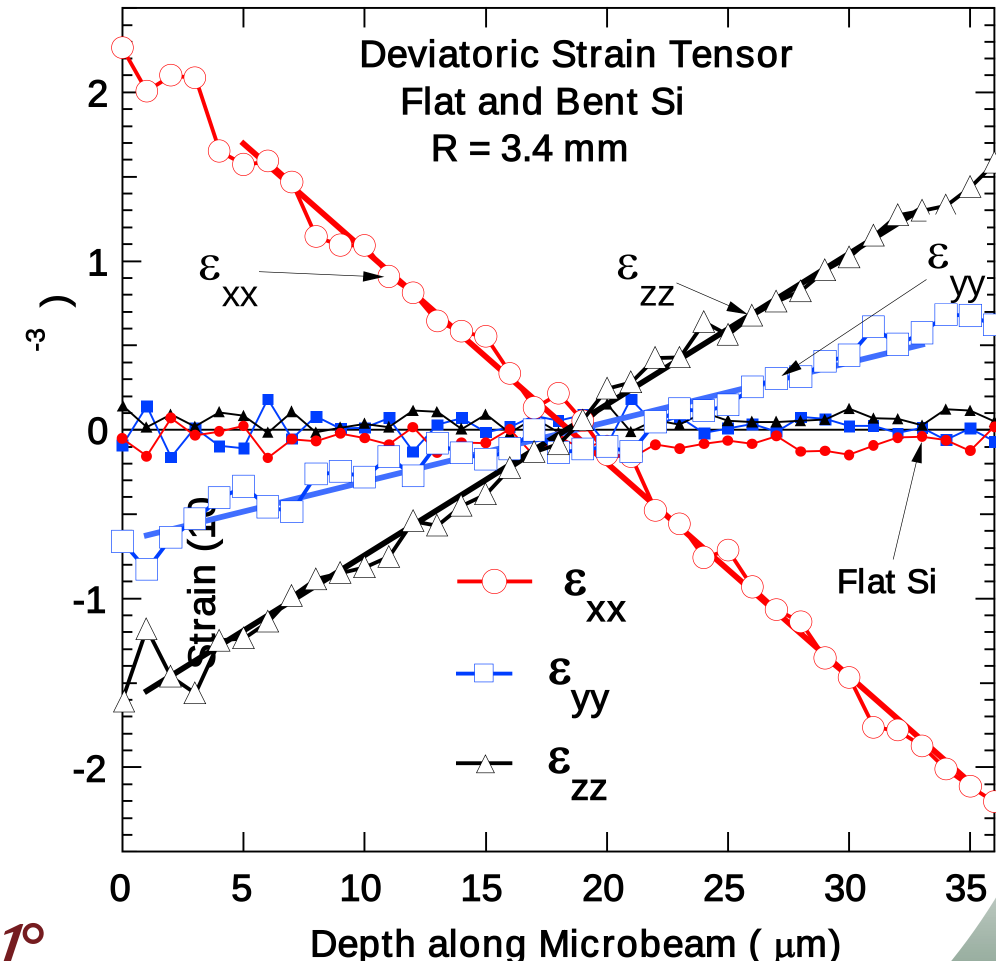
- **Near-surface fluorescence provides moving shadow**

- **Long scans needed for deep penetration**

QuickTime™ and a
Video decompressor
are needed to see this picture.

Precision measurements of strain tensor *inside* bent single crystal Si illustrate power of DAXM

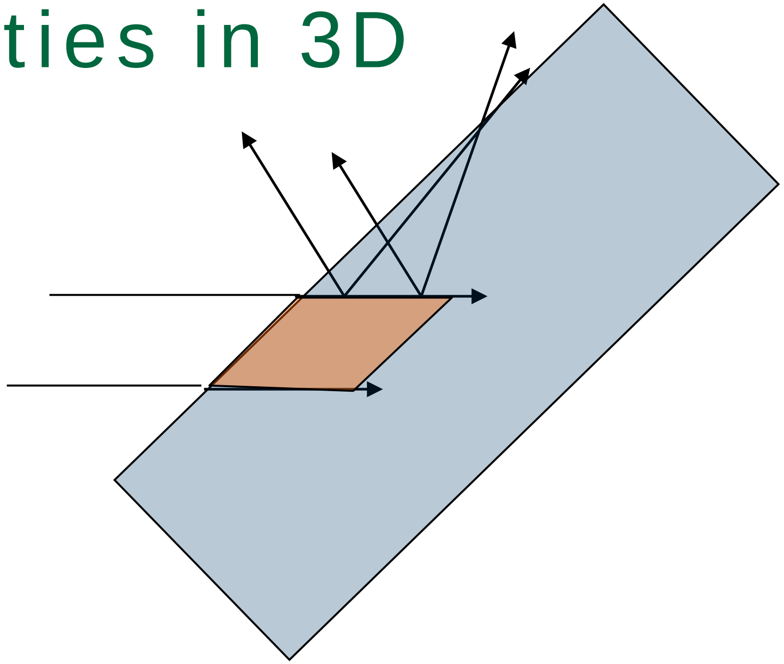


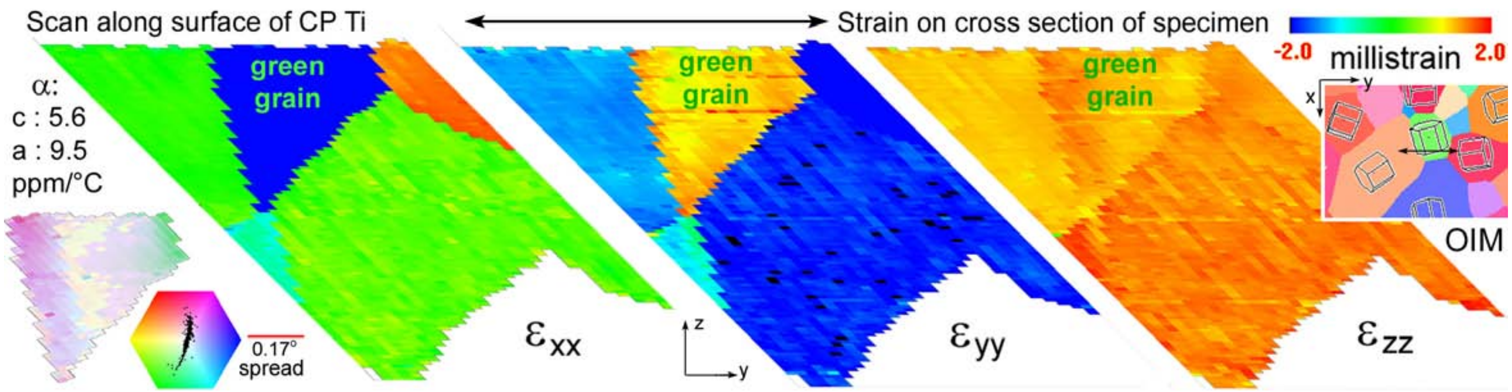


Orientations to 0.001°

Maps crystal properties in 3D

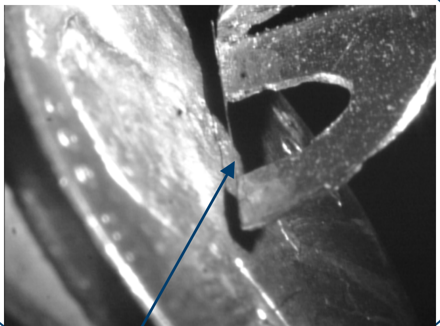
- **Phase**
- **Texture (orientation)**
- **Elastic strain tensor**
- **Nye tensor (deformation)**





T. Bieler et al.

Experimental Hutch 34ID-E at UNICAT, Advance Photon Source

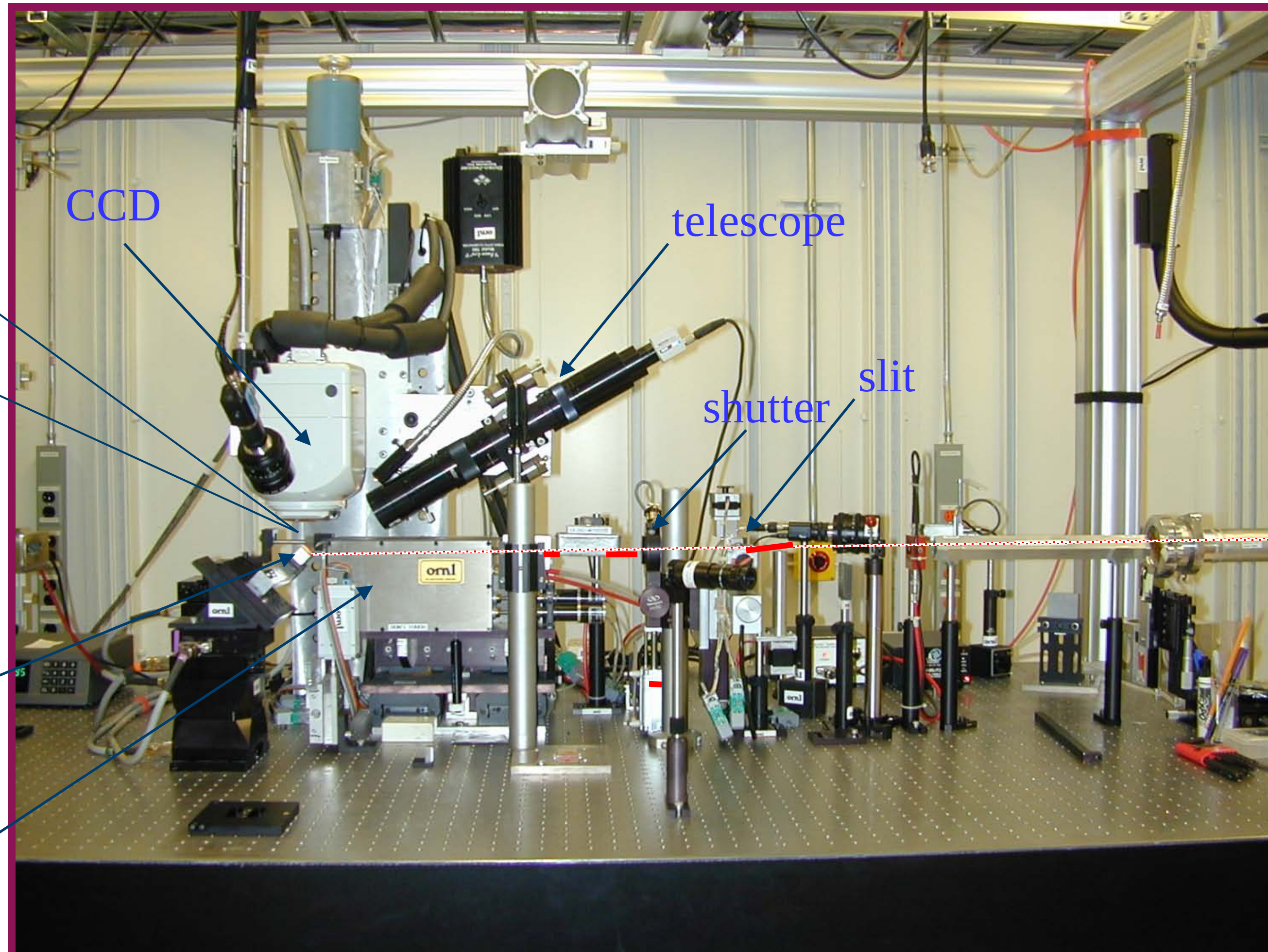


differential aperture

(wire scan, ~ 200 µm above sample surface)

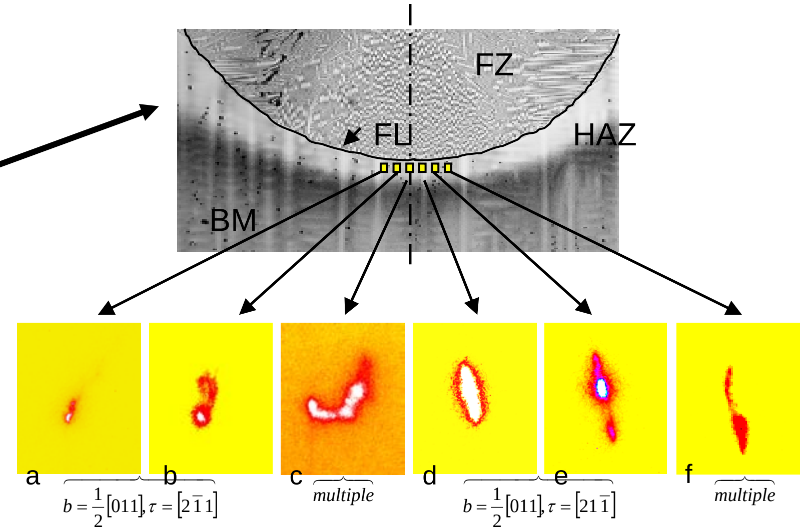
Sample stage

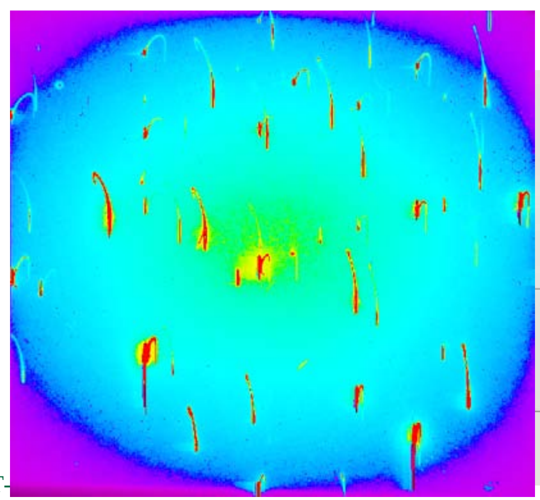
K-B Focusing mirrors

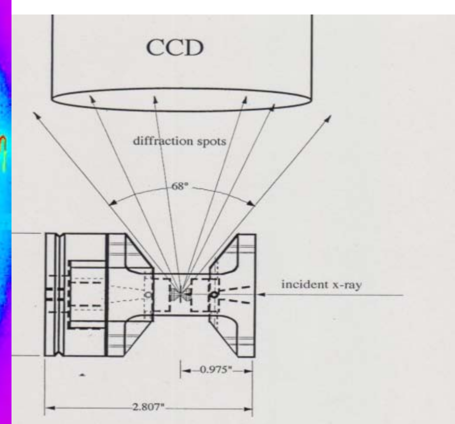


Ongoing research too extensive to cover

- **Fracture/stress localization in thin films**
- **Residual stresses/ deformation/ grain boundary network near welds**
- **Complex phase patterned materials**
- **Extreme environmental chambers**

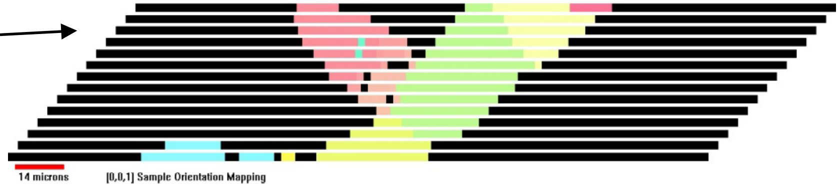




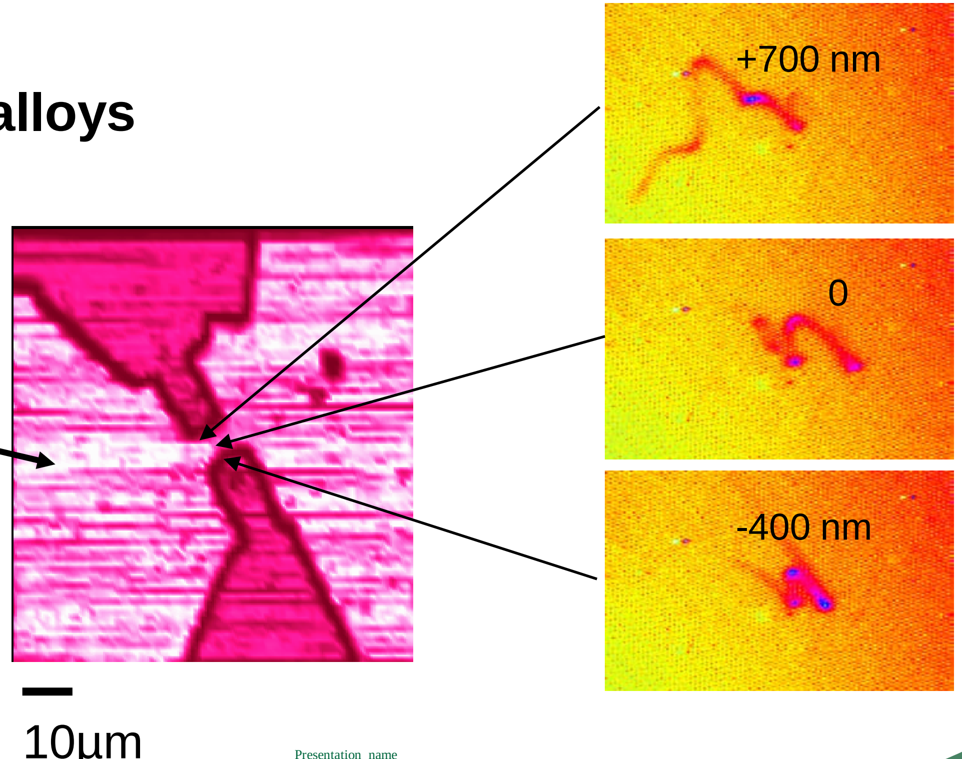


Ongoing too extensive continued..

- **Domain wall structure measurements**
- **Sn whisker growth**



- **High-performance alloys**
- **Nanomaterials**



Energy scans allow structure determination

- **Generalization of orientation software can identify phases**

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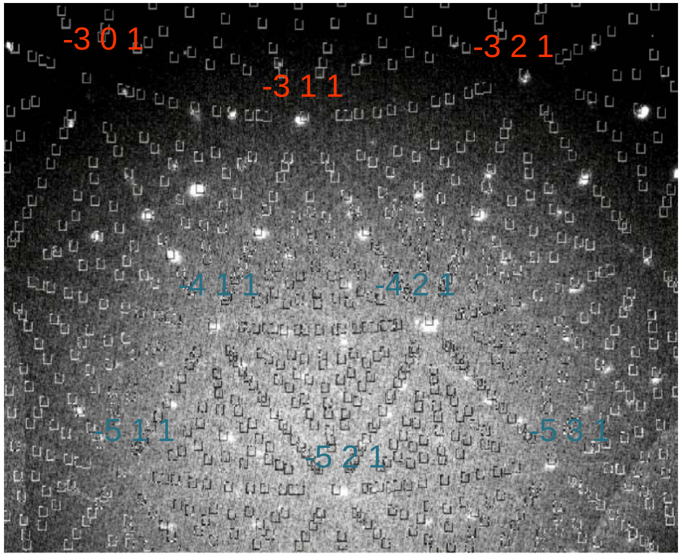
- **Energy scans provide integrated reflectivities.**

BAM braze $Pd_{40}Cu_{30}Ni_{10}P_{20}$

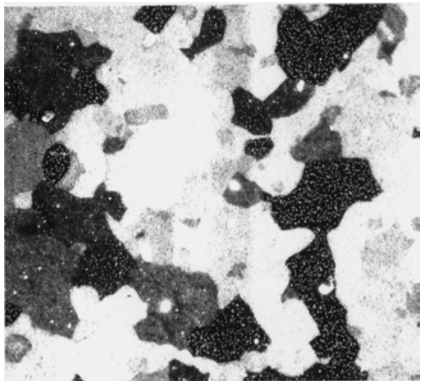
- **Identified two minor crystal phases tetragonal/hexagonal**



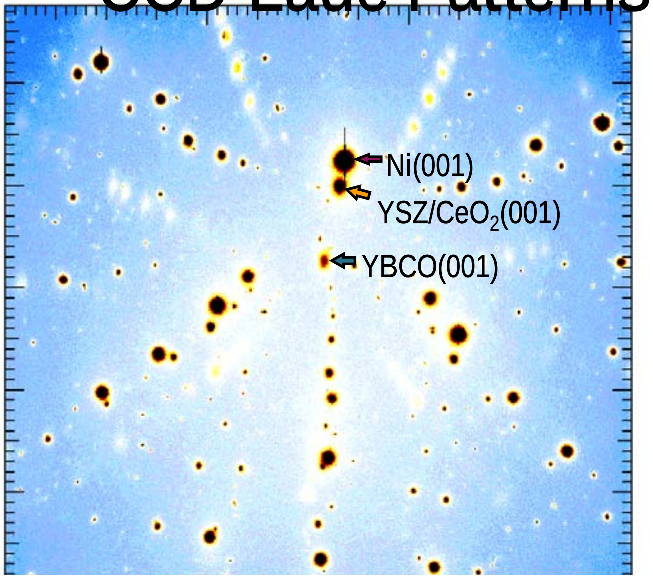
Cannot be found by powder

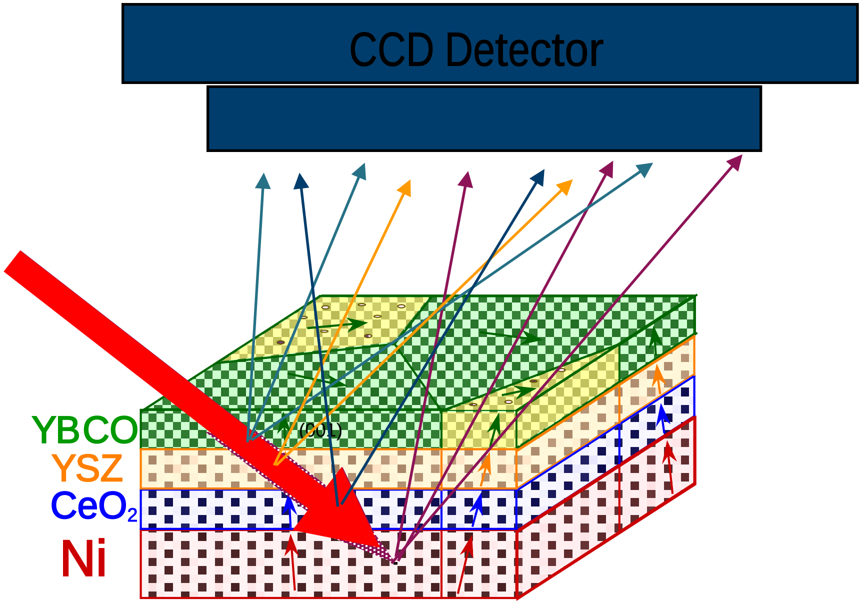
Grain-growth/ Budai et al. characterized epitaxial growth RABiTS

Optical: ~50μm grains



CCD Laue Patterns





CeO_2 Observation:

Exact epitaxy for growth at low T; lattice tilts at high T

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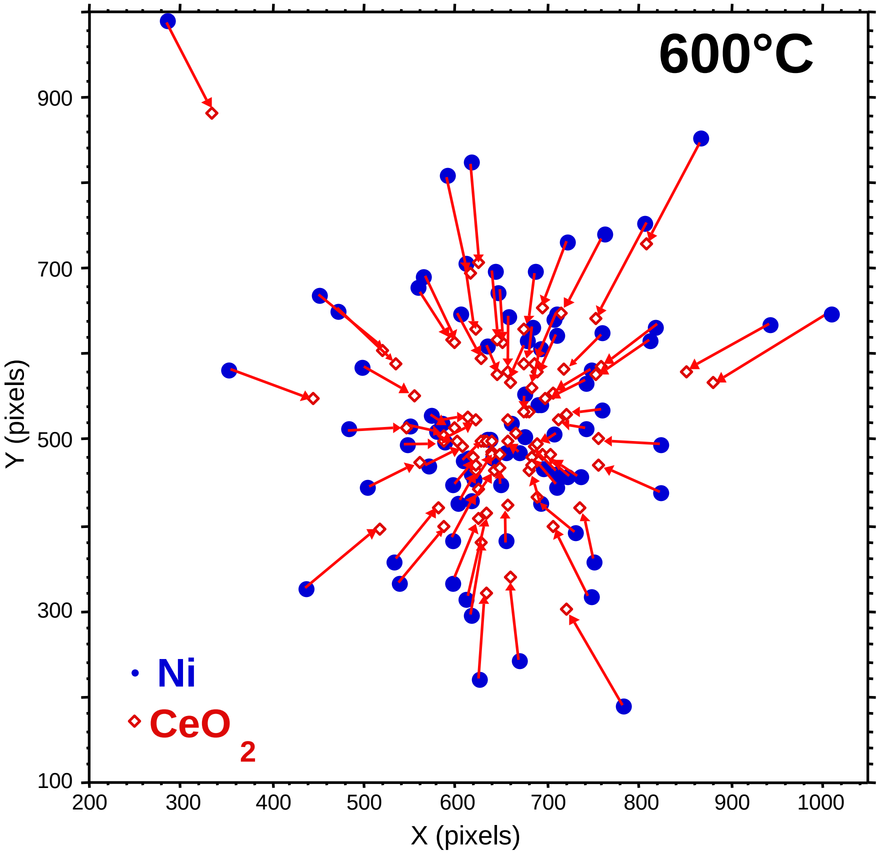
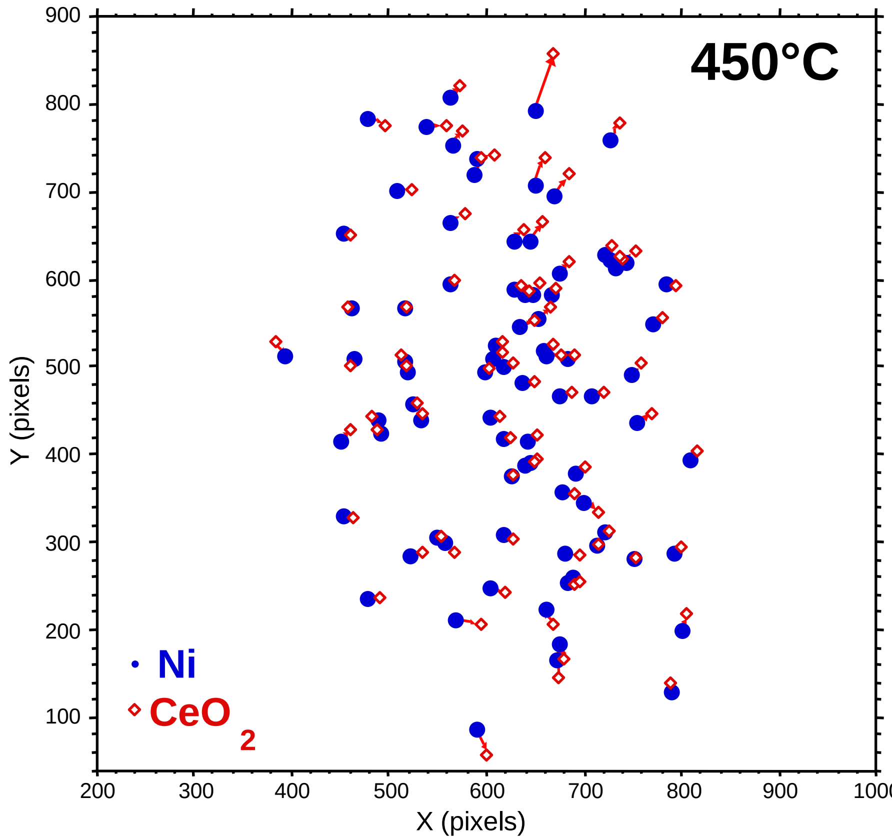
Relative CeO_2 orientation depends deposition temperature





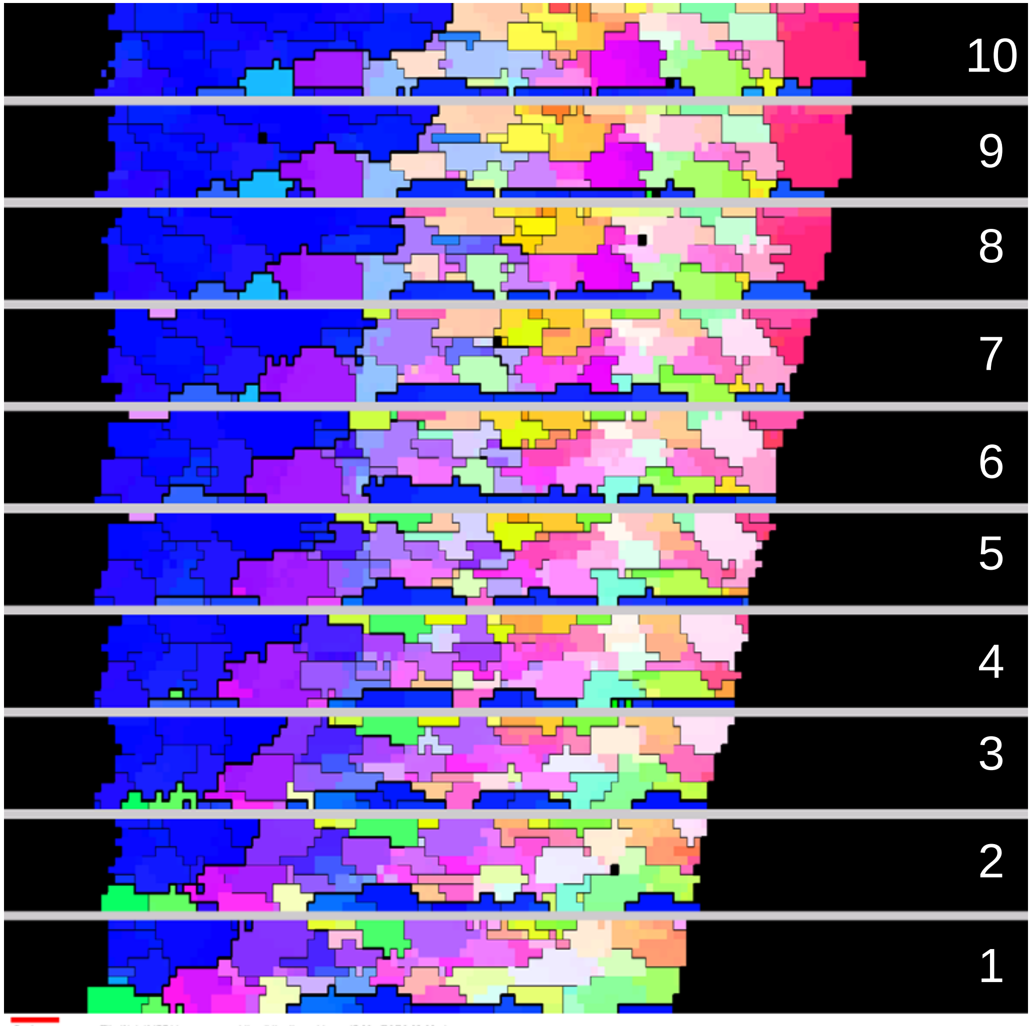
<u>Step edge growth- good</u>:

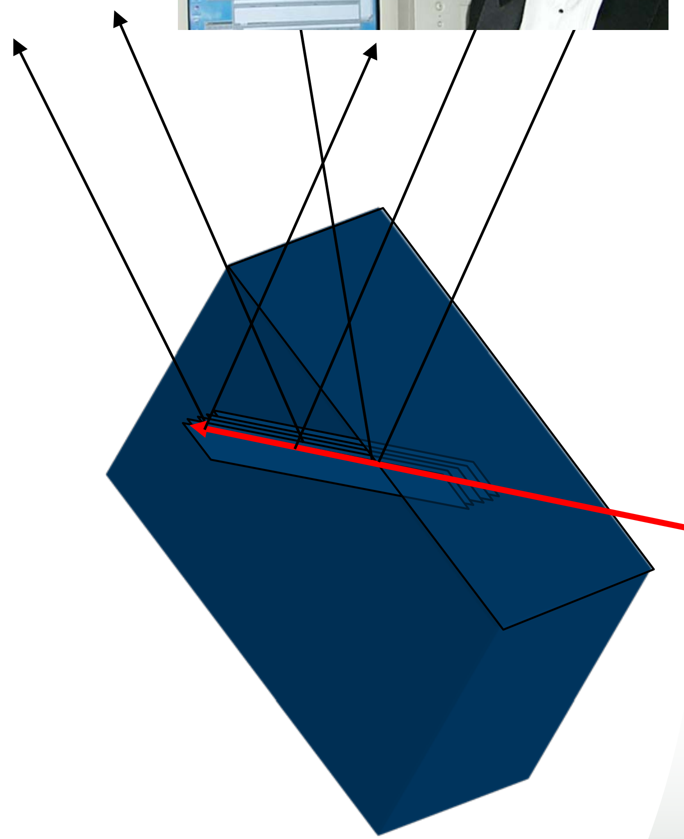
Crystallographic tilt towards ⊥

Tilt increases monotonically with miscut

<u>Island growth-bad</u>:

In-situ observations of 3D Grain Growth

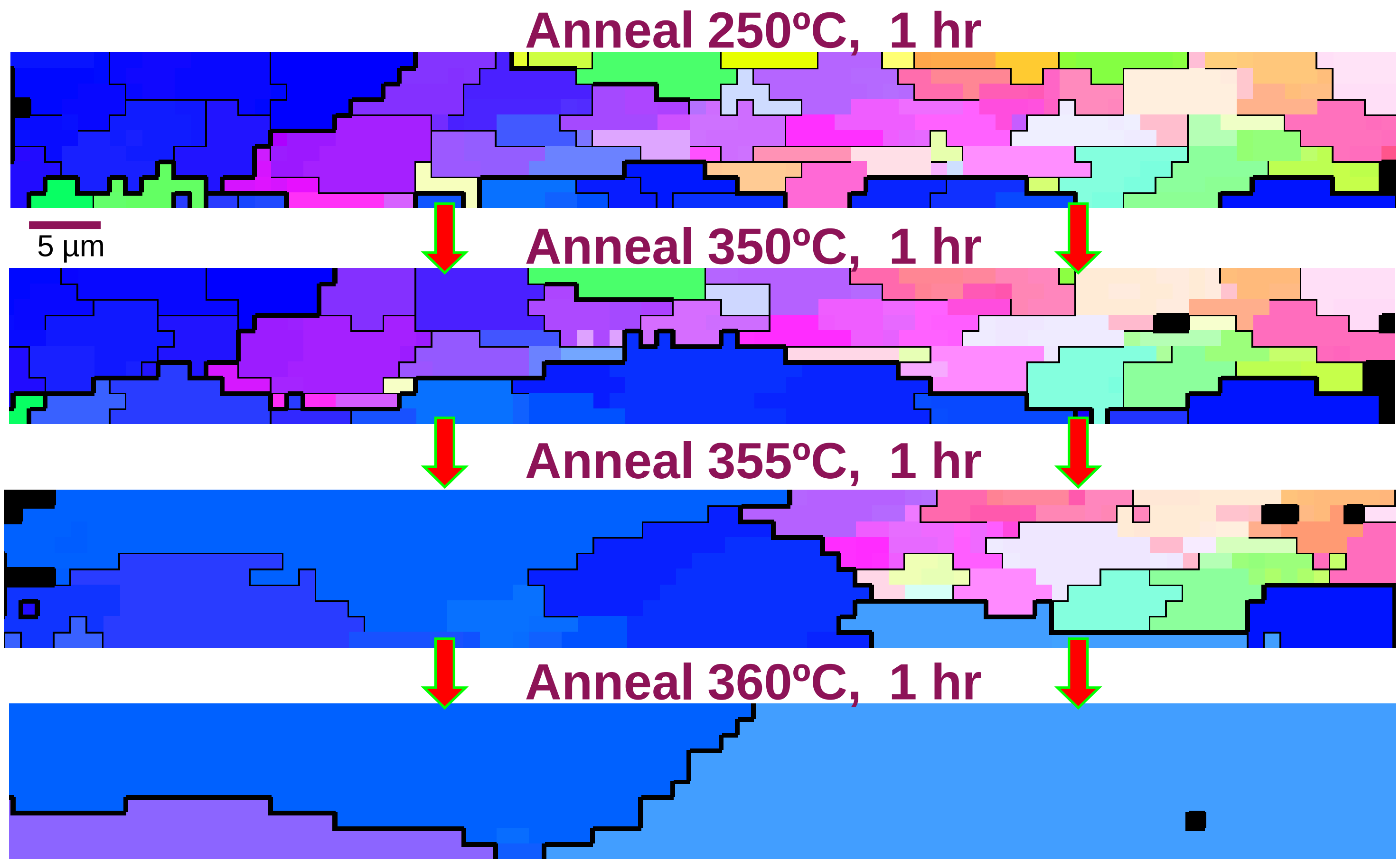




Hot-rolled (200°C
1xxx Al(~1%Fe,Si)
Alcoa Polycrystal

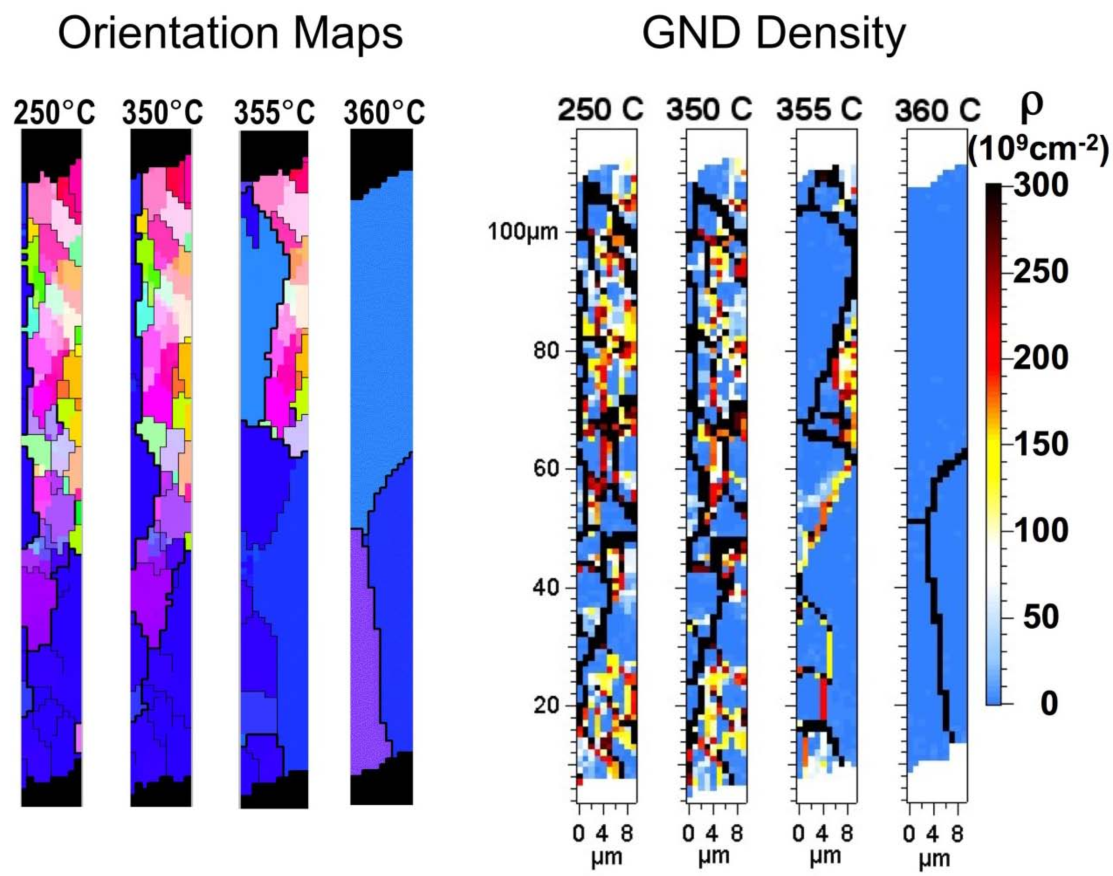
Thermal Grain Growth in Hot-Rolled Aluminum

1 µm pixels,   Boundaries: 5° & 20°



- **GB motions include both high-angle and low-angle boundaries**
- **Complete and detailed 3D evolution needed for validation of theories.**

Thermal Grain-Growth And Microstructure Refinement in Polycrystalline Al



- 3D X-ray Microscopy Measurements of Dislocation Density Finds Microstructure Refinement to Be Important

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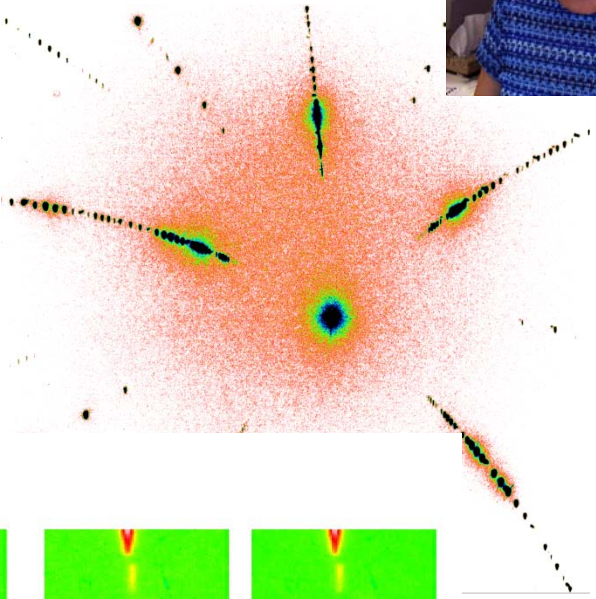
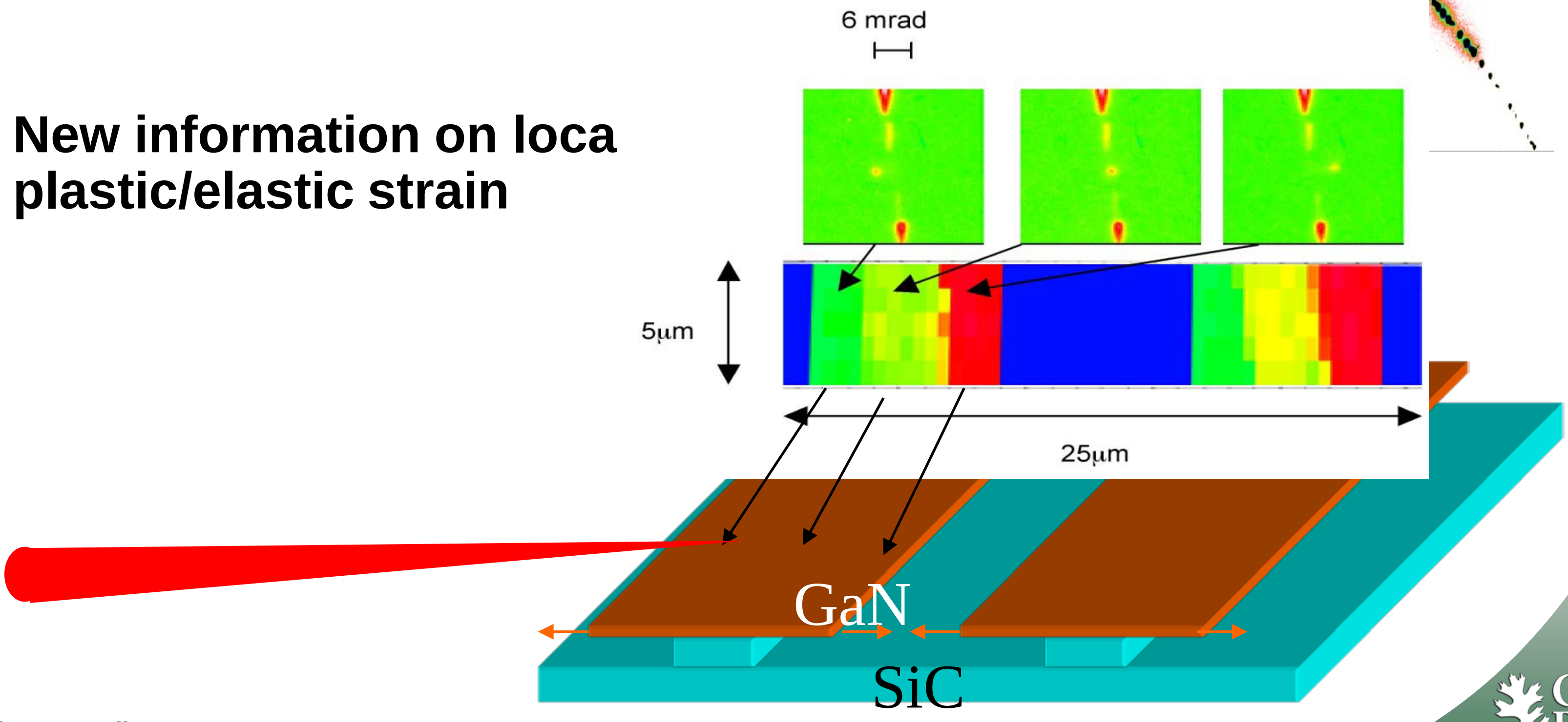
Local orientation and plastic/elastic deformation mapped in pendeoepitaxially grown GaN



- **Reduced dislocations**
- **Direct confirmation of tilted wing structures**
- **New information on loca plastic/elastic strain**



Spatial resolution critical photovoltaics

- **Chemical/ structural defects important**
- **Surfaces/Interfaces critical**
- **Novel materials depend on dimensionality**

QuickTime™ and a
TIFF (Uncompressed) decompressor
are needed to see this picture.

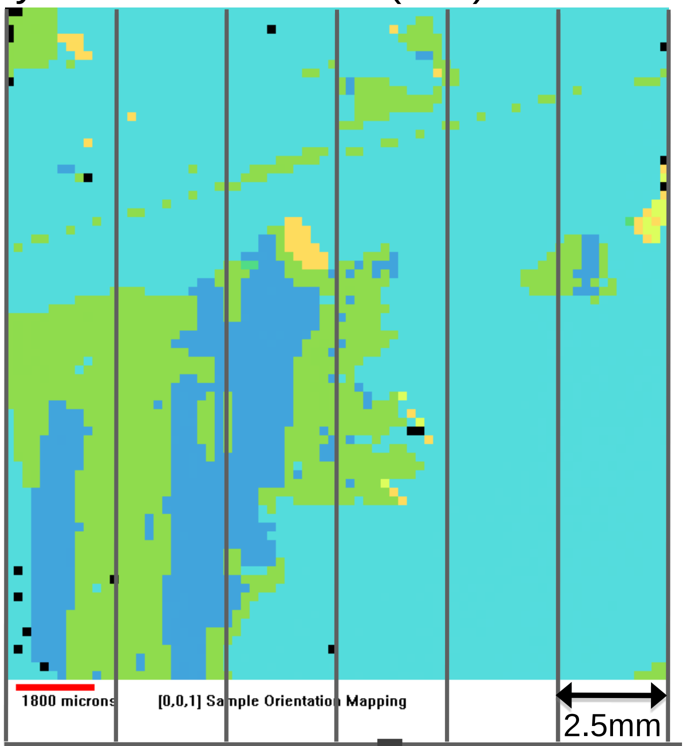
McHugo et al. ALS 1997
Chemical defect influence on Solar Cells

“Semicrystalline” Silicon for Cost-Effective Solar Cells

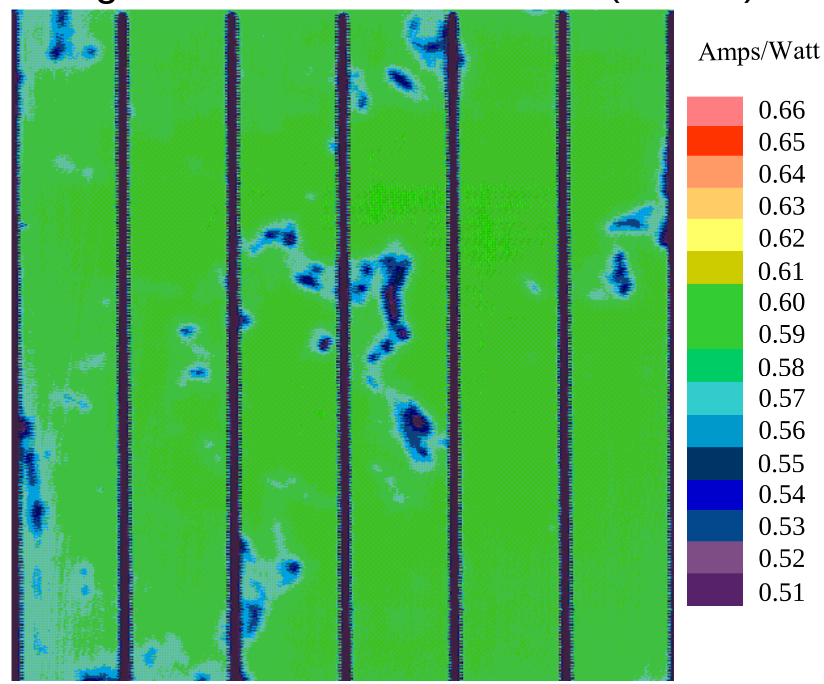
Collaboration between ORNL and Georgia Inst. of Technology

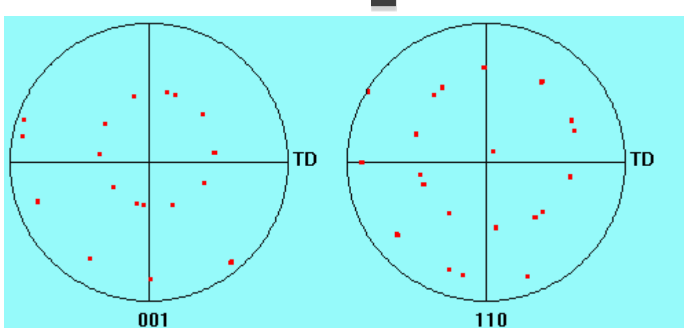
Correlate photovoltaic efficiency with local orientation and microstructure

X-ray Microdiffraction Si(001) Orientation



Light Beam Induced Current (980 nm)



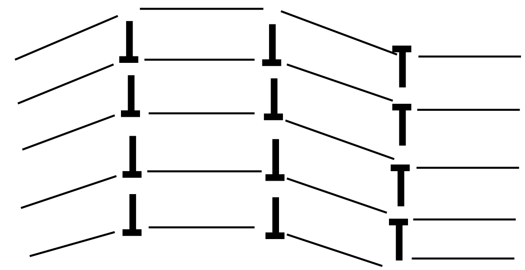


- Pole figures: Strong (110) fiber texture
- Goal: Understand and control structural origins of PV efficiency.

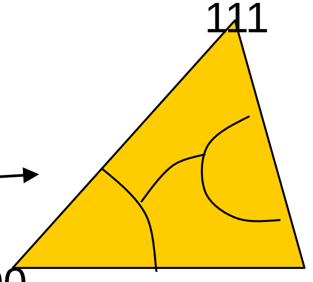
Deformation complicated

- **Polycrystals-grain boundaries/ inhomogeneous elastic parameters**

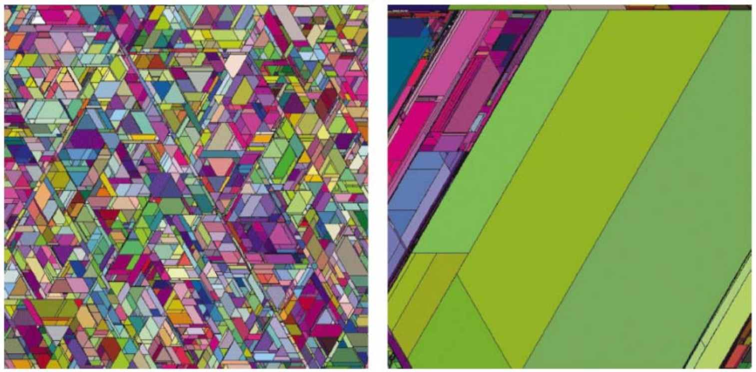
$$\langle |y(x) - y(x+L)| \rangle \propto L^H$$



- **Single crystals- Dislocation organize with universal scaling tominimize energy**



- **Deformation sensitive to crystallography**

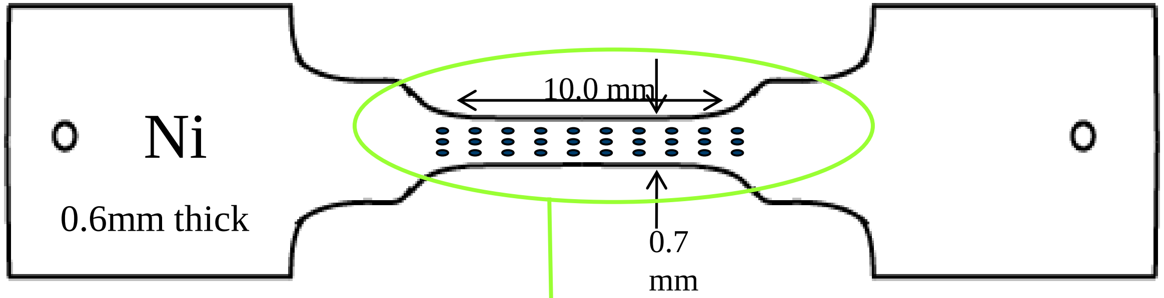


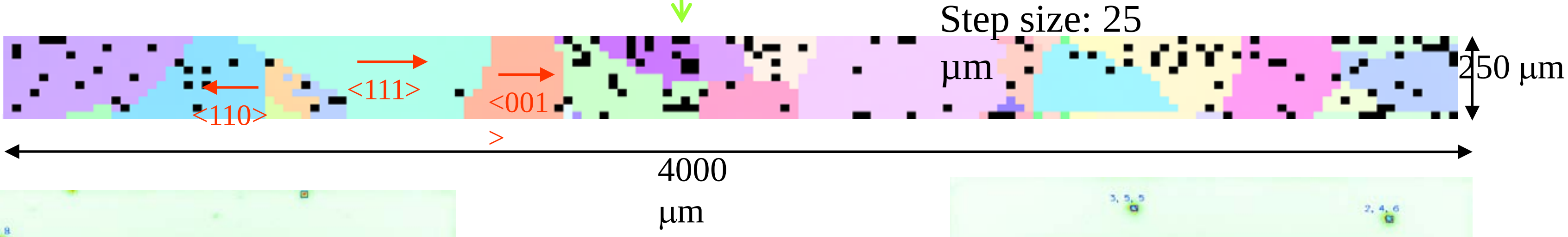
- **What controls spacing?**

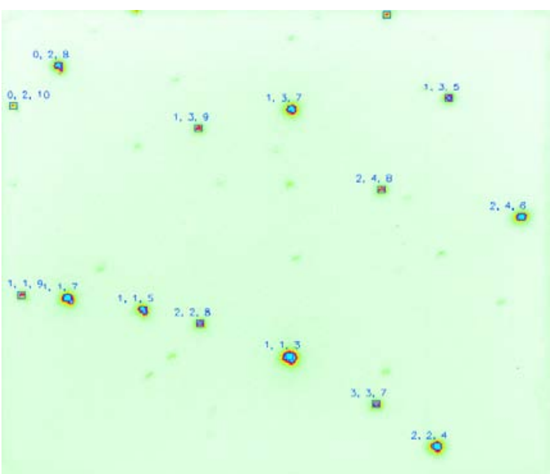
- **Surface morphology measurements (secondary effect) show self affine (fractal behavior) over four orders of magnitude!**
  - **Zaiser et al. Phys. Rev. Lett. (2004)**

Surface or bulk effect?

Deformation in polycrystals illustrates grain boundary behavior

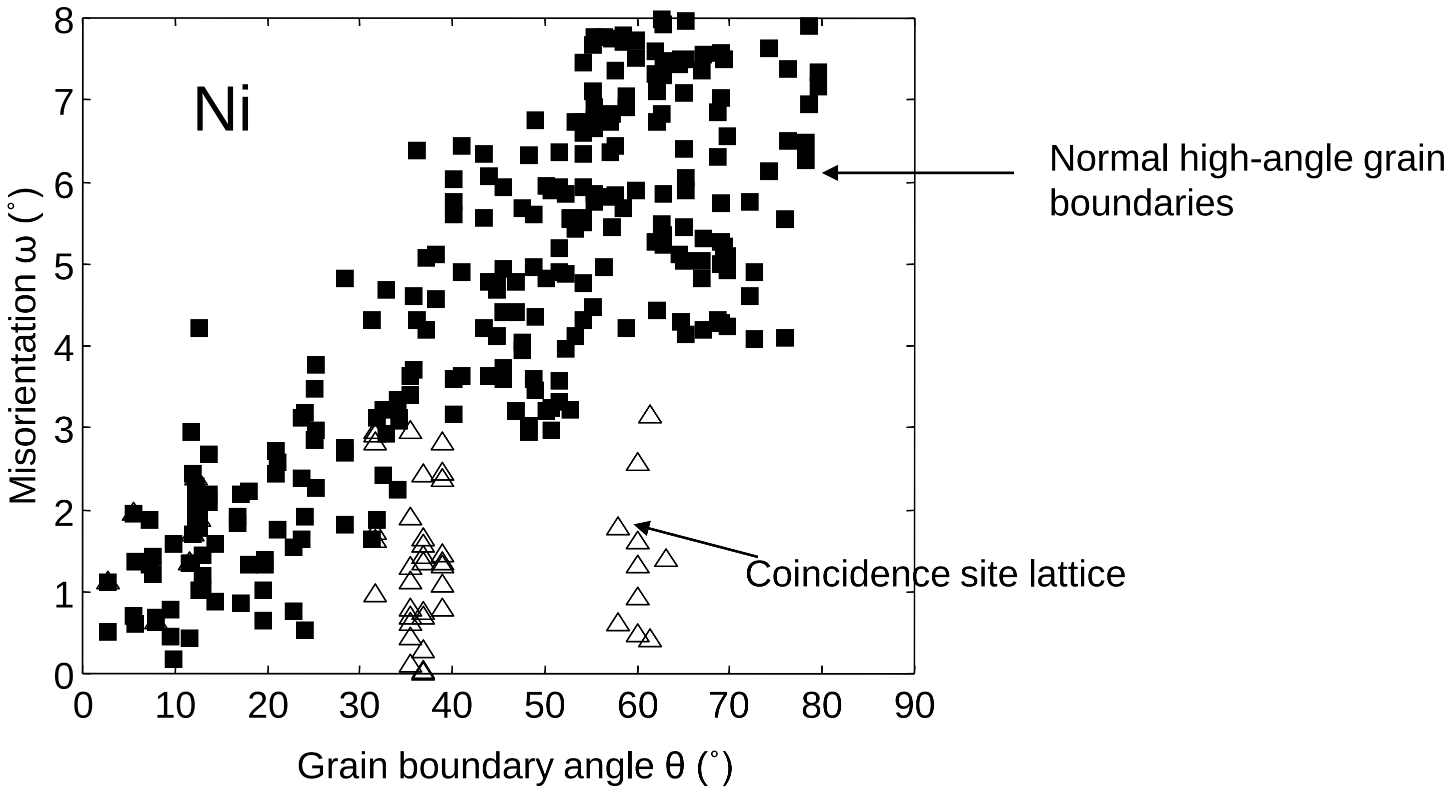




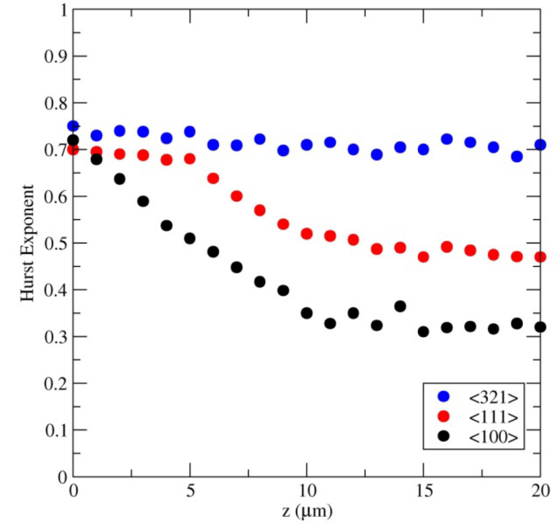


Indexation

Deformation induced rotations across grain boundaries sensitive to boundary type

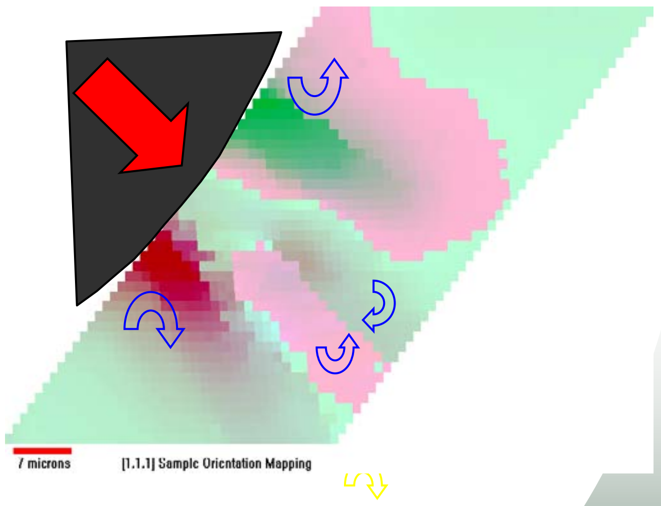


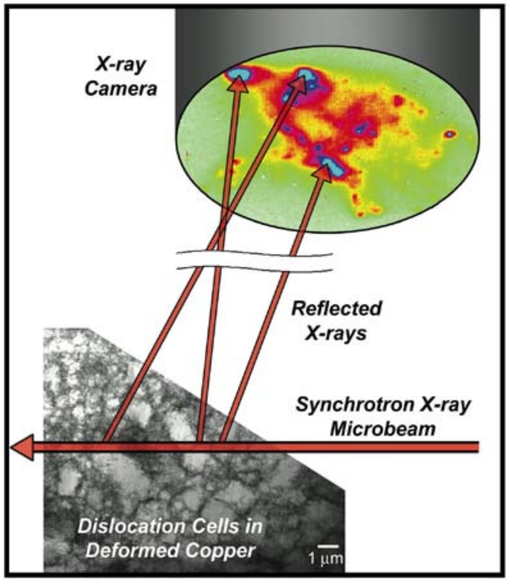
New information about deformation



- Near surfaces and interfaces
- In single and polycrystals
- Organization dynamics of defects (Krivoglaz defects of 2nd kind)

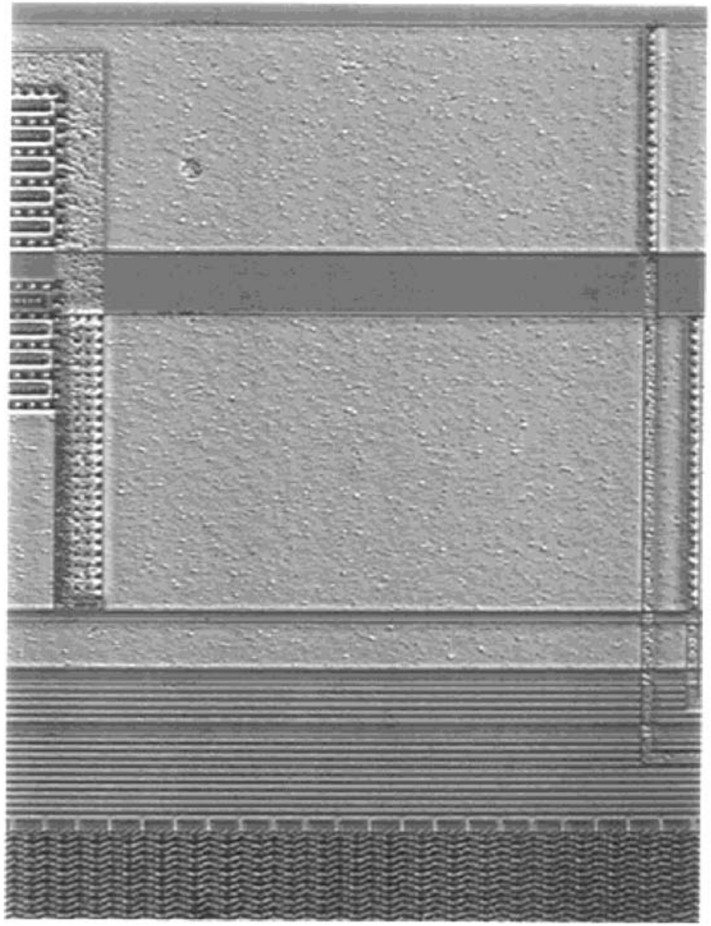
Lattice Rotation Map





Deformation found during electromigration

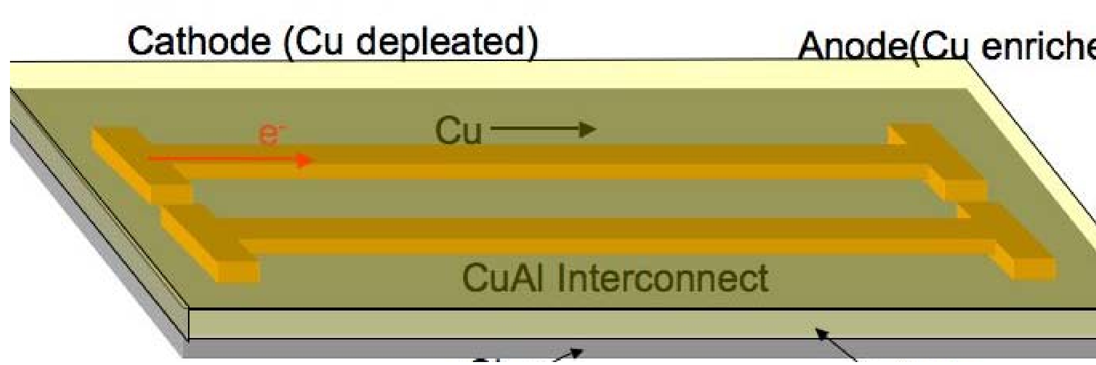
- **Device reliability linked to electromigration-MA/cm^2**
- **Theories assume stress development opposses mass transport**
- **Need to measure stress drove development of polychromatic microdiffraction**

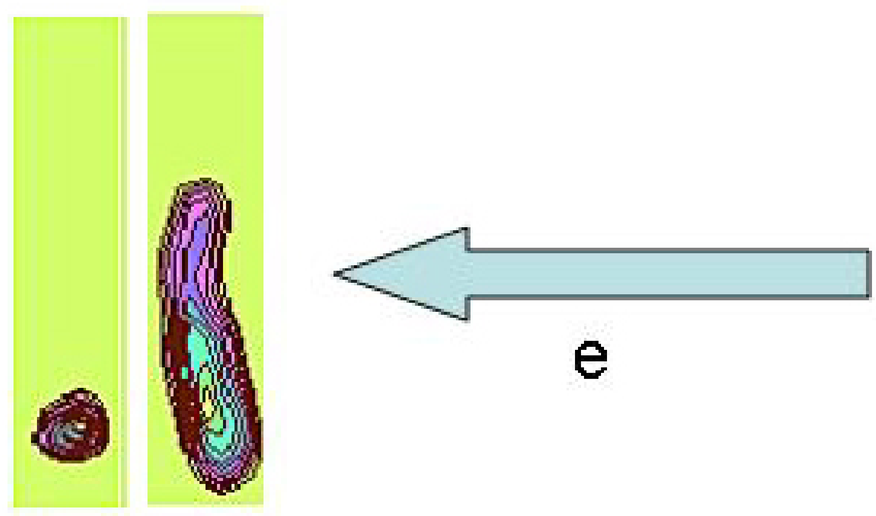


R. I Barabash et al. *J. Appl. Phys* **93** 5701 (2003).

Al (Cu) interconnects studied

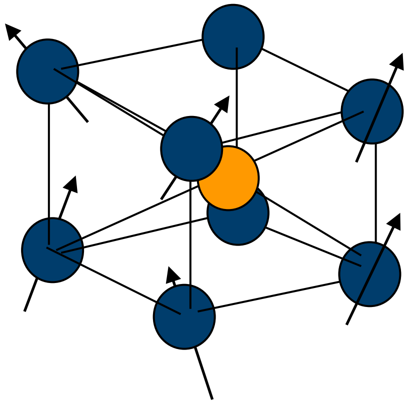
- **Deformation big grains.**
- **Small grains rotate.**
- **Deformation preceded Cu diffusion**
- **Unpaired dislocations develop parallel to the electrical field.**
- **Little dislocation activity near the anode end of the wire (Cu enhanced).**



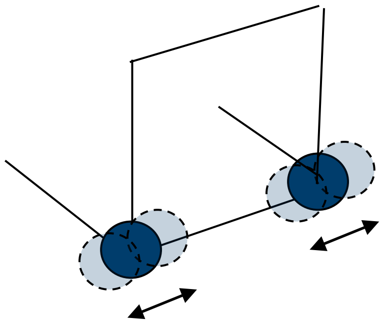


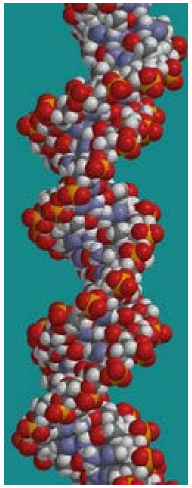
Neutron analog additional opportunities

Magnetism

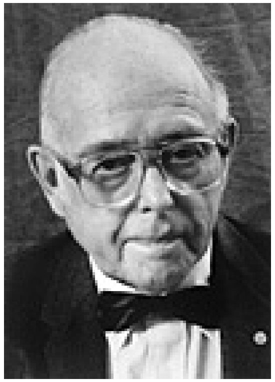


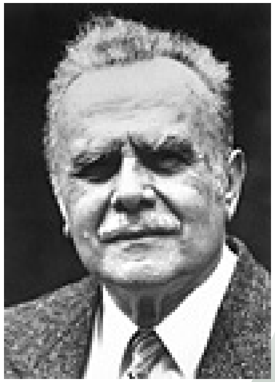
Atomic motions





Low Z materials





Nobel prize to Shull and Brockhouse

Even the most intense neutron source must be used efficiently

Neutron sources 10^{12} lower brilliance than advanced x-ray

Neutrons expensive 10^{13} more expensive!

10^{-16}\$ /x-ray

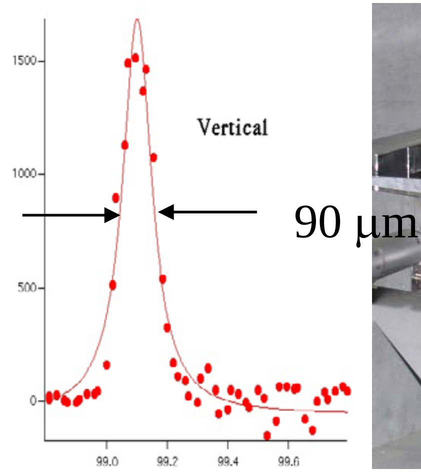
10^{-3}\$ /neutron

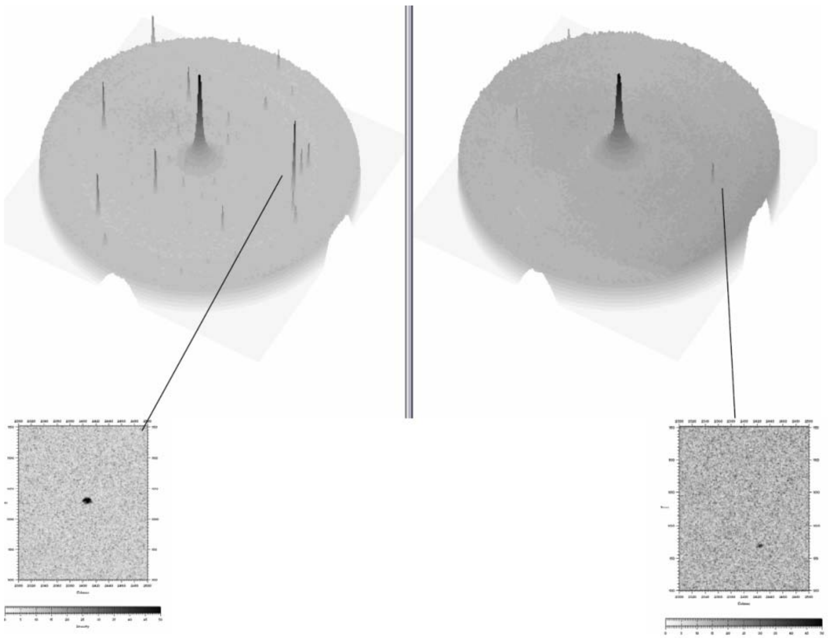
Increase divergence/bandpass

10^{-9}\$/ neutron

Neutron mirrors produce microbeams

- **Better signal-to-noise**
- **Resolve inhomogeneities**
- **Map crystal distributions**

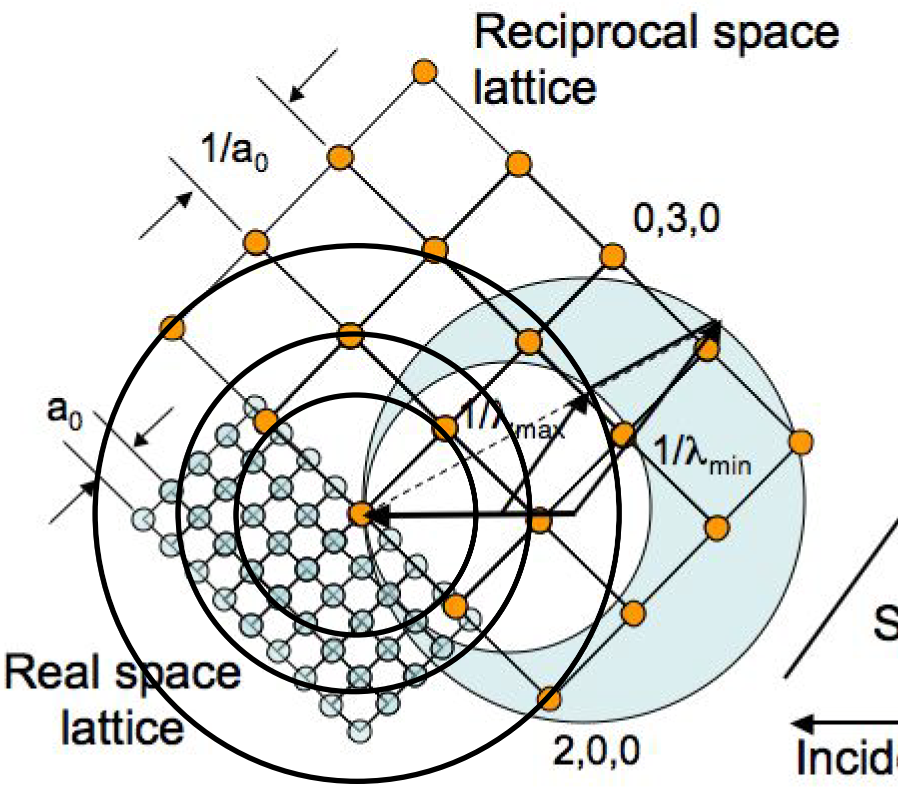




Useable 25 μm beams?

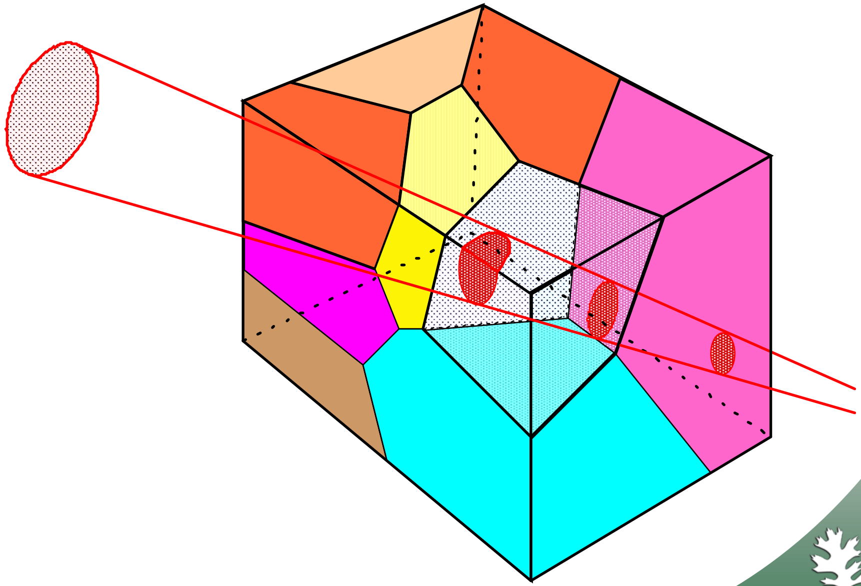
Spallation neutron science intrinsically polychromatic

- **Analogous to polychromatic X-ray microdiffraction-*but includes* energy**
- **Allows for structure determination**
- **Absolute strain measurements**



Will extend neutron science

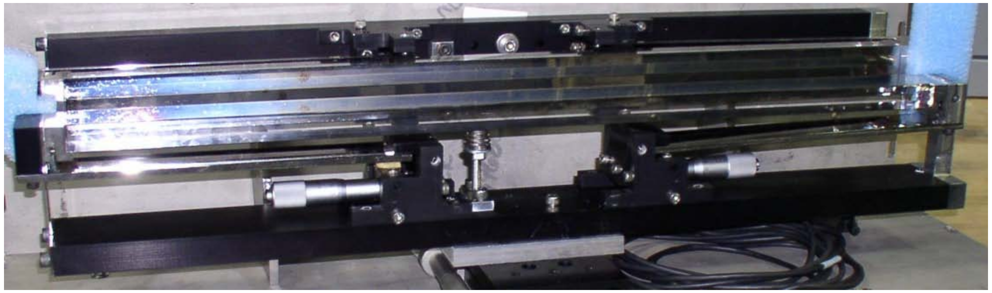
- **Inhomogeneous samples**
- **Small samples in environmental chambers**
- **Spatial resolved distributions deep in samples**

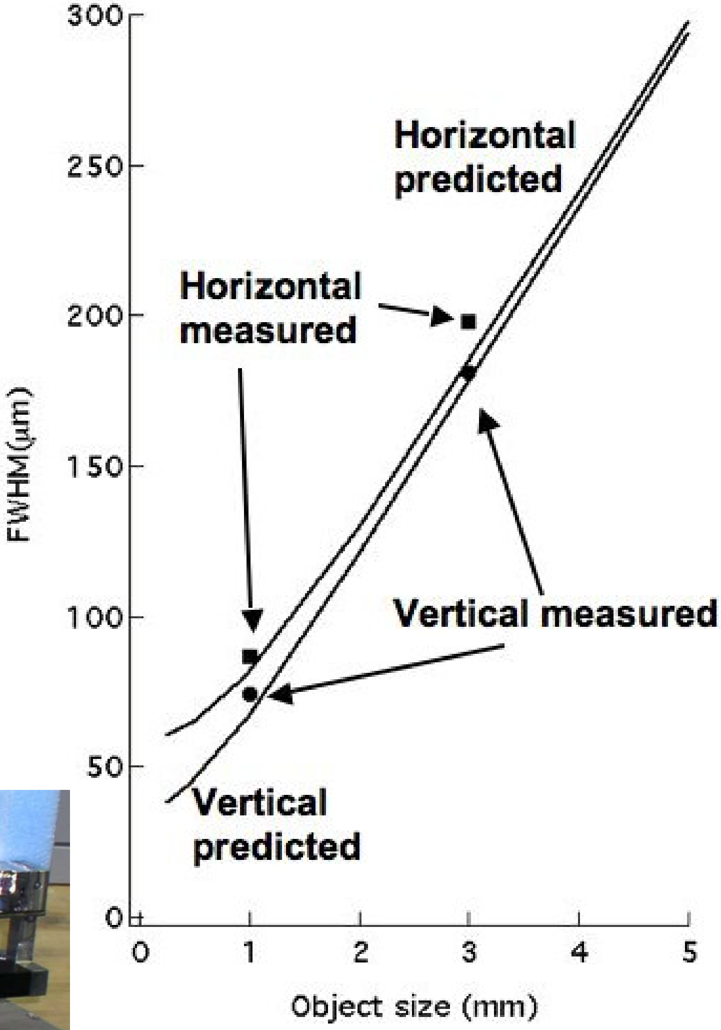


SNAP experiments diffraction high pressure cells

- **Focusing optics work near theoretical limit**

- **Minor improvements should enable 25 micron measurements**





OAK RIDGE
National Laboratory

Conclusion: Microdiffraction

- **Addresses long-standing issues materials physics**
- **Techniques and instrumentation rapidly evolving**
- **Answers specific questions about materials systems (Energy materials)**
- **Extend x-ray and neutron characterization to new classes of samples.**
  - **Dangerous**
  - **Inhomogeneous**
  - **Samples in extreme environments**

Advanced materials structures- intrinsically 3D
And spatial resolution- is needed urgently
X-ray and neutron beams- can penetrate to see
Materials crystal structures- and heterogeneity

CHORUS

Nondispersive - optics change what we can see
Mesostructure- resolved by crystallography
Atomic defects quantified - so that we can surmise
Emergent structures origins- at the mesoscopic size

New optics and new methods- extend what we can do
With spatial resolution- time resolution too
Nondestructive lets us watch- materials deep inside
Chambers or complex system - where once they could hide

Emerging applications- I've tried to show a few
Energy materials- have challenges quite new
With x-ray and neutron beams- we now are freed
To study these materials- on the scale that we need