X-ray and Neutron Microdiffraction

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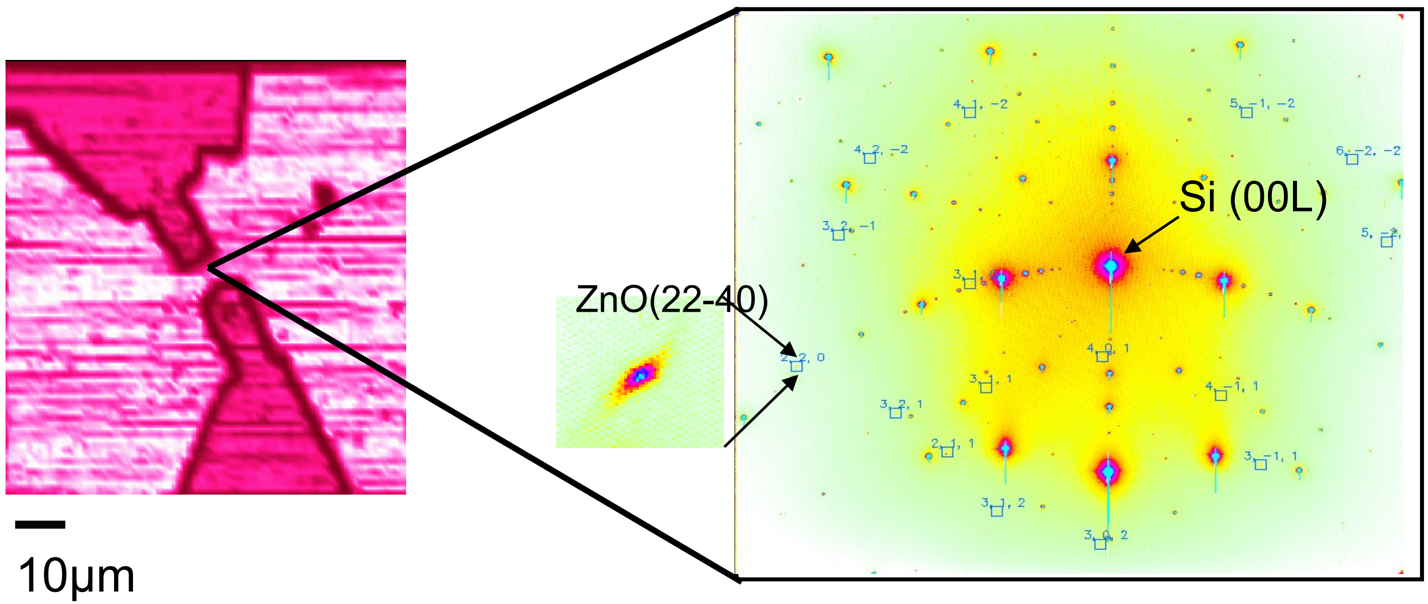


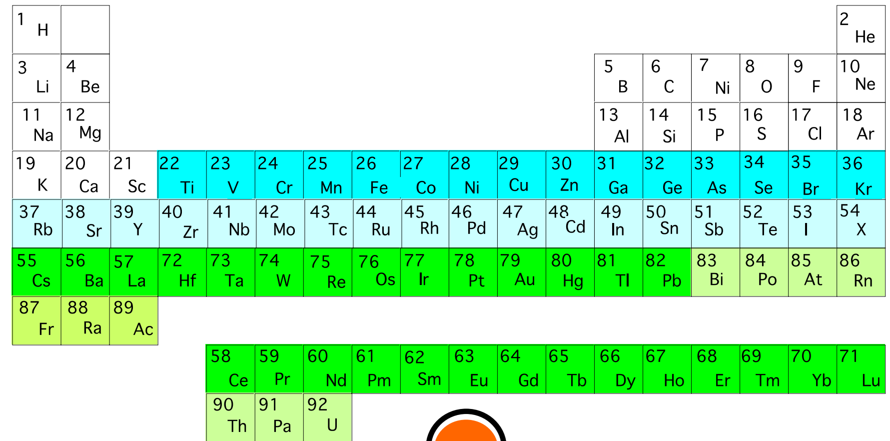
2012 Neutron X-ray Summer School

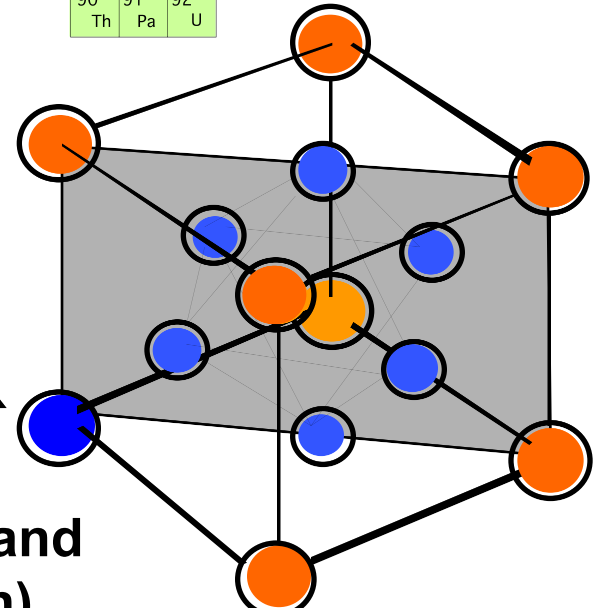
Two words

Spatial Resolution

Materials characterization begins 3 questions

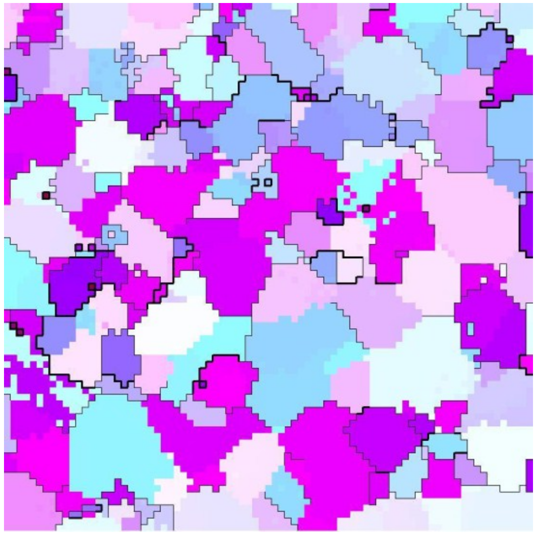
- What is the elemental composition?
- What is the crystal/local structure?
- What are the defects?



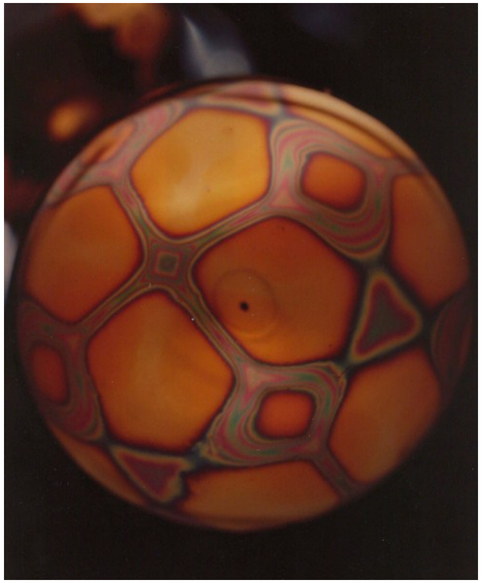


X-rays and neutrons probe structure and defects through scattering (diffraction)

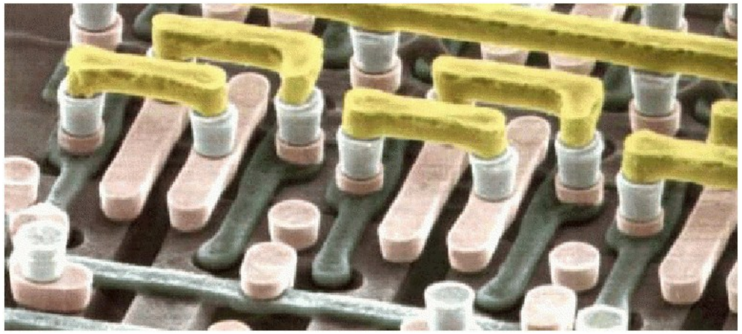
Spatial resolution essential!



- **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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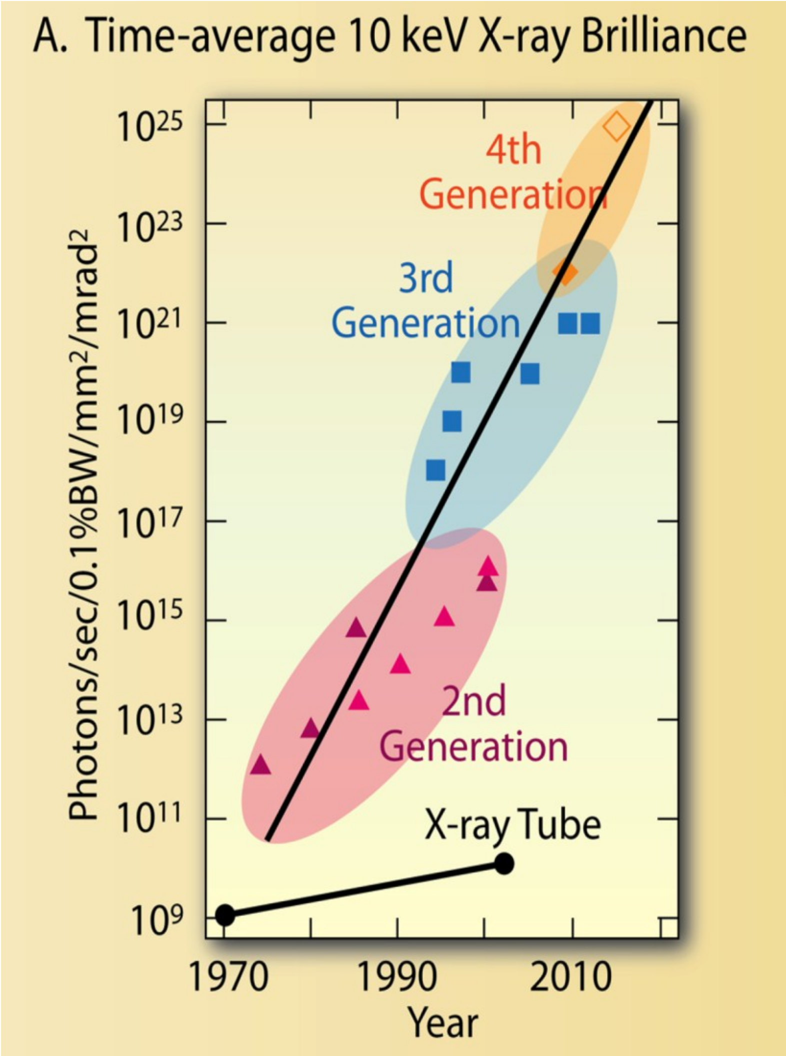
Spatial resolution essential for most advanced energy systems

New X-ray/ Neutron Sources Changing the Possible

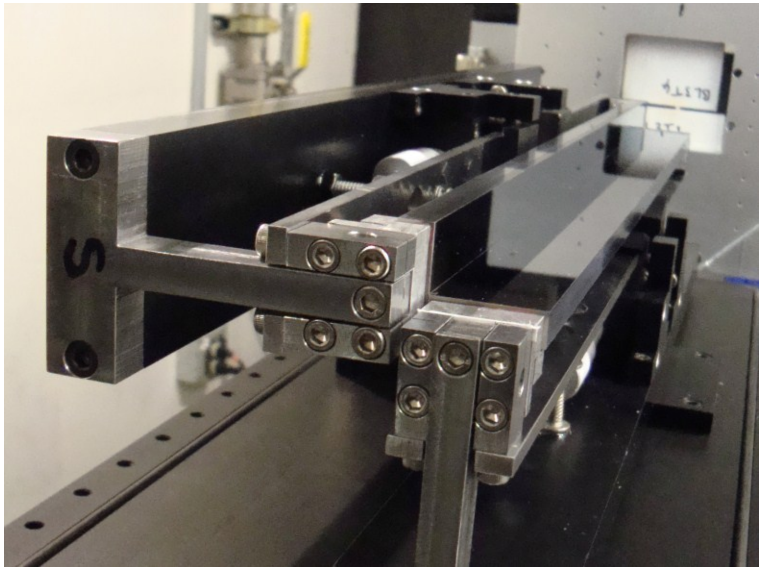
- Brilliance figure-of-merit for spatially-resolved exp.
- X-ray brilliance doubling faster than Moore's law
- SNS with 10x brilliance 100x more efficient detectors

TOPAZ/ SNS

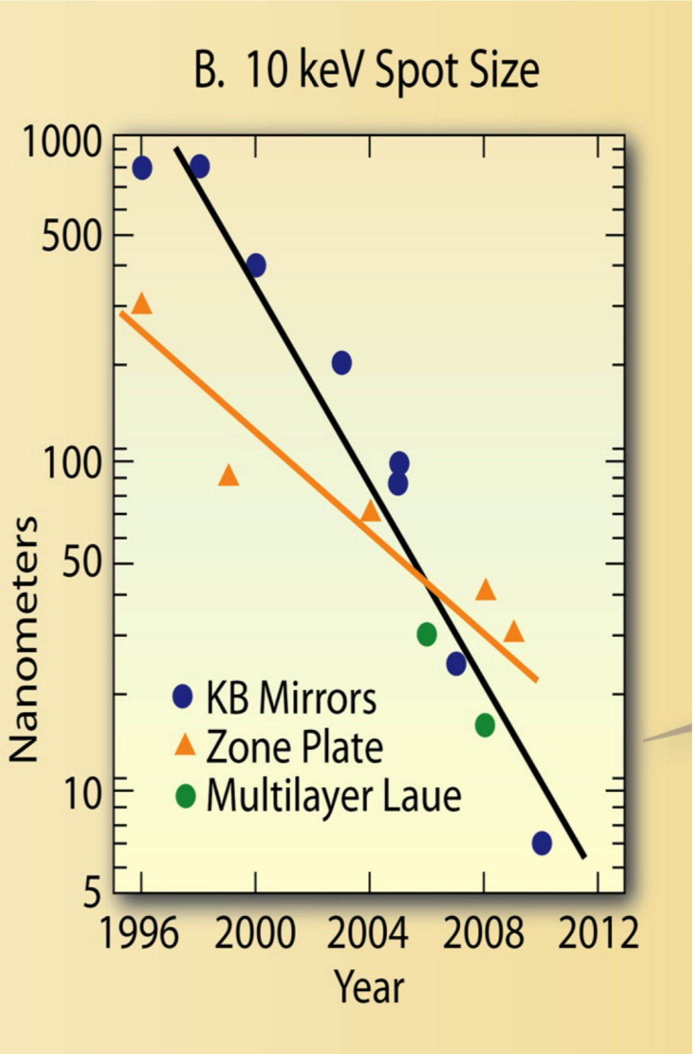


Spatial Resolving Optics Improving Rapidly

- **X-ray focal spot size routinely below 100 nm**
- **Neutron focusing optics below 100 μm**

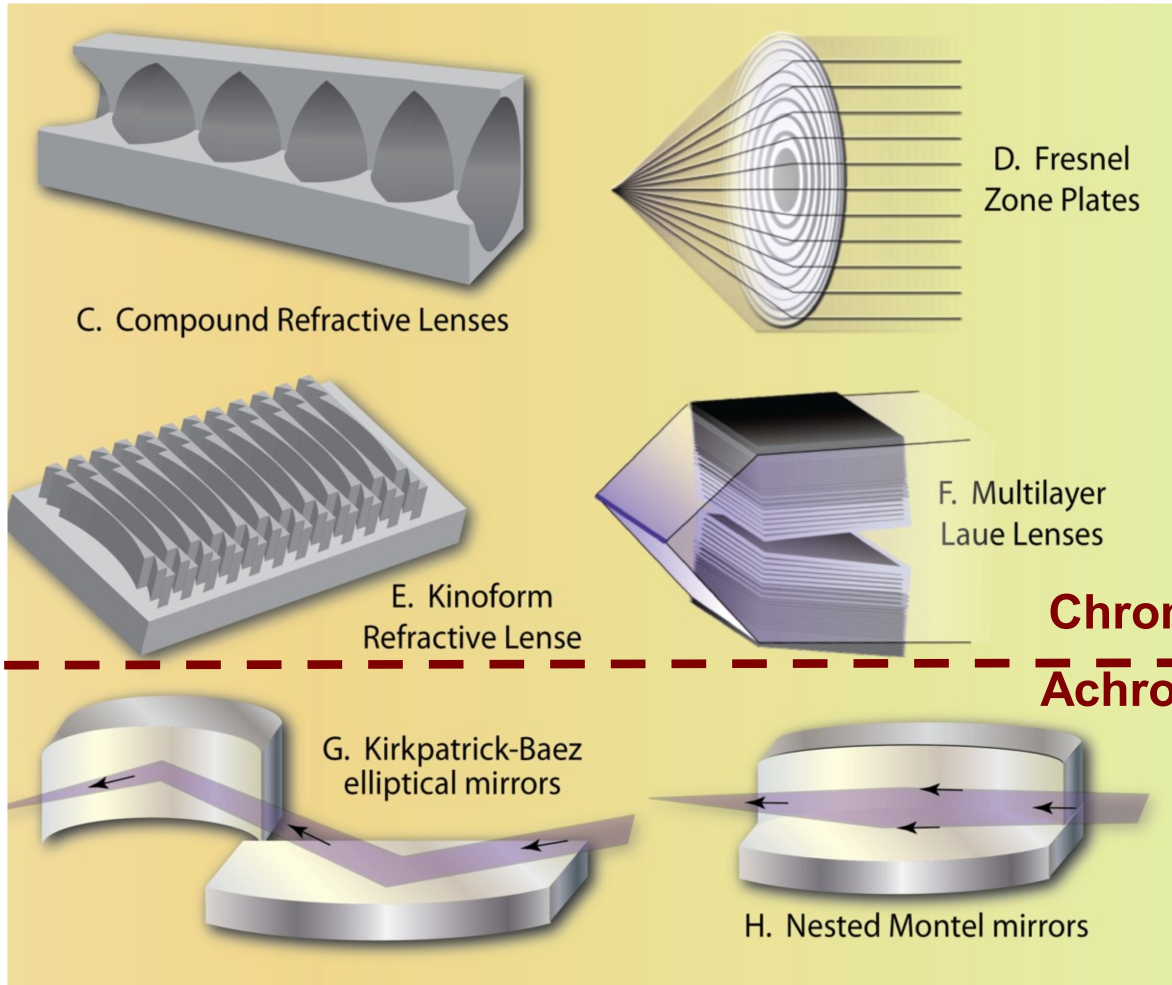


Neutron optics with <70 μm Focus



X-ray micro/nanofocusing optics rapidly evolving

- **CRL-50 nm**
- **FZP<30 nm**
- **Kinoform <70 nm**
- **MLL <15 nm**
- **KB <7 nm**
- **NMM<80 nm**

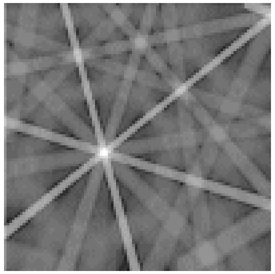


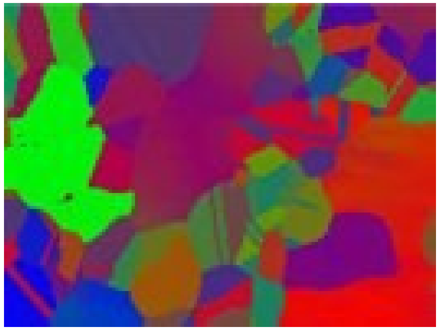
Chromatic

Achromatic

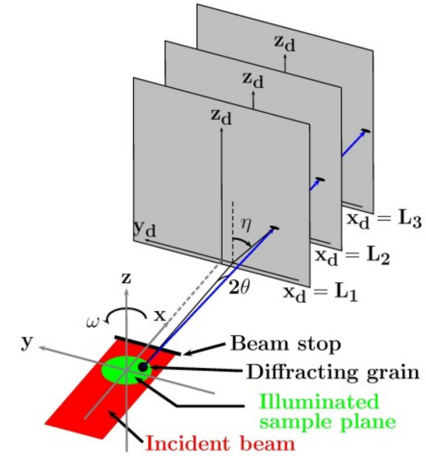
Diffraction mapping emerging area in electron and x-ray microscopy

- EBSD-transformed study of polycrystals
  - Surface phase
  - Surface orientation
  - FiB-3D mesoscale structure

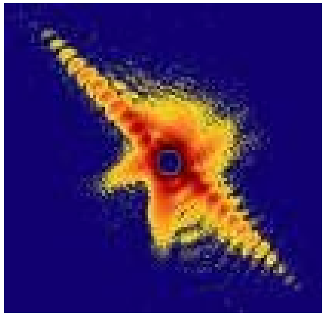




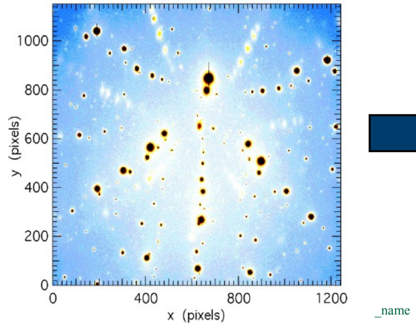
- 4D X-ray microscopy Lienert et al.
  - Time resolved
  - Deep penetration

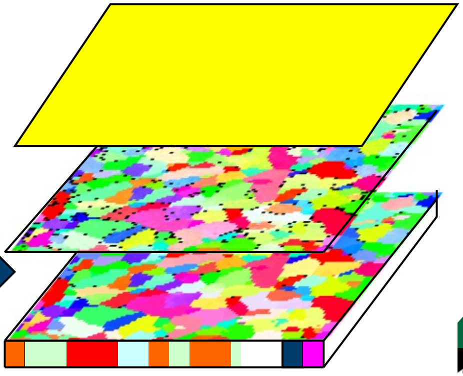


- Coherent X-ray Diffraction (Robinson et al.)
  - Simple structures



- Polychromatic X-ray microdiffraction
  - Phase/texture/***strain***/
  - ***Nondestructive***
  - ***Submicron***





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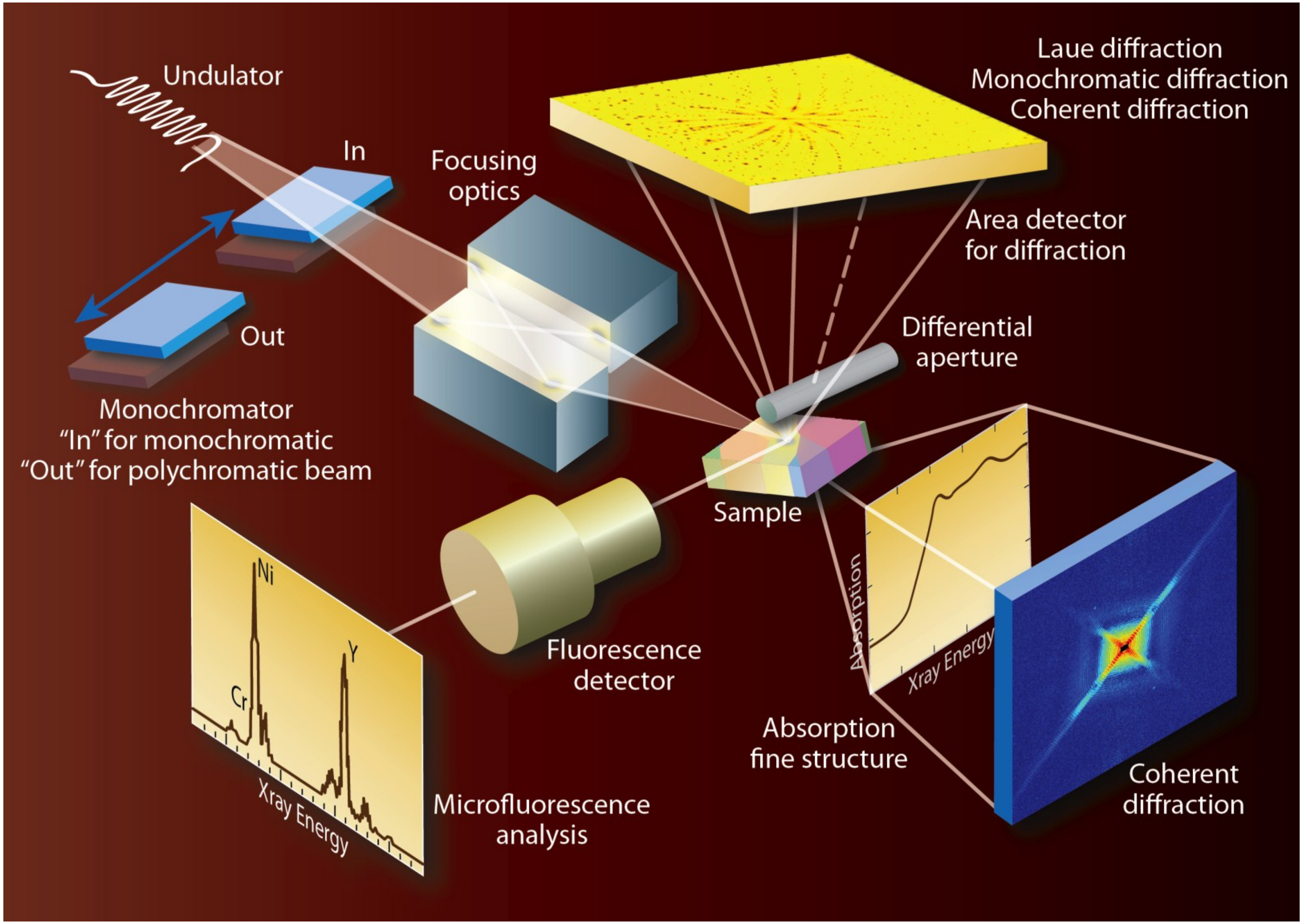
X-ray Micro/nanoprobe beams map chemistry and structure



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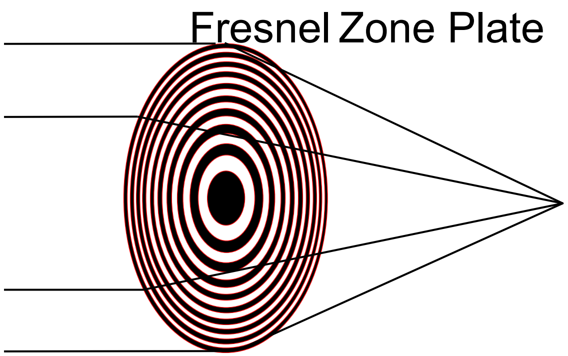
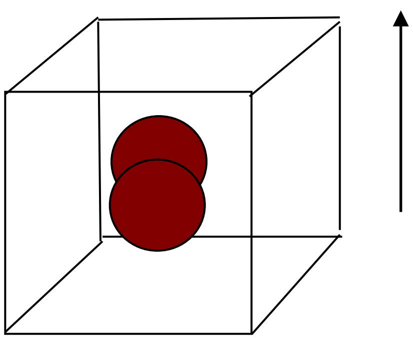
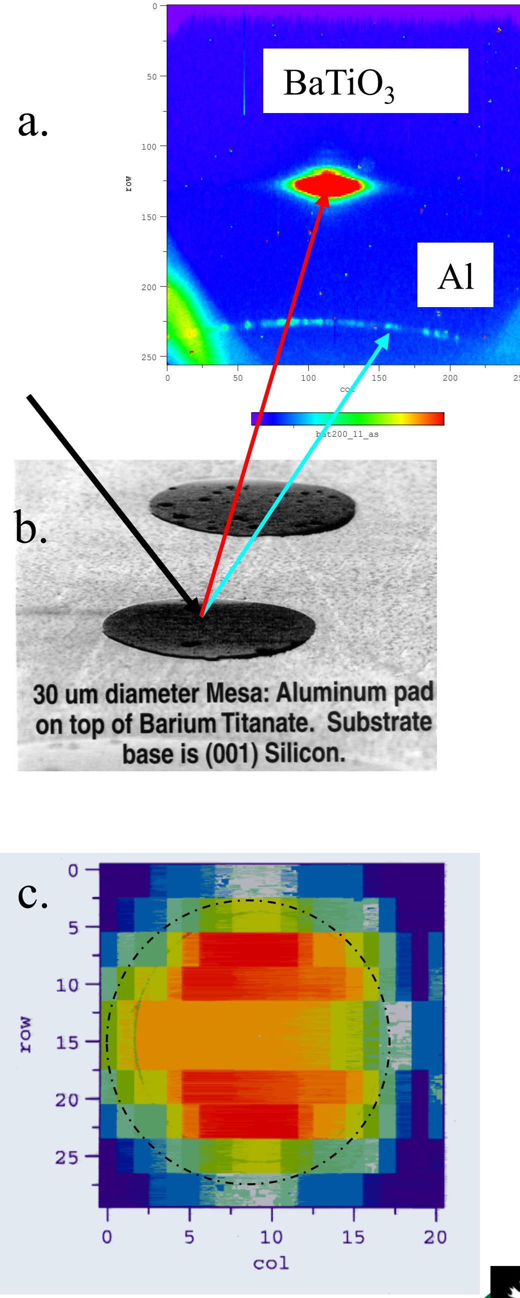
Monochromatic micro crystallography probes simple crystal systems



Wide-range of focusing choices



Ferroelectrics ideal samples



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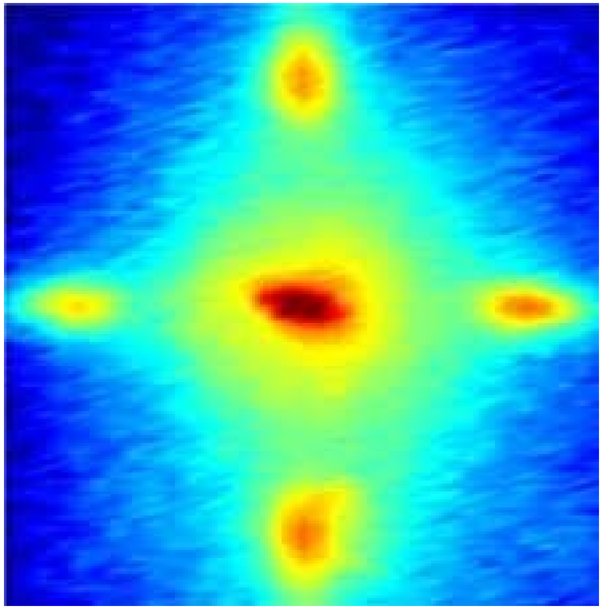
Thompson et al. study dimensionality of ferroelectricity

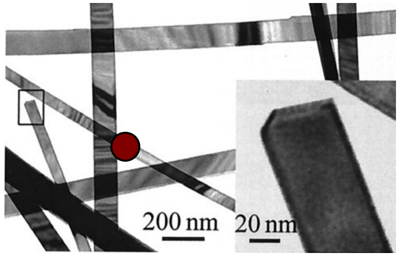
- Thickness
- Ribbons
- Dynamics



Diffraction from a ferroelectric stripe

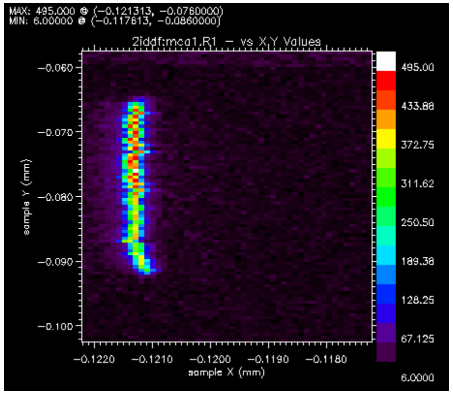
Ferroelectricity found with few unit cells

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

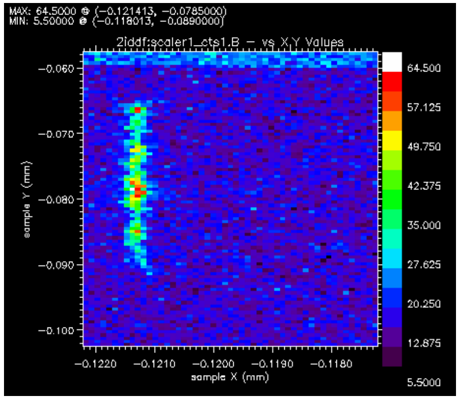


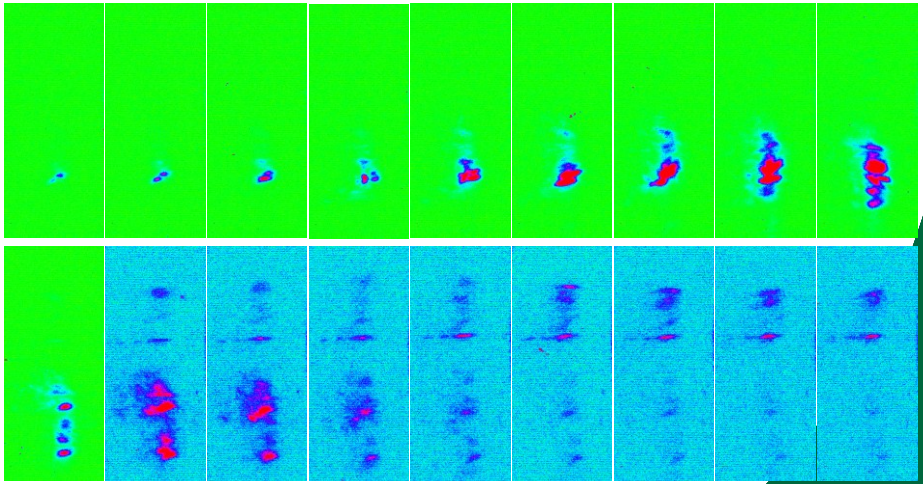
- 150 nm beam resolves crystalline substructure in individual Sn_2O_3 nanobelts

Fluorescence map



Diffraction map





APS *Nanoprobe*- opens new opportunities for spatially resolved

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

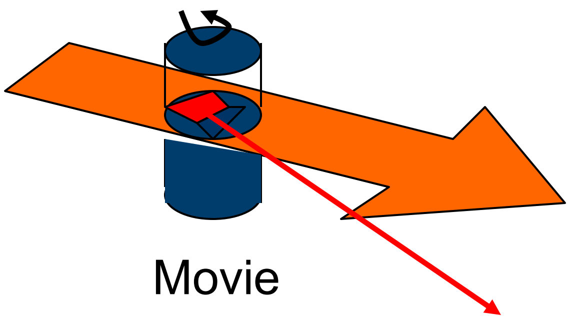
~30 nm now

<10 nm possible in future

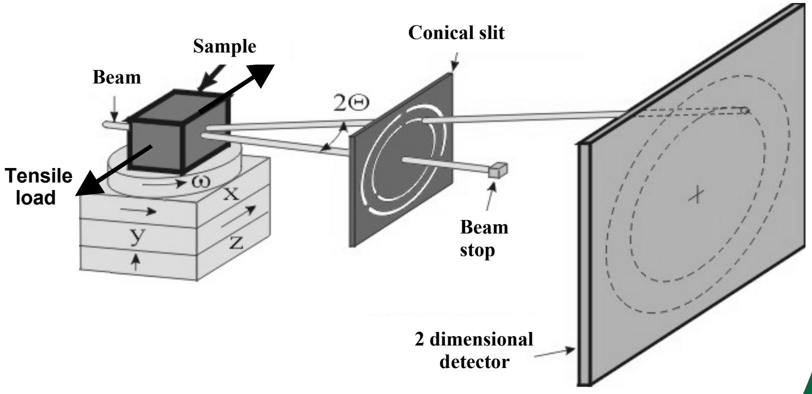
NSLSII ~1 nm!

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

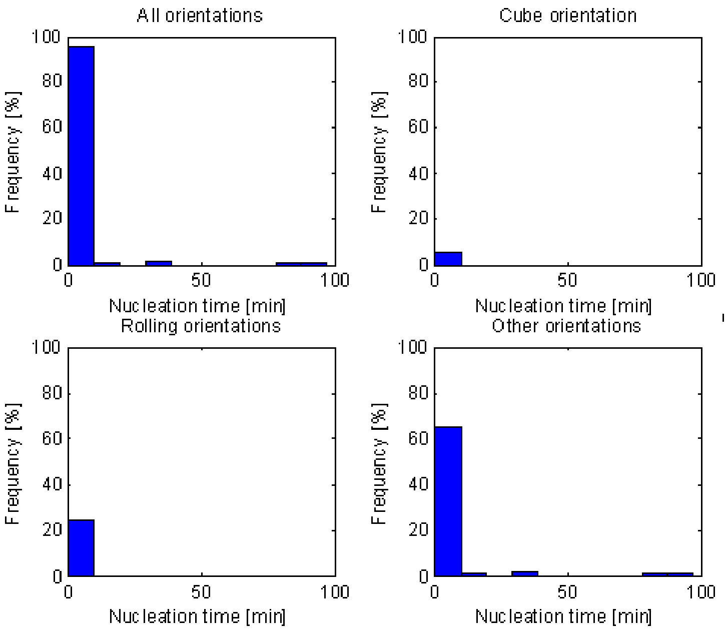


- E>50 keV allows deep measurements

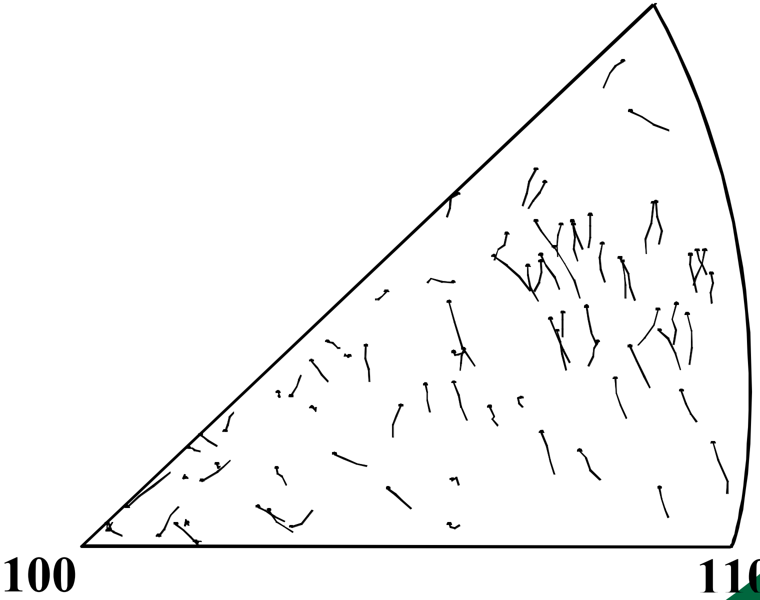
Best with high-energy beams/Beamline 1 at APS

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***

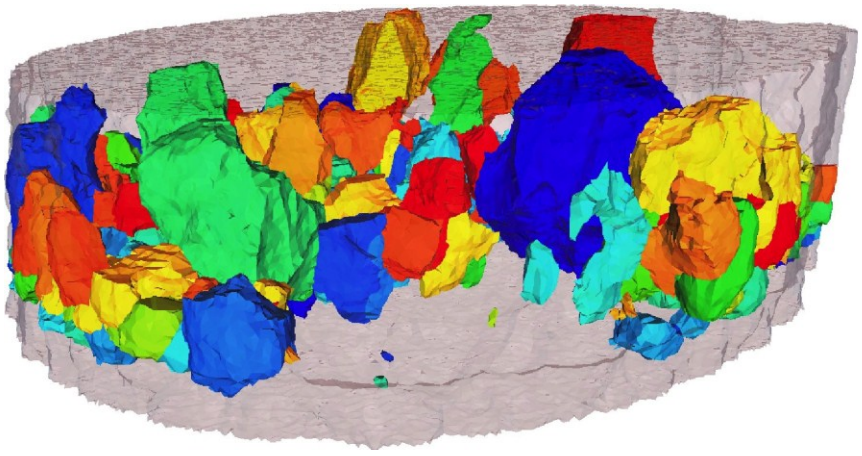


- **Rotations/texture evolution individual grains during deformation**
  - **Tests deformation models**
  - **L. Margulies, G. Winther and H.F. Poulsen, Science** 291**, 2392-2394 (2001).**

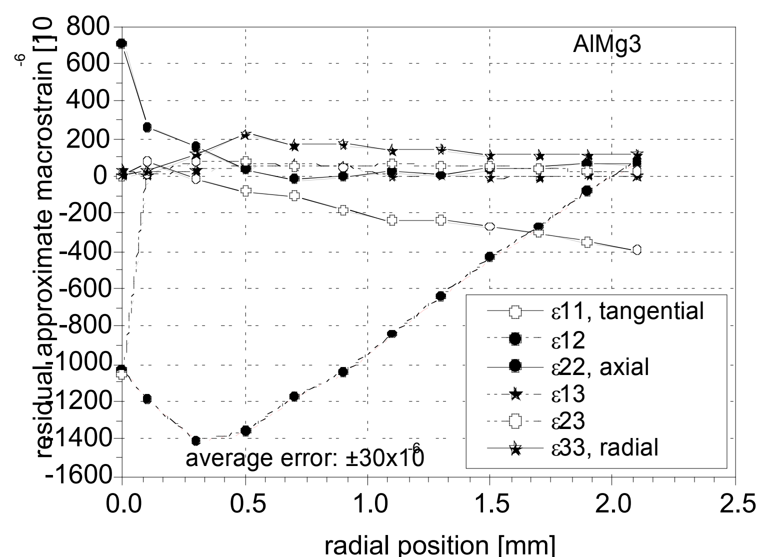


4DXRD Microscope provides additional powerful capabilities

- **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**



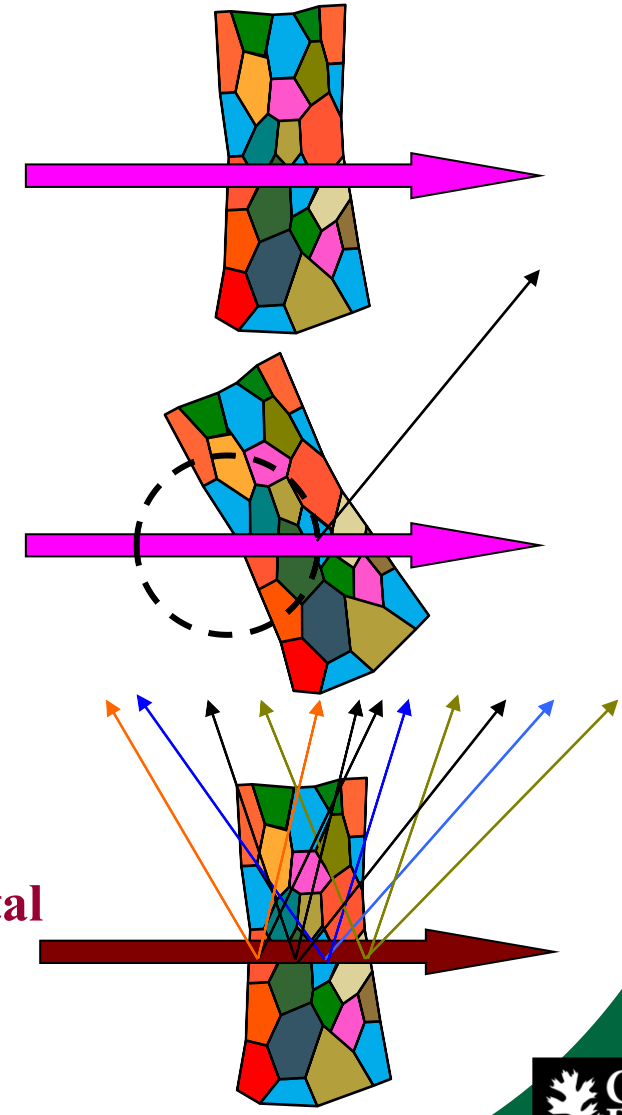
Ideal for neutrons! But needs high-resolution detectors!

Strain tensor elements in torsion sample

Polychromatic simplifies microdiffraction

Solves intrinsic problem with conventional microdiffraction-

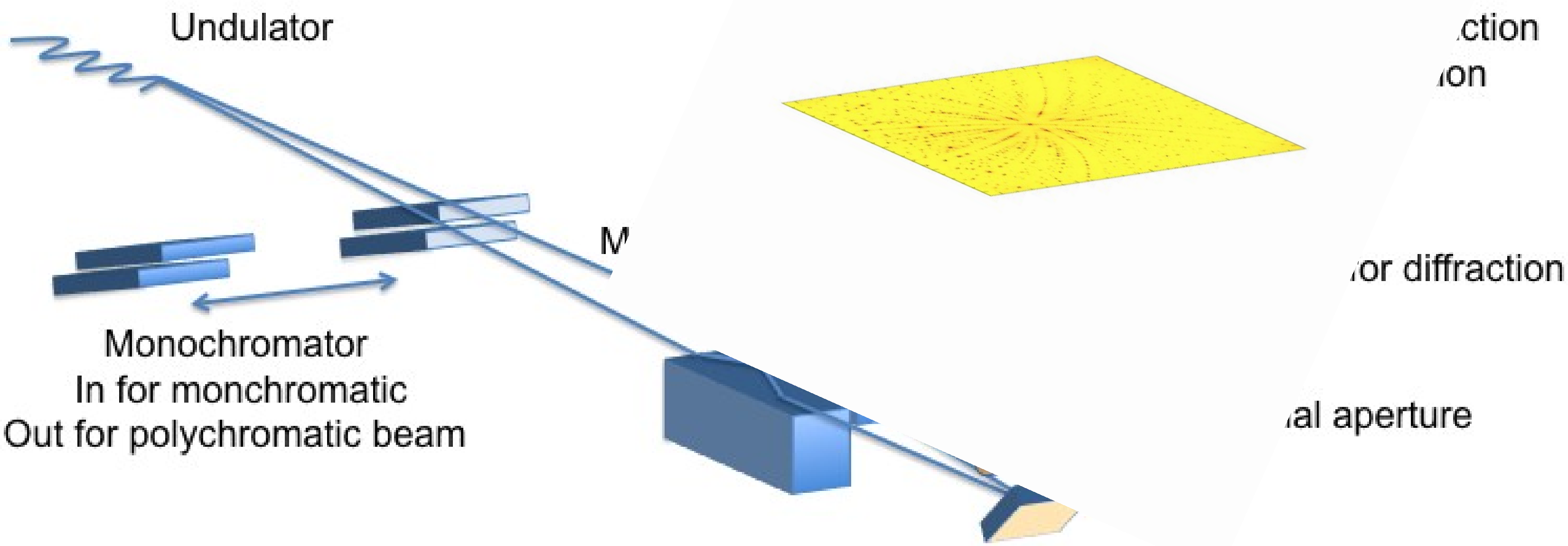
-Sample does not need to be rotated!

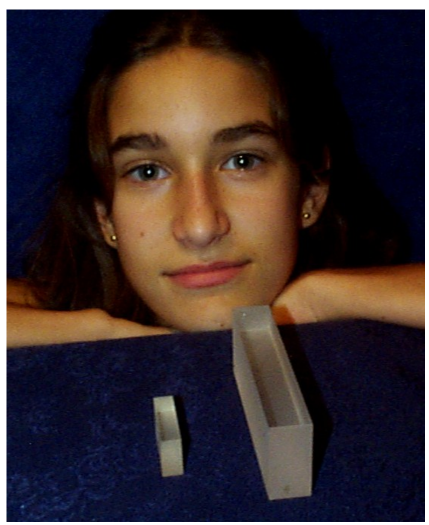


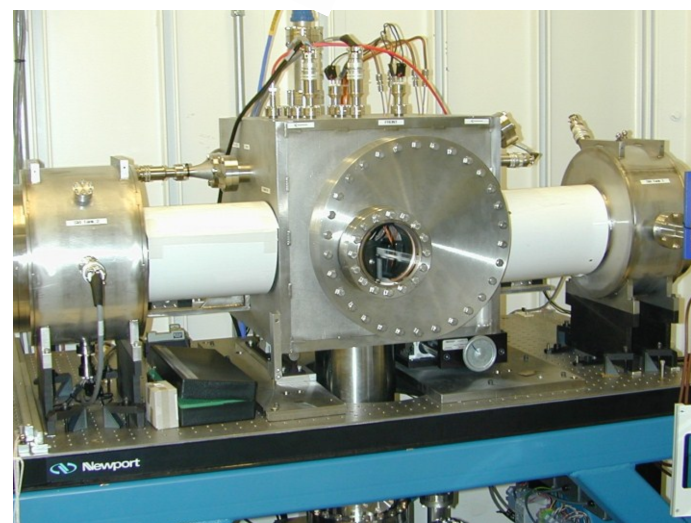
Special software required- Can index polycrystalline samples

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

3-D X-ray Crystal Micro pecialized elements



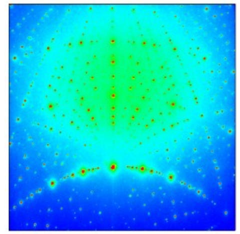


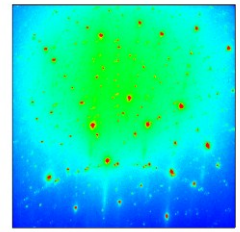


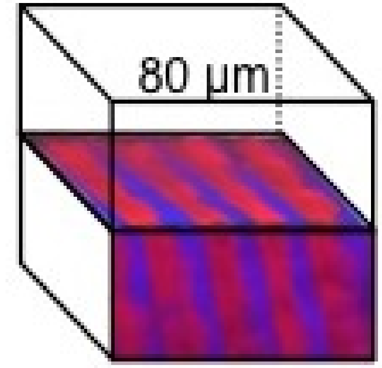
<0.25 x 0.25 x 0.5 μm^3
strain~10^{-4}-10^{-5}

Provides Submicron 3D Maps With New Information

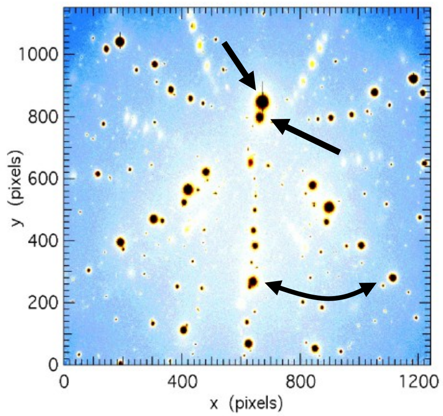
- **Phase boundaries**

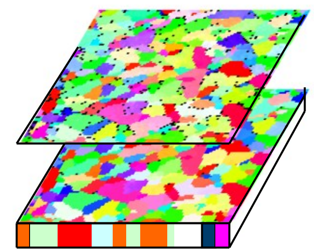




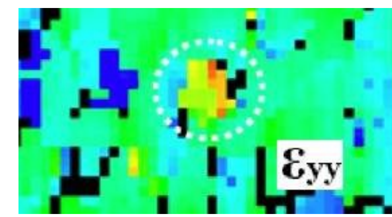


- **Grain boundaries(3D)**

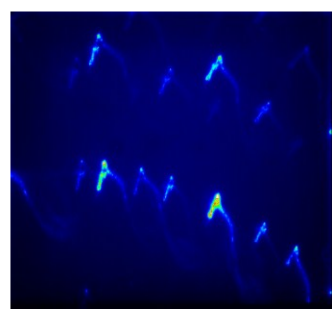


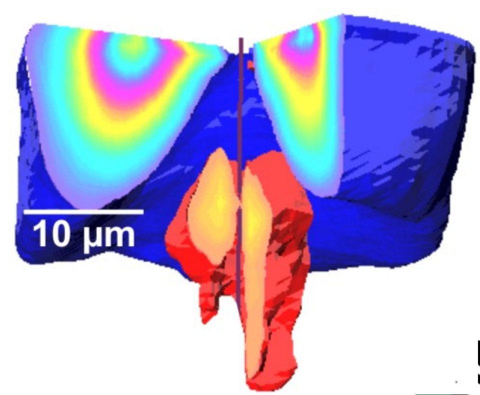


- **Elastic strain**



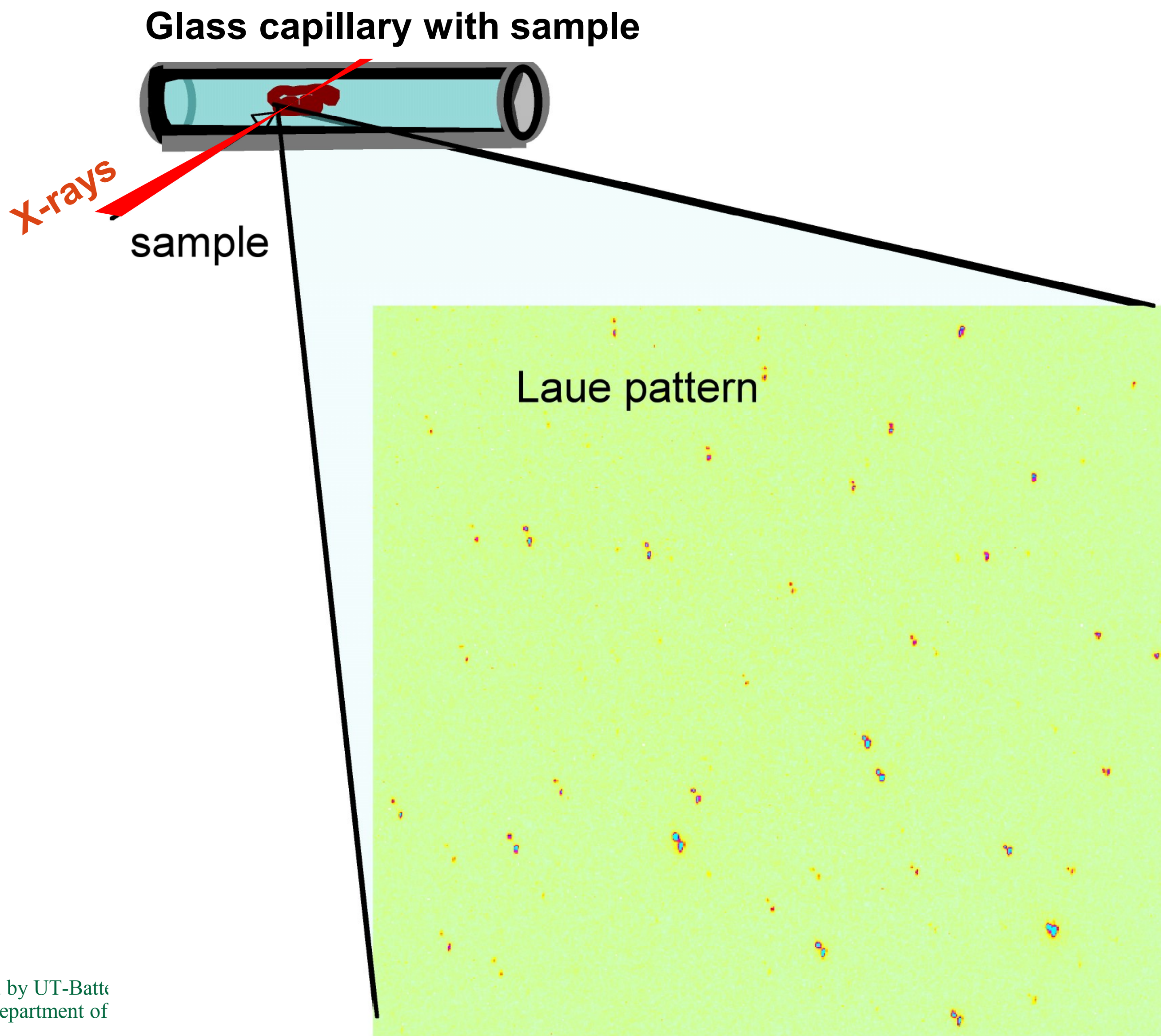
- **Deformation /Nye tensor**





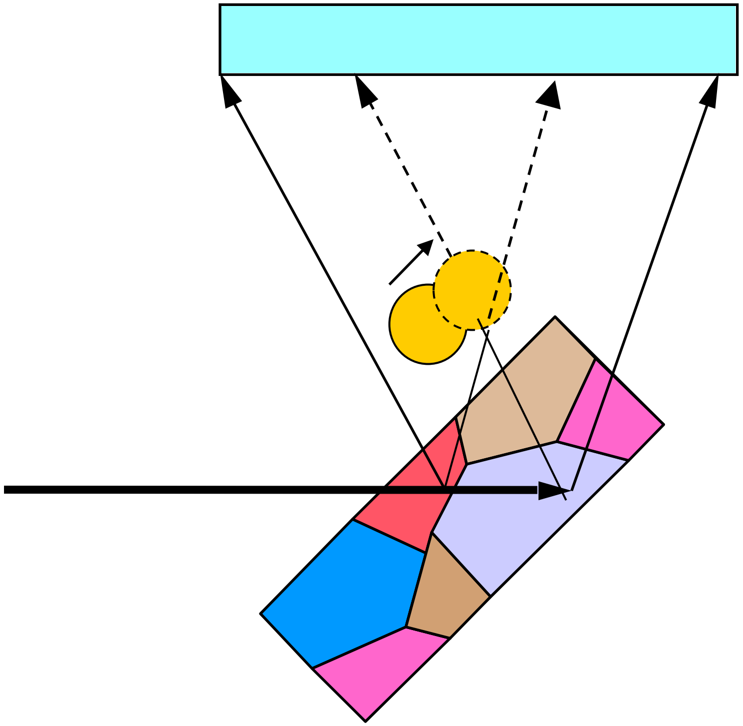
Nondestructive!

Laue methods essential for some samples



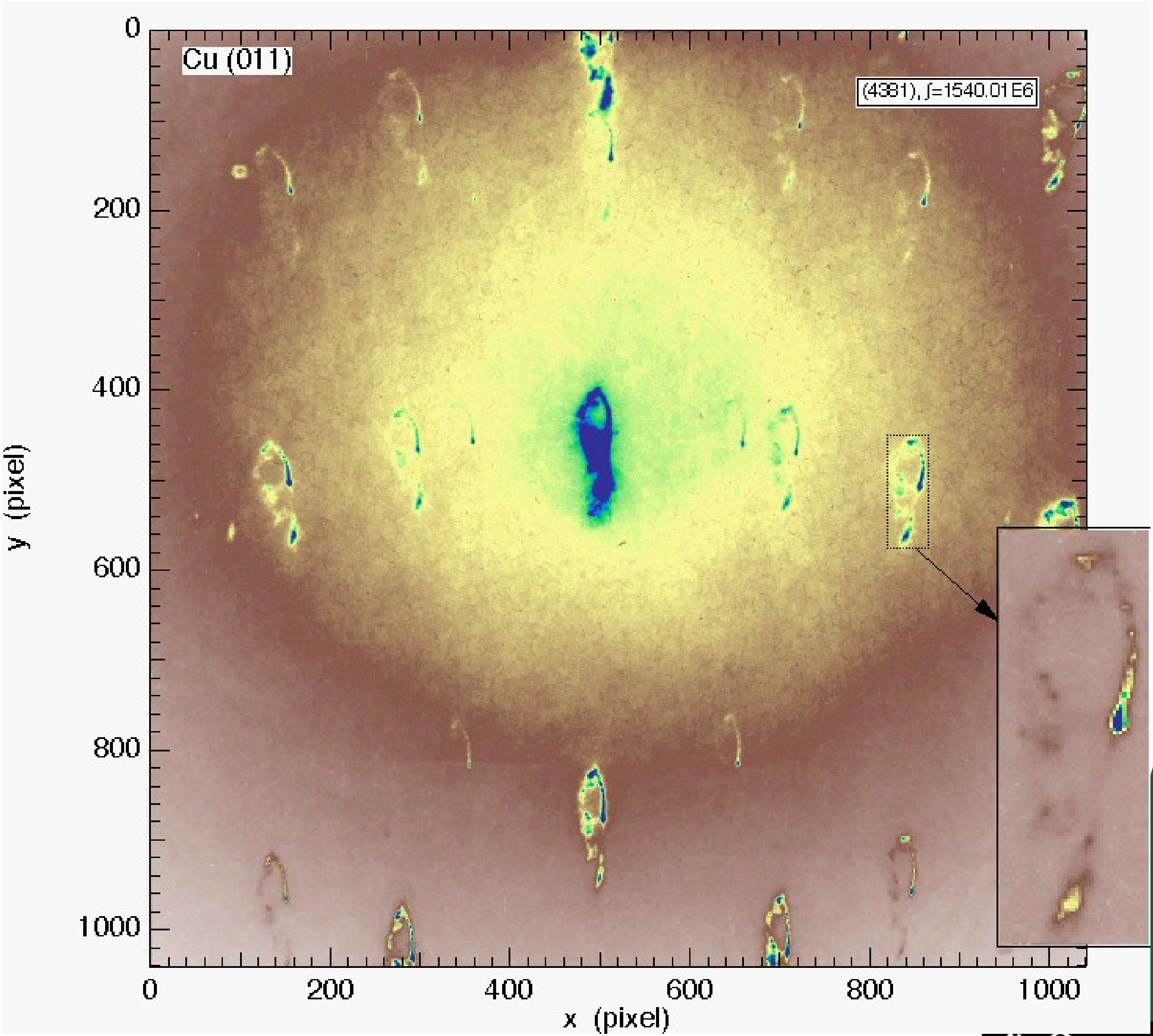
Differential aperture microscopy resolves submicron along incident beam!

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

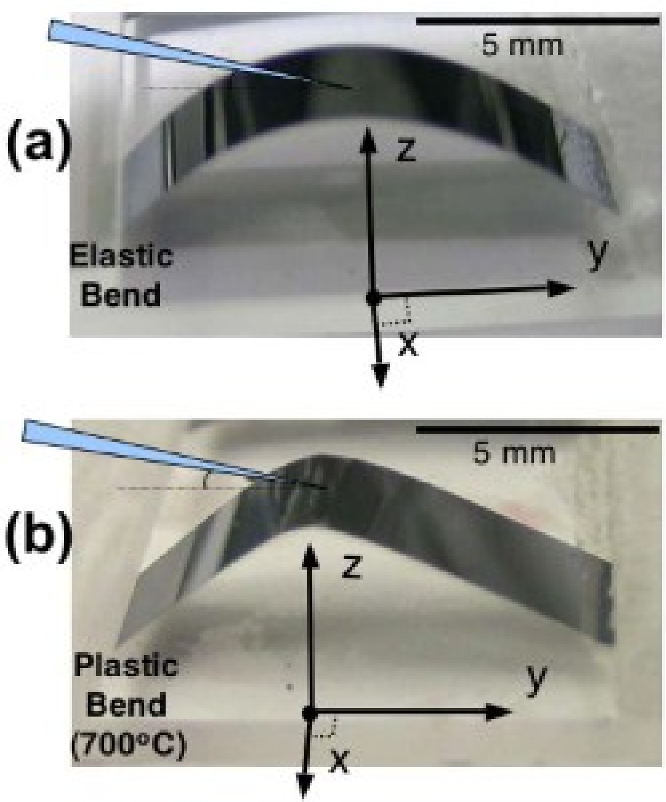


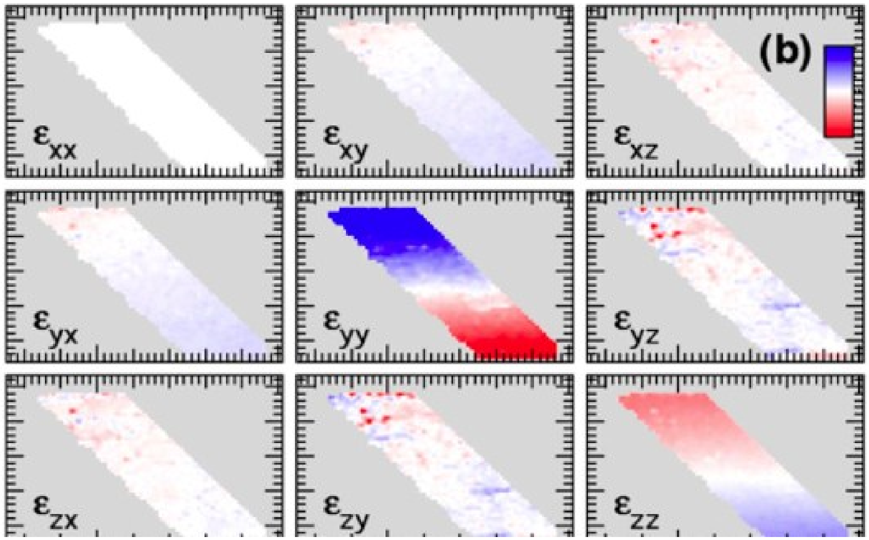
As wire moves its edge cuts through Laue spots

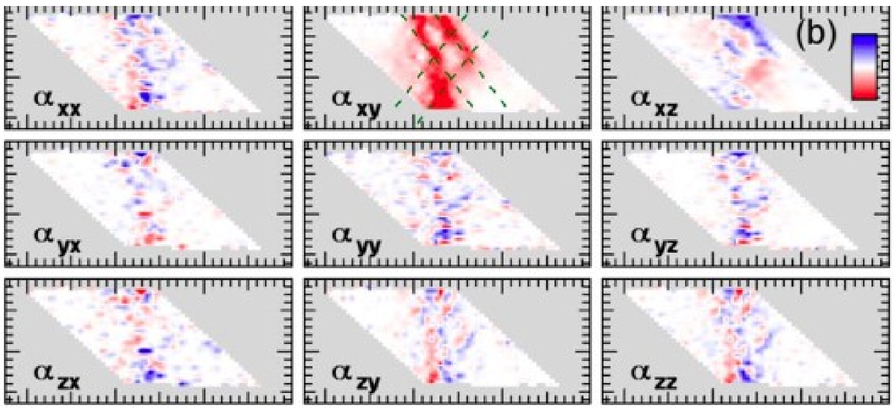
- **Near-surface fluorescence provides moving shadow**
- **Long scans needed for deep penetration**



Measurements of elastic strain tensor *inside* bent single crystal Si illustrate power of DAXM



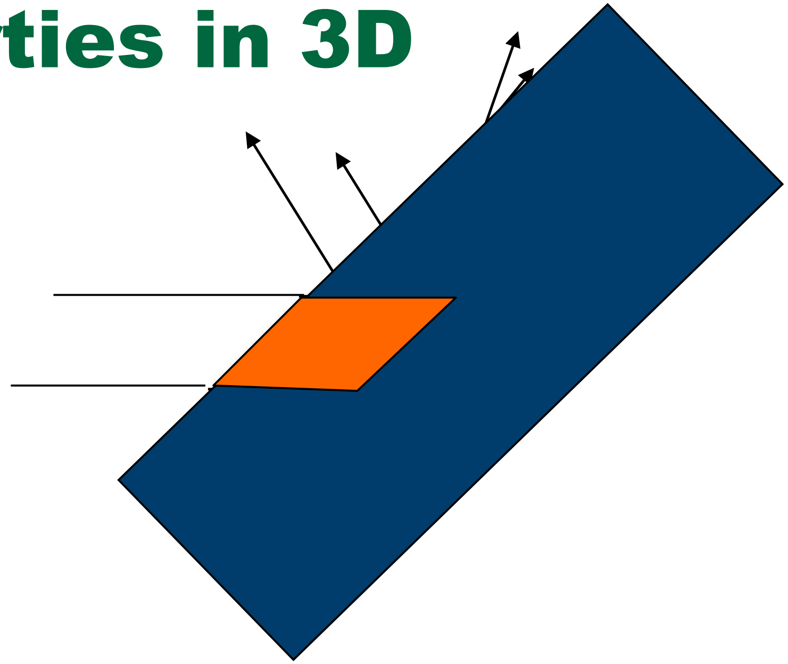




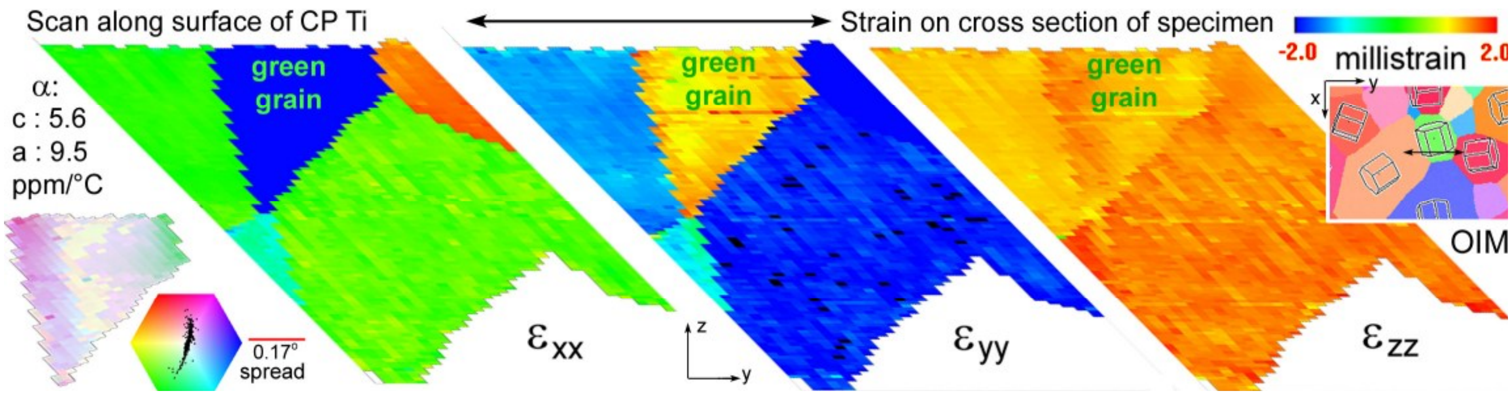
Orientations to 0.001°

Larson et al. J. Eng. Mat. and Tech. 130 021024 (2008) ORR award

Maps crystal properties in 3D



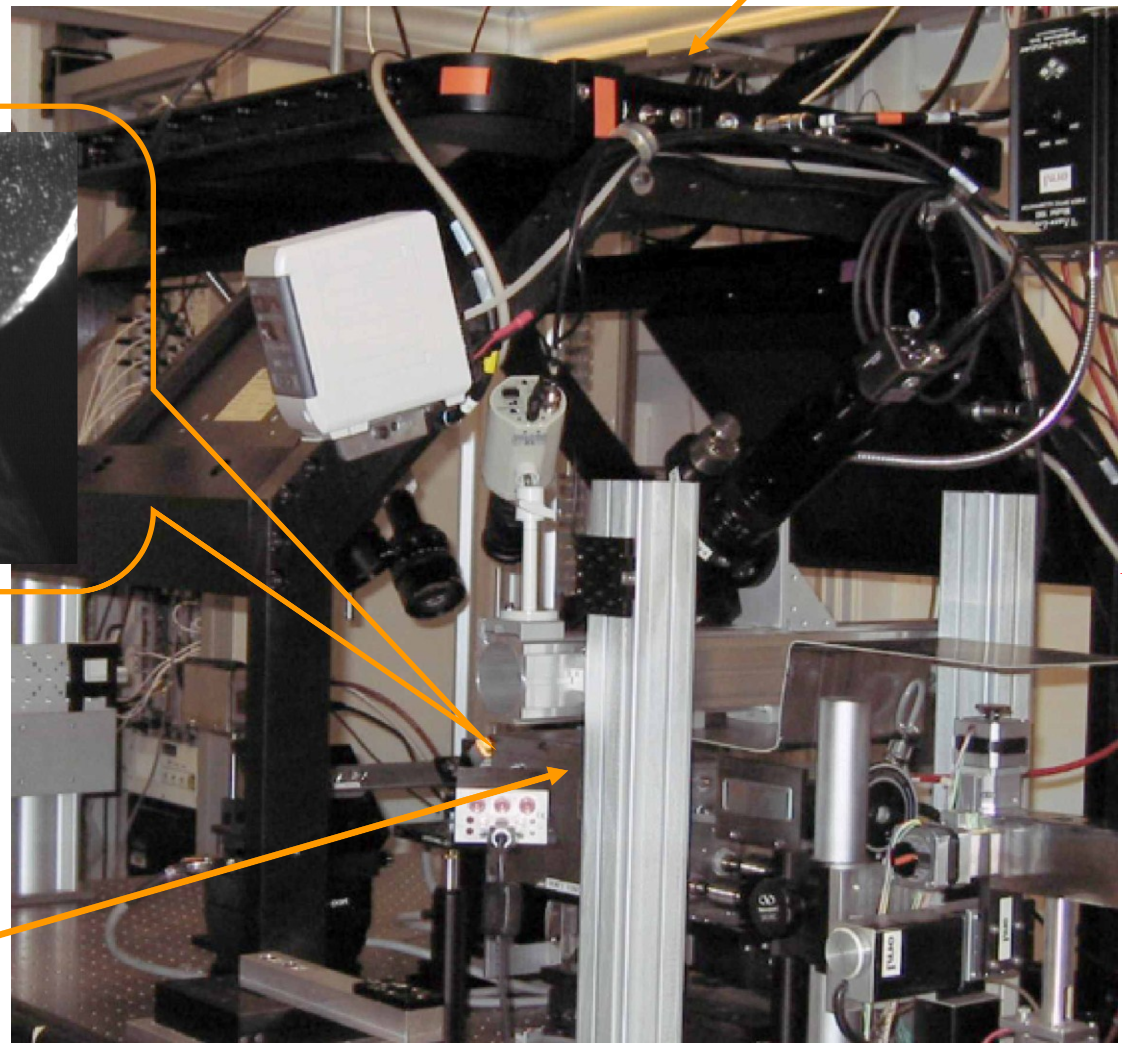
- Phase
- Texture (orientation)
- Elastic strain tensor
- Nye tensor (deformation)



T. Bieler et al.

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

Amorphous Si
Area detector



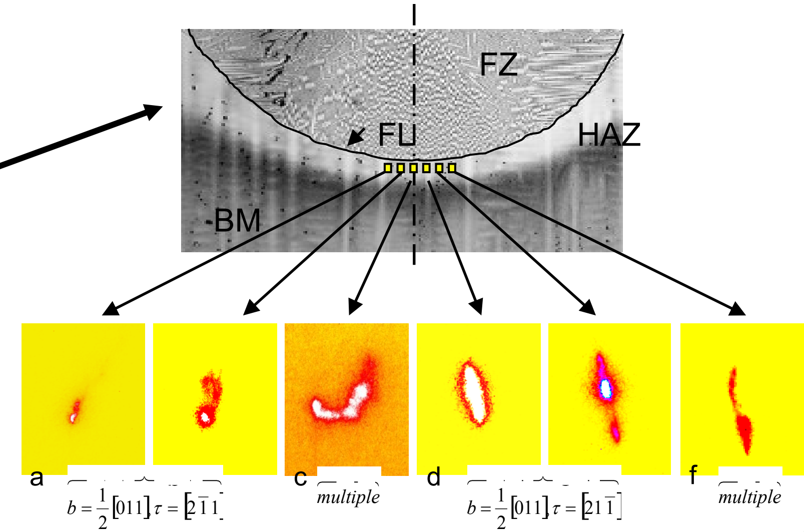
differential aperture

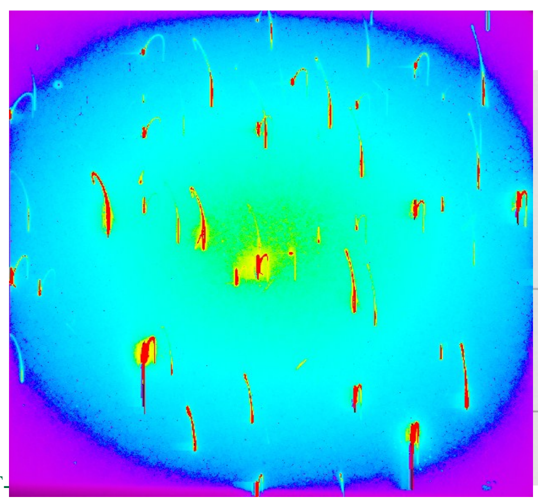
(wire scan, ~ 200 μm
above sample surface)

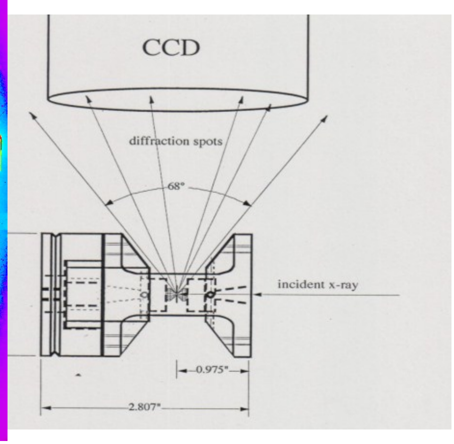
Mirror box

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**

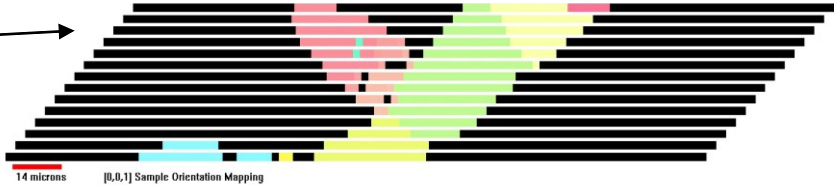




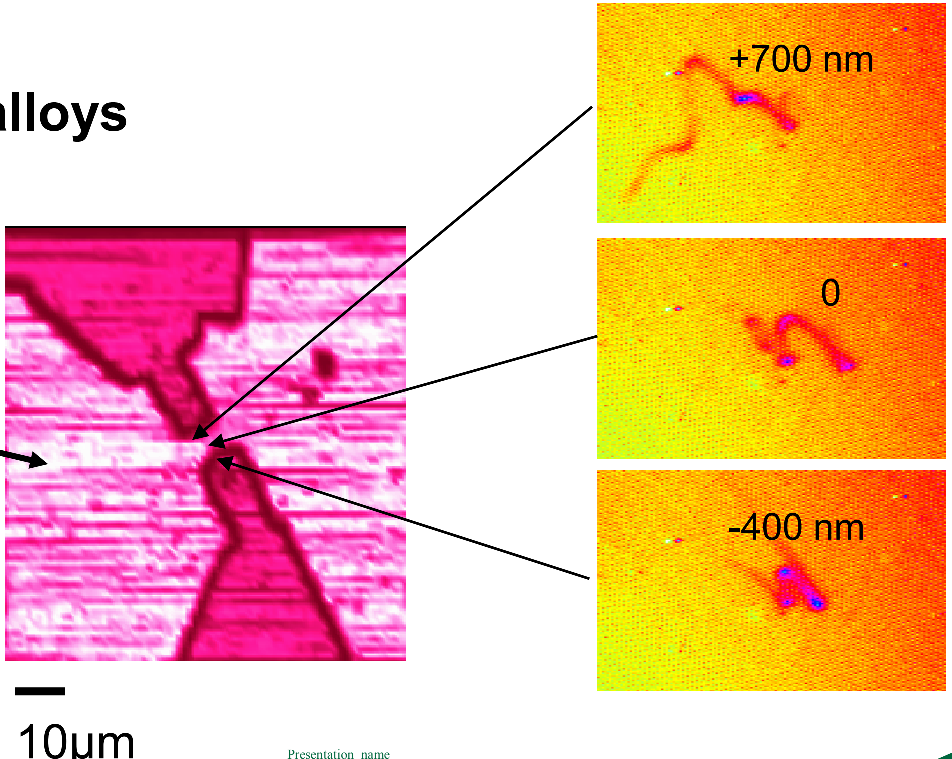


Ongoing too extensive continued..

- Domain wall structure measurements
- Sn whisker growth



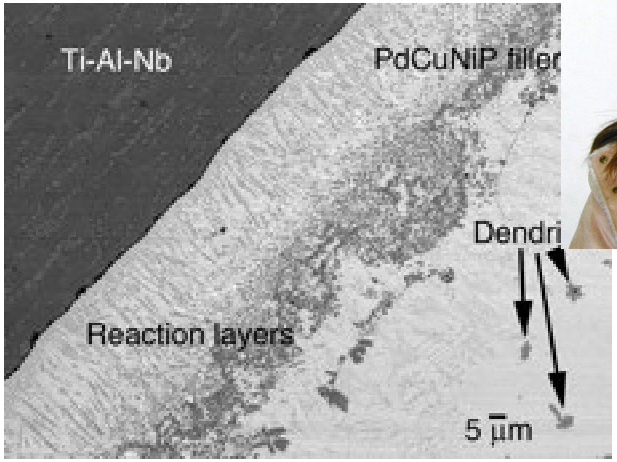
- High-performance alloys
- Nanomaterials



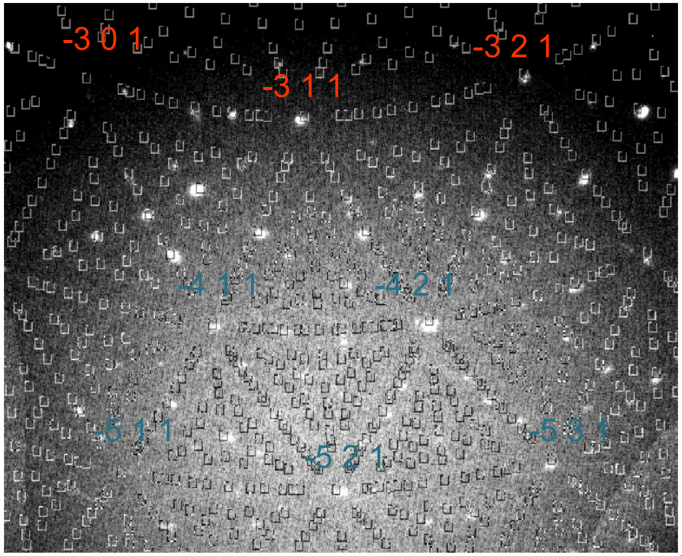
Energy scans allow structure determination

- Generalization of orientation software can identify phases
- Energy scans provide integrated reflectivities.
- Identified two minor crystal phases tetragonal/hexagonal

Cannot be found by powder

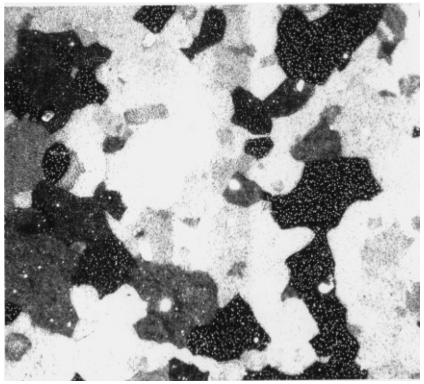


BAM braze $Pd_{40}Cu_{30}Ni_{10}P_{20}$

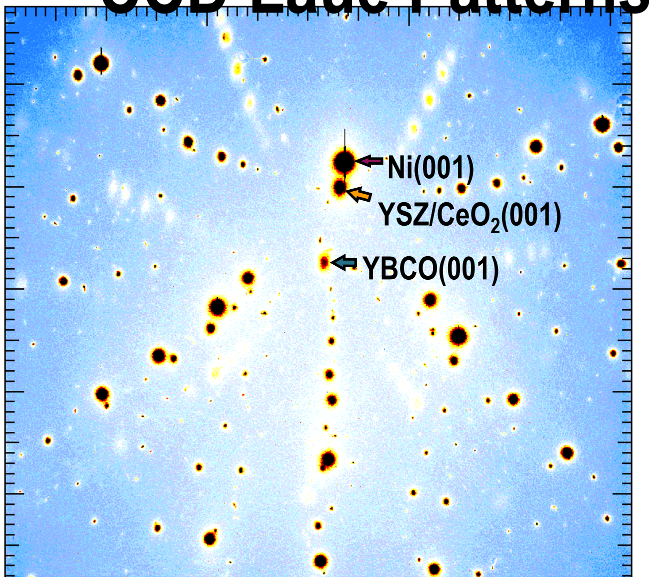


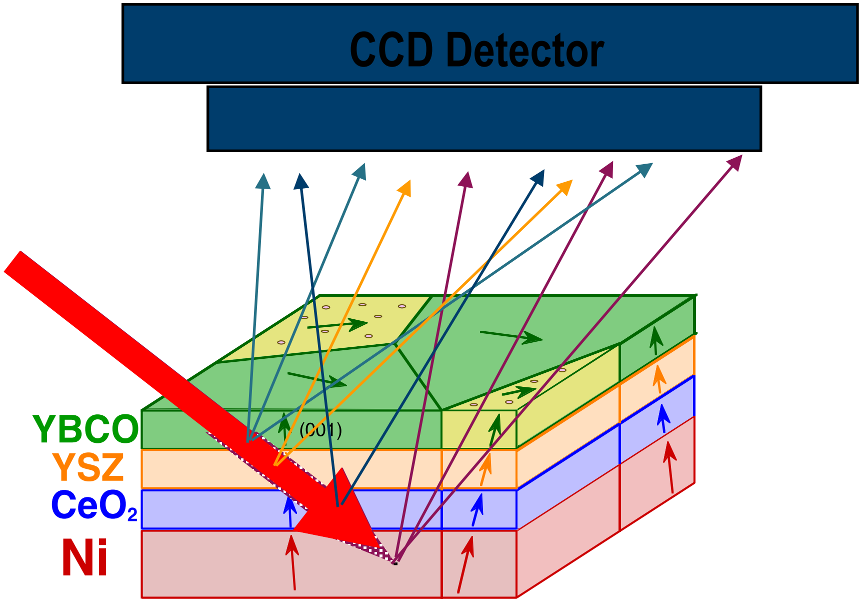
Grain-growth/ Budai et al. characterized epitaxial growth RABiTS

Optical: ~50μm grains



CCD Laue Patterns

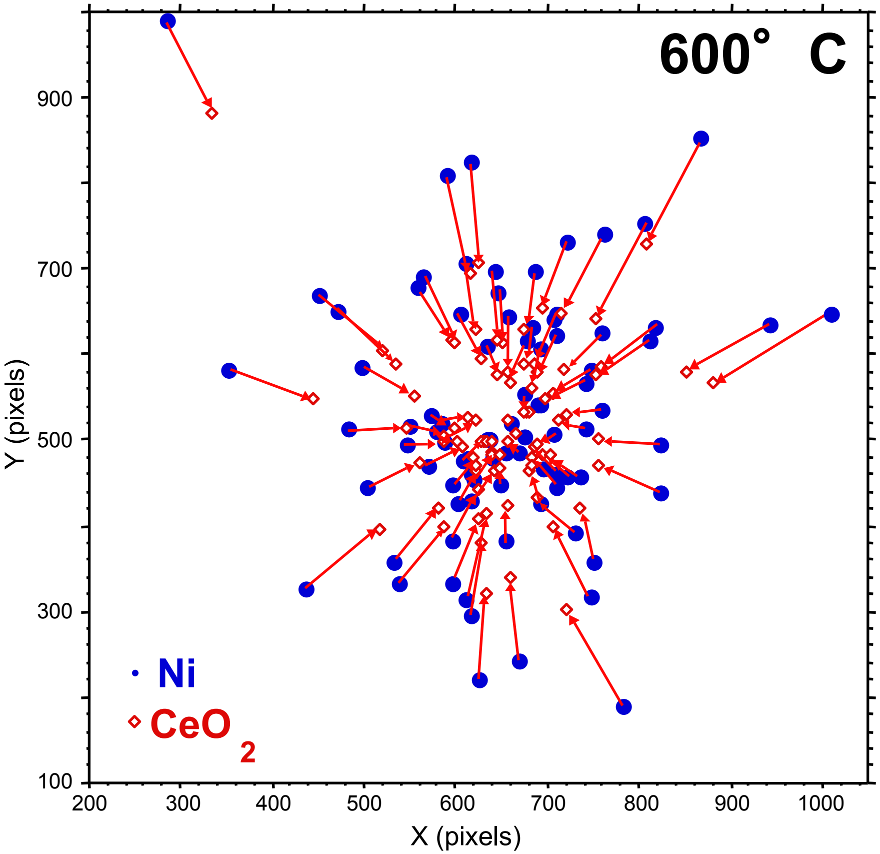


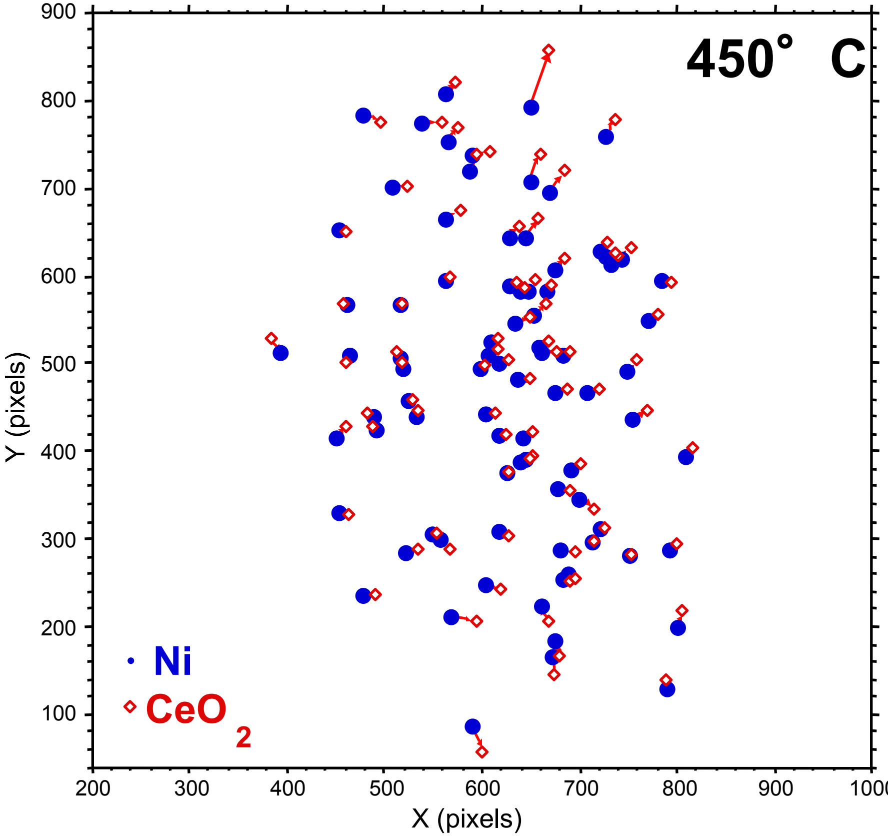


CeO$_2$ Observation:

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

Relative CeO_2 orientation depends deposition temperature





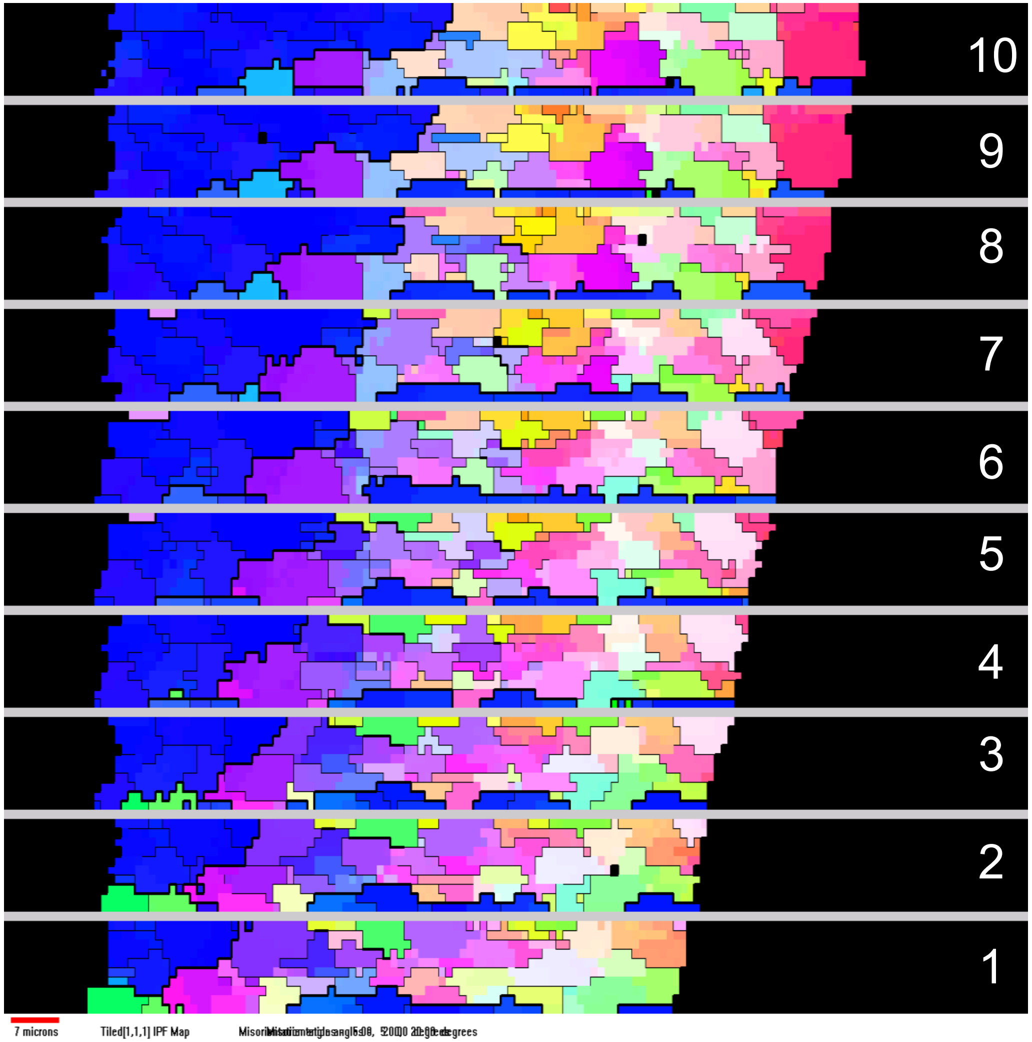
Step edge growth- good:

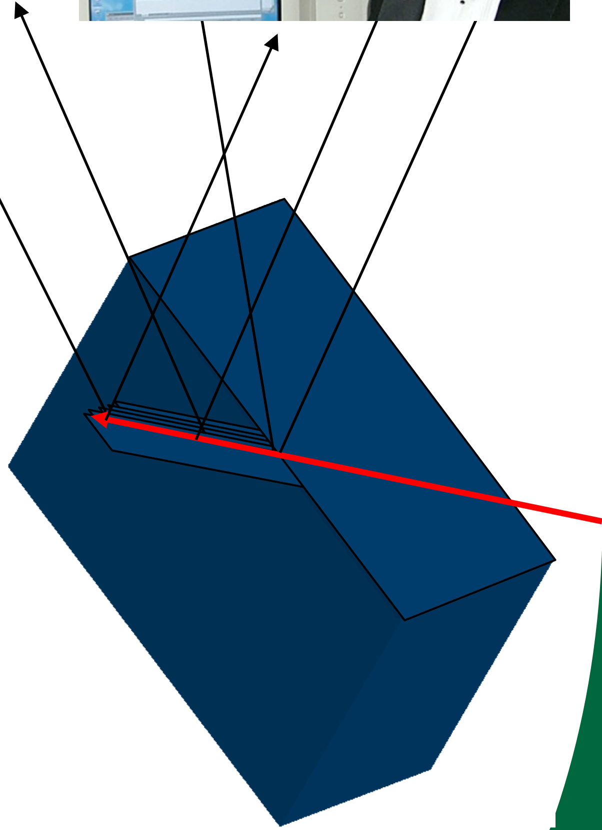
Crystallographic tilt towards ⊥

Tilt increases monotonically with miscut

Island growth-bad:

In-situ observations of 3D Grain Growth

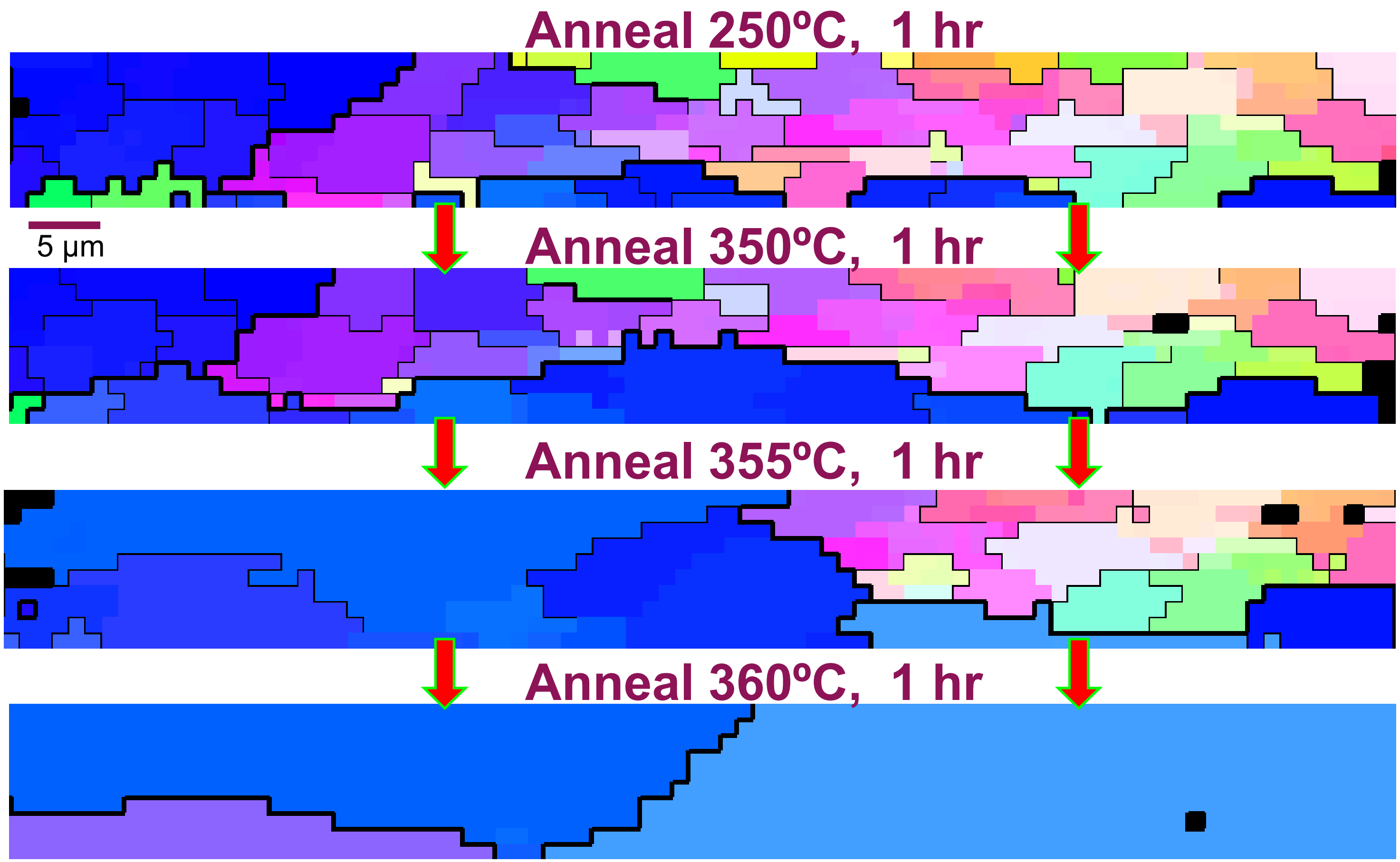




**Hot-rolled (200° C
1xxx Al(~1%Fe,Si)
Alcoa Polycrystal**

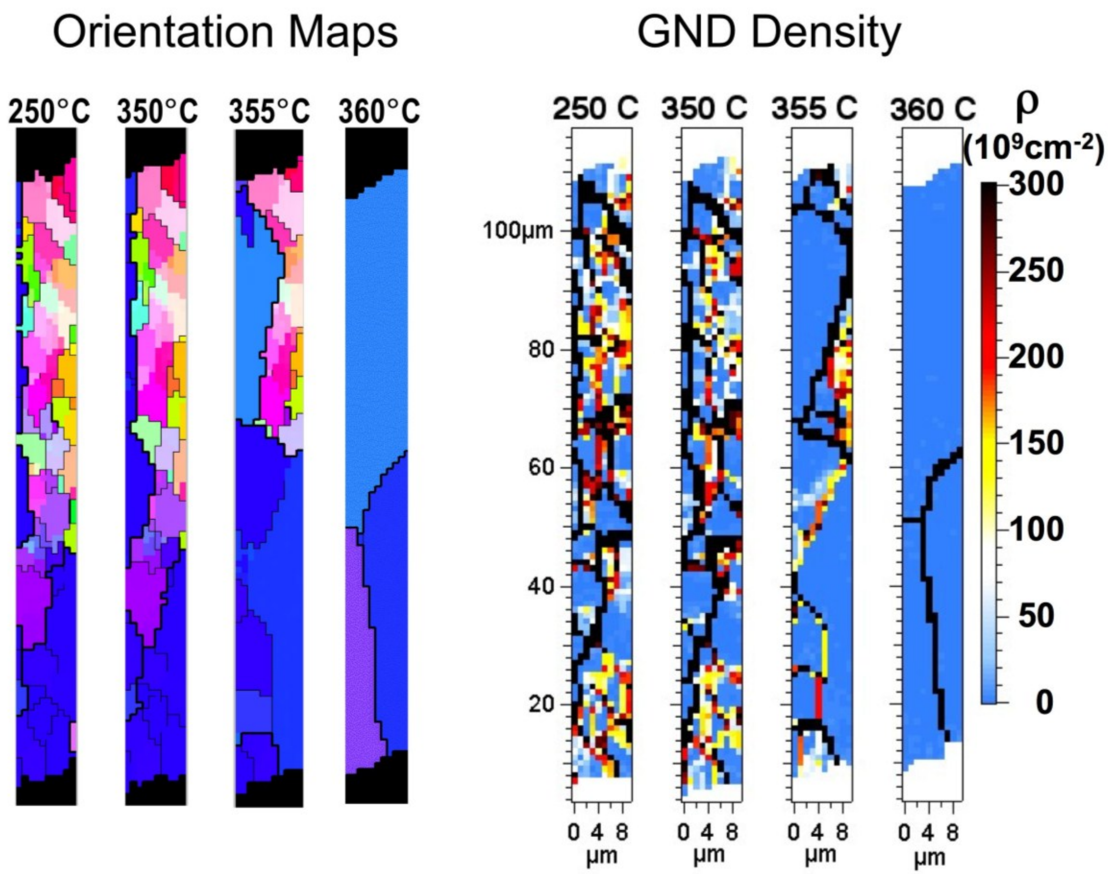
Thermal Grain Growth in Hot-Rolled Aluminum

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**

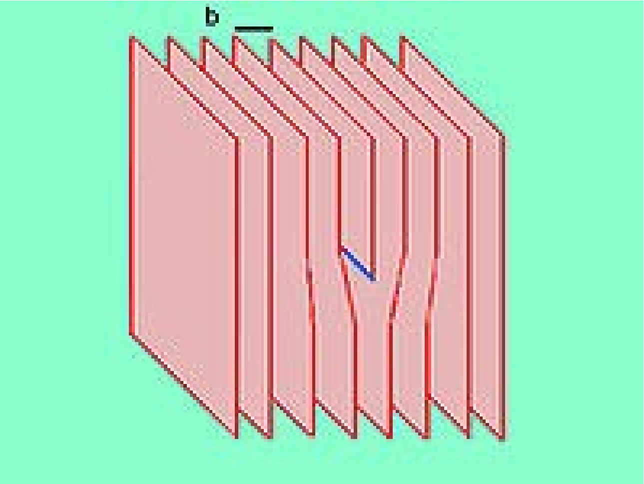
Deformation mediated by "dislocations"

- **Individual dislocations can be seen with TEM-but…**

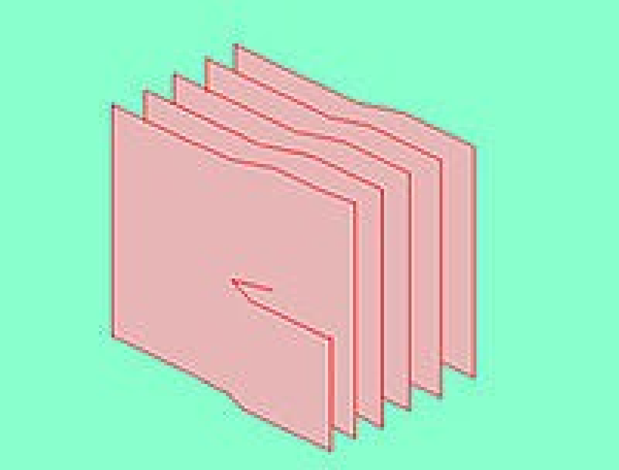
X-ray people complain thin electron samples Fundamentally different

Neutron people complain thin X-ray samples Fundamentally different

What is "thin" and "bulk"?

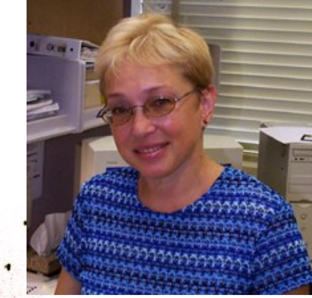


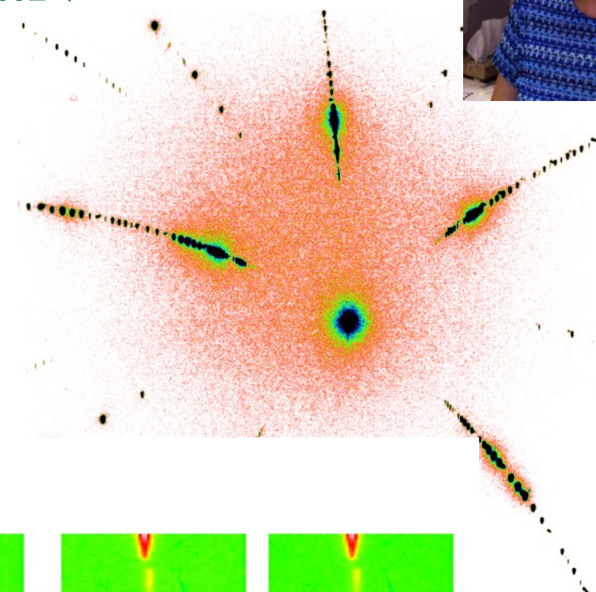
Edge dislocation



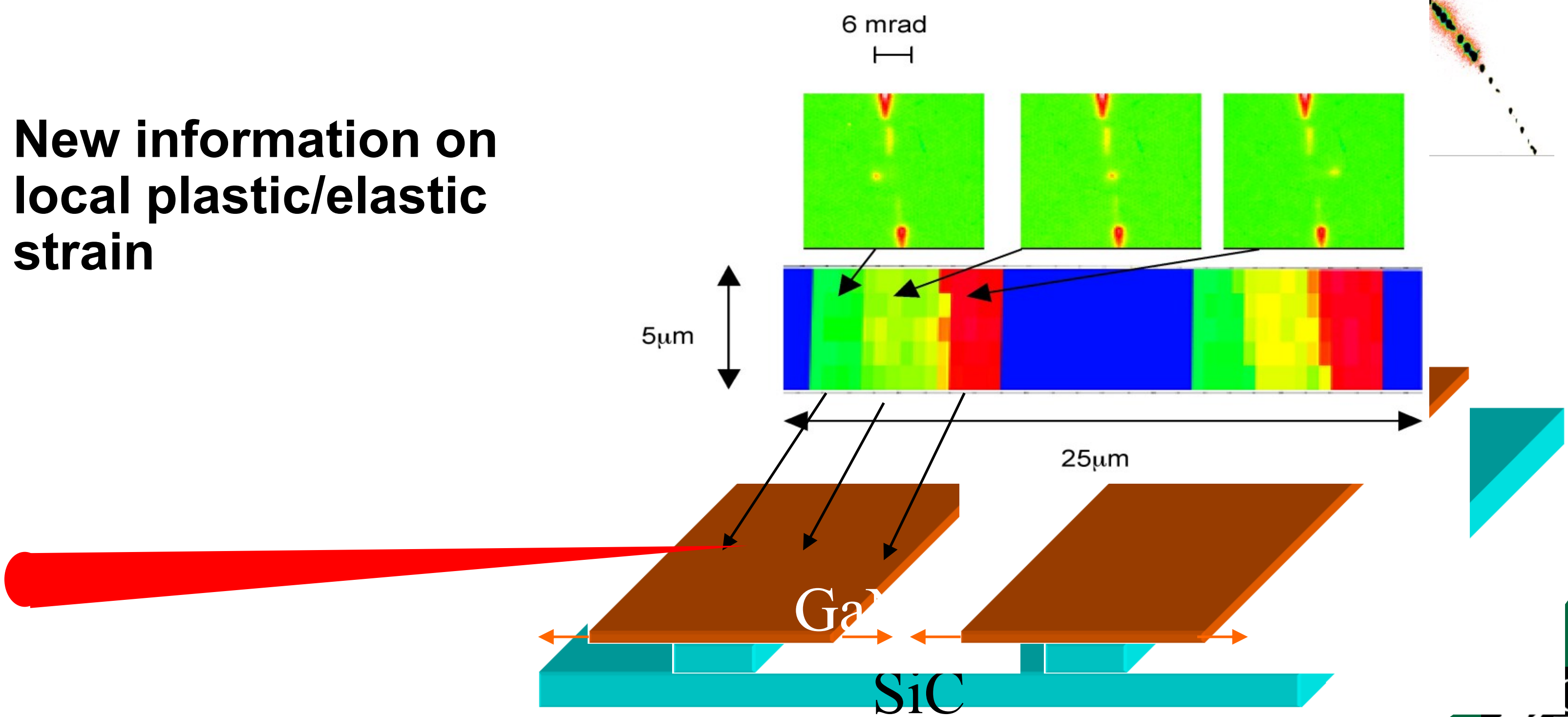
Screw Dislocation

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

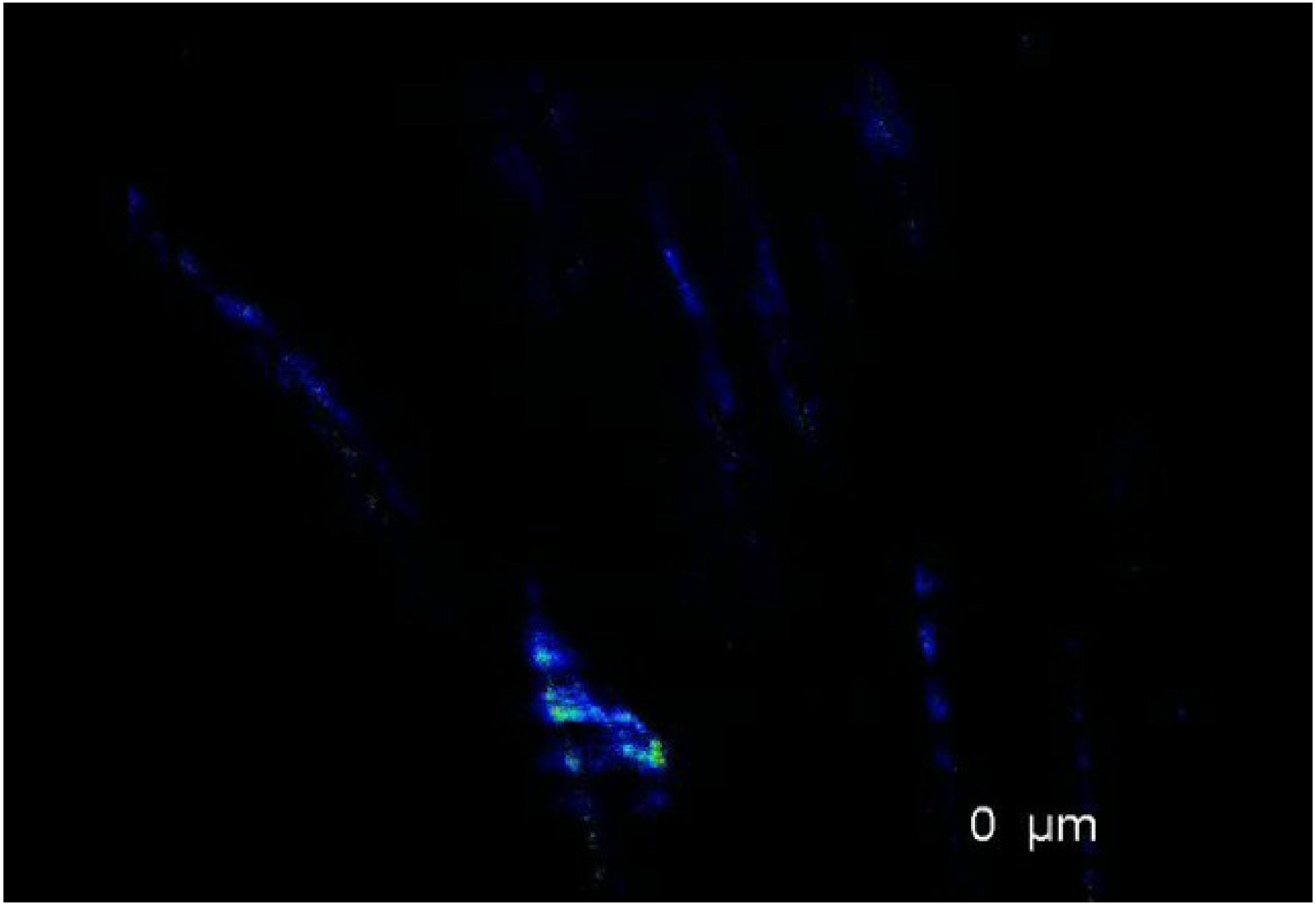




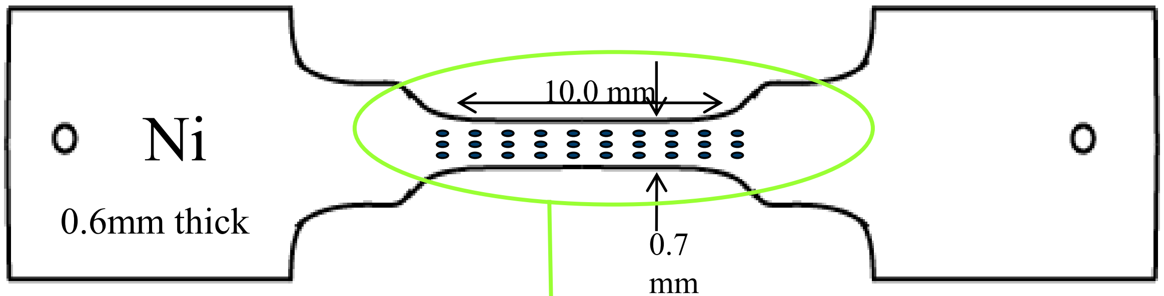
- **Reduced dislocations**
- **Direct confirmation of tilted wing structures**
- **New information on local plastic/elastic strain**

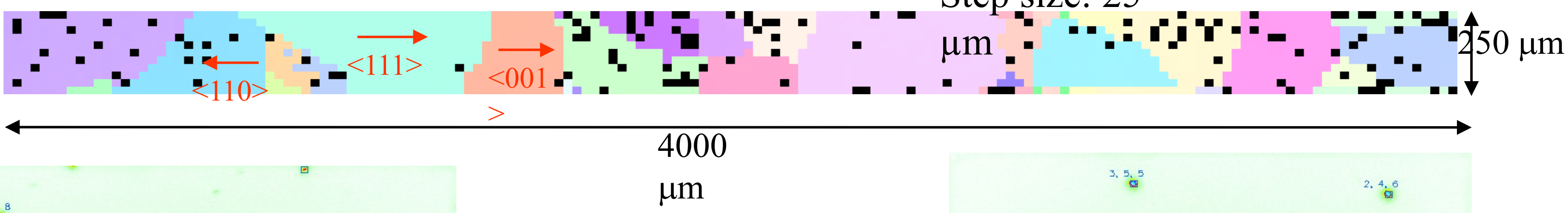


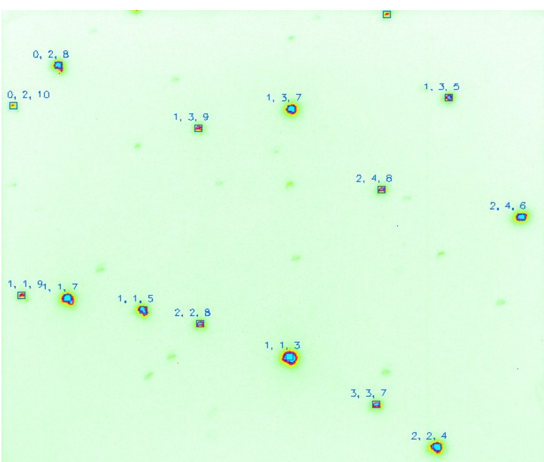
Deformation typically larger near surfaces/grain boundaries



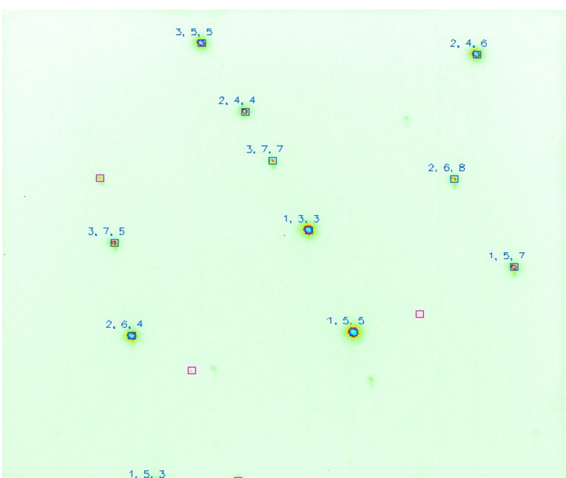
Deformation in polycrystals illustrates grain boundary behavior



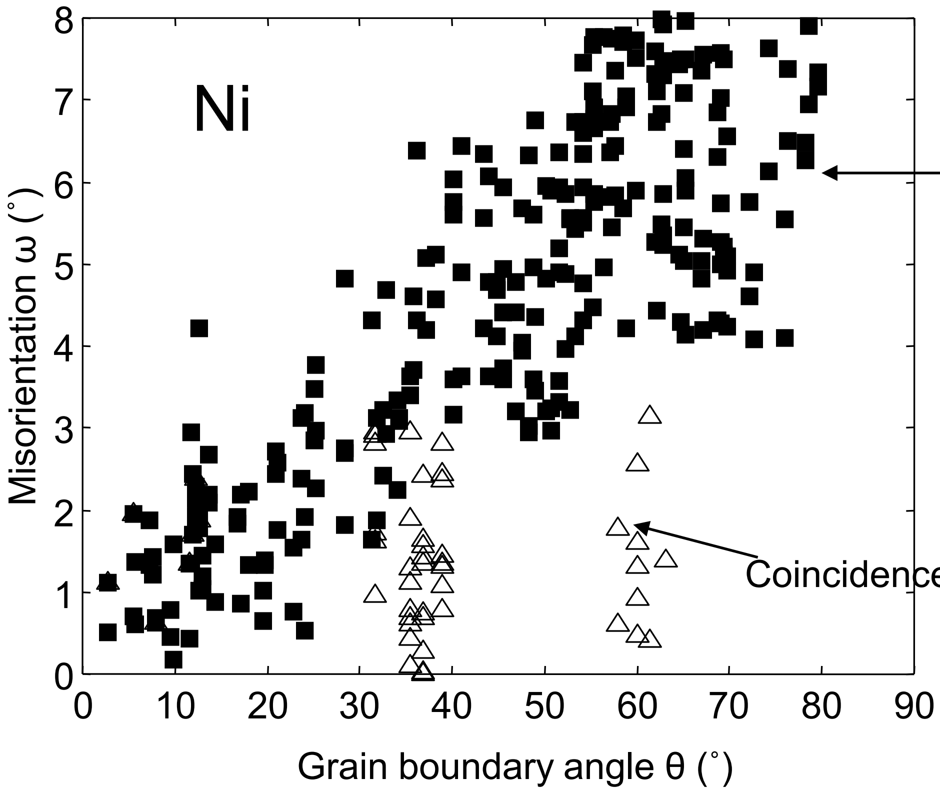




Indexation



Deformation induced rotations across grain boundaries sensitive to boundary type

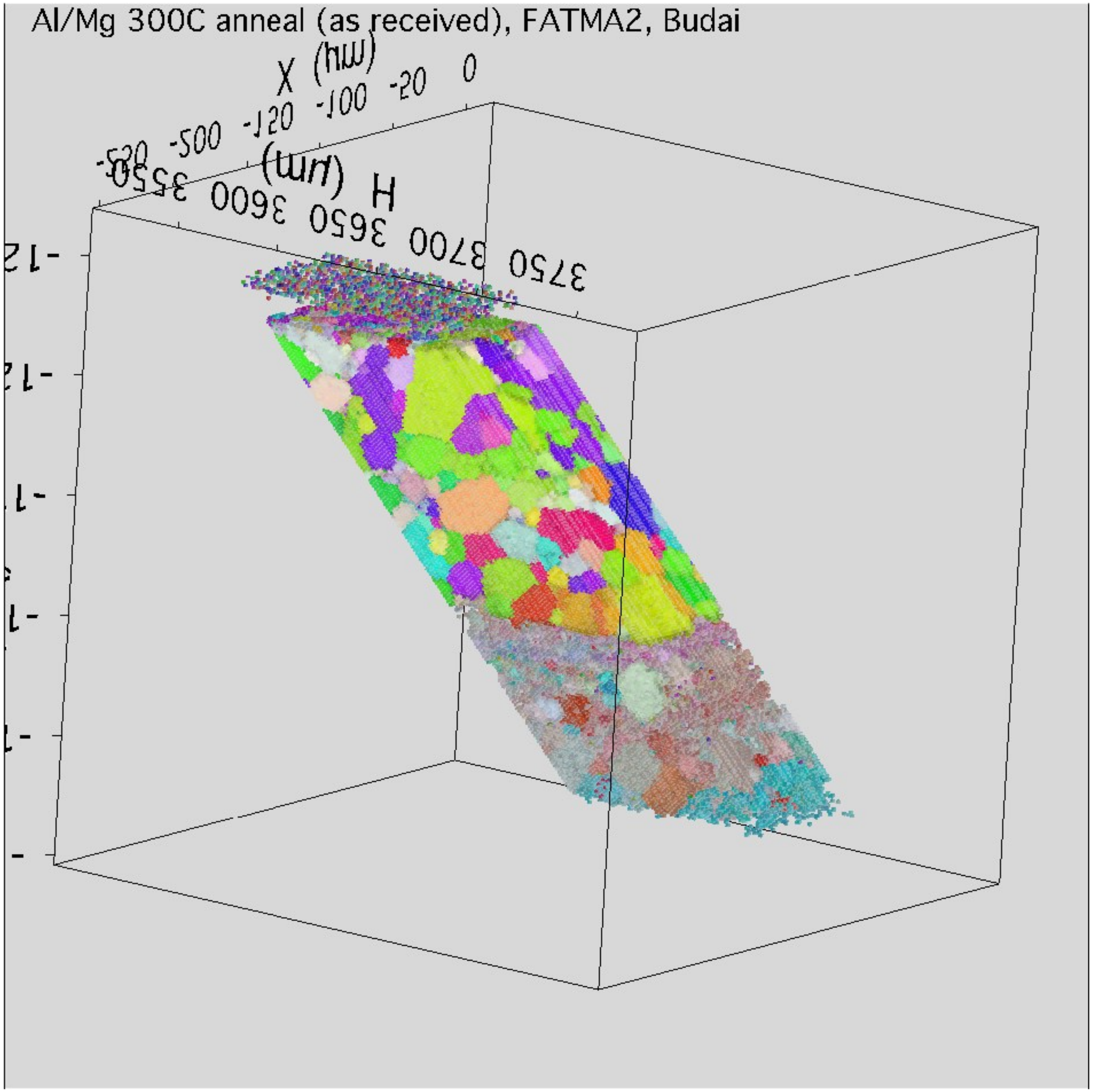


Normal high-angle grain boundaries

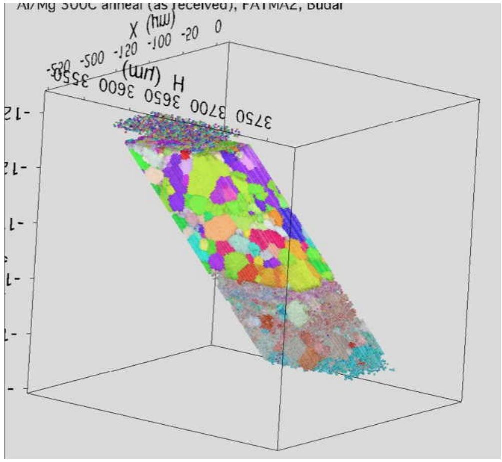
Coincidence site lattice

OAK RIDGE National Laboratory

Submicron spatially-resolved crystallography opens new opportunities



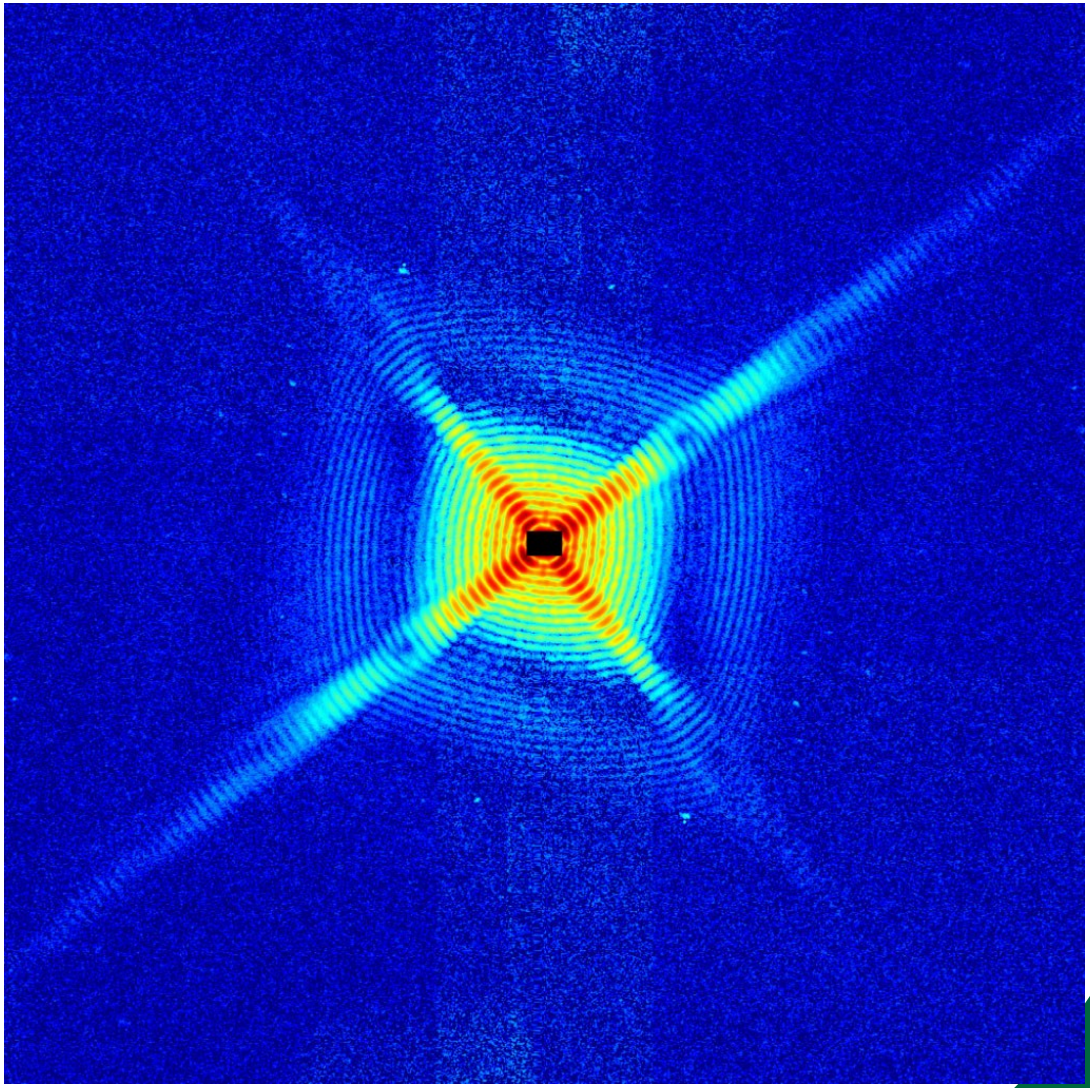
Do neural networks provide a path forward?



- **Visualization of 3D tensor data unsolved problem**
- **Human mind well adapted to 3D visualization based on binocular 3D data.**
- **Neural network not "limited" by streamlined processing of human brain**

Coherent diffraction offers promise for atomic resolution with focused beams

- Focusing
  - Better spatial resolution
  - Poorer field-of-view
- 2 nm with 3rd generation source and 1 µm focal spot
- 2 Å with 10 nm spot- or 4th generation source

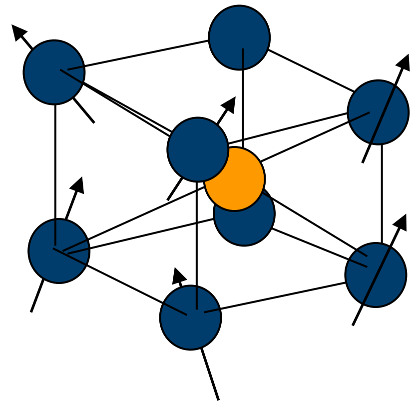


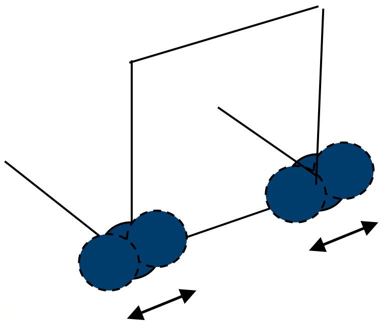
Neutron microdiffraction additional opportunities

Magnetism

Atomic motions

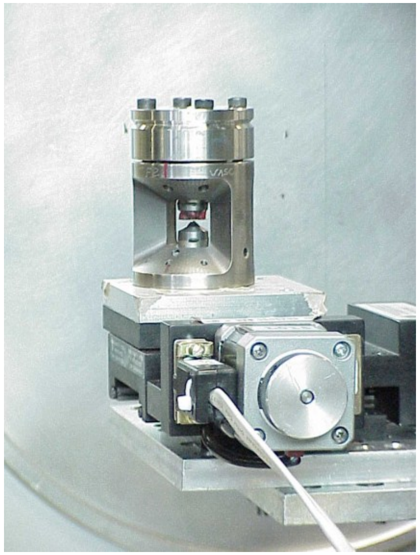
Low Z materials



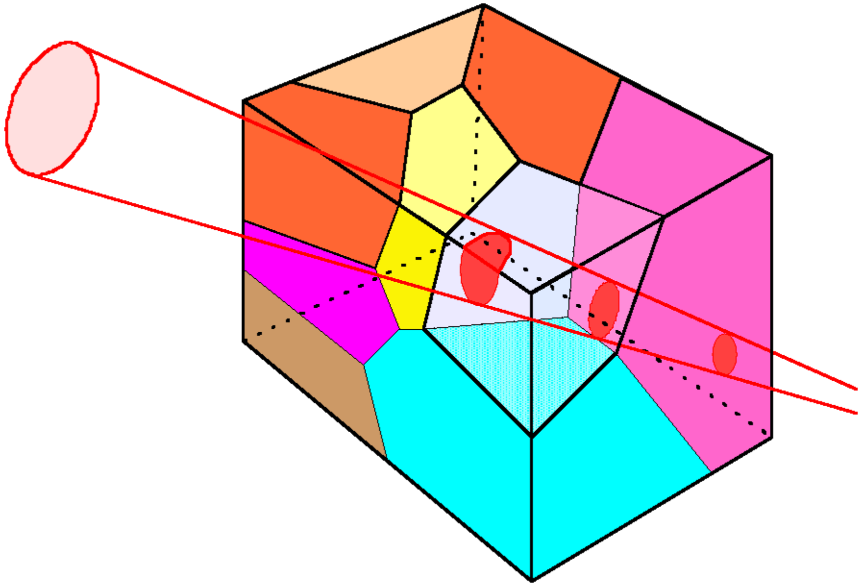


Nobel prize to Shull and Brockhou

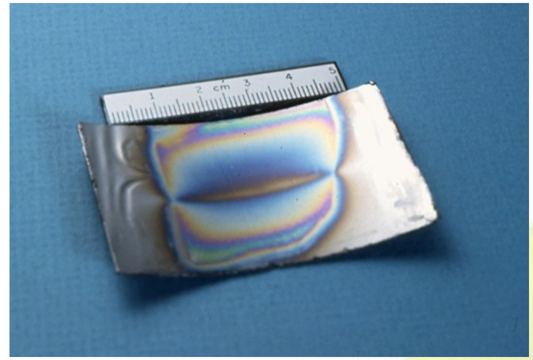
Focused beams extend neutron science

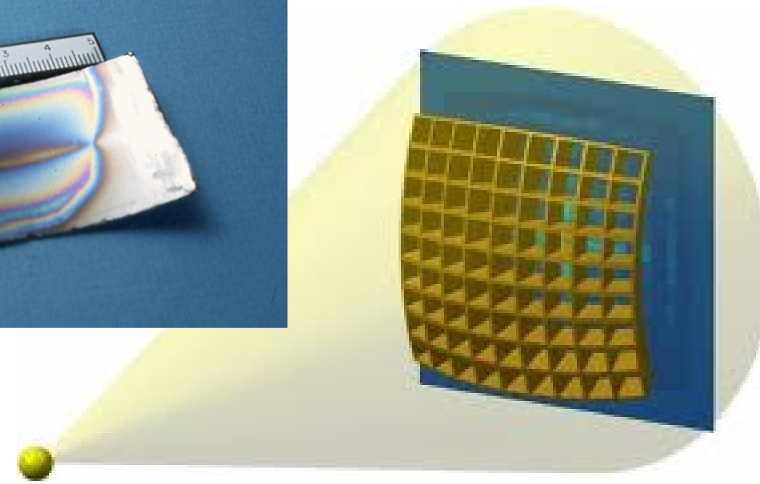


- Inhomogeneous samples
- Small samples in environmental chambers
- Spatial resolved distributions deep in samples

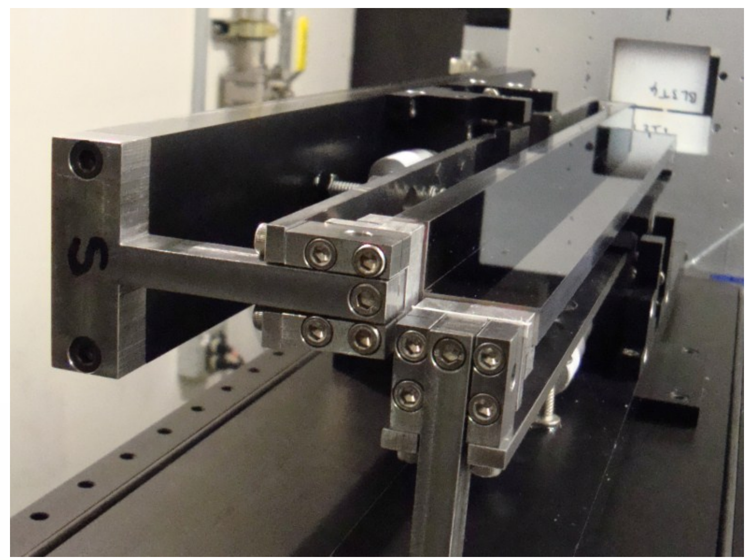


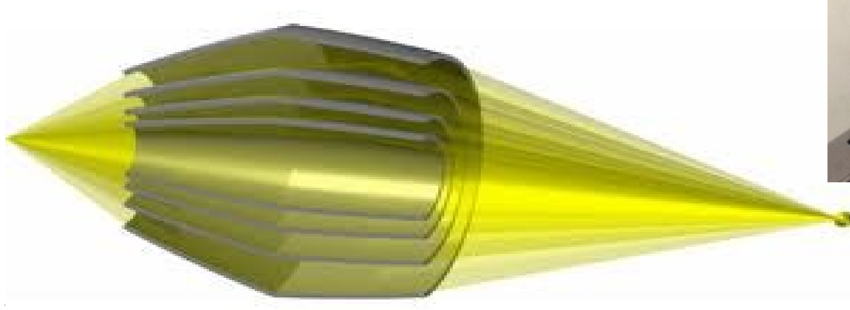
Neutron microfocusing optics also evolving





- Sagittal focusing optics < 300 μm
- Lobster eye optics ?
- NMM <100 μm
- Wolter optics<200 μm





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

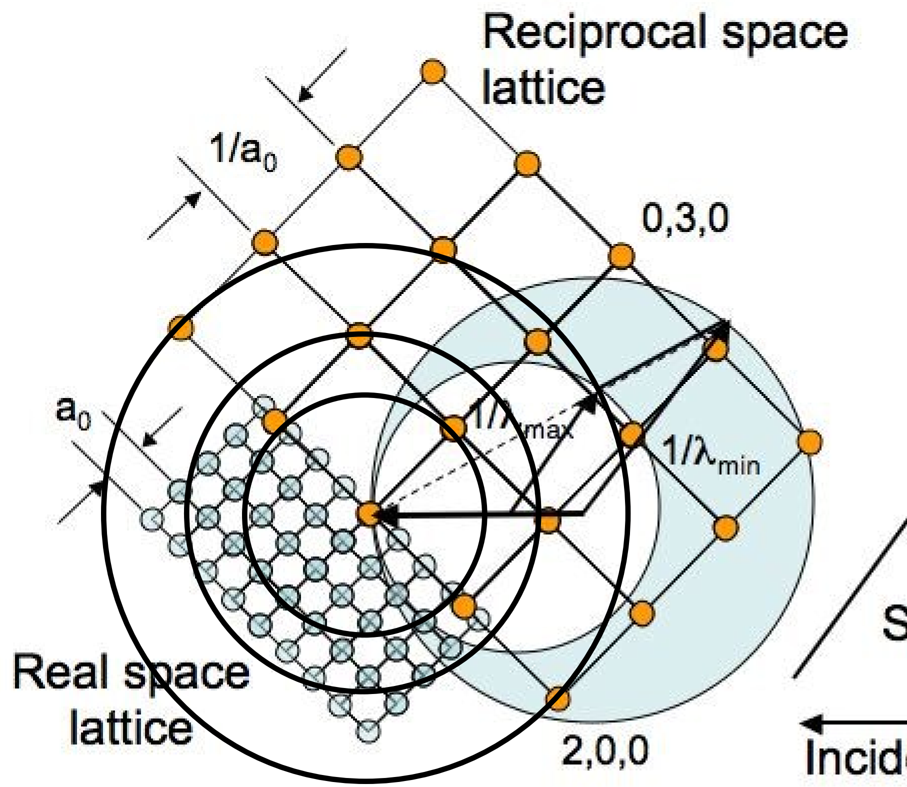
10^{-3}\$ /neutron

Increase divergence/bandpass

10^{-9}\$/ neutron

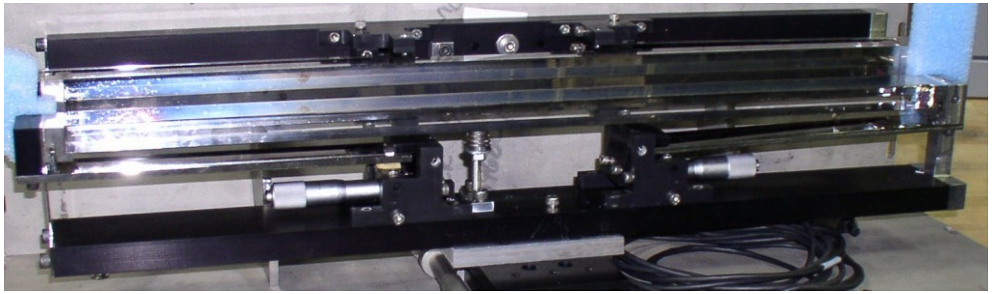
Spallation neutron science intrinsically polychromatic

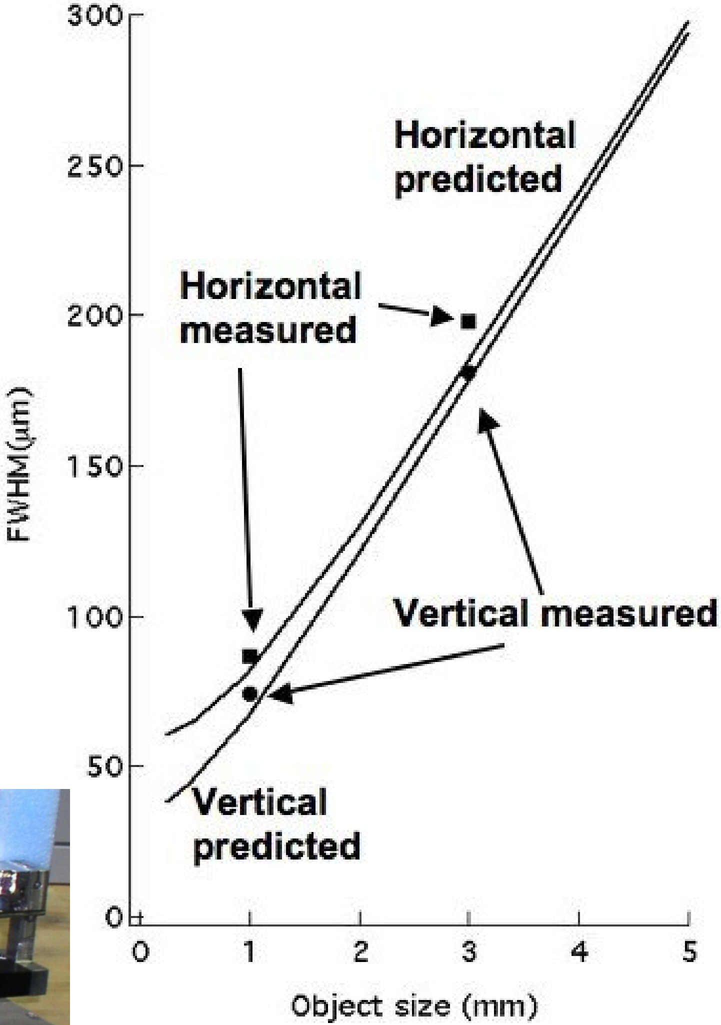
- **Analogous to polychromatic X-ray microdiffraction-*but includes* energy**
- **Allows for structure determination**
- **Absolute strain measurements**



SNAP experiments diffraction high pressure cells

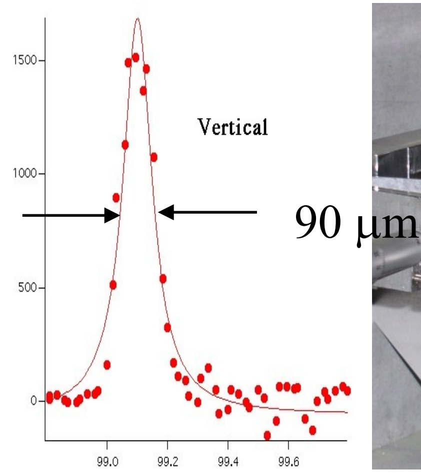
- **Focusing optics work near theoretical limit**
- **Minor improvements should enable 25 micron measurements**

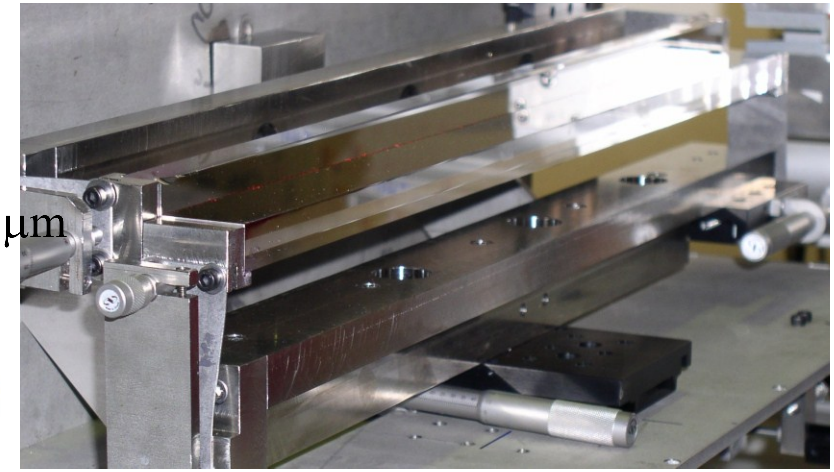


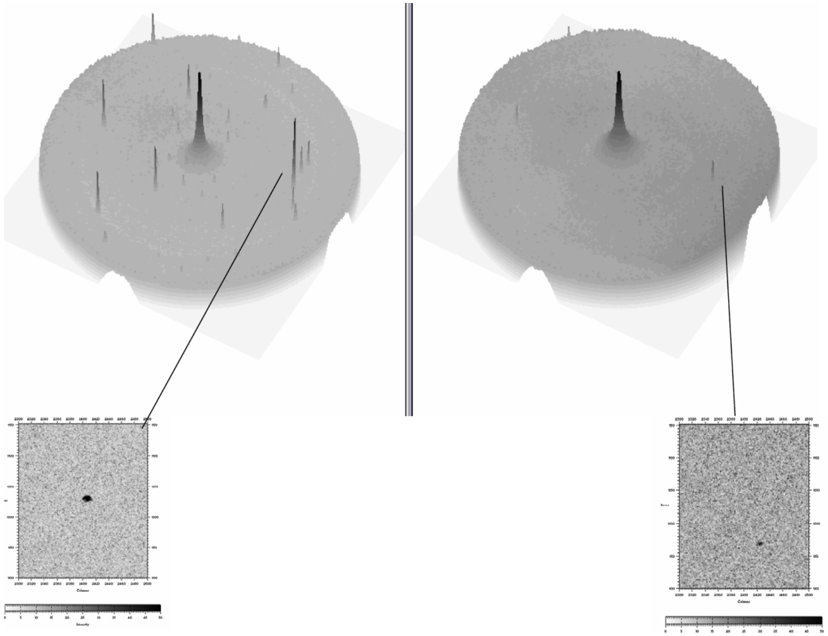


Neutron mirrors produce microbeams

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



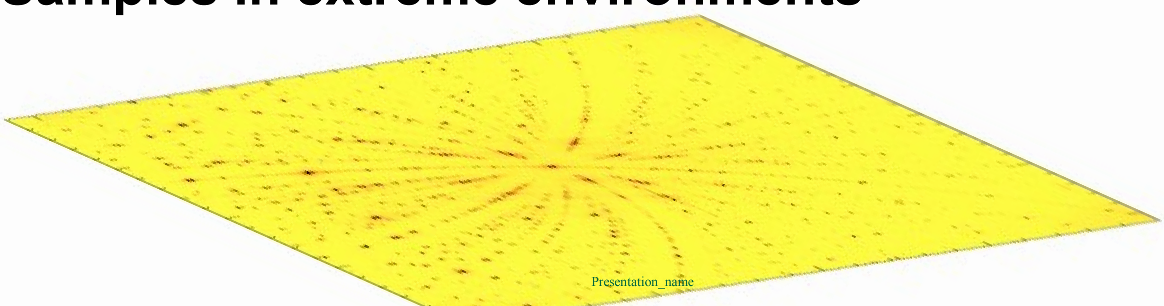




Useable 25 μm beams?

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**



Materials structure tiny- intrinsically 3D
And spatial resolution- is needed urgently
The frontiers moving quickly now-excitements in the air
Though ask the average person- they really couldn’t care

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