Engineering Neutron and X-Ray Diffraction

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Additive Manufacture is of Particular Interest to the Nuclear Weapons Complex

- AM is most efficient when applied to manufacture of small lots of complex geometries with minimal footprint and overhead investment…
  - …once the process is qualified.
- AM can be agile and produce single parts for specific applications or experiments.
- Can we design specific functionality into components?
- But how do you qualify components made with a completely new technology?
- Our team is trying to play its roll in this task by using neutron and x-ray scattering techniques to monitor microstructure throughout lifetime of a component.
- So called Process/Structure/Properties/Performance relationship.

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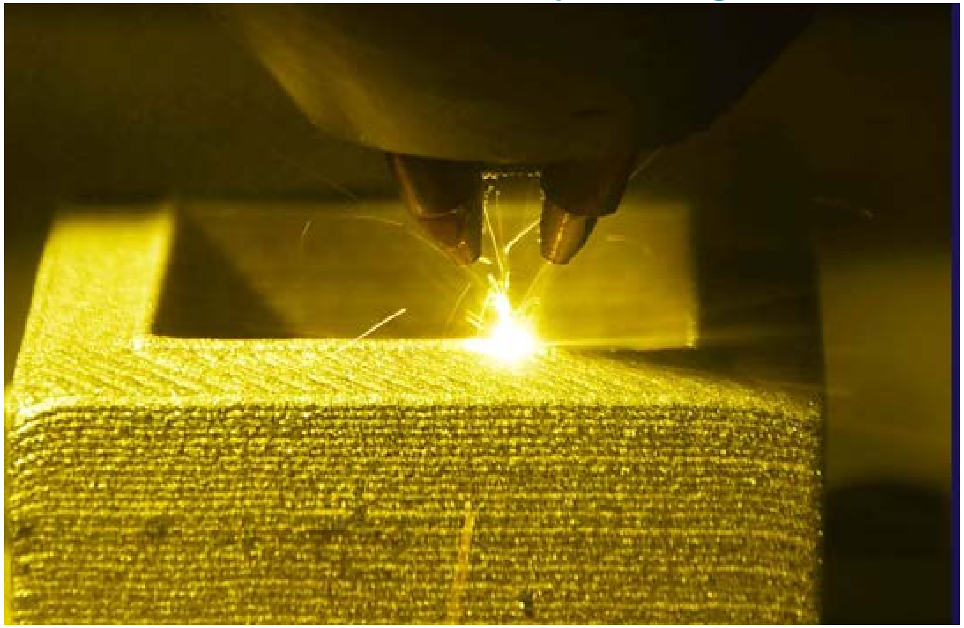
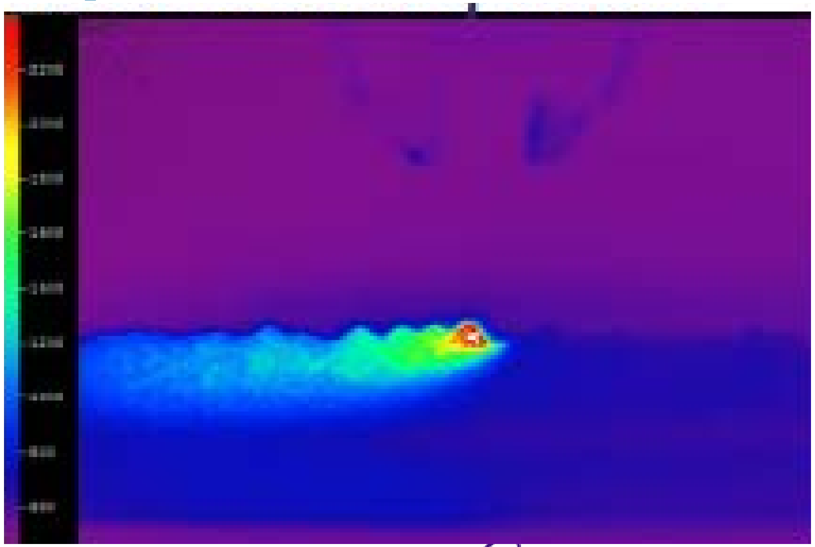
AM Inherently Spans Several Orders of Magnitude in Length and Time Scale.

- **Quench rates following deposition can reach ~10^6K/sec, pushing for phase quantification on µsec time scales.**
- **As material is subsequently added, temperature excursions can last as long as seconds.**
- **Post-build heat treatments last ~hour.**
- **Residual stresses build up over the duration of the build, hours.**
- **Their effect on performance can manifest in days-years.**
- **Dislocations, which control the material strength, exist on the atomic scale (Å).**
- **Phase topographies and grains are on ~µm scale.**
- **Powders are typically 10's of µm.**
- **Residual stresses are set by the length scale of the component, i.e. cm's.**





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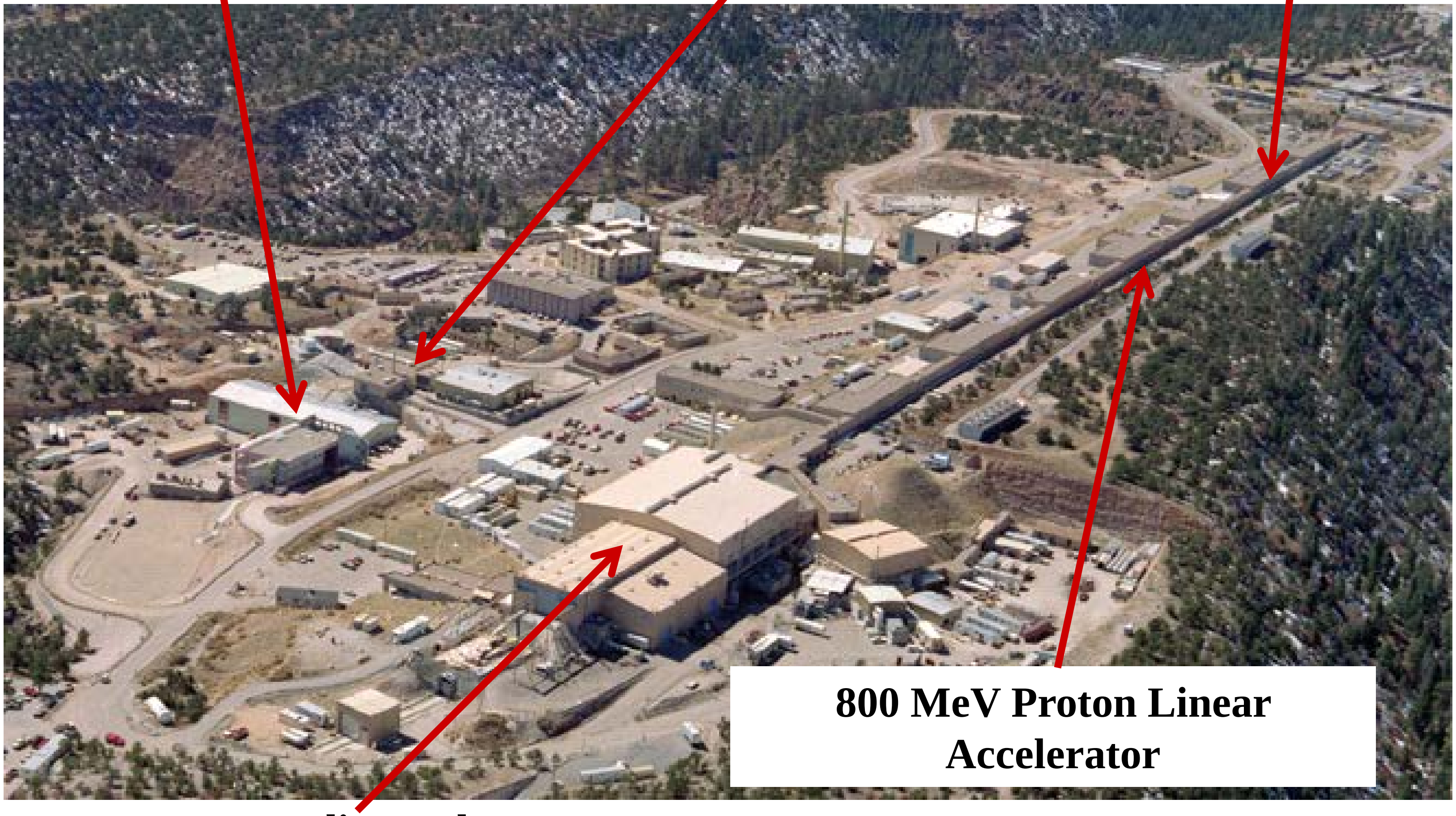
Neutron Sources Do Not Sit on Desktops !

Los Alamos Neutron Science Center : LANSCE

Lujan Neutron Scattering Center

Weapons Neutron Research Facility

Isotope Production Facility



Proton Radiography

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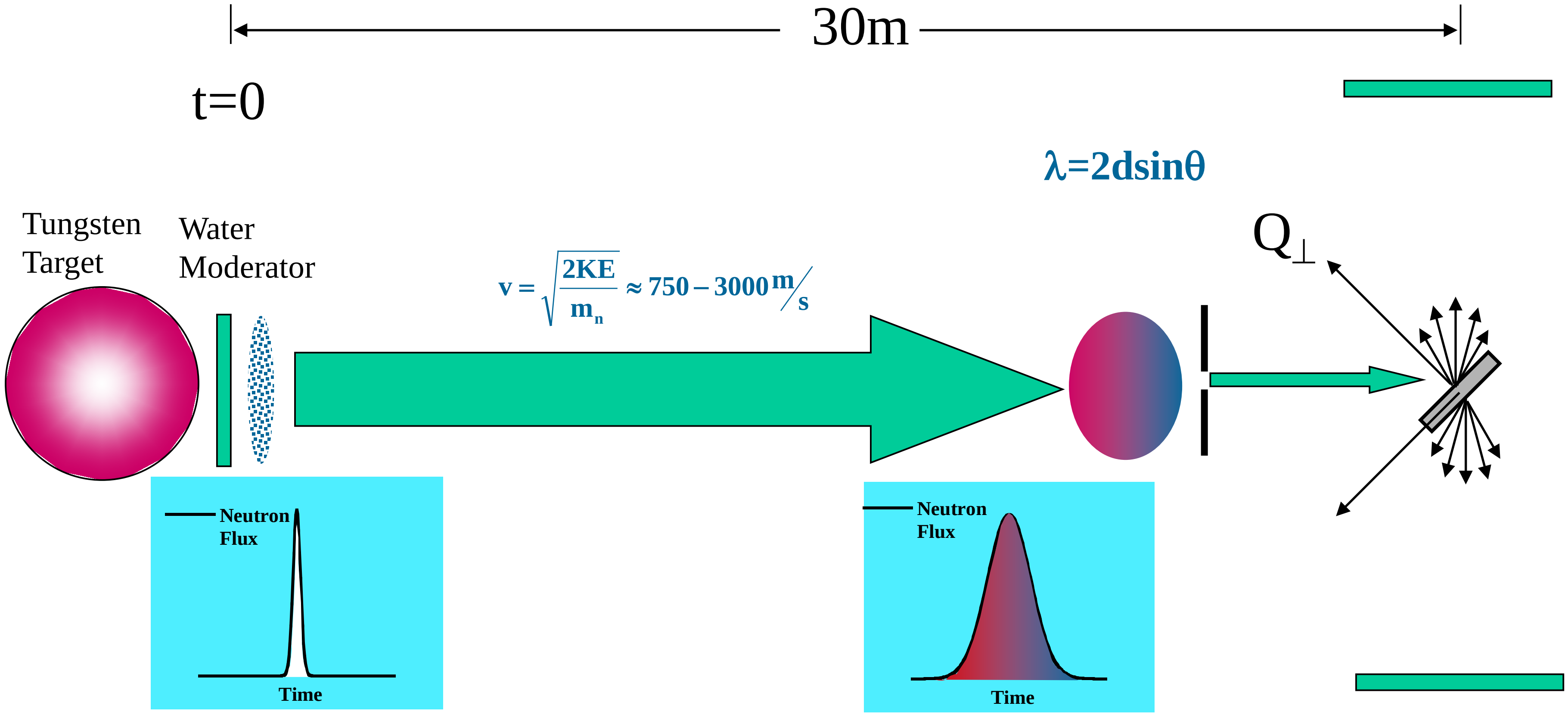
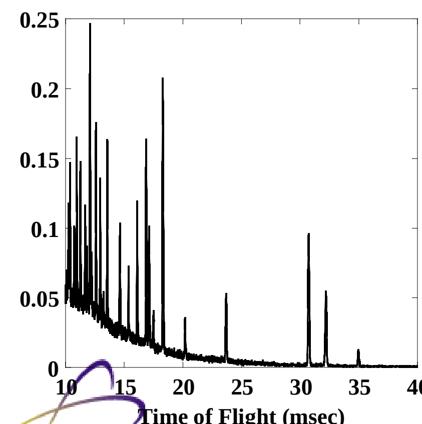
Time of Flight Diffraction at a Spallation Source



- Velocity (and λ) determined by the neutron time of flight.
- Entire diffraction pattern collected with unique diffraction vector.



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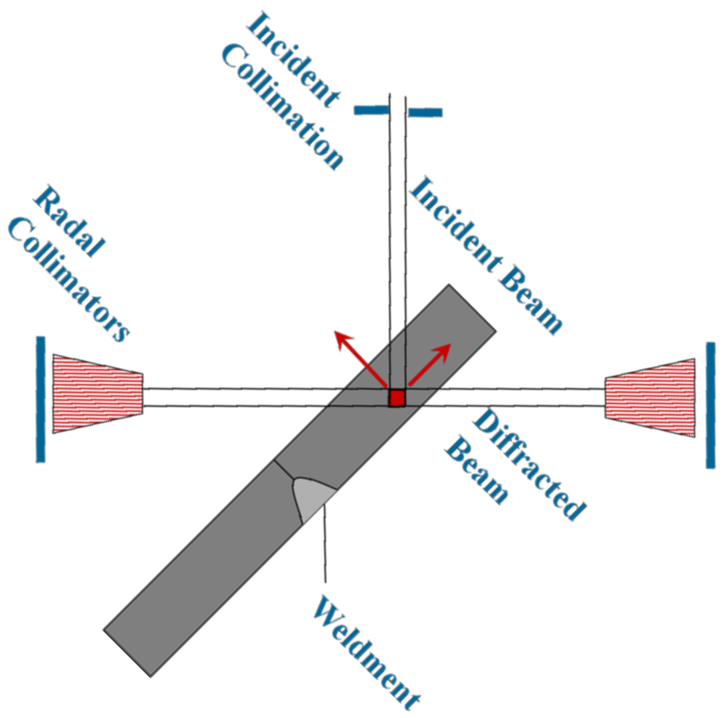
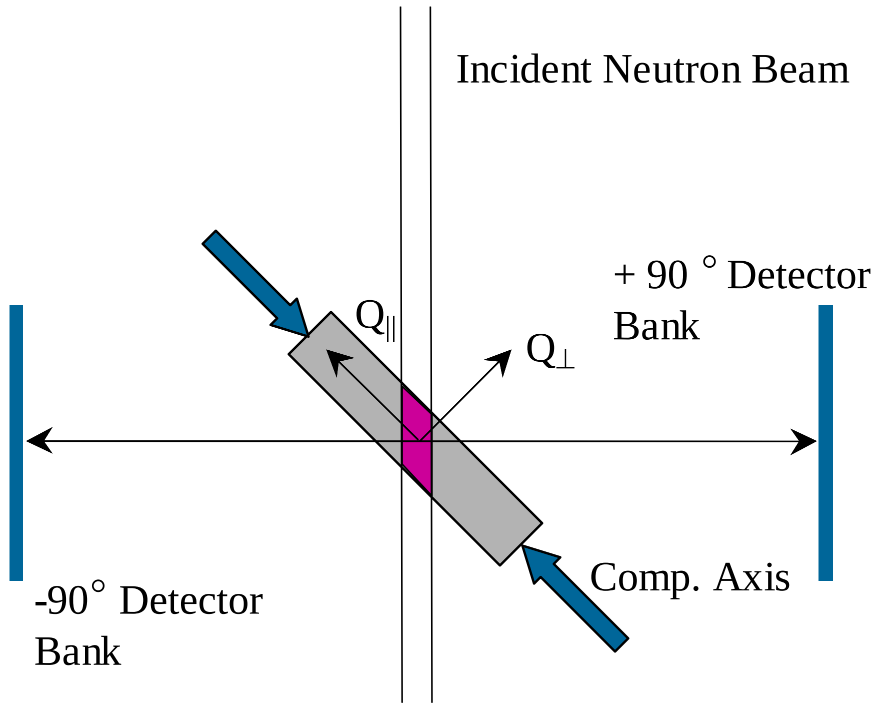
Two Types of Measurements





- **Residual stress measurement:**
  - **You know the stiffness tensor and want to determine unknown stress**
- **Lattice response to external stimulus:**
  - **You control the stresses and you want to learn about the springs (bonds)**

Lujan Scattering Team Utilizes Other Tools As Appropriate

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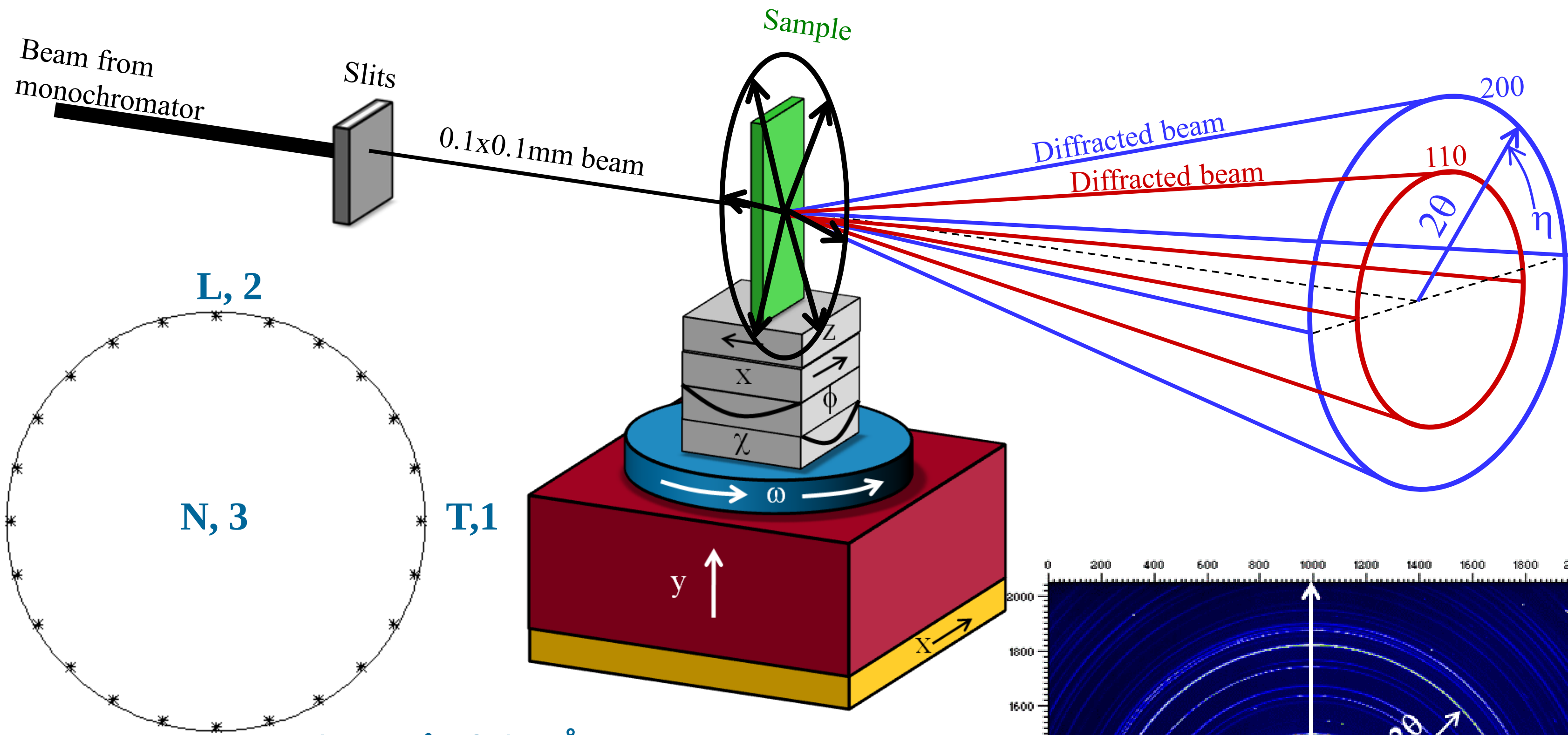
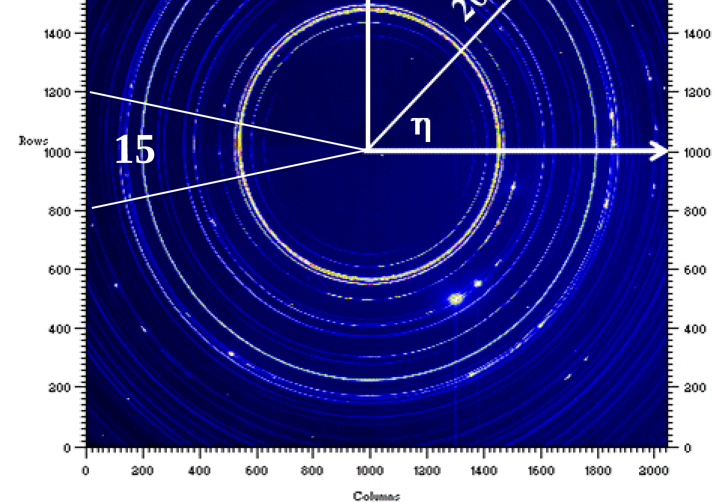
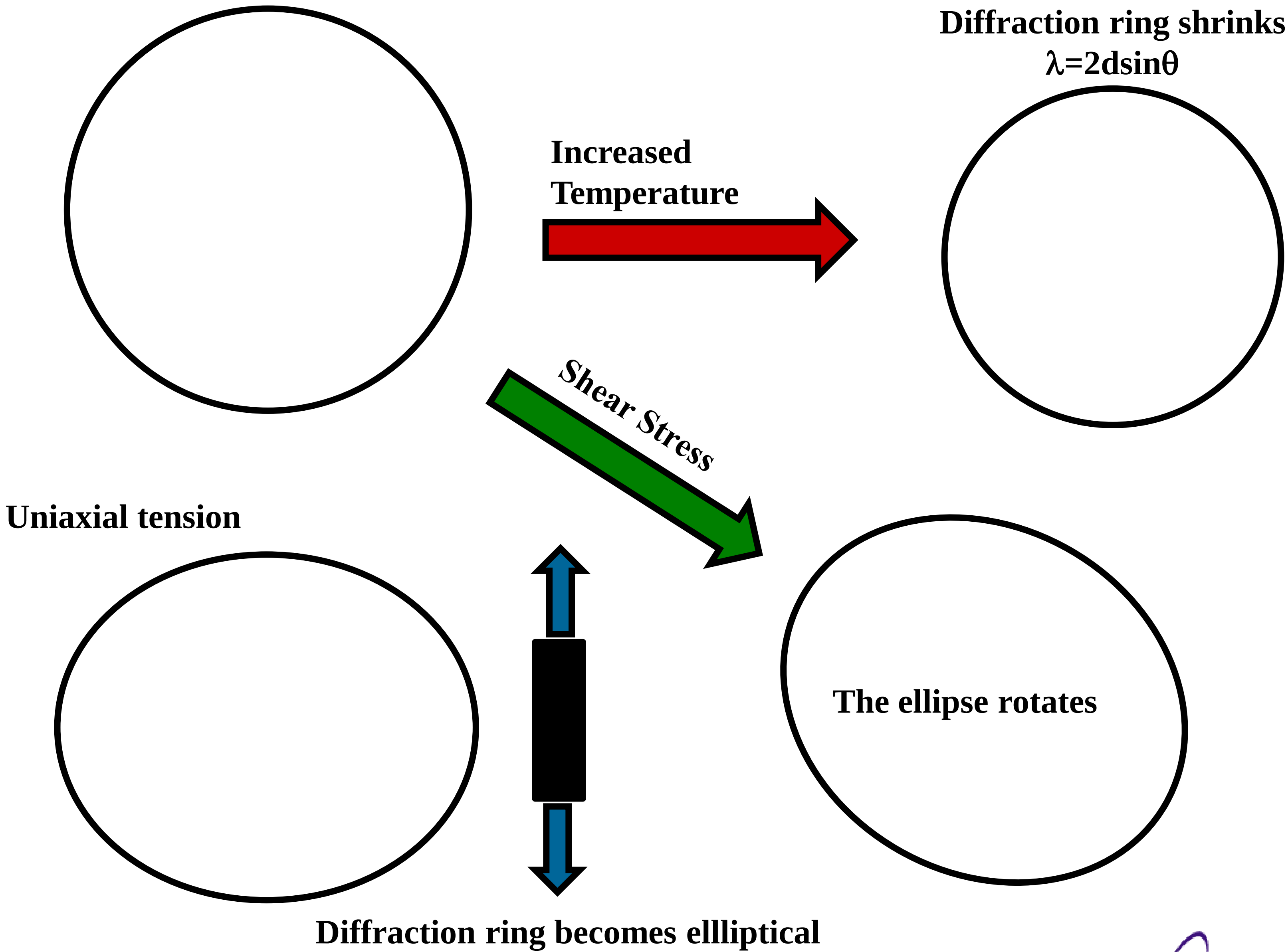
High Energy X-Ray Diffraction Probes the Bulk of the Material





- X-ray energy = 71KeV, λ=0.175Å.
- Debye cones provide diffraction vectors nearly normal to the incident beam.
- No depth resolution
- Integrate rings to get an average response.
- Bin (cake) the data over twenty-four 15deg intervals to provide orientation dependence.

Temperature And Stress Have Distinct Effects on the Diffraction Rings



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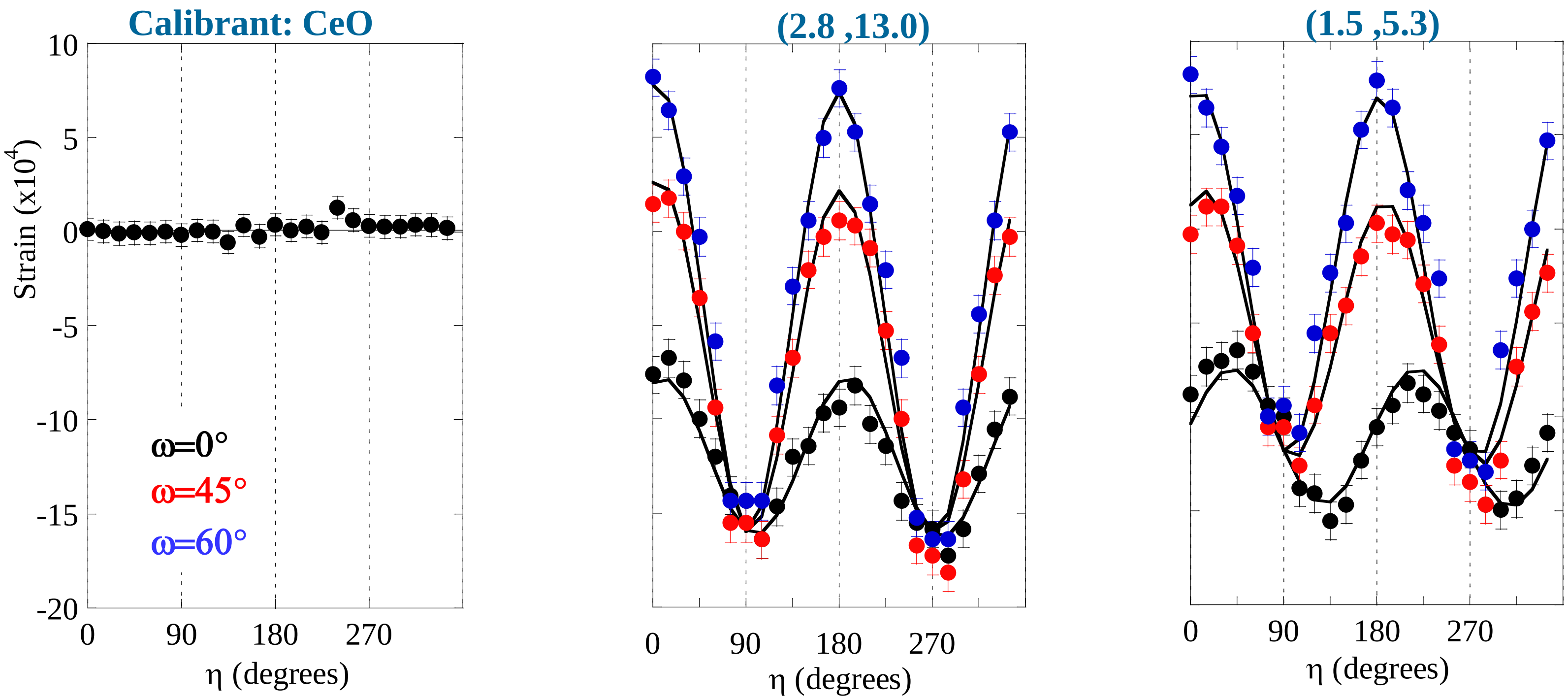
Example of Residual Stress in U10Mo Thin Foil



$$\varepsilon = \begin{Bmatrix} -8.1 & 1.4 & -0.5 \\ 1.4 & -16.0 & 1.0 \\ -0.5 & 1.0 & 12.8 \end{Bmatrix} \times 10^{-4} \pm \begin{Bmatrix} 0.3 & 0.3 & 0.3 \\ 0.3 & 0.2 & 0.3 \\ 0.3 & 0.3 & 0.4 \end{Bmatrix}$$

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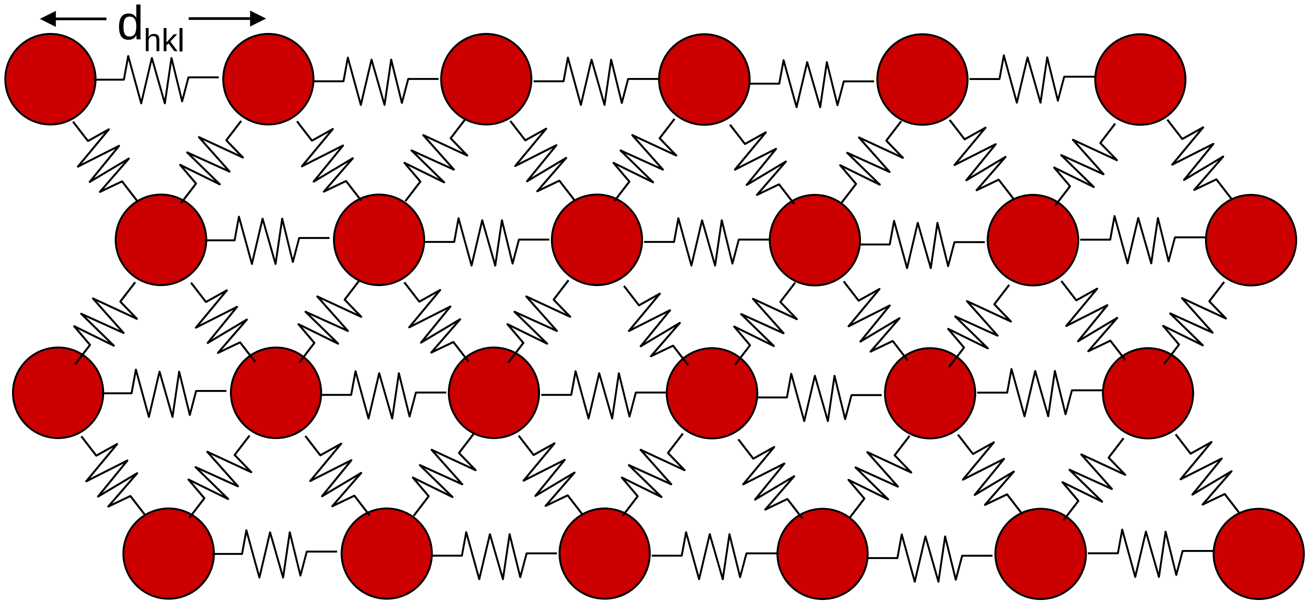
We Use Diffraction Like A Bathroom Scale



• We measure the spacing between atoms very precisely: ~1 part in 10^5.

• Calculate lattice strains (elastic) from change in atomic spacing.

$$\varepsilon = \frac{d_{hkl} - d_0}{d_0}$$

• Generalized Hooke's Law : $\sigma_{ij} = C_{ijkl}\varepsilon_{kl}$

• It is important to note that the lattice strain is necessarily proportional to the stress on the grain, not the macroscopic stress.

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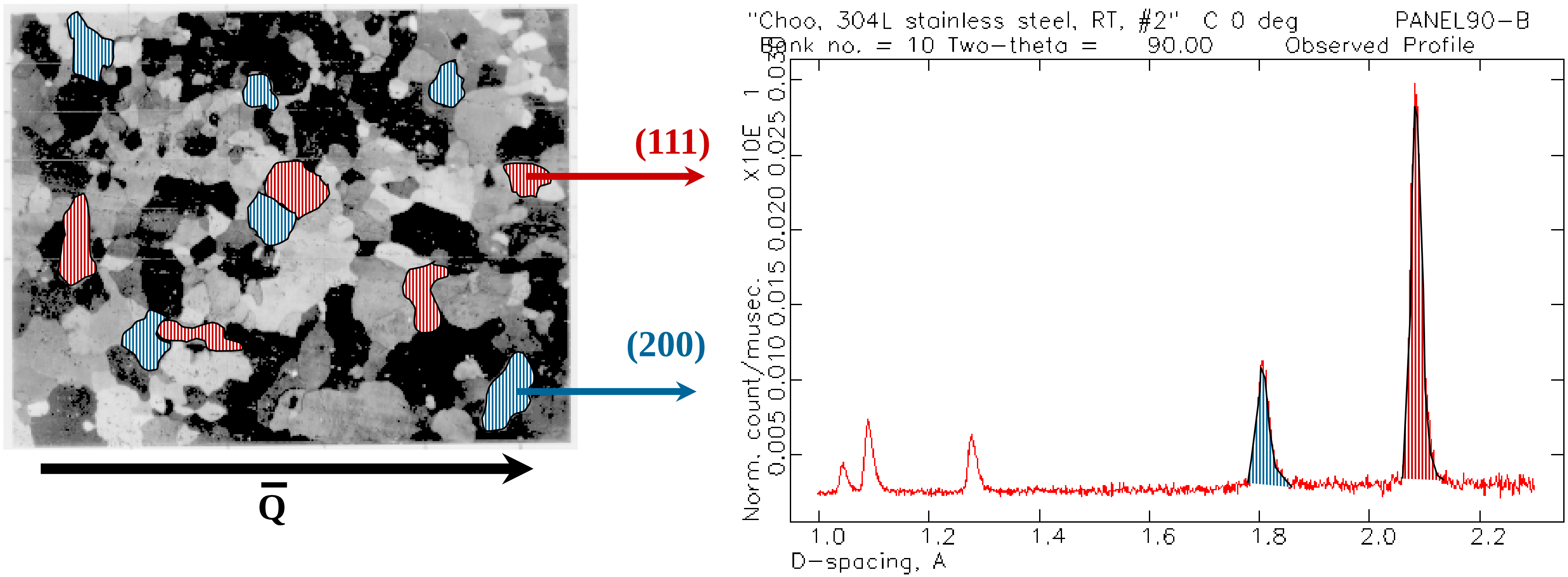
Diffraction Separates Response of Grain Orientations

Polycrystalline Aggregate | **Stainless Steel**



- **Grains with plane normals parallel to the diffraction vector defined by the instrument geometry diffract into a detector.**
- **Each grain orientation (hkl), or phase, contributes to a distinct peak, given by the interplanar spacing.**
- **We explicitly make the assumption that a family of grains can be used to represent the macroscopic stress field.**

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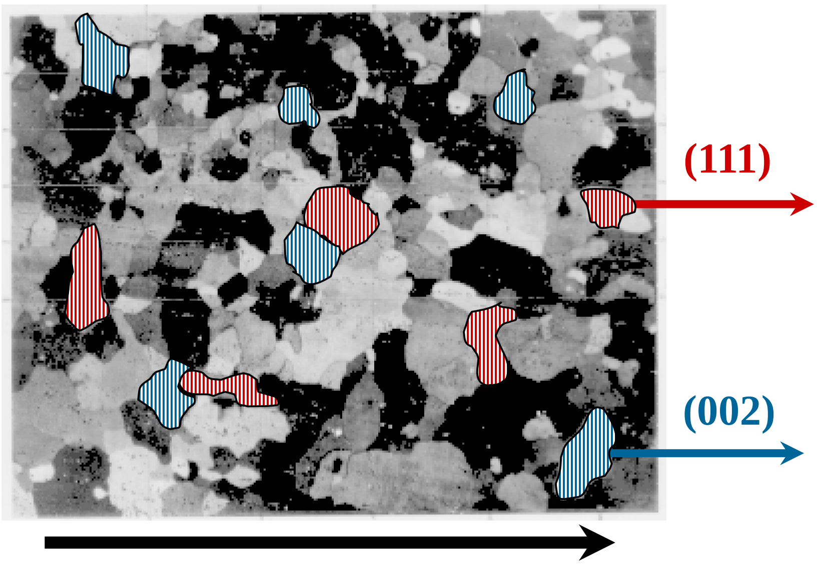
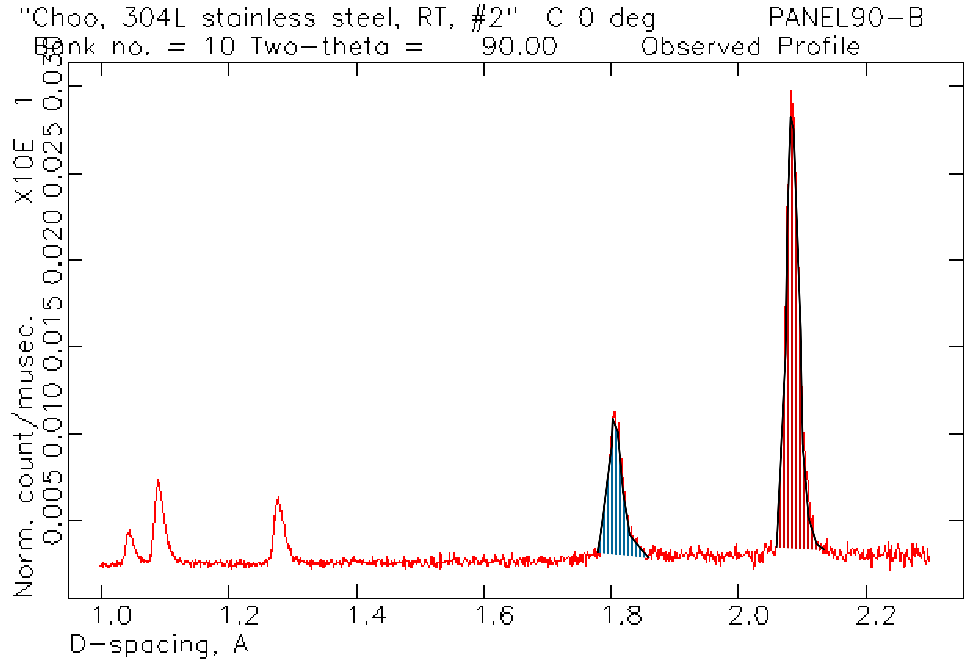
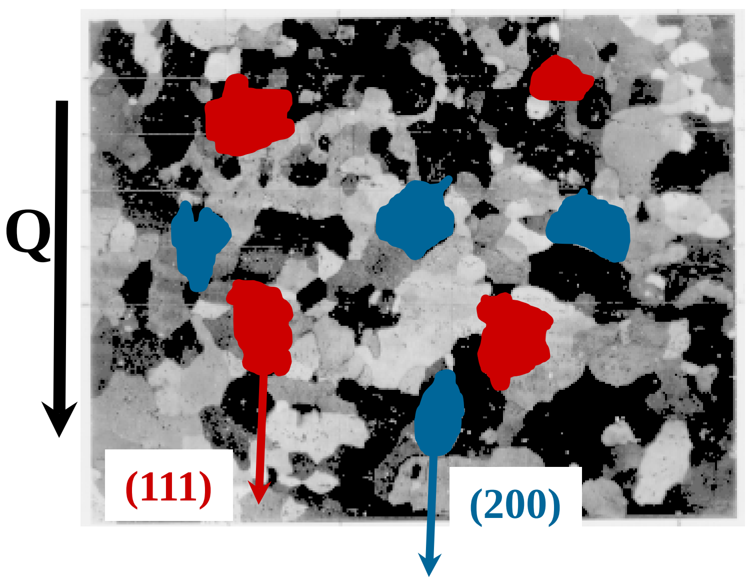
We Need to Step Back And Consider What Diffraction Tells Us

Polycrystalline Aggregate







- Each diffraction peak samples grains with a common orientation.
- Many sample and crystal directions can be monitored.
- But cannot link peaks from different sample directions to unique grain sets.
- To calculate stress, I must assume that the differently oriented grains are under the same stress.

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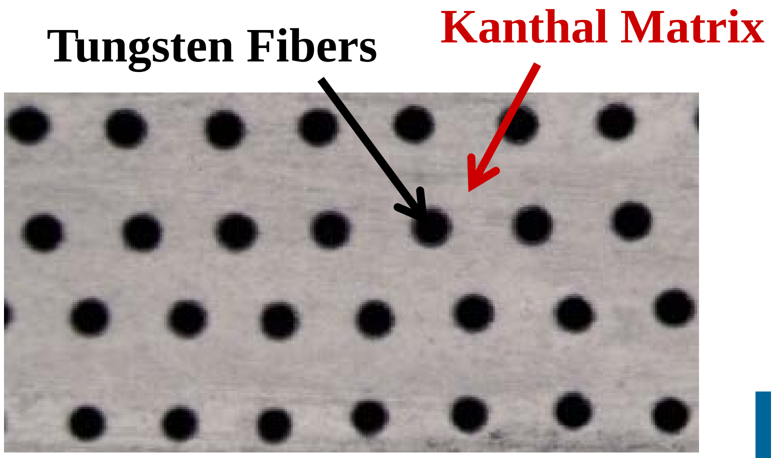
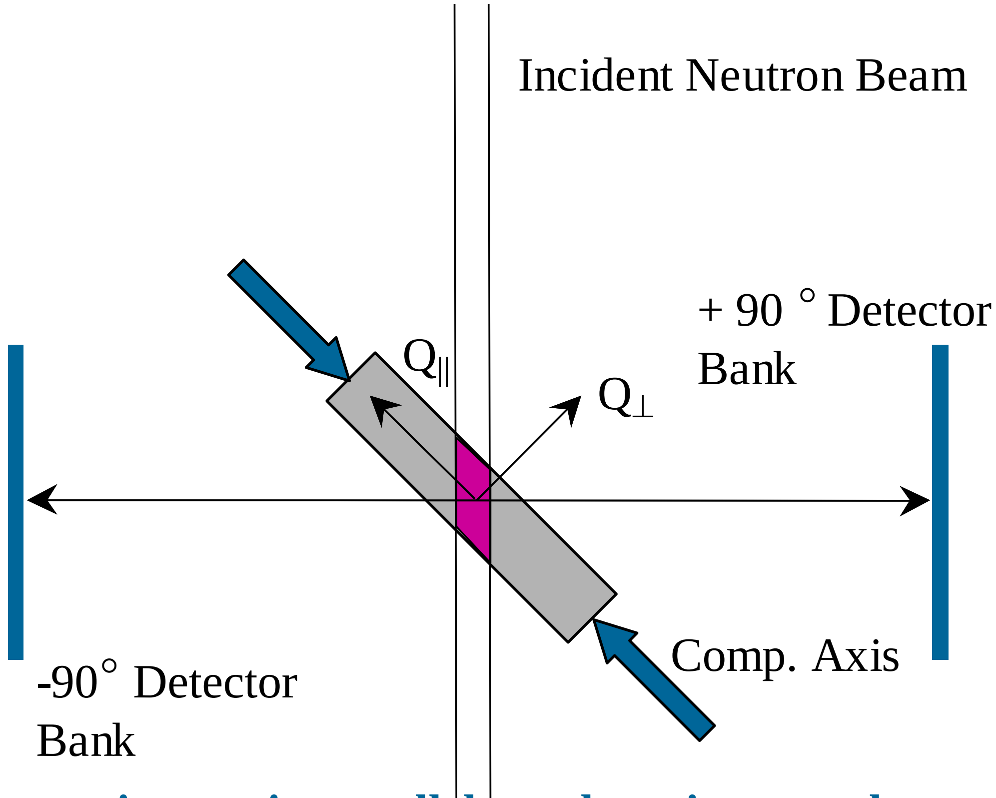
Lets Consider How a Composite Responds to Deformation



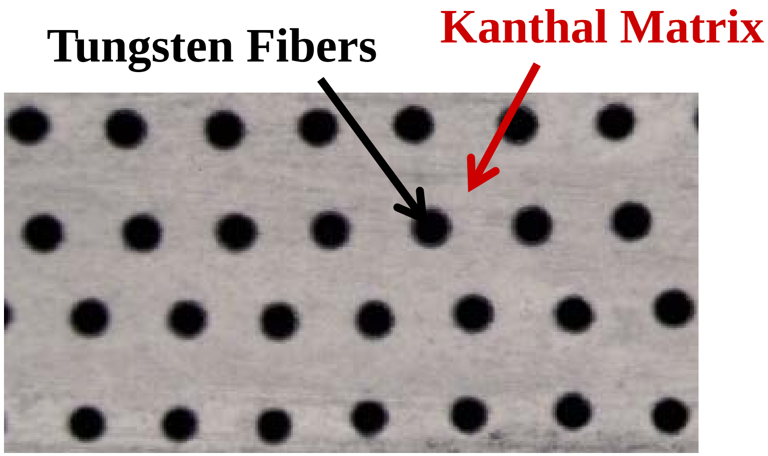


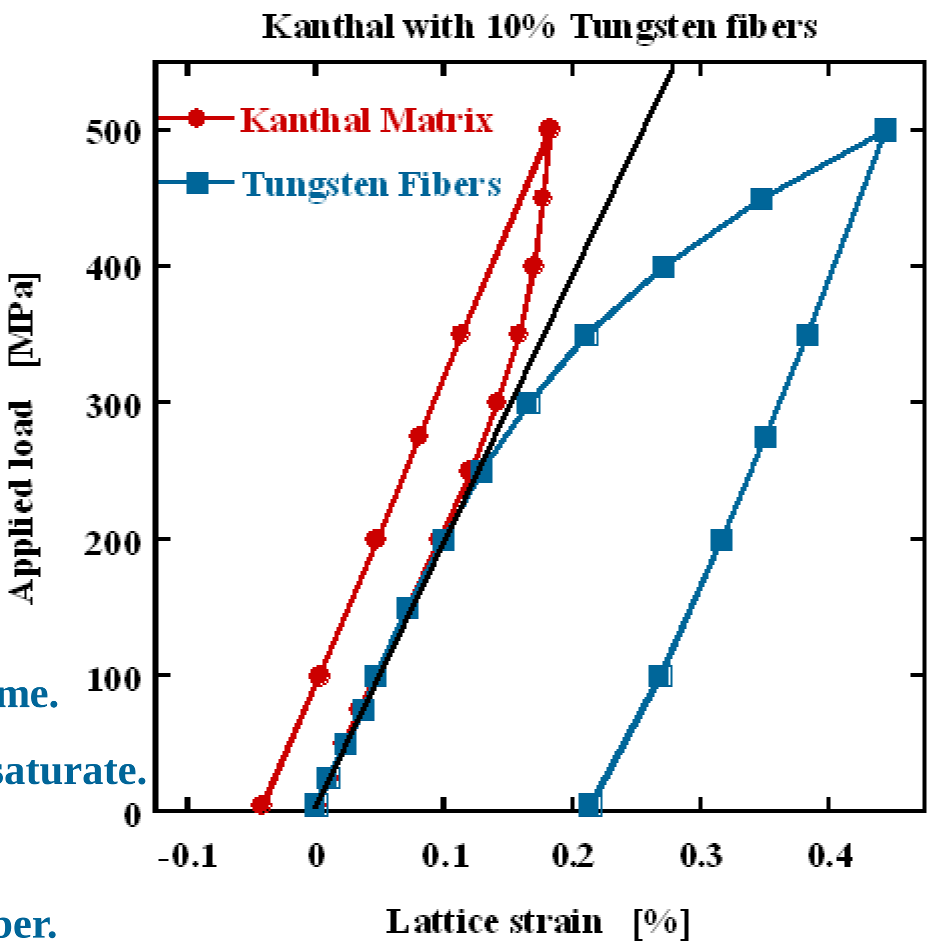
- Microstructure represents loading 2 constituents in parallel, total strains must be equal.

$$\varepsilon_T = \varepsilon_e + \varepsilon_p$$

- In elastic regime, lattice strains are equivalent.
- Once plasticity begins in one phase, the elastic lattice strains are no longer constrained to each other.

Understand Anisotropy in Terms of a Composite





- In elastic region, the lattice strains are the same.
- Above yield point, elastic strains in Kanthal saturate.
  - It is yielding.
  - Load is redistributed to the Tungsten fiber.
- This is how a composite is designed to work.
- With release of the macroscopic stress, there is a residual stress in each constituent.
  - The phases stresses are not representative of the macroscopic stress state.
  - However, a weighted average would be representative.

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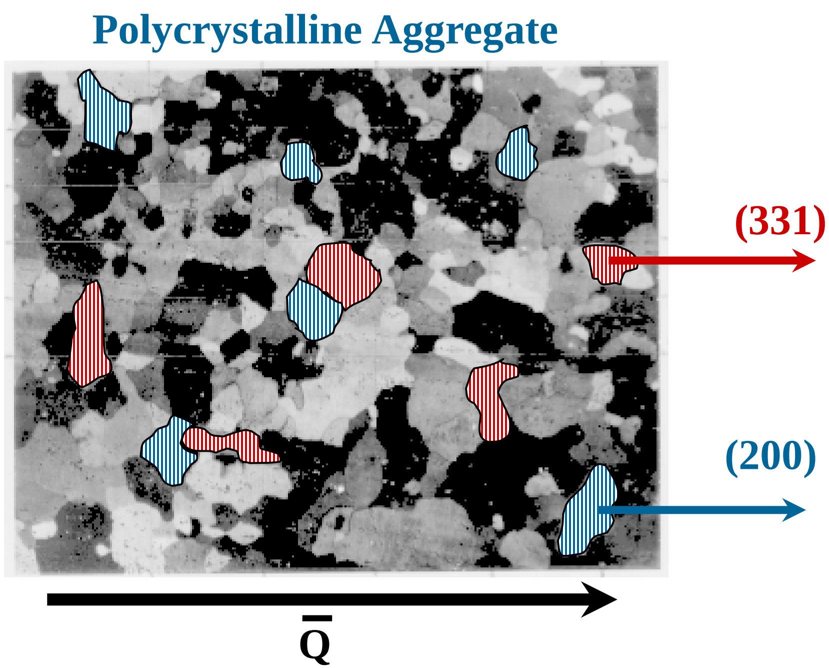
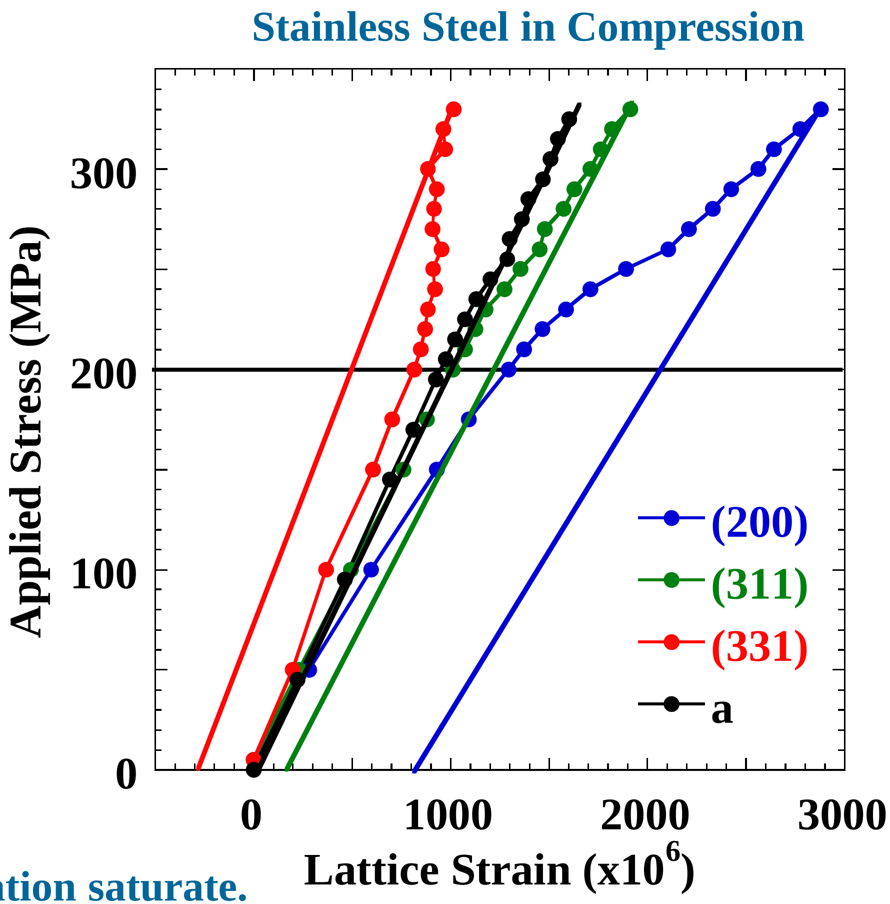
Polycrystalline Samples : "The Mother of All Composites".





- Above yield point, elastic strains in (311) orientation saturate.
  - Grains with (311) parallel to the load direction are yielding.
  - Load is redistributed to the (200) orientations.
- This is a general result seen in FCC metals.
  - Indicative of the specific deformation mechanism.

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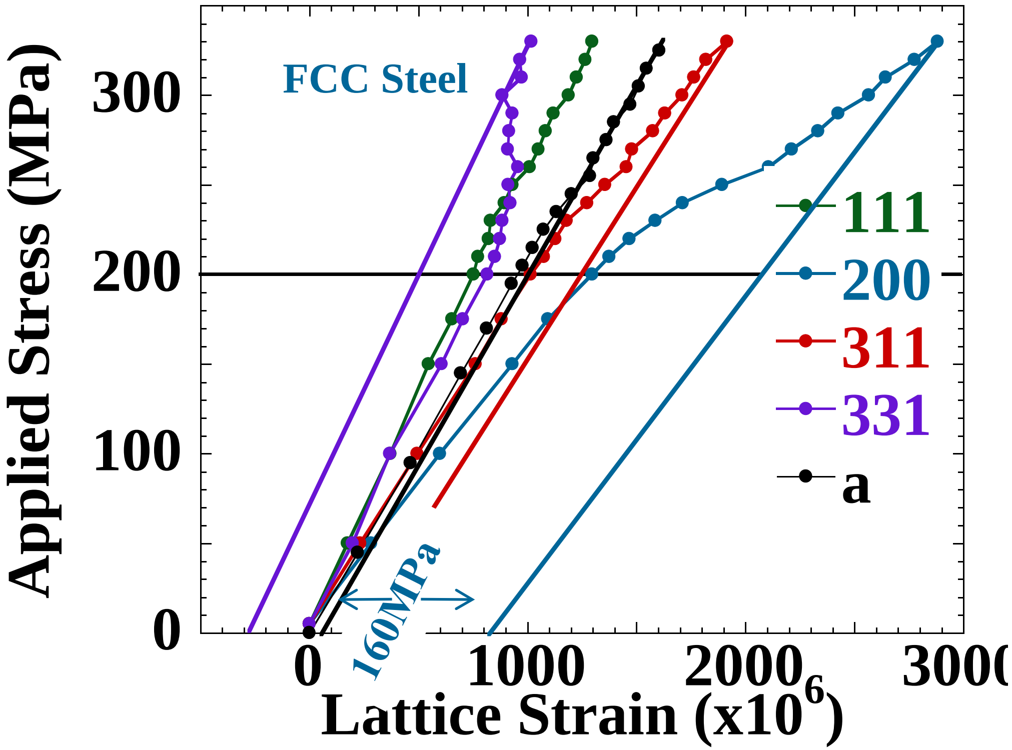
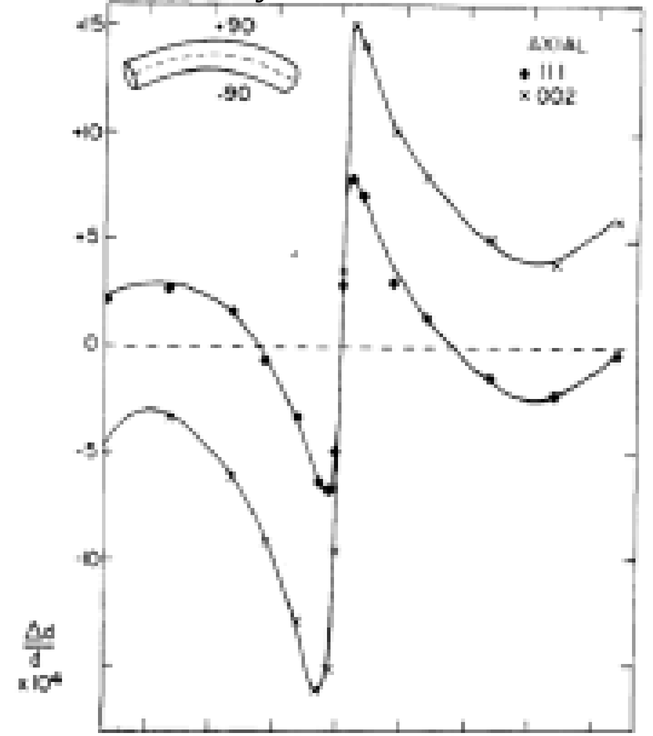
There is a Long Time Strategy to Deal With Anisotropy



Courtesy of Tom Holden



- Elastically anisotropic, but that just effects the stiffness you use.
- Plastic anisotropy will lead you to the wrong answer.
- Tom Holden has told us all not to use the (002) and (111) for FCC's.
- Solution : (311) reasonably represents the macroscopic stress state.
- The lattice parameter found from Rietveld refinement represents the bulk very well.

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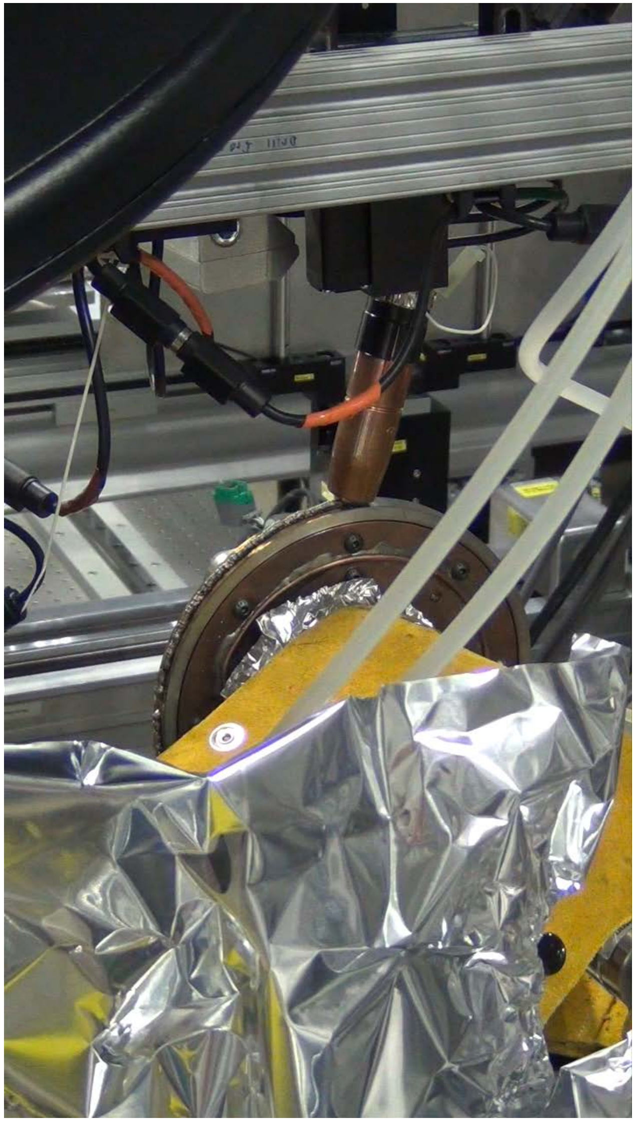
But What Can We Obtain From a Dynamic Measurement During Deposition?

$$\begin{pmatrix} \varepsilon_{11} & \varepsilon_{12} & \varepsilon_{13} \\ \varepsilon_{21} & \boxed{\varepsilon_{22}} & \boxed{\varepsilon_{23}} \\ \varepsilon_{31} & \boxed{\varepsilon_{32}} & \boxed{\varepsilon_{33}} \end{pmatrix} = \begin{pmatrix} \varepsilon_h & 0 & 0 \\ 0 & \varepsilon_h & 0 \\ 0 & 0 & \varepsilon_h \end{pmatrix} + \begin{pmatrix} \varepsilon_{11}-\varepsilon_h & \varepsilon_{12} & \varepsilon_{13} \\ \varepsilon_{21} & \varepsilon_{22}-\varepsilon_h & \varepsilon_{23} \\ \varepsilon_{31} & \varepsilon_{32} & \varepsilon_{33}-\varepsilon_h \end{pmatrix}$$

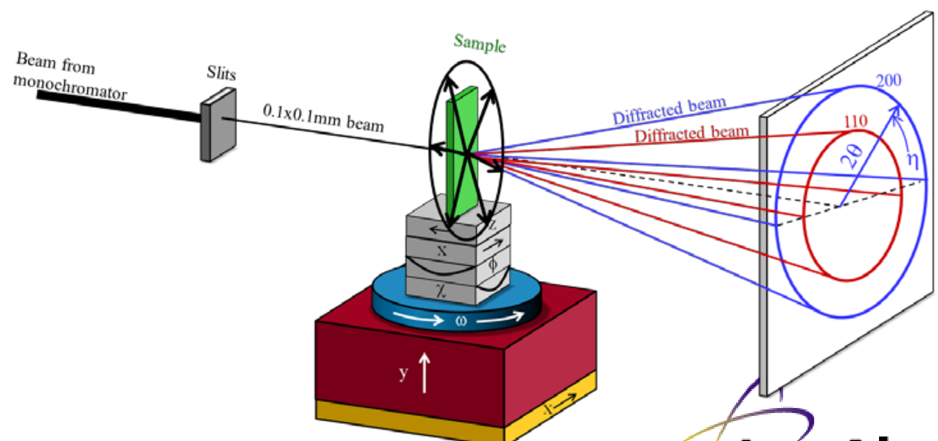
Temperature and Chemistry Change

Mechanical Strains

Portable Additive Manufacturing Platform Developed for In-Situ Studies on X-ray and Proton Beamlines

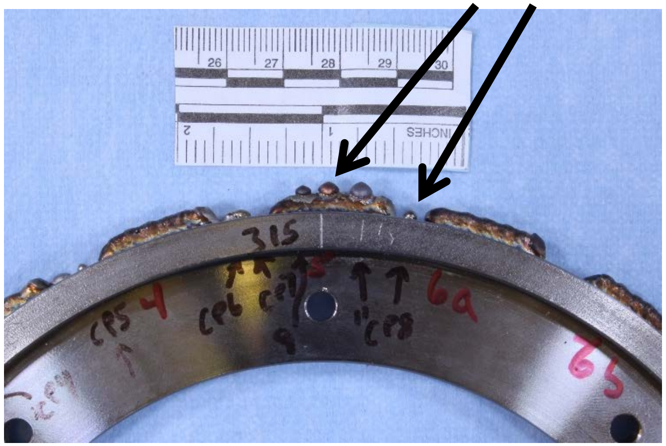


- Objective is to characterize the deposited material (and substrate) as the process is happening (in-situ).
- Cold Metal Transfer (CMT) weld process.
  - Like MIG but better controlled.
- 1ID @ the APS
  - E=71.67keV, λ=0.171Å
    - ~2 factors of e absorption.
  - Beam size 0.05-0.2mm for diffraction (could be smaller)
  - 1x2mm for imaging (largest FOV)
  - Rotating wheel speed ~1mm/sec

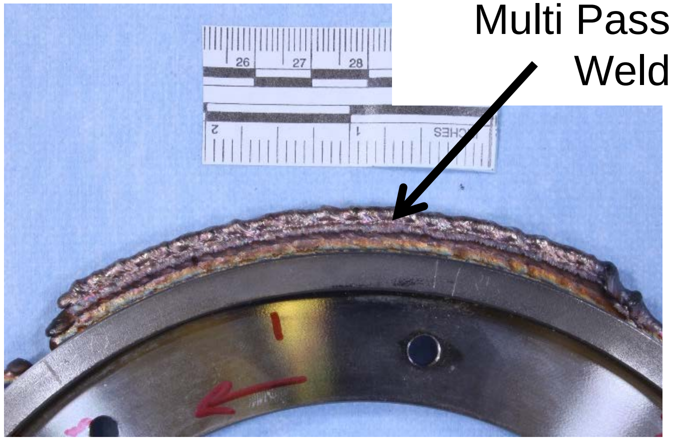


3 Types of In-Situ Measurements Were Completed on 1-ID

Cow Pats



Multi Pass Weld



Ordered in increasing complexity and increasing information…

- Stationary substrate (wheel) with ~1 second deposition.
- Rotating wheel with incident x-ray spot fixed.
    - Multiple tests with different lateral beam positions.
- Rotating wheel while moving x-ray spot.
    - Multiple tests at different beam heights.

Others you will not see today…

- Post deposition residual stress measurements
- Measurements in wheel below deposition
- Laser re-melting of previously deposited material.

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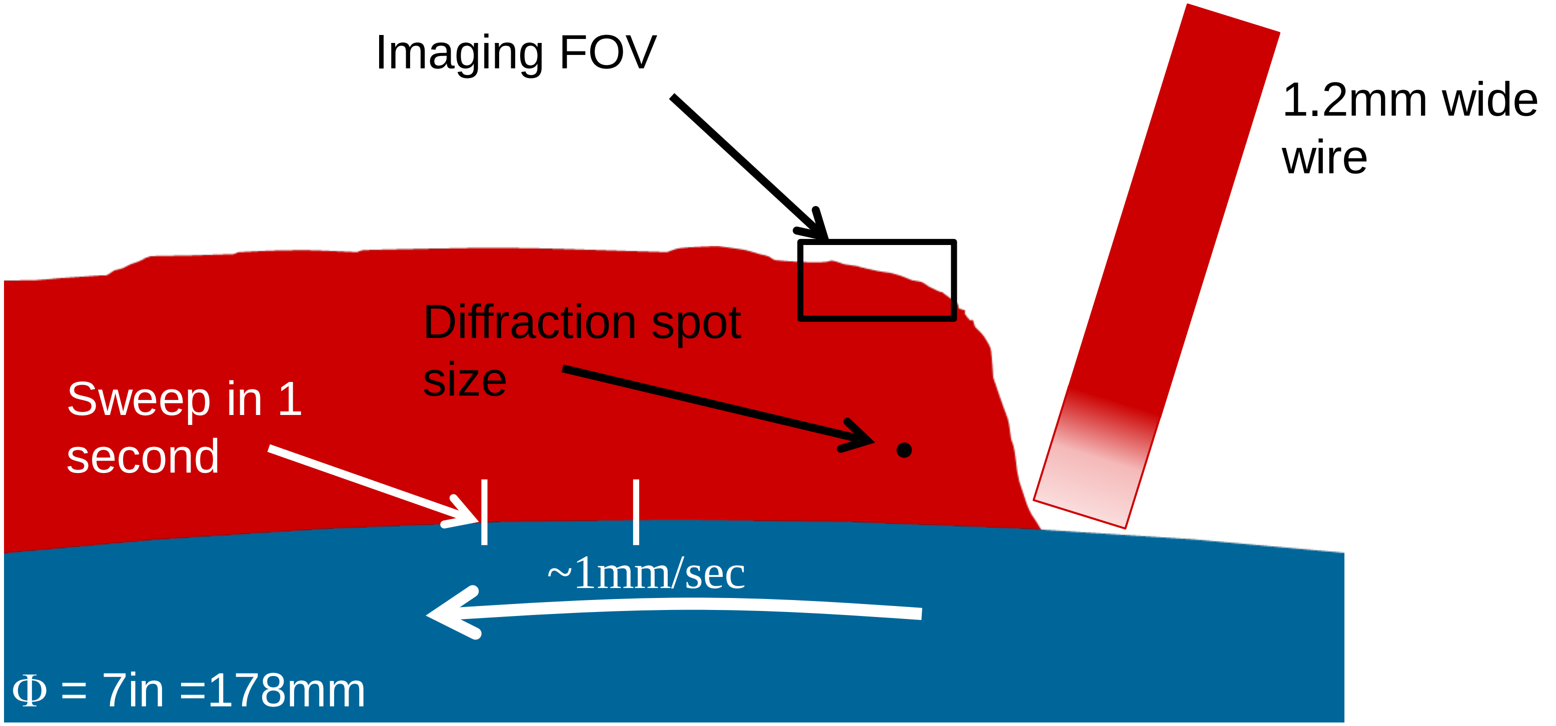
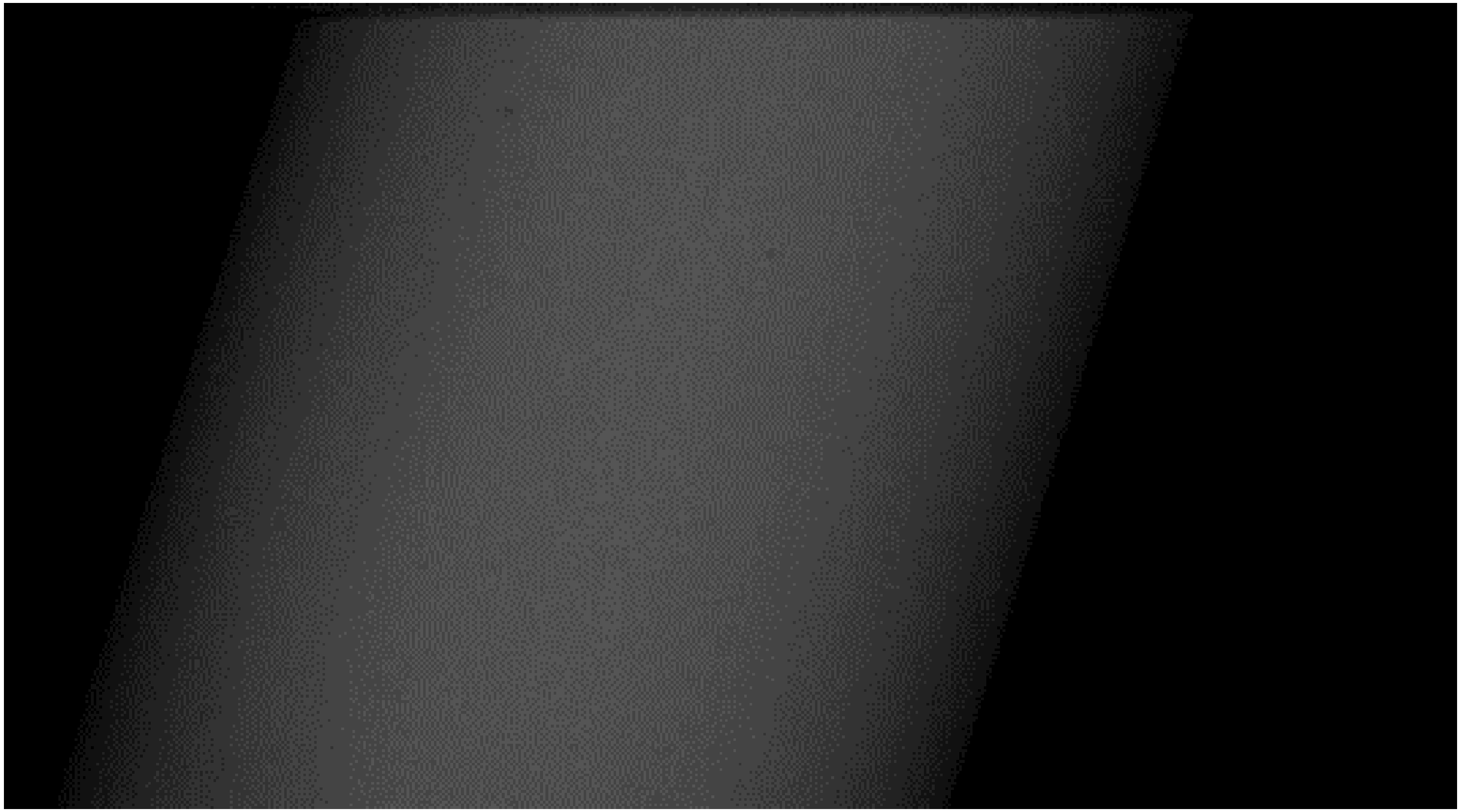
Scale Relationships Between Sample and Probe Size

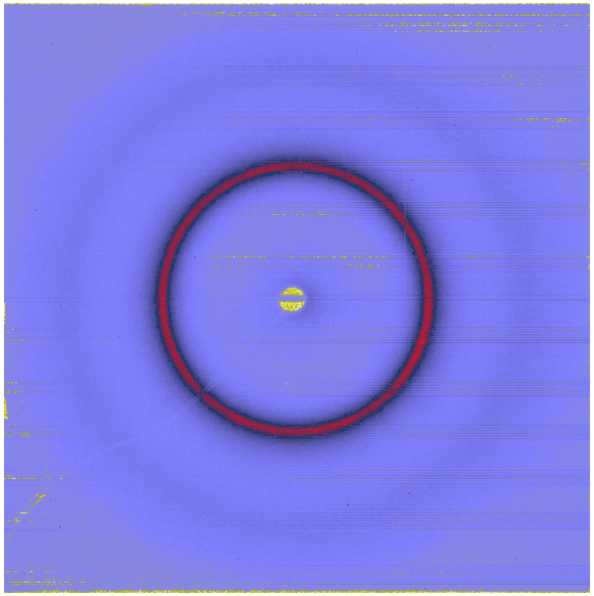


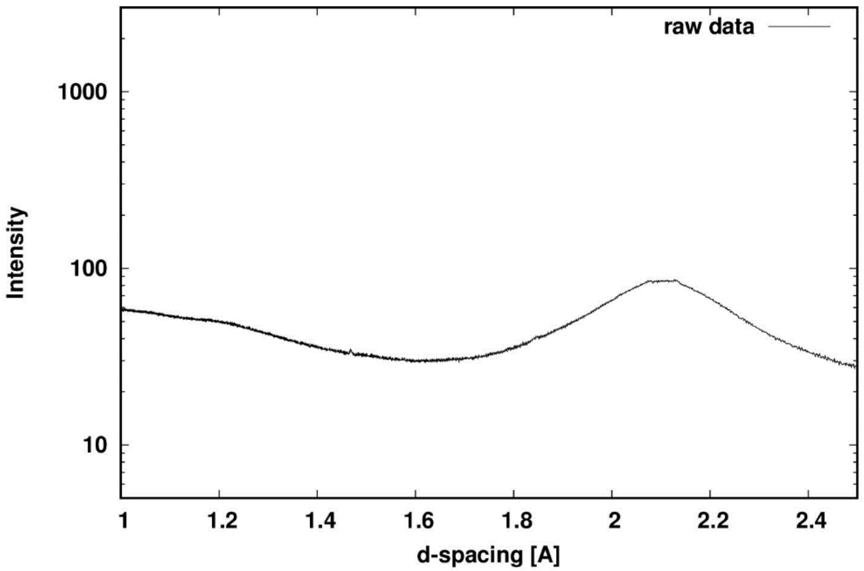
- Diffraction count time and spot size were dictated by statistical sampling of grains in integration volume.
    - Usually 1sec, sometimes 0.1sec (attenuators in beam).
- Imaging 0.1sec integration, limited by statistics.

Additive Manufacture

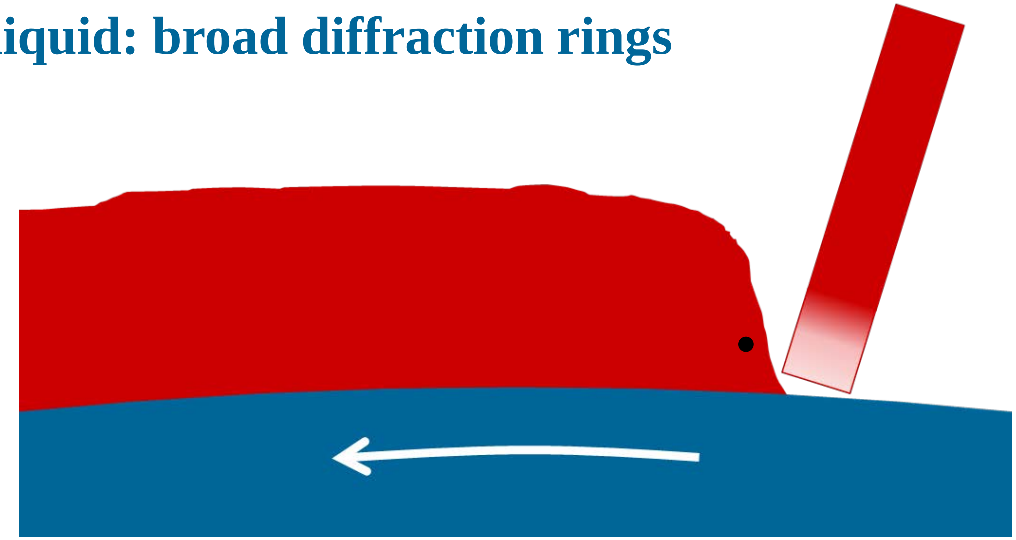


Raw Diffraction Images Tell You a Lot About Microstructure





- Pure liquid: broad diffraction rings



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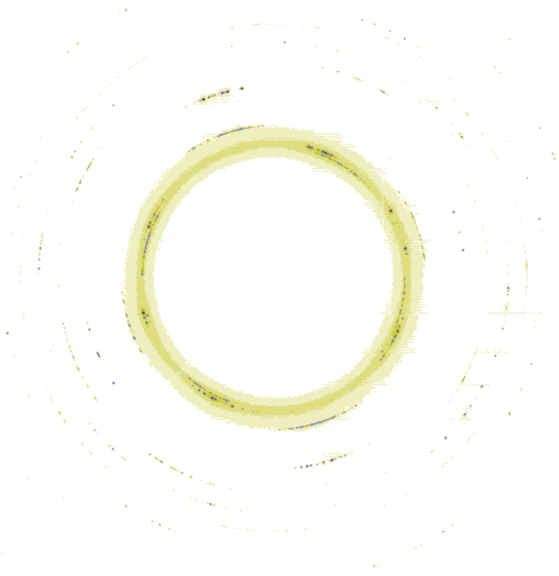
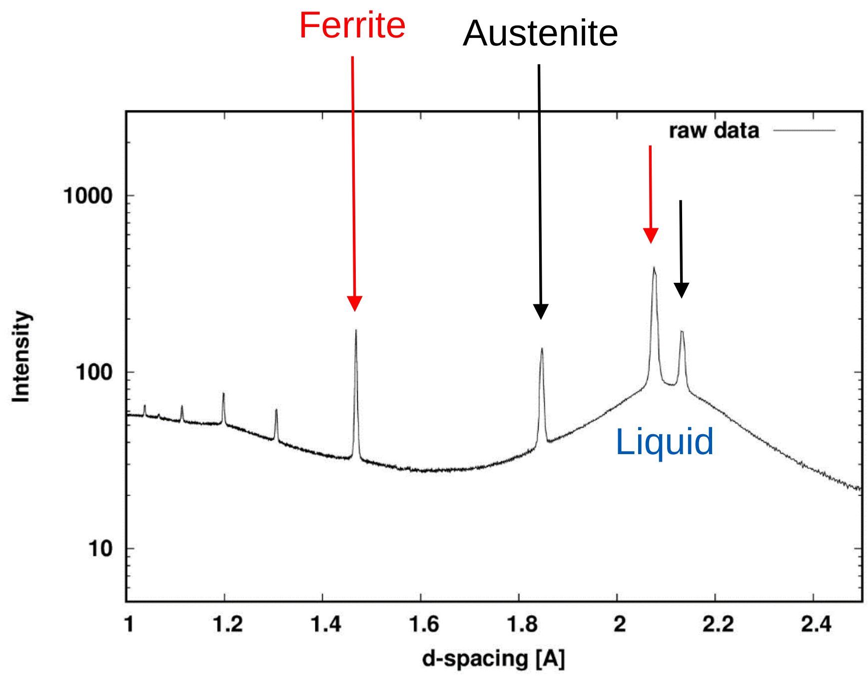
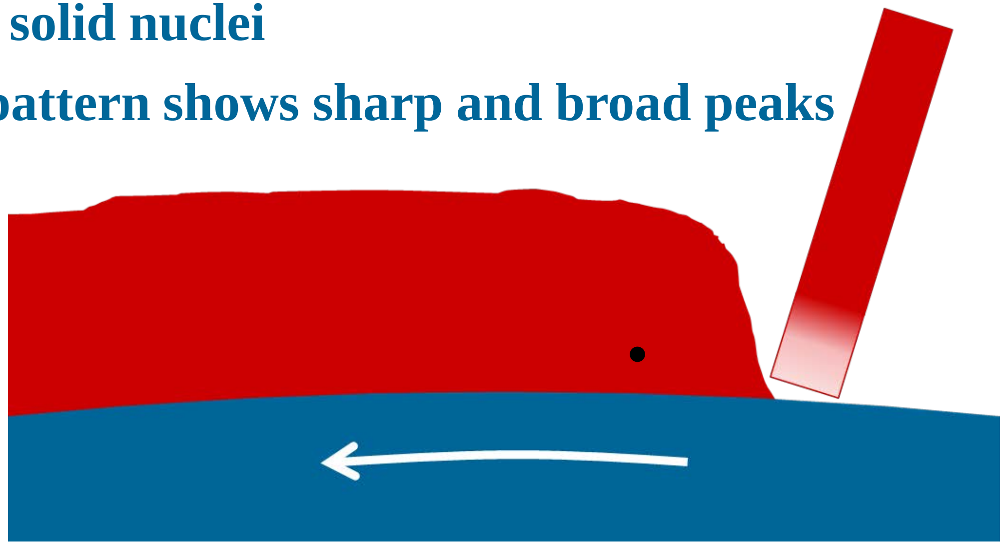




- Liquid with solid nuclei
- Integrated pattern shows sharp and broad peaks



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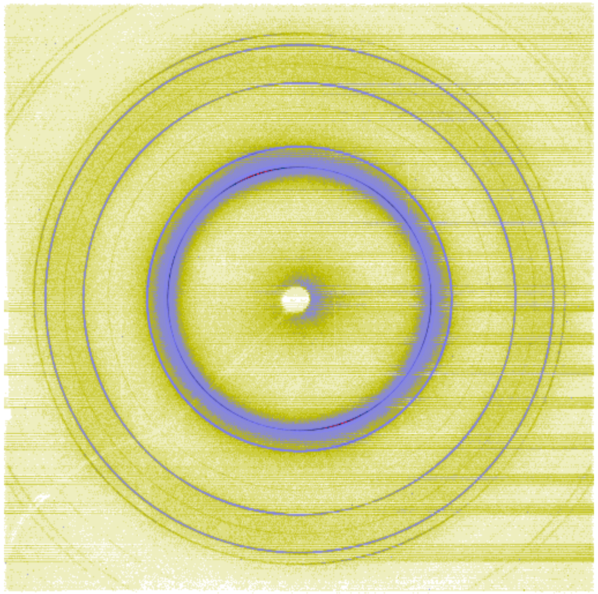
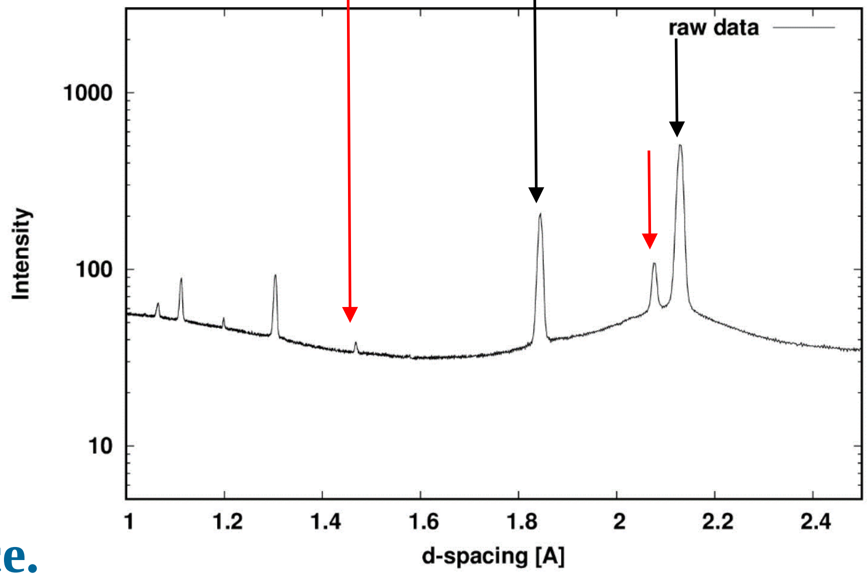
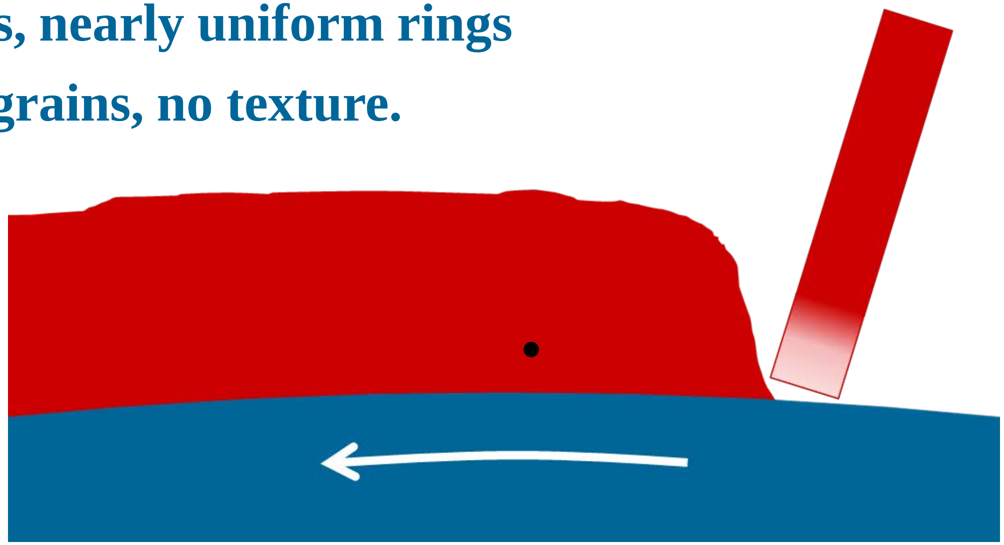




- Liquid and solid coexistence.
- Continuous, nearly uniform rings
  - Small grains, no texture.



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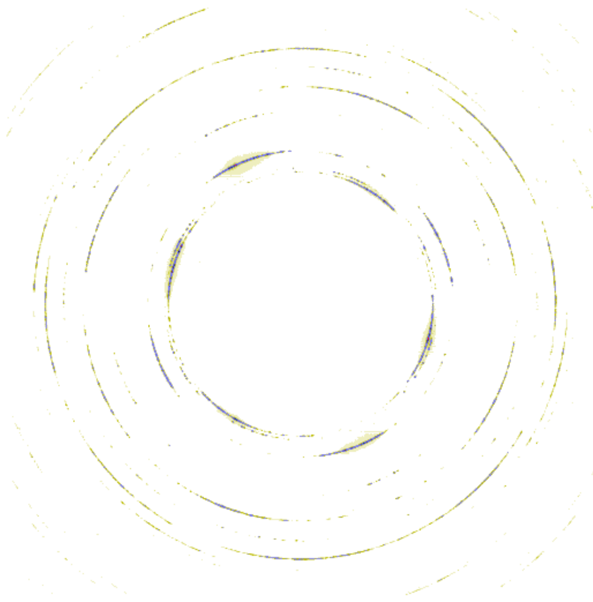
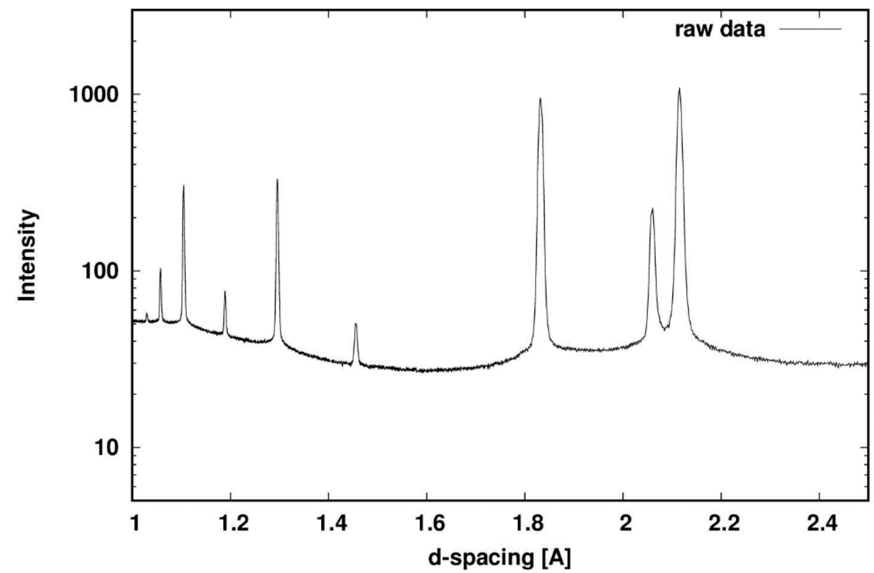
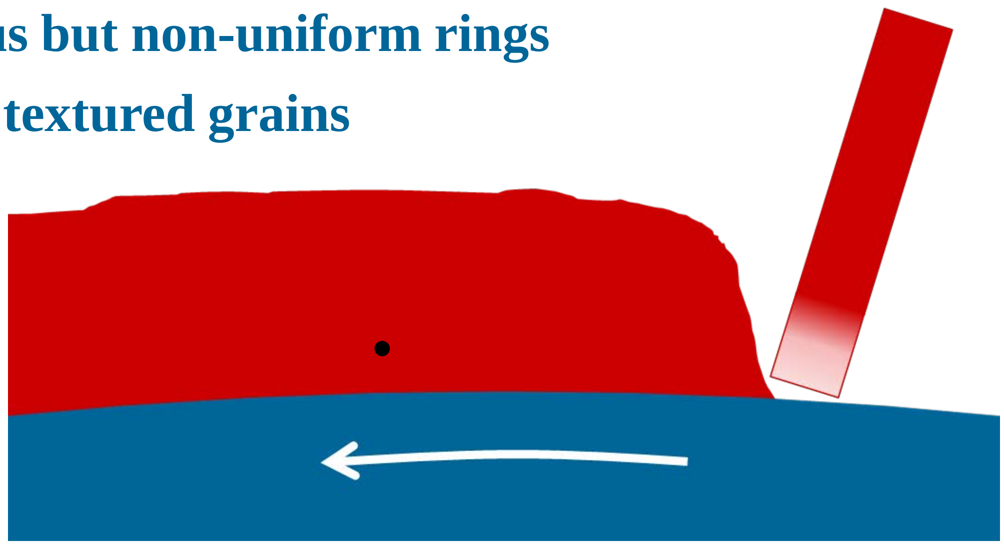
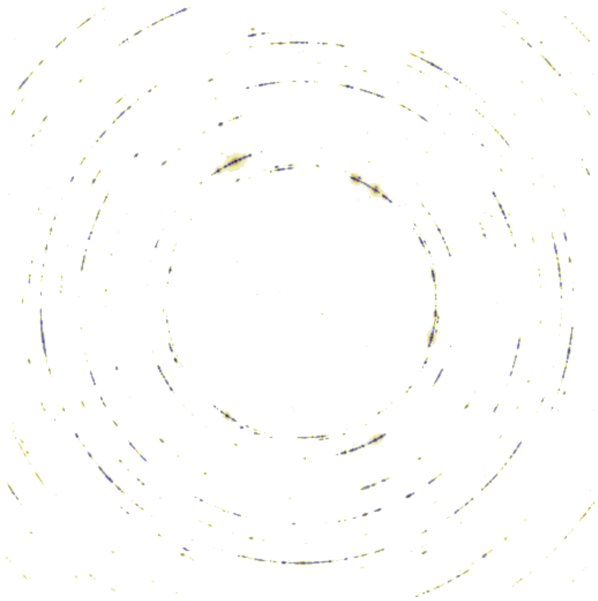


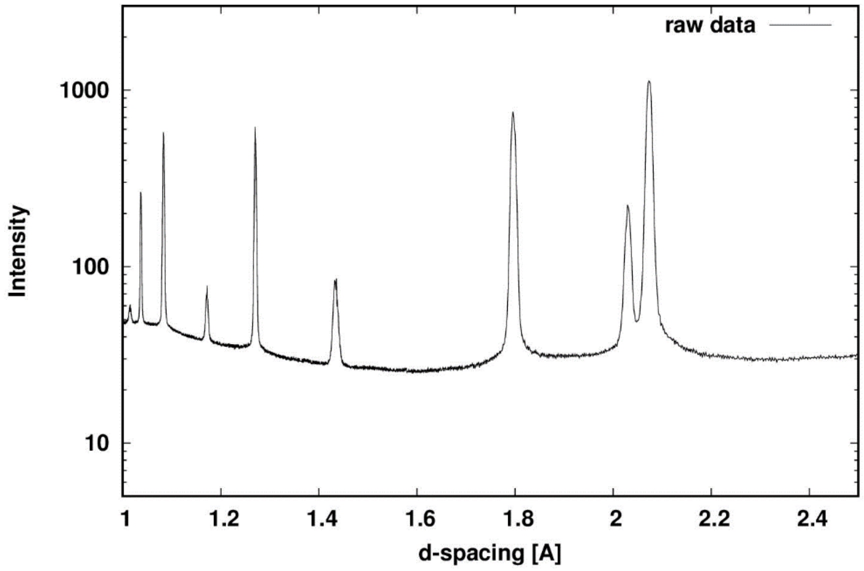


- Textured solid.
- Continuous but non-uniform rings
  - Small textured grains

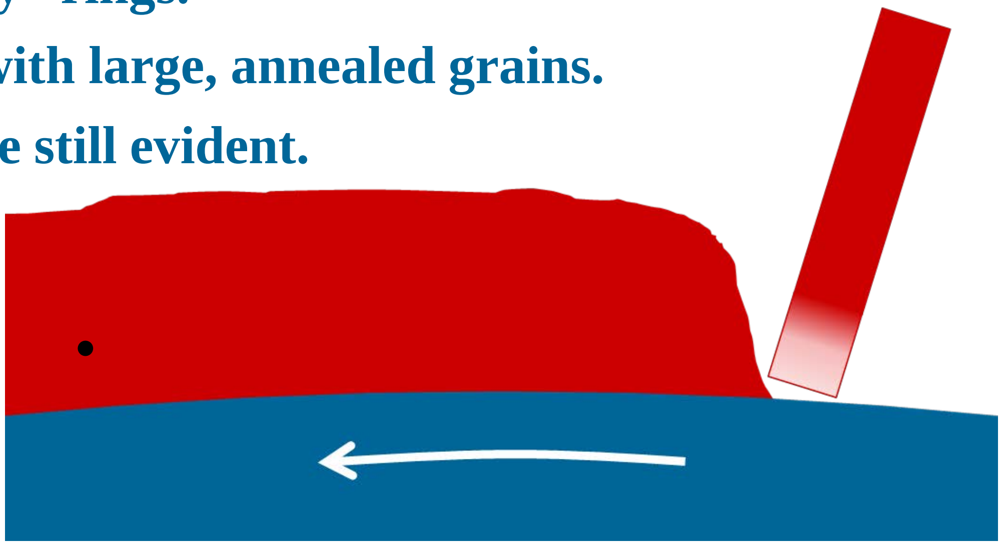


Raw Diffraction Images Tell You a Lot About Microstructure





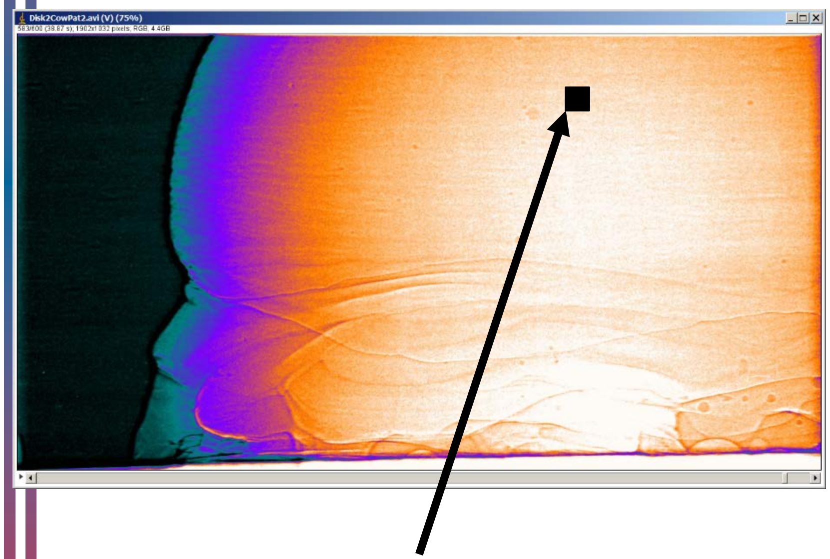
- “Spotty” rings.
- Solid with large, annealed grains.
- Texture still evident.



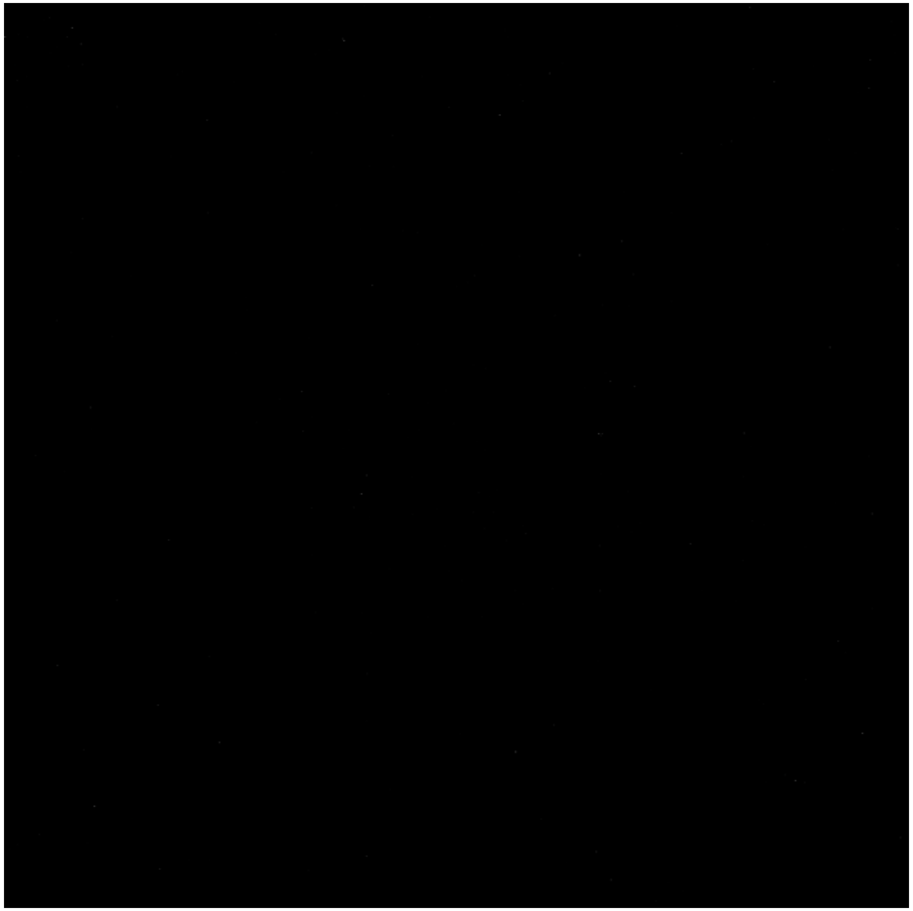
The Dropping of a Cow Pattie

Diffraction Used to Monitor the Microstructure of the Material During Deposition

- **In-situ diffraction probes phase evolution following deposition**
  - Liquid observed
  - Pattern is spotty, grains are "large".



Diffraction spot 0.8mm from rim



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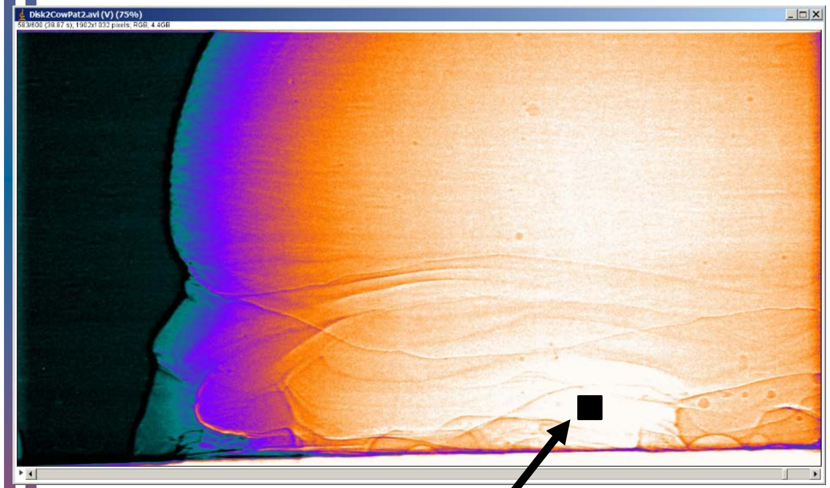
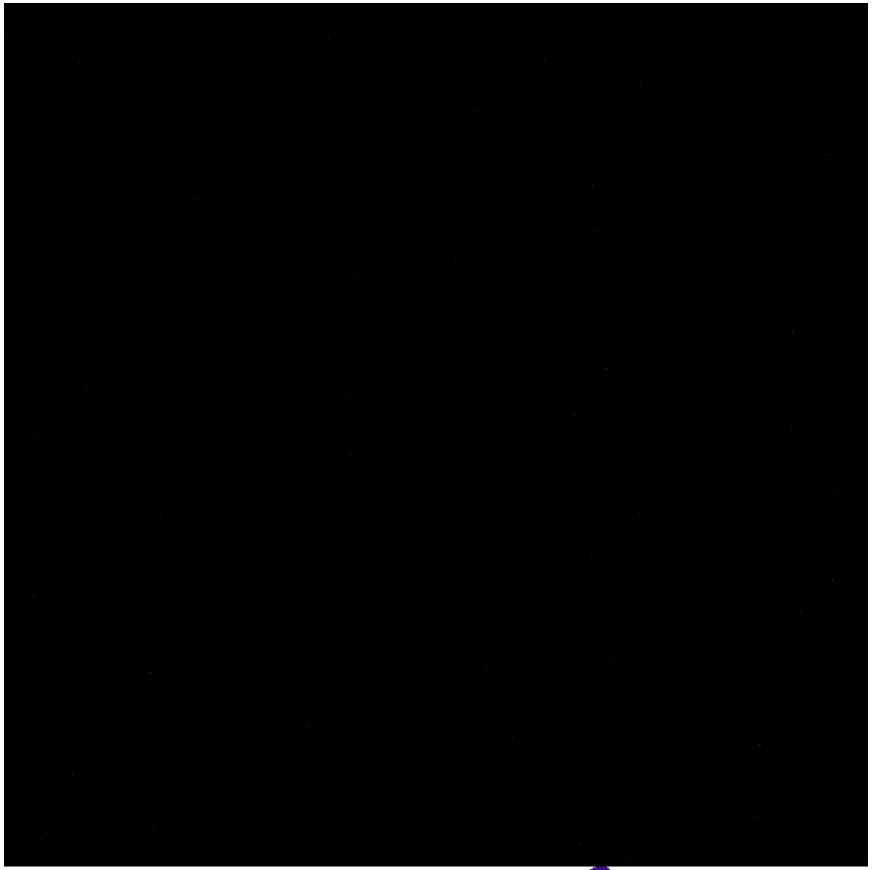
Diffraction Used to Monitor the Microstructure of the Material During Deposition

- In-situ diffraction probes phase evolution following deposition
  - No liquid observed, rapid solidification.
  - Rings are more continuous, grains are smaller



Diffraction spot 0.1mm from rim



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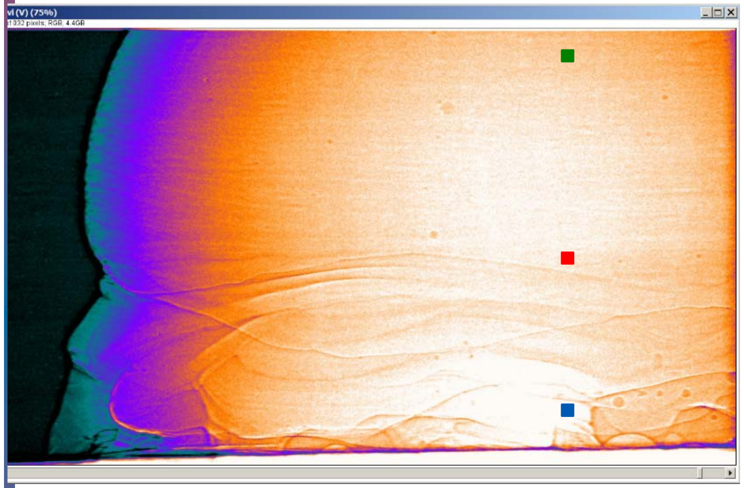
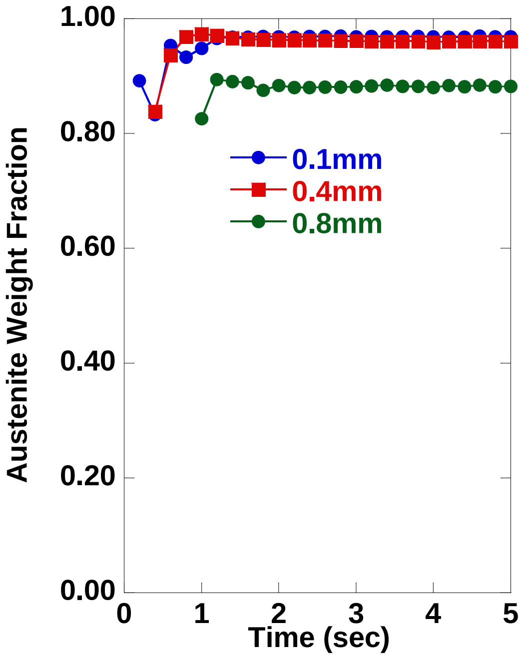
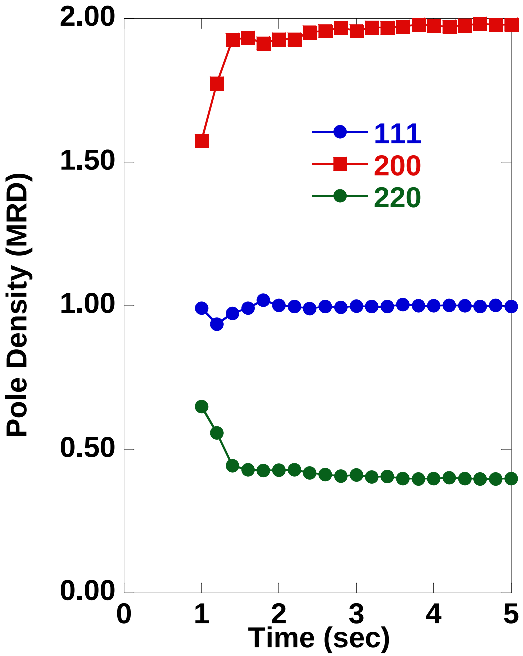
Quantitative Microstructural Information is Extracted From the Diffraction Pattern







- **Initially solidifies 80% Austenite (20% Ferrite)**
  - Quickly stabilizes, but is a function of distance above wheel.
- **Texture develops at highest measurement point, evolves in first ~0.5 second**

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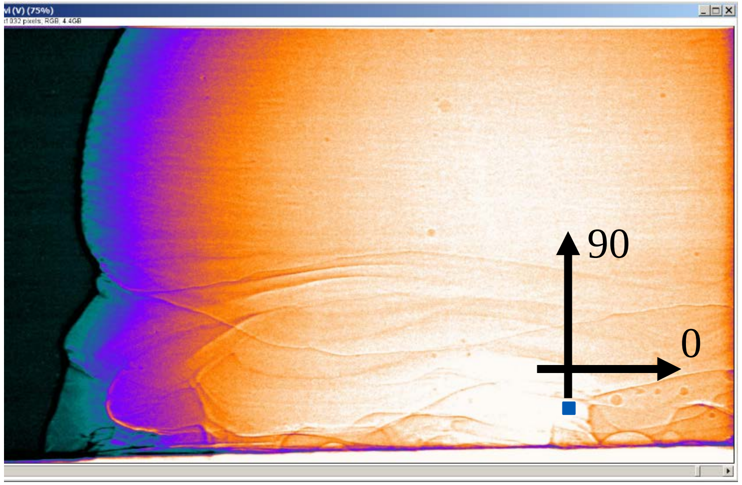
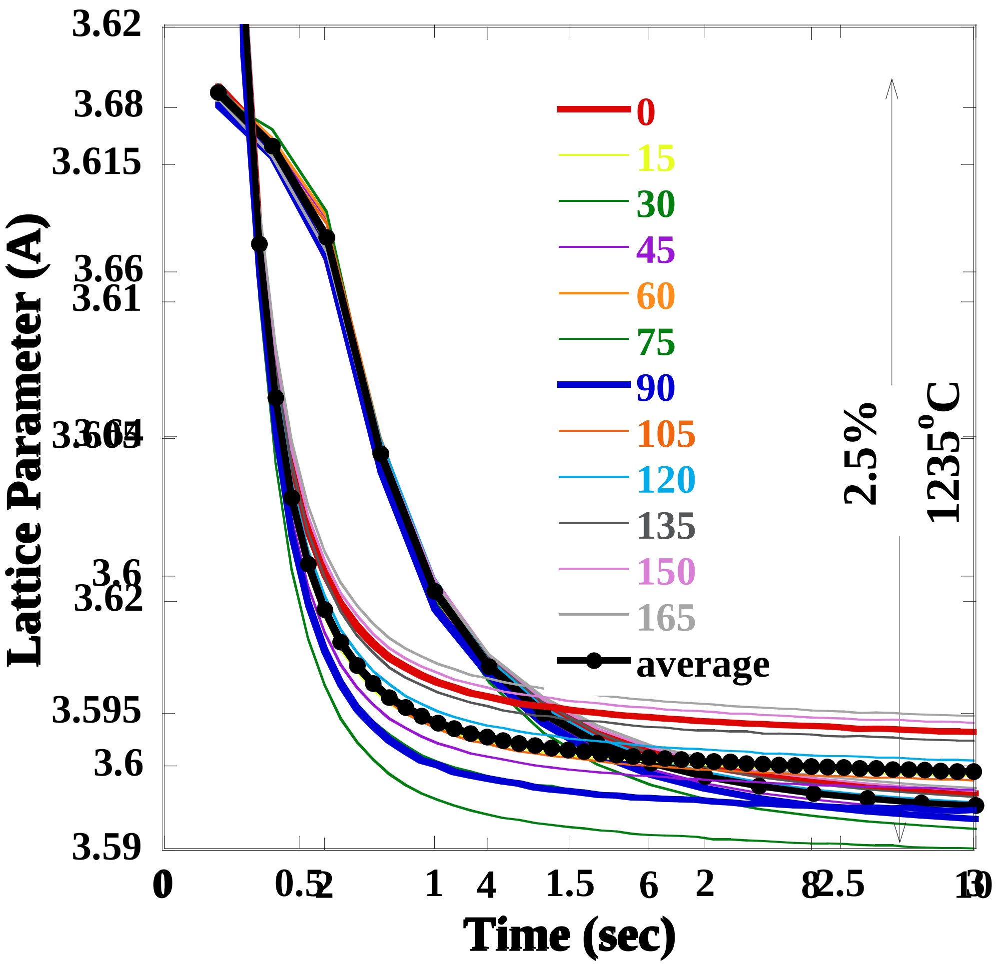
Temperature and Stress Are Evident in Lattice Parameter Evolution





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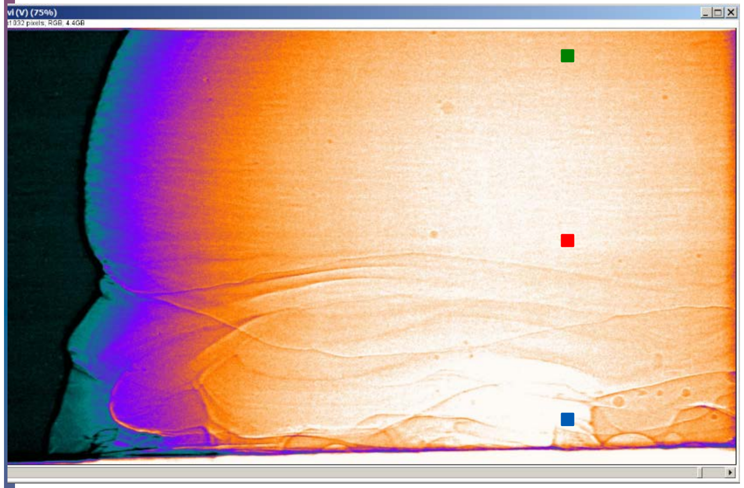
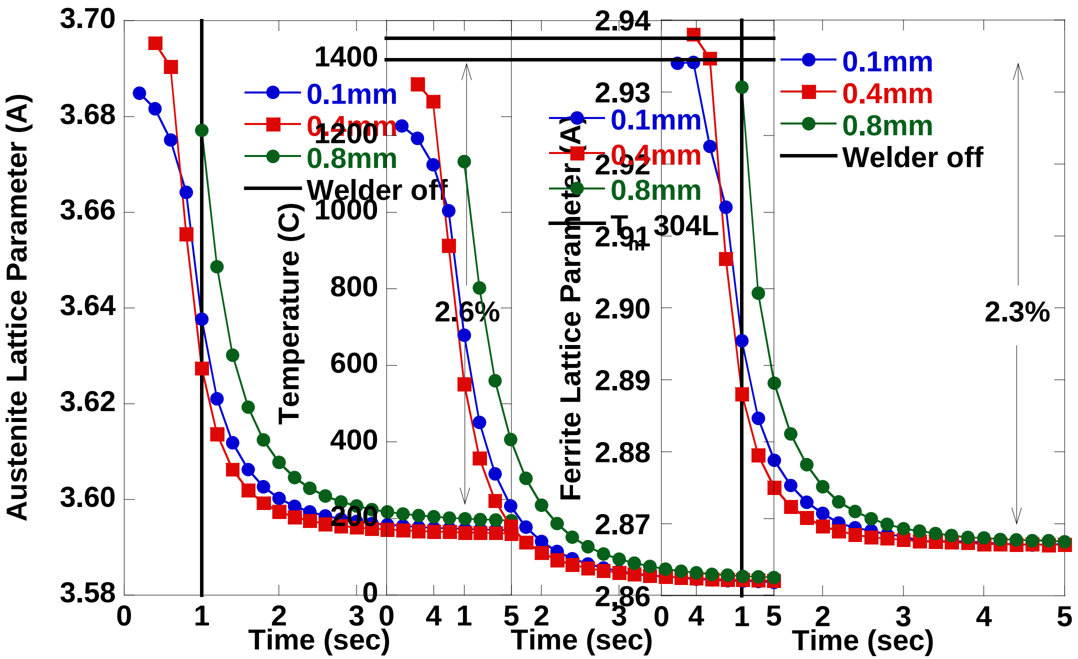
Quantitative Microstructural Information is Extracted From the Diffraction Pattern



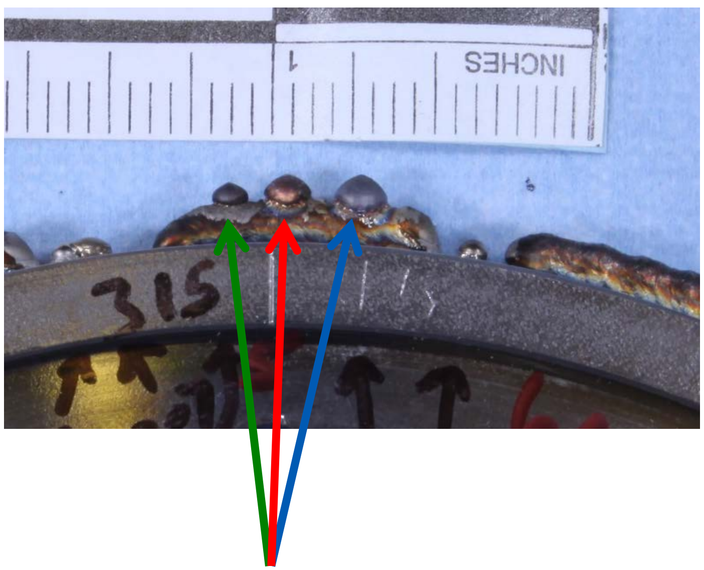


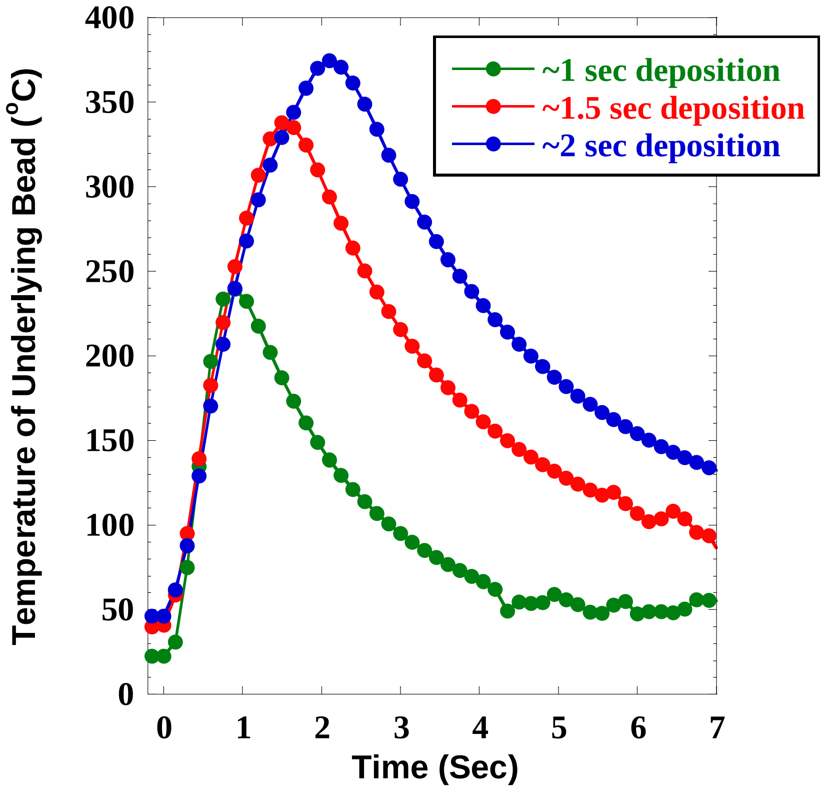
- **Track the lattice parameter of both phases during and following the weld.**
- **Lattice strain can be converted to temperature.**
  - Are there other factors in the lattice parameter, e.g. chemistry or stress?

Effect of Deposition on Previous Layer Observed

- Lattice parameter measured in-situ during welding used to determine temperature of material below deposition.

0.2mm spot size





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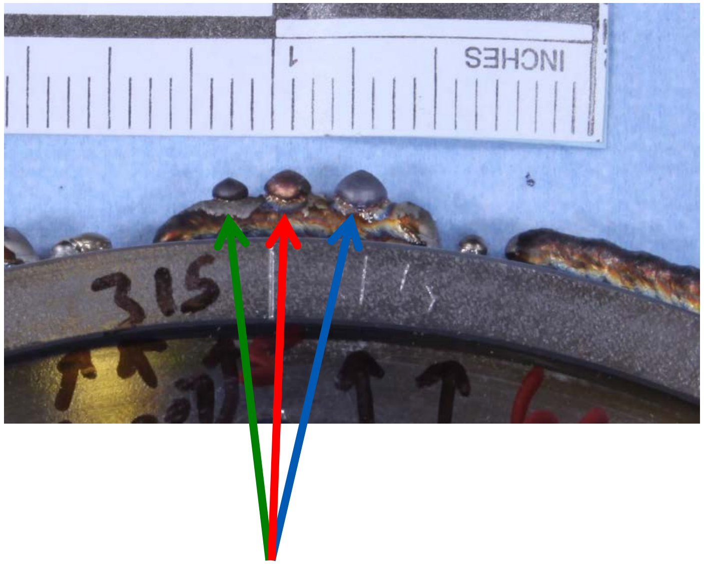
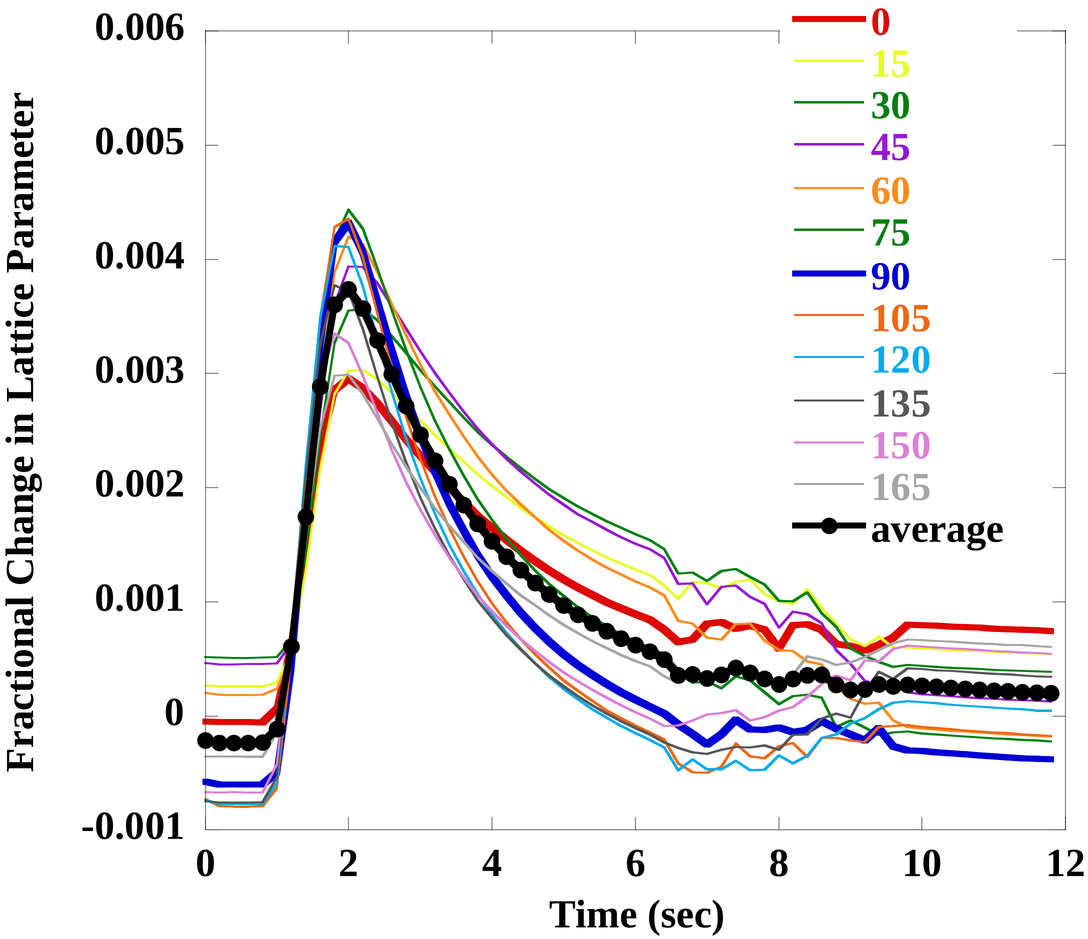
Effect of Deposition on Previous Layer Observed

0.2mm spot size





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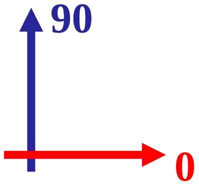
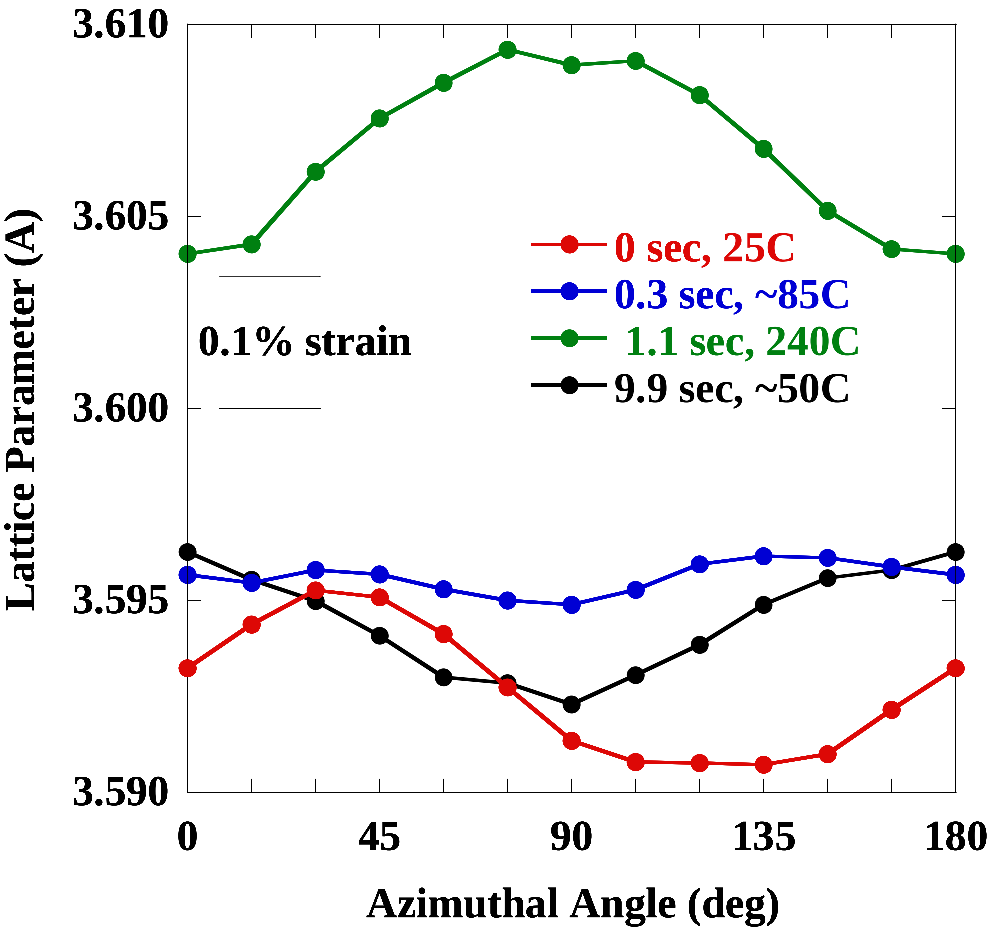
Evolution of Stress in Previous Layer is Evident

0.2mm spot size





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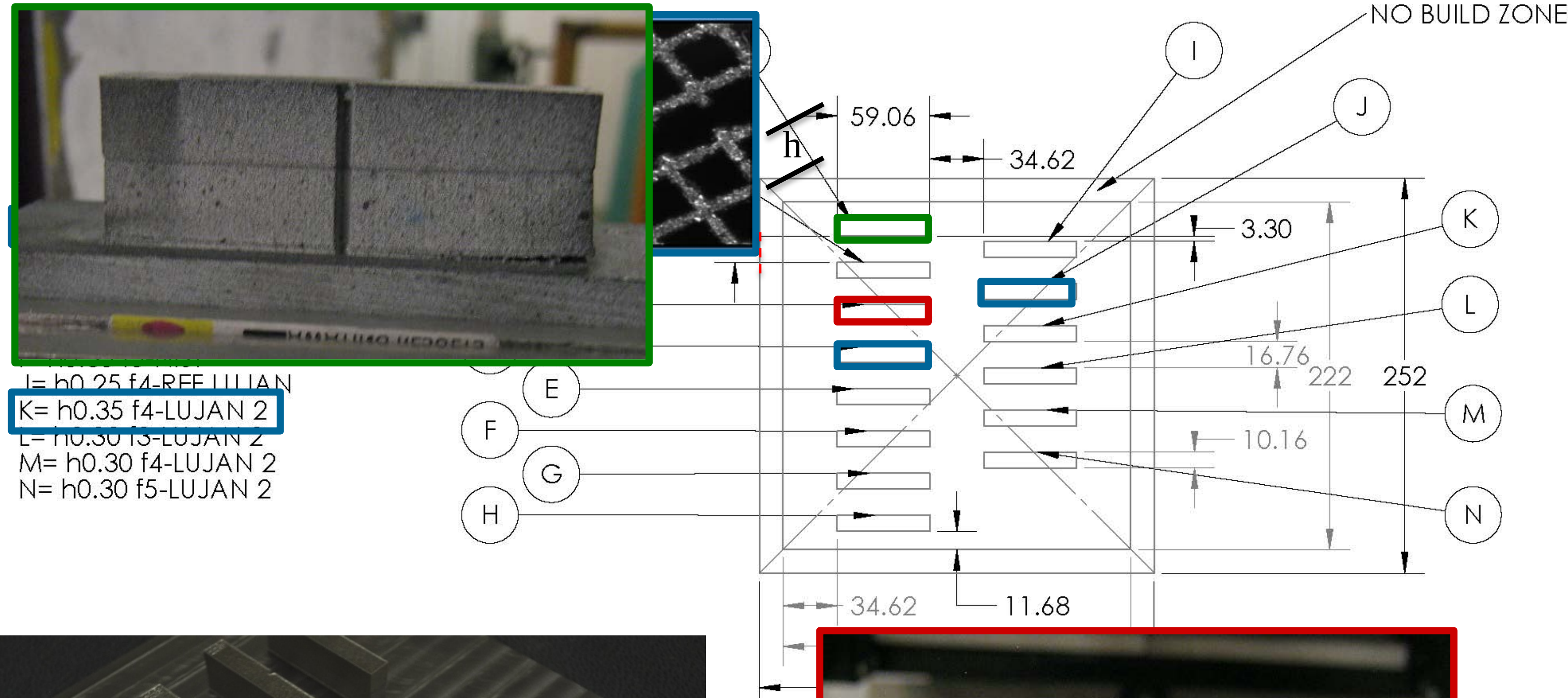
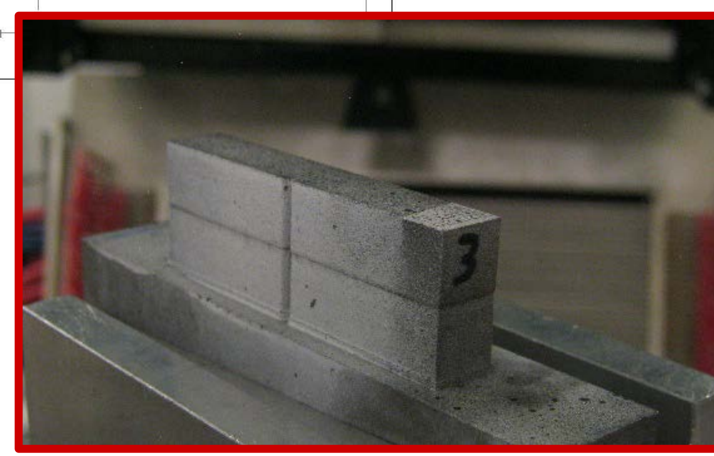
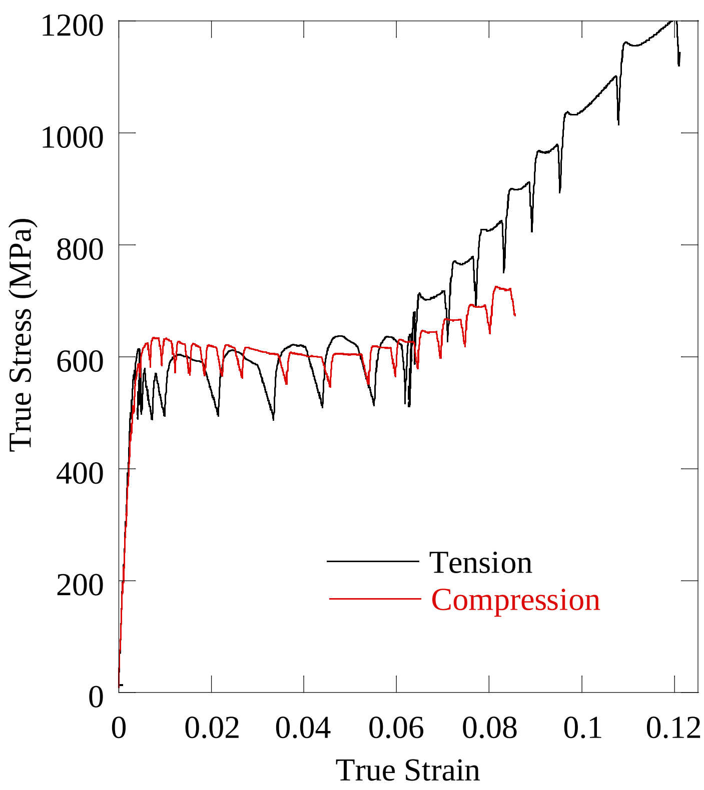
Charpy Specimen Grown From GP-1 to Study PSPP





Measured Mechanical Response of the As-Built Material



- Composition (corresponds to 17-4)
  - Cr (15 – 17.5 wt-%)
  - Ni (3 - 5 wt-%)
  - Cu (3 - 5 wt-%)
  - Mn (max. 1 wt-%)
  - Si (max. 1 wt-%)
  - Mo (max. 0.5 wt-%)
  - Nb (0.15 - 0.45 wt-%)
  - C (max. 0.07 wt-%)
- Mechanical properties of parts
  - Ultimate tensile strength ~1000MPa
  - Yield strength ~550MPa
  - Young's modulus 170 ± 20 GPa

Match EOS spec sheet (note non-typical flow curve).

Modulus of ~ 190GPa

Yield strength ~ 600MPa

UTS greater than 1.2GPa, looks like ~1.5GPa

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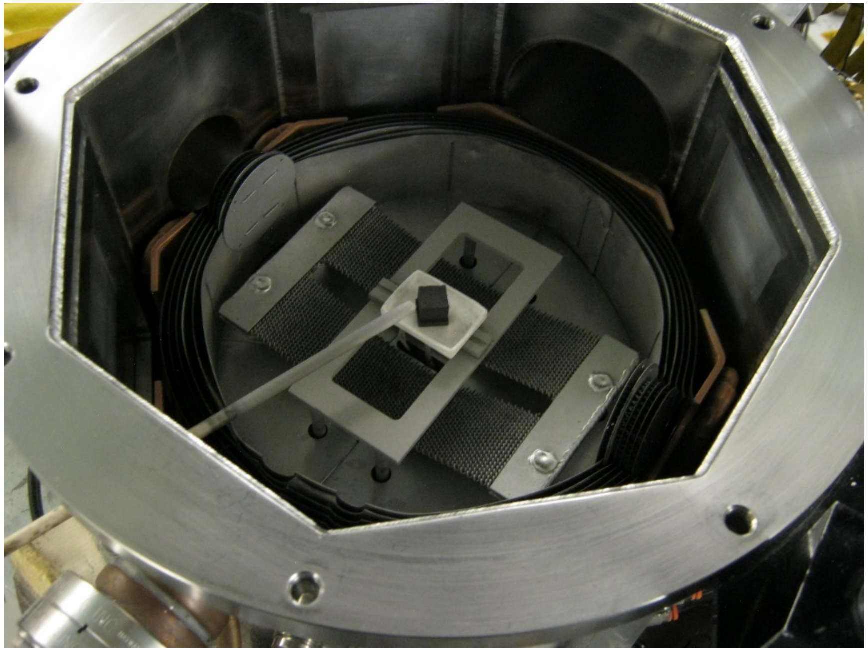
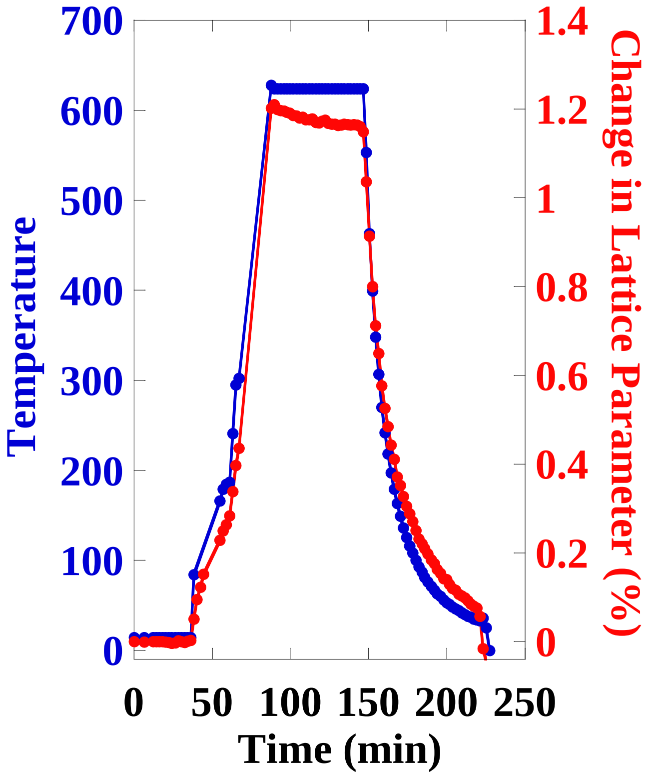
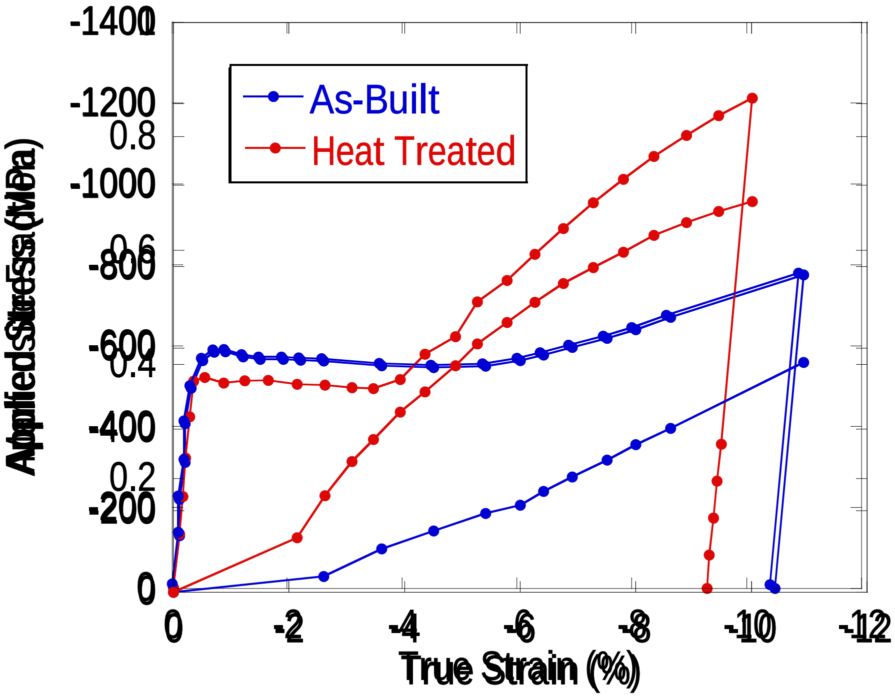
EOS Recommends a 1hr/650C "Stress Relief" Heat Treatment of GP-1 Builds

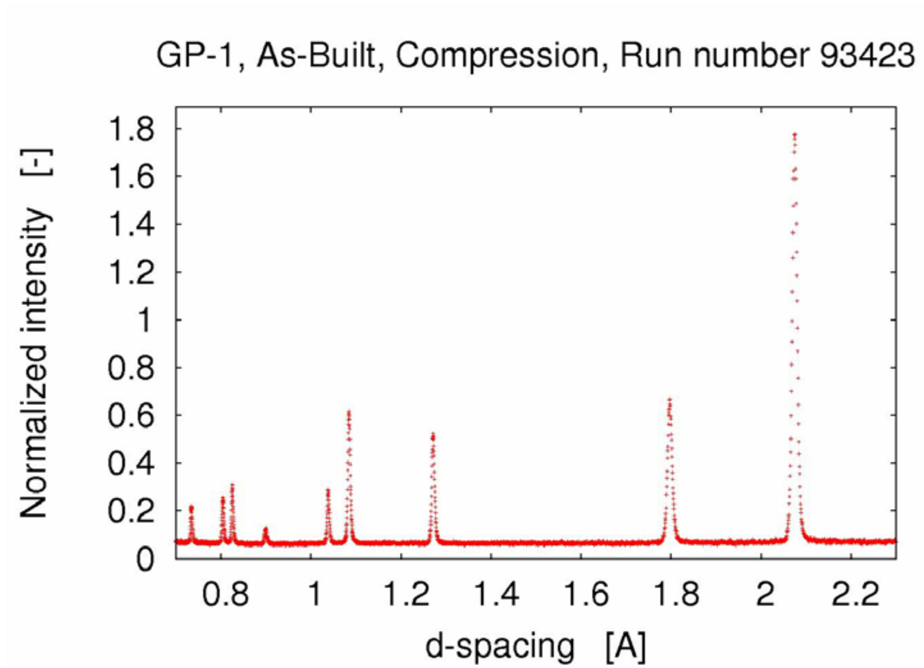




- Chemistry evolves during the heat treatment.
  - Analysis shows that interstitial nitrogen is lost during heat treatement.
- What effect does that process have on the structure/properties?

Heat Treatment Results in Large Difference in Structure and Properties





Loss of interstitial nitrogen increases propensity for martensitic transformation.

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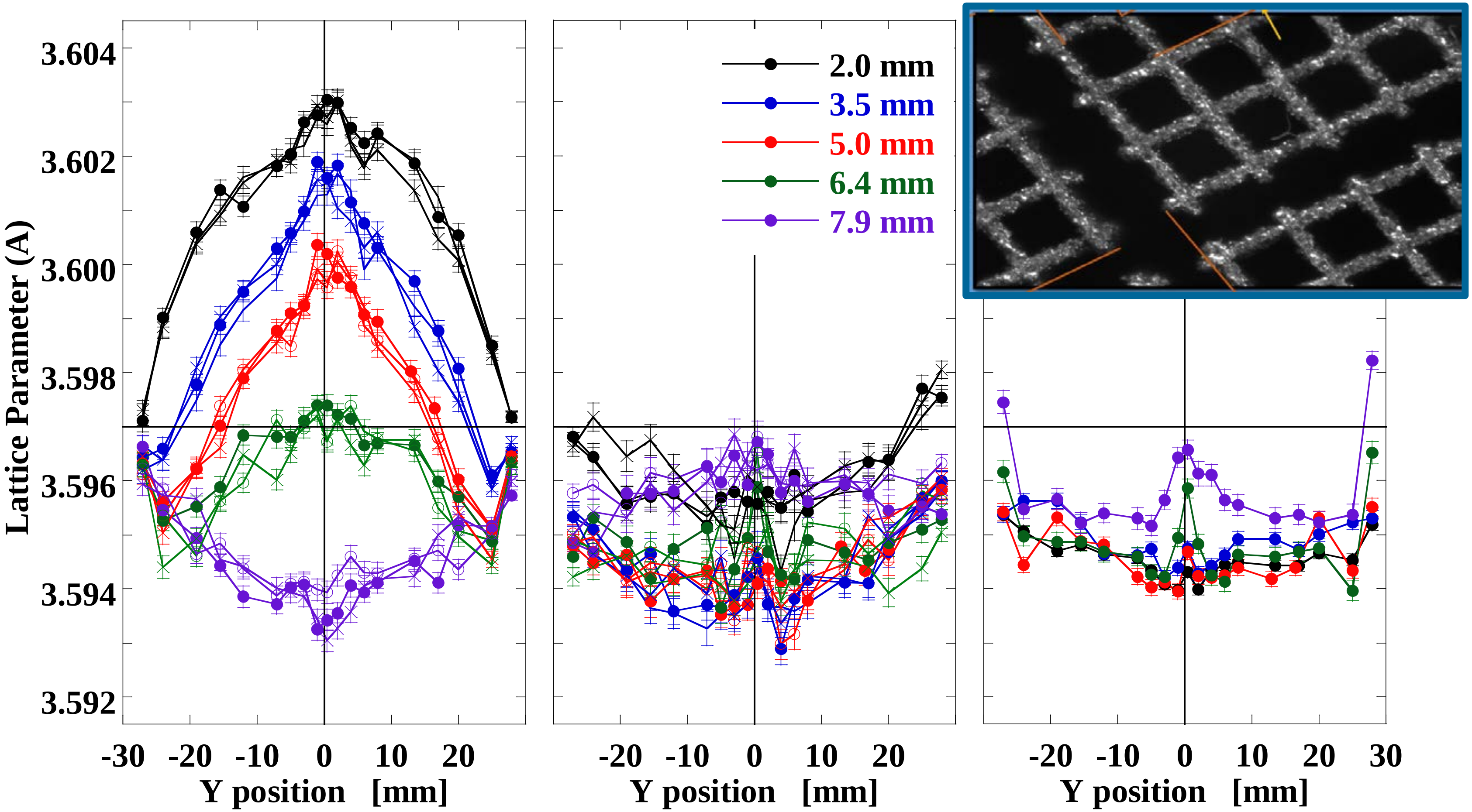
Stress is Independent of Hatching and Fragmentation



C: h = 0.25mm, f = 3mm
D: h = 0.25mm, f = 5mm
K: h = 0.35mm, f = 4mm

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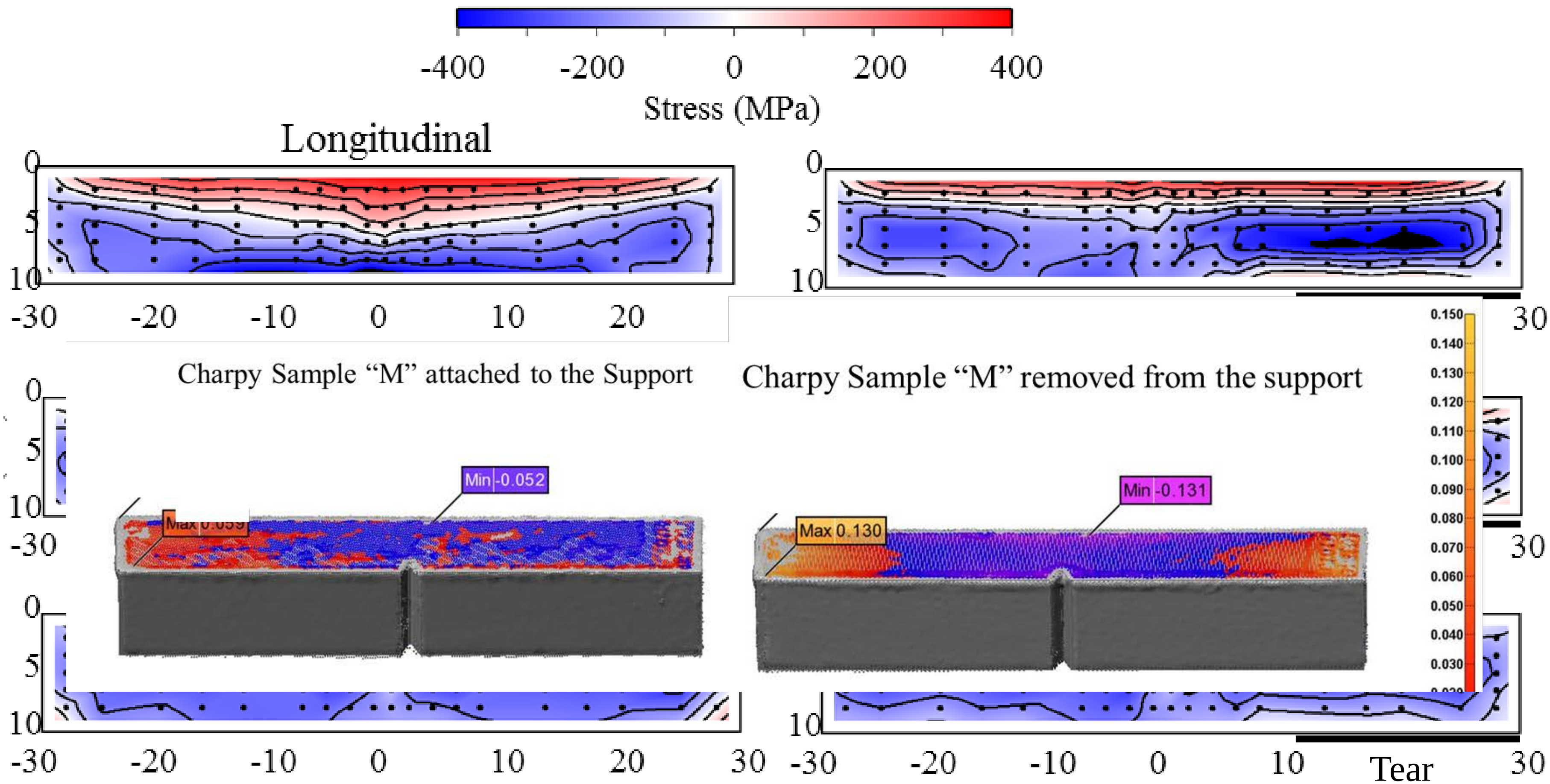
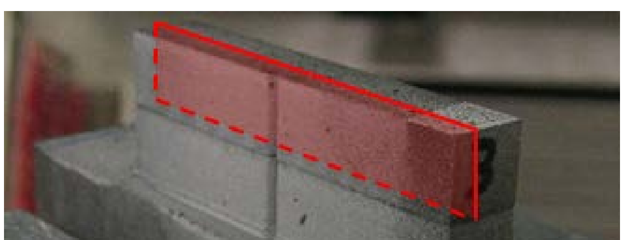
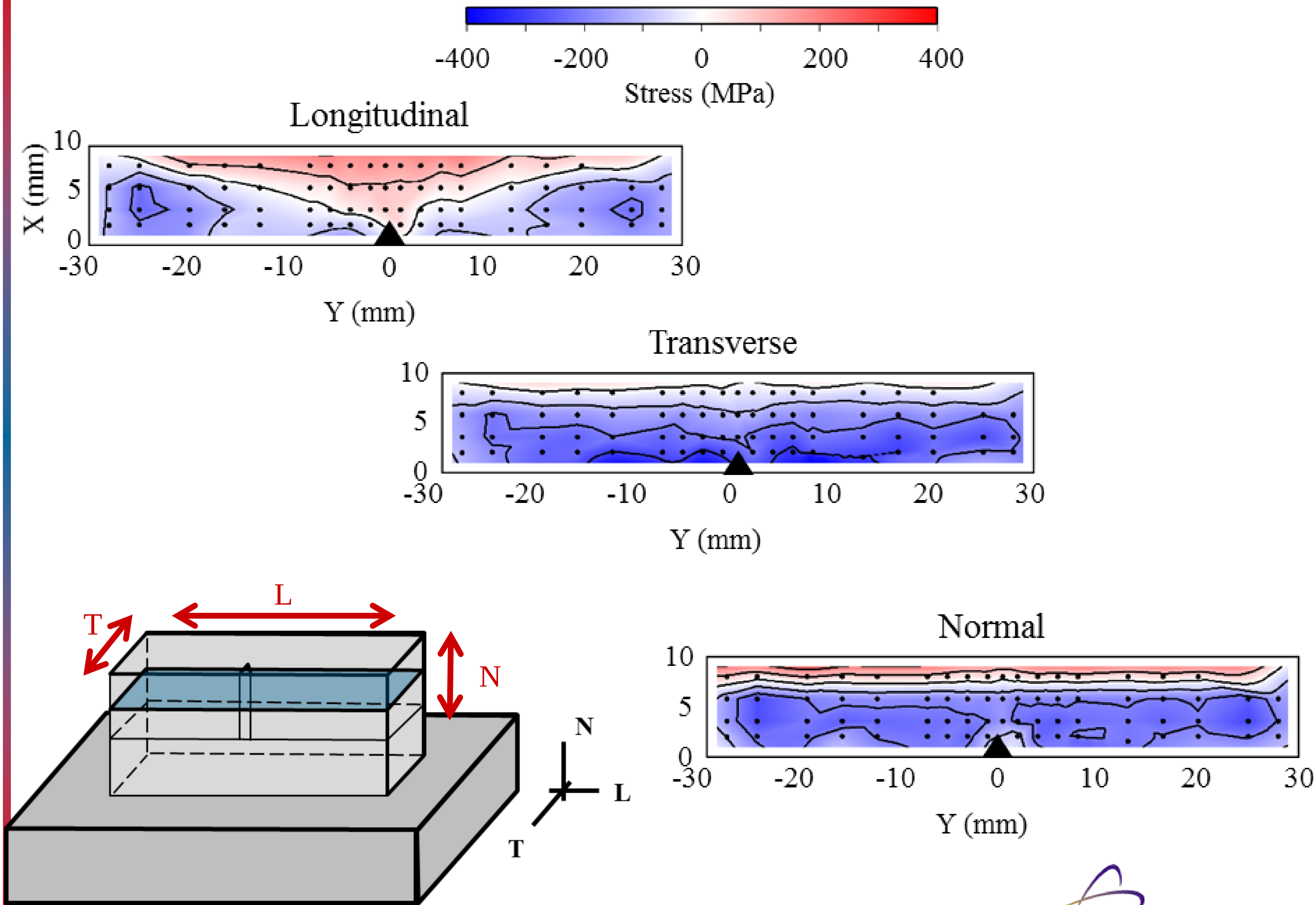
Stress is Strong and Predominantly Longitudinal





Tear During Deposition Has a Strong Effect on Residual Stress

Presence of Notch Results in an Asymmetry in the Stress Field



Conclusions

- Neutron and high energy synchrotron x-ray diffraction and imaging used to monitor microstructure and stress in engineering materials.
- Trying to understand Processing/Structure/Property/Performance relationship to that we can intelligently design and qualify novel materials processing.
  - Additive manufacture is a single example.
- X-ray diffraction has the promise for in-situ studies during deposition, with measurements in the millisecond time frame available today, microsecond or better in the future.
- Neutrons will be limited to post-processing and or operando or residual stress studies of material.
- Combined, they provide a window unto the entire PSPP relationship.

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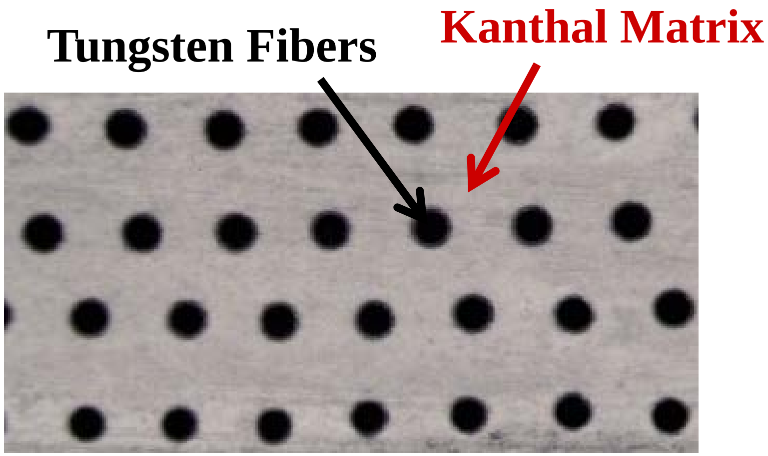
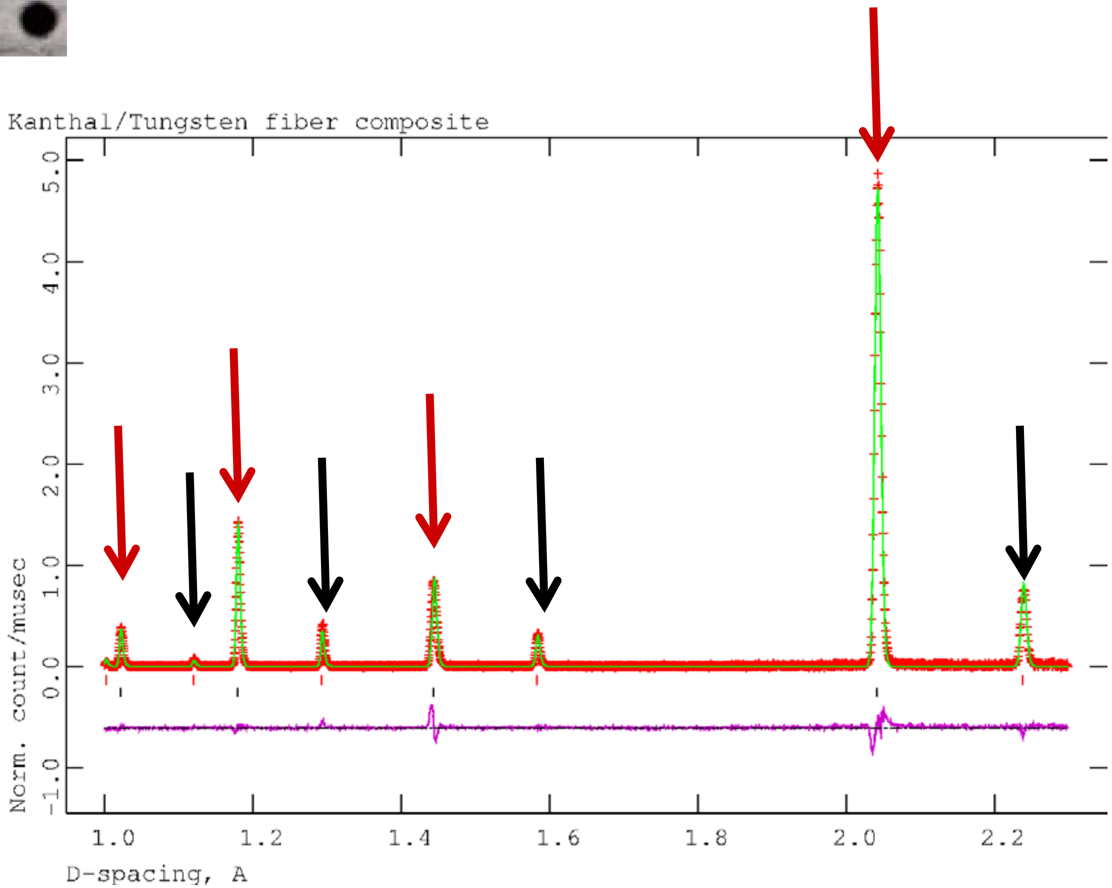
Diffraction Inherently Distinguishes Between Phases





Diffracted Peak Intensity is Directly Related to Pole Density

Extruded Magnesium