# Investigation of Grain Growth and Deformation in Nanocrystalline Metals: In-situ TEM Mechanical Testing and Crystallographic Orientation Mapping

SAND2017-8567C

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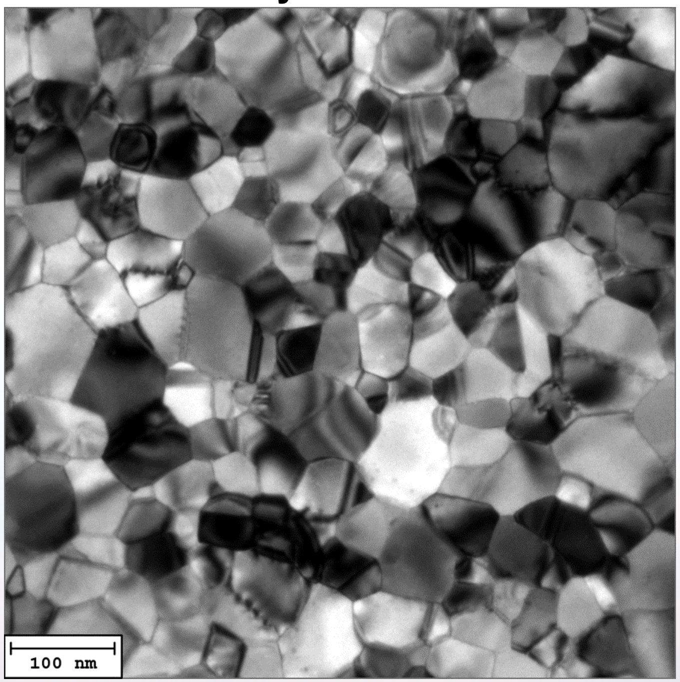
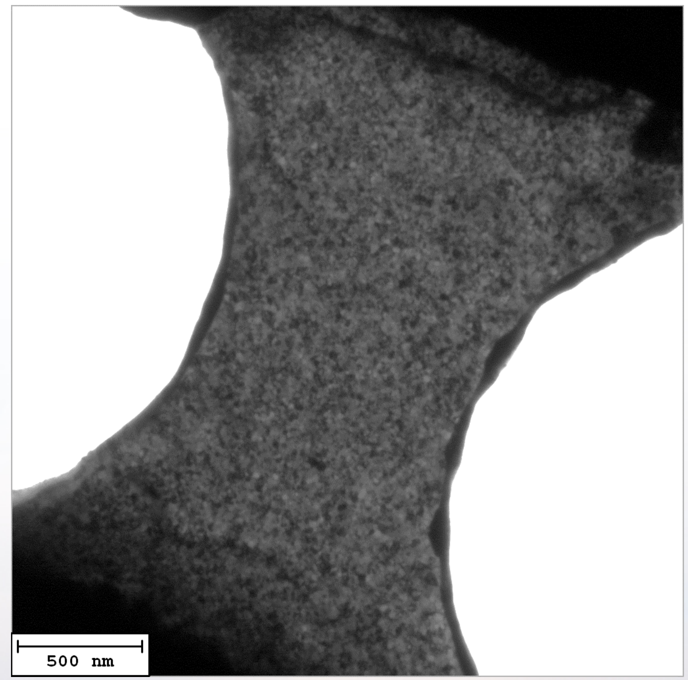
**Sandia National Laboratories**

**Key Project Collaborators: Douglas Stauffer and S.A. Syed, Hysitron, Inc.**

**Brad L. Boyce and William M. Mook, Sandia National Laboratories**




**Preliminary work to experimentally correlate the local grain orientation or grain boundary character in mechanical loading in the TEM**

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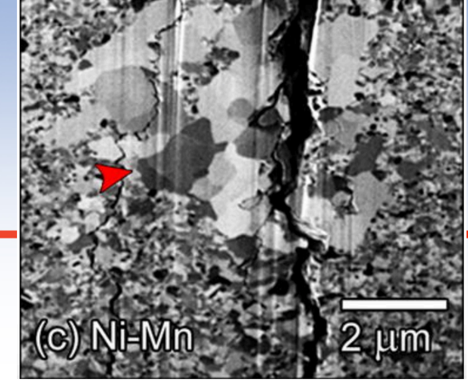
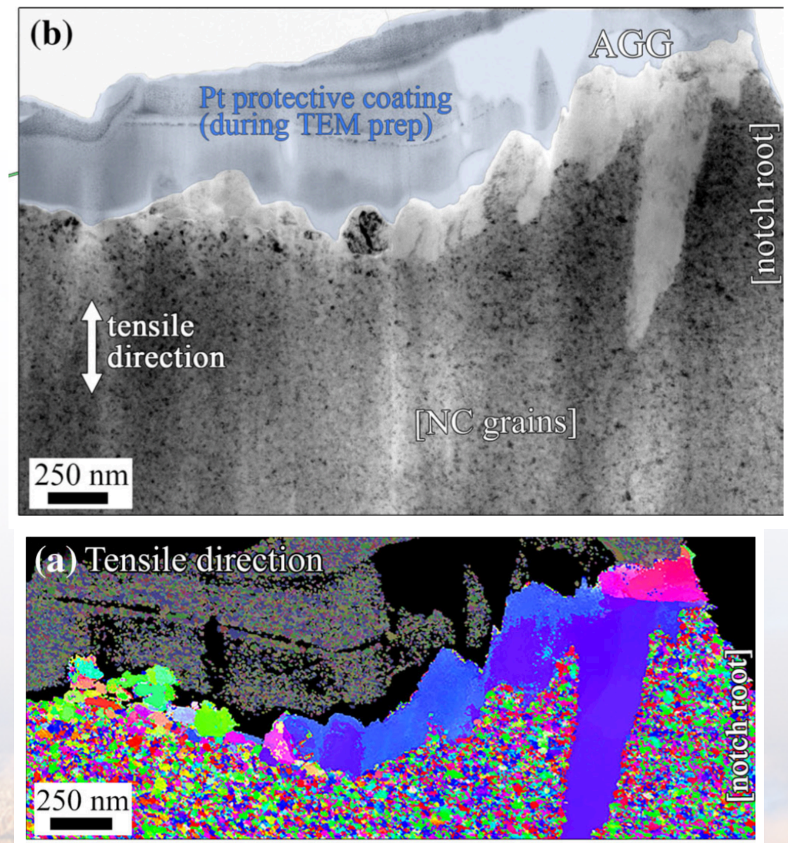
# Nanocrystalline Alloys: Fatigue and Fracture

- **Progressive microstructural changes with cyclic loading, often below yield stress**
- **Fatigue in nanocrystalline metals**
  - Grain boundary migration and grain growth
  - Crack initiation
- **What are the underlying mechanisms associated with these phenomena?**

- ***In situ* TEM deformation techniques provide the spatial resolution needed to investigate these questions**
- **Ideally coupled with bulk scale testing**




Boyce and Padilla, Met Trans A, **11** (2011)

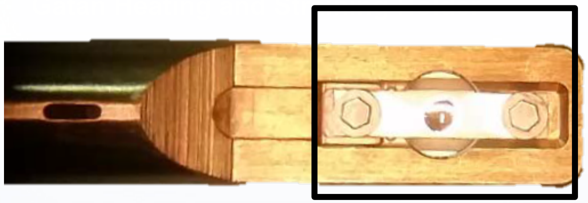



T.A. Furnish et al., J. Mat Sci **52**:46-59 (2017)

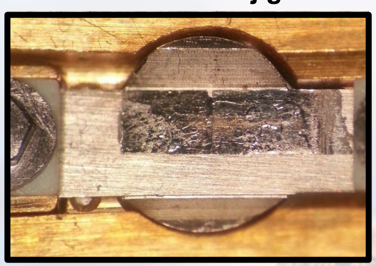
# *In situ* Qualitative Mechanical Testing

**Gatan straining TEM Holder**

- **Minimal control over displacement**
- **No out-of-box force information**
- **Successful in observing dislocation-GB interactions/mechanisms**

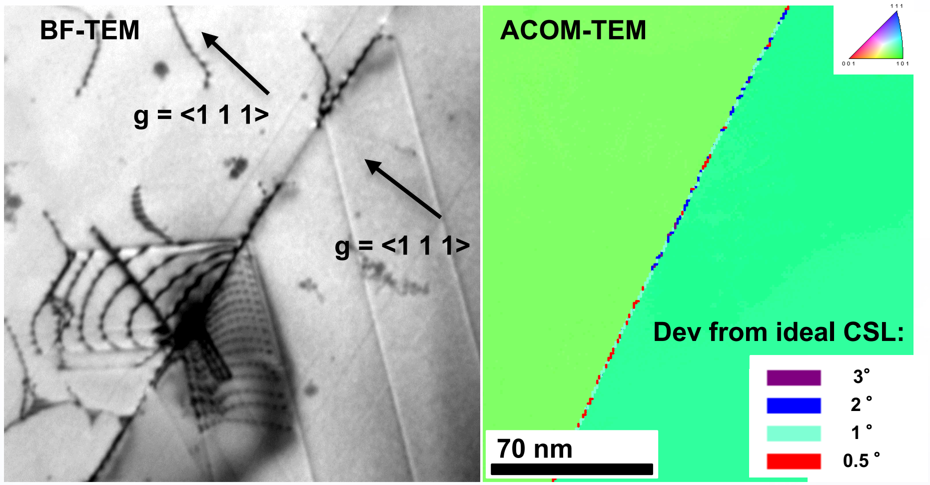


**Thin film tension "jig":**

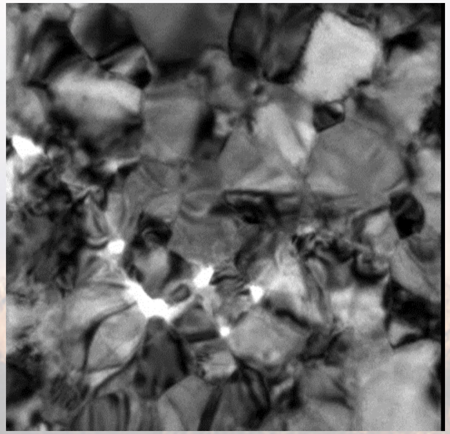


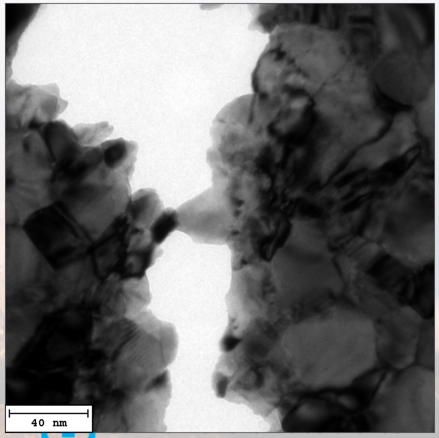
**Traditional jet thinned disk**

**Dislocation interactions as a function of GB character (Σ3 twin GB below):**



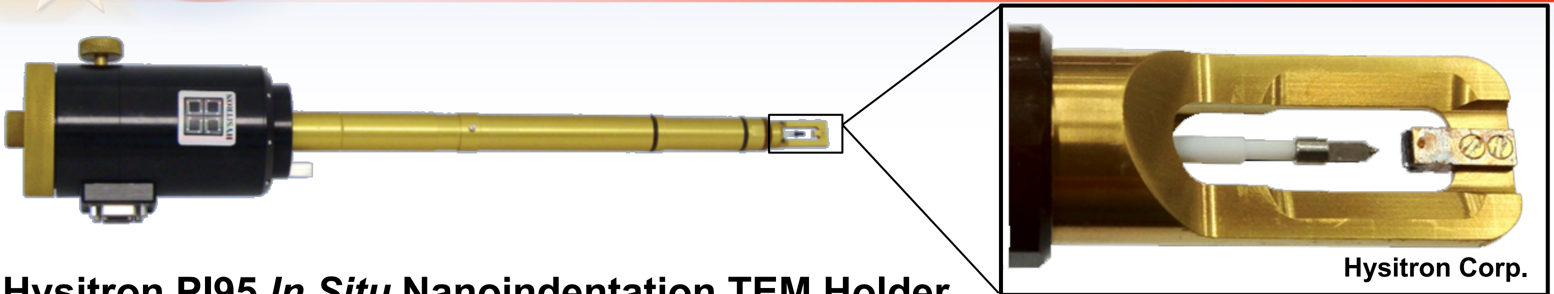

**Observe deformation mechanisms in nanocrystalline metals during tensile straining:**





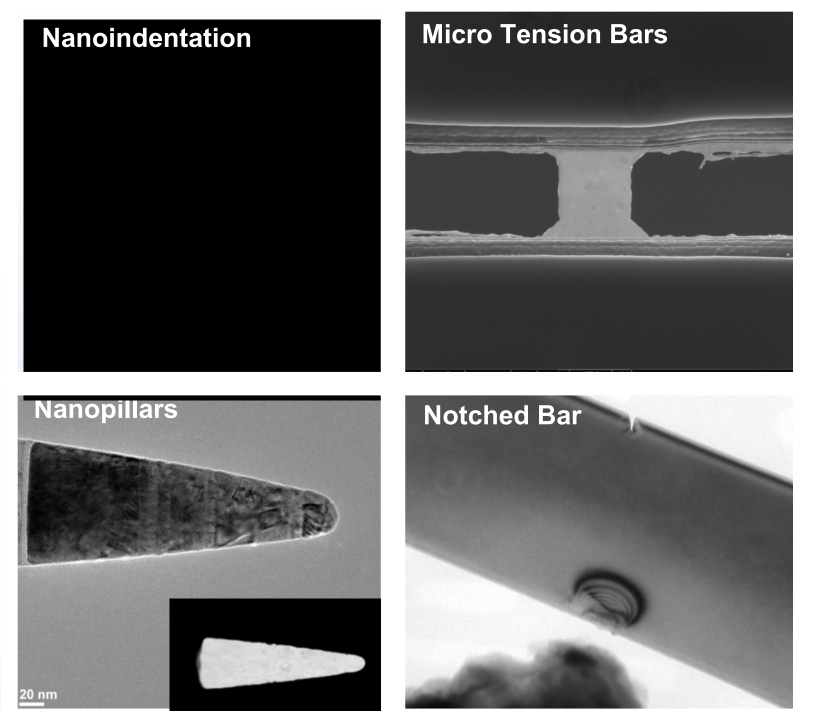
# *In situ* Quantitative Mechanical Testing

Collaborators: Douglas Stauffer and Eric Hintsala, Hysitron Inc.




**Hysitron PI95 *In Situ* Nanoindentation TEM Holder**

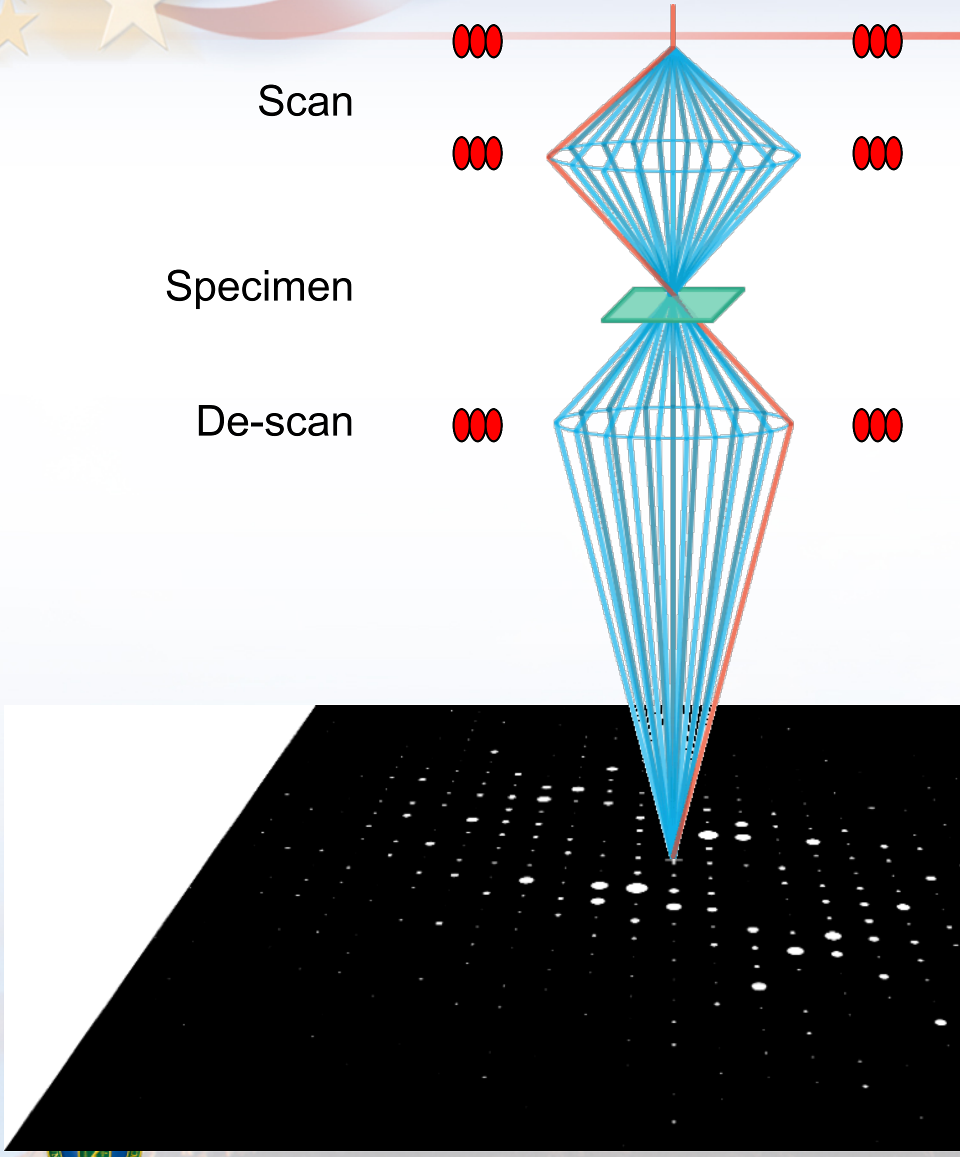
- **Sub nanometer displacement resolution**
- **Quantitative force information with μN resolution**
- **Concurrent real-time imaging**

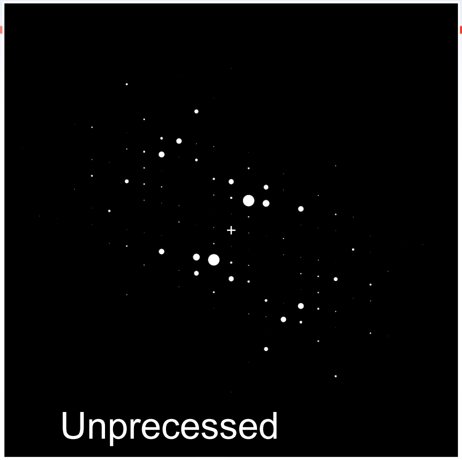


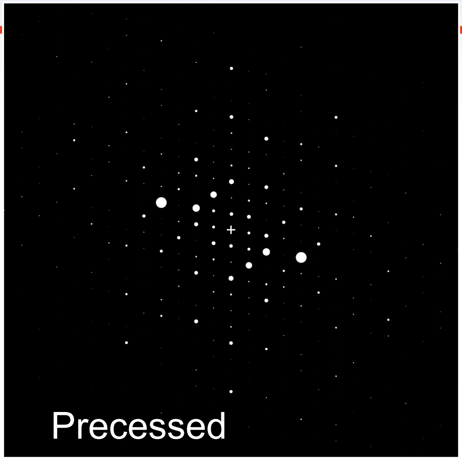

- A variety of sample geometries
- Load functions examined at I³TEM:
  1) Indentation
  2) Tension
  3) Fatigue
  4) Creep
  5) Compression

# Precession Electron Diffraction (PED) Microscopy

Collaborators: K.J. Ganesh, S. Rajasekhara



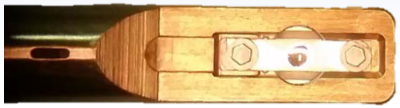

**Advantages**

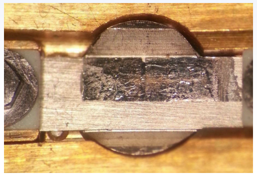
- **< 8 nm spatial resolution (LaB6)**
- **< 2 nm spatial resolution (FEG)**
- **Near kinematical electron diffraction**
- **Symmetry ambiguities are resolved**
- **Fast and automated acquisition**

Chris Own, PhD Dissertation, Northwestern University, 2004 Slide courtesy: K. J. Ganesh, K. Hattar

# In-Situ Mechanical Testing with ACOM: NC Pt

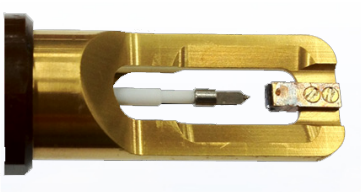
- **Qualitative Testing:**

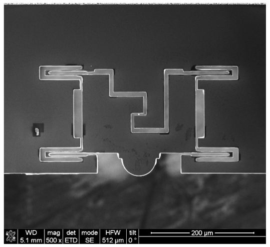




  - **Example 1:** Crack propagation in nanocrystalline Pt
    - Crack blunting and deformation mechanisms coupled with ACOM-TEM

- **Quantitative Testing:**

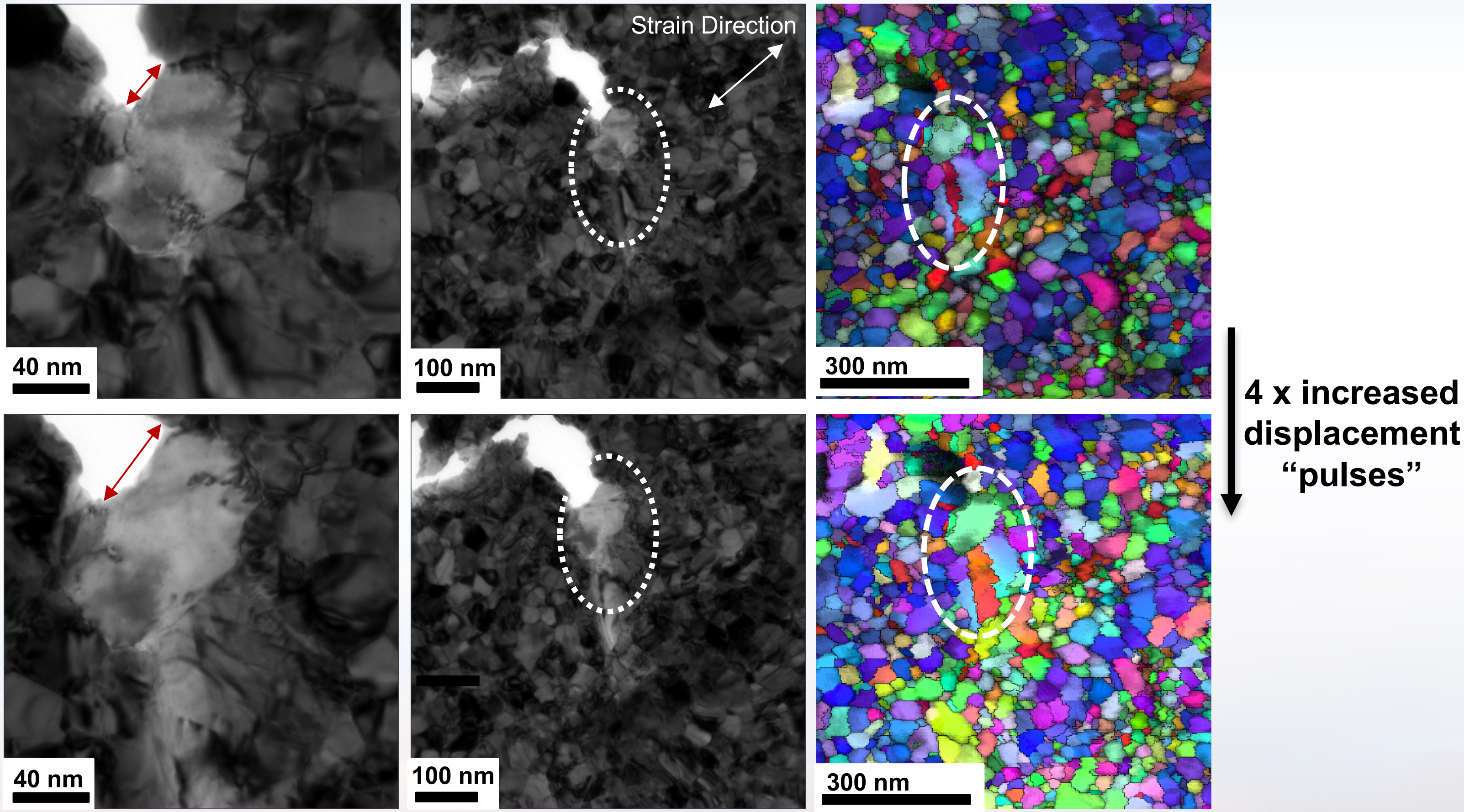




  - Example 2: “Push-to-pull” monotonic, cyclic loading, and notched fatigue of nanocrystalline Cu
    - Grain growth and crack propagation coupled with ACOM-TEM
  - Example 3: “Push-to-pull” cyclic loading (fatigue) of nanocrystalline Pt
    - Crack initiation and growth coupled with ACOM-TEM

# *In situ* Qualitative Mechanical Testing in NC Pt

1) Magnetron Pt Deposited on NaCl 2) Floated off onto tensile “jig” 3) 500$^{o}$C 20 minute anneal




- Crack tip blunting (95% increase in crack tip width) and clear grain elongation and thinning
- Observation of in-grain dislocation migration between displacement pulses
- ACOM confirms the grain growth/elongation in front of crack blunt

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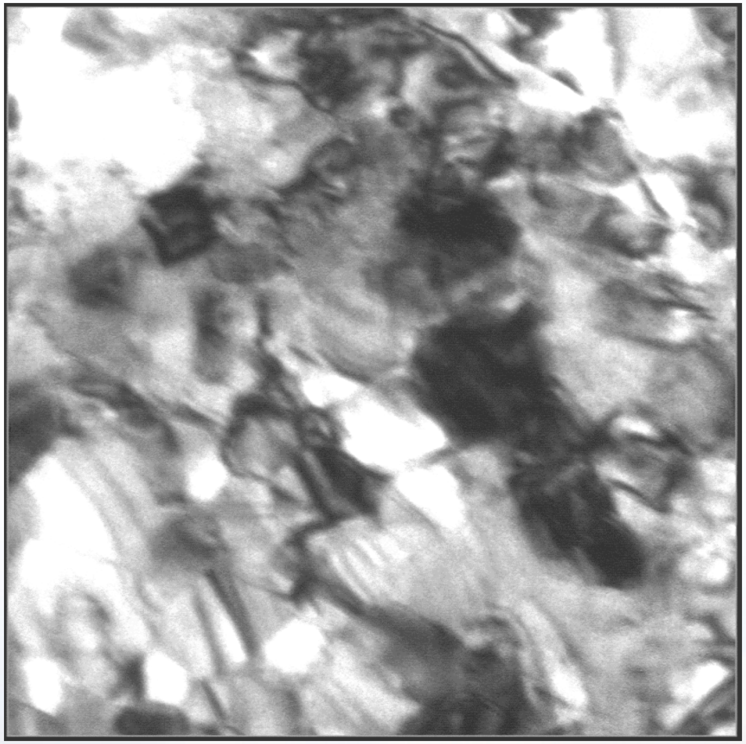
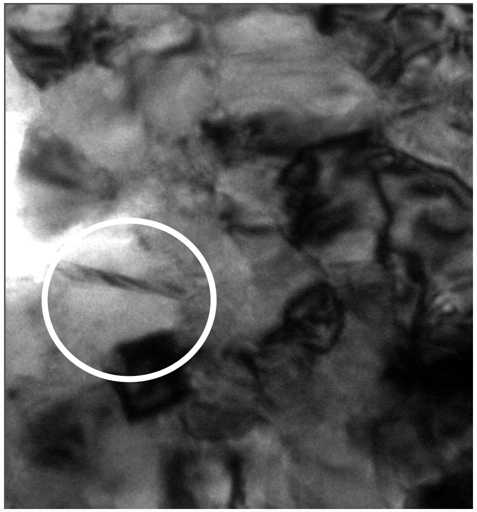
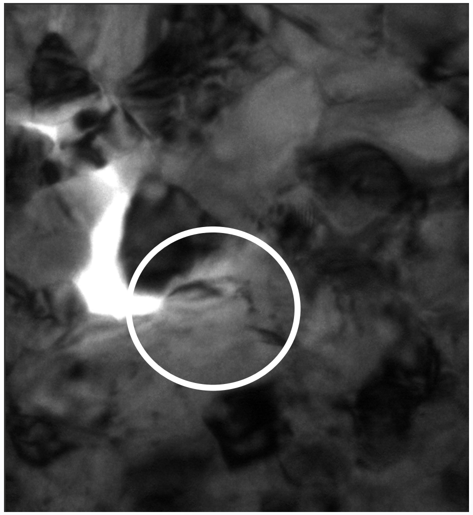
# *In situ* Qualitative Mechanical Testing in NC Pt

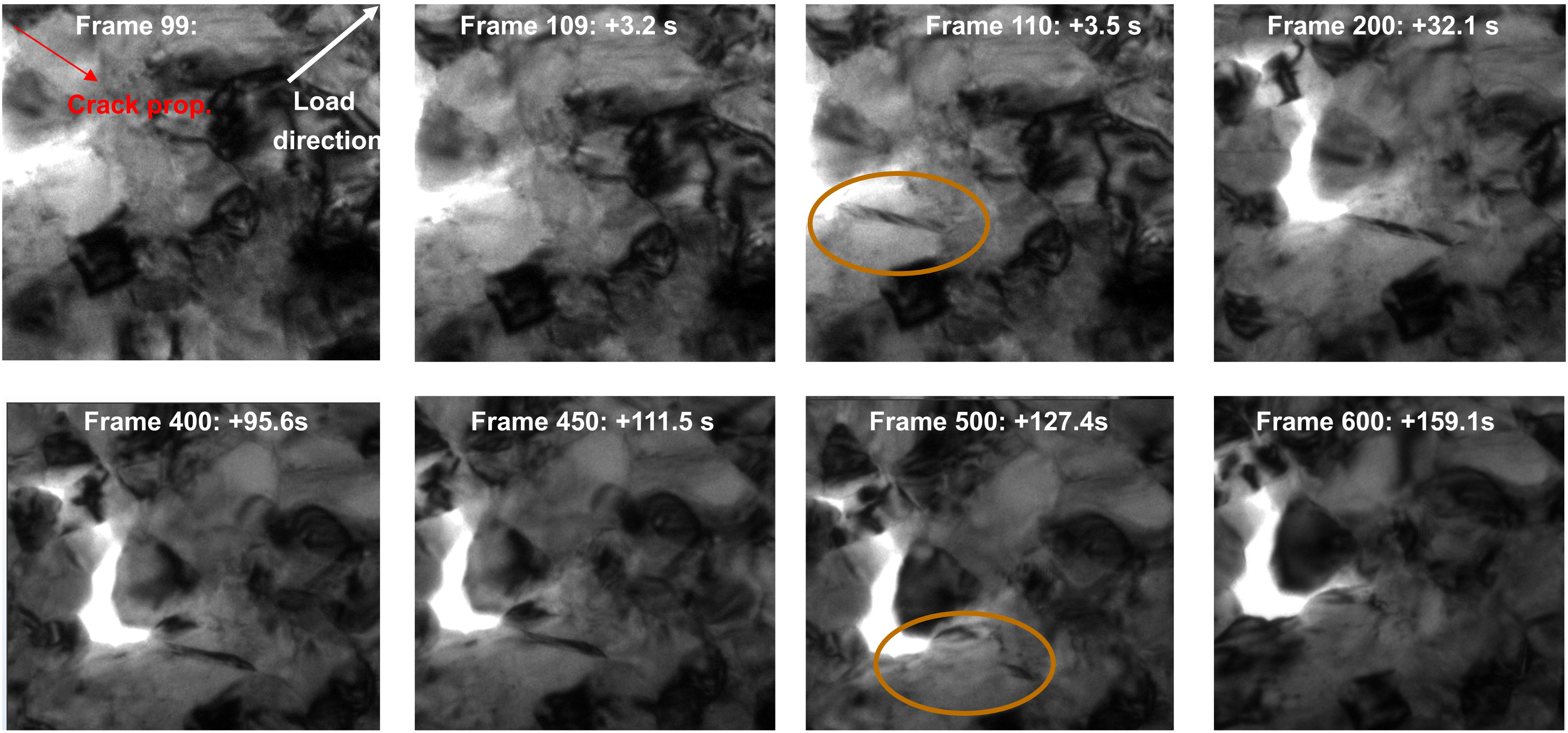






- **Observed the potential nucleation and de-twinning adjacent to a high strain region (crack tip blunt)**
- **Immediately after apparent de-twinning, crack propagates**

# NC Pt: Twin Boundary Interaction During Loading




- **Circled contrast: twin boundary “nucleates” and “de-twins” under individual pulse loading**

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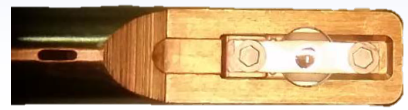
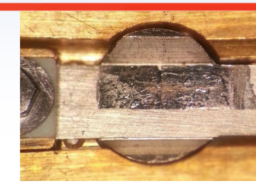
# In-Situ Mechanical Testing with ACOM: NC Cu

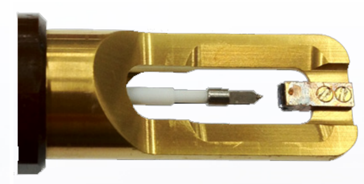
- **Qualitative ("Classical") Testing:**

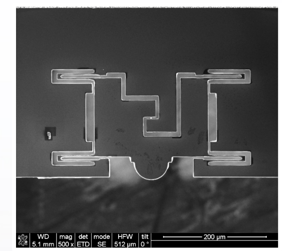




  - ✓ Example 1: Crack propagation in nanocrystalline Pt
    - Crack blunting and deformation mechanisms

- **Quantitative ("Modern" )Testing:**

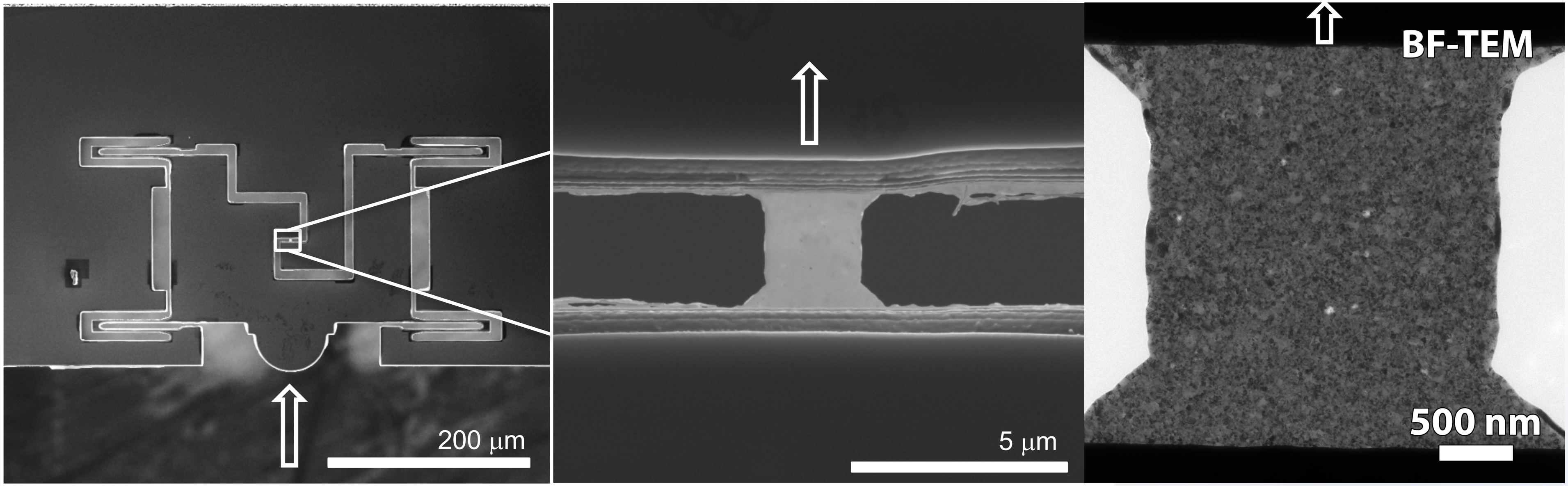




  - **Example 2:** "Push-to-pull" monotonic, cyclic loading, and notched fatigue of nanocrystalline Cu
    - Grain growth and crack propagation coupled with ACOM-TEM
  - Example 3: "Push-to-pull" cyclic loading (fatigue) of nanocrystalline Pt
    - Crack initiation and growth coupled with ACOM-TEM

# Tension Specimen Fabrication

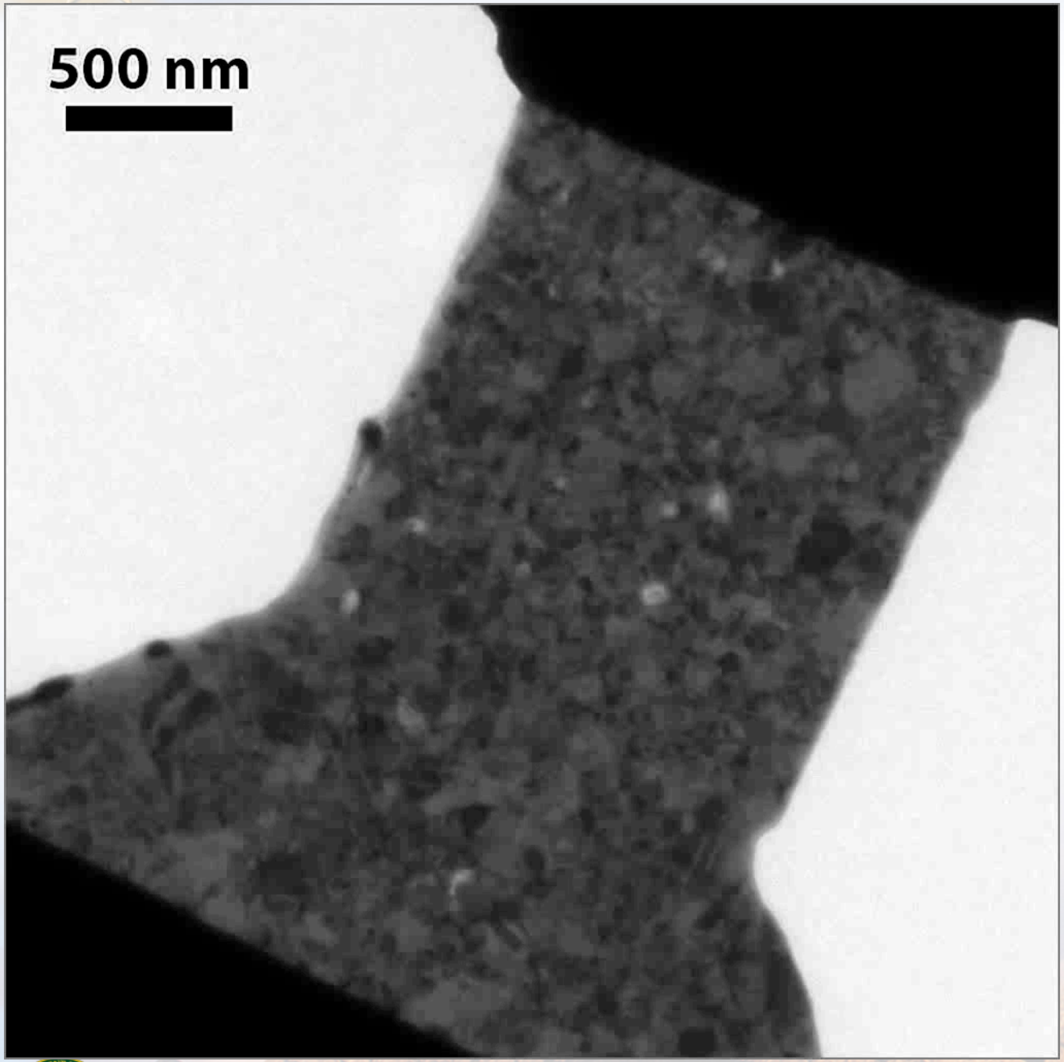
- **Hysitron "Push-to-Pull" devices**
  - **Microfabricated Si test frame**
  - **Cu film (75 nm) floated onto device, then FIB milled.  Final FIB cut: minimal or no I-Beam**

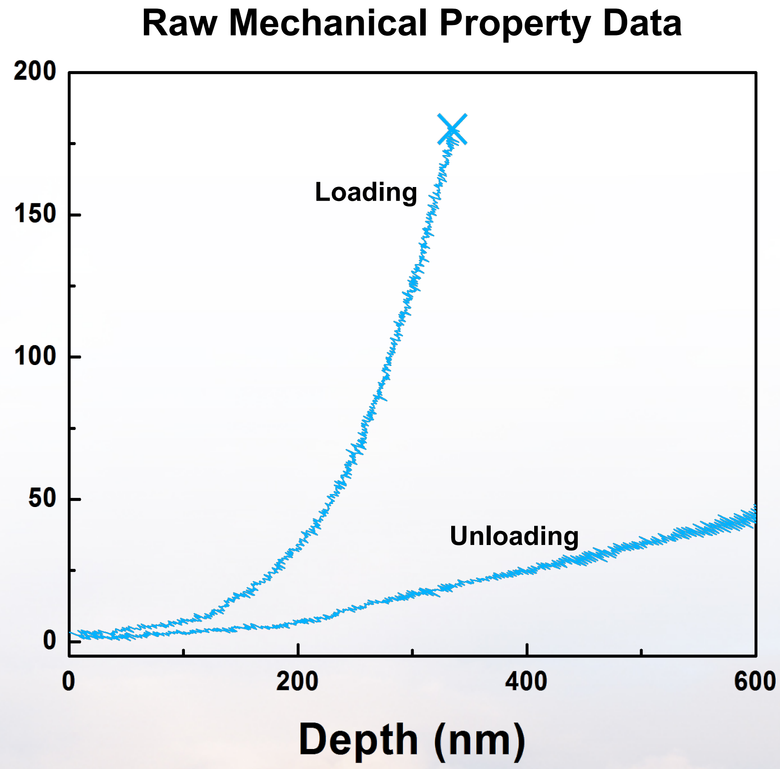



- **Nearly pure tension, uniform cross sectional area, stable load frame**
- **Sensitive to shape of edges, issues with magnetic materials**

# *In situ* TEM Monotonic Tension Testing: NC Cu

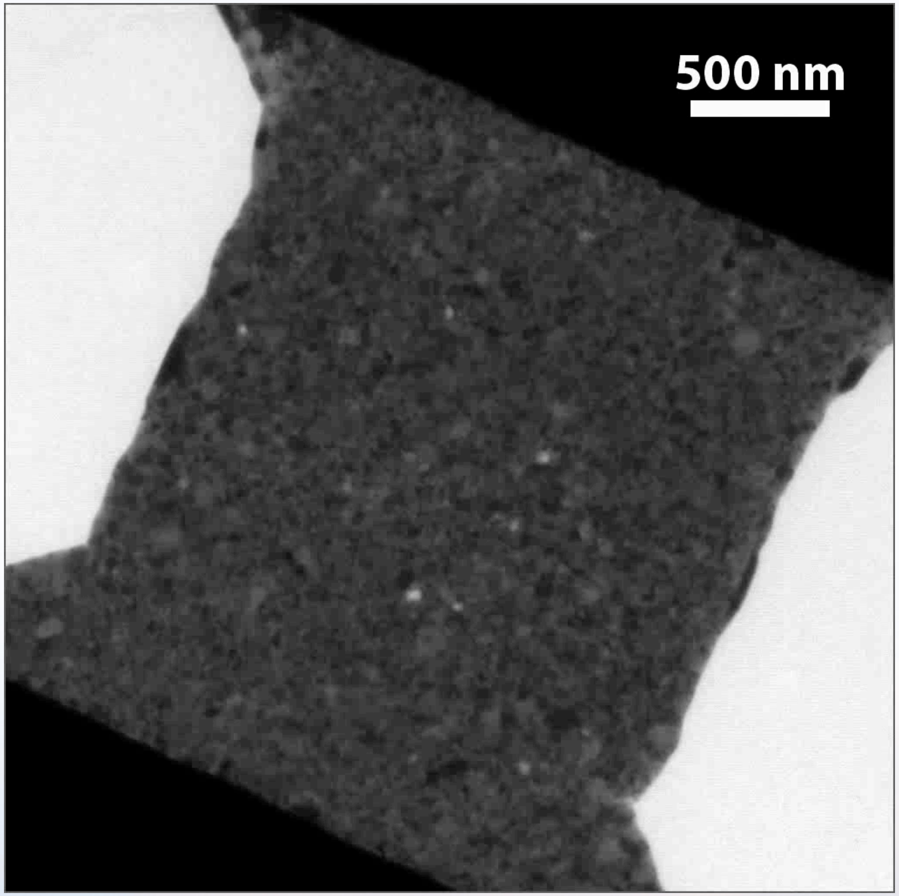
Video playback ×0.5



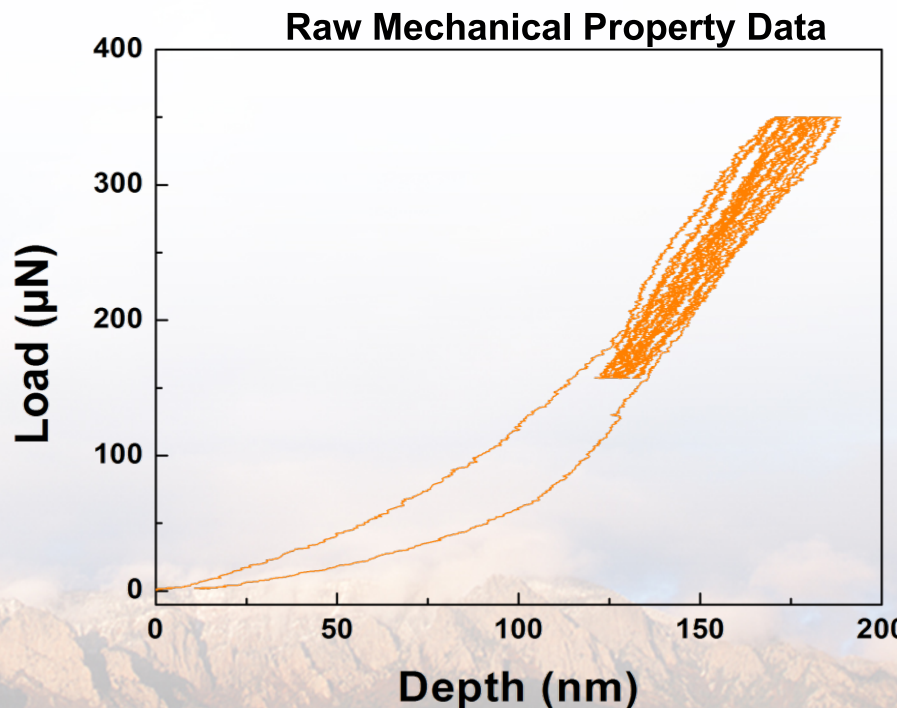
# *In situ* TEM Low Cyclic Tension Testing: NC Cu

Video playback ×10



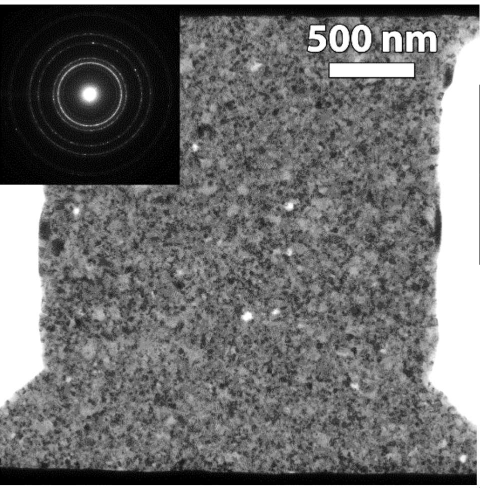

- Steady crack propagation
- Possible evidence of grain growth

- **Cyclic loading:**
  - Crack initiated in previous monotonic test
  - 9 cycles to ≈ 87.5% of that load
  - 50% unloading
  - Slow crack propagation

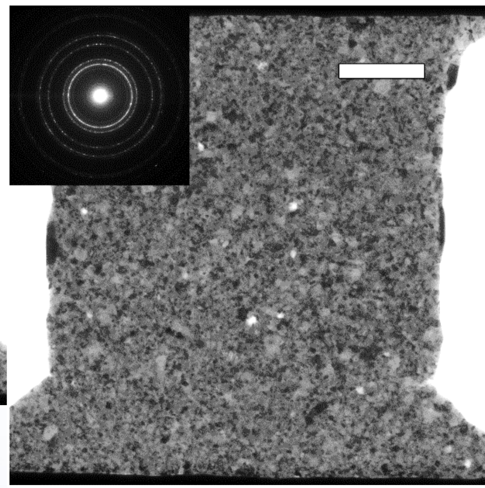


# Cyclic Tension *In Situ:* NC Cu

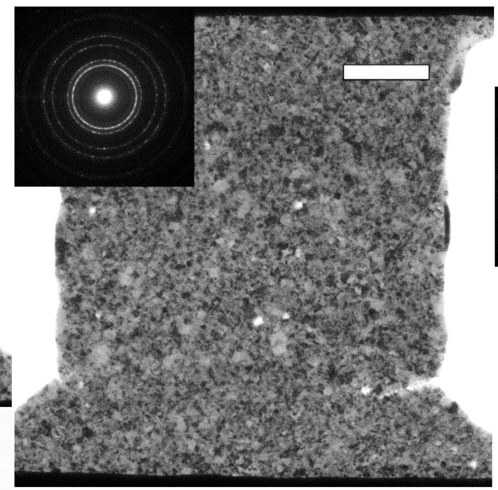
Before



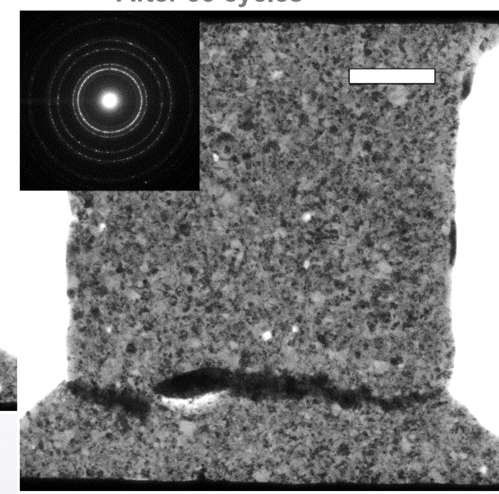

After 1 cycle



After 10 cycles



After 33 cycles

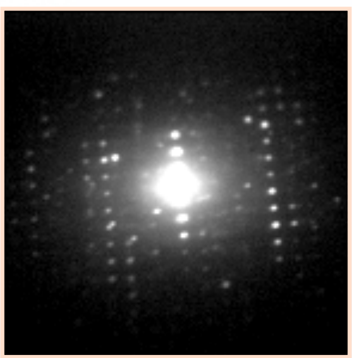


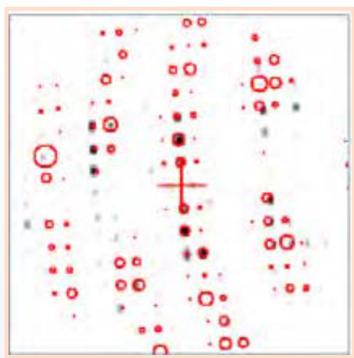
(Post-failure)

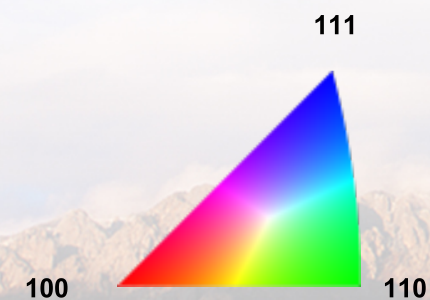
- **Wealth of information from one sample:**
  - Images and electron diffraction at each stage
  - Video and force/displacement during load cycles
- **Microstructural change still elusive**
  - Difficult to confirm and quantify

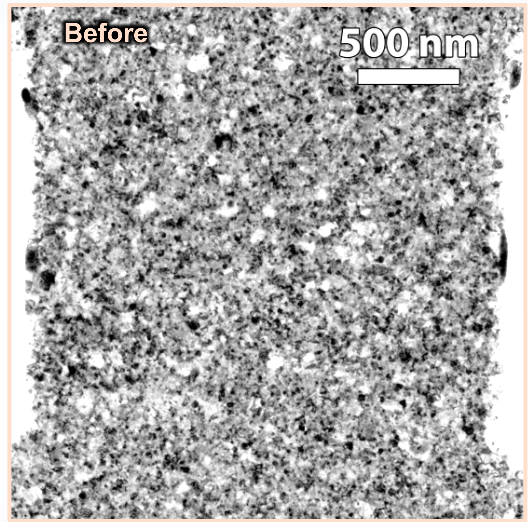
# Quantifying Microstructural Change

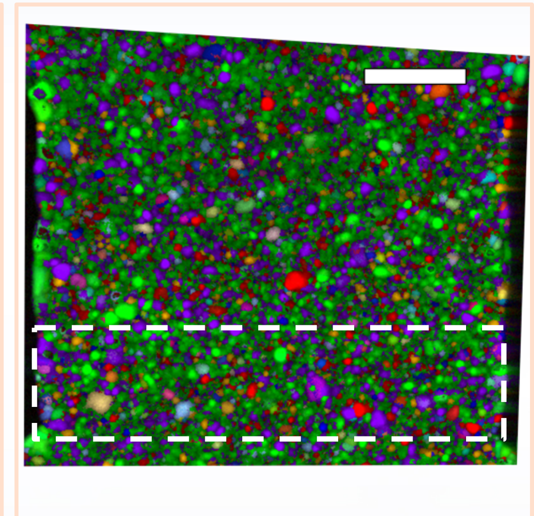
- Combining orientation mapping with deformation
- Provides a wealth of statistical data from the entire gauge section

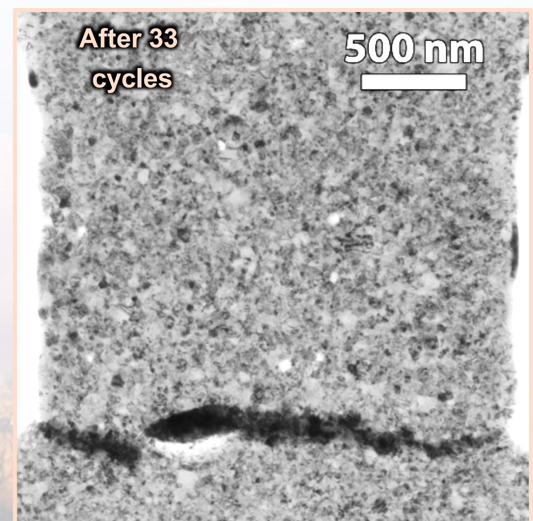


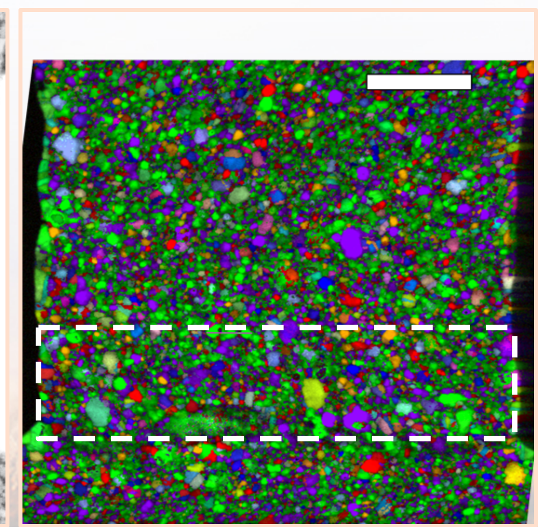




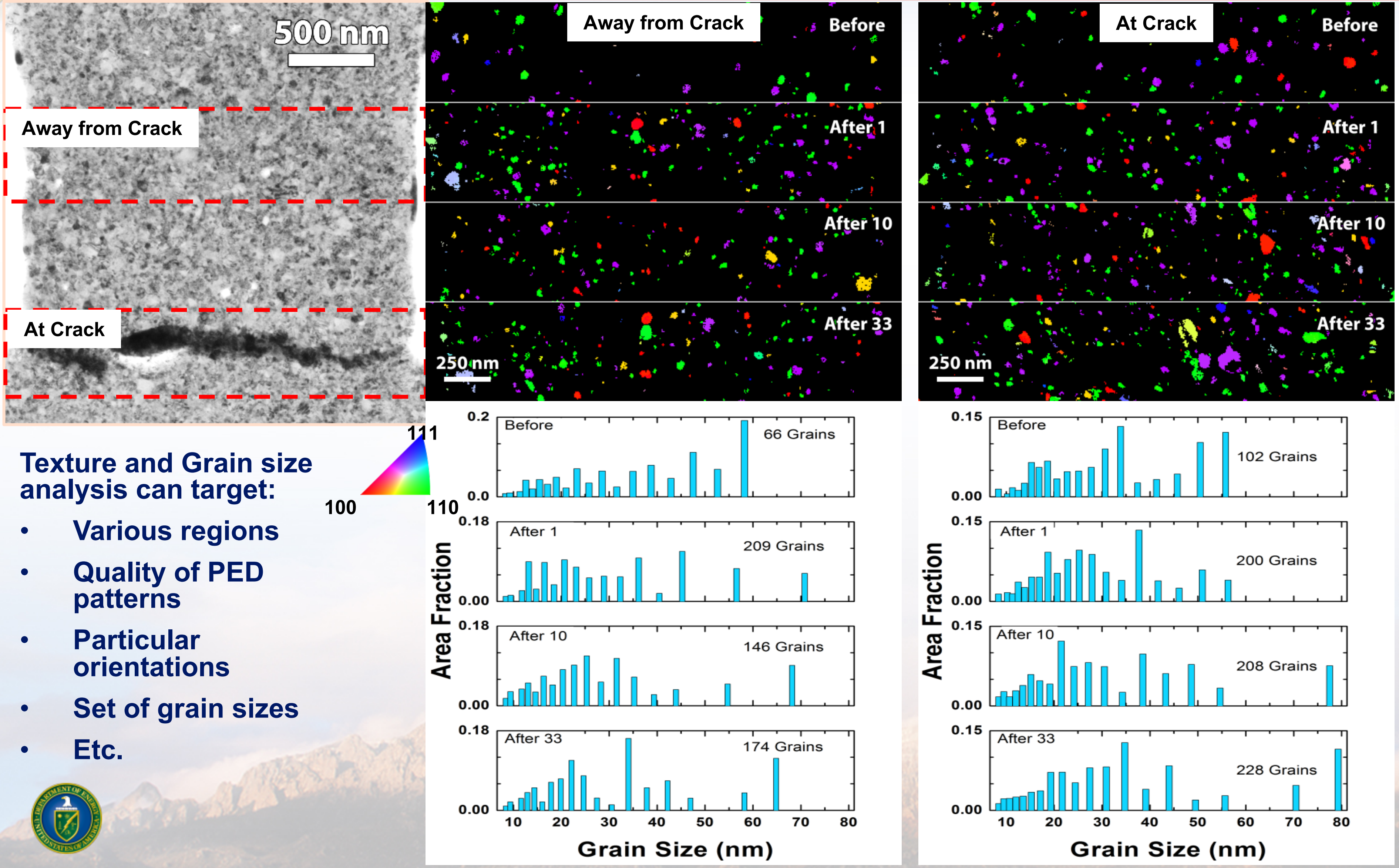





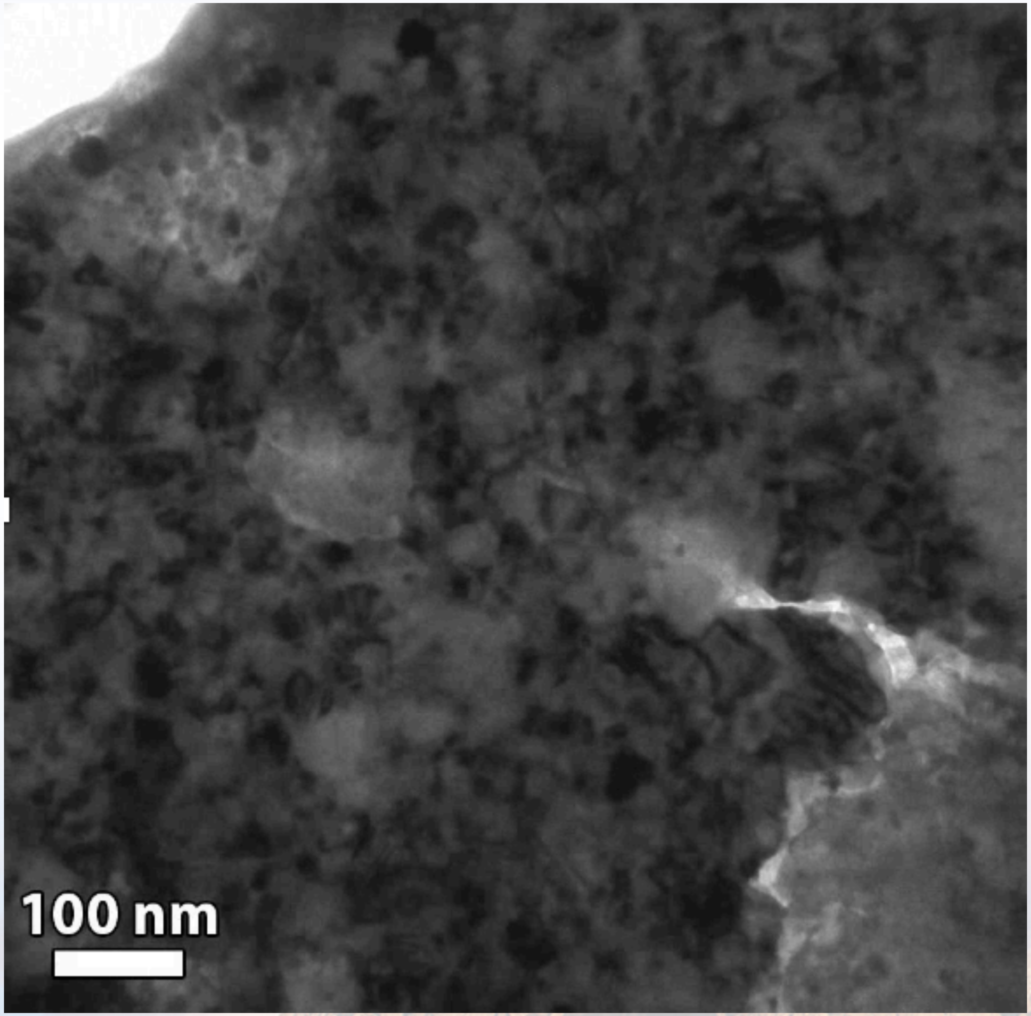
# Comparison of Grain Size Near and Far from the Fracture Surface




**Texture and Grain size analysis can target:**

- Various regions
- Quality of PED patterns
- Particular orientations
- Set of grain sizes
- Etc.

# Grain Growth and Crack Propagation during *In Situ* TEM Fatigue




- **200 Hz**
- Mean Load = 150 μN
- Load Amplitude = 101μN
- Sample drift rate of 0.04 nm/s

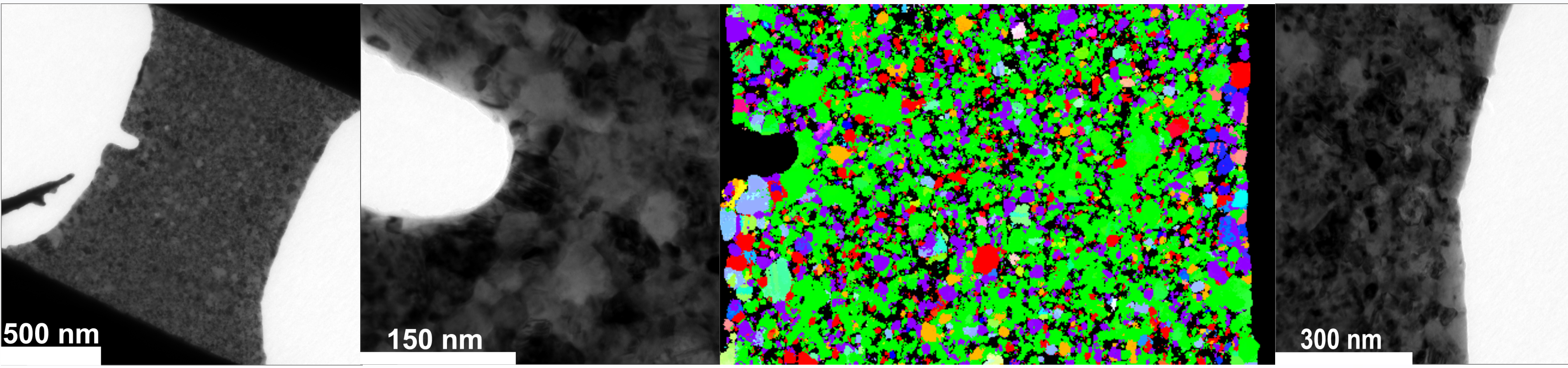
- Grain growth and crack propagation occur in 11,400 cycles (57 s)

- Crack growth rate of 10 nm can be measured over 113 nm over 20,000 or 6 pm/cycle!

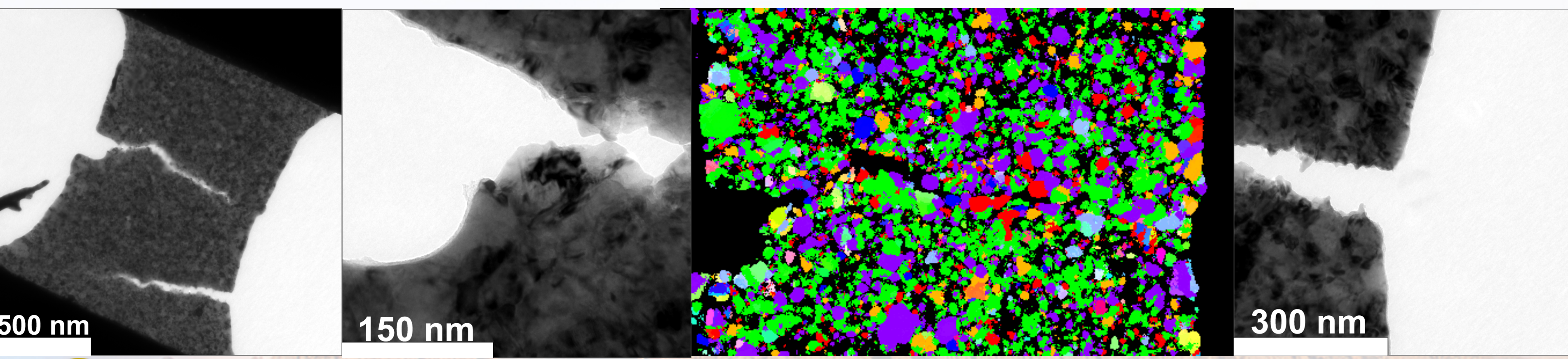
DC Bufford et al., Nano Letters 2016, 16 (8) 4946-4953

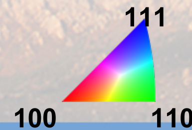
# Initial Investigation into Notched Fatigue Fracture

## Notch Prior to Testing




## After Crack Initiation during Fatigue Testing

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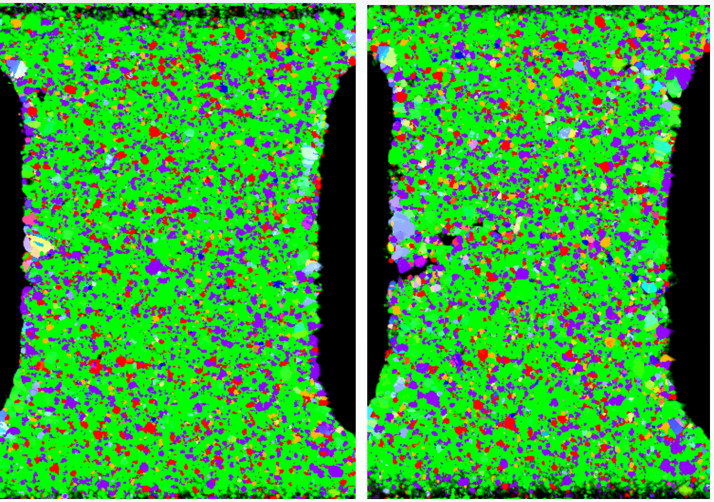
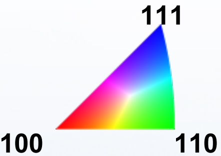
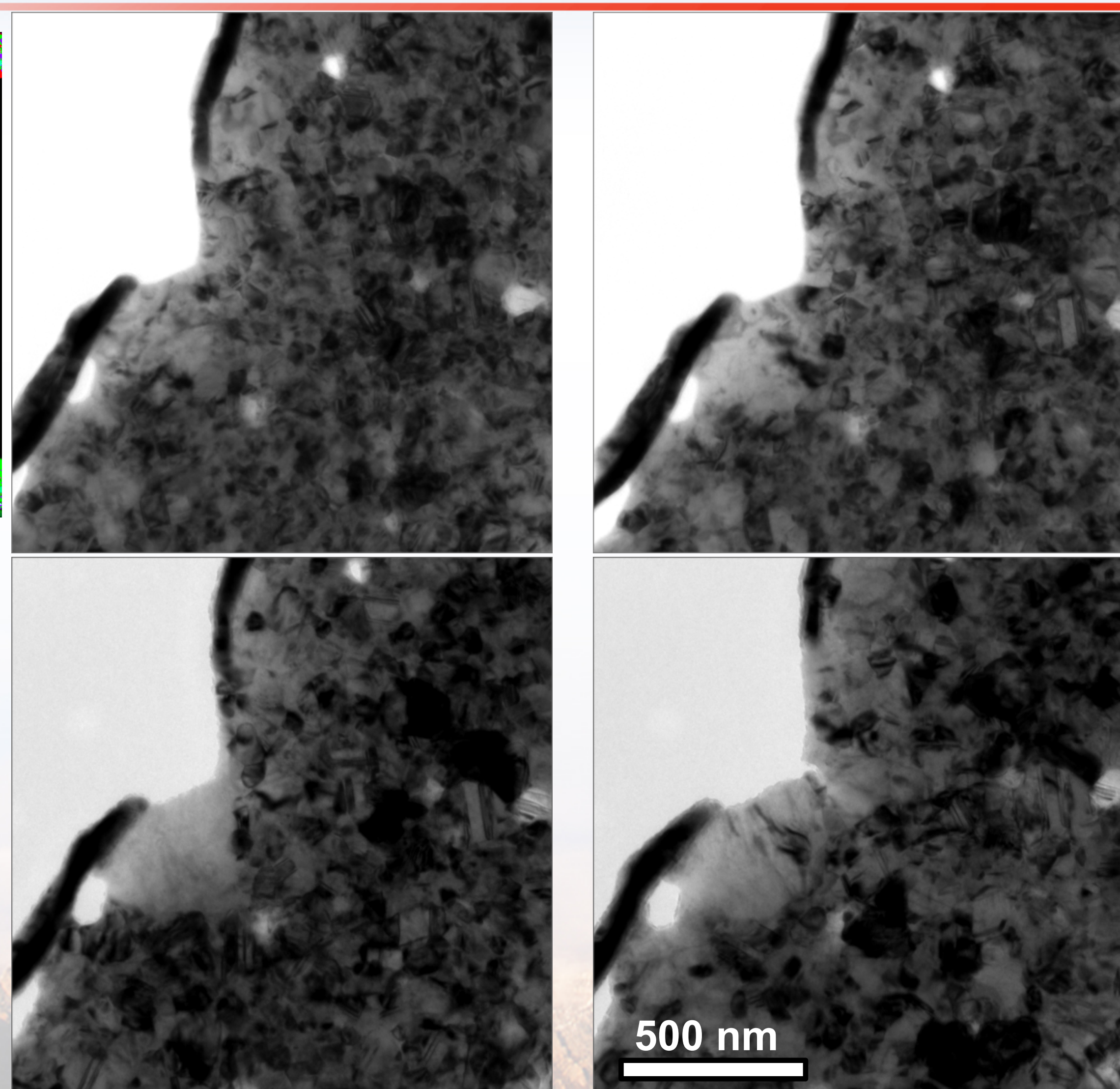
# Grain Growth Observed Directly at the Notch



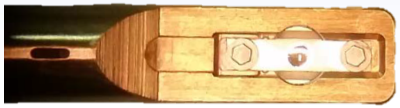


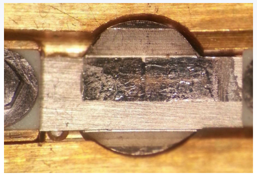

- Nucleation of the fatigue crack (10 nm) directly identified at the notch during high-cycle fatigue!
- Grain growth and thinning at the notch were clear.



# In-Situ Mechanical Testing with ACOM: NC Pt

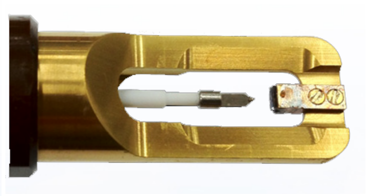
- **Qualitative Testing:**

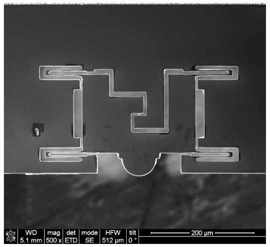




  - ✓ Example 1: Crack propagation in nanocrystalline Pt
    - Crack blunting and deformation mechanisms

- **Quantitative Testing:**



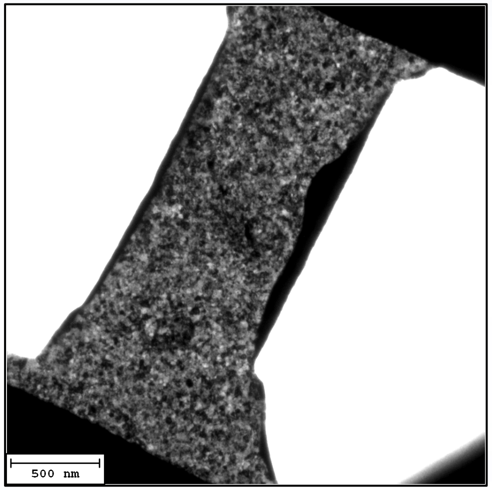


  - ✓ Example 2: "Push-to-pull" monotonic, cyclic loading, and notched fatigue of nanocrystalline Cu
    - Grain growth and crack propagation coupled with ACOM-TEM

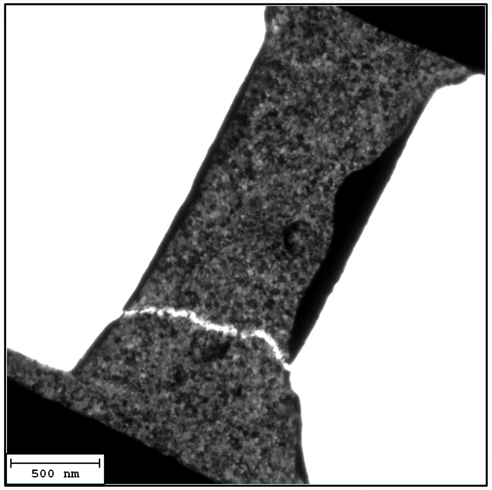
  - **Example 3**: "Push-to-pull" cyclic loading (fatigue) of nanocrystalline Pt
    - Crack initiation and growth coupled with ACOM-TEM

# In-situ Cyclic Fatigue: NC Pt

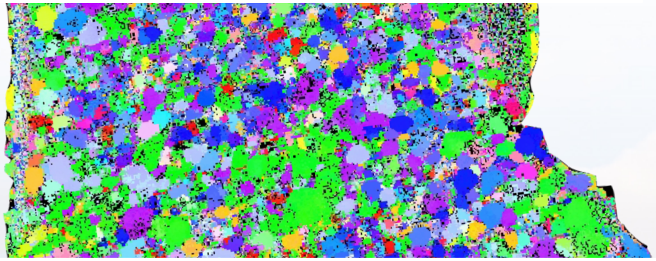
Pre-Test



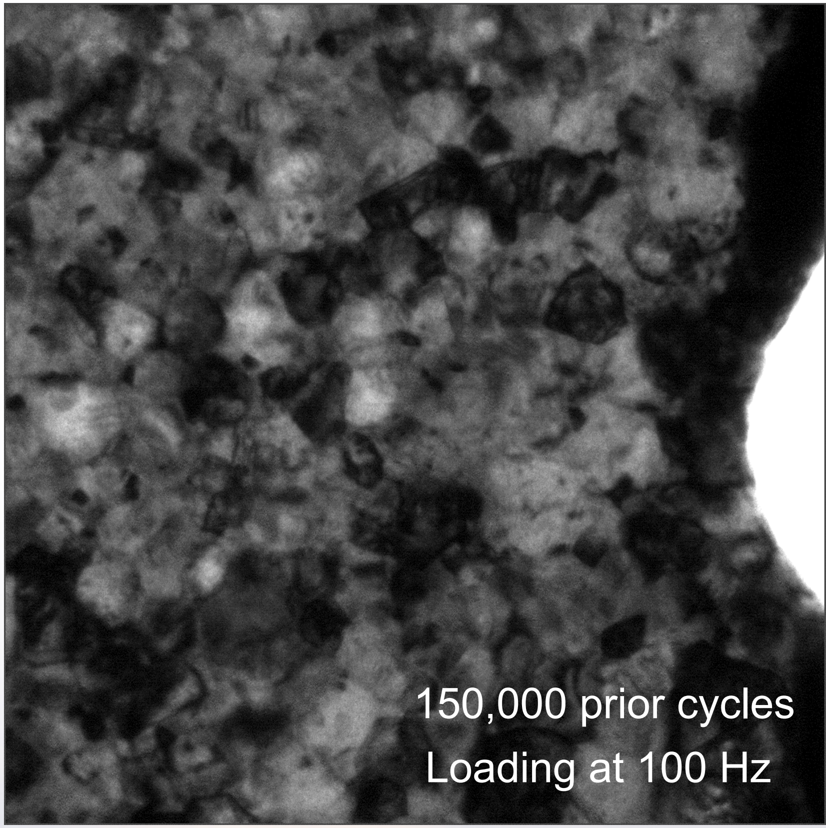

Post-Test




ACOM Pre-Test in failure region

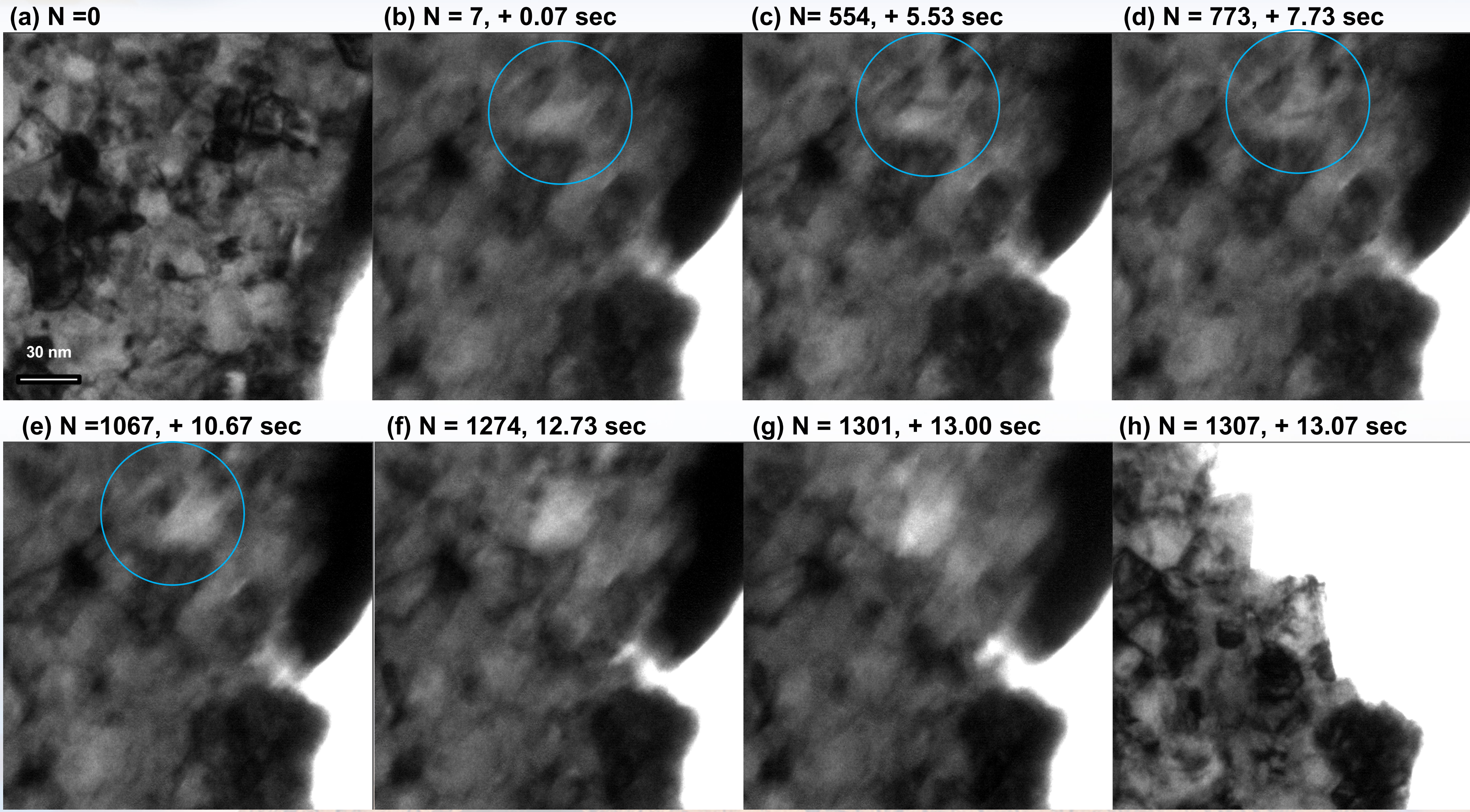


Mean Load: 110 uN, Oscillating Load: 35 uN




- Observation of likely dislocation activity prior to crack opening
- Crack growth as a function of cycles (N)
- Dislocation/GB activity observed in front of migrating crack

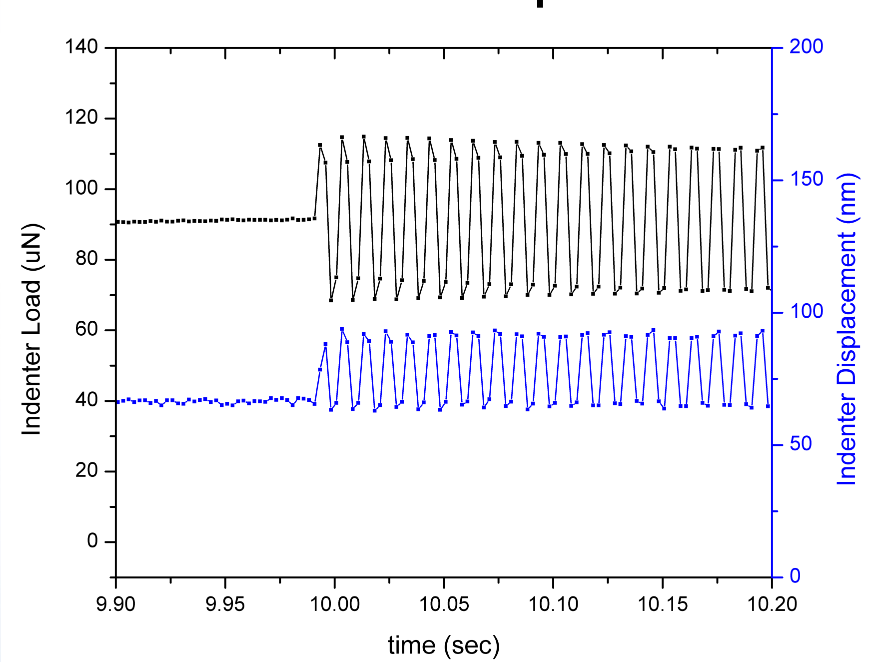
# In-situ Crack Tip Evolution Until Failure



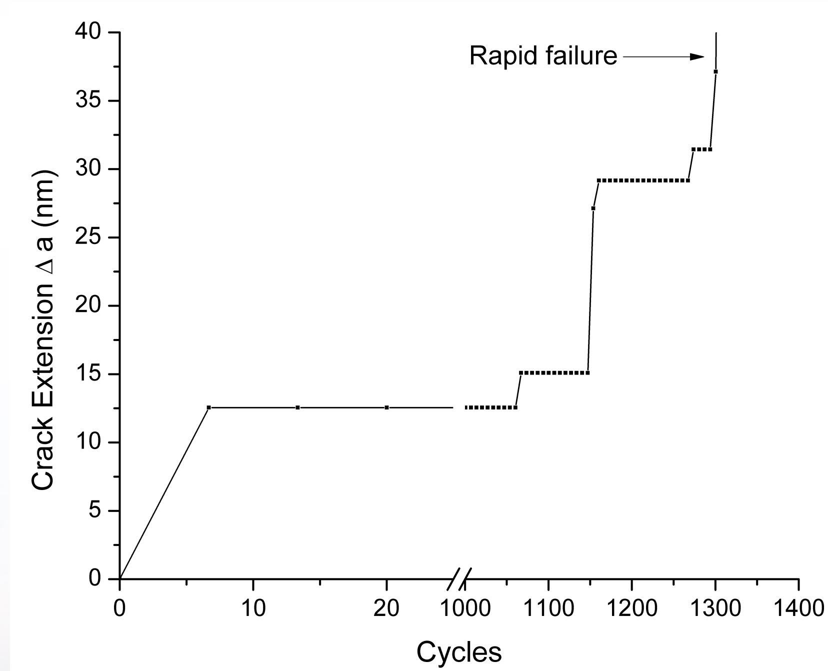

- Previously N = 150,000; Sequence shows failure over  additional ~1300 cycles
- Circle track motion of dislocation activity prior to crack prop.

# Quantification of Crack Extension in NC Pt

## Indenter load and displacement

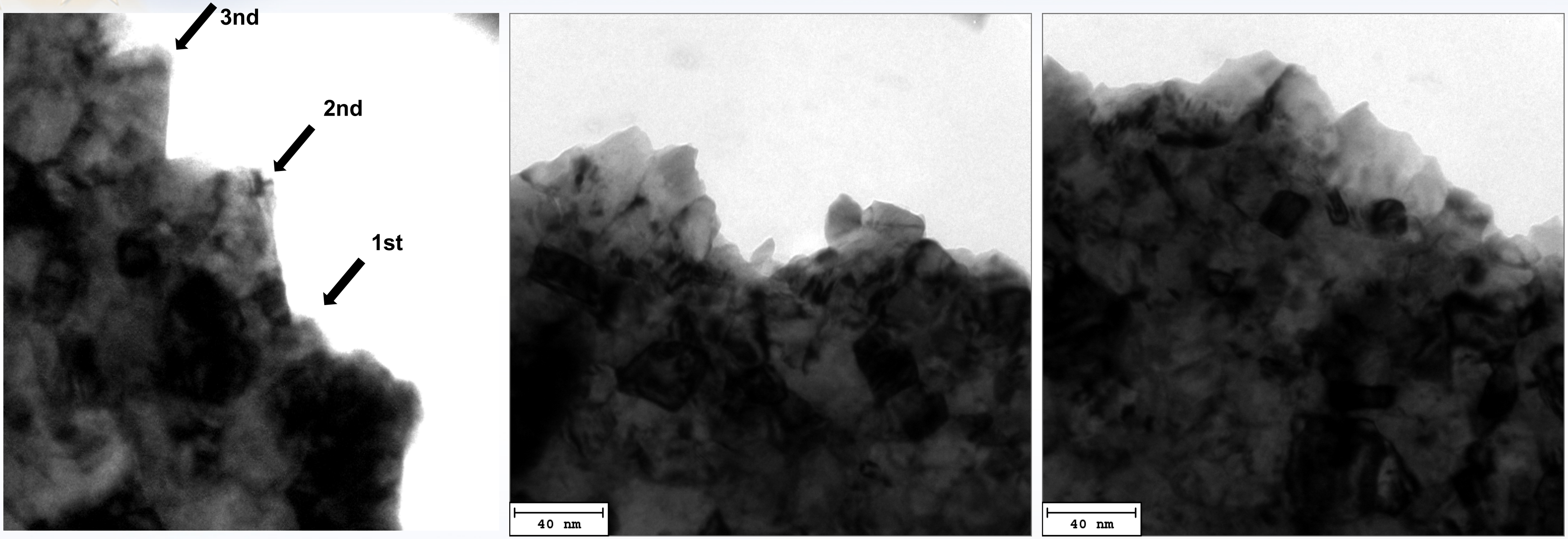



## Crack extension




- Initial crack tip forms during "first" cyclic frame (6.67 cycles per image frame; 15 fps collection)
- Crack blunts until approximately 1050 cycles and further extends until rapid failure
- Crack extension evolution with cycles provides additional detail on cyclic failure mechanisms of free standing thin films

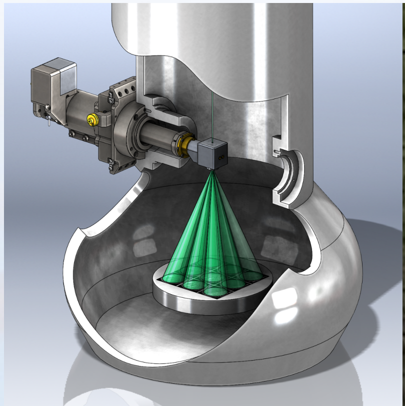
# Localized Plasticity during Cyclic Loading

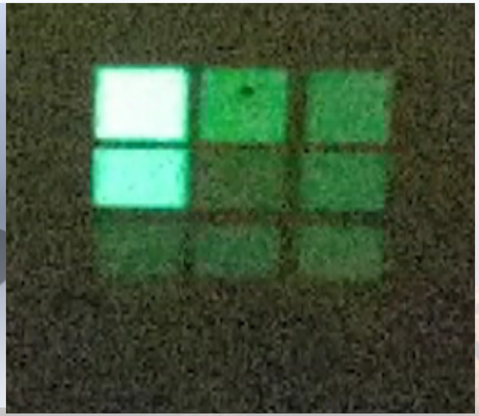



- Arrows indicate "saw tooth" localized plasticity observed in the in-situ crack blunting
- Grain thinning observed across the fracture surface
- Initial dislocation activity directly observed at N= 7, N= 554, and N= 1067 cyclic load frames correspond to 2nd "saw tooth"

# Future In-Situ Directions at $I^3$TEM Facility (PI Khalid Hattar):

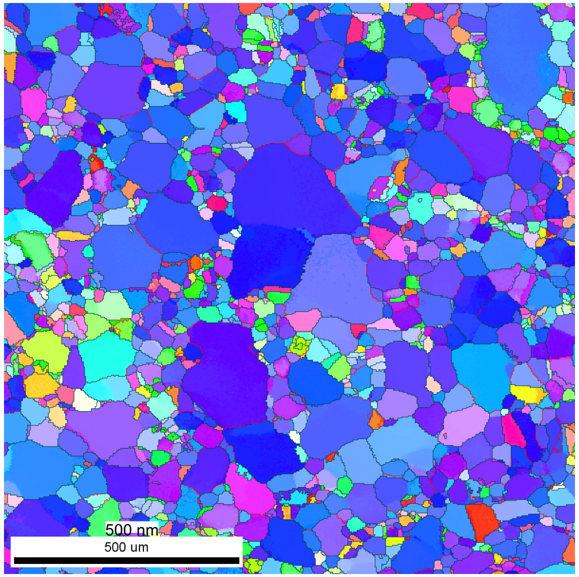
- High cycle fatigue – more detailed quantification of grain growth
- Creep, radiation-induced creep
- TEM notched three point bend
- Under development capabilities:
  - DTEM/Movie mode with crack propagation for improved temporal resolution under 200 Hz cyclic loading

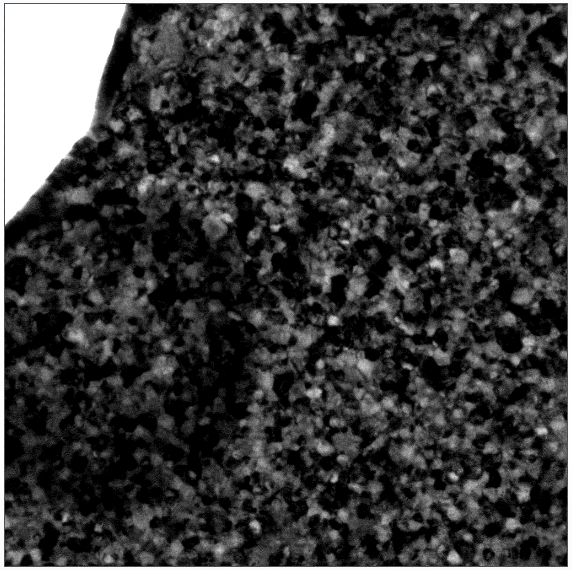


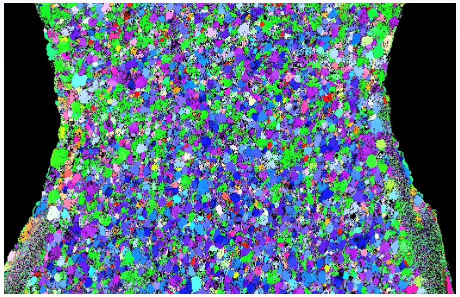


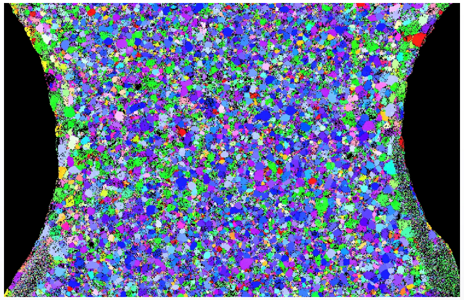
**Combining the precision of Hysitron's Pico-indenter with harsh environments capable in Sandia's In-situ Ion Irradiation TEM a wealth of previously impossible experiments are now feasible.**

# Summary







**Combining precession electron diffraction with quantitative mechanical testing provides new correlations between structure-property relationships with unprecedented orientation and mechanical property information.**

Work performed by C.M.B, D.C.B., and K.H. was fully supported by the Division of Materials Science and Engineering, Office of Basic Energy Sciences, U.S. Department of Energy. FIB sample preparation was performed, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science.

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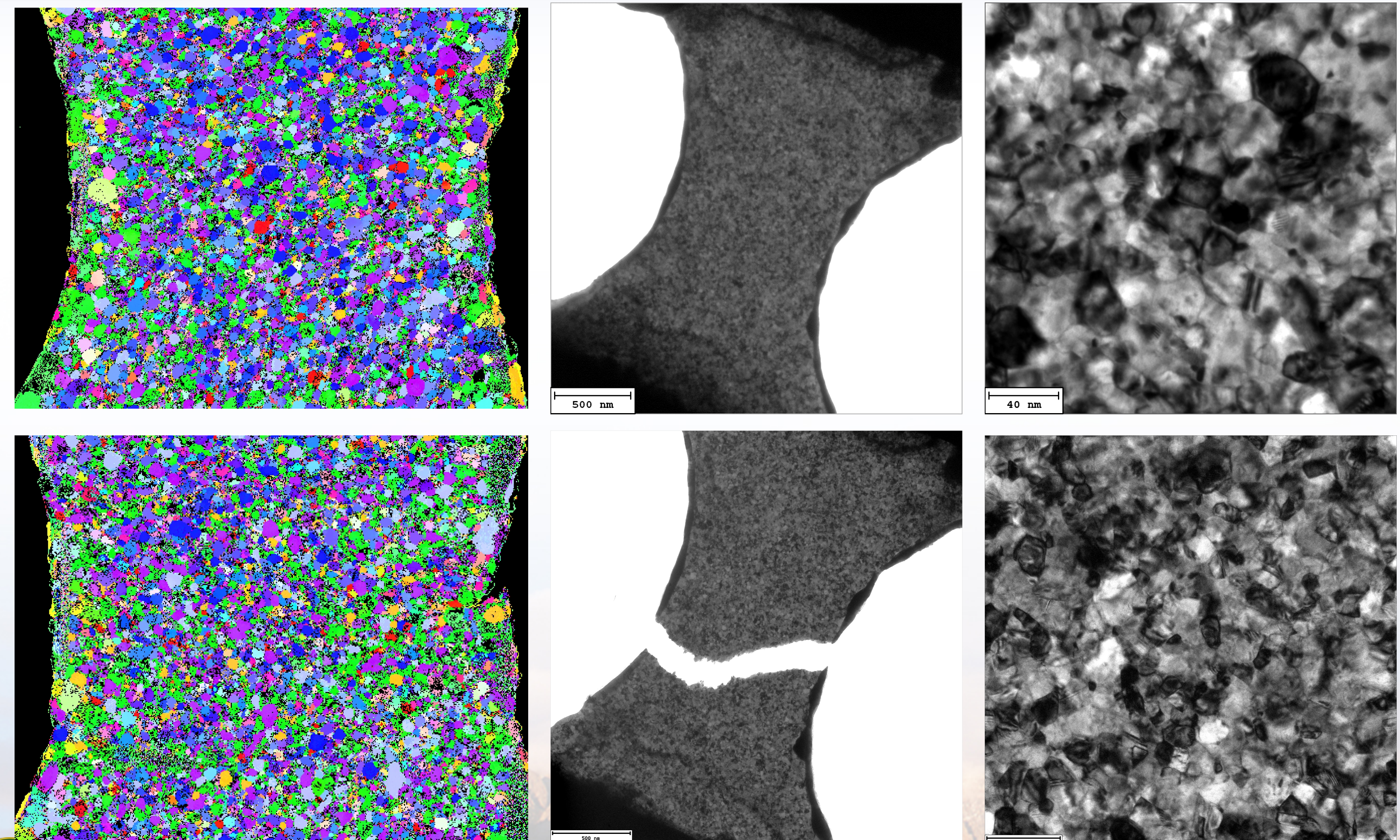
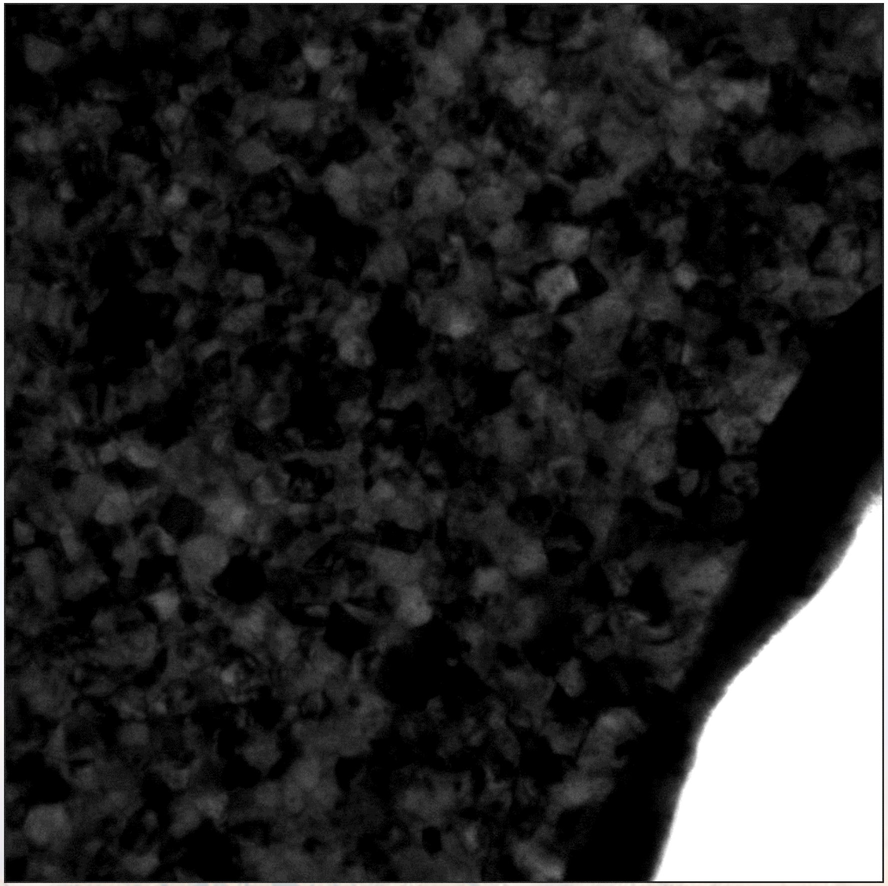
# Extra Slides Beyond This Point

# NC Pt Sample 4 Failure



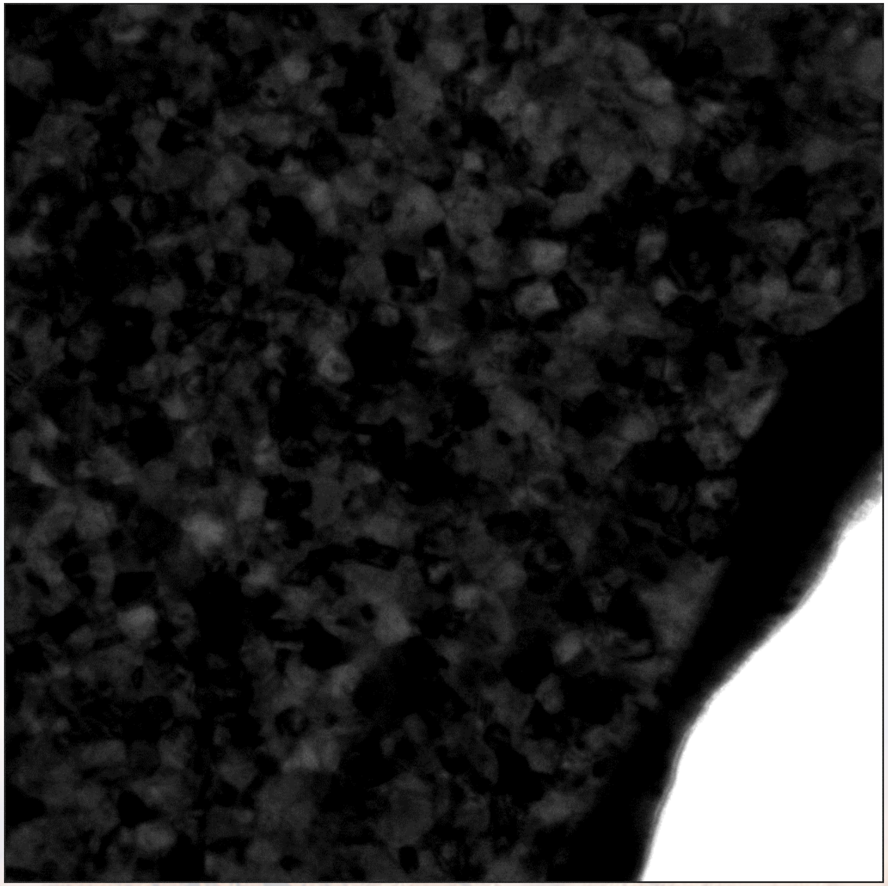
# NC Pt Sample 4 Failure

**Pt4(5): 110 uN mean load, 45 uN cyclic load, 200 Hz, 100 seconds of cyclic loading,2x speed**



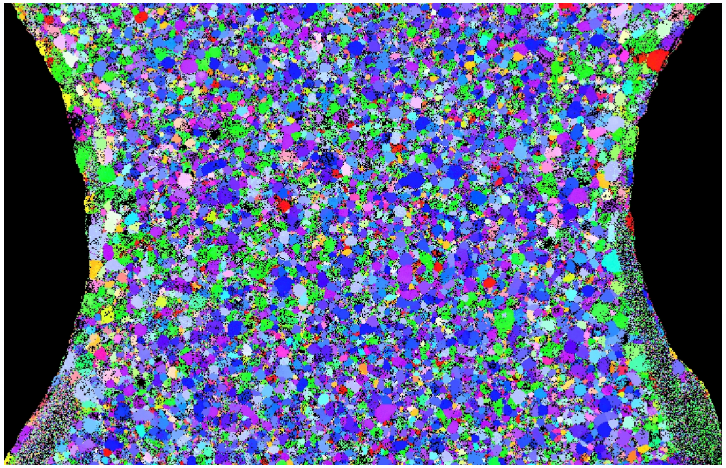
# NC Pt Sample 4 Failure

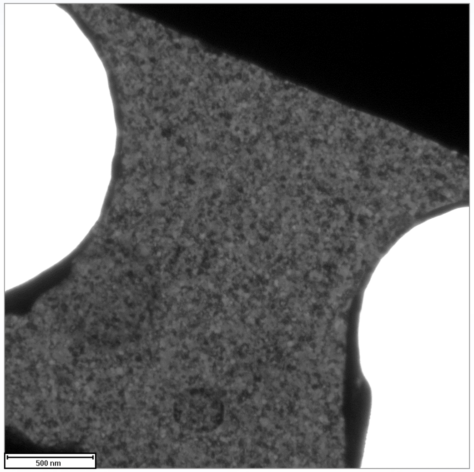
**Pt4(6): 110 uN mean load, 45 uN cyclic load, 200 Hz,  first cyclic loading failure 2x speed**

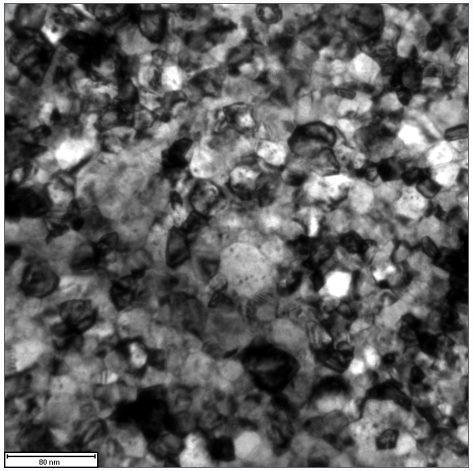


# NC Pt Sample 3 Failure

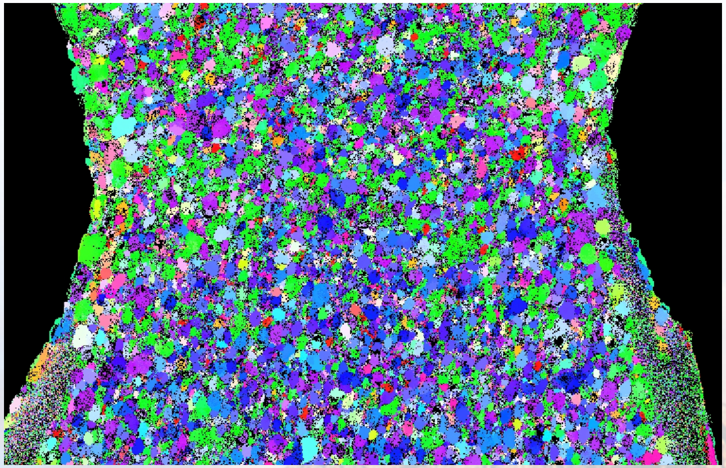
**Pre-test**

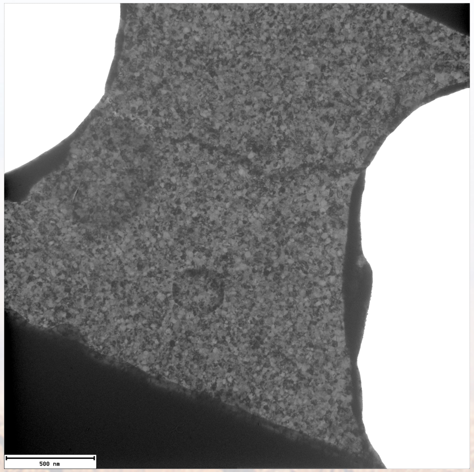


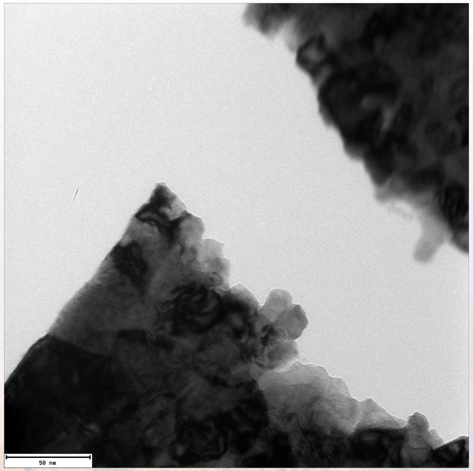




**Post-test: Mean Load 200 uN, Cyclic Load: 60 uN, Failure on first cycle**

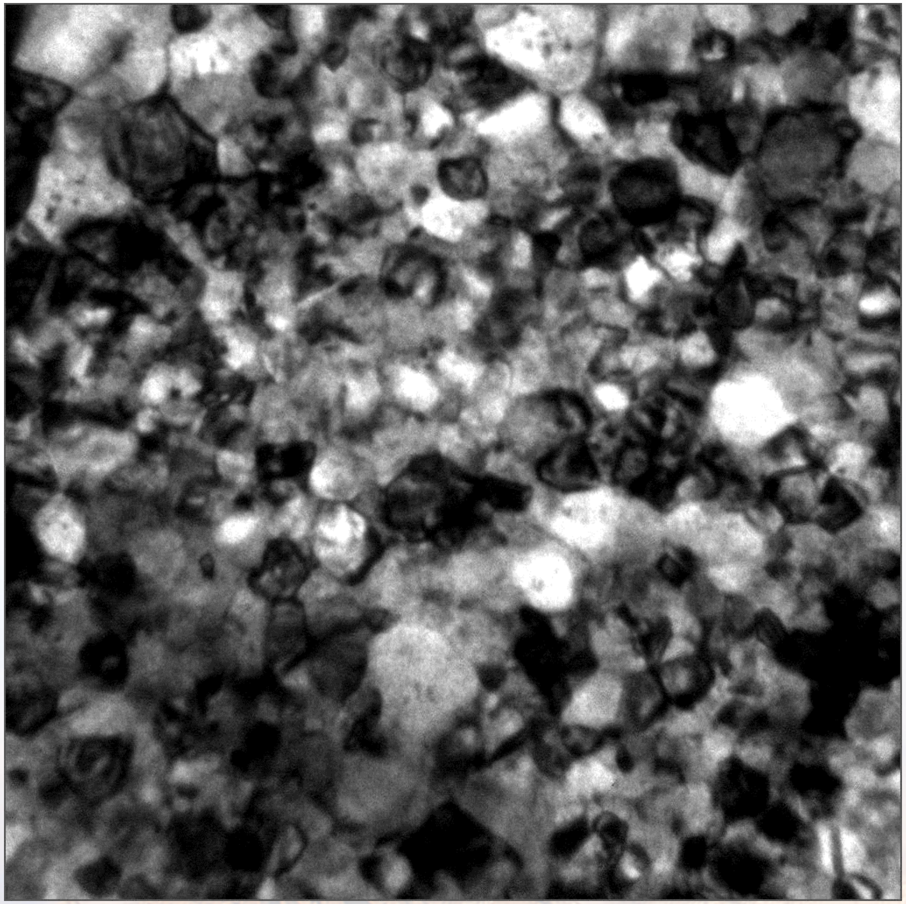






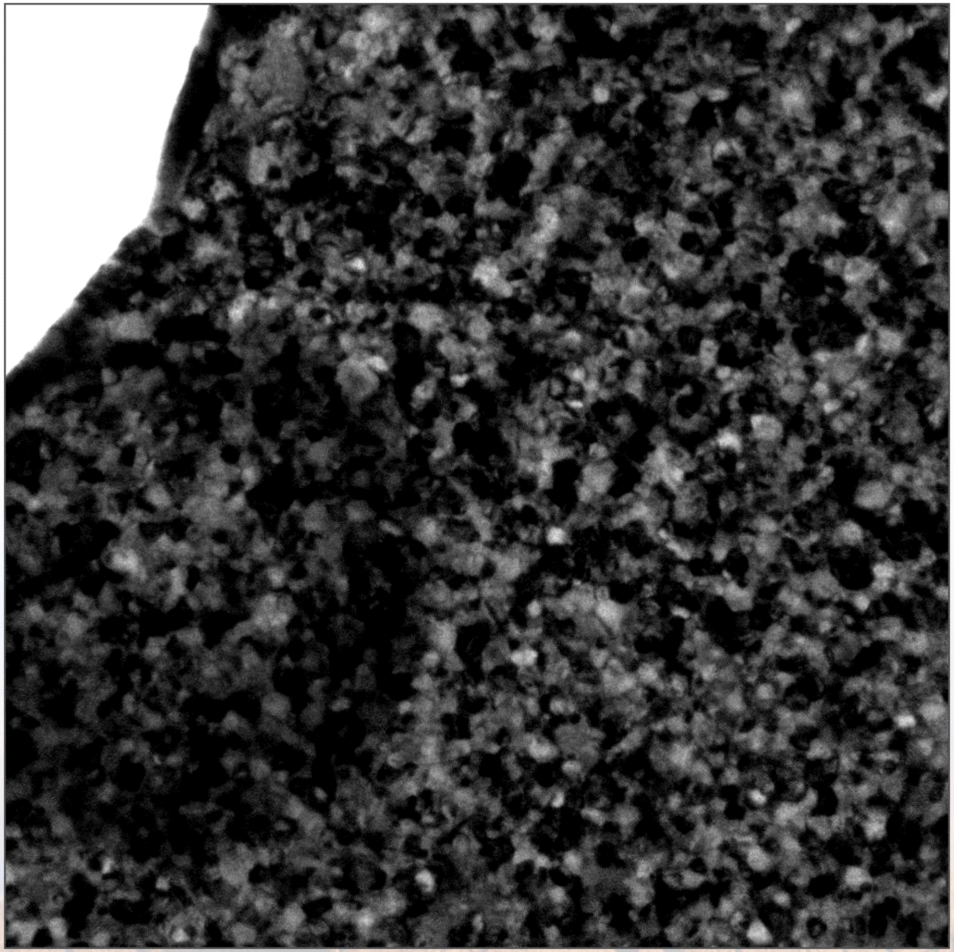
# Pt Sample 3 Observation of In-grain dislocation activity

**Pt (20): 150 uN mean load, 50 uN cyclic load, 100 Hz, 200 seconds of cyclic loading,2x speed**



# Pt Sample 3 Failure on First Cycle

**Pt (20): 200 uN mean load, 60 uN cyclic load, 100 Hz, 2x speed**



# Discussion

- **Localized plasticity observed in the nc Pt fatigue after crack initiation consistent with Gatan straining experiments**
- **Local deformation consistent with local in-grain plasticity confirmed from saw tooth appearance of fracture surface**
- **Current efforts of ACOM-TEM coupled with fatigue experiments limited by spatial resolution of ACOM on LaB6 TEM.  Grain size of Pt is 20 nm.**
  - Low reliability creates challenge for example quantifying grain size evolution after cyclic and fatigue loading
  - Post sample on unload can hit fracture surface and complicate crystallography reconstructions.
  - 2.5 nm step size