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Evidence that abnormal grain growth precedes crack initiation in nanocrystalline metals

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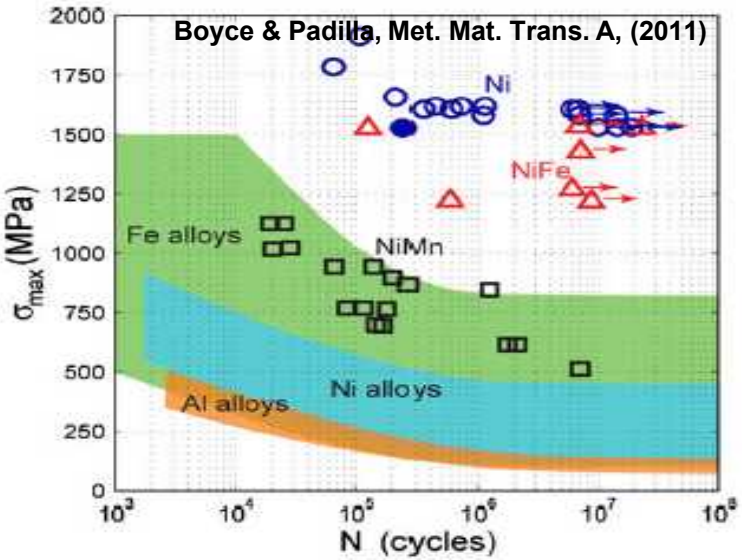
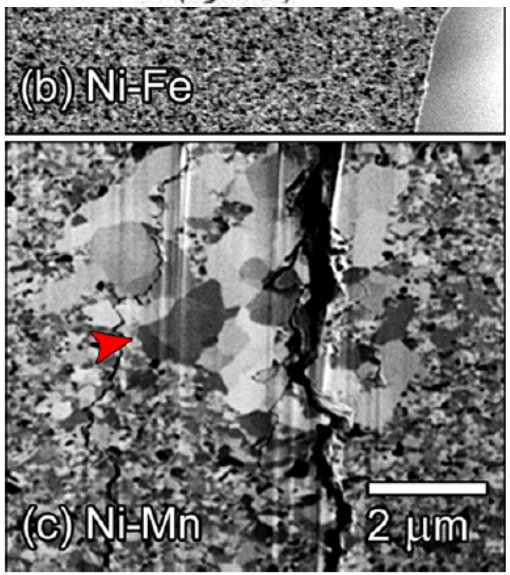
Nanocrystalline metals show unique mechanical behaviors, including a general propensity for mechanically-induced grain growth

- Typical properties of NC metals include:
  - Ultra-high strength & hardness
  - High wear-resistance
  - **Ultra-high fatigue strengths**
- When grains are at the nanoscale, unique mechanical properties and plastic responses arise, including **mechanically-induced grain growth**
- In fatigue, ***abnormal* grain growth** has been suspected to lead to **crack initiation**



- Post-fracture techniques leave us with a "chicken and egg" problem (fracture or AGG?)
- Additionally, are these abnormally large grains pre-existing (grown during deposition) or cyclically-induced?



Post-fracture analysis can only tell us so much – to understand the role of abnormal grain growth on the fatigue deformation and crack initiation, non-destructive *in-situ* techniques are required.

Synchrotron XRD can provide non-destructive means to detect abnormally large grains occupying < 0.0001% of the sampled volume

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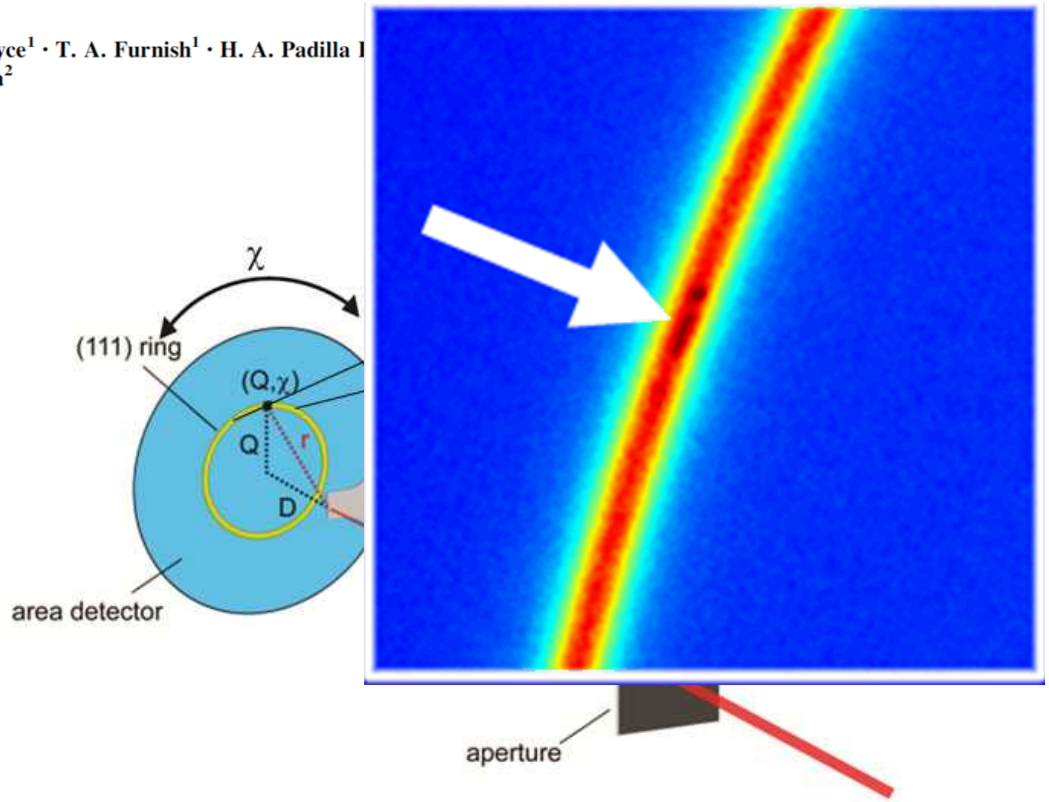
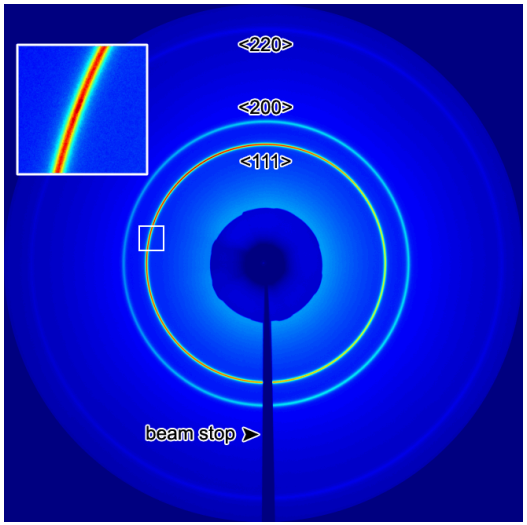
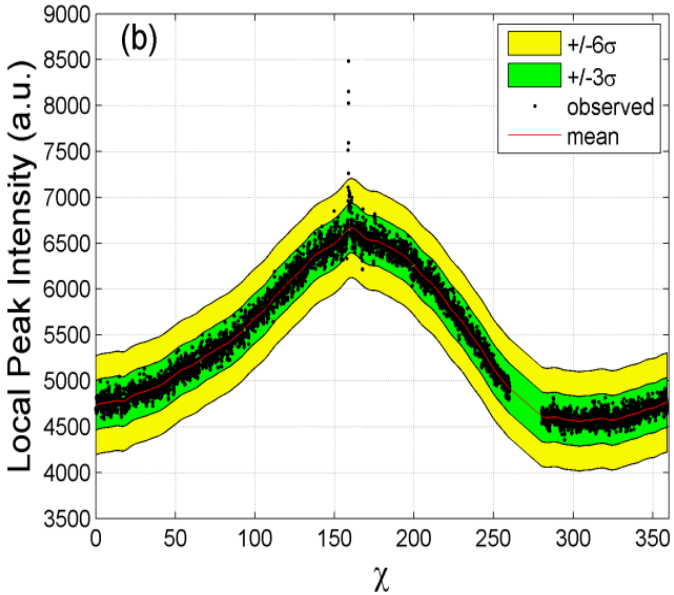
CrossMark

Detecting rare, abnormally large grains by x-ray diffraction

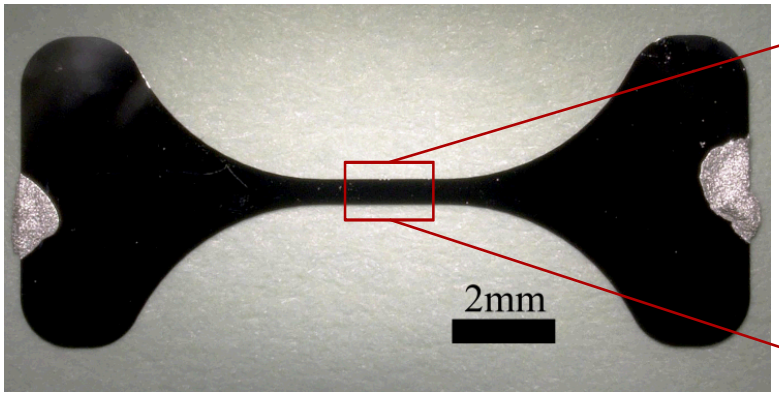
B. L. Boyce[1] · T. A. Furnish[1] · H. A. Padilla I
A. Mehta[2]



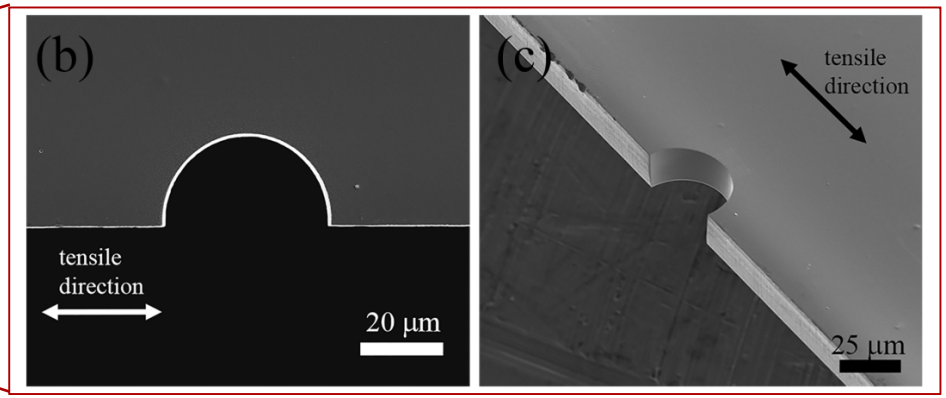




X-ray beam was centered on a FIB-milled semi-circular notch (used to prescribe crack initiation to a known location)

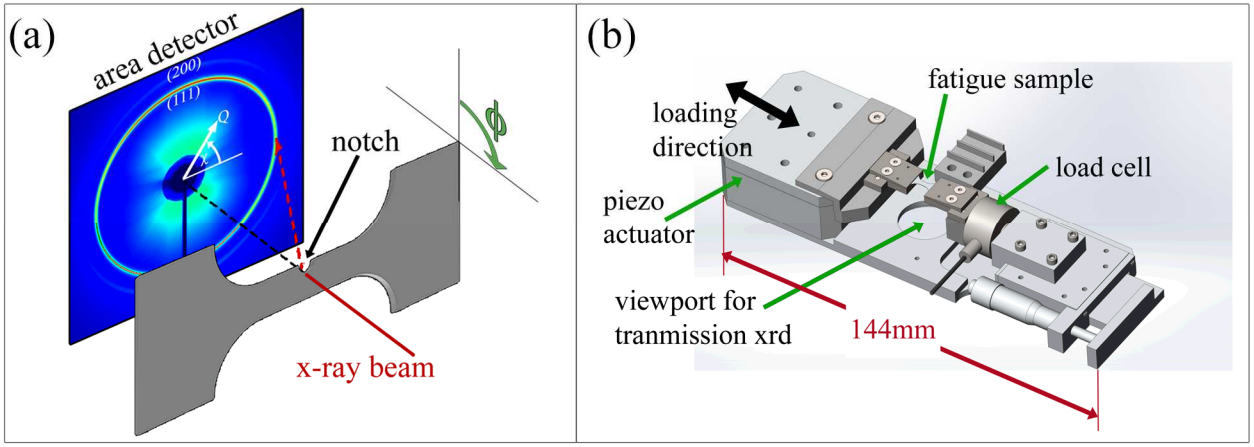


Notched electrodeposited NC Ni-Fe tensile specimens. Gage: 0.5 x 2.4 mm, 10-20 µm thick. Notch radius: 10 µm. Grain size: ~16 nm.



Schematic representation (a) and SEM micrographs (b,c) of the semi-circular notches FIB-milled at 30kV, 3nA. Elastic stress concentration factor: Kt=2.8.

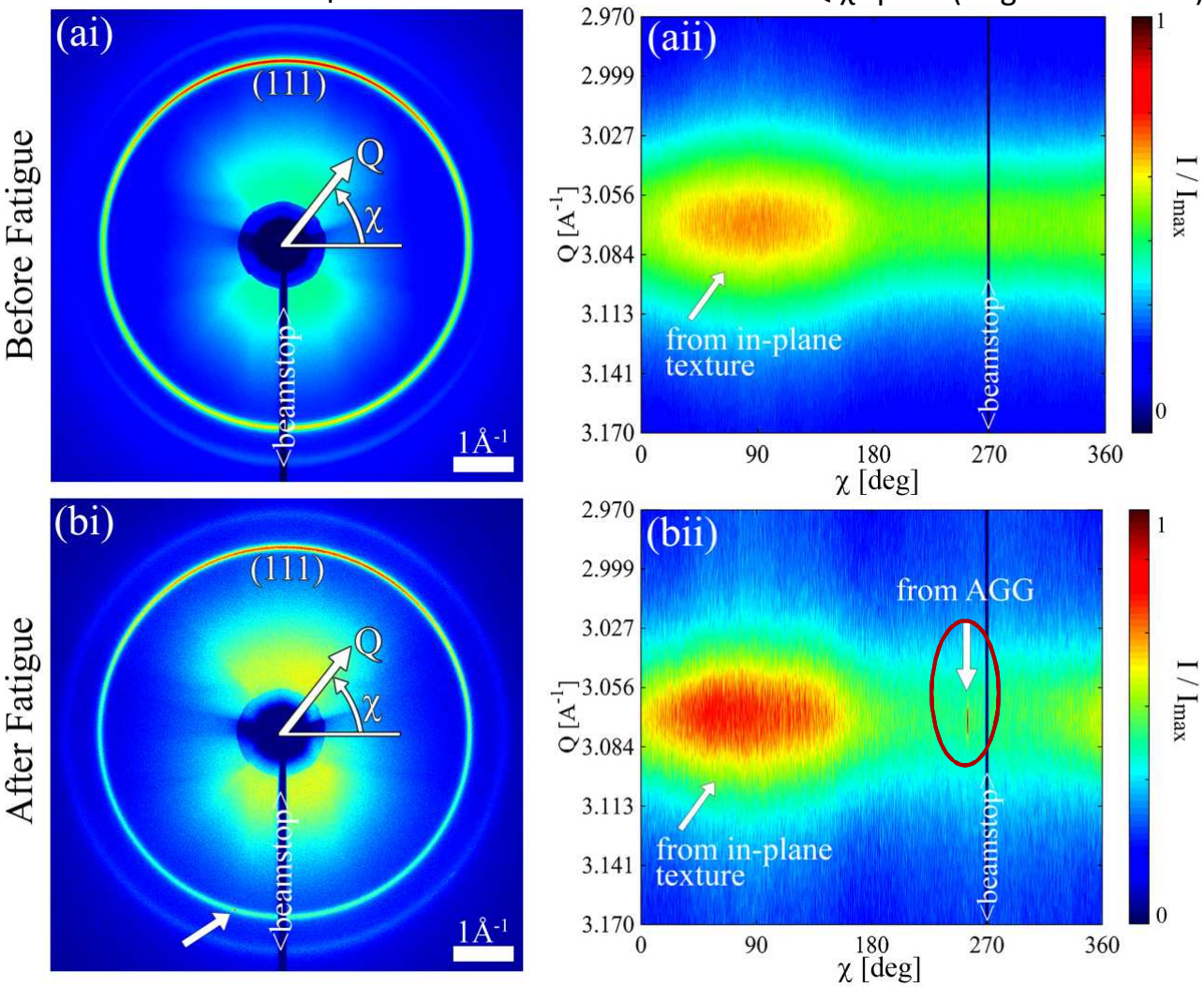
(a) Piezo-actuated in-situ micro-fatigue tester; (b) transmission xrd setup used in this study.

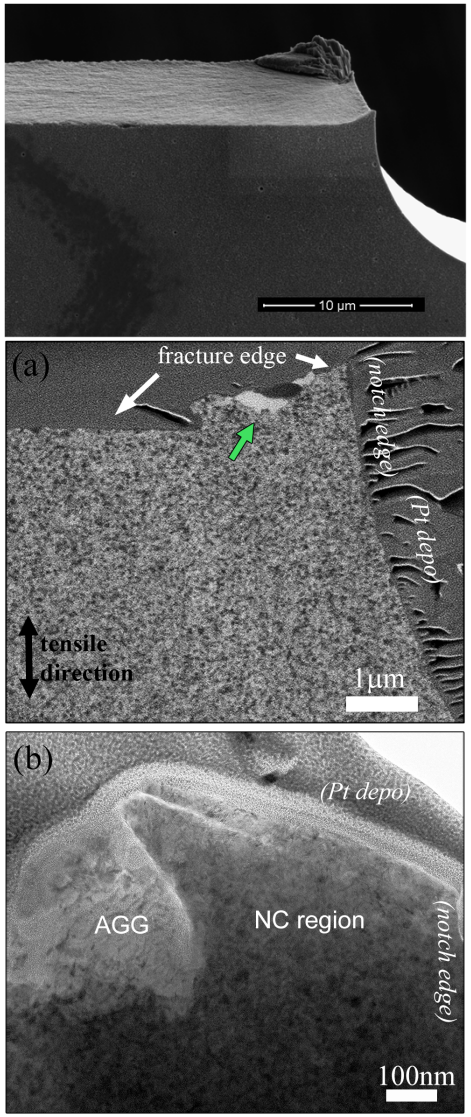


XRD revealed the *onset* of AGG (indicated by anomalous intensity "hotspots")

Raw diffraction patterns

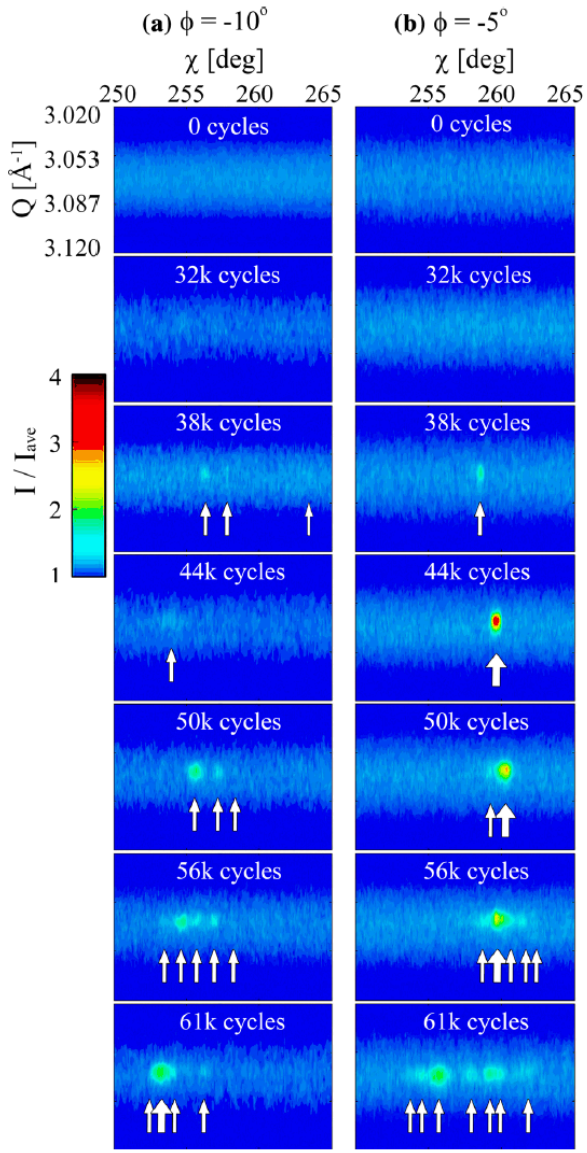
Transformed to Q-χ space (rings "unfolded")





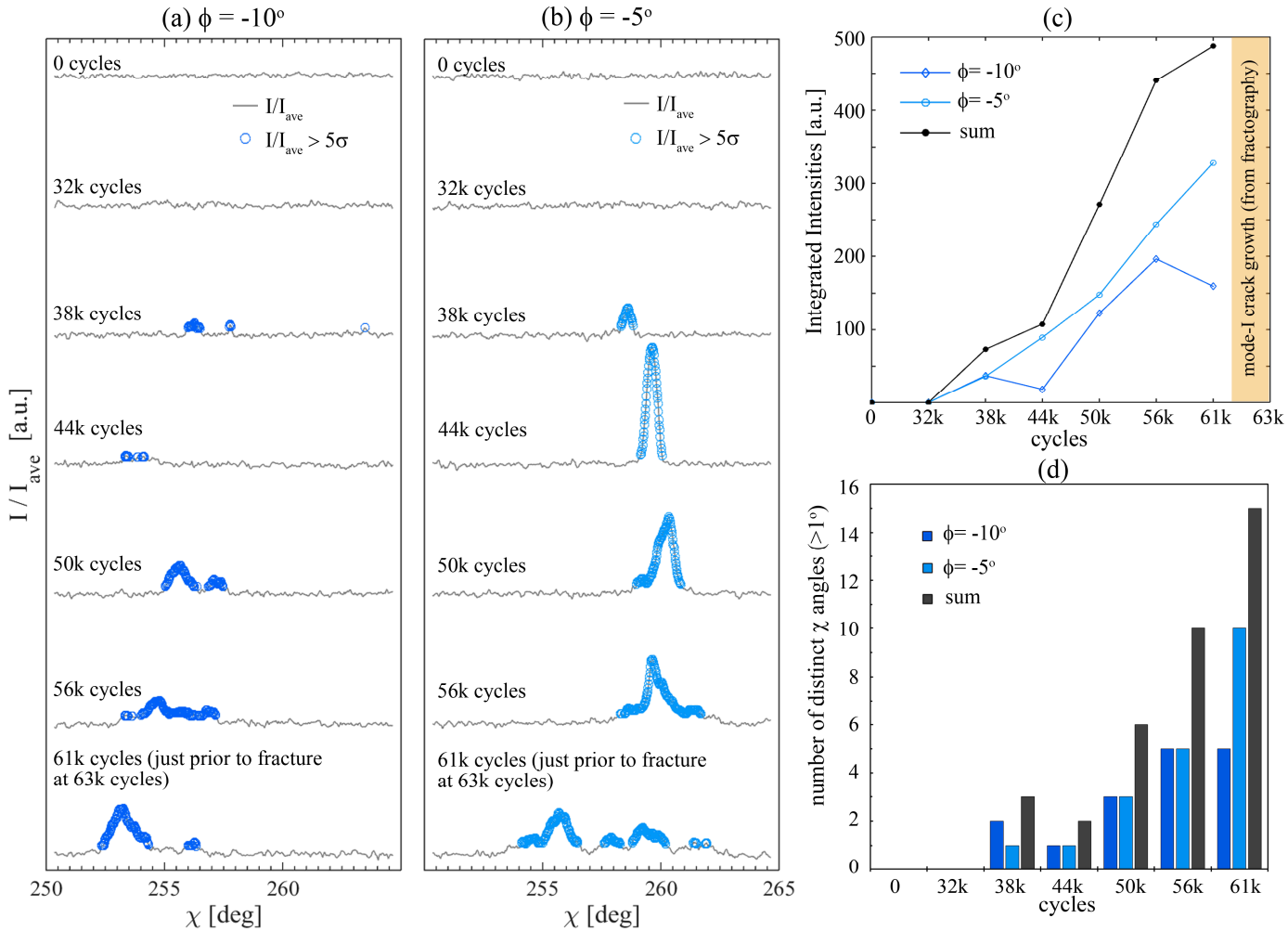
Fensin, S. et al., J. Mater. Sci. (2016 in prep.)

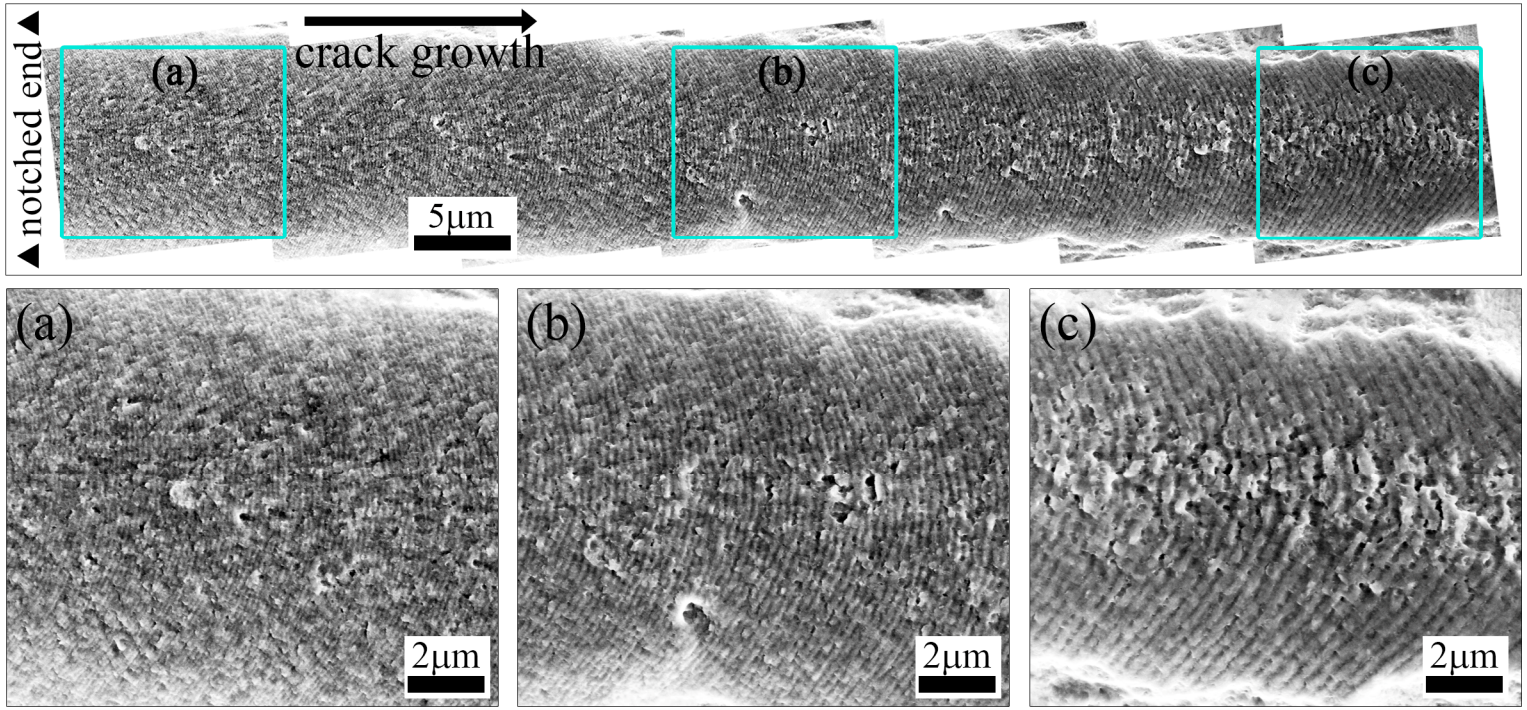
Looking at the in-situ XRD data reveals that AGG developed **well before final fatigue fracture!!**



- We first detect the anomalous intensity spike at 38k cycles (25,000 cycles <u>before final fracture</u>!)
- The "hotspot" is actually multiple intensity peaks with very similar orientation
  - Attributed to formation of subgrain boundaries
- The predominant peak intensity continuously increases, while many other smaller peaks form
  - dislocation accumulation → grain rotation
- If only taking this data, it appears that the grain gradually grows with subsequent cycling…
- But, if we do a side-by-side comparison of a different tilt angle, the intensity is very large at first, then gradually decreases
  - The abnormally large grains are not necessarily growing continuously, but they are rotating
- It appears that the abnormal grain growth occurs relatively quickly, reaches a critical size to support dislocation activity, then subsequently cyclically deforms until initation a crack

Growing intensities of the anomalous intensities suggests continuous growth of the ALGs, in addition to the collection of small angle misorientations



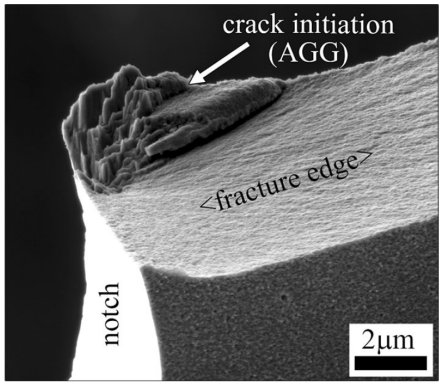


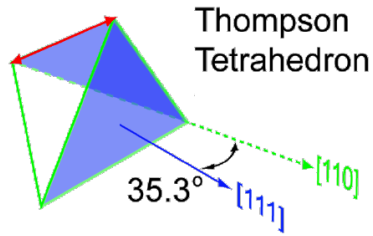
$$\frac{da}{dN} = C\Delta K^m$$

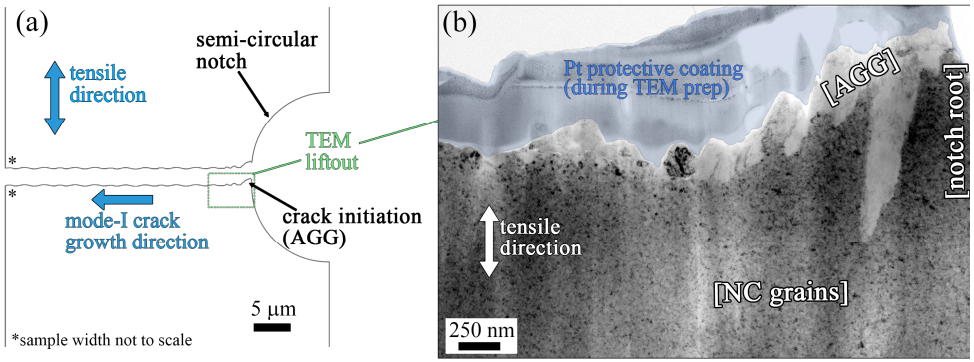
Paris Power Law exponent, m, was calculated to be 3.4, which is consistent with other reports on crack growth rates in NC metals.

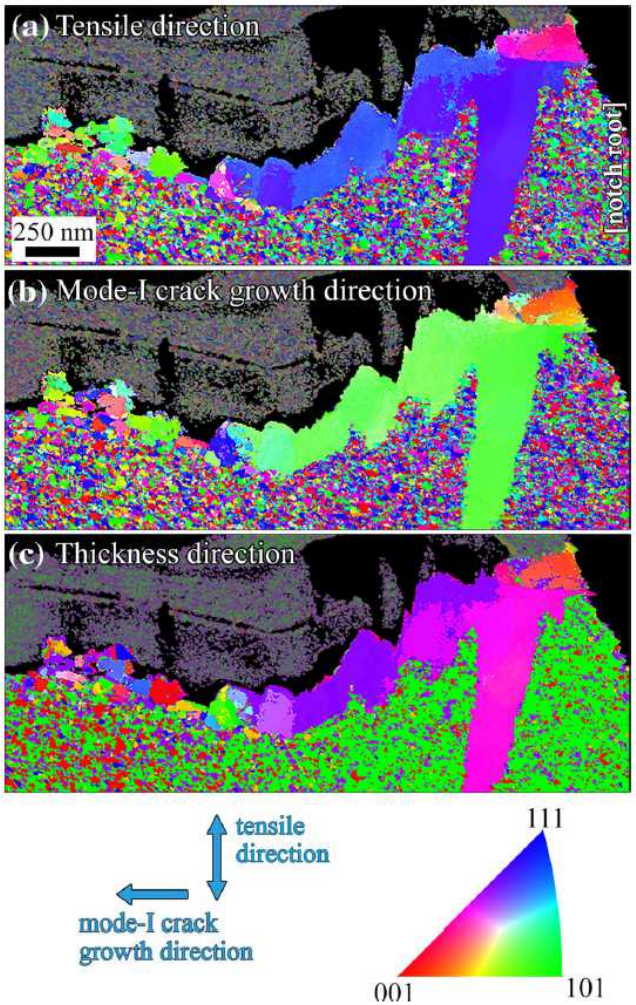
- **approximately 38,000 cycles (60% of total fatigue life) was spent fatiguing the NC structure without obvious AGG (at least non-detectable AGG)**
- **23,800 (38% of total fatigue life) was spent cyclically deforming and further evolving the AGG region**
- **1,200 cycles (2% of total fatigue life) was spent growing the Mode-I fatigue crack until final fracture.**

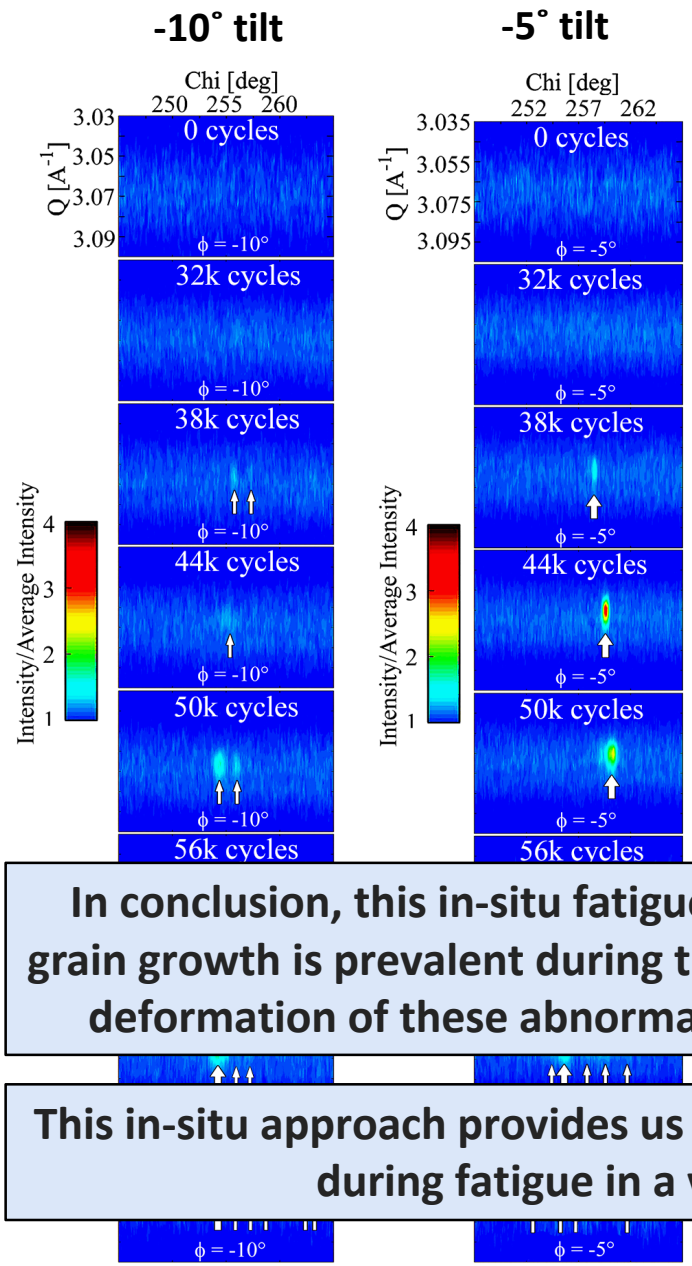
TEM and Precession Electron Diffraction (PED) confirmed sub-grain formation and dislocation-based slip protrusions











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- The predominant peak intensity continuously increases, while many other smaller peaks form
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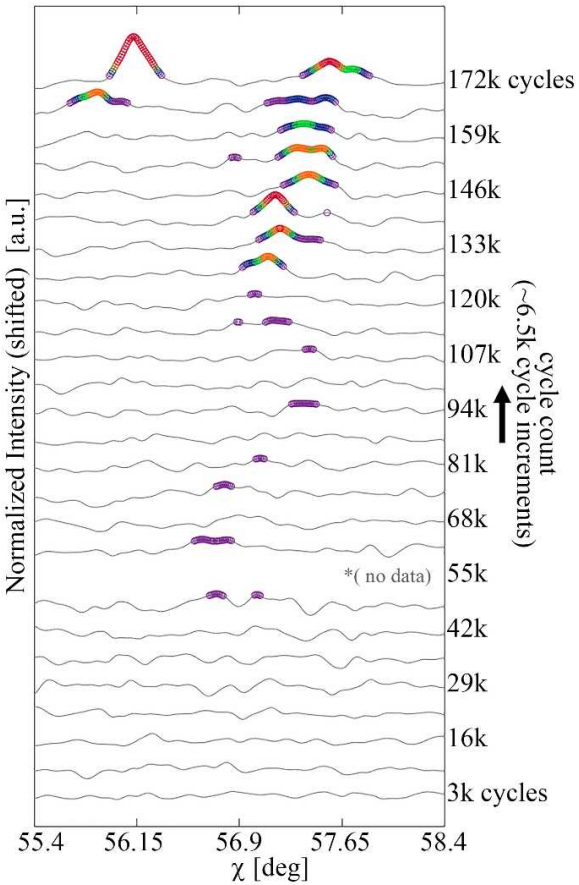
- If only taking this data, it appears that the grain gradually grows with subsequent cycling…
- But, if we do a side-by-side comparison of a different tilt angle, the intensity is very large at first, then gradually decreases

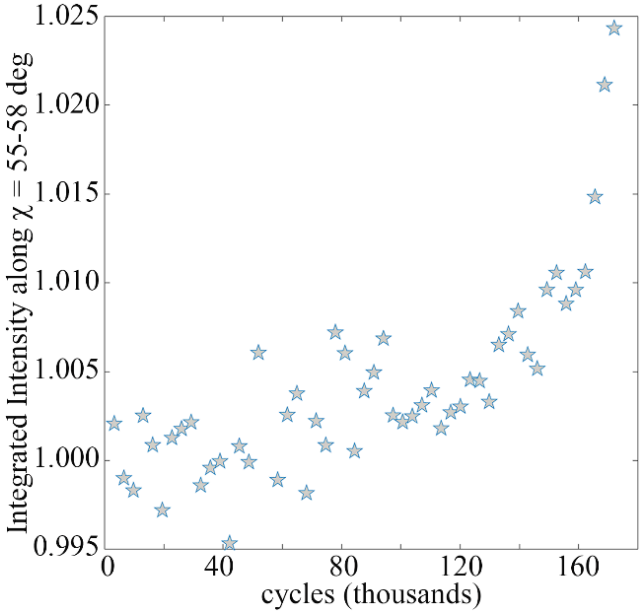
In conclusion, this in-situ fatigue study corroborates our initial hypothesis that abnormal grain growth is prevalent during the high cycle fatigue in NC metals, and that the subsequent deformation of these abnormally large grains is likely the precursor for crack initiation.

This in-situ approach provides us with a valuable tool to continue to explore the role of AGG during fatigue in a variety of NC systems and testing conditions.

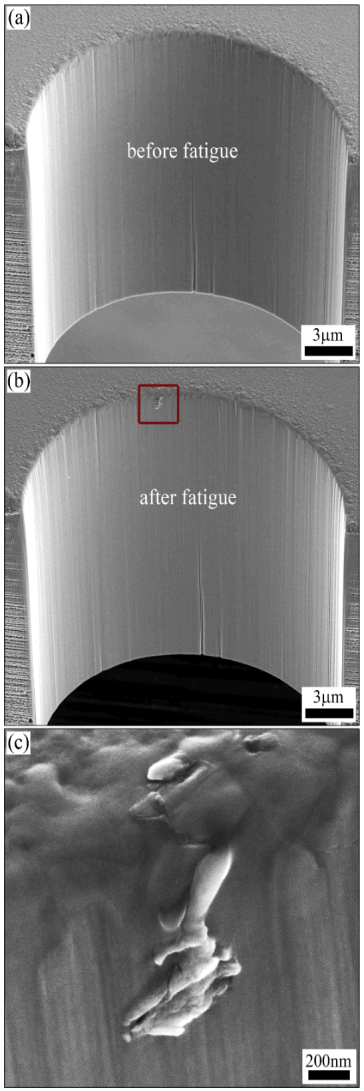
Taking this a step further…

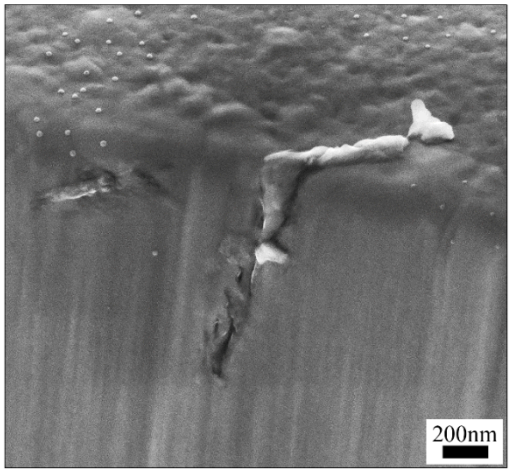


analyzed in near real-time during cyclic loading, allowing us to stop the fatigue process upon detection of AGG.



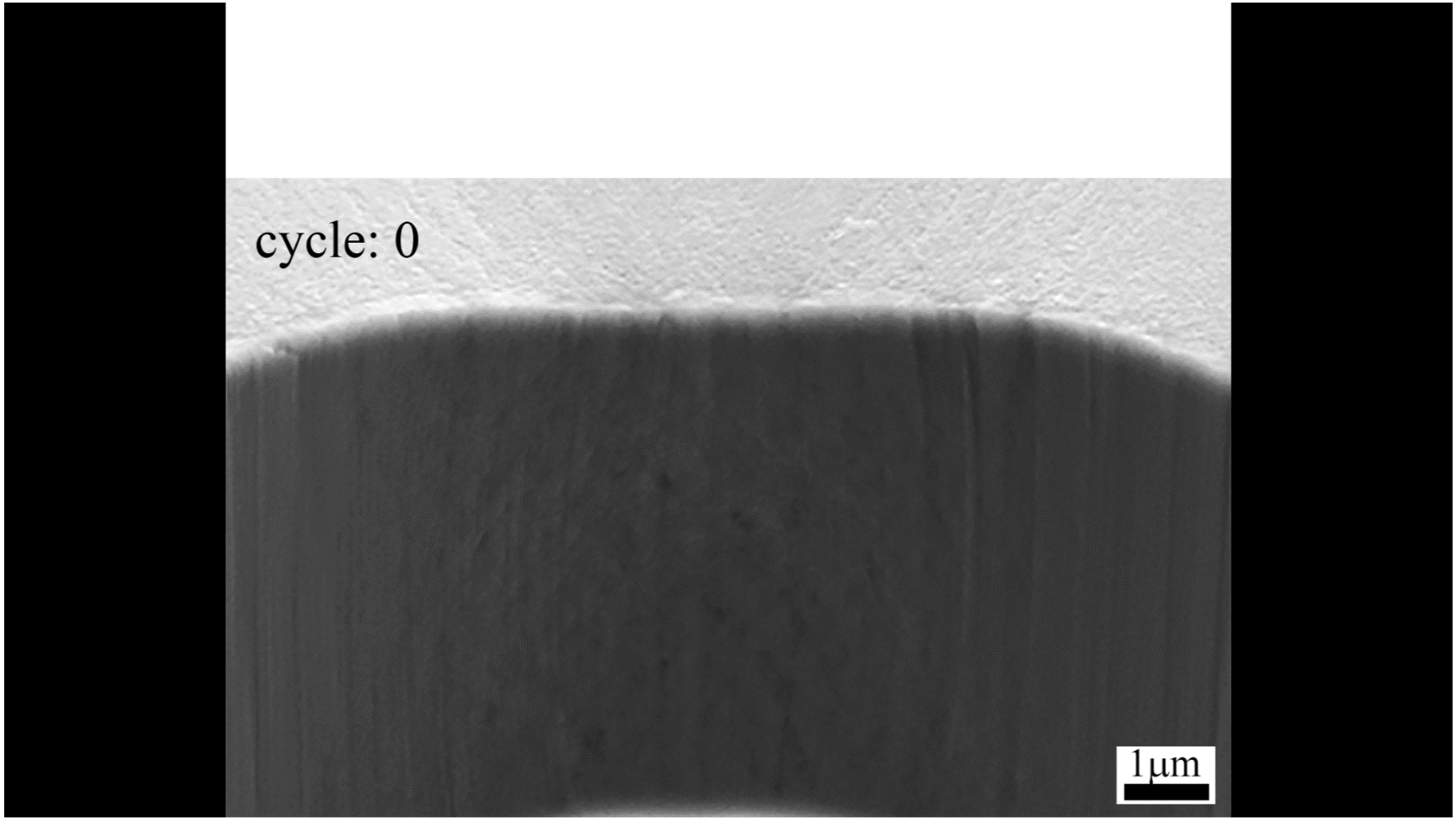
Despite XRD evidence of AGG during fatigue, a fatigue crack is notably absent! Additionally, slip protrusions were found to emanate from the ALGs.



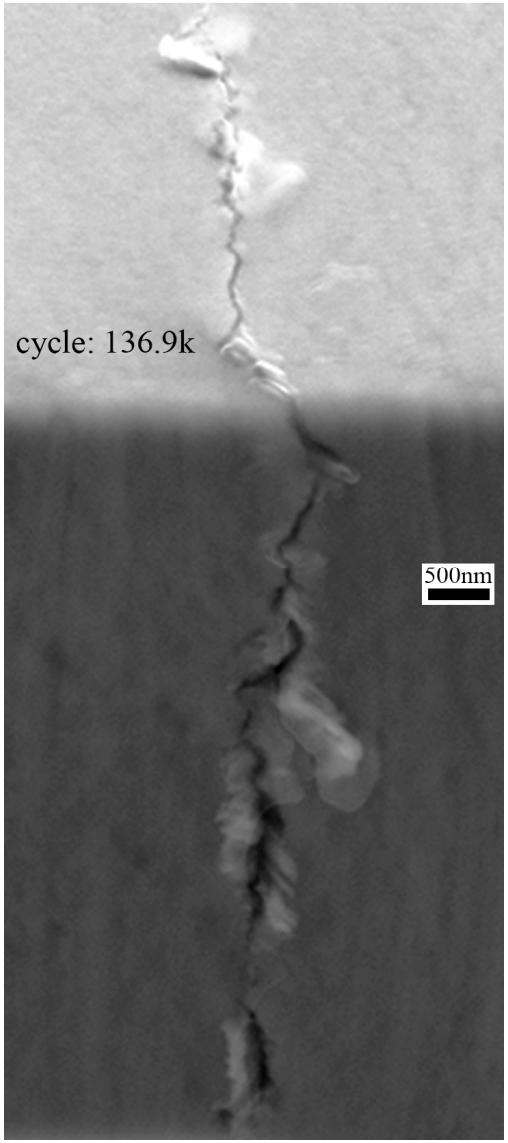


Fatigue crack embryo in a second fatigued NC specimen showing first stages of crack initiation following AGG and slip band formation at the sample surface.

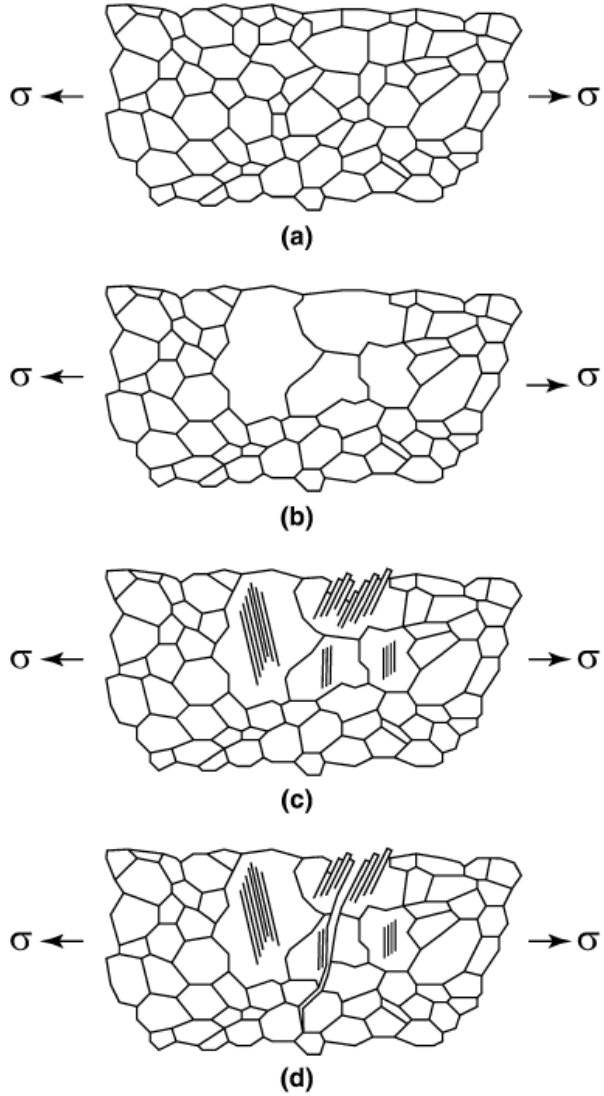
Can we repeat this in-situ in the SEM??



< Movie of in-situ fatigue >


cycle: 136.9k
500nm

Proposed mechanisms for crack initiation in NC metals



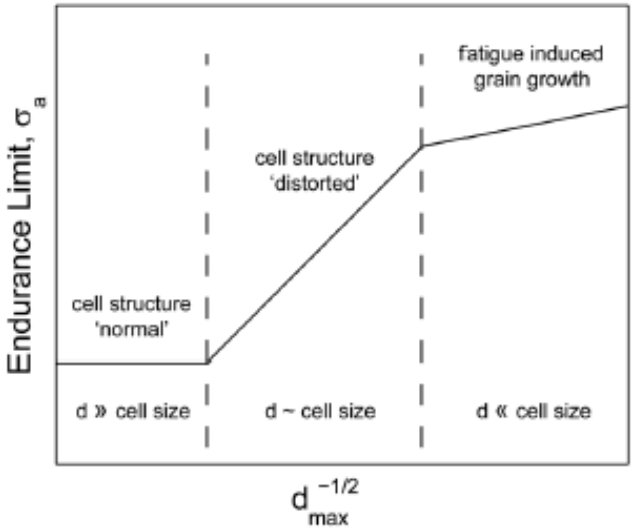


Figure 14. Schematic showing different scaling regimes for fatigue behavior with respect to the grain size.

Acknowledgments

- This work was funded by the U.S. DOE, Office of Basic Energy Sciences, Division of Materials Science and Engineering
- A portion of the microscopy and FIB work was performed at the Center for Integrated Nanotechnologies (CINT)
- Synchrotron work was performed at beamline 11-3 at Stanford Synchrotron Radiation Lightsource (SSRL)

Extra Slides

Conclusions:

- In-situ xrd fatigue corroborates our initial hypothesis that abnormal grain growth is prevalent during high cycle fatigue, and that the subsequent deformation of these abnormally large grains is likely the precursor for crack initiation.
- Despite XRD evidence of AGG during fatigue, a fatigue crack is notably absent. Finally, the chicken-or-egg question is answered: indeed, AGG preceded crack initiation
- The current data shows that AGG and subsequent deformation of the ALGs precede fatigue crack initiation in these metals!

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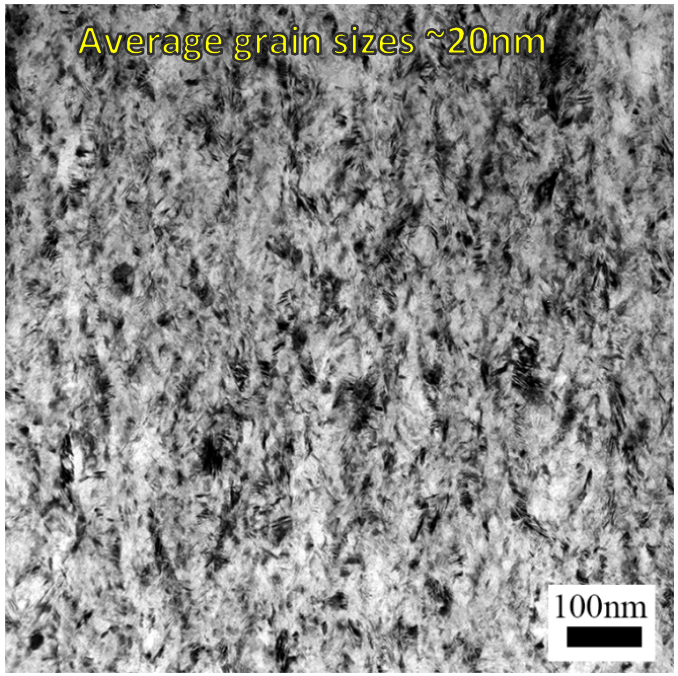
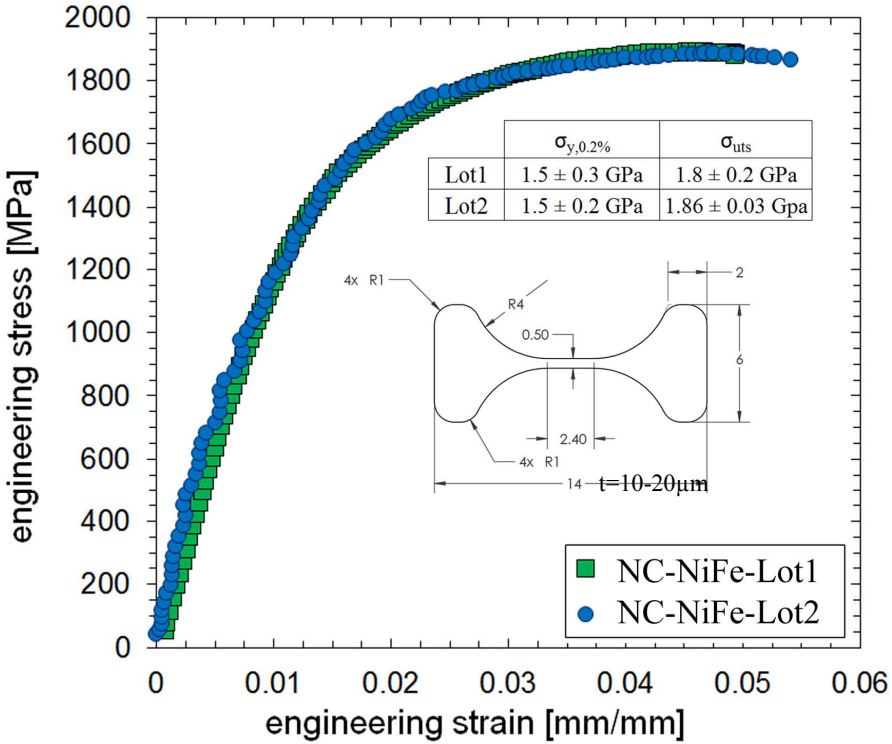
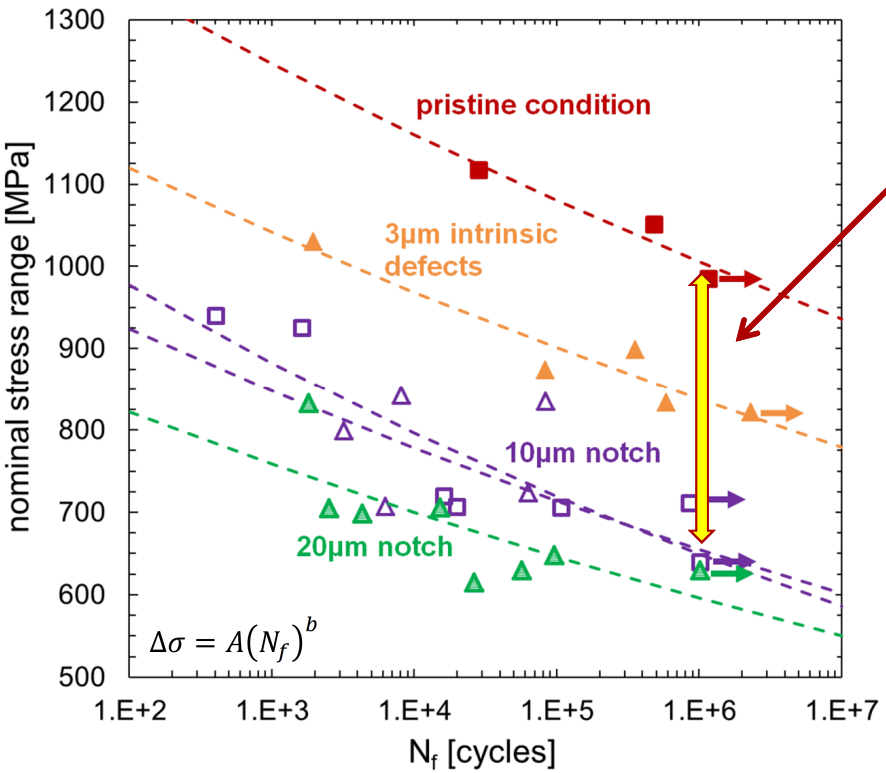




	$\sigma_{y,0.2\%}$	σ_{uts}
Lot1	1.5 ± 0.3 GPa	1.8 ± 0.2 GPa
Lot2	1.5 ± 0.2 GPa	1.86 ± 0.03 Gpa

- …evidence "suggest that the NC matrix undergoes AGG during cyclic loading, allowing dislocation activity to persist over length scales necessary to initiate a fatigue crack by traditional fatigue mechanisms." [Boyce and Padilla] also, "grain growth process that appears to be a necessary precursor to crack initiation". "insufficient space for the collective dislocation interactive mechanisms found in CG metals such as pileups and subgrain formation." "fatigue mechanisms may be influenced more by the evolved grain structure than by the initial structure". Ni-Fe shows the <112> pole aligned with the stress axis, which is the well-known orientation that any fcc single crystal rotates toward during uniaxial deformation to take full advantage of available slip systems. In NC metals, neighboring grains under an external stress can rotate to a common orientation, eliminating the HAGB allowing for collective dislocation motion
- *Stress concentrations are relevant for NC metals!* Most likely dependent on notch-to-grain size ratios…
- Abnormal grain growth (AGG) is a predominant precursor for crack initiation, the onset of which is likely dependent on the stress concentrations
- Understanding the AGG/crack initiation process will be paramount to devising means of improving the notch sensitivity in NC metals

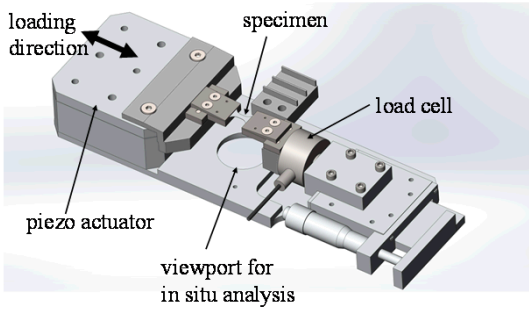
Notched fatigue tests revealed dramatic fatigue stress concentrations and high notch sensitivity!



$$K_f = \frac{\Delta\sigma_{unnotched,lim}}{\Delta\sigma_{notched,lim}}$$

$$q = \frac{(K_f - 1)}{(K_t - 1)}$$

q = 0 (totally notch insensitive)
q = 1 (fully notch sensitive, K_f=K_t)

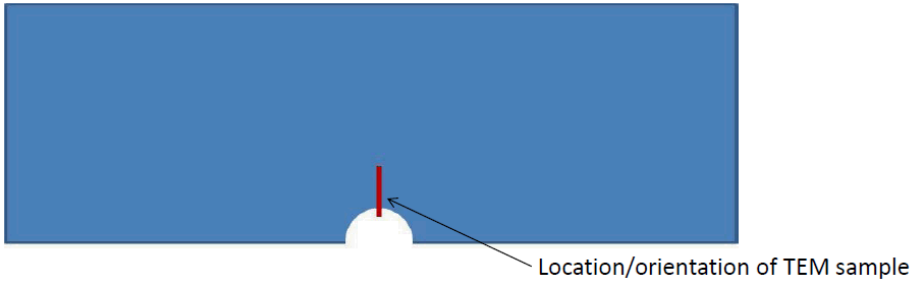


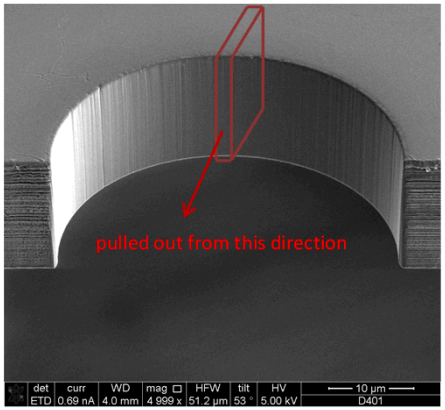
- Custom built piezo actuated *in situ* fatigue tester for thin foil testing
- Constant-load, 4-6 Hz (sine) used in this study

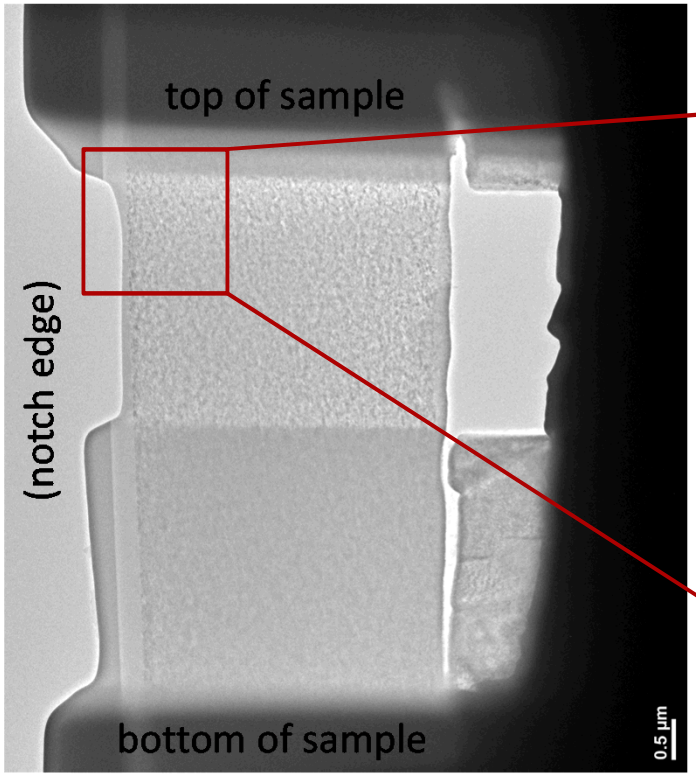
- Fatigue stress concentrations:
  - 1.2 (3μm intrinsic defects)
  - 1.5 (10μm notch)
  - 1.7 (20μm notch)
- Notch sensitivity factors, q:
  - 0.28 (10μm notch)
  - 0.37 (20μm notch)

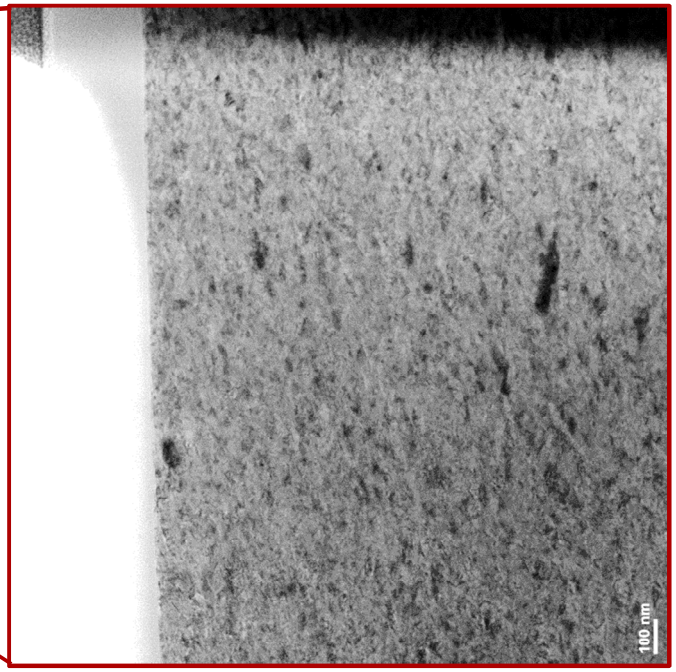
*Furnish, Sharon, Arrington, Pillars, Clark, and Boyce *submitted*

No obvious FIB-induced abnormal grain growth under notches



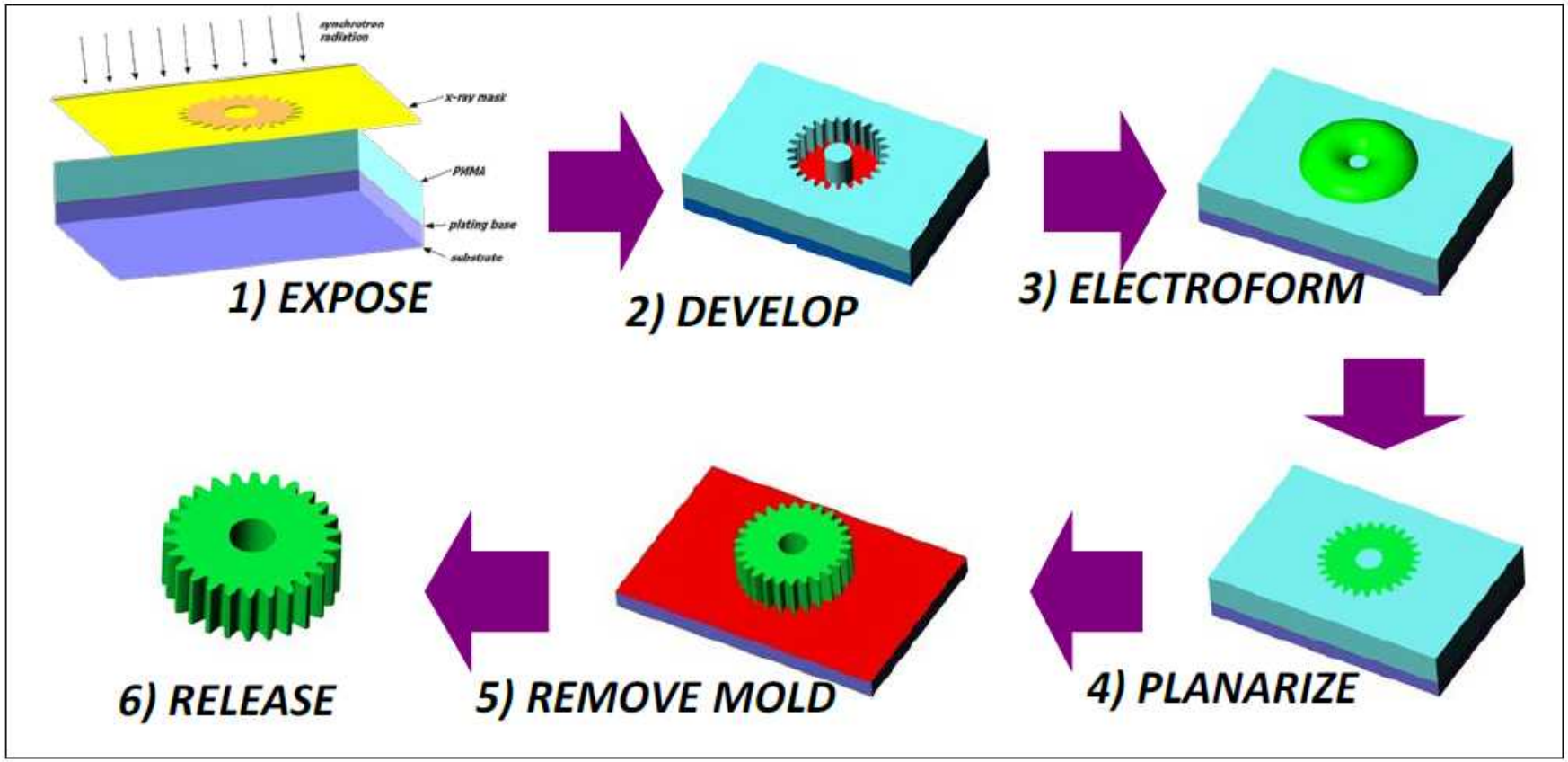






Sample Preparation

Electrodeposited nanocrystalline metal is lithographically patterned for fatigue testing



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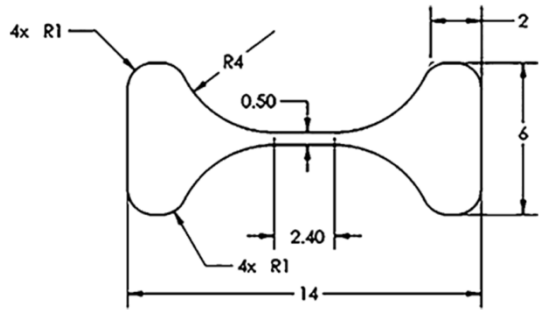
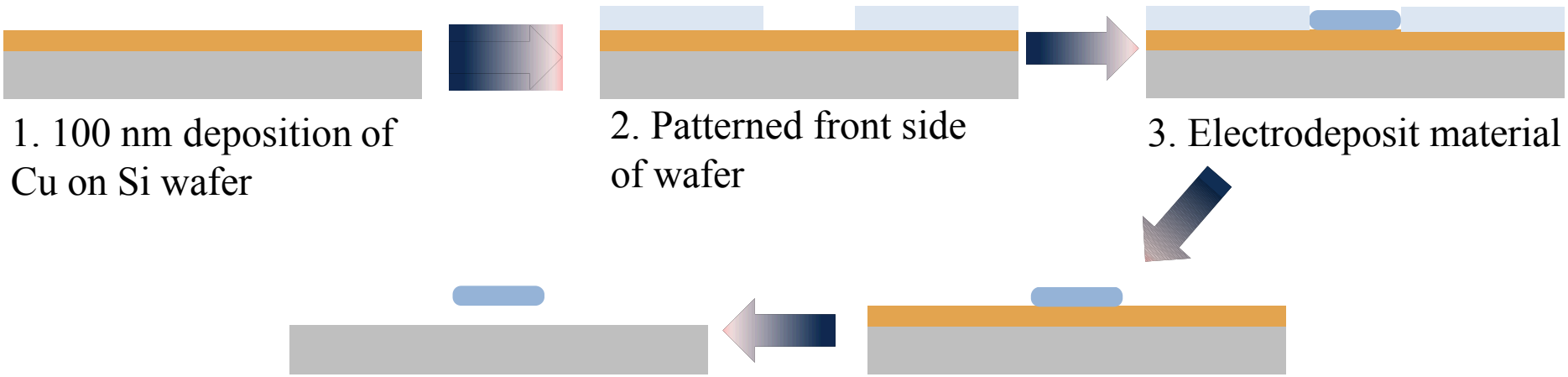
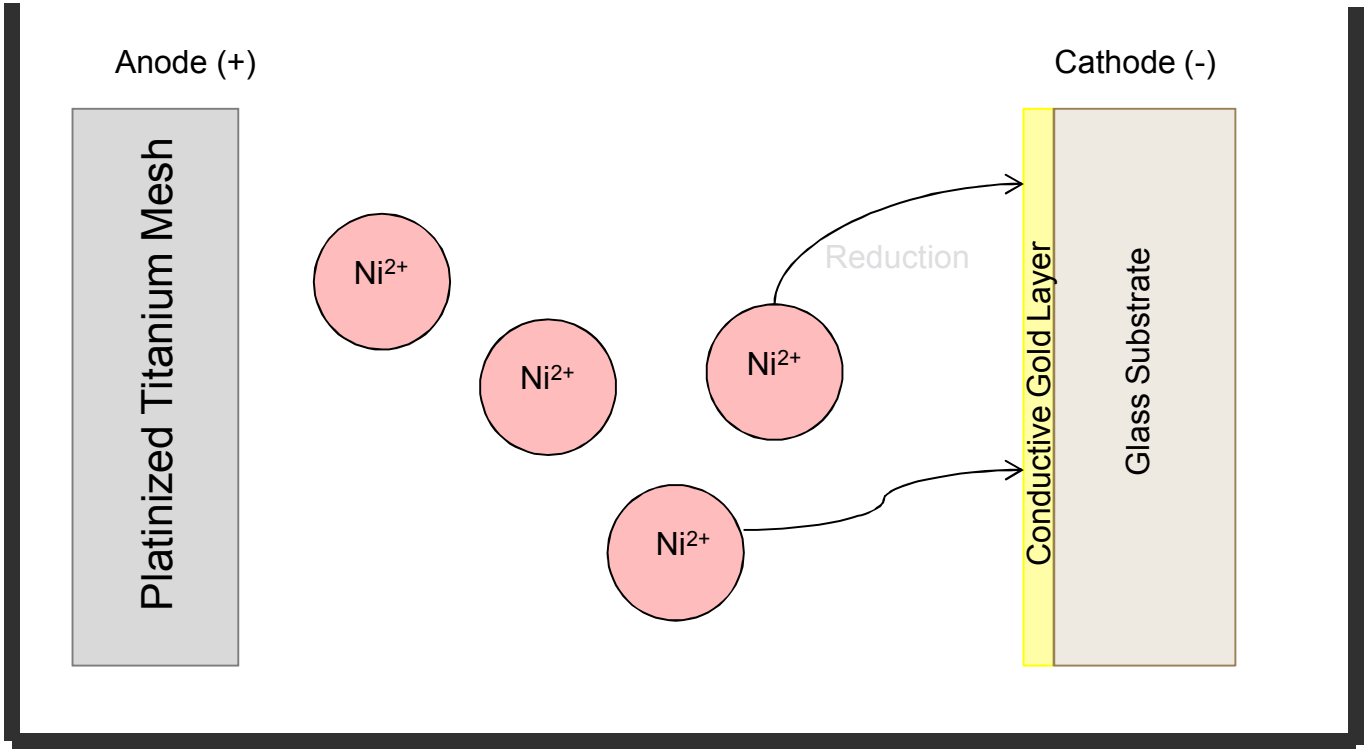


Nickel/iron chemistry make-up	
$NiSO_4$ Nickel Sulfate	112 g/L
$FeSO_4$ Iron sulfate	5 g/L
$Na_3C_6H_5O_7$ Sodium Citrate	75 g/L
KSO_4 Potassium Sulfate	1.5 g/L
$C_7H_5NO_3S$ Saccharin	1 g/L




Anode (+)
Cathode (-)
Platinized Titanium Mesh
Ni2+
Ni2+
Ni2+
Ni2+
Reduction
Conductive Gold Layer
Glass Substrate

efficiency tradeoff…

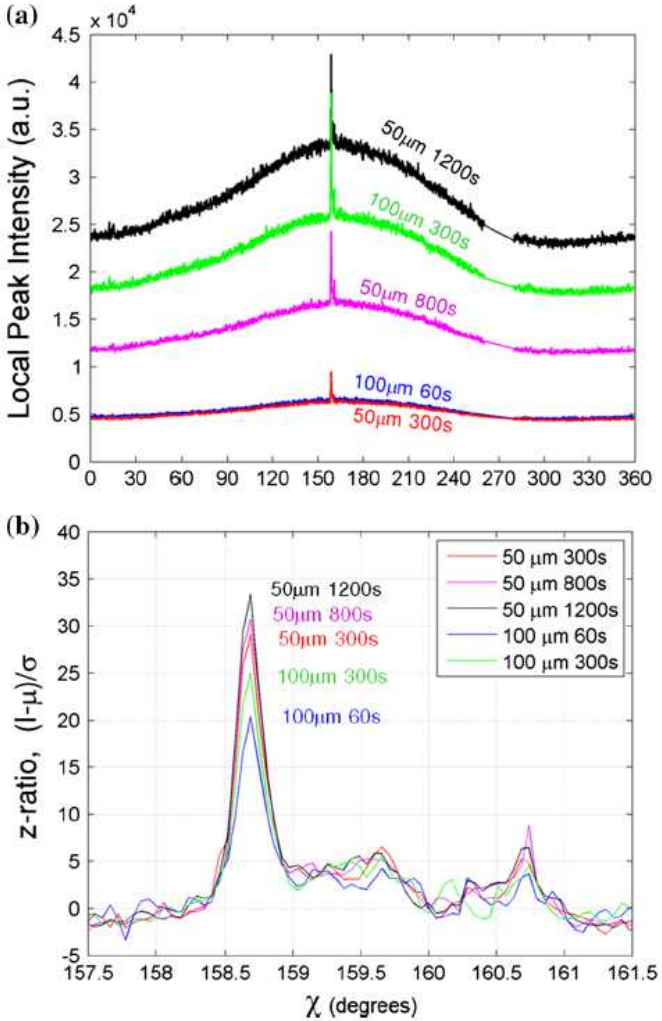
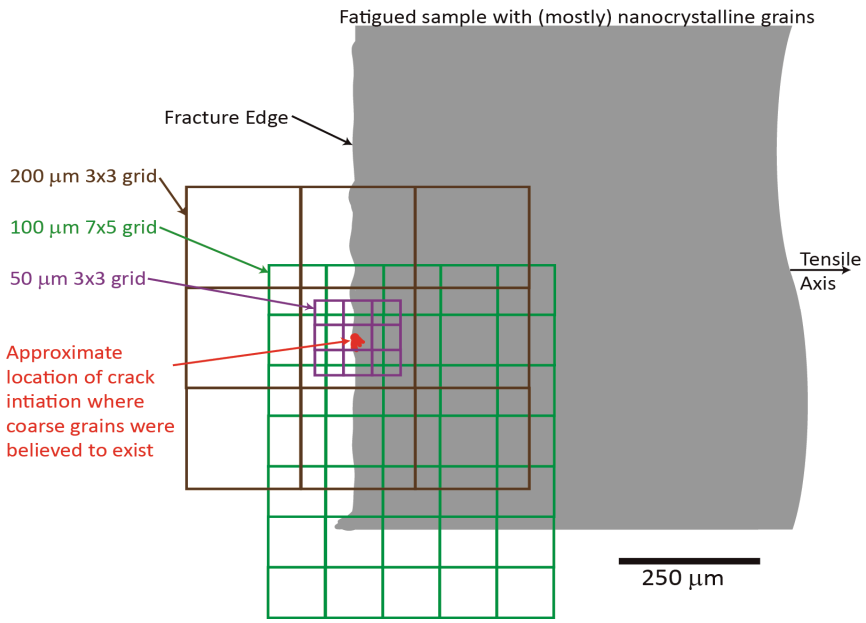
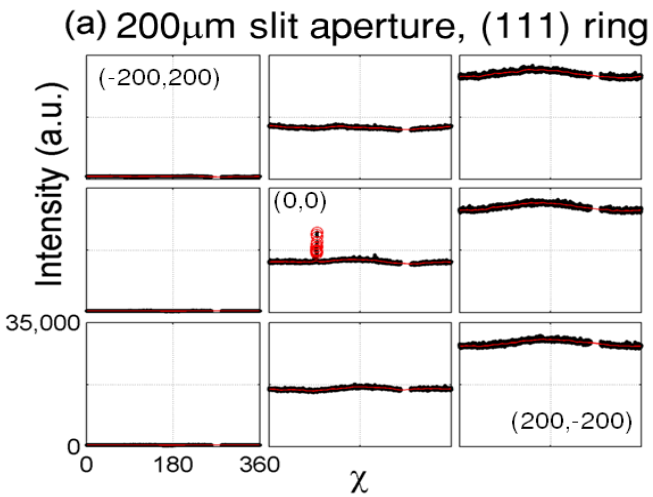


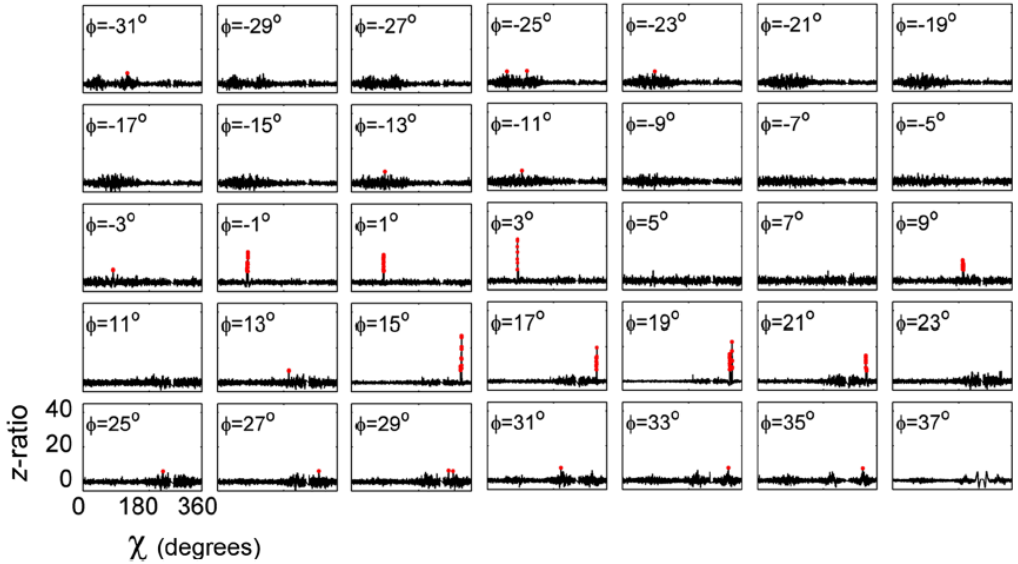
Table 1 Q data for the (111), (200), and (220) peaks

Peak	Q (Å^{-1})	Q range analyzed (Å^{-1})
(111)	3.08	2.7–3.3
(200)	3.56	3.3–3.9
(220)	5.03	4.7–5.3

This x-ray technique identified the AGG region, despite only occupying < 0.0001% of the sampled volume!

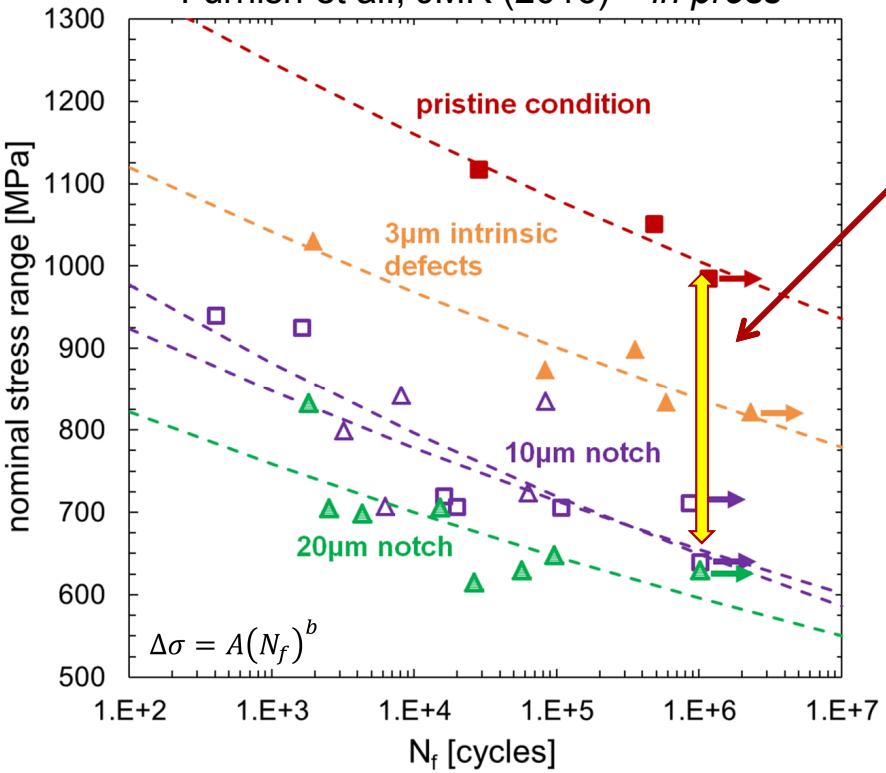






Notched fatigue tests revealed dramatic fatigue stress concentrations and high notch sensitivity!

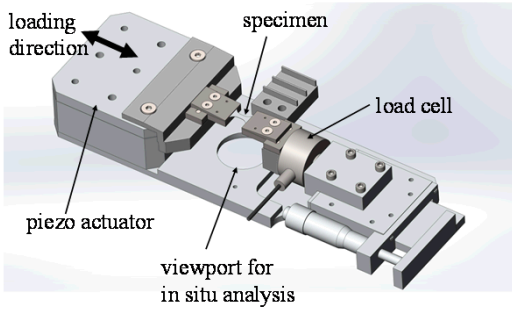
*Furnish et al., JMR (2016) – *in press*



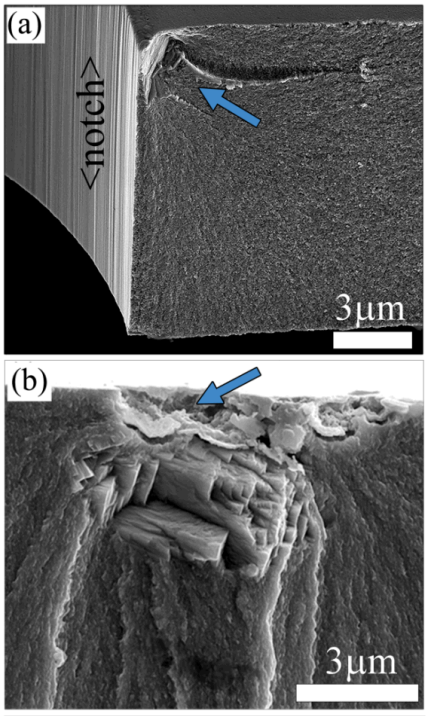
$$K_f = \frac{\Delta\sigma_{unnotched,lim}}{\Delta\sigma_{notched,lim}}$$

$$q = \frac{(K_f - 1)}{(K_t - 1)}$$

q = 0 (totally notch insensitive)
q = 1 (fully notch sensitive, K_f=K_t)



Custom built piezo actuated *in situ* fatigue tester for thin foil testing



- K_f and q values:
  - 20 μm notch: 1.7 / 0.37
  - 10 μm notch: 1.5 / 0.28
  - 3 μm intrinsic defects: 1.2 / na

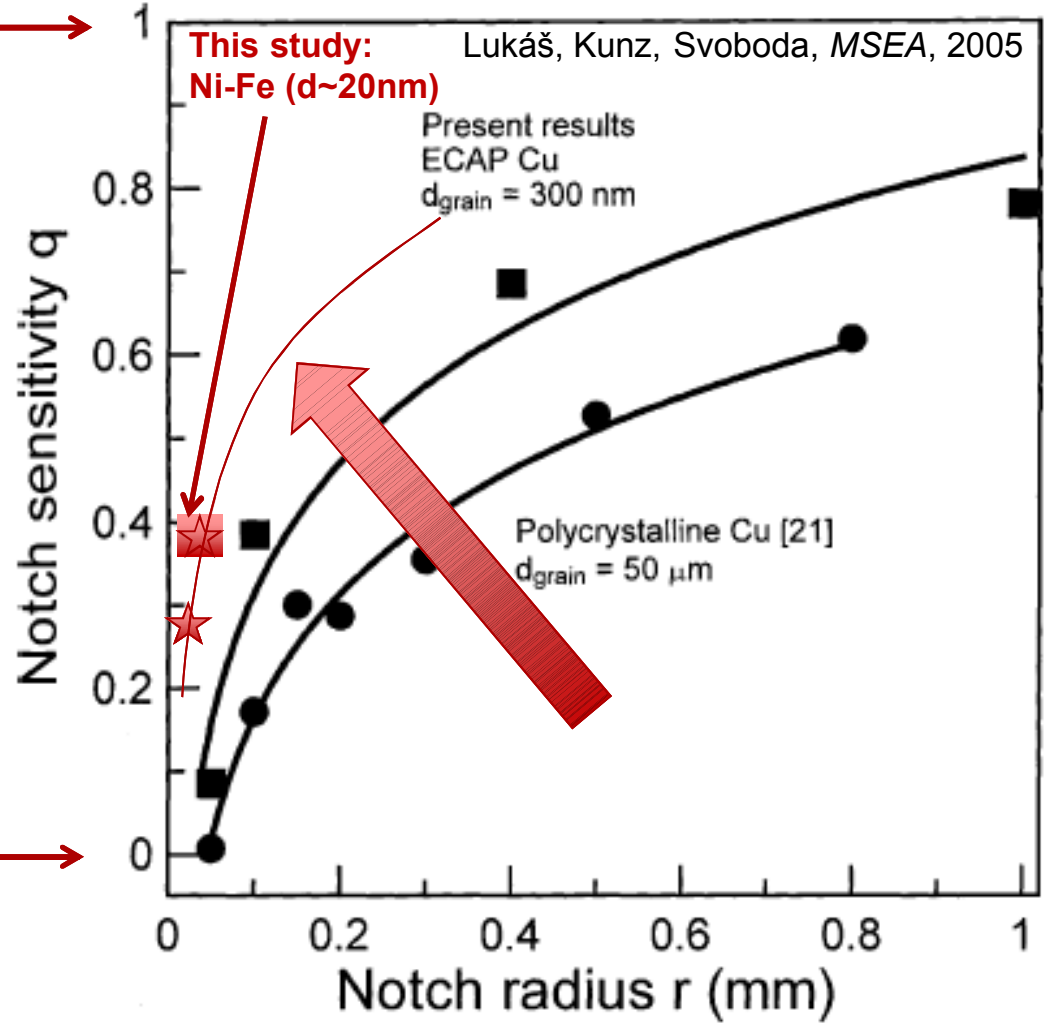
In all cases, the fatigue crack initiation was found to occur directly below both the intrinsic and extrinsic defects!

Notch effects diminish as two length scales converge

Notches obey elastic stress concentration factor

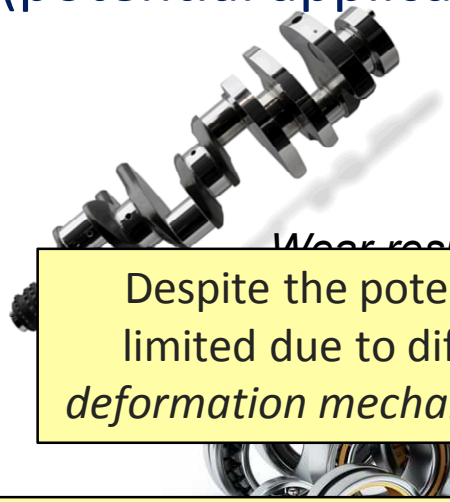
$$q = \frac{(K_f - 1)}{(K_t - 1)}$$

Notches are Irrelevant!!!



Nanocrystalline metals are *more* sensitive to stress concentrations than their coarse-grained counterparts!

Why do we care about fatigue in NC metals? (potential applications….)

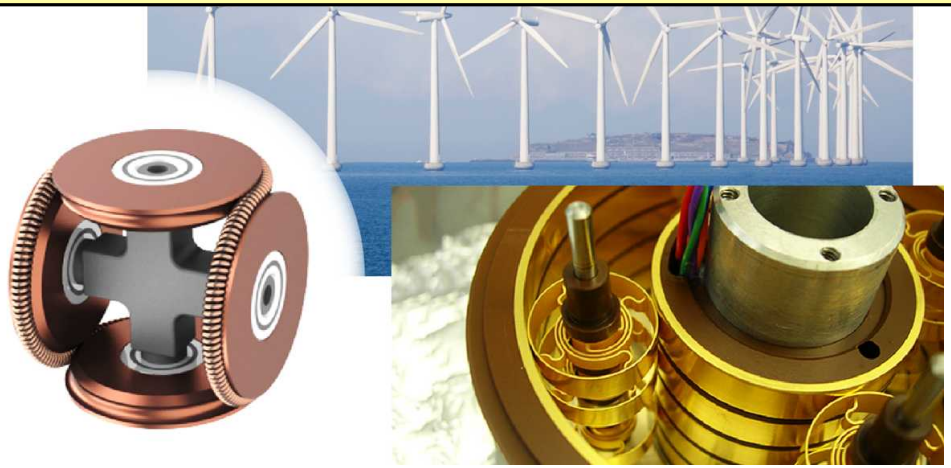


Ultra-hard thin films

Wear resistant coatings

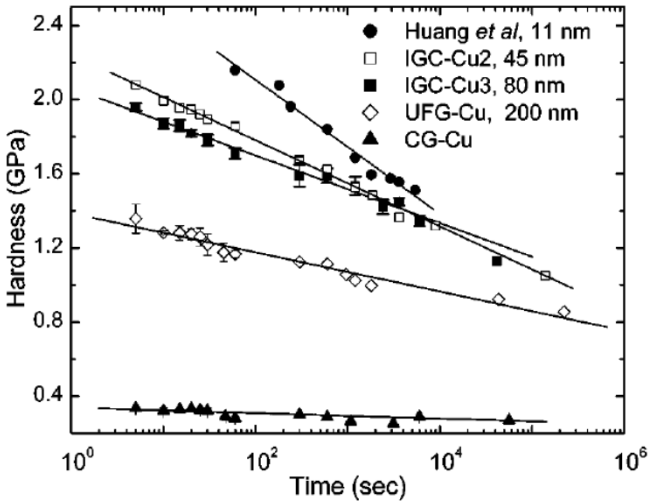
Despite the potential of NC metals for advanced applications, their use has been limited due to difficulties in *material synthesis* and lack of understanding of their *deformation mechanisms*, particularly during complex loading scenarios (e.g., fatigue).

In this study, the fatigue and fracture behaviors of *electroplated* NC metals were evaluated to ***assess the role of the microstructure on the macro-scale mechanical properties*** and to lay the foundation for long-term reliability.



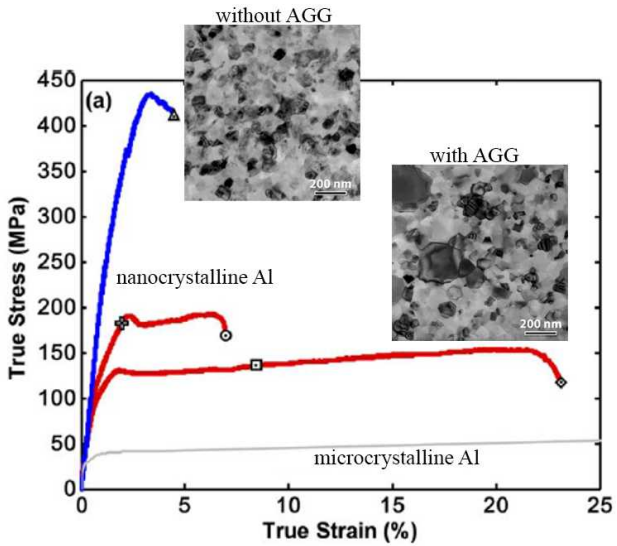
Mechanically-induced grain growth as a potentially *life-limiting process* for NC metals…

Hardness evolution of NC Cu during indentation



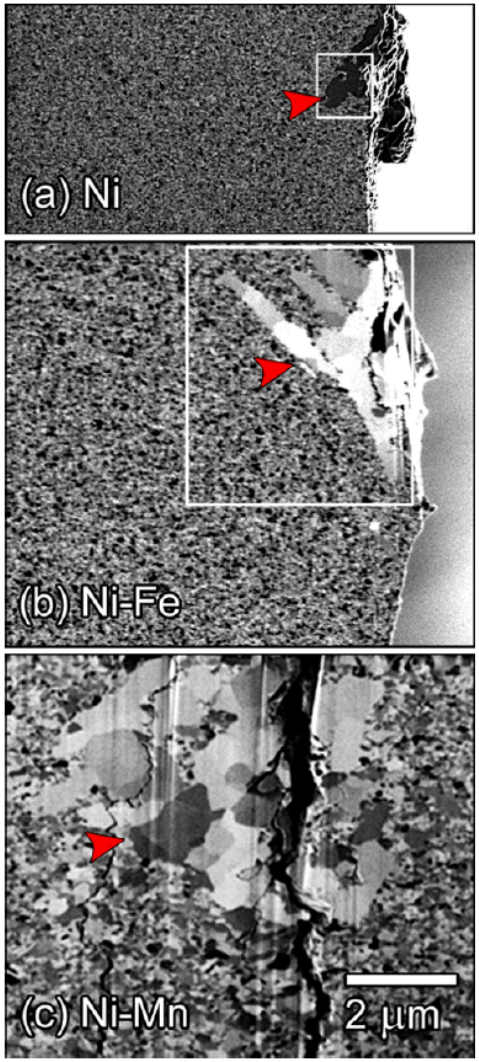
K. Zhang, APL (2004)

Grain growth during mononotic loading leading to decreased strength



Gianola et al., Acta Mater. (2006)

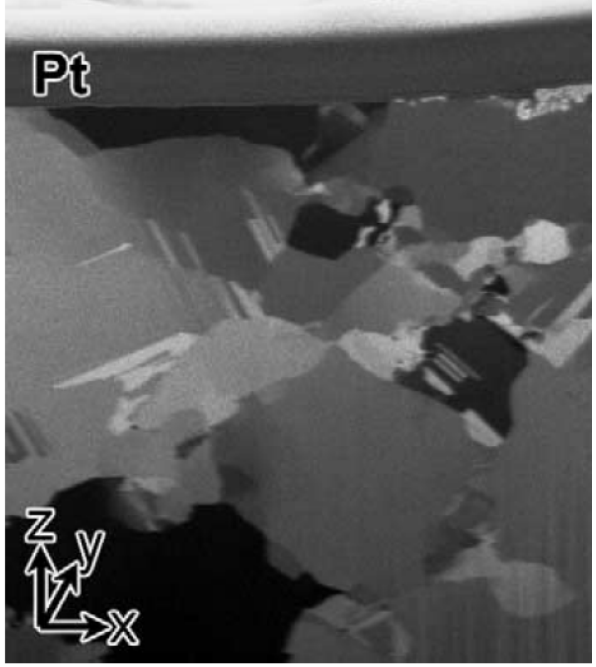
Fatigue-induced grain growth leading to crack initiation

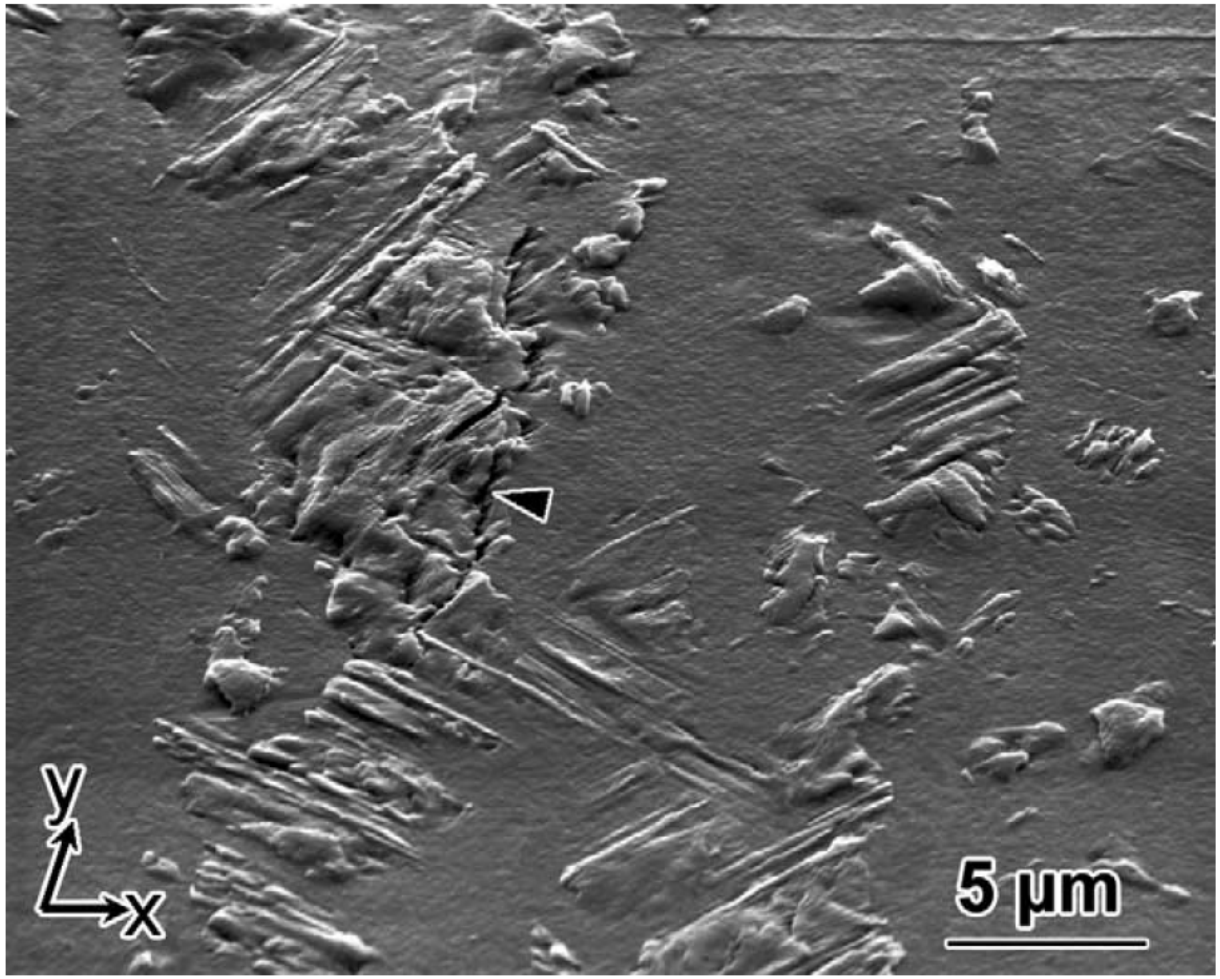


Boyce, Met. Mat. Trans. A (2011)

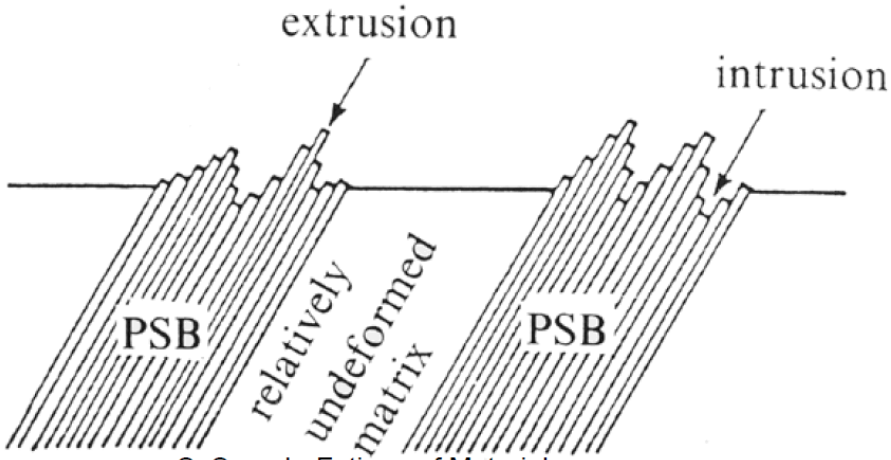
The propensity for mechanically-induced grain growth in NC metals has been suspected to play a major role in the macro-scale mechanical properties! But, the necessity for *post-fracture* characterization has led to only speculation…

Deformed
Zone

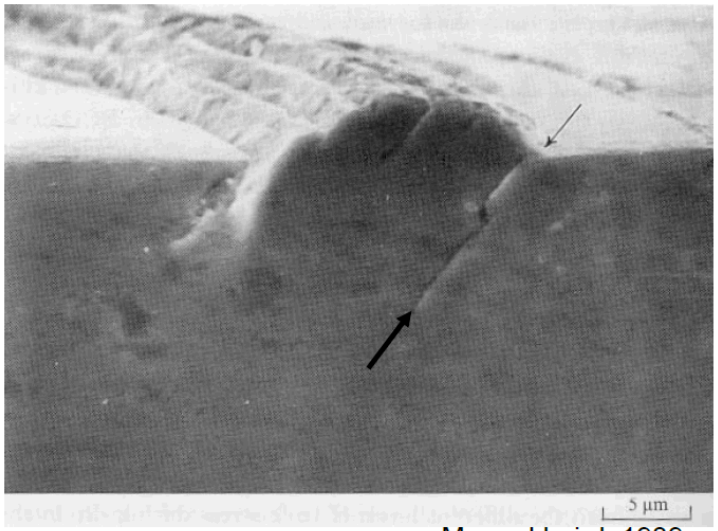

Pt
z
y
x


y
x
5 µm

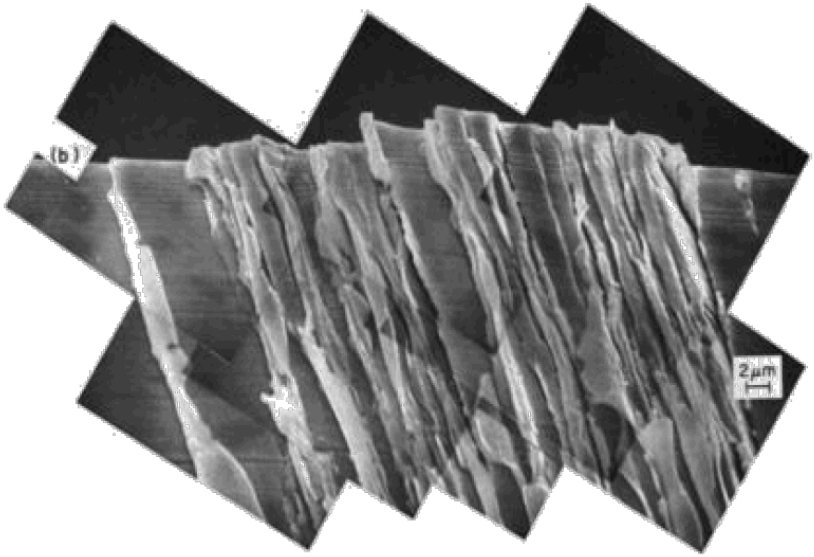
Is the fatigue mechanism suppressed in nanocrystalline metals?



S. Suresh, Fatigue of Materials



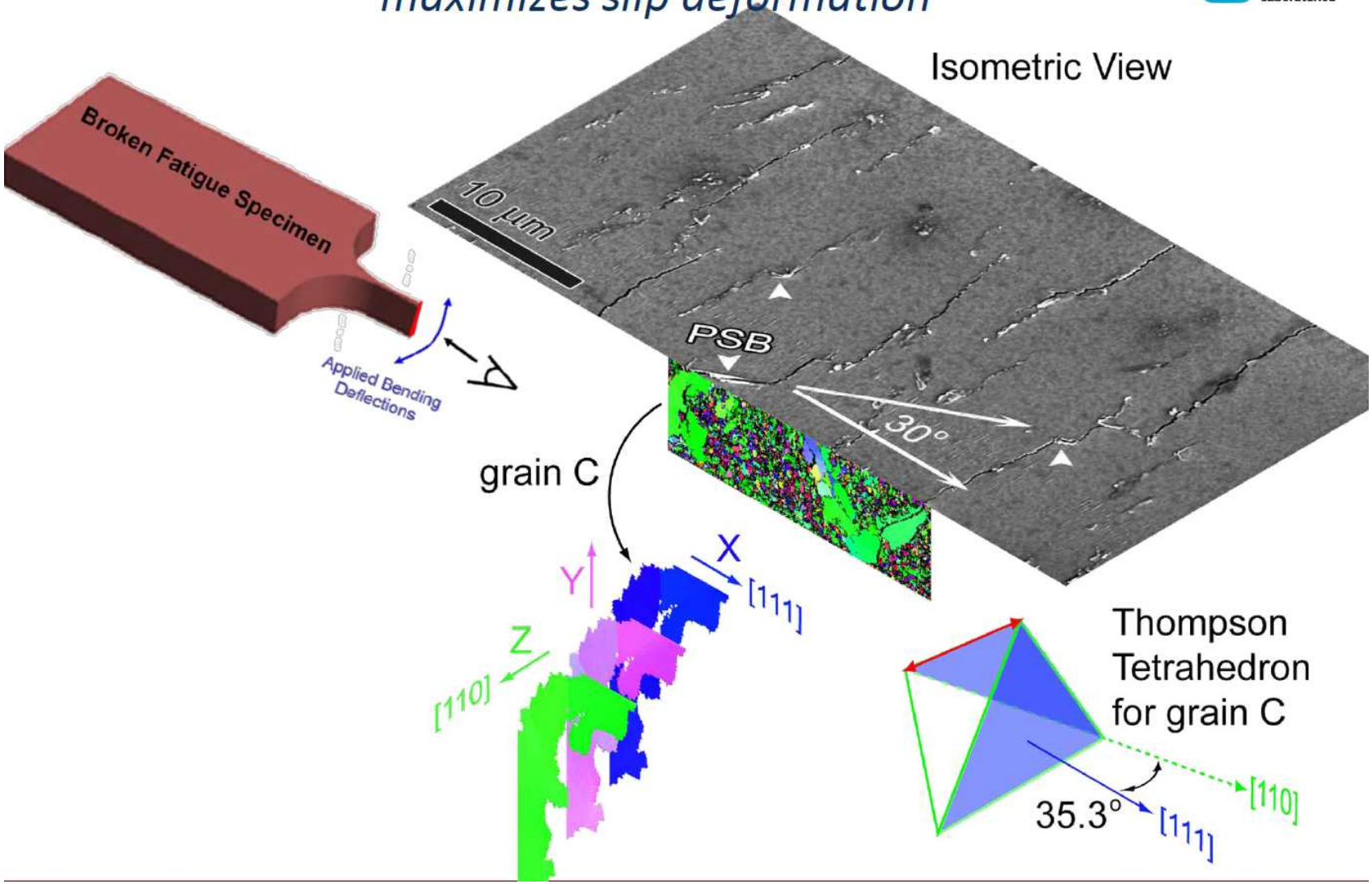
Ma and Laird, 1989



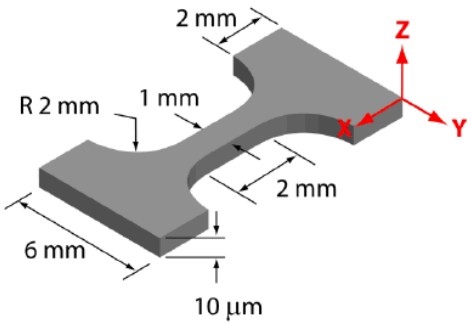
A billion nanocrystalline grains would fit inside a single microcrystalline grain of a traditional structural alloy.

Does the fatigue mechanism change for such small grain sizes?

The crystalline orientation of the large grains maximizes slip deformation

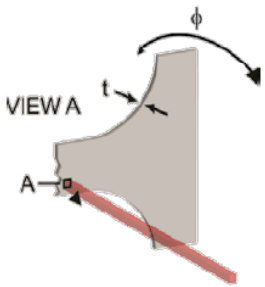


the needle-in-a-haystack challenge…
Rapidly detecting 1 abnormal grain in 10^{12}



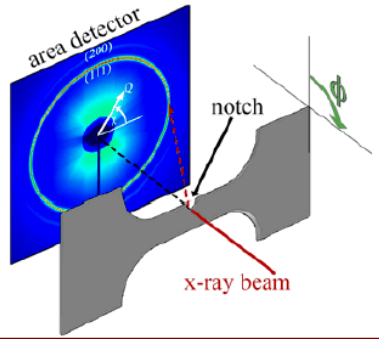
The gage section contains

~1 x10^{12} grains



A 100x100 µm x-ray spot interrogates

~1 x10^{9} grains



A 10 µm notch localizes the peak stress to

<<1 x10^{7} grains

(fatigue test terminated here)

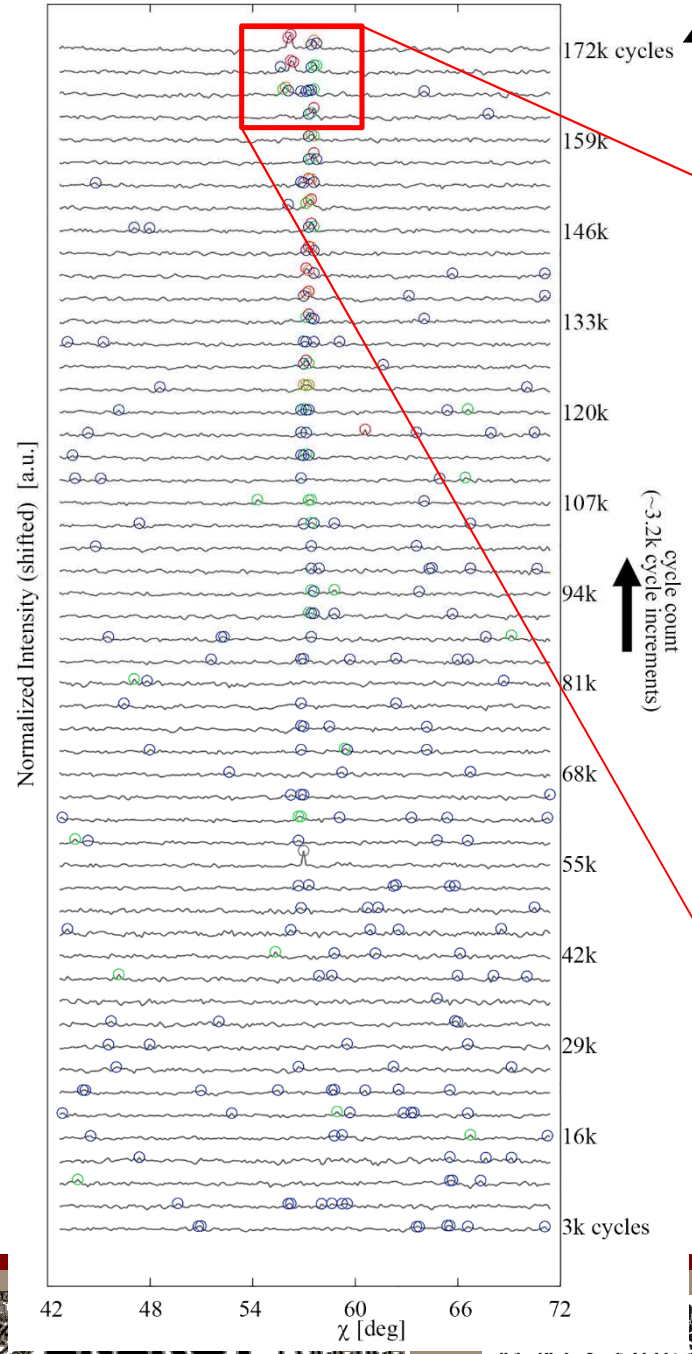
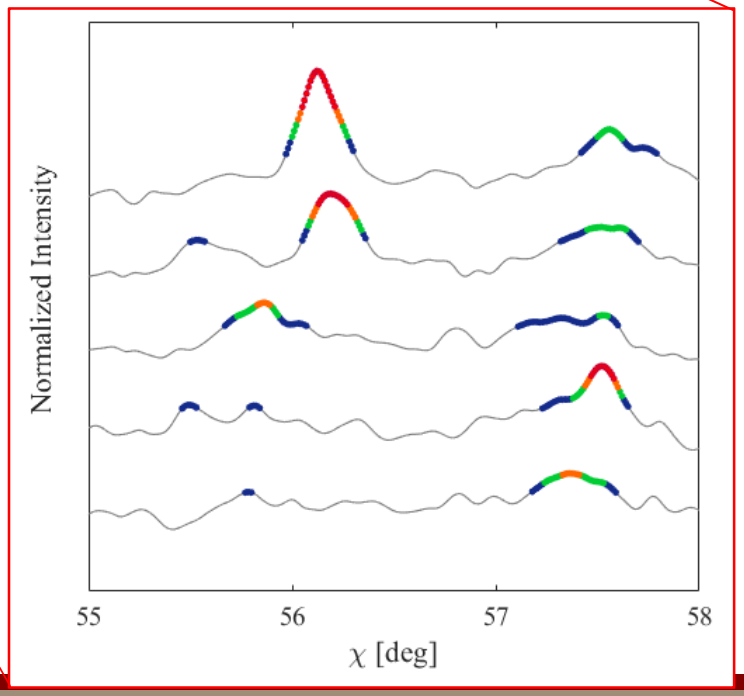


Figure. XRD intensities normalized by their means at regular intervals (~3k cycle increments) of fatigue revealing the development of statistically anomalous intensities associated with fatigue-induced abnormal grain growth. Open circles indicate intensities of different standard deviations above the local mean with 1.5-2σ (purple), 2σ-3σ (blue), 3σ-4σ (green), 4σ-5σ (orange), and greater than 5σ.

Ultimately, the signal from the AGG becomes indistinguishable from noise. However, by viewing the additional 1.5σ-3σ data point, it can argued that the detection of AGG actually began closer to 42k cycles. It is difficult to say based on this data that the AGG did not actually begin at the beginning of the fatigue. To fully address this, the signal-to-noise ratio of the diffraction of the AGG must be further improved (e.g., decreasing the spot size, thus increasing the volumetric ratio of the AGG compared to the surrounding NC grains).



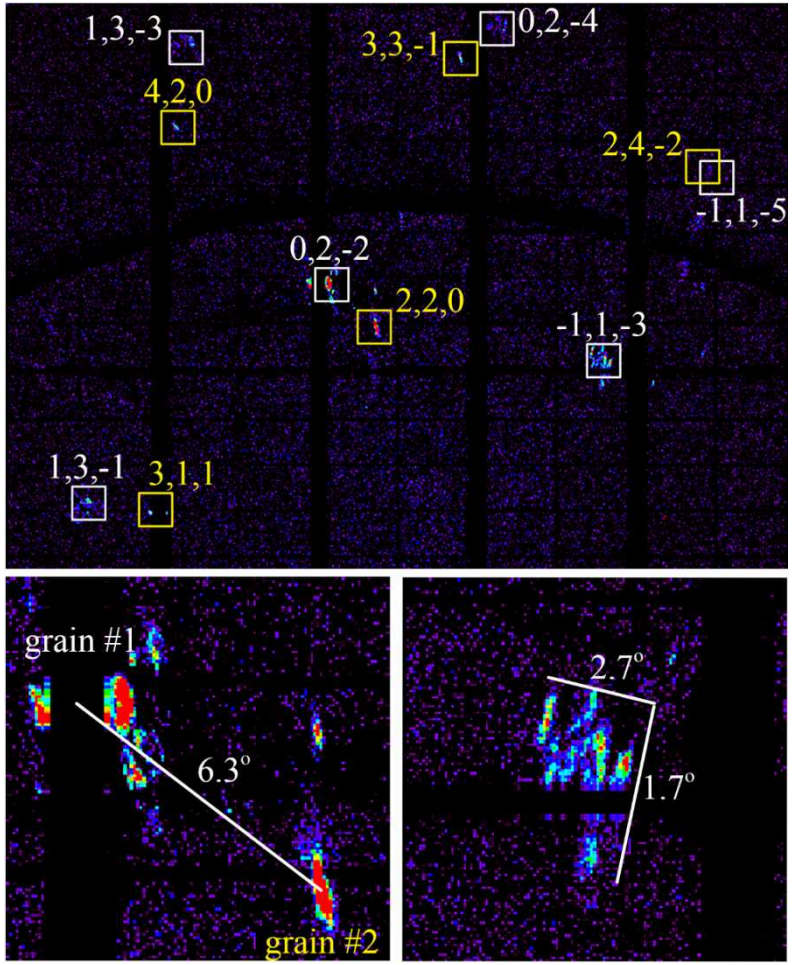
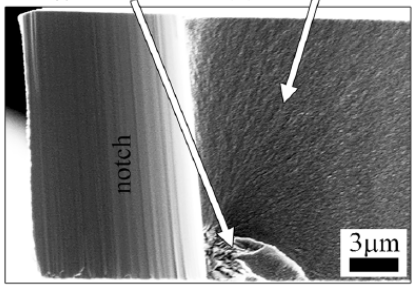


Figure. Whitebeam Laue diffraction from a second fatigued NC Ni-Fe specimen in which case additional diffraction spots were resolved, enabling indexation of the AGG region. Iin this case, two separate grains were observed with ~6° orientation difference (i.e., a relatively low-angle grain boundary between two grains). (Advanced Light Source BL 12.3.2, whitebeam spectral range ~6-22 keV, DECTRIS Pilatus 1M Pixel array detector).

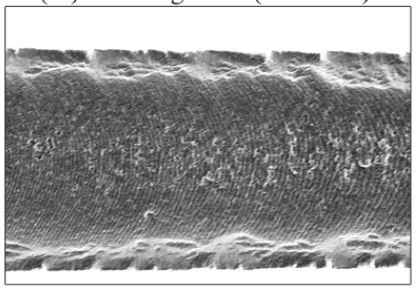
(note, this diffraction data is from sample W4DB15).

In the specimen used in this study, the whitebeam diffraction conditions were not optimized to detect more than a few spots. However, in a similar specimen in which the fatigue was halted after detection of abnormal grain growth, the whitebeam conditions were more ideal to produce multiple spots capable of indexation. This sample experienced more than xx cycles after the first detection of AGG but prior to fatigue failure when the test was stopped. …

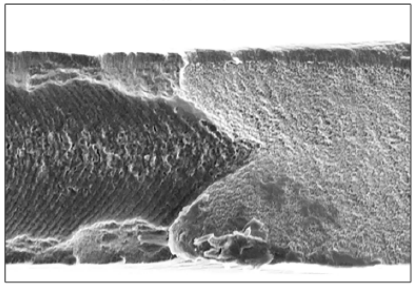
(i) initiation (ii) initial growth



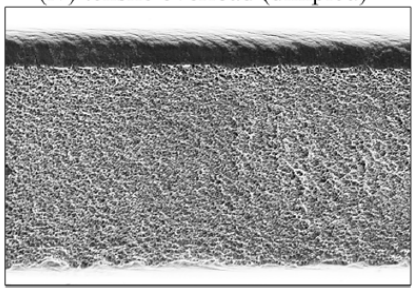
(iii) mode-I growth (striations)



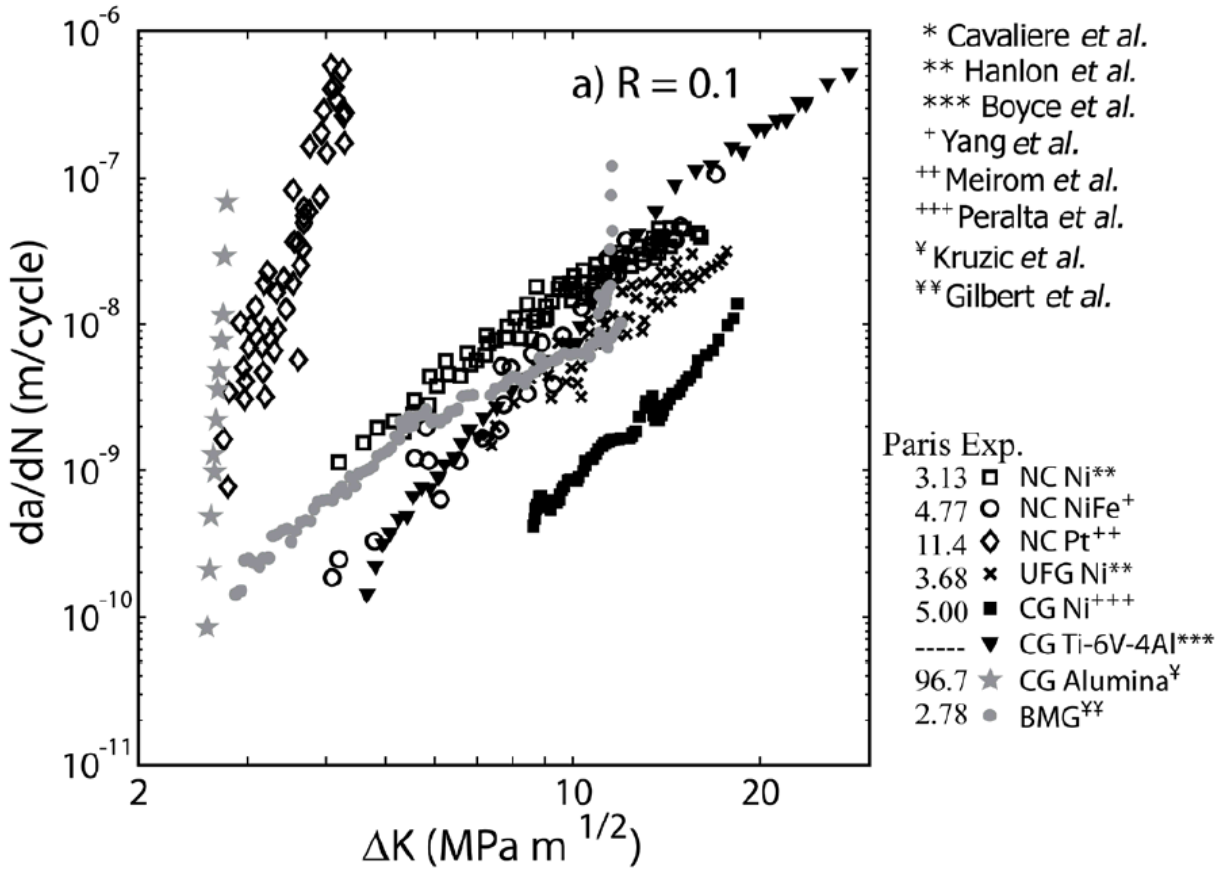
transition



(iv) tensile overload (dimpled)



Crack growth resistance is worse than coarse-grained Ni, as expected



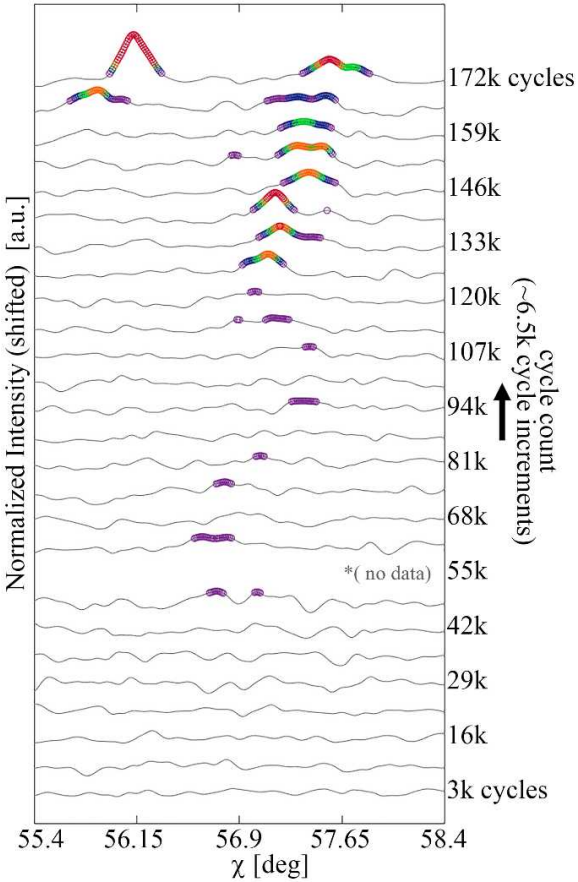


Figure 2. XRD intensities (normalized by their averages and shifted for visualization) in the χ range that anomalous high intensities associated with ALGs were observed in-situ during cyclic loading. The fatigue was intentionally interrupted upon first detection of ALGs at 172k cycles, though more detailed data analysis after the fact revealed anomalous intensities before 55k cycles. Peak shifts in this case is attributed to in-plane rotation of the growing ALGs while the broadening and subdivision is consistent with subgrain formation.

analyzed in near real-time during cyclic loading, allowing us to stop the fatigue process upon detection of AGG.

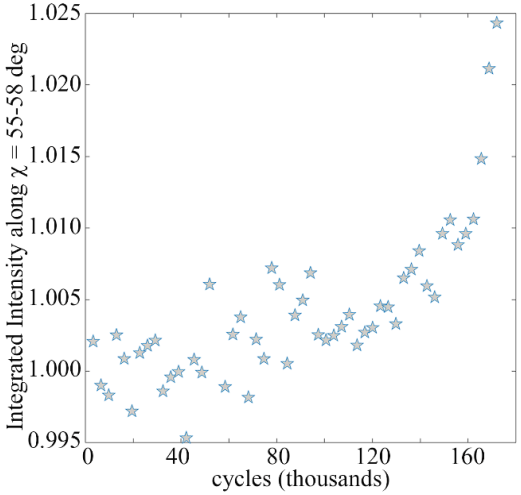


Figure 3. Total intensity across the χ range of interest (cumulative intensity from χ=55.4° to 58.4°) as a function of the number of cycles indicating continuous growth of the ALGs during cyclic loading.

despite XRD evidence of AGG during fatigue, a fatigue crack is notably absent. Finally, the chicken-or-egg question is answered: indeed, AGG preceded crack initiation