Sandia National Laboratories

6th International Conference on Recrystallization and Grain Growth
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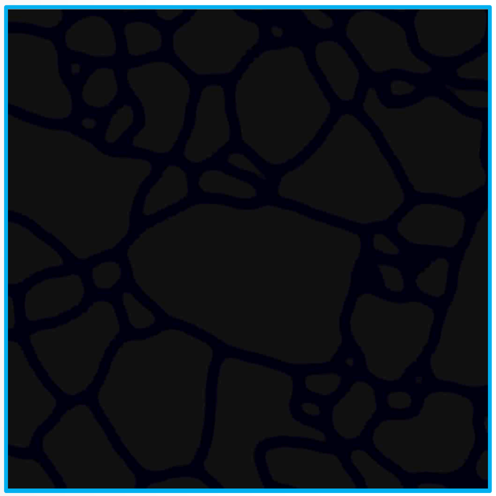
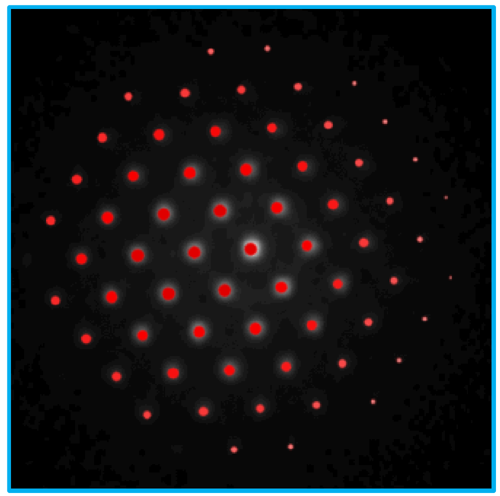
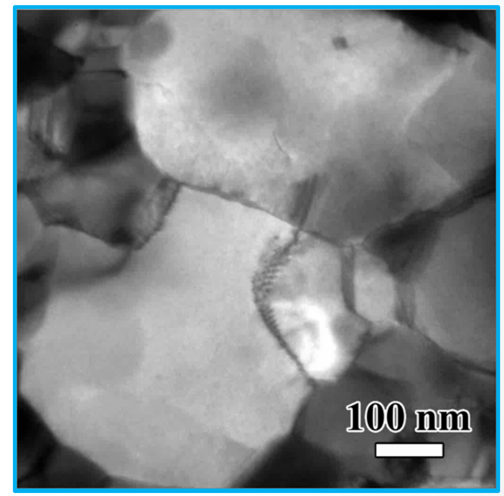
Pittsburgh ReX GG 2016

SAND2016-7168C

Coupling *in situ* TEM and phase field modeling to better understand grain growth in nanocrystalline metals







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U.S. DEPARTMENT OF ENERGY
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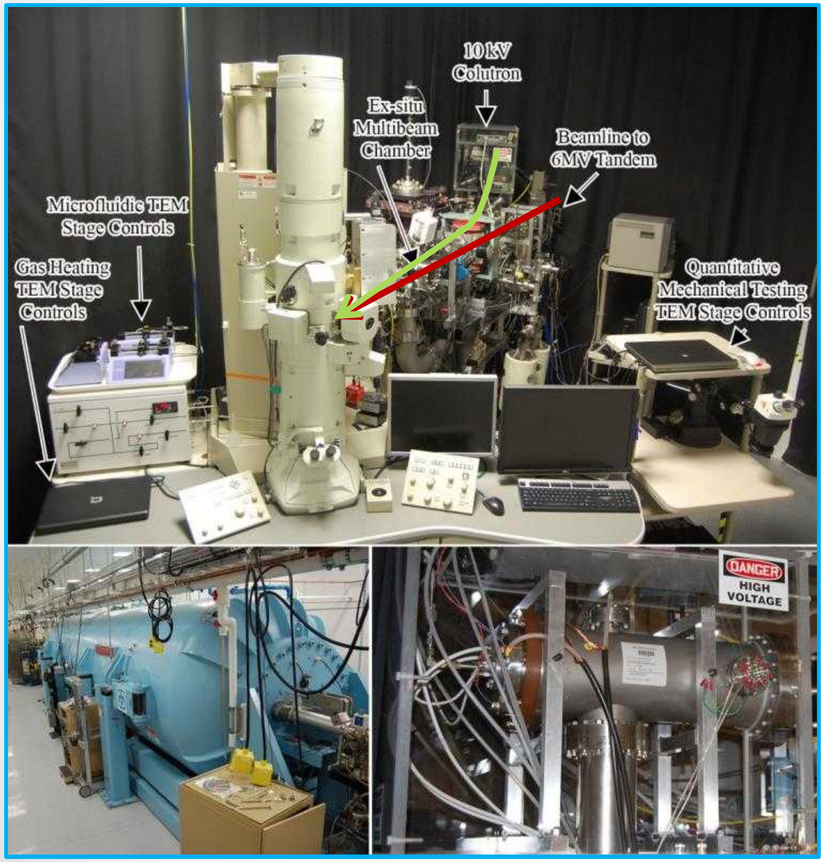
18 February 2016

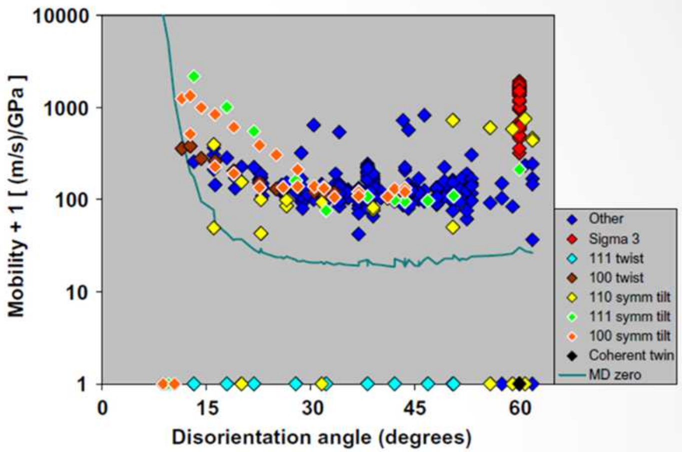
Sandia's *In situ* Ion Irradiation TEM (I^3TEM)

Collaborator: D.L. Buller

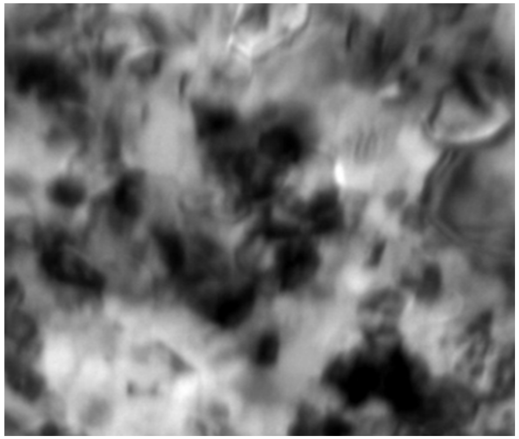
- 10 kV Colutron - 200 kV TEM - 6 MV Tandem

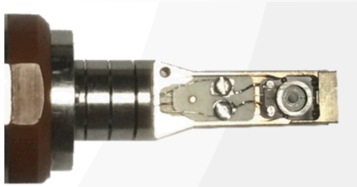


Hattar, *et al*, Nucl Instr Meth Phys Res B, 2014.



Olmstead, *et al*, Acta Mater, 2009.

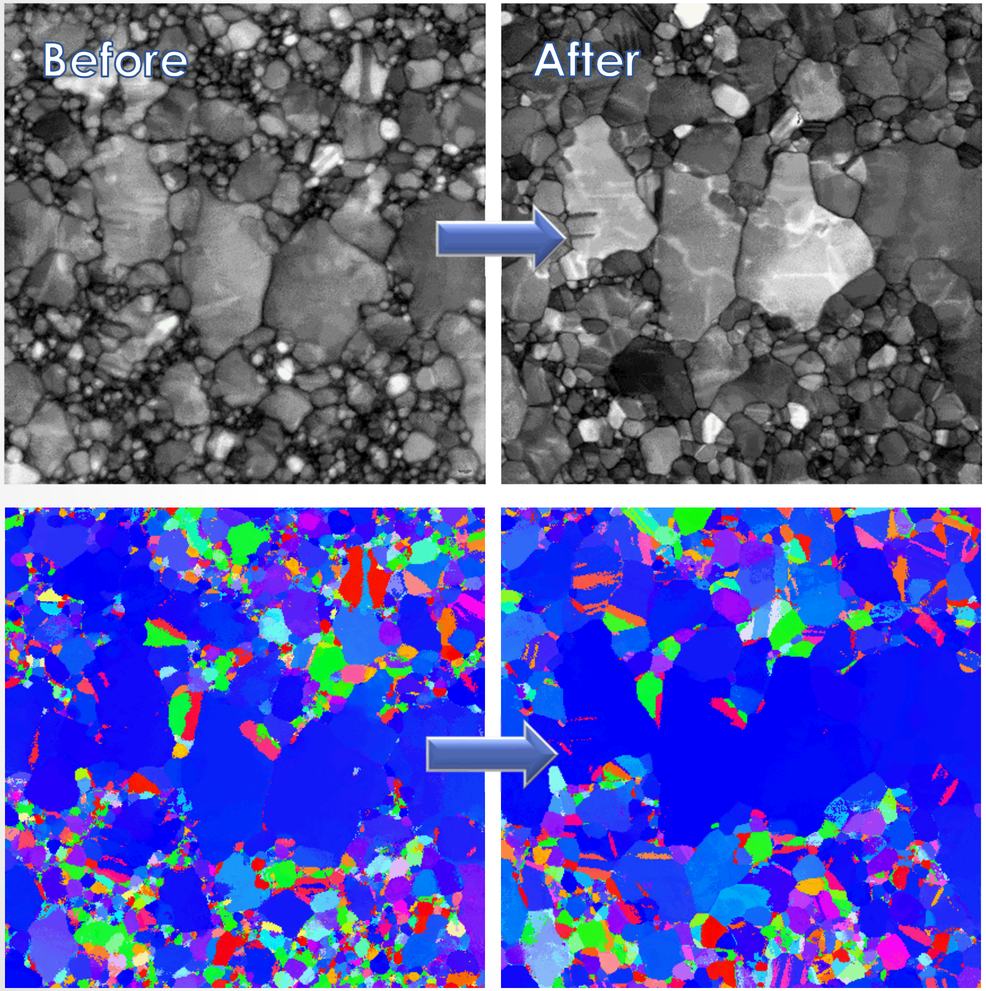




- Heating to 800 °C

Real-time observation during heating and irradiation at length scales accessible by models.

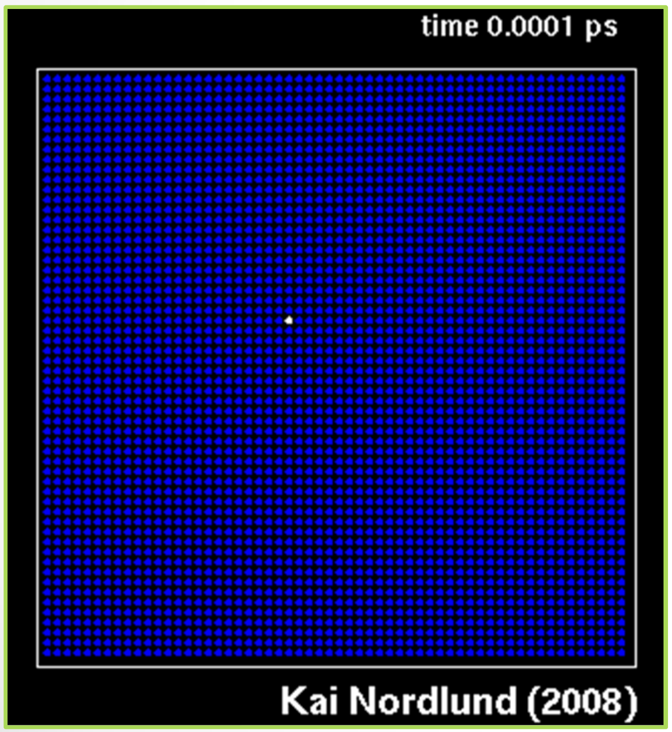
Annealing



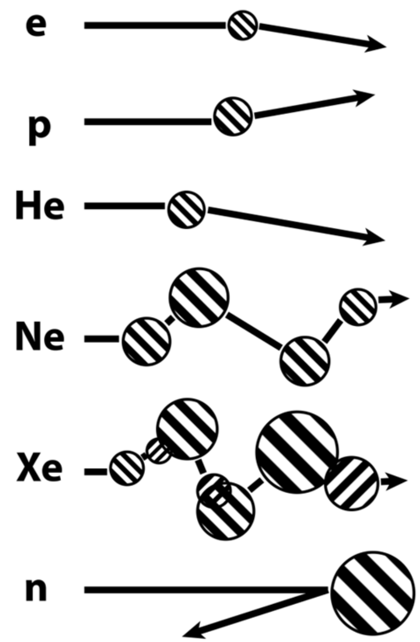
- Freestanding Au foil
  - 40 nm thickness
  - Annealed to 300 °C
- PED orientation mapping to digitize the microstructure
  - <10 nm resolution
- Captures changes in structure
  - Orientation
  - Grain size
- Direct input for use in computational models

Radiation-Solid Interactions

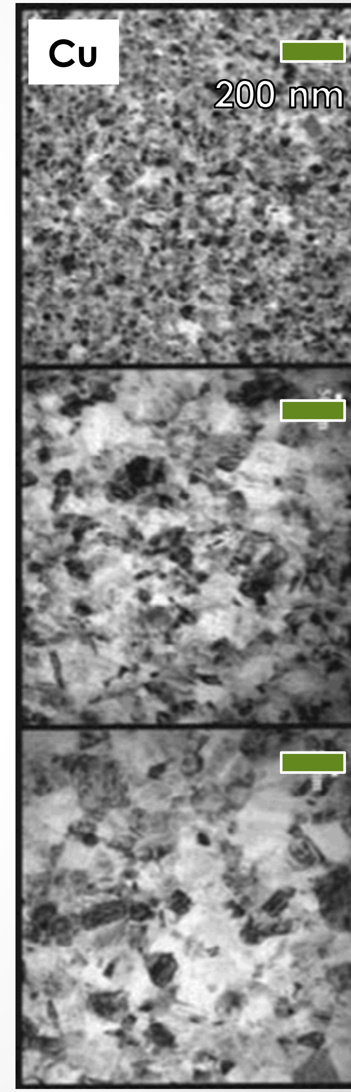
- Energetic ion displaces one or more target atoms
  - Frenkel (vacancy-interstitial) pair
  - Collision cascade
  - Nuclear and electronic interactions



10 keV Au in Au, *via* Wikimedia Commons.



Schematic recoil spectra for 1 MeV particles in Cu. Sizes represent recoil energies. After Averback, J Nucl Mater, 1994.



Kaoumi, *et al*, J ASTM Intl, 2006.

Highly temporally and spatially localized energy transfer drives microstructural change.

In Situ Irradiation

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- Au foil during bombardment with 10 MeV Si^{3+}
- ~22 s of 4000s total experiment time

In situ ion irradiation TEM: 10 MeV Si into nanocrystalline Au.

Playback at 2 × real time.

2× real time

Locations of single ion strikes and resulting microstructural change captured.

Quantification: Overall

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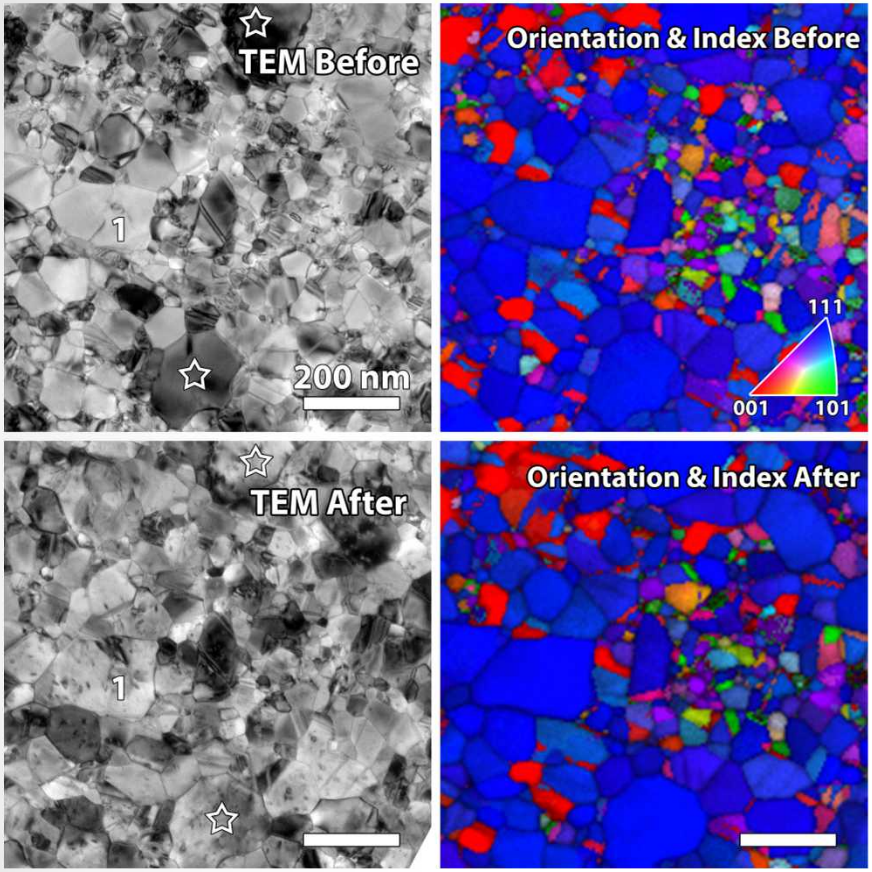

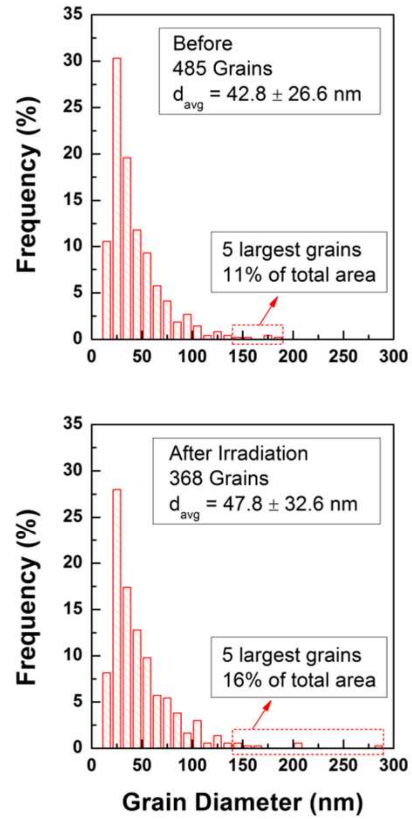

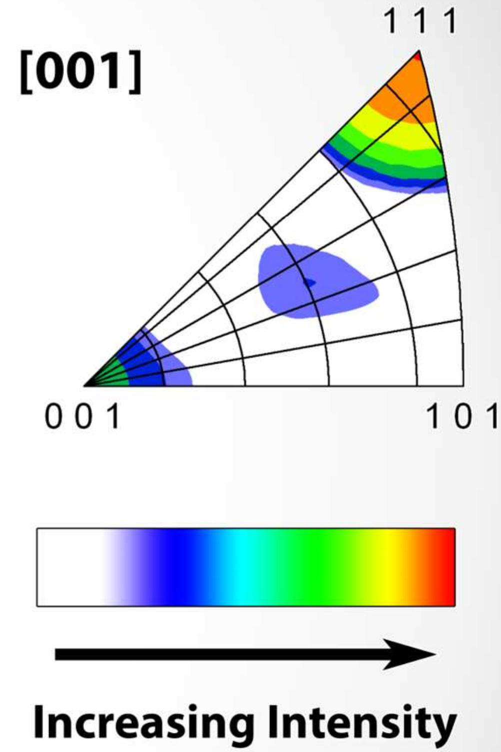

Bufford, *et al*, J Appl Phys, 2015.

- Same area characterized before and after irradiation.
  - Local grain size, orientation, boundary character
  - Hundreds of grains counted in minutes

Rapid quantification of statistically relevant numbers of grains and boundaries.

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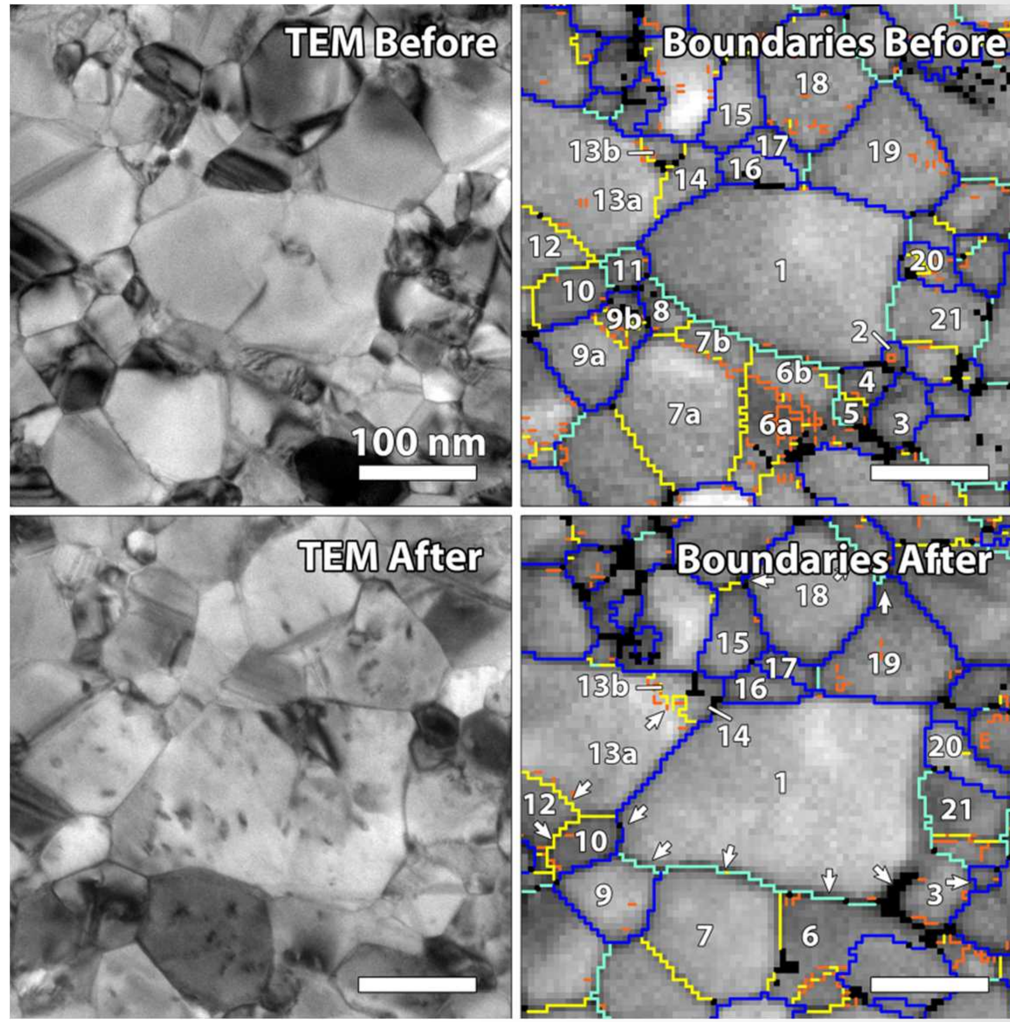
Quantification: Local

- The same grains identified before and after irradiation
- Individual grain boundary misorientation angles and axes quantified
- Correlation of GB properties and radiation-induced changes

$\phi < 3°$
$3° \leq \phi < 15°$
$15° \leq \phi < 30°$
$30° \leq \phi$



Bufford, *et al*, J Appl Phys, 2015.

Individual grain boundary misorientation angle and axes quantified

Simulated Irradiation

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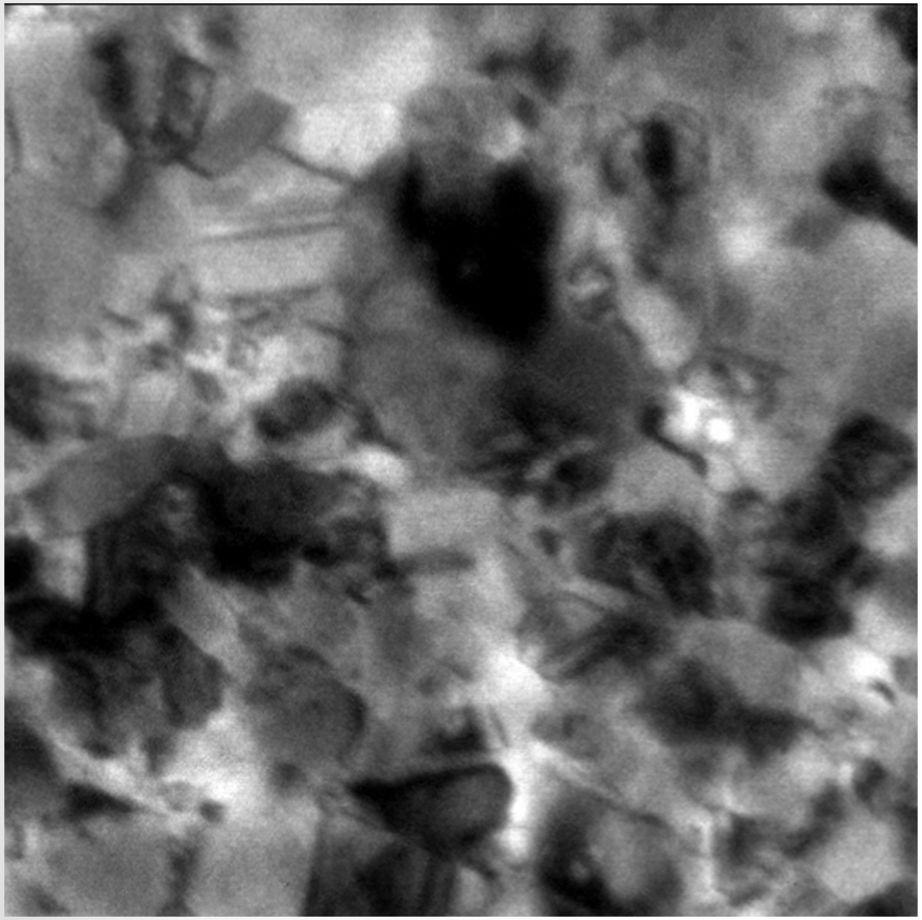
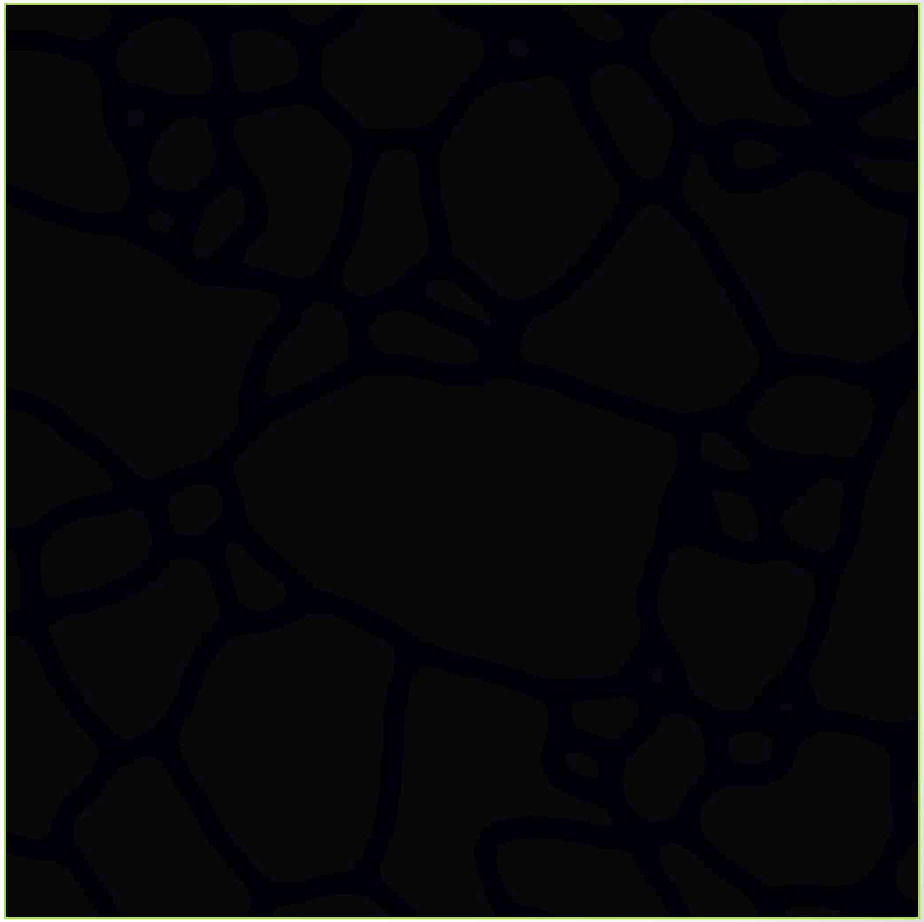
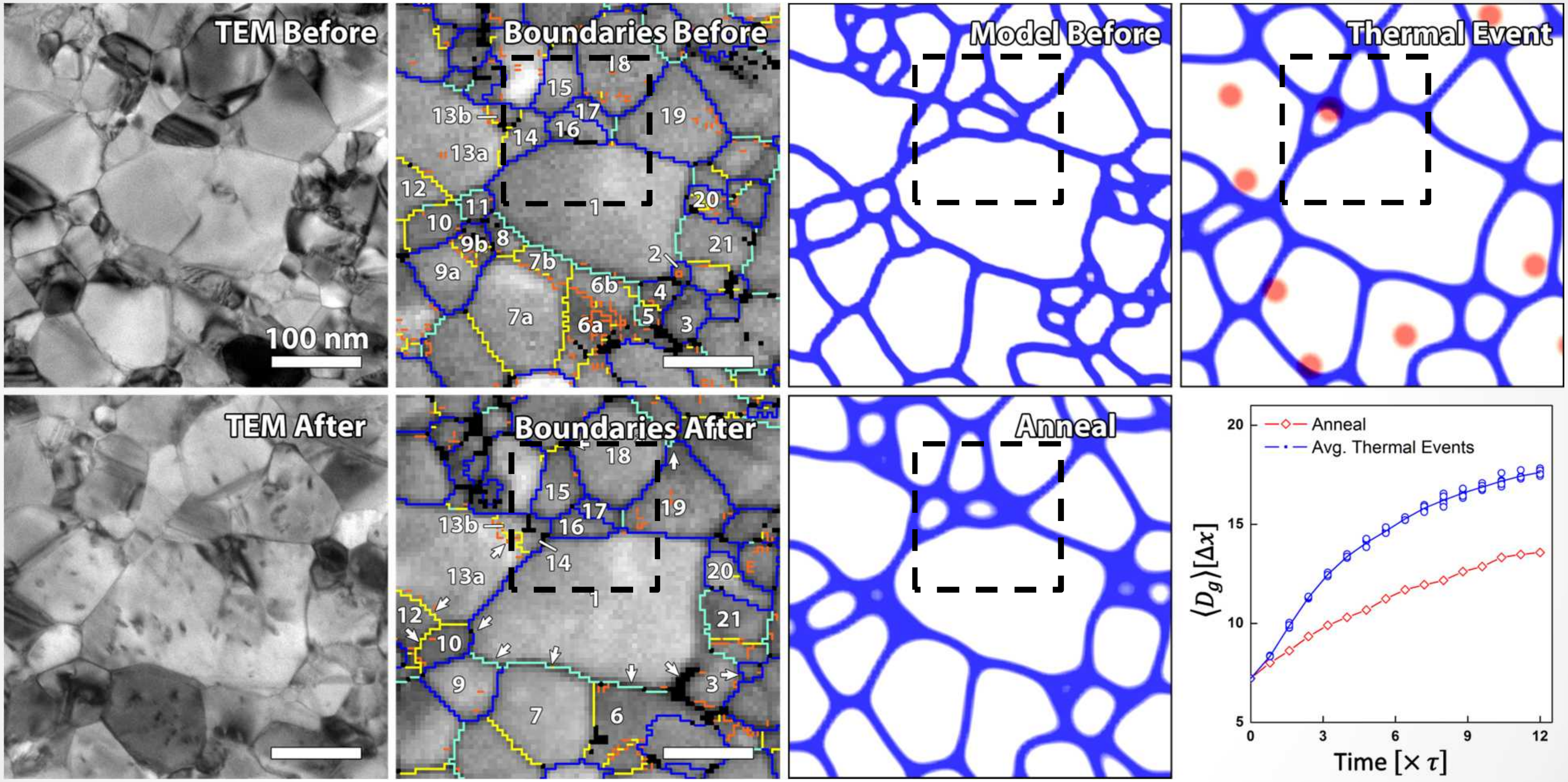




2× real time

Exp. & Model Comparison



Bufford, *et al*, J Appl Phys, 2015.

- Overall scaling laws appear consistent
- Subtle deviations from homogenous grain growth

Immobile boundaries suggest importance of non-thermally activated mobility

Model Improvements

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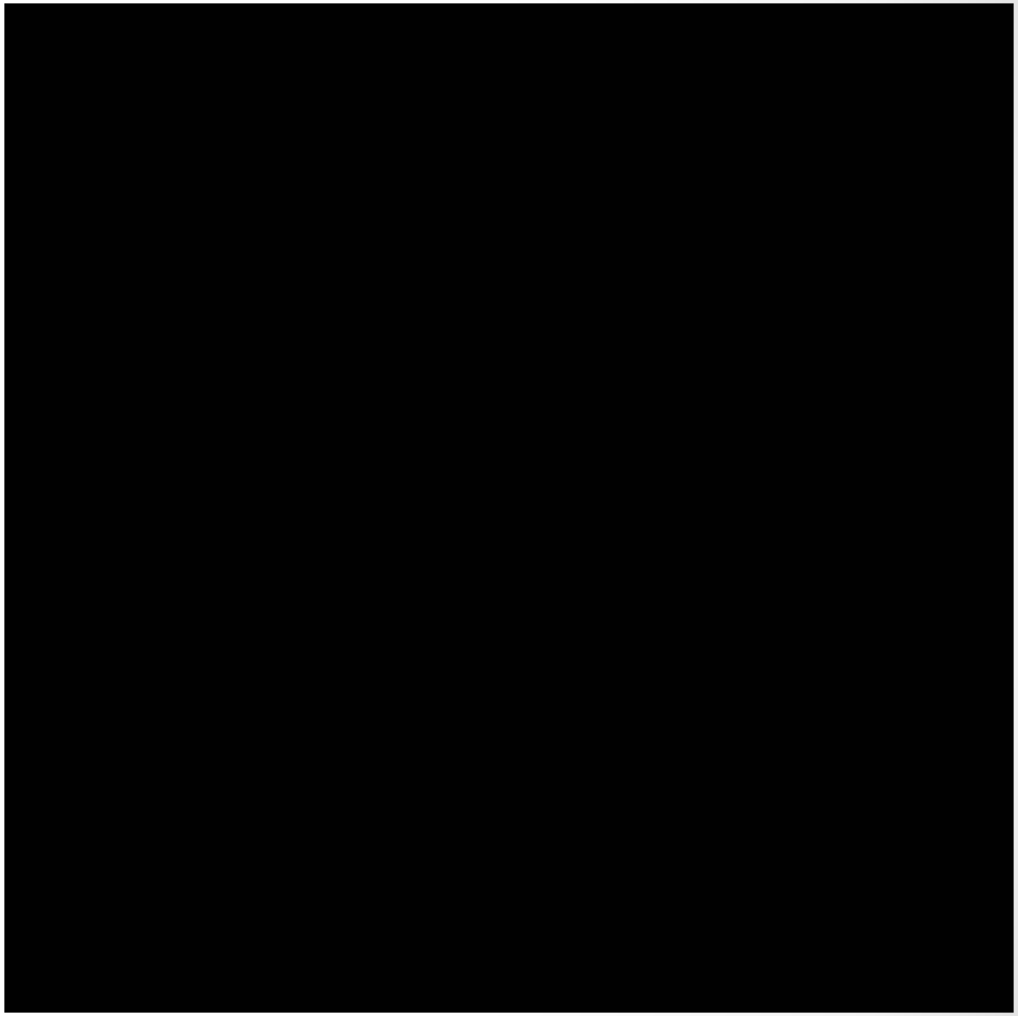
- Grain boundary mobility dependent on φ
  - Step function

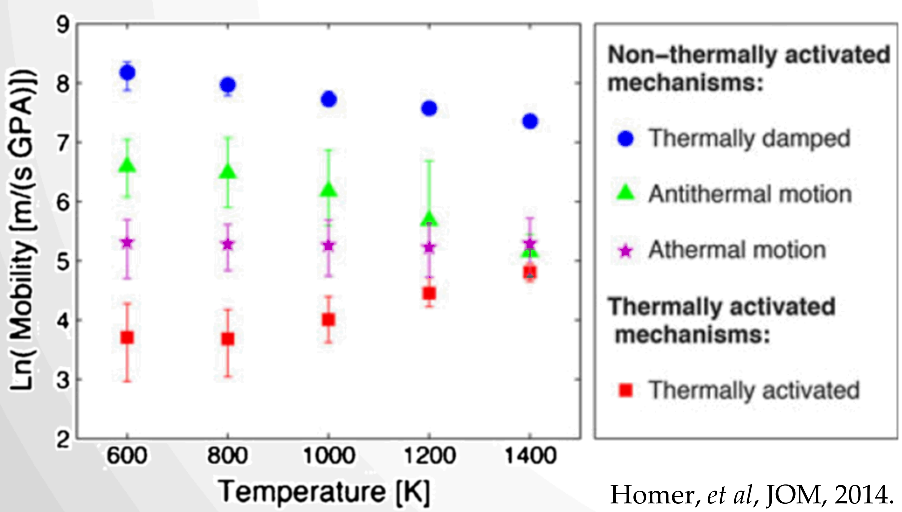
$3° \le \phi < 15°$ - $M = 0.01$
$15° \le \phi < 30°$ - $M = 0.1$
$30° \le \phi$ - $M = 1$



Implementation of heterogeneous boundary mobility

Future Directions

- Nature of Ion Interactions
  - Nuclear and electronic stopping
  - Point defects and defect clusters
- Better informed GB mobility in model
- Relationships to other driving forces for grain growth

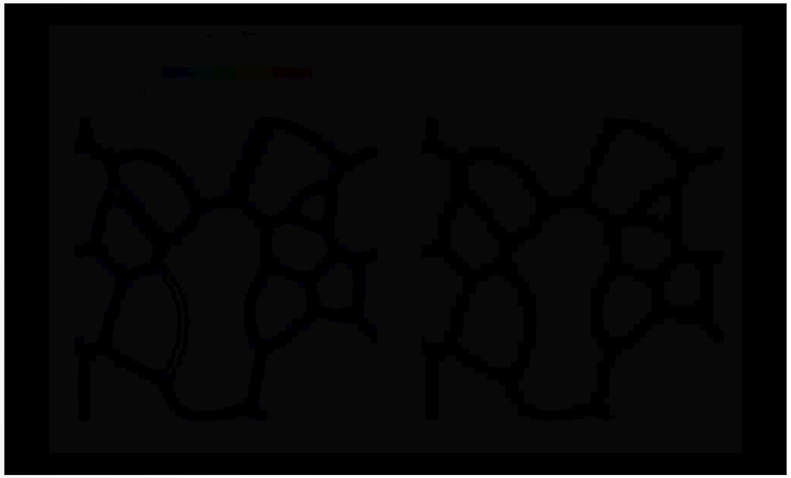


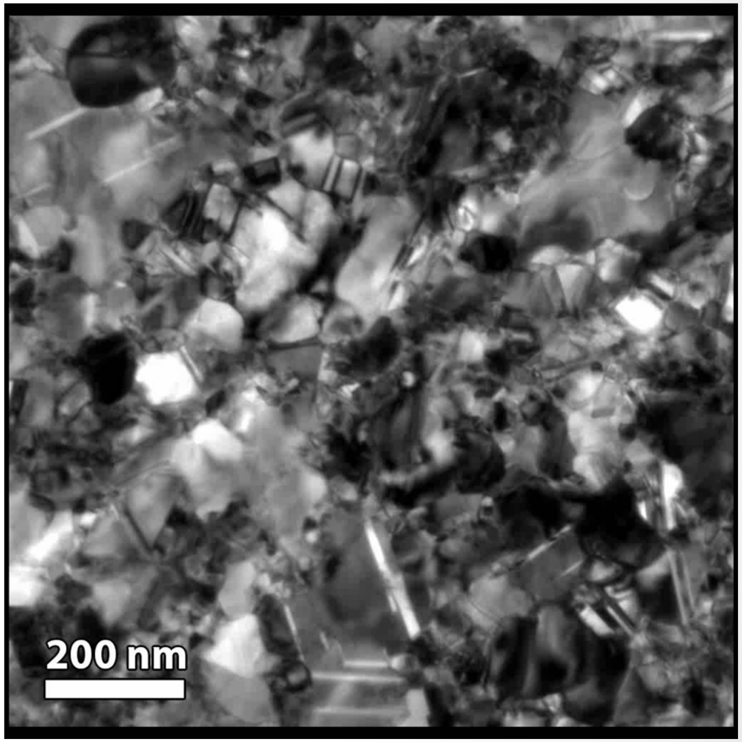
Homer, *et al*, JOM, 2014.

Bufford, *et al*, Nano Lett, 2016.

Summary

- Heating and irradiation *in situ* combined with PED orientation mapping
  - Analyzed and used as direct input for a phase field model
- Stable grains are characteristic of known low mobility grains
- Discrepancies between experimentally observed and modeled grain growth attributable to GB character





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Approach: Experimental

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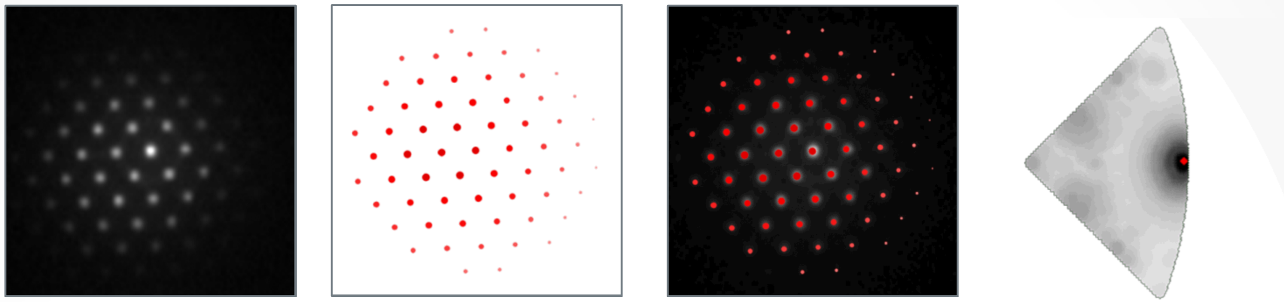
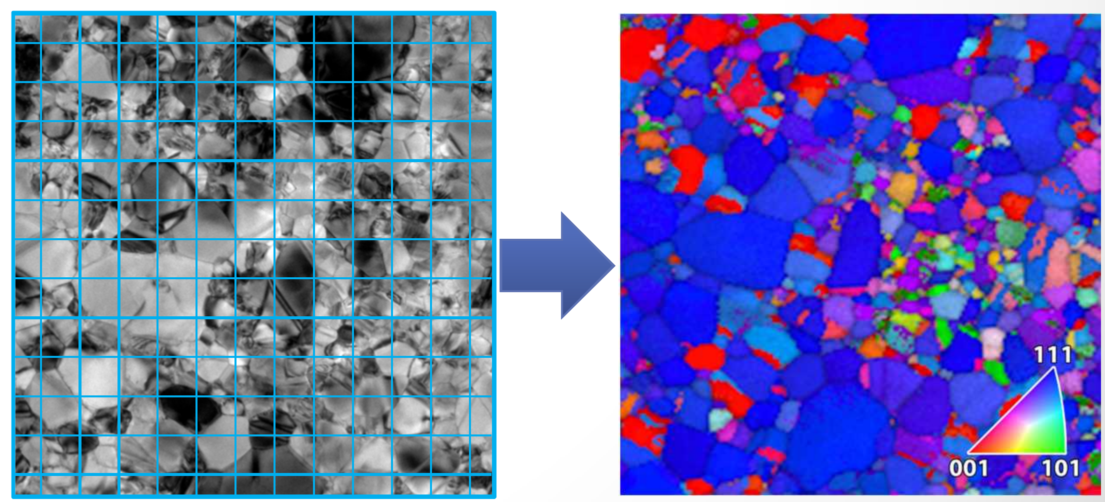


Experimental Pattern | Theoretical Template | Template Matched | Point Mapped To IPF

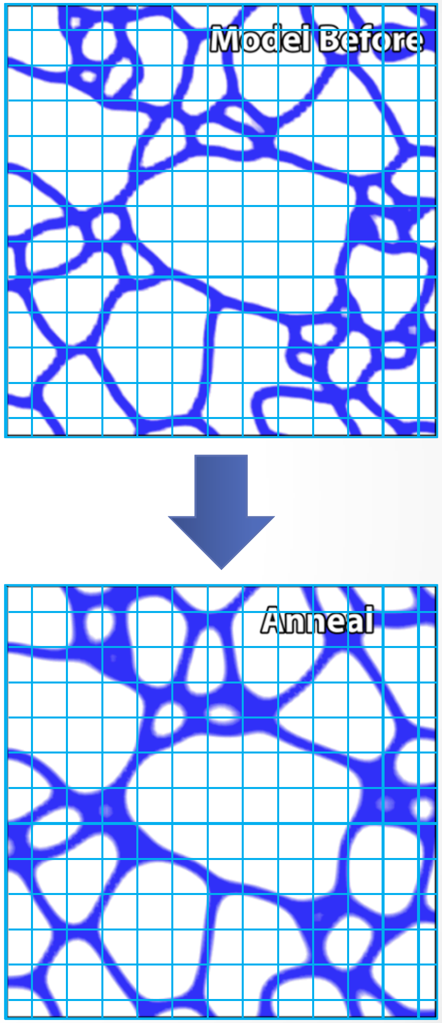
- Automated diffraction orientation mapping
  - Point by point grid of orientations mapped
  - 5 nm resolution
- Analogous to EBSD



Point diffraction data

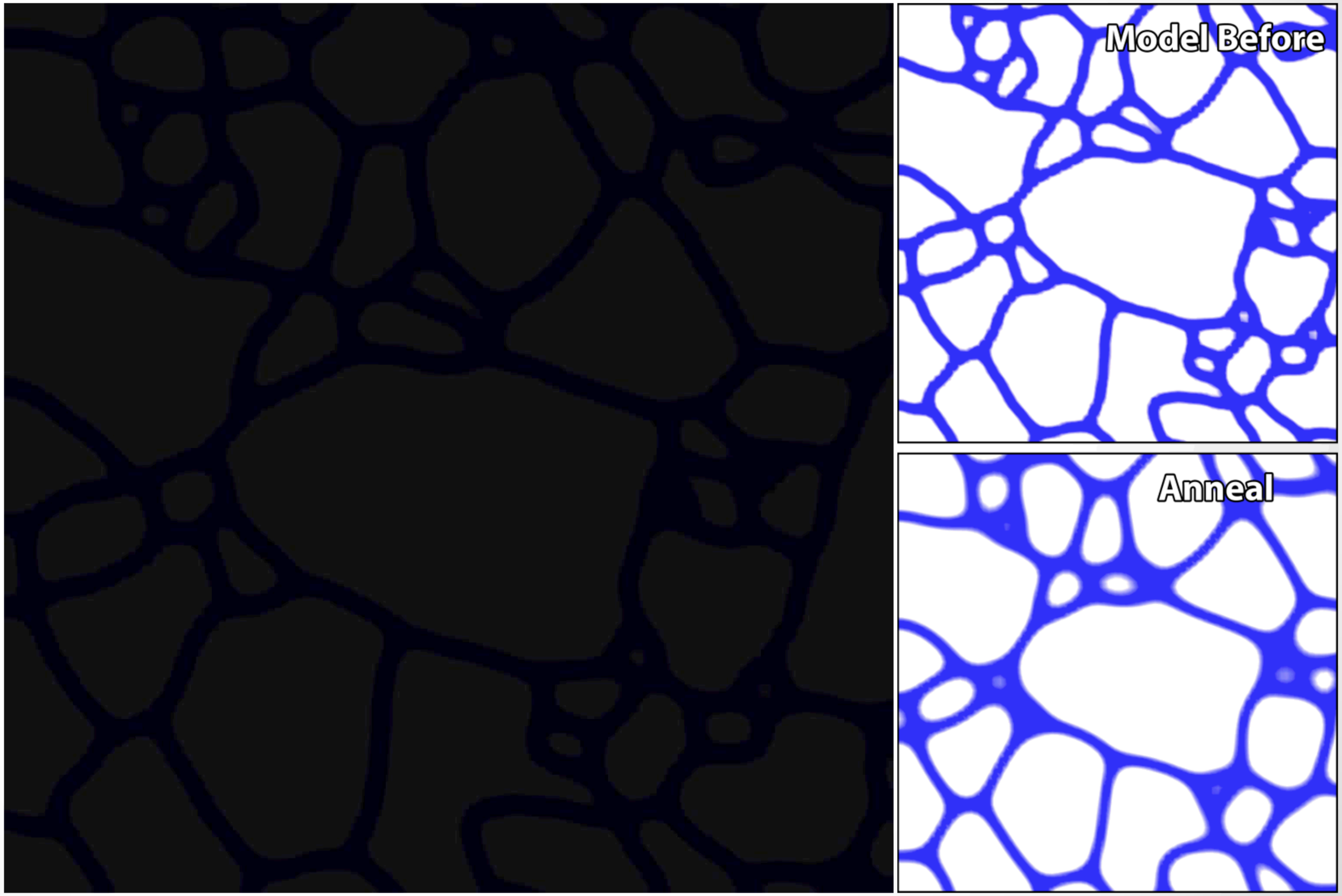
Approach: Modeling

- What is phase field modeling?
  - Mathematical model for solving interfacial problems, like solidification, growth, etc.
- Example grain growth model
  - Thermodynamic free energy function
    - $dF = d(\gamma A) = \gamma dA$ (γ: GB energy, A: GB area)
  - Model for kinetics
    - $V = M\gamma h$ (M: GB mobility, h: GB curvature)
  - Solve at each pixel for a predetermined timestep
- See Abdeljawad and Foiles, Acta Mater, 2015 for more information



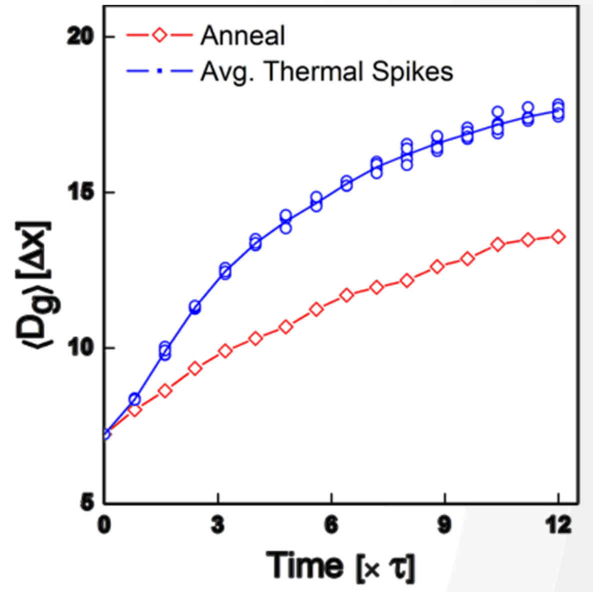
Can directly use experimental maps as input structures, and then compare evolutions!

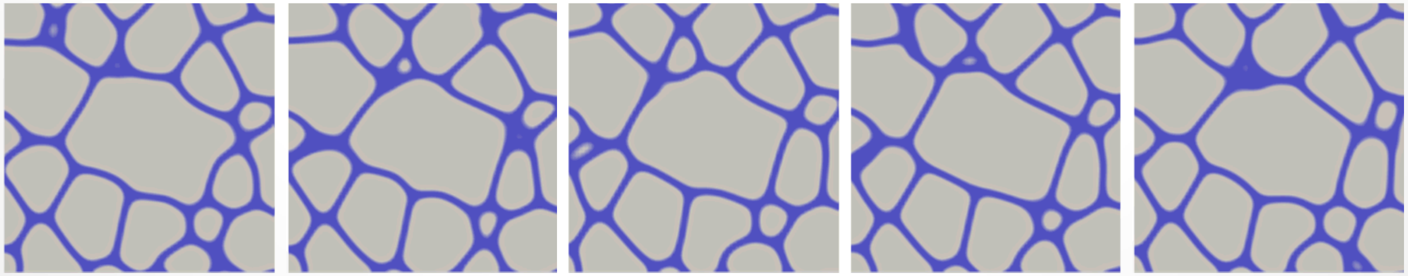
Simulated Anneal



Model Data Analysis

- During simulated annealing grain growth scales approximately with $T^{1/2}$
  - Expected for homogenous grain growth
- During simulated irradiation, grain growth scales with $T^{1/n}$, where $n \approx 3$
  - Initially faster, but stagnates sooner





Time scaling

Fraction of ion strikes that intersect grain boundaries (thus contributing to grain growth):

$$f_{\mathrm{GB}} = \frac{\pi D^2 \frac{d_{\mathrm{spike}}}{2}}{\frac{\pi D^3}{6}} = \frac{3d_{\mathrm{spike}}}{D},$$

- Incorporation of this *D* term leads to scaling proportional to $t^{(1/3)}$.
- Consistent with experimental observations.

$$\frac{dD}{dt} = \frac{\Phi\chi\delta}{N_{\mathrm{at}}}\frac{3d_{\mathrm{cas}}}{D}\left[\frac{4\gamma V_{\mathrm{at}} N_{\mathrm{at}} \nu}{D k_B}\frac{\sqrt{\frac{3}{5}}\Gamma\left(\frac{8}{3}\right)k_B^{8/3}}{10\pi C_0^{2/3}\kappa_0}\frac{Q^{5/3}}{E_a^{8/3}}\right].$$

$$D^2 dD = \left[12\gamma d_{\mathrm{spike}}\Phi\chi\delta\frac{V_{\mathrm{at}}\nu\sqrt{\frac{3}{5}}\Gamma\left(\frac{8}{3}\right)k_B^{5/3}}{10\pi C_0^{2/3}\kappa_0}\frac{Q^{5/3}}{E_a^{8/3}}\right]dt.$$

$$D^3 - D_0^3 = \left[36\gamma d_{\mathrm{spikes}}\chi\delta\frac{V_{\mathrm{at}}\nu\sqrt{\frac{3}{5}}\Gamma\left(\frac{8}{3}\right)k_B^{5/3}}{10\pi C_0^{2/3}\kappa_0}\frac{Q^{5/3}}{E_a^{8/3}}\right]\Phi t$$

$$= K\Phi t.$$

Kaoumi, *et al*, J Appl Phys, 2008.