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14. ABSTRACT

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RPPR Final Report
as of 01-Apr-2022

Agency Code: 21XD

Proposal Number: 72834MSYIP **Agreement Number: W911NF-18-1-0162**

INVESTIGATOR(S):

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Submitted By: Ashwin Shahani Email: shahani@umich.edu
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Major Goals: The long-term objective of this project is to understand the interaction between grain boundaries and second phase particles during abnormal grain growth (AGG). AGG is often seen in multi-phase materials, yet its origins and mechanisms have remained an enigma. For instance, what are the microstructural markers associated with particle-assisted AGG? How do the boundaries of the abnormal grain propagate through the particle cloud? What are the most significant driving forces associated with grain growth in the presence of particles? Resolving these open questions requires nondestructive imaging in order to follow the evolution of grain boundaries upon annealing. In particular, Shahani's team has integrated two nondestructive imaging modalities — absorption-based and diffraction-based X-ray computed tomography on a laboratory microscope — to capture the full microstructural details in real-time. From the three-dimensional tomographic (3D) data, they extract grain boundary characteristics, particle locations, and their correlations. The insights gained can be used by researchers to design polycrystalline materials with targeted grain size and shape distributions.

Accomplishments: Year 1: A critical need existed to firstly devise the infrastructure for processing high-dimensional and multimodal datasets collected through X-ray tomographic microscopy. To this end, in year one of this project, Shahani and his team have developed a one-of-a-kind function package (PolyProc, https://github.com/shahaniRG/PolyProc) that is compatible with a range of data shapes, from planar sections to time-evolving and 3D orientation data. All data is stored in a MATLAB structure format which provides better organization and versatility, increases computational efficiency, and eases retrieving saved data. The package comprises functions to import, filter, analyze, and visualize the reconstructed grain maps that are so essential to understand the progression of grain growth. To accelerate the computations in the data processing workflow, a number of computationally efficient approaches are utilized, e.g., genetic and combinatorial optimization. Details on its content, implementation, and performance are given in a publication to Integr. Mater. Manuf. Innov. in the 3D Materials Science thematic section (2019). It is anticipated that the function package will become the de facto route by which investigators process their multi-modal and multi-dimensional datasets.

Leveraging this toolbox, Shahani and his team conducted X-ray imaging experiments in year one of this project to characterize the grain structure evolution and particle distribution in an Al-3wt%Cu alloy as a model system. This work was done in early 2019 on a laboratory X-ray microscope at the University of Michigan. The team observed via diffraction-contrast tomography AGG after prolonged annealing. Meanwhile, the distribution of Al_2Cu particles is highly non-uniform, as determined via absorption-contrast tomography. By connecting the two datasets, Shahani and his team determined that abnormally large grains are found in regions with low particle density. That is, a non-uniform distribution of particles can trigger AGG even in initially normal grain structures. Once the abnormal grain develops its size advantage, it parasitically takes over the microstructure owing to a size advantage over the normal grains.

To better understand the experimental observations, Shahani's team has developed an analytical model that takes into account the competing capillary and particle pinning pressures. More generally, the model sheds light on why the abnormal grain should run away from the grain size distribution. It can be used to predict under what conditions (e.g., grain size advantage) AGG will occur, which is of scientific interest and technological relevance to those in the field of metals processing. A manuscript on this combined experimental-theoretical work was published in Acta Mater. (2020).

Year 2: The above study left several questions unanswered, grounds for exploration in year two of the project: For instance, how and why does the particle distribution become non-random, even after homogenization? Secondly, what is the critical degree of non-randomness in the particle distribution above which abnormal grain growth is triggered? To gain some preliminary clues, Shahani's team characterized the particle evolution and grain growth behavior in an Al-Cu alloy as a function of annealing time and temperature (below the solvus), via SEM. They observed a transition of the particle distribution from random to non-random and a concomitant transformation from normal to abnormal grain growth with increasing annealing temperature and/or increasing annealing time. For quantitative analysis, they codified the degree of inhomogeneity in the particle distribution through various statistical metrics and linked them to the critical grain size necessary for AGG to take over. With this information, the team constructed a tentative temperature-time- (structural) transformation (TTT) diagram that identifies the onset of abnormal grain growth. By doing so, they draw an important parallel between structural transformations (grain growth) and phase transformations (nucleation), in a way that has never been done before. This work was published in Metall. Mater. Trans. A (2020).

Year 3: Due to the COVID-19 pandemic, Shahani and his team were unable to visit the synchrotron in year three to conduct higher resolution experiments on the interaction mechanism between particles and grain boundaries. Instead, they shifted their focus to answering other fundamental scientific questions related to the proposed effort. Notably, the team has visualized in 3D the connectivity of the grain boundary network, which is thought to play a dominant role in not only grain growth, but also liquid metal embrittlement (LME) and intergranular damage propagation. This effort was made possible thanks to the multimodal X-ray imaging platform (developed by Shahani's team in the first year of the ARO award) that enables precise characterization of microstructures that are simultaneously polyphase and polycrystalline. Fortunately, they were able to use the experimental facilities at the University of Michigan for this purpose. The wealth of data collected via 3D X-ray microscopy enabled Shahani's team to test directly the validity and applicability of geometric percolation theory, as they report on in their publication in Acta Mater. (2021). The result of this paper not only improves understanding of (i) the structure of grain boundary networks, (ii) the dynamics of LME, and (iii) the correlations between the two, but also provides insight into (iv) the design of LME-resistant materials via grain boundary engineering.

Inspired by these most recent results, Shahani and his team have quantified the connectivity of the grain boundary network more rigorously, at the end of year three. Specifically, they have measured the percolation threshold of high-angle random grain boundaries and compared to recent simulations (e.g., idealized models by Schuh and others). Whereas such models typically assume a constant topology of the grains (i.e., 14 nearest grain neighbors), real microstructures are much more complex (i.e., there exist a broad range of grain neighbors). To test the validity of models, the team has used their existing X-ray imaging data stockpile as input. The calculations of percolation thresholds from the experiments agrees very well with those from the simulations in the literature, provided that the thresholds are normalized by the number of grain neighbors. This work is currently in preparation for submission to Acta Mater. (2022).

Training Opportunities: Direct educational benefits include (1) the training of graduate students in state-of-the-art characterization techniques (e.g., X-ray tomographic microscopy). Furthermore, owing to the large datasets collected through these experiments, the students are (2) actively learning data science methods (e.g., machine learning) to uncover hidden patterns in their data. Expertise in data science is crucial to their success in this project and beyond. To this end, the students have participated in professional development activities, such as the workshop on "Machine Learning in X-ray Imaging and Microscopy Applications" held virtually in summer 2020, where they interfaced with leaders in the field. Last but not least, the students are (3) learning topics fundamental to microstructure evolution, such as thermomechanical processing and grain growth.

Results Dissemination: Publications: A description of the function package developed by Shahani's group has been published in Integr. Mater. Manuf. Innov. (2018), https://doi.org/10.1007/s40192-019-00147-2. Its inclusion in the "3D Materials Science" thematic section will ensure that it reaches a wide audience in the field of multidimensional imaging. The results from the in situ experiments enabled by this function package have been published (2020) in Acta Mater. (impact factor of 8.2), http://10.1016/j.actamat.2020.04.049. The reviewers commented that the manuscript "has the depth and originality to be an important paper on a long-standing puzzle, even though it leaves plenty of room for additional work. The combination of recently developed 3D characterization methods is also significant." Shahani and his team have a third paper that follows on the heels of this work, published in Metall. Mater. Trans. A (2020), http://doi.org/10.1007/s11661-020-06125-0. The last published paper concerns the connectivity of grain boundary networks in 3D, see Acta. Mater. (2021), http://doi.org/10.1016/j.actamat.2021.117145.

Presentations: Shahani was the lead organizer of two symposia: "Leveraging 3D Imaging and Analysis for New Opportunities in Materials Science" at M&M (2019) as well as "In Situ Characterization of Dynamic Phenomena during Materials Synthesis" at MRS (2019). In both of these symposia, his own students presented on ARO-supported research: For instance, PhD student Jiwoong Kang gave a talk on "The Dynamics of Abnormal Grain Growth in a Particle-Containing System — New Insights from Multimodal Three-Dimensional X-ray Imaging" at MRS (2019). Furthermore, post-doc Dr. Ning Lu from the Shahani group presented the same work at M&M (2019). The MRS symposium garnered international attention and was well-received by the materials community, culminating in Shahani serving as a guest co-editor (with Amy Clarke) for a thematic issue titled “Processing Metallic Materials far from Equilibrium” for MRS Bull. (impact factor of 6.6). The issue was published in November 2020. Beyond these two conferences, Shahani has presented the ARO-funded research at the REX&GG meeting in summer 2019 and Jiwoong Kang at TMS in 2021.

Invited seminars: Shahani has given five invited seminar talks based on ARO YIP-funded research, and specifically on his team’s in situ observations of colossal grain growth stimulated by particles. These were organized by SIAM MS21 (2021), Cornell University (2021), Department of Defense Basic Research Forum (2020), Zeiss on your Campus Webinar Series (2020), and Ohio State University (2019). Some of these presentations were delivered virtually owing to the COVID-19 pandemic.

Workshops: Shahani's team was invited to lead a virtual workshop at Brookhaven National Laboratory's National Synchrotron Light Source II, through the Users' Meeting (2020). The goal was to share our codes with the synchrotron community, thereby opening the 'black box' of diffraction-contrast X-ray tomography. Over 100 took part.

Honors and Awards: In 2021, Shahani received the "Outstanding Faculty Achievement Award" from the Department of Materials Science and Engineering at the University of Michigan. The award is the top honor for tenured and tenure-track faculty members in a given academic year. It recognizes "stellar performance in materials research, teaching, and service to the department." In addition, PhD student Jiwoong Kang won the 2nd place prize in the "Best Student Oral Presentation Competition" at TMS 2021. His talk is based on ARO-funded research and is titled, "Dynamics of Abnormal Grain Growth in a Particle-Containing System uncovered by Multimodal Three-dimensional X-ray Imaging." Jiwoong was also selected for the highly prestigious and selective “National School on Neutron and X-ray Scattering,” which was held remotely in 2020. This was a unique opportunity afforded only to the top 5% of PhD applicants.

Protocol Activity Status:

Technology Transfer: Improved understanding of the conditions in which abnormal grain growth is favorable may lead to new routes to fabricate single crystals, all in the solid-state.

PARTICIPANTS:

Participant Type: PD/PI
Participant: Ashwin J Shahani
Person Months Worked: 2.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Ning Lu
Person Months Worked: 15.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Jiwoong Kang
Person Months Worked: 13.00 **Funding Support:**
Project Contribution:
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Participant Type: Undergraduate Student
Participant: Isaac Loo
Person Months Worked: 2.00 **Funding Support:**
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Article Title: PolyProc: A Modular Processing Pipeline for X-ray Diffraction Tomography
Authors: Jiwoong Kang, Ning Lu, Issac Loo, Nancy Senabulya, Ashwin J. Shahani
Keywords: 3D data processing, X-ray diffraction contrast tomography, grain mapping, microstructure evolution
Abstract: Direct imaging of three-dimensional microstructure via X-ray diffraction-based techniques gives valuable insight into the crystallographic features that influence materials properties and performance. For instance, X-ray diffraction tomography provides information on grain orientation, position, size, and shape in a bulk specimen. As such techniques become more accessible to researchers, demands are placed on processing the datasets that are inherently “noisy,” multi-dimensional, and multimodal. To fulfill this need, we have developed a one-of-a-kind function package, PolyProc, that is compatible with a range of data shapes, from planar sections to time-evolving and three-dimensional orientation data. Our package comprises functions to import, filter, analyze, and visualize the reconstructed grain maps. To accelerate the computations in our pipeline, we harness computationally efficient approaches: for instance, data alignment is done via genetic optimization; grain tracking ...
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Acknowledged Federal Support: **Y**

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Volume: 195 Issue: First Page #: 1
Date Submitted: 7/29/21 12:00AM Date Published:
Publication Location:
Article Title: Dynamics of particle-assisted abnormal grain growth revealed through integrated three-dimensional microanalysis
Authors: Ning Lu, Jiwoong Kang, Nancy Senabulya, Ron Keinan, Nicolas Gueninchault, Ashwin J. Shahani
Keywords: grain growth, abnormal grain growth, Smith-Zener pinning, 3D X-ray microscopy, X-ray diffraction-contrast tomography, laboratory X-ray microscopy
Abstract: Secondary-phase particles are routinely dispersed in metals and ceramics to prevent grain growth and take full advantage of the small grain size in the mechanical properties of polycrystals. Somewhat surprisingly, the preferential or abnormal growth of a few grains is observed in particle-containing systems at relatively high temperature, which will limit the lifetime of the material. The origins and mechanisms of particle-assisted abnormal grain growth are widely contested. Here, we employ integrated three-dimensional X-ray imaging to throw new light on the complex interactions between grain boundaries and particles in an Al-Cu alloy as a model system. We observe abnormal grain growth in the presence of a highly non-random distribution of particles. The incipient grain size is set by the local distribution of particles such that the larger grains come from particle-poor regions. Subsequently, grains with a size advantage may "run away" from the grain size distribution, in agreement...
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Acknowledged Federal Support: **Y**

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Metallurgical and Materials Transactions A
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Date Submitted: 1/27/21 12:00AM Date Published: 1/1/21 5:00AM
Publication Location:
Article Title: Origins of Non-random Particle Distributions and Implications to Abnormal Grain Growth in an Al-3.5 Wt Pct Cu Alloy
Authors: Ning Lu, Jiwoong Kang, Ashwin J. Shahani
Keywords: Grain growth, abnormal grain growth, particles, particle distribution
Abstract: The mechanisms of abnormal grain growth (AGG) in particle-containing systems have long been a mystery. Recently, we reported that a non-random particle distribution can induce a grain size advantage and trigger AGG. However, the processing conditions leading to a non-random particle distribution are far from being understood. Here, we investigate the particle distribution and concomitant grain growth behavior at different annealing temperatures and times in an Al-3.5 wt pct Cu alloy by scanning electron microscopy (SEM). At high temperatures and long times, the particle distribution evolves from random to non-random, with an accompanying transition from normal grain growth (NGG) to AGG. Analytical calculations suggest that a non-random particle distribution is introduced by residual Cu segregation even after homogenization. In short, the corresponding fluctuation of ?-Al2Cu phase distribution is amplified at elevated temperatures via particle dissolution. We quantify the spatial inhom
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Acknowledged Federal Support: **Y**

RPPR Final Report
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Article Title: Dynamics of Ga penetration in textured Al polycrystal revealed through multimodal three-dimensional analysis
Authors: N. Lu, S. Moniri, M.R. Wiltse, J. Spielman, N. Senabulya, A.J. Shahani
Keywords: Liquid metal embrittlement; Three-dimensional characterization; X-ray diffraction tomography; Strong texture; Grain boundary diffusion; Percolation theory
Abstract: We investigate in situ the penetration of liquid Ga in a polycrystalline Al rod at a temperature of 31.9°C. Our study is made possible with the aid of a novel, multimodal laboratory-based X-ray imaging platform, which enables us to characterize the three-dimensional (3D) grain structure, Ga wetting behavior, and correlations of the two. Our sample shows a strong texture along [001] and a higher fraction of low-angle grain boundaries (LAGBs) compared to a random distribution, signifying a poor connectivity of high-angle grain boundaries (HAGBs). We detect channels of Ga along HAGBs after 1-hour of exposure to the liquid metal, but such channels gradually disappear upon prolonged annealing. We interpret this phenomenon in the lens of geometric percolation theory and diffusion theory, which point to the synergistic effects of a limited HAGB connectivity and the enhanced Ga leakage from HAGBs into the bulk through dislocation-pipe diffusion. This work provides detailed insight into the ...
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Acknowledged Federal Support: **Y**

Partners

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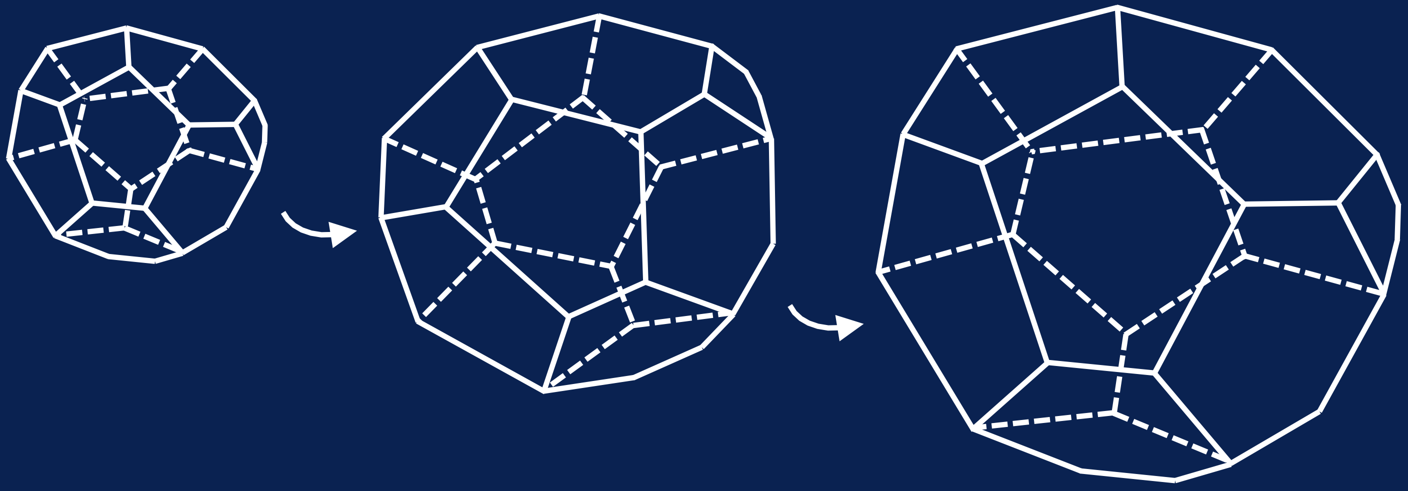
I certify that the information in the report is complete and accurate:
Signature: Ashwin J Shahani
Signature Date: 3/30/22 3:29PM



Revealing new modes of grain growth at elevated temperatures *via* 3D x-ray microscopy

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Assistant Professor
Materials Science and Engineering
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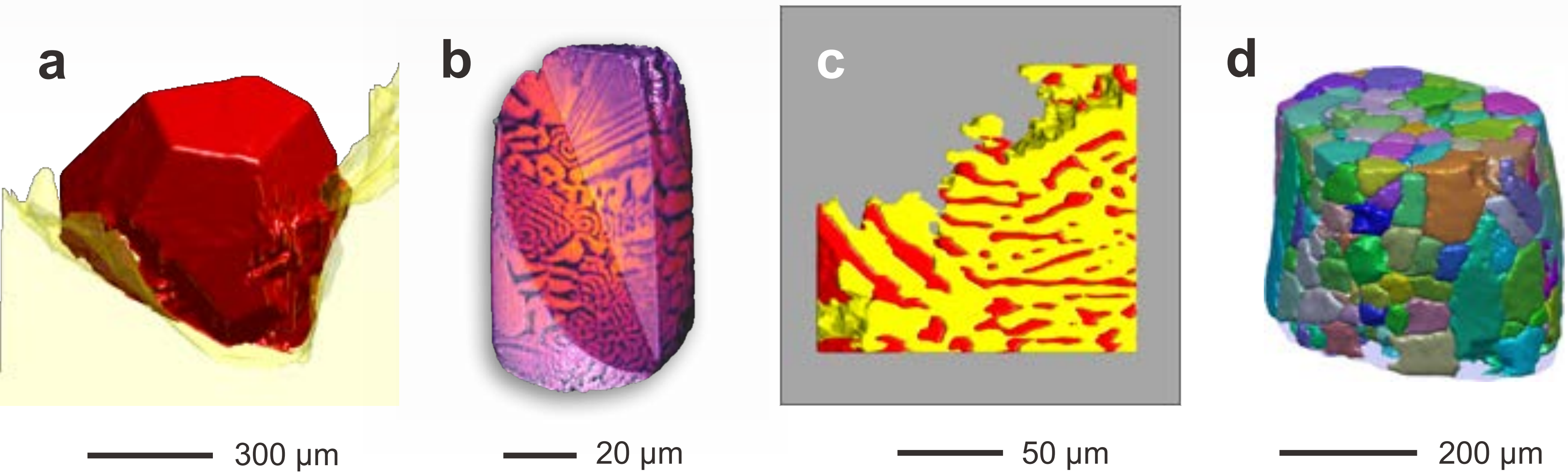
ARO YIP award no. W911NF-18-1-0162

Shahani group @ UM

- **Research focus:** Understand solidification and processing pathways of materials through real-time and 3D imaging
- Current interests span quasicrystals to eutectics to polycrystals



Quasicrystals **Eutectics** **Polycrystals**

a N. Senabulya et al., *Scripta Mater.* **146**, 218 (2018).
b (cover) S. Moniri et al., *Small* **16**, 1906146 (2020).
c S. Moniri et al., *Sci. Rep.* **9**, 3381 (2019).
d N. Lu et al., *Acta Mater.* **195**, 1 (2020).

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- **Army Research Office**
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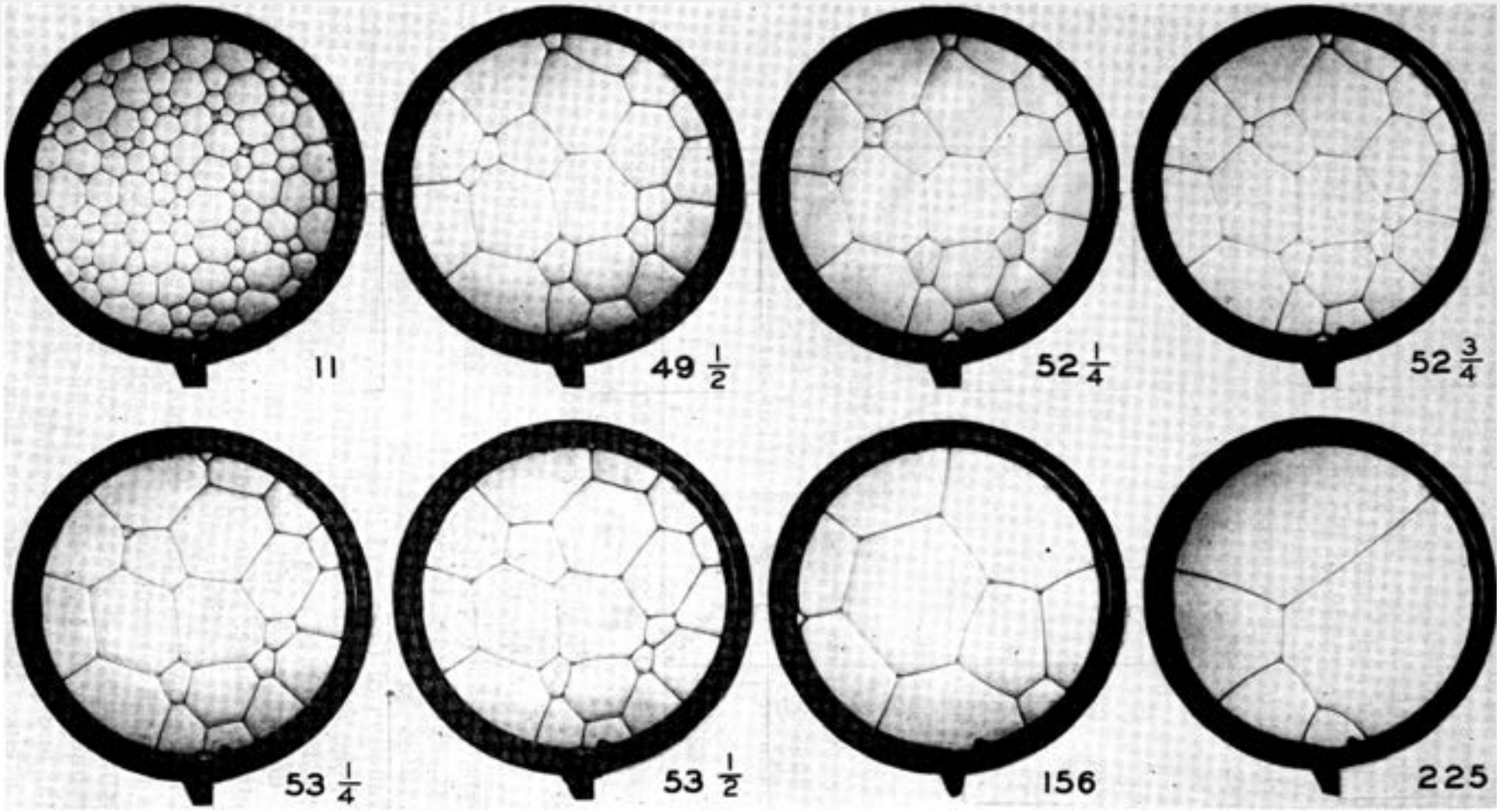
Agenda

- **Background**
  - Normal *vs.* abnormal grain growth
  - Driving forces for grain boundary motion
- **Approach**
  - X-ray diffraction tomography
  - X-ray absorption tomography
  - Integrating the two imaging modalities
- **Results and discussion**
  - Abnormal grain selection and growth
  - Grain migrations driven by strain energy

R. Keinan, et al., A.J. Shahani, *Acta Mater.* **148**, 225 (2018).
J. Kang, et al., A.J. Shahani, *Integr. Mater. Manuf. Innov.* **8**, 388 (2019).
N. Lu, et al., A.J. Shahani, *Acta Mater.* **195**, 1 (2020).
N. Lu, et al., A.J. Shahani, *Metall. Mater. Trans. A* **52**, 914 (2021).

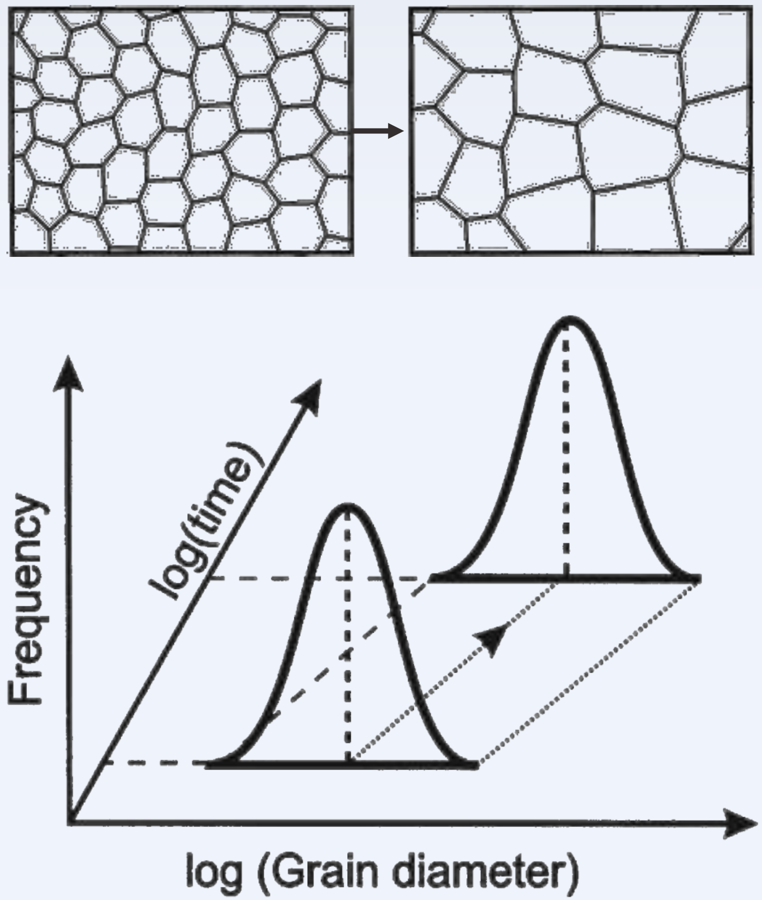
Soap bubbles: a proxy for granular materials



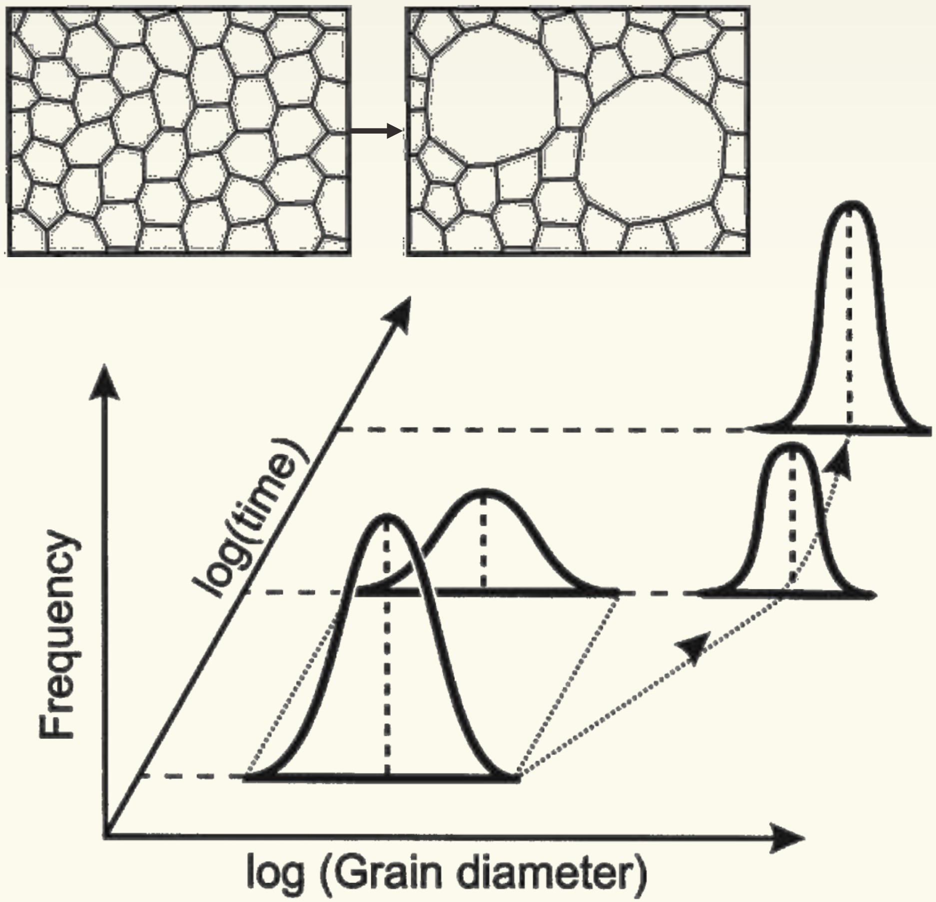
C.S. Smith, in *Metal Interfaces*, 65 (1952).

Two routes for grain growth

Normal (continuous)



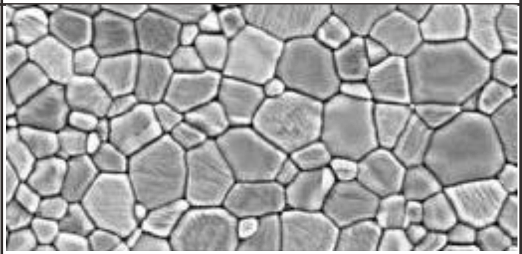
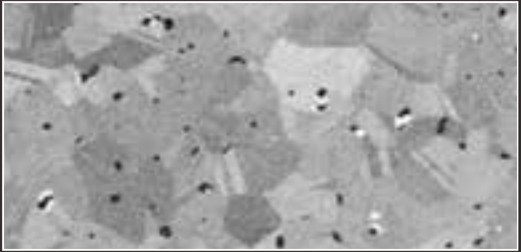
Abnormal (discontinuous)



F.J. Humphreys and M. Hatherly, Pergamon, 2nd ed. (1996).

Why do grains grow?

G. Gottstein and L.S. Shvindlerman, *Grain boundary migration in metals* (2010).
J. von Neumann, in C. Herring, ed., *Metal Interfaces*,108-110 (1952).
W.W. Mullins, *J. Appl. Phys.* **27**, 900-904 (1956).
R.D. MacPherson and D.J. Srolovitz, *Nature* **446**, 1053-1055 (2007).
C. Zener, quoted by C.S. Smith, *Trans. AIME* **175**, 15-51 (1948).

Constitutive model	Microstructure	Driving force
Recrystallization Growth driven by lattice defect energy		$\frac{1}{2}\rho G b^2$
von Neumann-Mullins Growth driven by capillarity in 2D		$\frac{\pi}{3}\gamma(n-6)$
MacPherson-Srolovitz Growth driven by capillarity in 3D		$\frac{\pi}{3}\gamma\left(\sum_{i=1}^{n} e_i(\mathbf{D}) - 6\mathcal{L}(\mathbf{D})\right)$
Zener-Smith Retardation by second-phase particles		$-\frac{3f\gamma}{2r}$

Limiting grain size

- The point at which grain growth stops is (approximately) determined by a balance of the capillary driving force for growth and the particle drag force

$$\frac{3f\gamma}{2r} \approx \frac{\gamma}{R_0} \qquad \rightarrow R_0 = \frac{2r}{3f}$$

- **The effect of the particles is to slow down and eventually stop further grain growth**

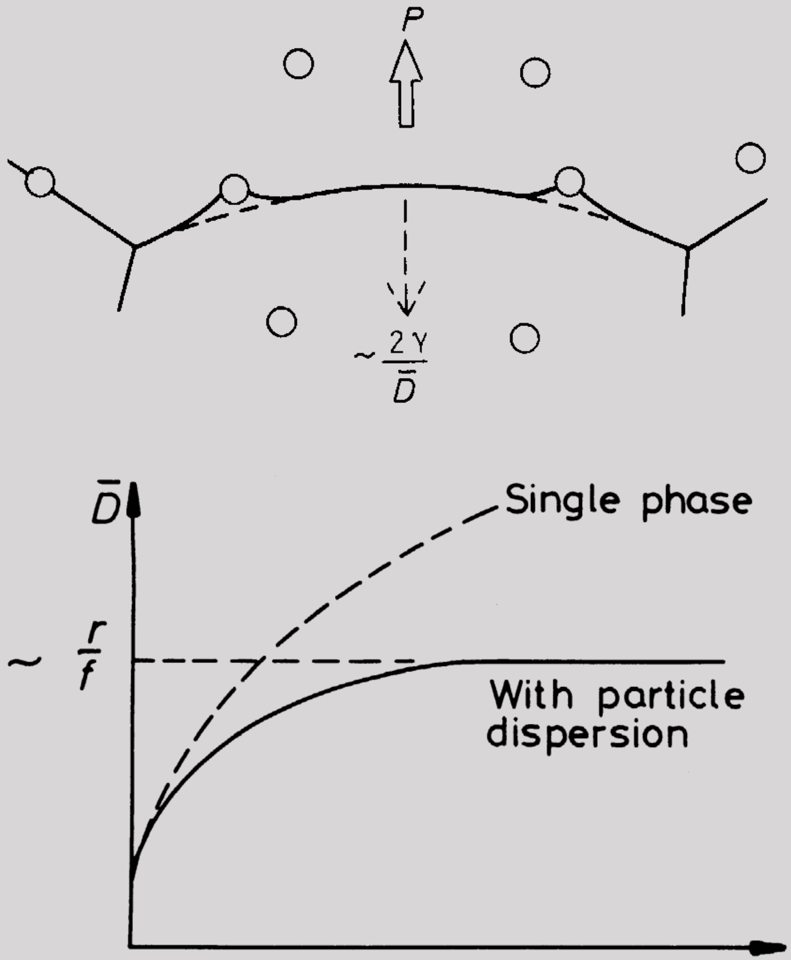
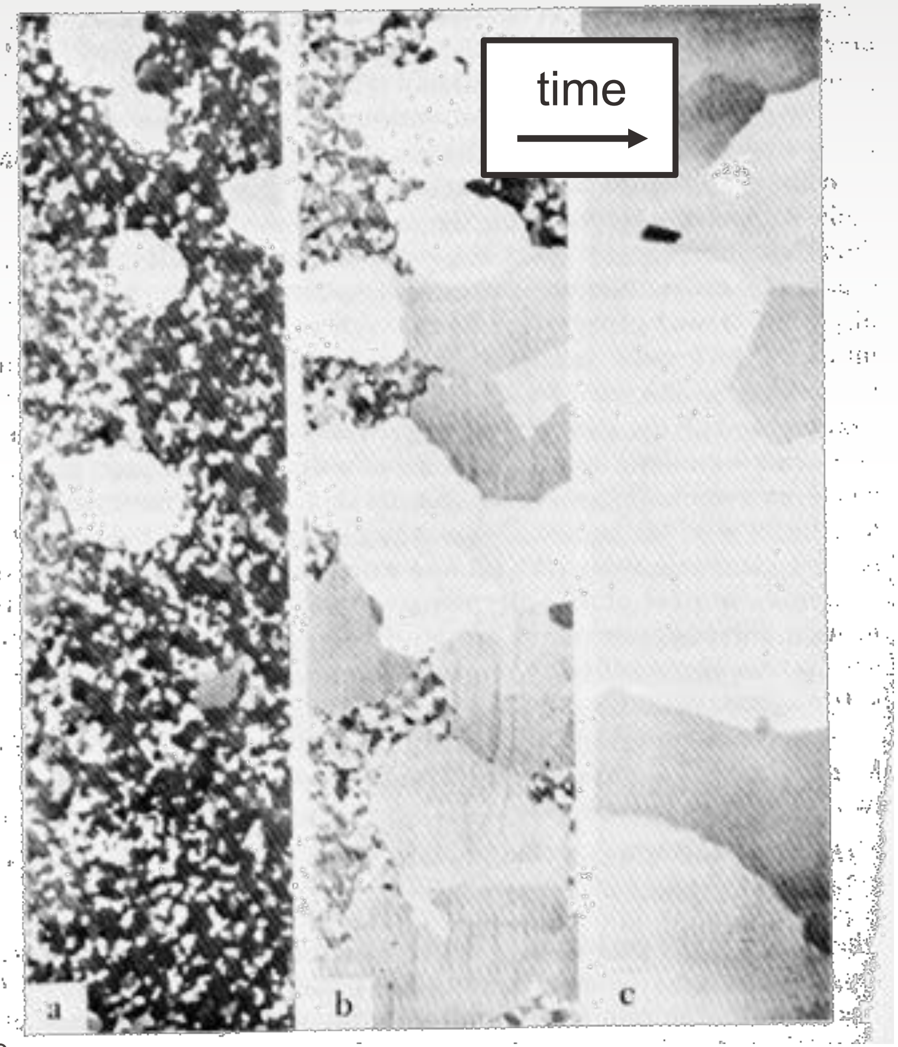


Fig. 3.31 Effect of second-phase particles on grain growth.

D.A. Porter, K.E. Easterling, and M.Y. Sherif, CRC Press, 3rd ed. (2009).

Abnormal growth in transformer steels

- AGG dramatically obvious in Fe-5wt%Si steels
- {110}<001> “Goss” grains are much larger than matrix grains (speckled regions)
- Alloy contains MnS and AlN particles (not pictured), which may pin matrix grains
- **Where does AGG initiate and how does it persist to consume microstructure?**

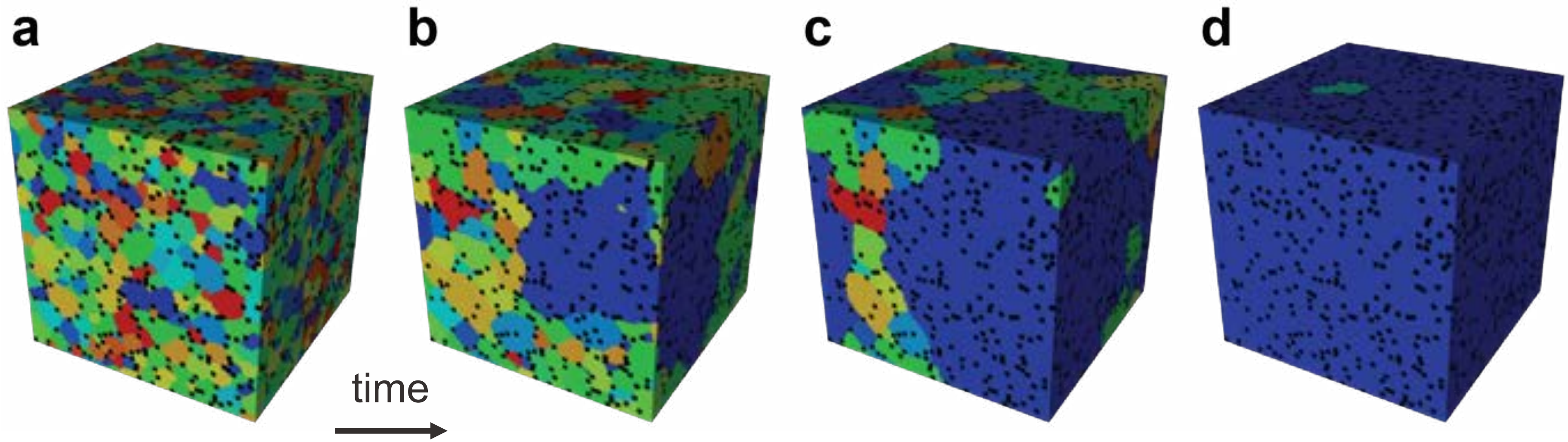


N.P. Goss, *Trans. Met. Soc. AIME* **23**, 511-544 (1935).
C.D. Graham, Jr., Academic, Chap. XV (1969).

Insights from MCP simulations

- **Initial condition:** isotropic boundaries, random particle locations
- Blue grain thermally fluctuates from its pinned configuration (**b**)
- Free boundaries then grow at expense of neighboring grains (**c**)

Our goal: harness ***in situ*** **3D imaging** to verify this model



E.A. Holm, T.D. Hoffmann, A.D. Rollett, and C.G. Roberts, in S. Fæster et al., eds., *Proc. 36th Risø Intl. Symp. Materials Sci.* **89**, 012005 (2015).

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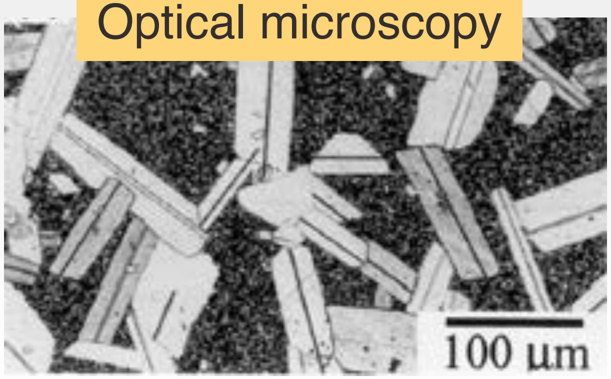
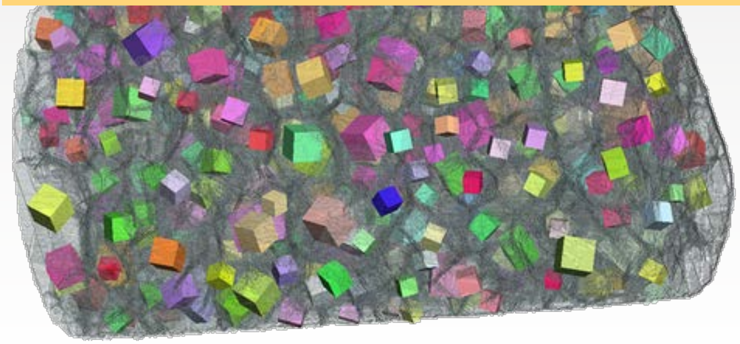
N. Lu, et al., A.J. Shahani, *Metall. Mater. Trans. A* **52**, 914 (2021).

Approaches for orientation mapping

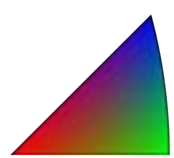
Optical microscopy



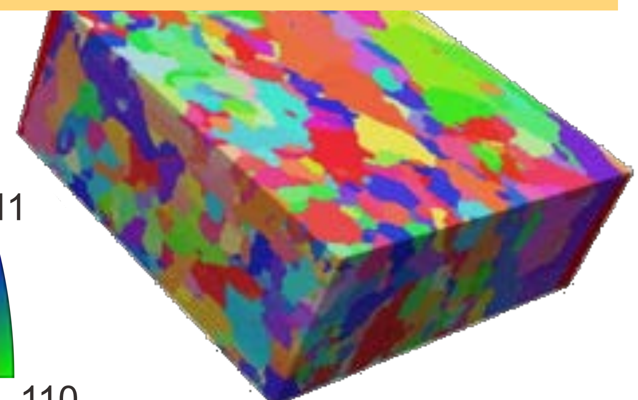
Diffraction contrast tomography



Electron backscatter diffraction



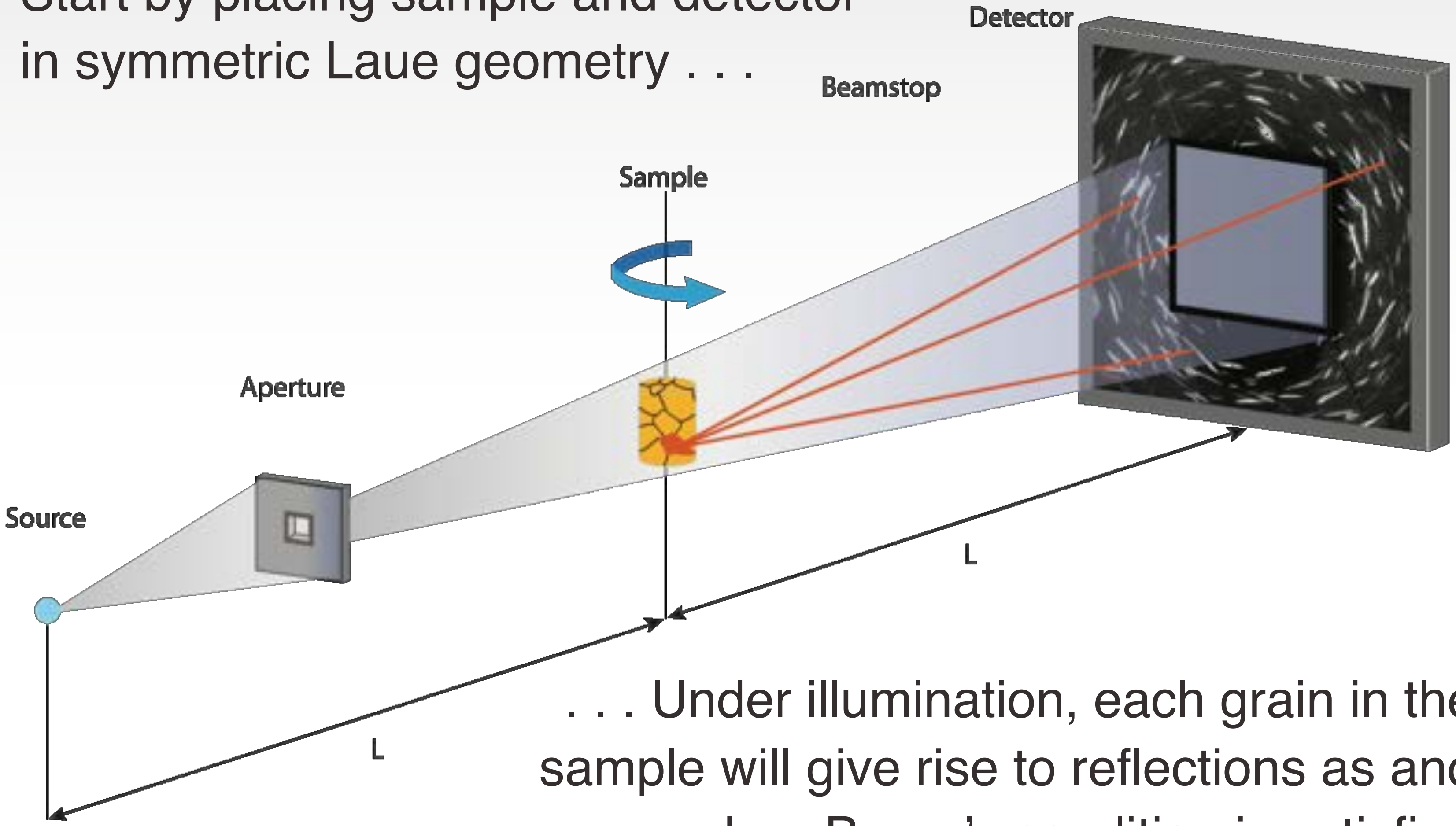
EBSD + focused ion beam



B.-K. Park et al., *J. Am. Ceram. Soc.* **84**, 2707 (2001).
H. Beladi and G. S. Rohrer, *Acta Mater.* **61**, 1404 (2013).
S.A McDonald et al., *Sci. Rep.* **5**, 14665 (2015).

LabDCT: how does it work?

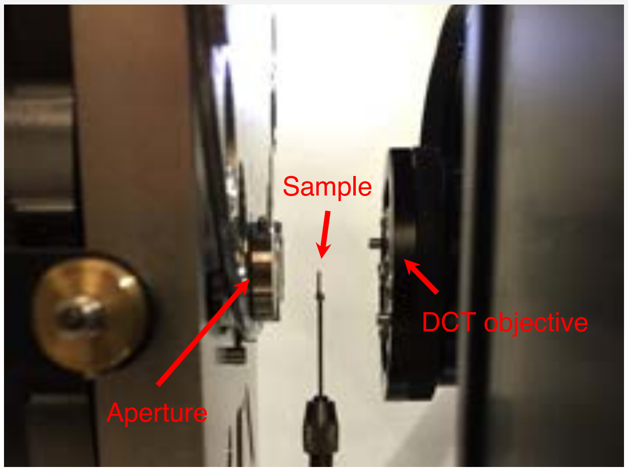
Start by placing sample and detector in symmetric Laue geometry . . .

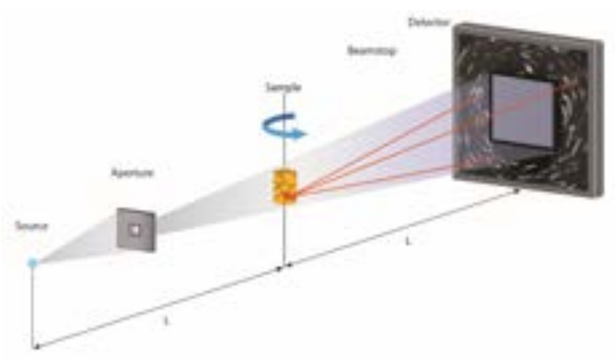


. . . Under illumination, each grain in the sample will give rise to reflections as and when Bragg's condition is satisfied

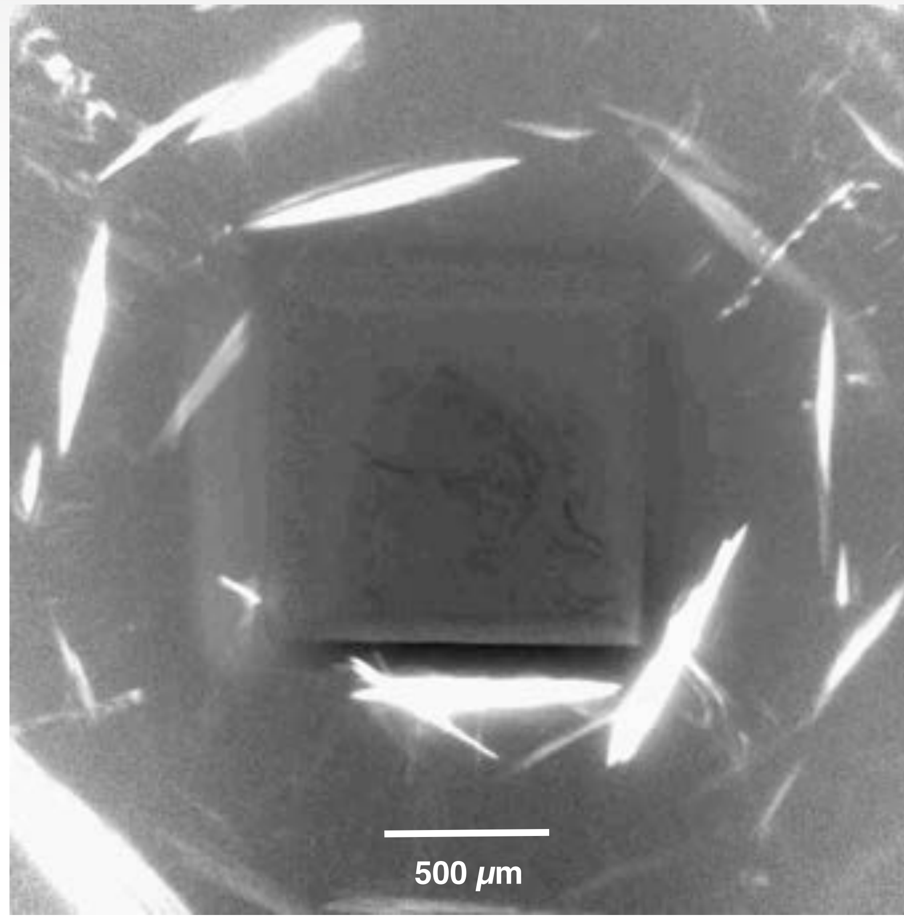
LabDCT in action at $(MC)^2$ @ UM

Experimental setup





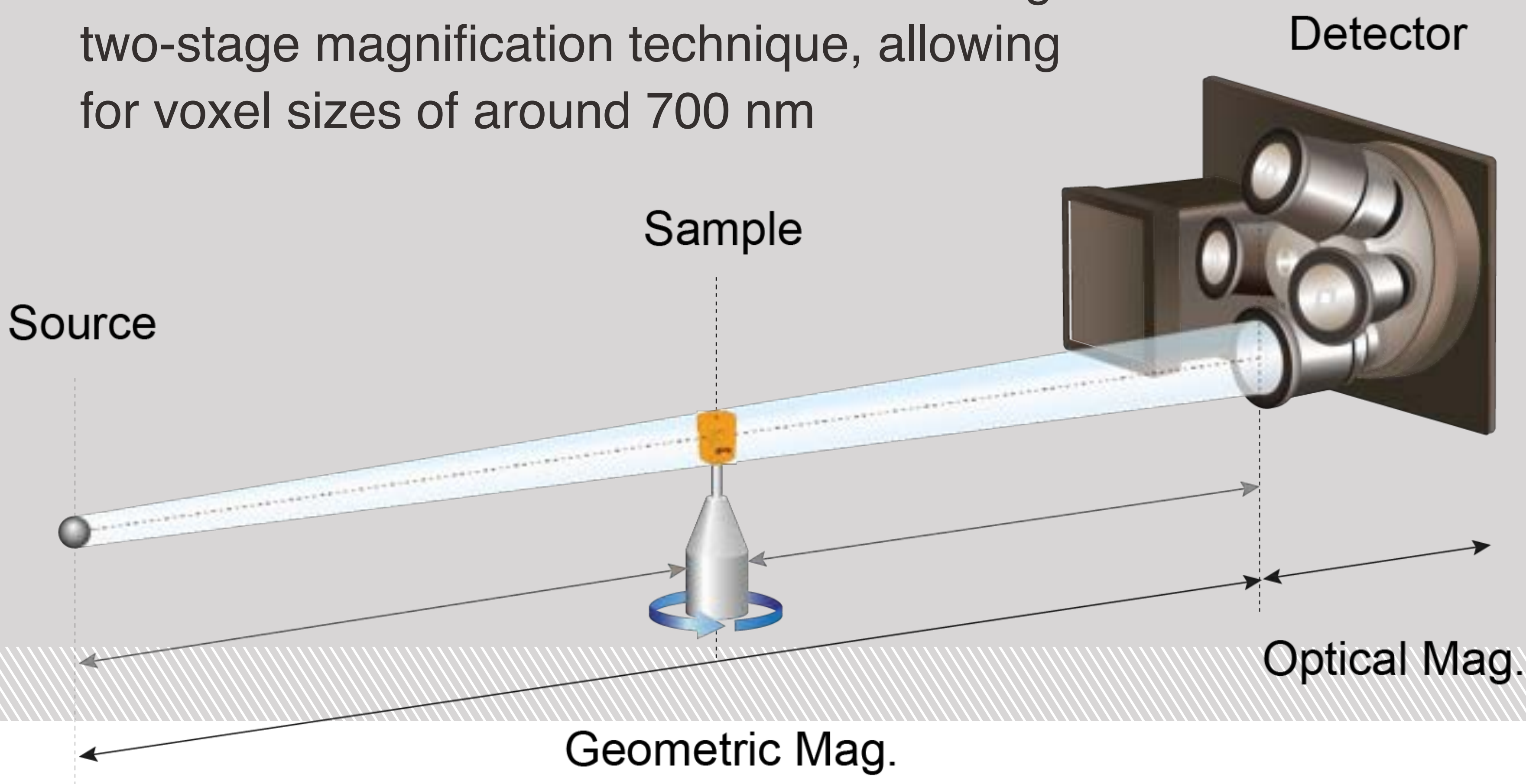
Output diffraction pattern



Other imaging modalities of interest

Absorption-contrast X-ray tomography

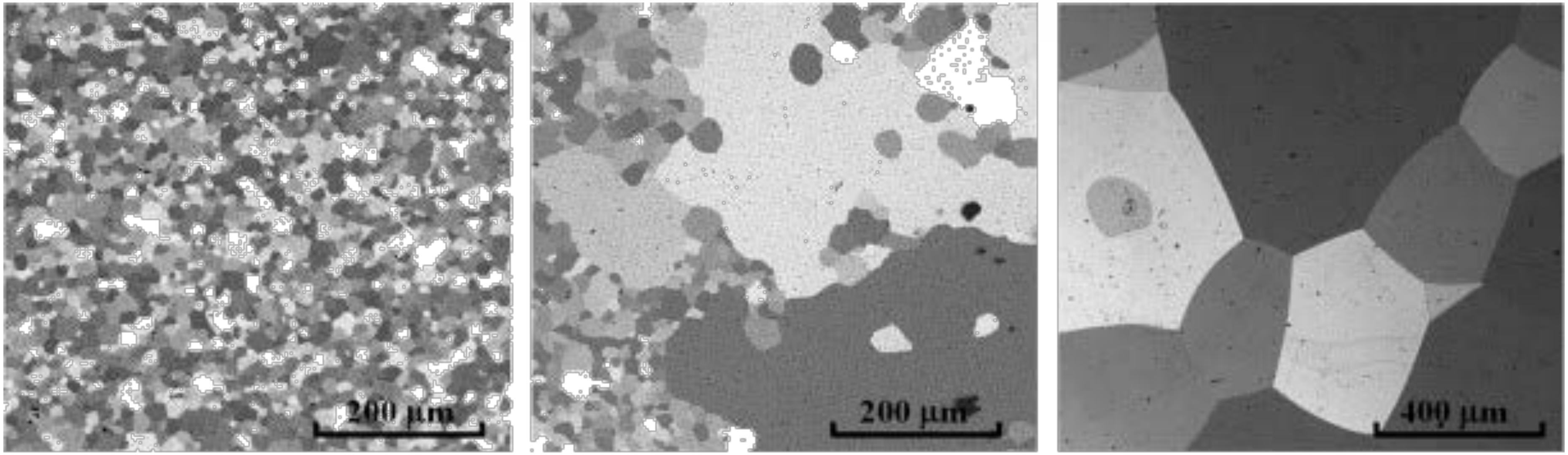
- Sub-micrometer resolution achieved through two-stage magnification technique, allowing for voxel sizes of around 700 nm



System-of-interest: Al-3.5wt%Cu

Known to exhibit AGG below solvus (491 C)

- Consists of θ-Al_2Cu particles in Al matrix
- Fully recrystallized at 400 C for 30 min.
- XRT done in interrupted manner during annealing at 485 C



460 °C 485 °C 500 °C

J. Dennis, P.S. Bate, and F.J. Humphreys, *Acta Mater.* **57**, 4539-4547 (2009).

Agenda

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  - Driving forces for grain boundary motion
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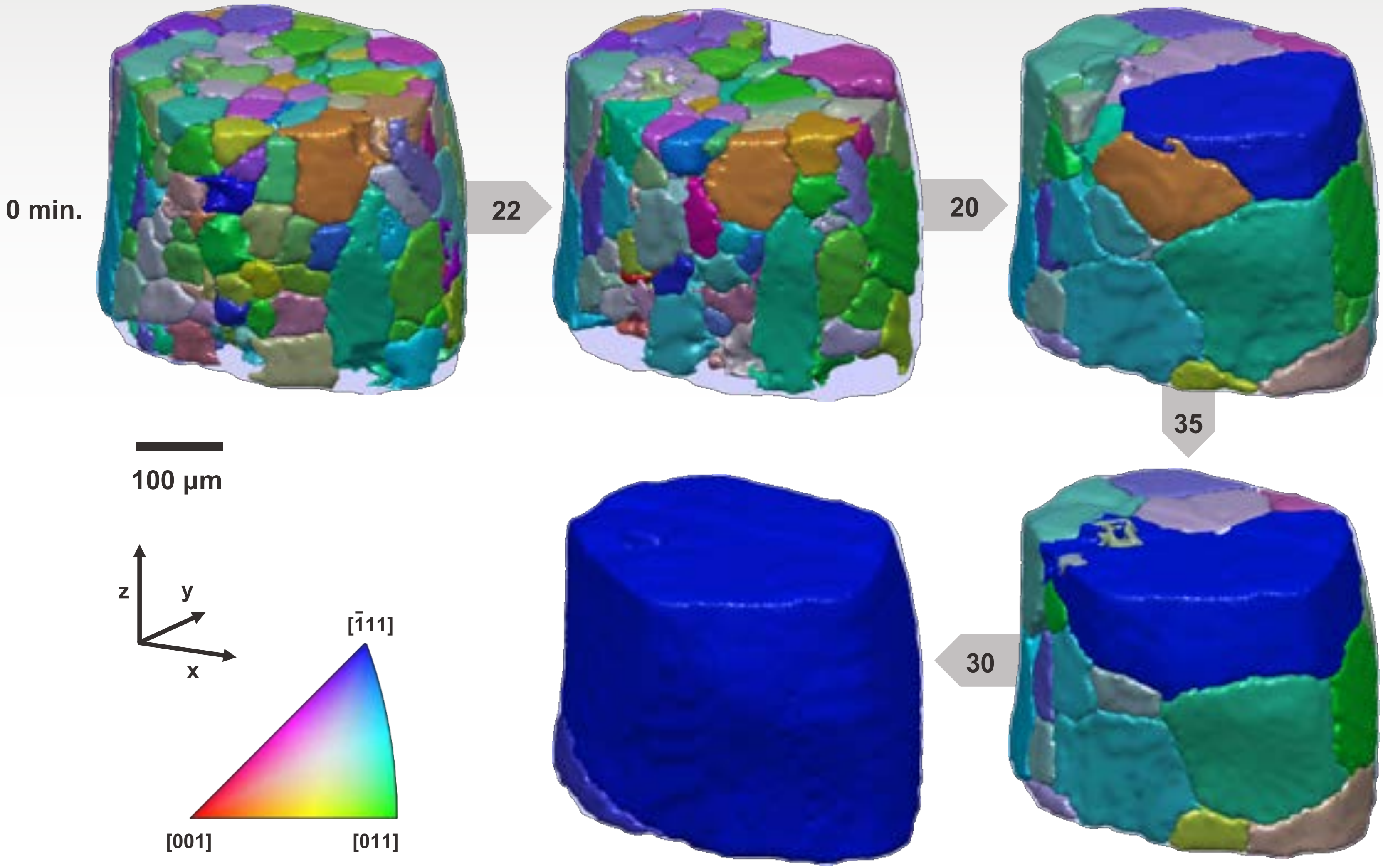
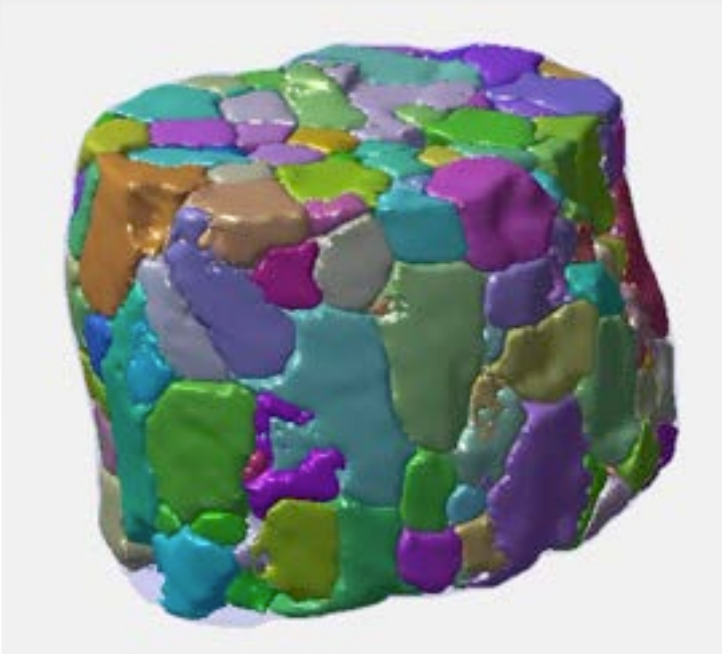
  - X-ray diffraction tomography
  - X-ray absorption tomography
  - Integrating the two imaging modalities
- **Results and discussion**
  - Abnormal grain selection and growth
  - Grain migrations driven by strain energy

R. Keinan, et al., A.J. Shahani, *Acta Mater.* **148**, 225 (2018).
J. Kang, et al., A.J. Shahani, *Integr. Mater. Manuf. Innov.* **8**, 388 (2019).
N. Lu, et al., A.J. Shahani, *Acta Mater.* **195**, 1 (2020).
N. Lu, et al., A.J. Shahani, *Metall. Mater. Trans. A* **52**, 914 (2021).

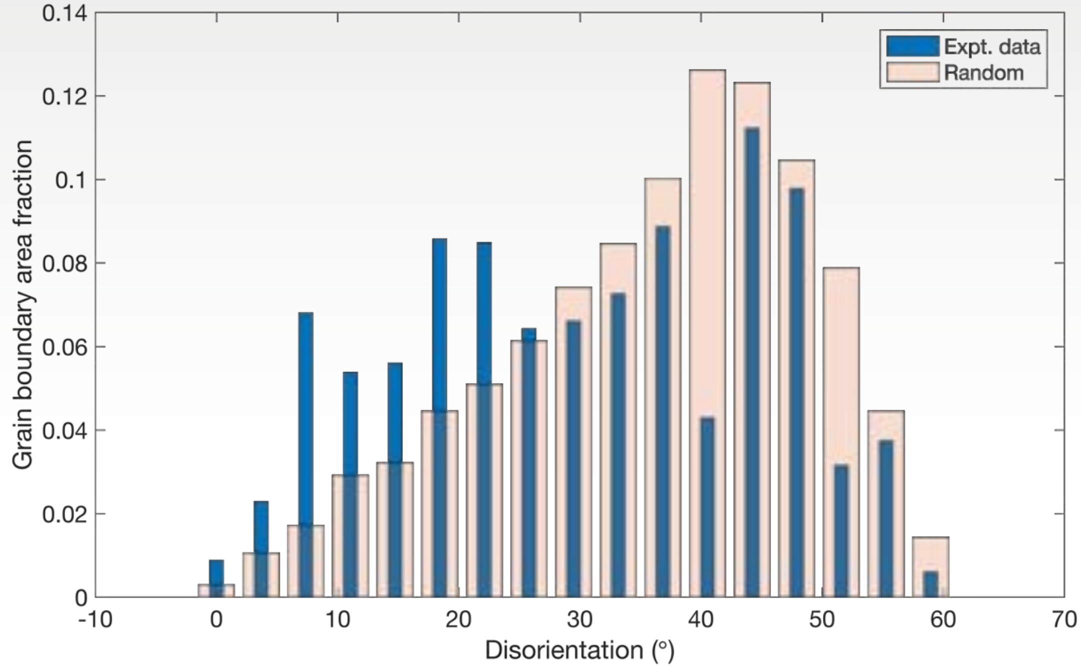
Orientation mapping during AGG

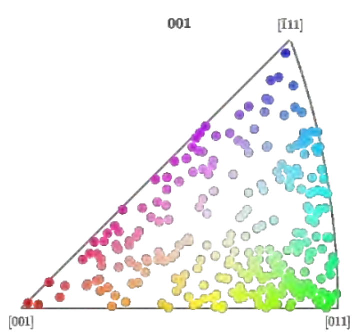


Initial condition



100 μm

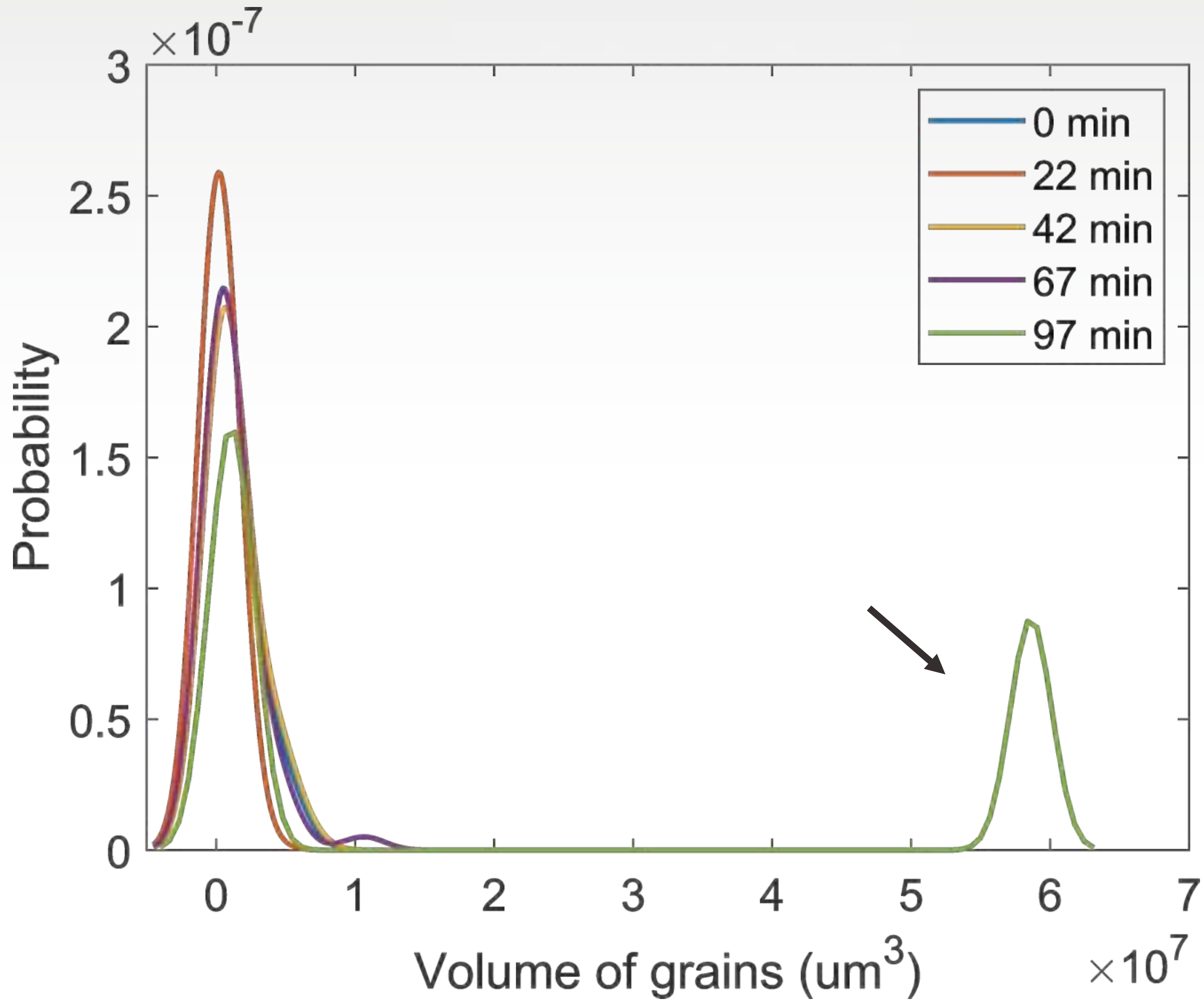




- Sample has no distinct texture
- Grain boundary distribution is random
- Grains are 30 μm in size (on average)
- 267 grains captured within field-of-view

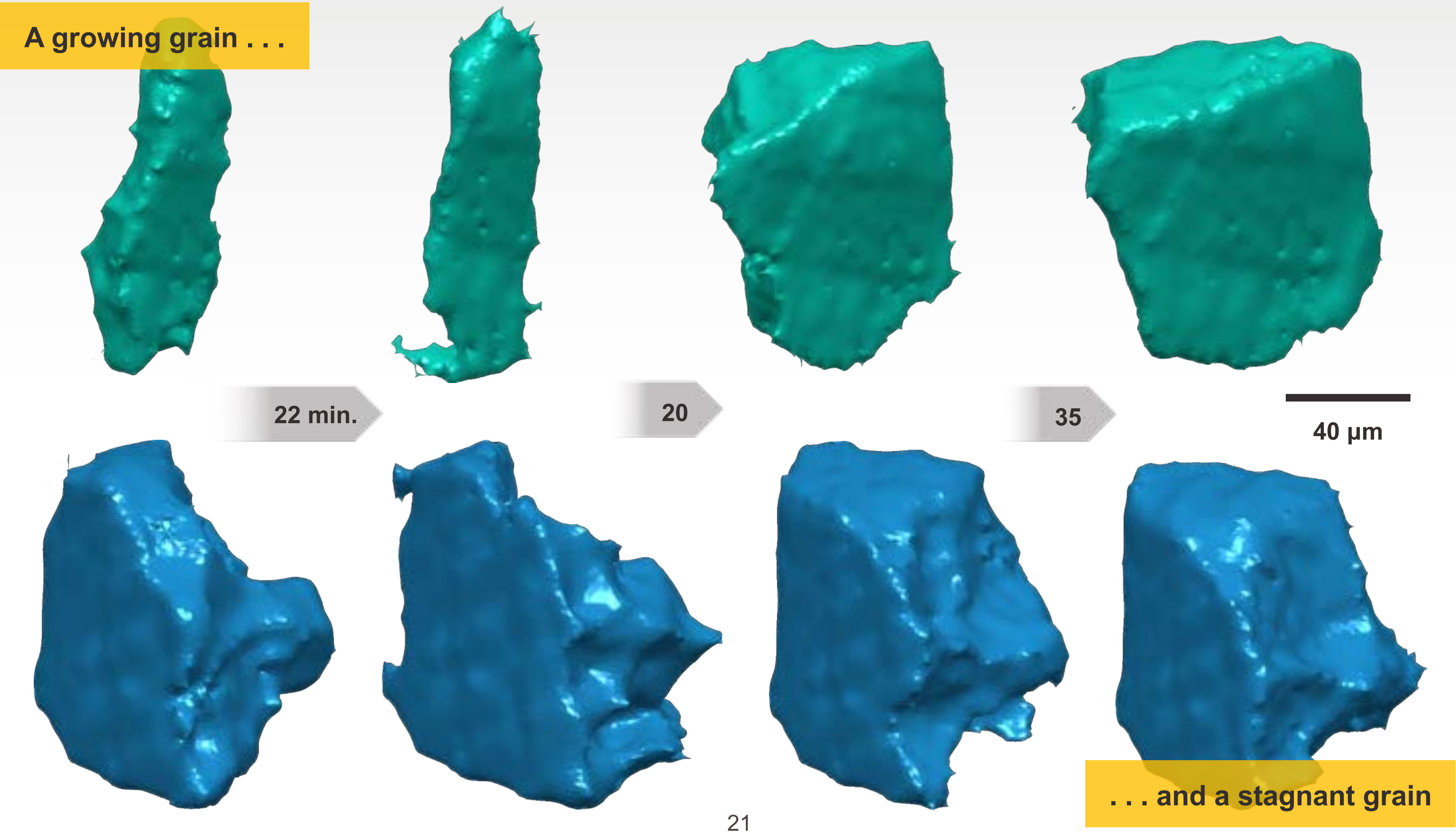
Evolution of grain size

- Bimodal distribution of grain size seen at long anneal times, consistent with AGG
- **Which of the grains becomes abnormal?**
- Requires the ability to track individual grains as a function of annealing time

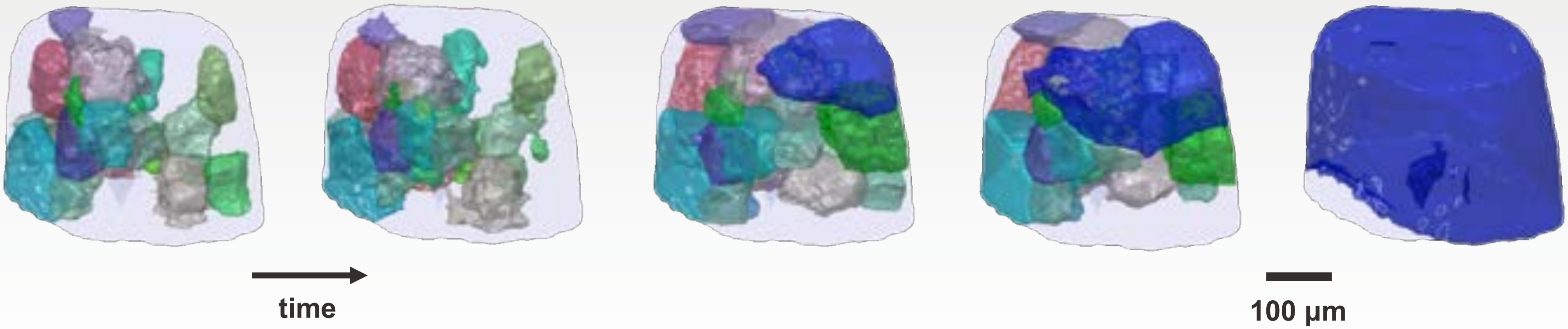


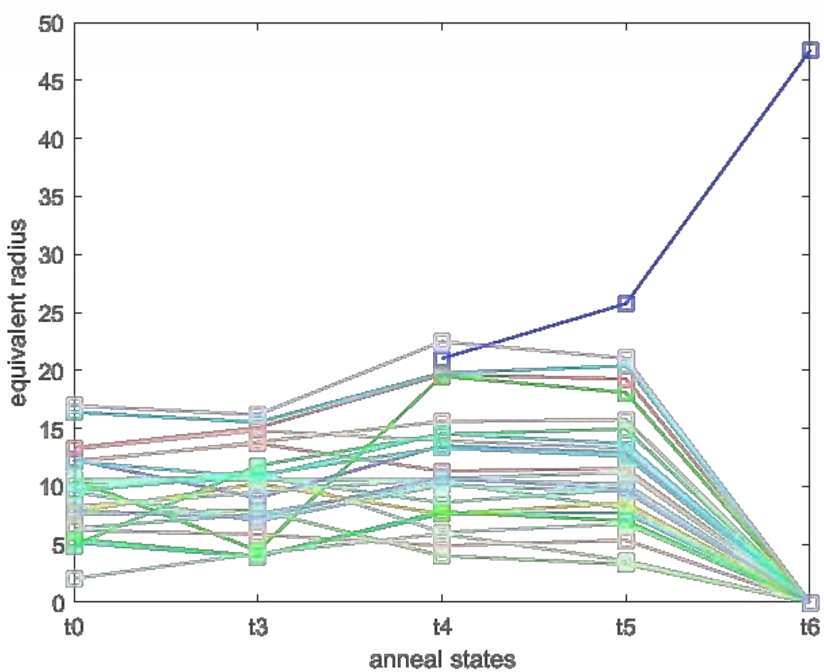
J. Kang, et al., A.J. Shahani, *Integr. Mater. Manuf. Innov.* **8**, 388 (2019).

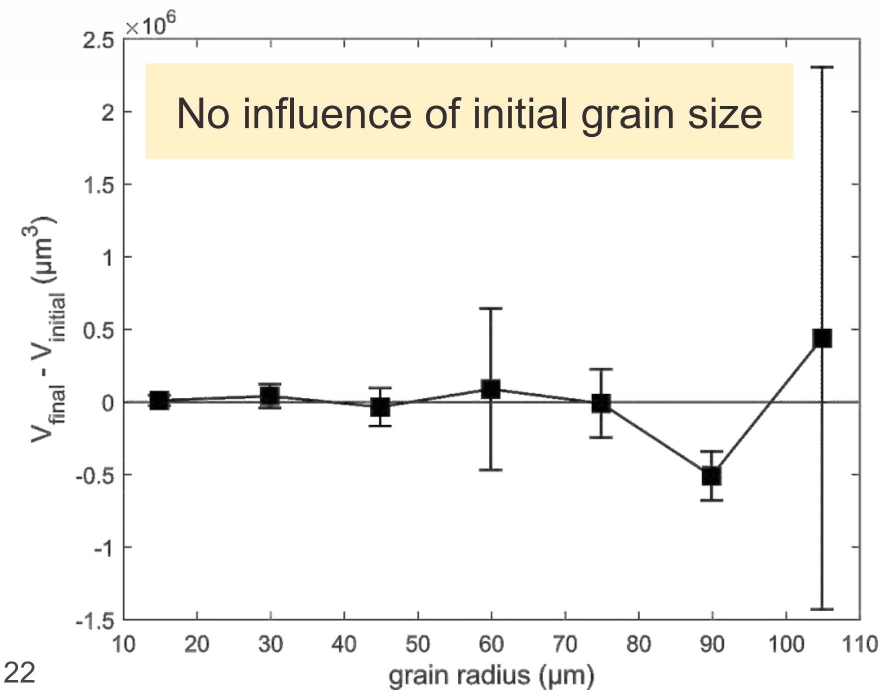
Individual grain trajectories



Some basic statistics: Grain size

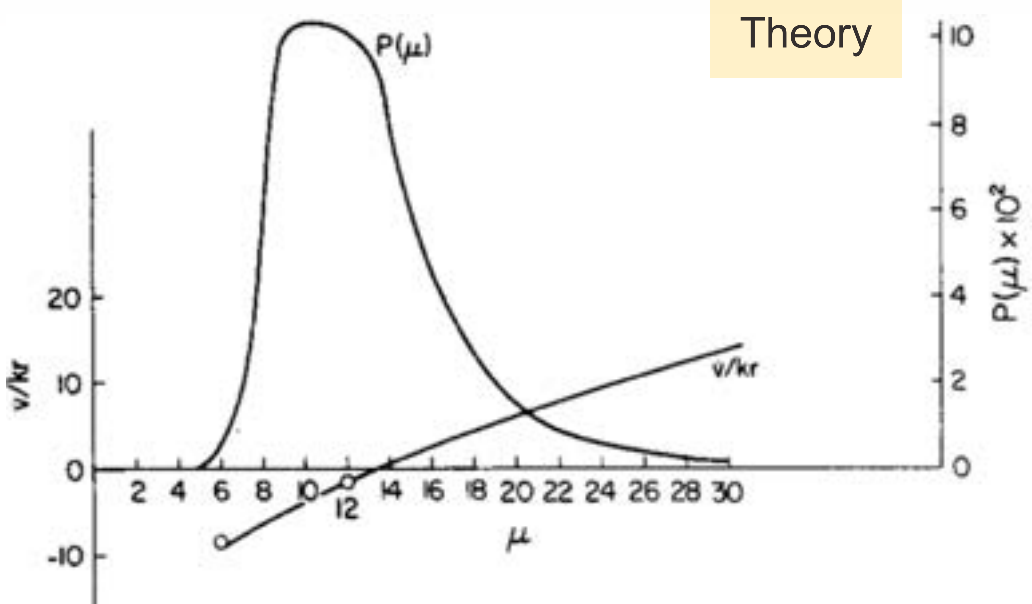


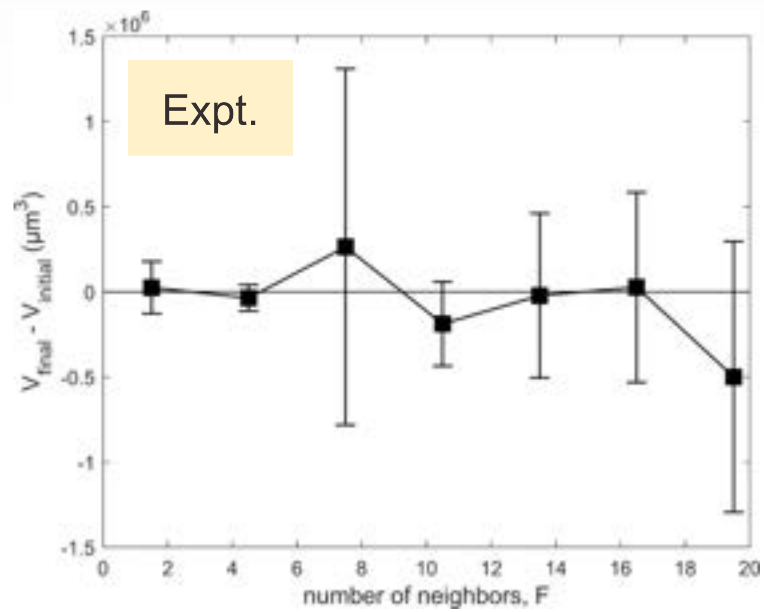




Some basic statistics: Grain topology

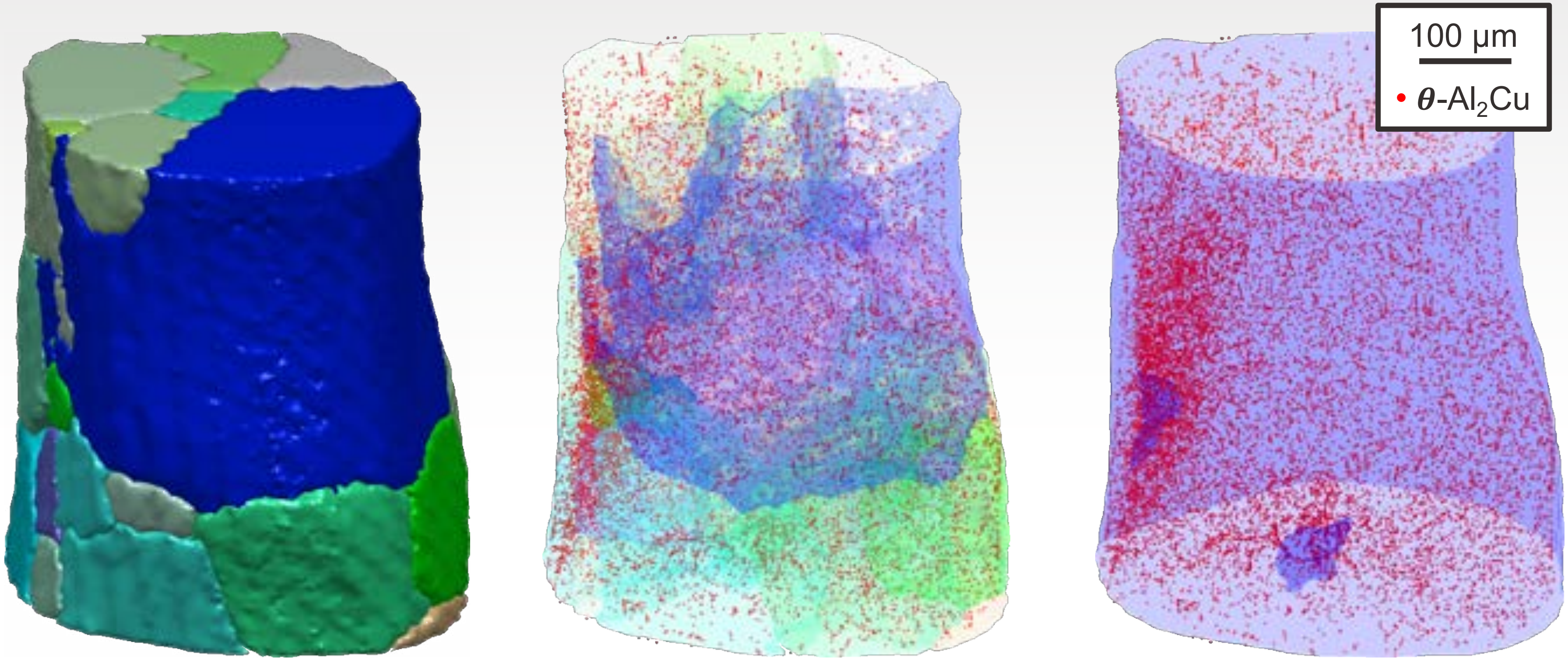
- According to W.W. Mullins, “grains with less than 13 neighbors (*F*) shrink, and those with greater than 14 neighbors grow; the mathematical point of zero growth is *F* = 13.3.”
- **No such effect of grain topology observed here**





W.W. Mullins, *J. Appl. Phys.* **27**, 900 (1956).
W.W. Mullins, *Acta Metall.* **37**, 2979 (1989).

Integrating absorption and diffraction data

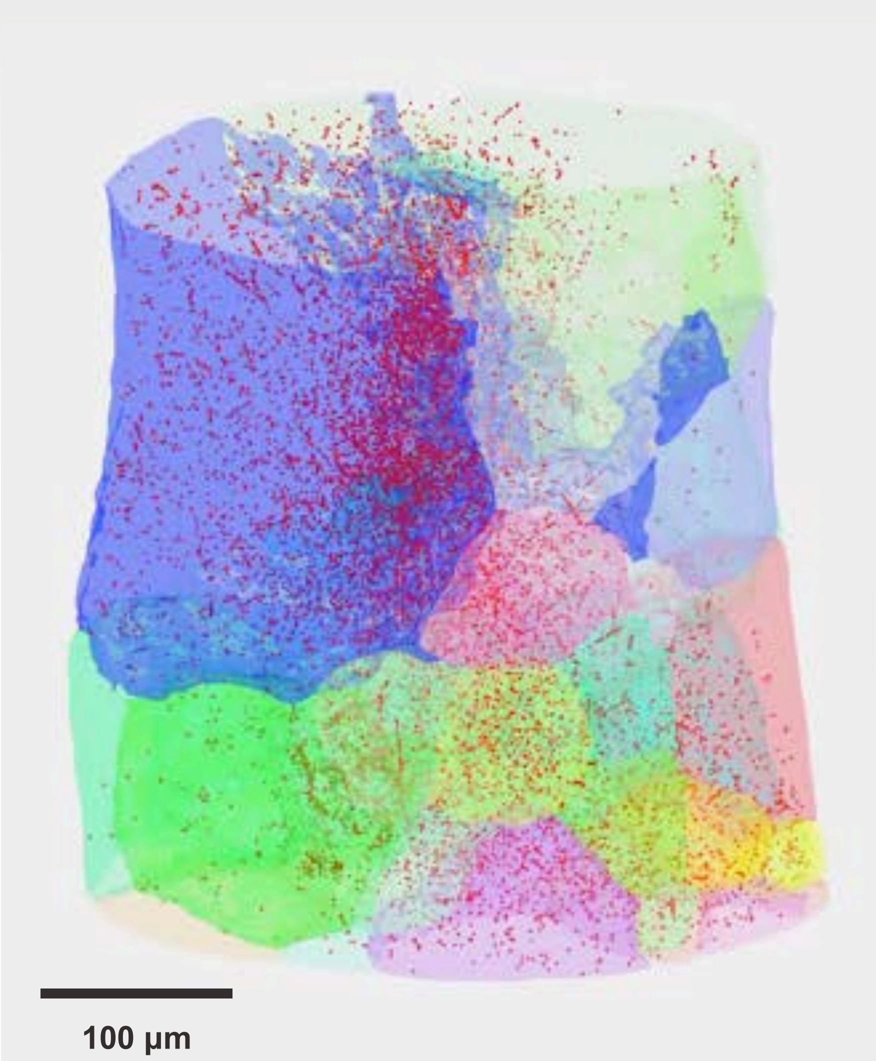


Diffraction XRT | **Hybrid** | **Absorption XRT**

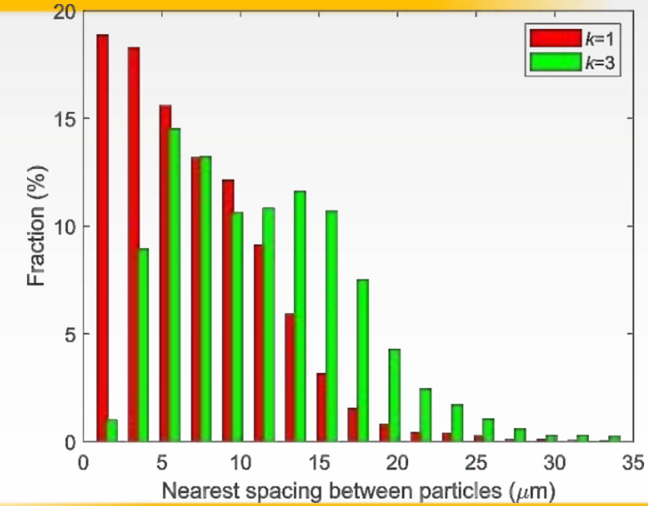
Both particle locations and grain sizes are **non-uniform** @ 67 min.

R. Keinan, et al., A.J. Shahani, *Acta Mater.* **148**, 225 (2018).

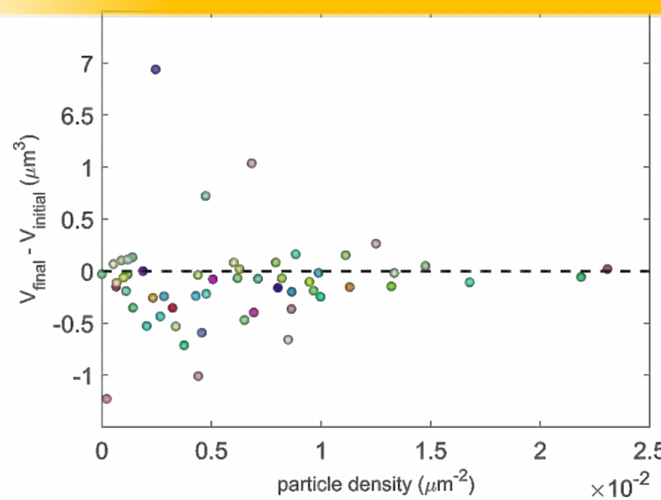
Effect of the particle cloud



Particle dispersion is nonuniform . . .



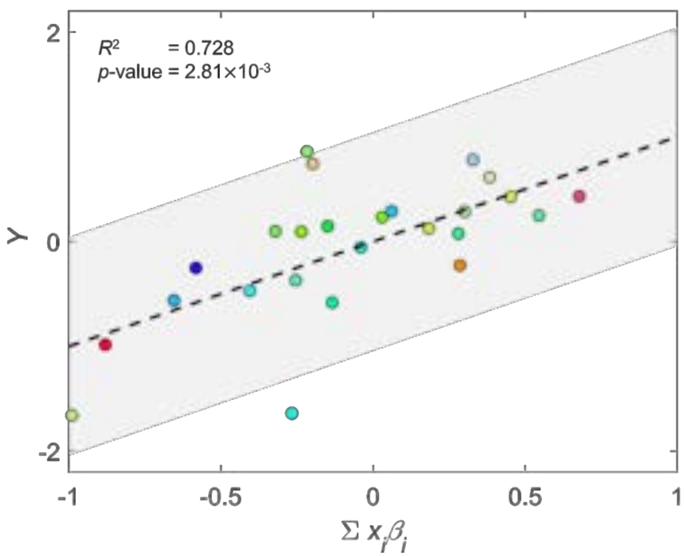
. . . and leads to a distribution of grain growth rates



Which variable is most important?

- **Multiple linear regression** (MLR) models relationship between multiple variables ("structure") and a response variable ("evolution")

- Only statistically significant predictor is the local distribution of particles ⇒ **AG selected from particle-poor region**



	p	R
Neighbors, N	0.97	−0.14
Grain size, V	0.73	−0.38
Particle density, ρ	0.19	−0.07
$N \times V$	0.99	−0.31
sgn(−V) × ρ	**<0.01**	**+0.66**

N. Lu, et al., A.J. Shahani, *Acta Mater.* **195**, 1 (2020).

Why does AGG persist?

1. **Growth rate (or driving force) of abnormal grain is positive**

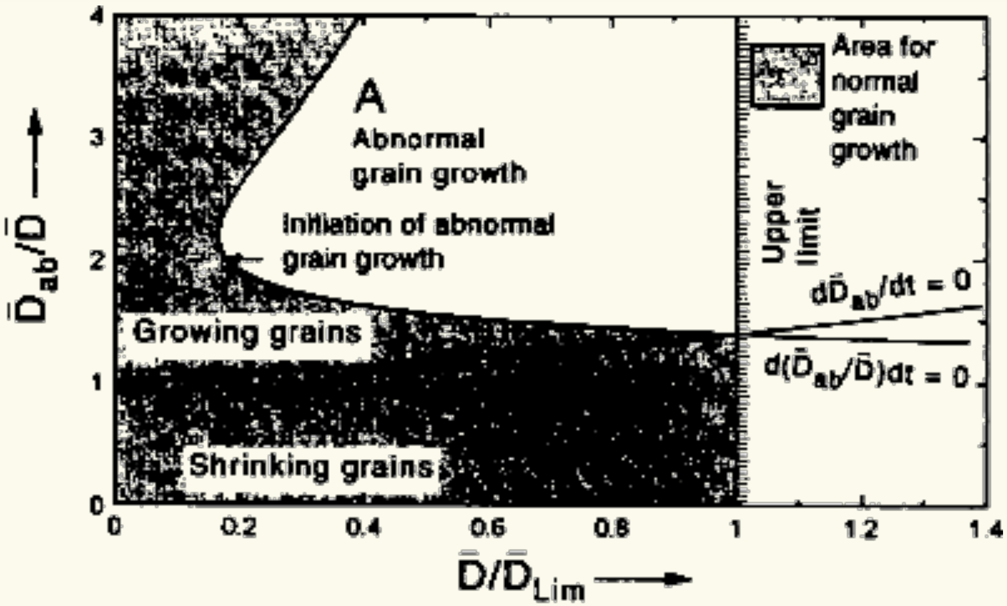
$$\frac{dR_{ab}}{dt} > 0$$

2. **Abnormal grain must grow faster than other, normal grains**

$$\frac{d}{dt}\left(\frac{R_{ab}}{R_n}\right) > 0$$

Mechanism map for 2D GG

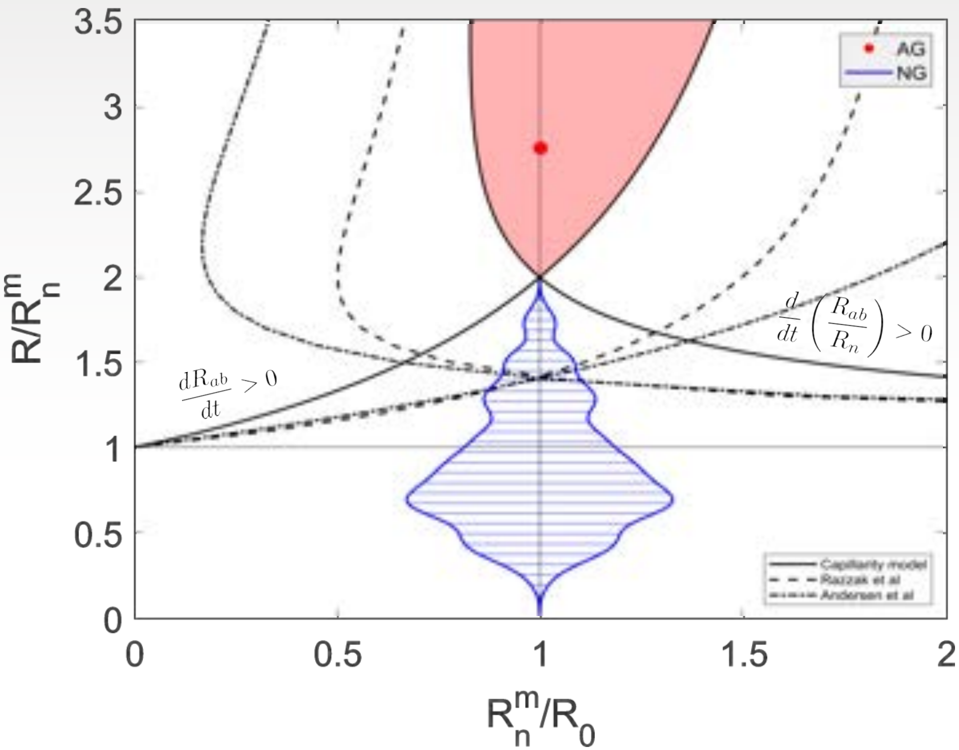
- AGG favored when matrix strongly pinned ($D/D_{lim} = 1$) and the abnormal grain has a size advantage ($D_{ab}/D \geqq 1.4$)
- In all other regions, normal grain growth occurs



I. Andersen et al., *Acta Metall. Mater.* **43**, 2689 (1995).

Mechanism map for grain growth

- Extended the mechanism map from 2D to 3D: Narrower regime for AGG
- Superimpose grain size data *right before* AGG consumes sample volume
- **Abnormal grain falls into region where its abnormal growth should persist, while normal grains below this region**



R = equivalent radius of abnormal grain

R_n^m = equivalent radius of normal grain

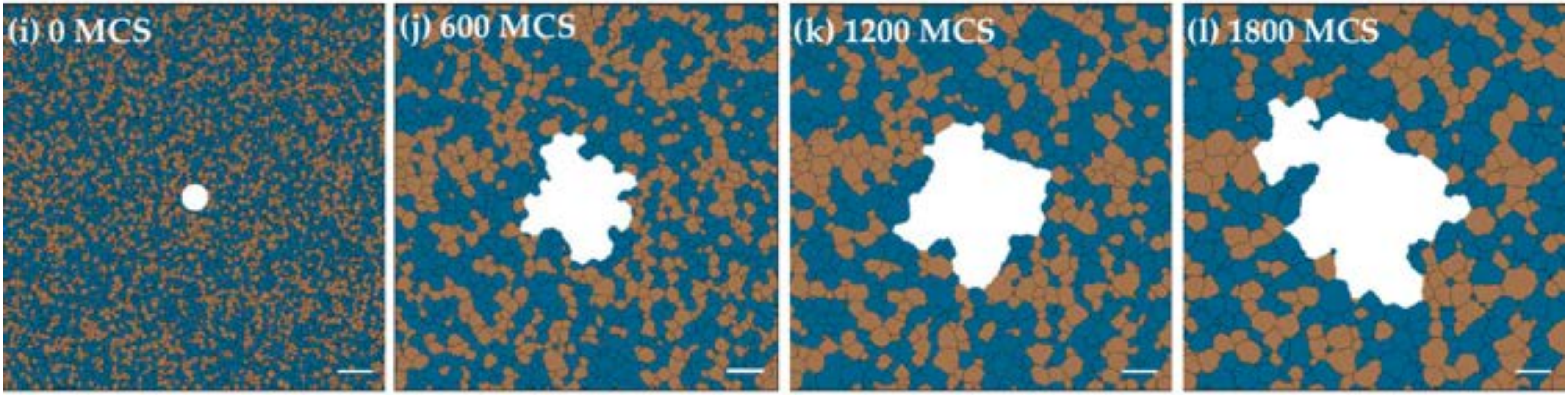
R_0 = limiting grain size (equilibrium)

M. Razzak et al., *ISIJ INT*. **52**, 2278 (2012).
I. Andersen et al., *Acta metal. Mater*. **43**, 2689 (1995).

Pathway of abnormal grain

Insights from MCP simulations

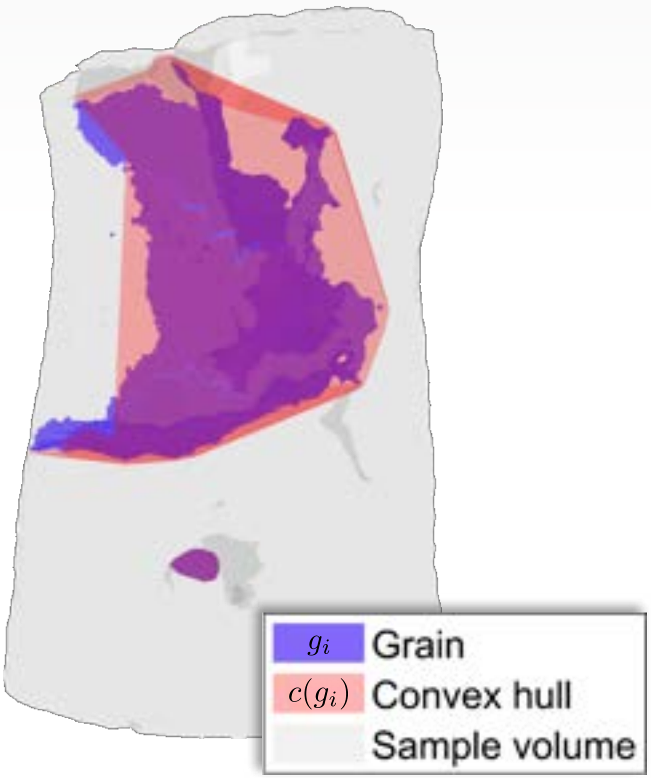
- According to Holm, AGG proceeds through “rapid localized growth events” which give abnormal grain its irregular shape
  - Candidate grain encounters clusters of high mobility boundaries (which may represent here particle-free regions)

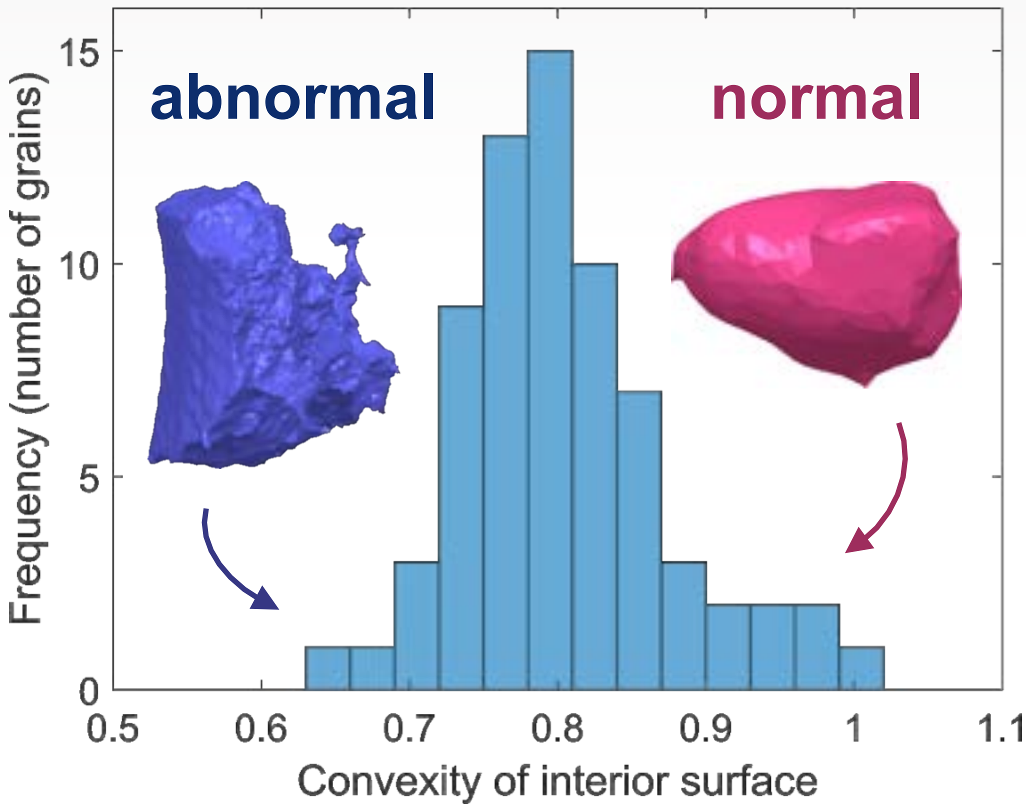


B.L. DeCost and E.A. Holm, *Metall. Mater. Trans. A* **48**, 2771 (2017).

Grain morphology may offer clues

- Quantify shape of *i* through convexity $\psi = A[c(g_i)]/A(g_i)$
- Right before abnormal grain consumes volume, it shows $\psi \ll 1$





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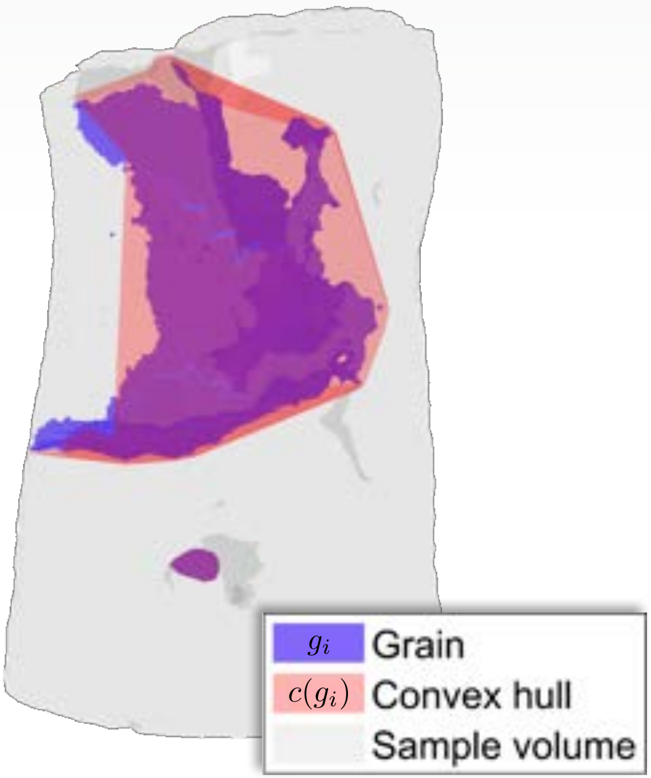

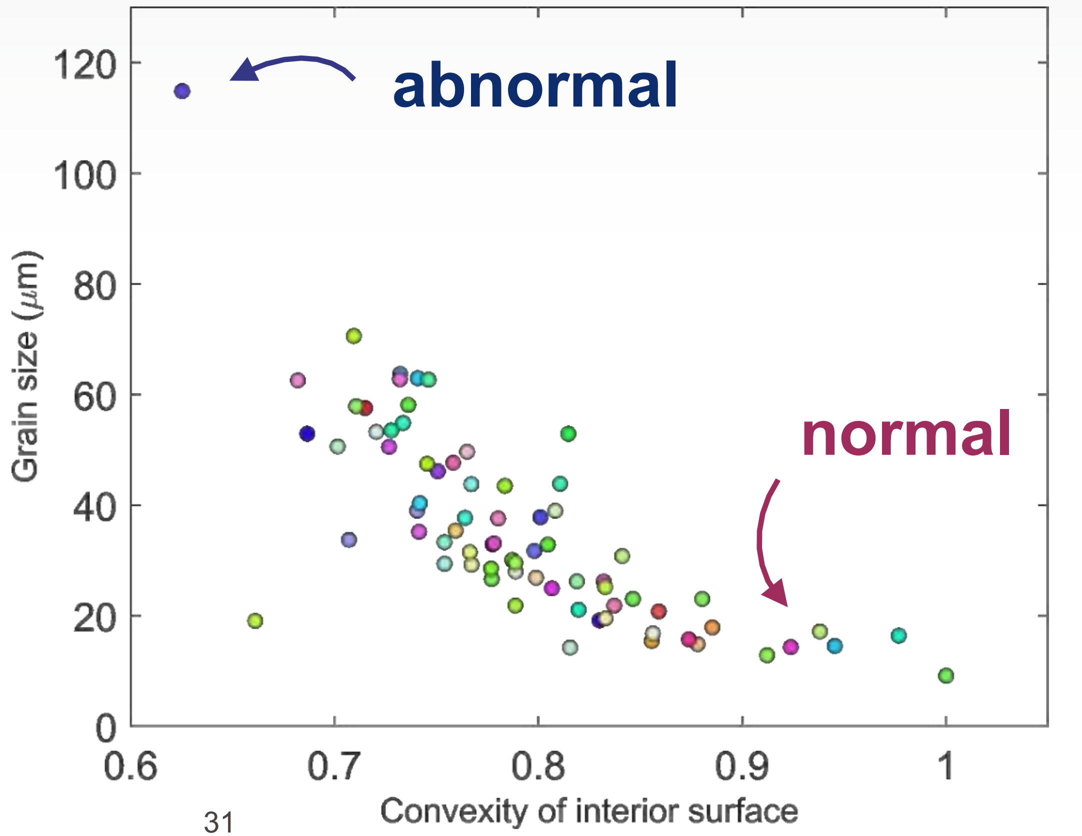

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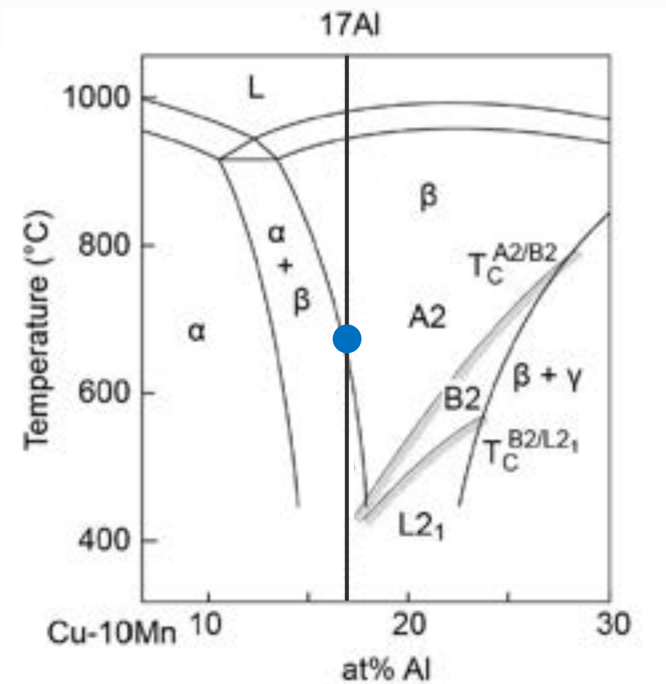
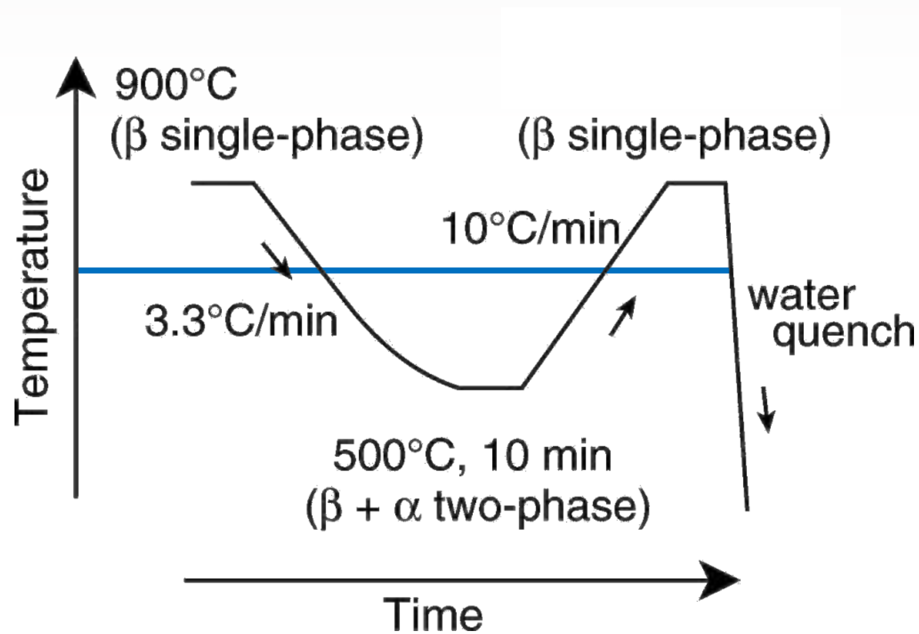
R. Keinan, et al., A.J. Shahani, *Acta Mater.* **148**, 225 (2018).
J. Kang, et al., A.J. Shahani, *Integr. Mater. Manuf. Innov.* **8**, 388 (2019).
N. Lu, et al., A.J. Shahani, *Acta Mater.* **195**, 1 (2020).
N. Lu, et al., A.J. Shahani, *Metall. Mater. Trans. A* **52**, 914 (2021).

Grain evolution upon 'dynamic annealing'

- Consider alloy of composition Cu-17at%Al-11.4at%Mn
- Oscillate above and below solvus temperature for FCC-α phase
- Subgrains accommodate transformational strains between FCC-α particles and BCC-β matrix *via* dislocations



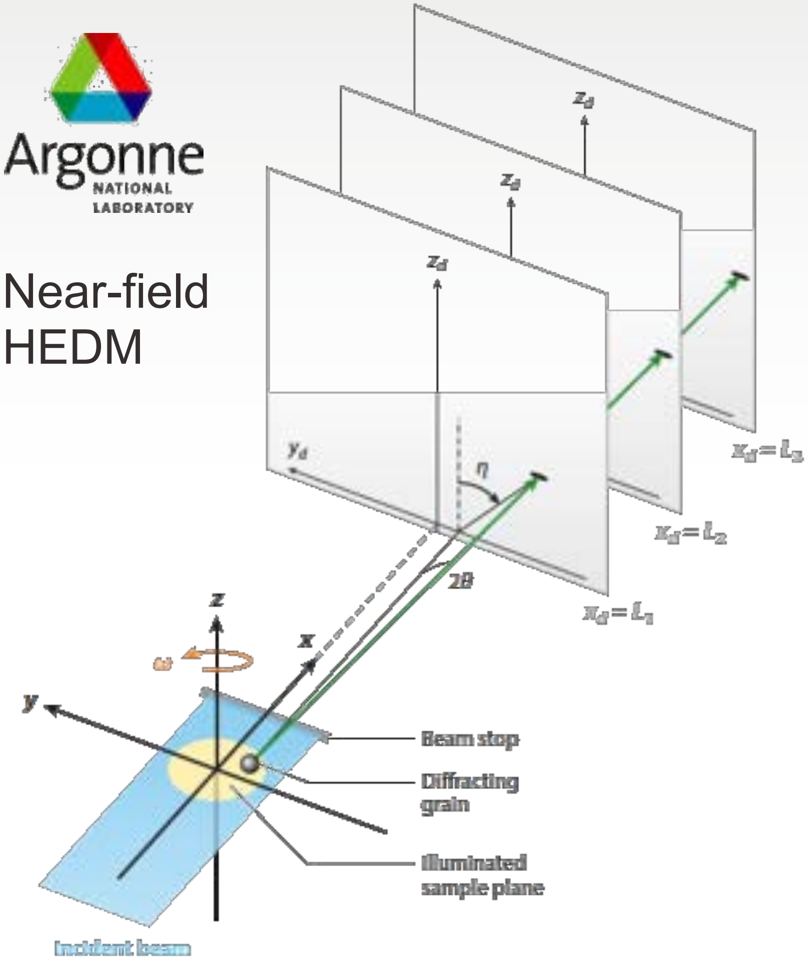


T. Omori et al., *Science* **341**, 1500 (2013).

Resolving intragranular structure

Argonne NATIONAL LABORATORY

Near-field HEDM

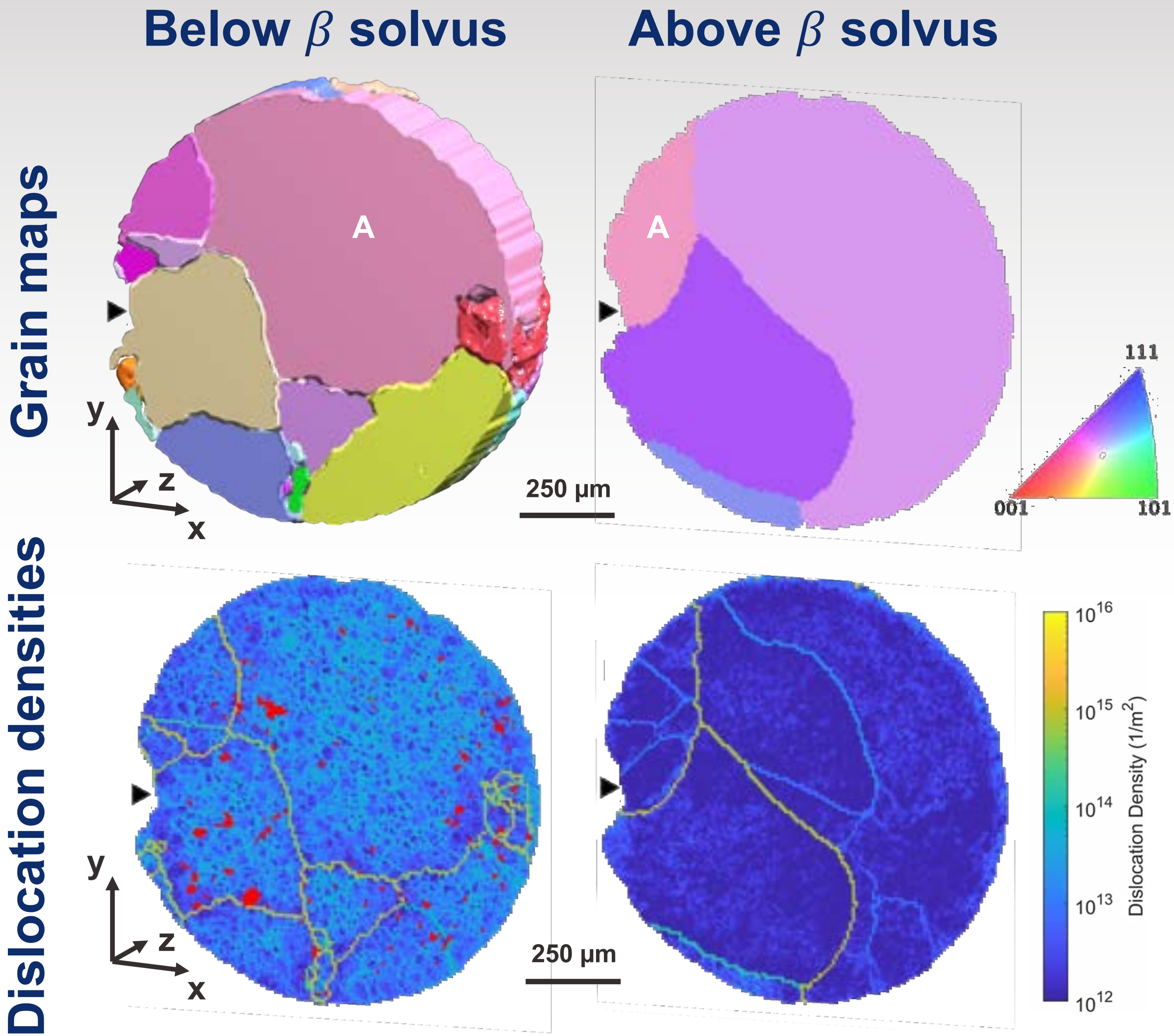


Synchrotron high energy X-ray diffraction microscopy

- High energy (≥ 40 keV) line-focused beam penetrates and diffracts from sample
- Affords high spatial resolution of 1 µm, allowing us to measure lattice rotations and hence, dislocation densities
- *In situ* furnace enables grain mapping at temperature

J.V. Bernier et al., *Annu. Rev. Mater. Res.* **50**, 396 (2020).
R.M. Suter et al., *Rev. Sci. Instrum.* **77**, 123905 (2006).

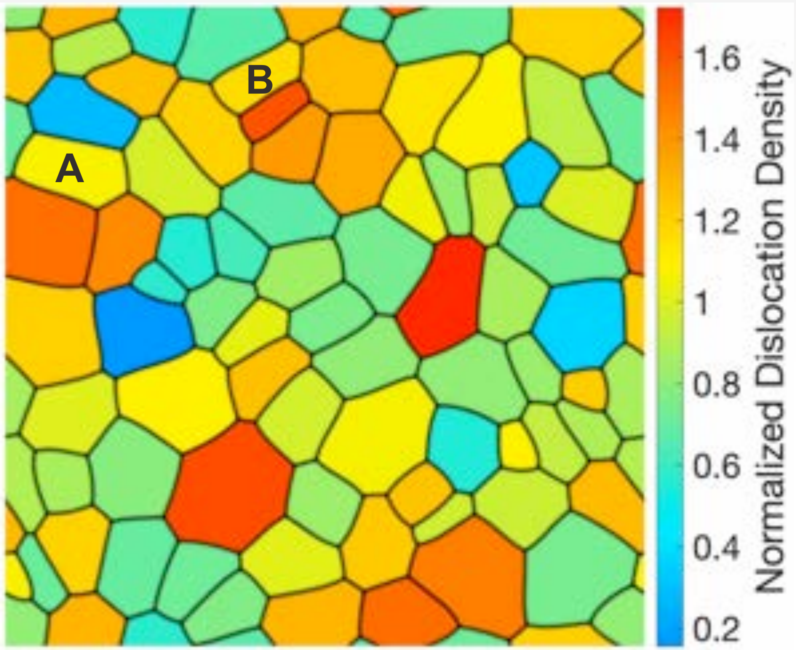
Anomalous grain migrations

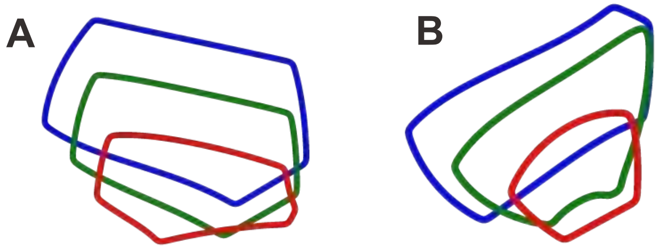


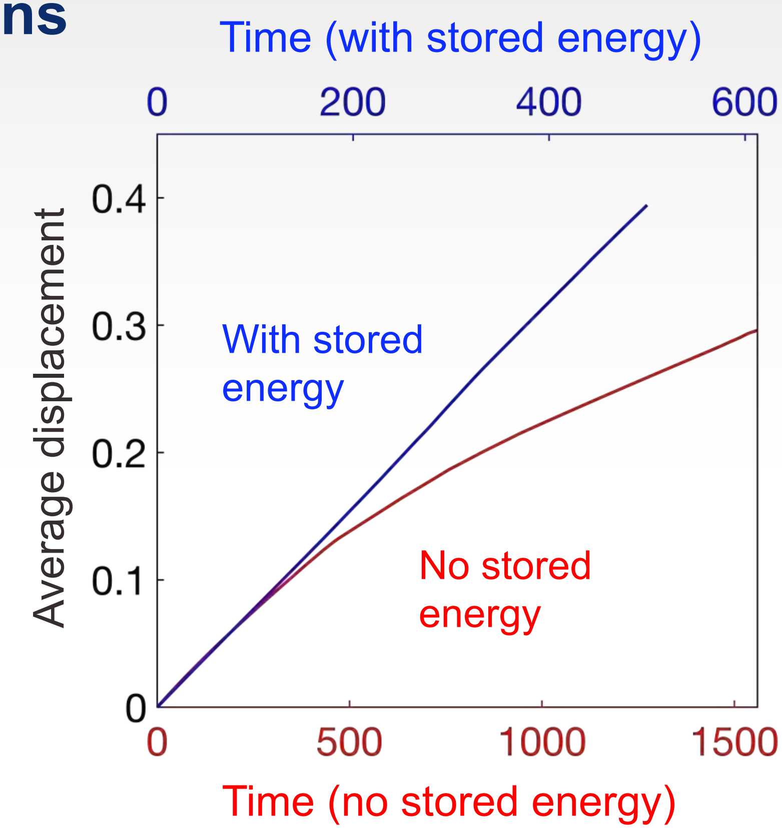
M. Higgins et al., K Thornton, A.J. Shahani, in review (2021).

Why do grain centers migrate?

Insight from phase-field simulations







Grain with medium value of stored energy may grow on one side and shrink on the other side, resulting in effective translation

M. Higgins et al., K Thornton, A.J. Shahani, in review (2021).

Conclusion and outlook

4D imaging provides new vision on microstructure evolution

- Diffraction-based X-ray tomographic microscopy

***In situ* observations of grain boundary motion**

- Abnormal grains form in relatively particle-free regions
- They grow if they have a size advantage
- Irregular grain morphology due to localized “growth spurts”
- Grains may “migrate” due to gradients in strain energy

Next steps: use our data to inform, parametrize simulations

R. Keinan, et al., A.J. Shahani, *Acta Mater.* **148**, 225 (2018).
J. Kang, et al., A.J. Shahani, *Integr. Mater. Manuf. Innov.* **8**, 388 (2019).
N. Lu, et al., A.J. Shahani, *Acta Mater.* **195**, 1 (2020).
N. Lu, et al., A.J. Shahani, *Metall. Mater. Trans. A* **52**, 914 (2021).

The next time you stare into a beer, contemplate the bubbles!

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Materials Science and Engineering
University of Michigan

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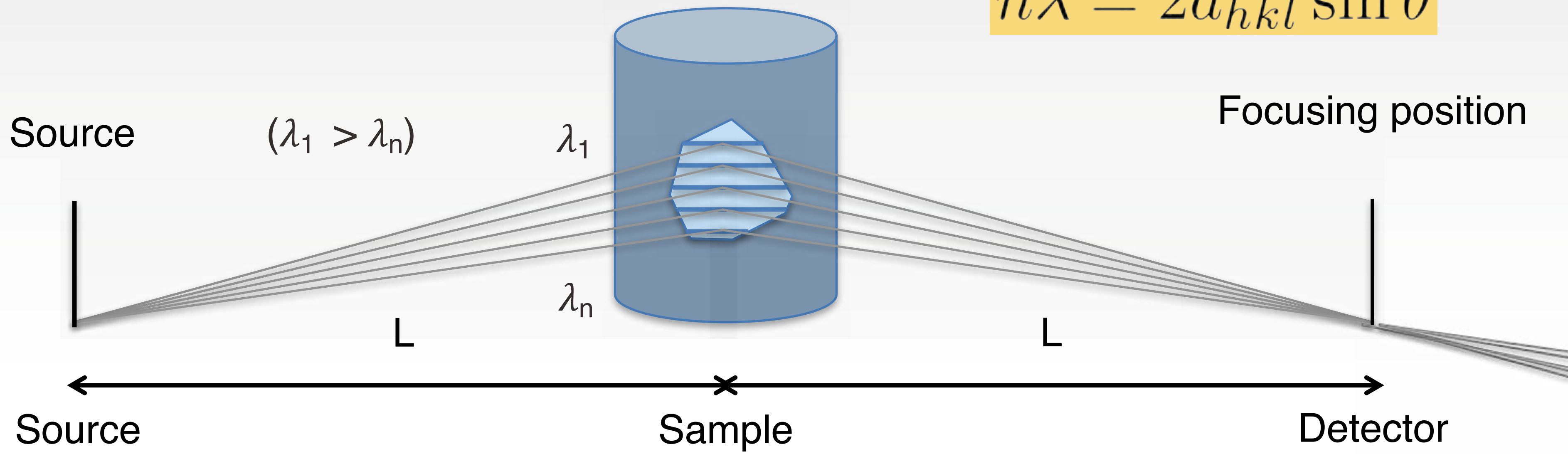
ARO YIP award no. W911NF-18-1-0162

Laue focusing geometry

$$n\lambda = 2d_{hkl}\sin\theta$$



This geometry offers a few key advantages:

1. Increased signal-to-noise due to focusing
2. Increased flux due to small source-detector distance
3. Substantially less overlap of reflections

Typical parameters

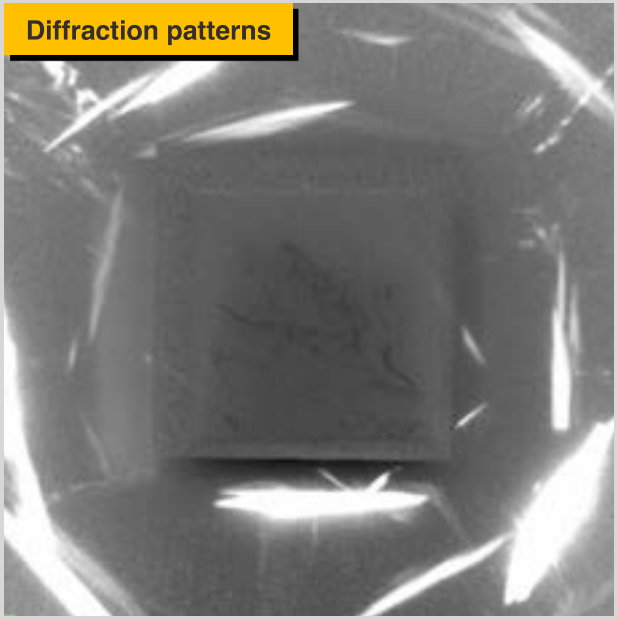
- Energy: 80 keV
- Working distance: 15 mm

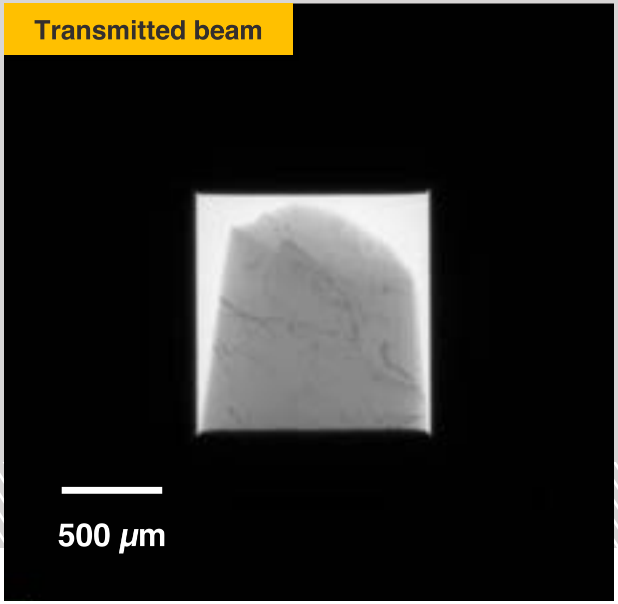
Diffraction contrast XRT

- Aperture: 750^2 μm^2
- Beamstop: 2 mm
- # Projections: 181 $\in (0,2\pi)$
- Exposure: 180 s

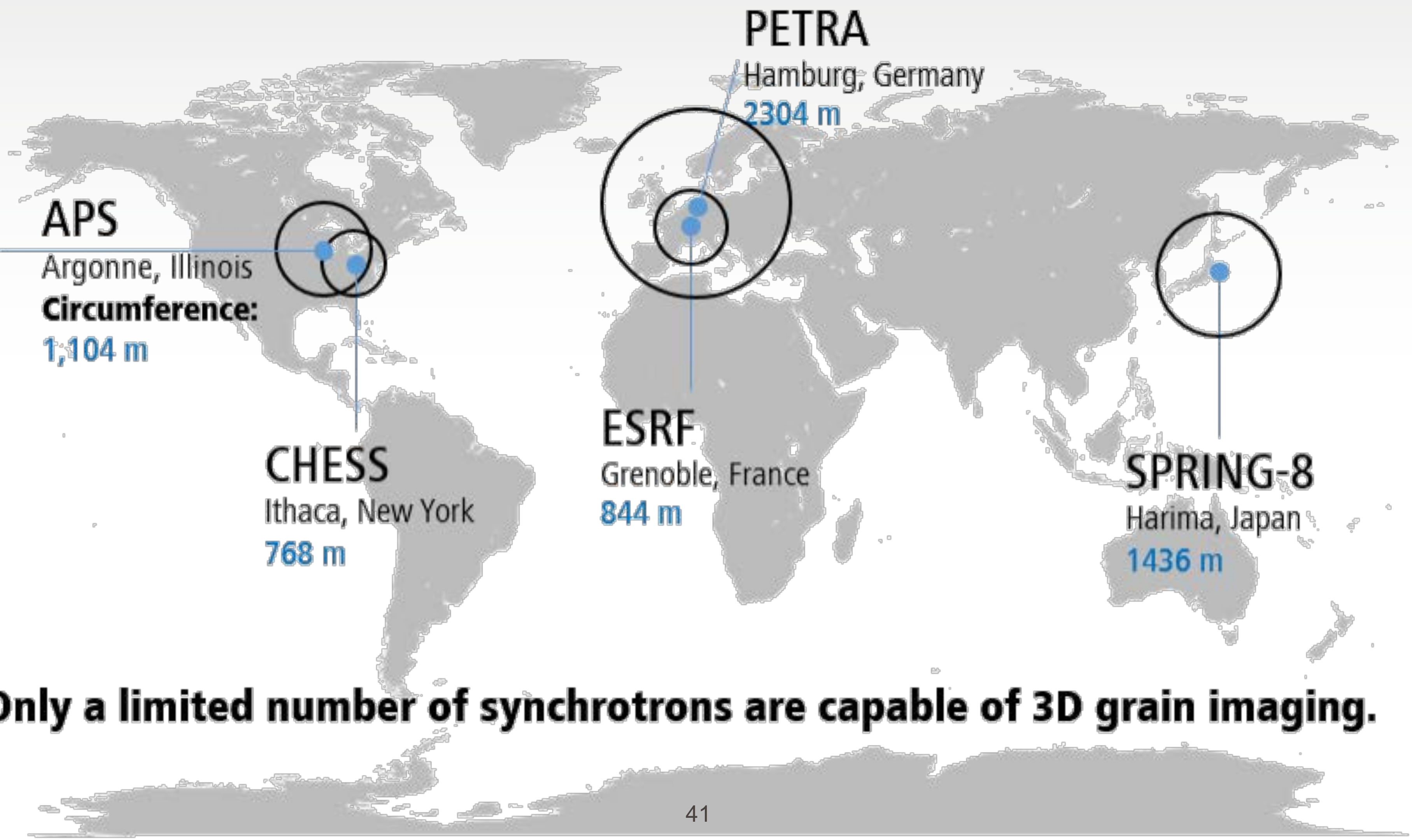
Absorption contrast XRT

- # Projections: 721 $\in (0,\pi)$
- Exposure: 2 s





Grain mapping at synchrotrons

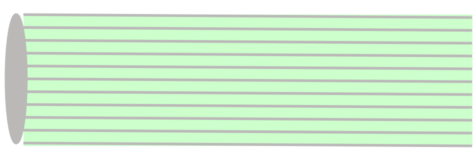


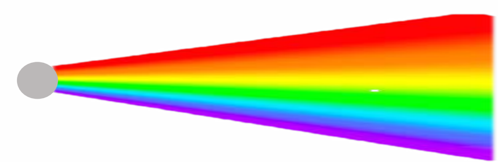
Only a limited number of synchrotrons are capable of 3D grain imaging.

Implementing DCT in the lab

Challenges and opportunities

vs.



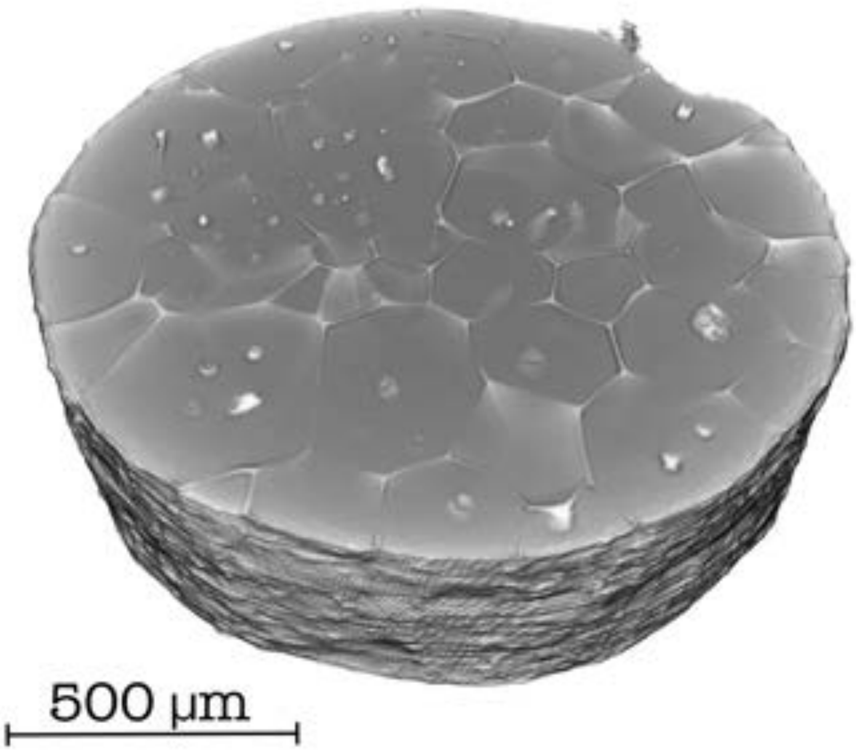


- Monochromatic, parallel *vs.* polychromatic, divergent sources
- Lab sources have **orders-of-magnitude less intensity**

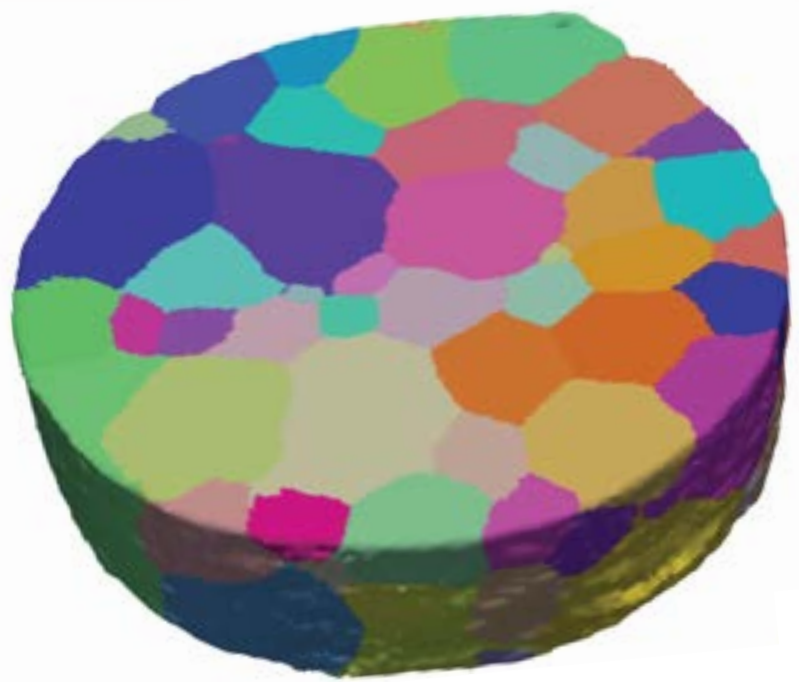
DCT reconstruction

- Cross-checked on an Al-5wt%Cu alloy where the Cu wets the grain boundaries $\Longrightarrow$ acts as a "marker"

Absorption reconstruction



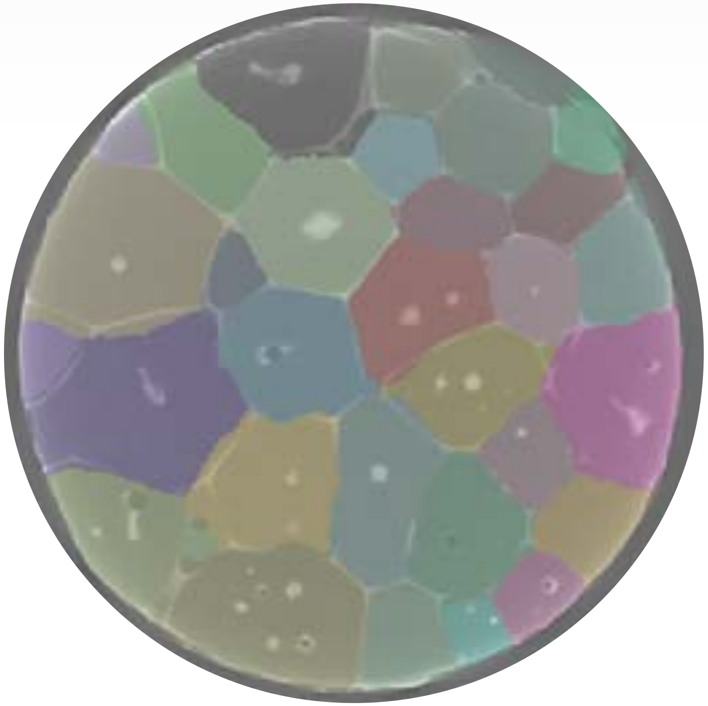
Diffraction reconstruction



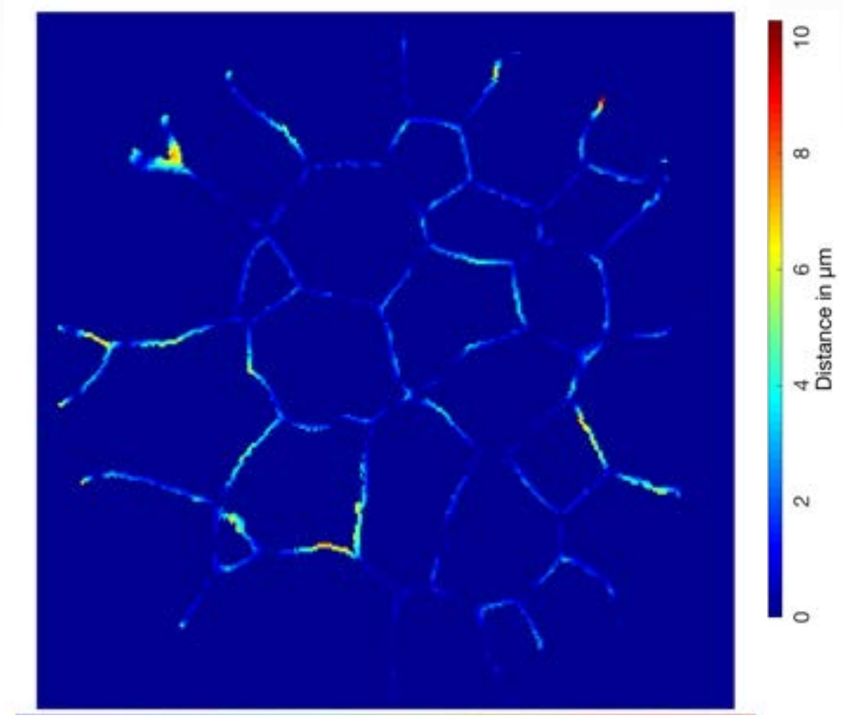
DCT reconstruction

- Computed **distance** between grain boundaries in absorption and diffraction volumes ⟹ “error” of 5 µm

Composite reconstruction

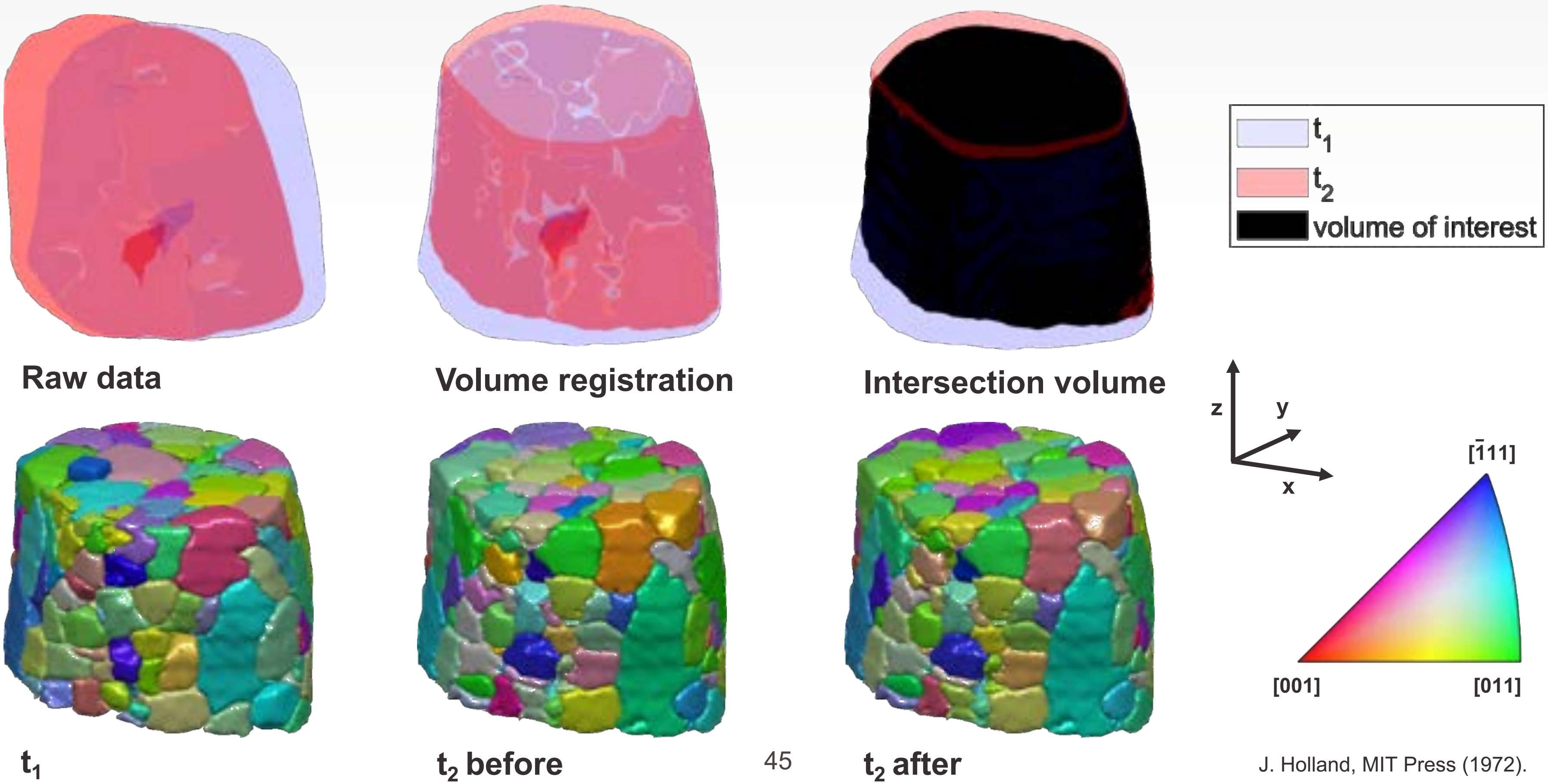


Grain boundary distance map



Tracking grains: Volume registration

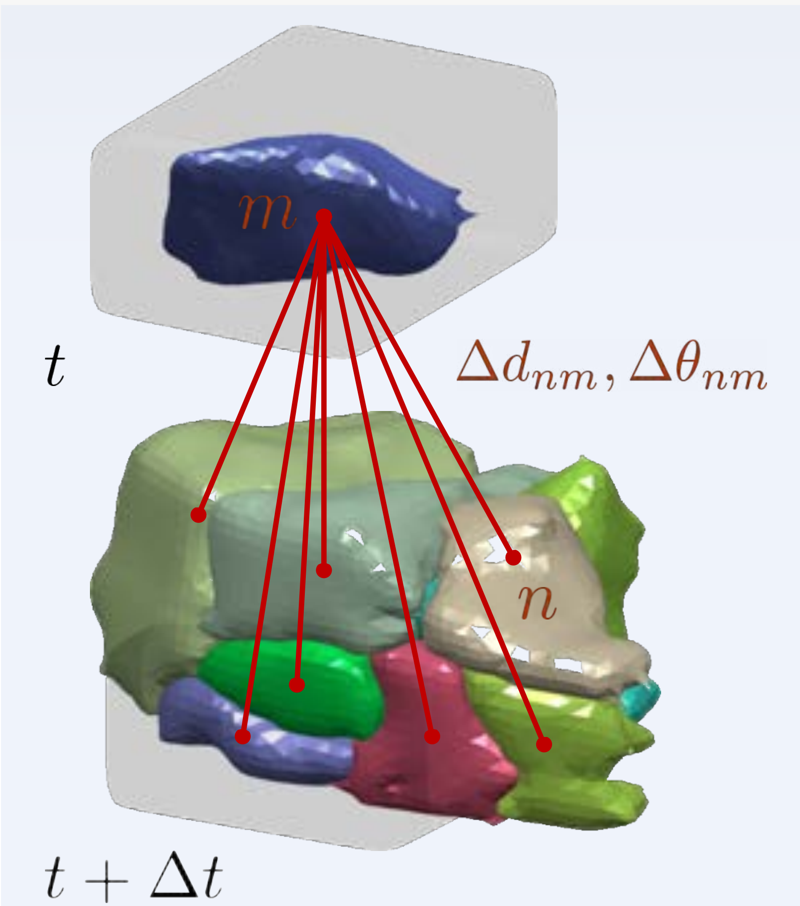
- Alignment of sample volumes from each time-step accomplished *via* machine learning (genetic algorithms)



J. Holland, MIT Press (1972).

Tracking grains: Bipartite matching

- Matching a grain from one time-step to the next in registered data accomplished *via* Kuhn-Munkres algorithm



$$\text{Cost} = c_1 \Delta d_{nm} + (1 - c_1)\Delta\theta_{nm}$$

m^{th} grain from time t

n^{th} grain

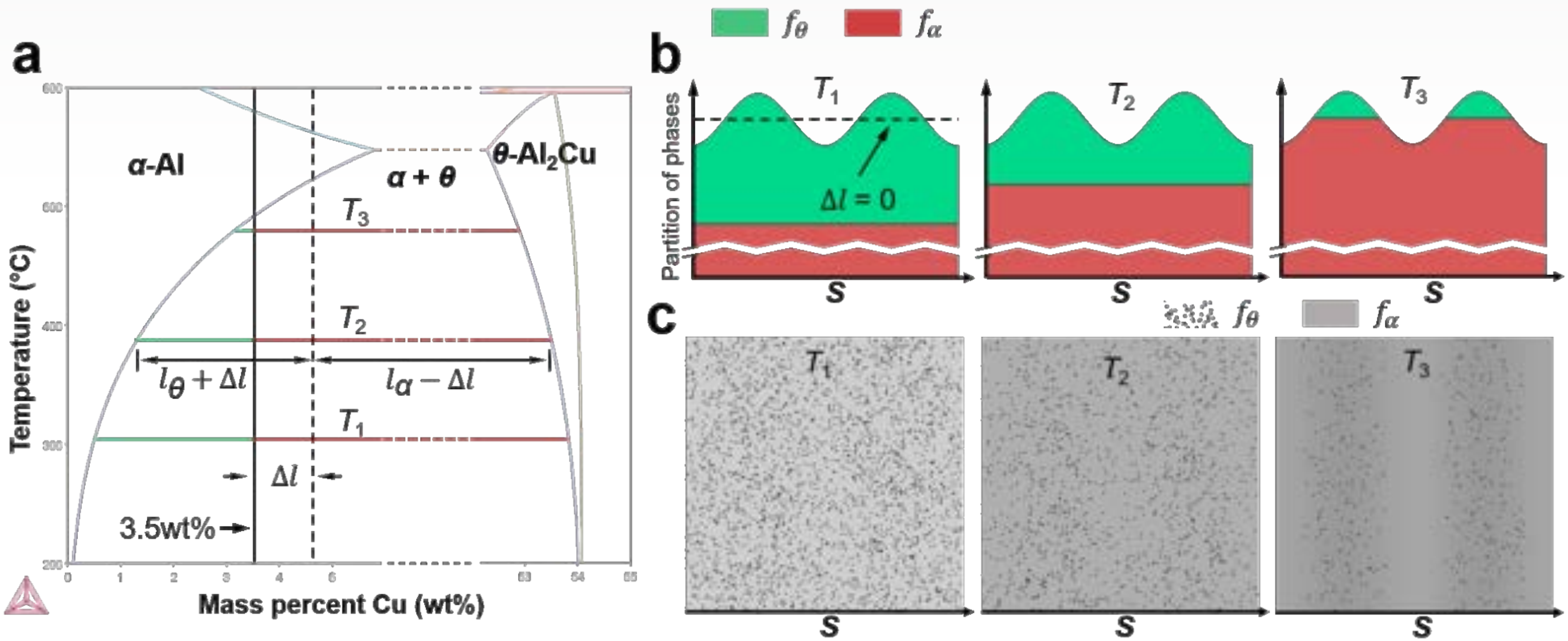
$$\begin{bmatrix} 0.1 & \infty & \infty & \infty & \infty & \cdots \\ 0.8 & \infty & 0.5 & 0.1 & \infty & \cdots \\ \infty & 0.6 & \infty & \infty & 0.3 & \cdots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \ddots \end{bmatrix}$$

- Approx. 80% of grains are matched, out of total no. grains

H.W. Kuhn, *Nav. Res. Logist. Q.* **2**, 83 (1955).
J. Munkres, *J. Soc. Ind. Appl. Math.* **5**, 32 (1957).

Origins of non-random particle distribution

- Experiments and modelling show that a non-random θ-Al_2Cu particle distribution induced by residual Cu segregation
- It is amplified at elevated temp. due to particle dissolution



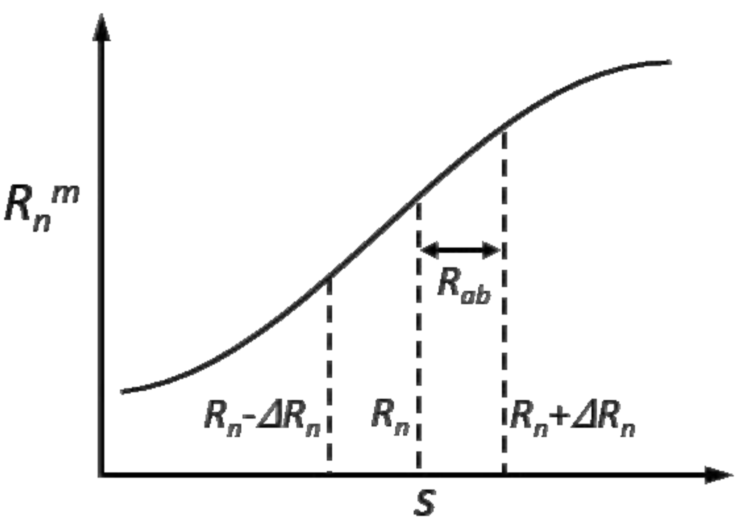
N. Lu, et al., A.J. Shahani, *Metall. Mater. Trans. A*, in press (2020).

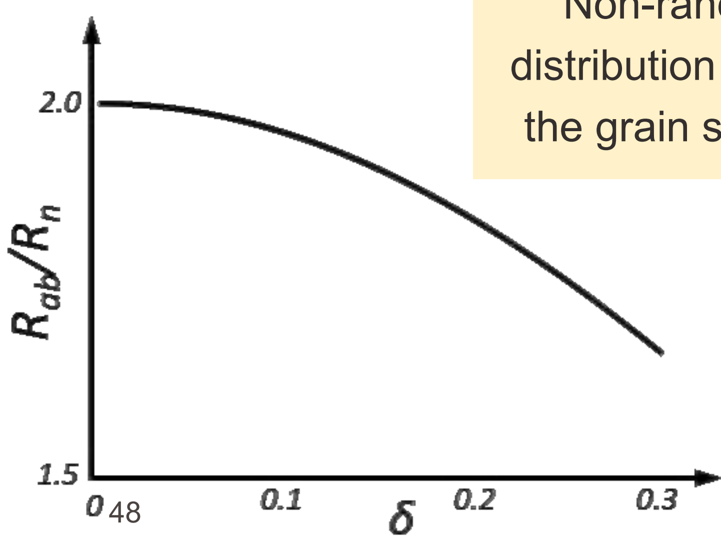
Extending the mechanism map

- **We can incorporate the influence of gradient in particle density**

$$\frac{dR_{ab}}{dt} = M\gamma\left(\frac{1}{R_n + \Delta R_n} + \frac{1}{R_n - \Delta R_n} - \frac{1}{R_{ab}} - \frac{1}{2R_0}\right)$$

- When matrix grains are pinned, *i.e.*, $\frac{R_n}{R_0} = 1$, and $\frac{dR_{ab}}{dt} > 0$, the size advantage for AGG is ≤2, and is a function of $\delta = \frac{\Delta R_n}{R_n}$





Non-random particle distribution (δ > 0) *relaxes* the grain size advantage

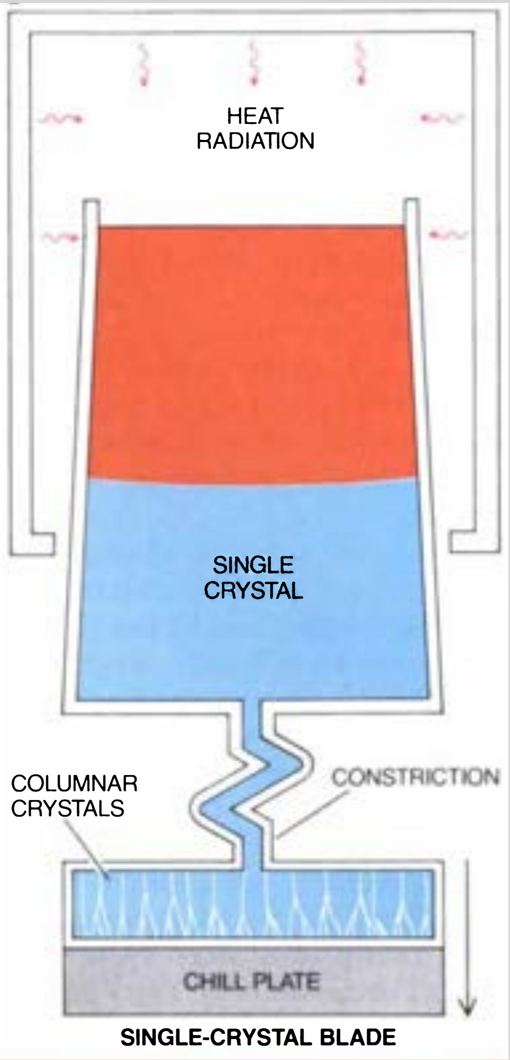
Manufacturing single crystals

Current approach

- Directional solidification with “bottleneck”
- Complicated process which requires optimization over several parameters

Proposed approach

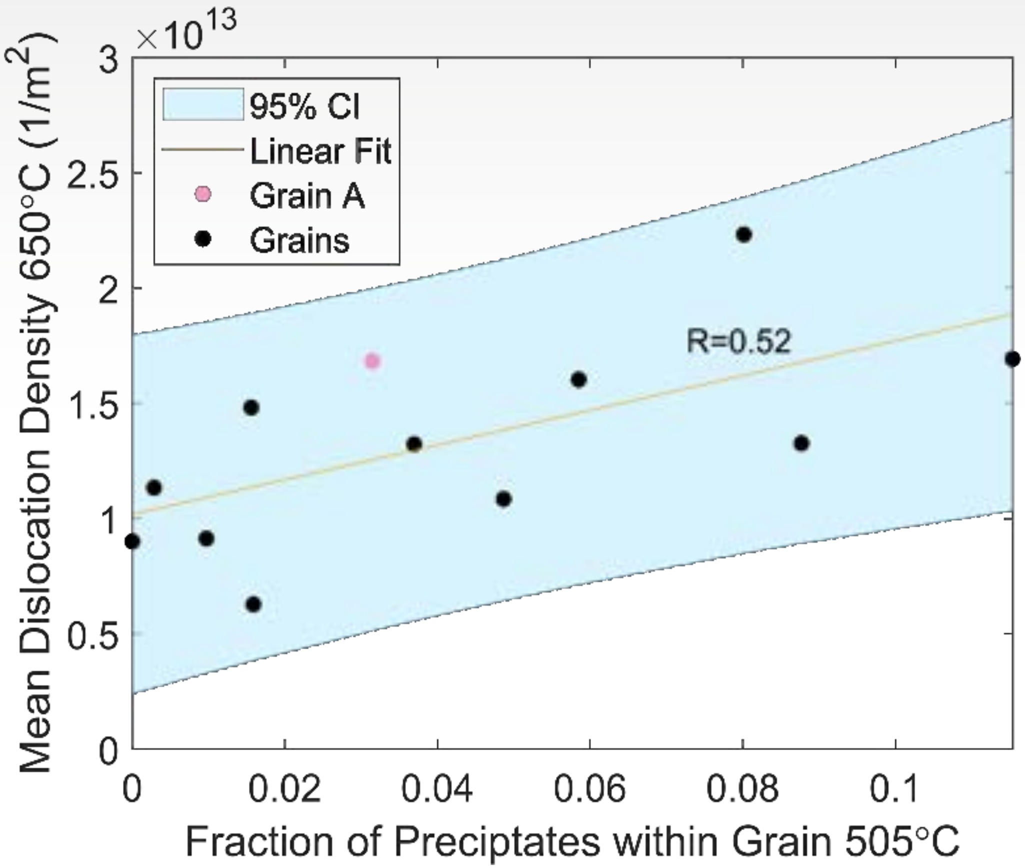
- Solid-state conversion involving particle-stimulated abnormal grain growth
- Generally lower capital costs since only simple equipment is needed



B.H. Kear, *Scientific American,* **255**, 159 (1986).

Effect of non-uniform precipitation

- Plot grain-averaged dislocation density *versus* FCC-α particle density at preceding temperature point
- Dislocations generated by semi-coherent α-β interface upon dissolution of α phase
- **Grains with high particle density have correspondingly high dislocation density**



M. Higgins et al., K Thornton, A.J. Shahani, in review (2021).