



Managing the Mosaic of Microstructure

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Final Report

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14. ABSTRACT The MURI program entitled 'Managing the Mosaic of Microstructure' was a joint research project between groups from seven major universities: Carnegie Mellon University (lead institution), CalTech, Georgia Tech, Northwestern University, Purdue University, University of Michigan, and the University of Minnesota. This research program created break-through concepts and methodologies for elucidating the micro-structure-properties link to enable materials design by bringing together cuttingedge theories and techniques from materials science, mathematics and information science -- many of which are unknown to researchers outside individual disciplines. The main objectives of our research plan were stated in terms of three Grand Challenges (GCs) around which our efforts were built: GC1: to establish a standardized methodology, grounded in sound mathematics, for acquiring, storing, analyzing, modeling, and querying ``beyond 3-D'' materials data, taking full account of the potential sparsity of such data as well as the associated uncertainties and variabilities;</p> <p>GC2: to employ advanced stochastic/probabilistic models that allow not only for the description of the ``average'' microstructure, but also for the inclusion of rare events (large deviations), and to set up the proper data structures to enable such a stochastic description;					
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MURI: Managing the Mosaic of Microstructure

Final Project Report

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1 Introduction

The MURI program entitled *Managing the Mosaic of Microstructure* started officially on September 30, 2012 and ended (with a no-cost extension) on November 30, 2018. The program was a joint research project between groups from seven major universities: Carnegie Mellon University (lead institution), Cal-Tech, Georgia Tech, Northwestern University, Purdue University, University of Michigan, and the University of Minnesota. BlueQuartz Software acted as a software consultant on the project.

This research program aimed to create break-through concepts and methodologies for elucidating the microstructure-properties link to enable materials design by bringing together cutting-edge theories and techniques from materials science, mathematics and information science – many of which are unknown to researchers outside individual disciplines. The main objectives of our research plan were stated in terms of three *Grand Challenges* (GCs) around which our efforts were built:

- **GC1:** *to establish a standardized methodology, grounded in sound mathematics, for acquiring, storing, analyzing, modeling, and querying “beyond 3-D” materials data, taking full account of the potential sparsity of such data as well as the associated uncertainties and variabilities;*
- **GC2:** *to employ advanced stochastic/probabilistic models that allow not only for the description of the “average” microstructure, but also for the inclusion of rare events (large deviations), and to set up the proper data structures to enable such a stochastic description;*
- **GC3:** *to employ advanced data mining approaches, constrained by accurate mathematical models and accounting for variability, to instantiate large numbers of digital microstructures to search for an optimal microstructure and its process path, to achieve a desired property combination.*

The first two challenges required the creation of several new methodologies, and the third challenge represented an overarching goal of the research program. The ability to predict what a microstructure should look like in order to achieve a given property or performance would be transformational to the field of materials science and engineering.

To address these three Grand Challenges, we implemented a multi-pronged approach divided into three main thrust areas:

- **Thrust Area 1** – [T1] **Representation** of structure and time evolution in microstructures: What can we measure and how can we compactly represent microstructure?
- **Thrust Area 2** – [T2] **Mathematical quantification** of microstructures for bridging scales: What information about a microstructure is relevant for its properties?
- **Thrust Area 3** – [T3] **Multi-scale materials design** using informatics: How can we use microstructure information for design?

The thrust areas were selected so that each team member, through his/her own relevant expertise, could contribute to addressing each of the three Grand Challenges; they were also formulated to be consistent with discovery-based university style research. The MURI project produced a variety of deliverables, some in the form of algorithms, some in the form of mathematical models, and others in the form of methodologies or frameworks for microstructure design and microstructure-property relations modeling:

— *algorithms for:* optimal design of magnetoelastic and energy conversion devices; optimization of multiferroic microstructure; reconstruction and generalized forward projectors for focused ion beam serial sectioning, vector field electron tomography, and time-dependent X-ray computed tomography; reconstruction code capable of detecting/identifying rare events; extraction of accurate interface curvatures from 3D time-dependent data sets; data storage and retrieval, compatible with high-end data mining approaches; optimal sampling of microstructure space; robust importance samples for stochastic modeling of rare events.

— *mathematical models for:* microstructure-property relations in multiferroics; time-dependent tomographic reconstructions; thin-manifold description of microstructures; probabilistic graphical model adapted to materials microstructures and data acquisition;

— *methodologies/frameworks for:* microstructure-sensitive design in multiferroic alloys, Ni-based superalloys and α - β Ti alloys; robust process design of polycrystalline materials for customized performance; experimental validation of process designs.

In the remainder of this document, we will present a few statistics related to the program, followed by a

description of 10 selected joint research results. The document concludes with a list of all publications that acknowledge MURI support.

1.1 Program Participants

During the six years of this program, a total of 30 graduate students and 9 post-doctoral researchers participated in various research projects. Table 1 lists all students and post-docs along with their current occupation, if known.

Graduate Students	
Ruoqian Liu	Research Scientist at Uber AI Labs
Arindam Paul	EECS, Northwestern University (expected to graduate end of 2019)
Ahmet Cecen	Exxon-Mobil Chemicals, Houston TX
<i>Yuksel Yabansu</i>	Research Engineer, Georgia Tech
Xian Chen	Assistant Professor, Hong Kong University of Science and Technology
Yintao Song	Principal Software Engineer, Lattice Engines, San Mateo, CA
Amartya Banerjee	Assistant Professor, Materials Science and Engineering, UCLA
<i>Vivekanand Dabade</i>	Postdoctoral Fellow, Ecole Polytechnique
Abhishek Kumar	Asst Prof at Northeastern University
Pinar Acar	Asst Prof at Virginia Tech
Siddhartha Srivastava	Current PhD student at U Michigan
Michael Chapman	Air Force Research Laboratory (Dayton OH)
<i>Saransh Singh</i>	Post-doctoral researcher (CMU)
Ashwin Shahani	Assistant Professor (U. Michigan)
Yue Sun	JP Morgan Chase
Chun-Jen Hsueh	Currently working at Amazon 126
Dingyi Sun	postdoc at Brown University
Jin Yang	Currently a post-doc at University of Wisconsin at Madison
Md. Zubaer Hossain	Currently Assistant Professor at University of Delaware
G. M. Dilshan Godaliyadda	Texas Instruments
Suhas Sreehari	Wells Fargo
<i>Kadri Aditya Mohan</i>	Lawrence Livermore National Laboratory
<i>Singanallur V. Venkatakrishnan</i>	Oak Ridge National Laboratory
Maliha Hossain	Graduate Student Purdue University
Soumendu Majee	Graduate Student Purdue University
Benjamin Foster	Lockheed Martin Corporation
Emma Reid	Graduate Student Purdue University
Huixi Zhao	Technical Lead, Zoom Video Communications
Dae Woo Kim	Programmer Analyst, Washington University at St. Louis
Camilo G Aguilar Herrera	Current PhD student, Purdue University
Tianyu Li	Current PhD student, Purdue University
Postdoctoral Researchers	
Vijay Srivastava	Lead Materials Scientist, GE Global Research
Amy Wang	(The Netherlands)
Farangis Ram	Carnegie Mellon University
Emine Begum Gulsoy	Northwestern University
Gal Shmuel	Currently Assistant Professor at Technion, Israel
Stella Brach	Continuing as a postdoc at Caltech
Dong Hye Ye	Assistant Professor, Marquette University

Table 1: Graduate students and post-doctoral researchers who participated in the MURI program, along with their current employer (if known). Students in italics continued as post-doctoral researchers after graduation, students/post-docs in bold became faculty members.

1.2 Reviews and Team Meetings

1.2.1 Program Review Meetings

The MURI program held annual review meetings at a number of locations (Table 2); details of the presentations given at those reviews can be found on the MURI website.

Meeting	Dates	Locations
MURI Kickoff Meeting	10/18-19/2012	Carnegie Mellon University
Sayir Portfolio Review Presentation	2/18/2013	Arlington, VA
MURI Review 1	1/21-22/2014	Dayton, OH
Sayir Portfolio Review Presentation	5/13/2014	Arlington, VA
MURI Review 2	6/22-23/2015	Arlington, VA
Sayir Portfolio Review Presentation	5/21/2015	Arlington, VA
Final Program Review	12/18-19/2017	Arlington, VA

Table 2: Dates and locations of all official meetings associated with MURI kickoff, annual reviews and final review.

1.2.2 Team Summer Meetings

The MURI team organized four work meetings at different host institutions. The purpose of these meetings was two-fold: (1) to make sure that the graduate students and post-doctoral researchers from different groups would get to know each other, and (2) to present recent research results to the complete group, to facilitate “cross-pollination” across research fields. The meeting locations and dates are listed in Table 3.

Meeting	Dates	Locations
Team Meeting 1	7/1-2/2013	Northwestern University
Team Meeting 2	8/14-15/2014	Purdue University
Team Meeting 3	8/17-18/2015	Carnegie Mellon University
Team Meeting 4	8/22-23/2016	CalTech

Table 3: Dates and locations of all Summer MURI Team Meetings.

1.3 Scientific Advisory Committee

The MURI program benefitted from the input from our Scientific Advisory Committee, consisting of:

- Jeff Simmons (AFRL);
- Tresa Pollock (UCSB);
- Shlomo Ta’asan (CMU);
- David Furrer (PW);
- Peter Chung (UMD).

The committee provided detailed written feedback, in particular early on in the program during the first two years, that helped us focus our research efforts into areas that would benefit strongly from multi-disciplinary approaches, instead of more individual efforts.

1.4 Team Member Awards and Recognitions

- ***S. Kalidindi***: International Khan Plasticity Medal, 2019; Vannevar Bush Faculty Fellow, 2018; Vajra Faculty, Department of Science and Technology, India, 2018; Advisor of the Best Doctoral Dissertation 2018, Sigma Xi, Georgia Institute of Technology Chapter; First place winner in the national 2016

Materials Science and Engineering Data Challenge along with Joshua Gomberg and Andrew Medford; Runner up winner in the national 2016 Materials Science and Engineering Data Challenge along with Eva Popova, Theron Rodgers, Xinyi Gong, Ahmet Cecen, and Jonathan Madison; Elected Fellow of TMS, 2015; Alexander von Humboldt Award, 2013.

- **R. James:** Simons Fellow, Issac Newton Institute for Mathematical Sciences, Cambridge, 2018; Theodore von Karman Prize, SIAM, 2014; Department of Mechanical Engineering Distinguished Lecture, Colorado School of Mines, 2015; Plenary Lecture, ESOMAT 2015 (Antwerp); Panorama of Mathematics Lecture (Hausdorff Institute: Bonn), 2015; Keynote Lecture: 2nd Midwest Workshop on Mechanics of Materials and Structures (Northwestern), 2016; Timoshenko Lecture (Stanford University), 2017; Golden Medallion Lecture, College of Science and Engineering (UMN), 2017; Plenary lecture: SMST-2017, San Diego; Distinguished Lecture at IAS, Hong Kong University of Science and Technology, 2017; Invited lecture, International Congress of Mathematicians, Rio de Janeiro, 2018; The Hooke Public Lecture (Oxford University, https://www.youtube.com/watch?v=IBvHw_5mlRc), 2018; Plenary: AMS Current Events Bulletin, Joint Mathematics Meetings, San Diego, 2018; Tedori-Callinan Distinguished Lecture, Department of Mechanical Engineering, University of Pennsylvania, 2018.
- **K. Bhattacharya:** Distinguished Alumnus Award, Indian Institute of Technology, 2019; Outstanding Achievement Award, University of Minnesota, 2018; Warner T. Koiter Medal for distinguished contributions to the field of Solid Mechanics, American Society of Mechanical Engineering, 2015.
- **V. Sundararaghavan:** 2018 AFRL Summer faculty fellow; 2017 Aerospace Engineering Department Award, University of Michigan Ann Arbor; Pinar Acar: Amelia Earhart Fellow 2016; Abhishek Kumar: Ivor McIvor Award for Applied Mechanics, 2014; Richard and Eleanor Towner Prize for Distinguished Academic Achievement, 2014
- **A. Agrawal and A. Choudhary:** Ankit Agrawal and Alok Choudhary, Best Paper Finalist, HiPC 2017; Alok Choudhary, Best Tech Manager, Third Annual Timmy Awards, 2017; Ankit Agrawal and Alok Choudhary, Best Paper Award at IEEE Cluster 2016; Alok Choudhary, BITS Pilani Distinguished Alumni Award, 2016; Ruqian Liu, EECS/TGS conference travel grant for KDD 2016; Arindam Paul, EECS/TGS conference travel grant for KDD 2016; Ruqian Liu, Runner-up, poster presentation at EECS Student Poster Fair Award, 2015; Ankit Agrawal and Alok Choudhary, Outstanding Paper Award awarded by Tata Consultancy Services for joint work published in IMMI, 2014; Ruqian Liu, Runner-up, poster competition at the Symposium of Multidisciplinary Computer-Aided Design and Simulation-Based Optimization - Recent Applications & Future, Evanston IL, December 2014.
- **C. Bouman:** Showalter Professor of Electrical and Computer Engineering and Biomedical Engineering; Member of the National Academy of Inventors; Fellow of the IEEE, a Fellow of the American Institute for Medical and Biological Engineering (AIMBE); Fellow of the society for Imaging Science and Technology (IS&T); Fellow of the SPIE professional society; 2014 Electronic Imaging Scientist of the Year award; IS&Ts Raymond C. Bowman Award; Editor-in-Chief for the IEEE Transactions on Image Processing; Distinguished Lecturer for the IEEE Signal Processing Society; Vice President of Technical Activities for the IEEE Signal Processing Society; Lead creation of the IEEE Transactions on Computational Imaging; Vice President of Publications and a member of the Board of Directors for the IS&T Society; Founding and Co-Chair of the IS&T conference on Computational Imaging.
- **P. Voorhees:** Moore Distinguished Scholar, California Institute of Technology, 2018; Member, American Academy of Arts & Sciences, 2016; Fellow, The Minerals, Metals and Materials Society, 2013; J. Willard Gibbs Phase Equilibria Award ASM International, 2013
- **M. De Graef:** 2016 DoD Vannevar Bush Faculty Fellowship (formerly known as the National Security Science and Engineering Faculty Fellow program); Saransh Singh: Paxton Award for Best Doctoral Dissertation, Carnegie Mellon University, 2018; Best paper Award (Software and Instrumentation), Microscopy and Microanalysis, 2018; Cover Article, Microscopy and Microanalysis, 2018; Student Scholar Award, Microscopy and Microanalysis, 2017; Student Award, EBSD Meeting, May 2016; Bertucci Fellowship, Carnegie Mellon University, 2016.

Selected Joint Research Results

The final program review took place in Arlington, VA, on December 18-19, 2017 and consisted of joint presentations by the co-PIs. The main body of this report (sections 2 through 11) mimics the structure of the final review and provides 10 sections describing important results from our joint research, i.e., research that would likely never have happened if it were not for this MURI program. The final list of publications that acknowledge MURI support can be found in reverse chronological order at the end of this report starting on page 58.

2 Microstructure in Three and Four Dimensions

C. Bouman, M. Comer, P.W. Voorhees

Abstract: Using new signal processing algorithms solidification structures from dendrites to irregular eutectic growth structures are investigated. The combination of state-of-the-art algorithms and high brightness X-ray synchrotron radiation has allowed unprecedented insights into the dynamics of solidification processes. We present a few vignettes of this groundbreaking research.

Dendrites exist almost everywhere, from industrial metal castings to Li metal anodes in Li-ion batteries. Not only are dendrites ubiquitous, they also have a profound effect on the properties of materials; for example, their morphology has a major influence on materials properties as a result of the coupling between the dendrite arms and the chemical non-uniformity of the solid and in Li-ion batteries, dendritic growth is a major failure mechanism. This combination of prevalence and influence on material performance means that the dendrite growth kinetics and morphology have profound technological implications. In addition, dendrites are also an example of an out-of-equilibrium pattern forming system. Here the focus is on understanding the processes underlying the formation of the complicated dendrite morphology from a featureless liquid.

Far from equilibrium, solids may crystallize into highly ordered patterns, or eutectics, of remarkable complexity. Eutectic systems are ubiquitous in nature, and have been discovered in a vast array of organic, metallic and semi-metallic alloys. Such materials can exhibit outstanding mechanical and electrical properties because their microstructures act as natural or in situ composite materials. To tune the eutectic microstructures to technological demands, we must understand the fundamental processes underlying their formation from a featureless liquid. An exploration of this crystallization process has the potential to provide the models necessary to accelerate the design of new advanced alloys, thereby fulfilling the promise of the Materials Genome Initiative. The eutectic morphologies that may arise during solidification can be classified as either regular or irregular. Regularity refers to the periodic arrangement of lamellae, and is typical of non-faceted systems. The situation is more complex when irregular eutectics are considered, in which one of the phases is faceted (for example, Si and Ge) and the other non-faceted (for example, Al and Ag). The faceted phase does not easily change direction due to its atomic structure, covalent bonding and defect-mediated growth mechanism. Due to this inherent a “stiffness” of the faceted phase, the microstructure is non-periodic with varying interphase spacing.

While great insights have been gained into the structure and crystallography of irregular eutectics and dendritic growth, their interfacial dynamics have remained an area of great interest. Previous studies have been limited to “quench-and-look” experiments wherein a completely solid alloy sample is analysed post mortem. However, it is well known that the quenching needed to convert the liquid to solid can distort the morphology of the solid-liquid interface from that present during crystallization. Thus, it is only through in situ experiments that interfacial dynamics can be tracked with a high degree of precision. To circumvent these challenges, these processes have been investigated via optical microscopy where solidification can be observed using transparent organic films sandwiched between two glass slides. Yet many details of the microstructural evolution remain unclear because of the effects of the constraint imposed by the thin film on an inherently 3D phenomenon. Furthermore, the ability of organic materials to accurately mimic the growth process of faceted phases has not been ascertained.

For these reasons, we employ a fully 4D analysis in order to investigate the dynamics of irregular eutectic and dendritic growth. X-ray microtomography (XRT) is a nondestructive method to determine the evolution of a microstructure in 3D and as a function of time. In this work, we have succeeded in tracking in situ the growth dynamics of an irregular eutectic alloy and dendritic growth via synchrotron based XRT. The predominant challenge with the tomographic imaging of eutectic solidification is associated with the length scale of the lamellae, as well as the rate at which these lamellae evolve in time.

To this end, we meet the necessary spatial and temporal resolution requirements by using the time-interlaced model-based iterative reconstruction methodology, described by Mohan and coworkers [26, 21, 22]. In this research, we introduced a time-interlaced model-based iterative reconstruction (TIMBIR) method, which is a synergistic combination of two innovations. The first innovation, interlaced view sampling, is a novel method of data acquisition, which distributes the view angles more evenly in time. The second

innovation is a 4-D model-based iterative reconstruction algorithm (MBIR), which can produce time-resolved volumetric reconstruction of the sample from the interlaced views. In addition to modeling both the sensor noise statistics and the 4-D object, the MBIR algorithm also reduces ring and streak artifacts by more accurately modeling the measurement non-idealities. Our experimental results on both simulated and real X-ray synchrotron data indicate that TIMBIR can improve temporal resolution by an order of magnitude relative to existing approaches.

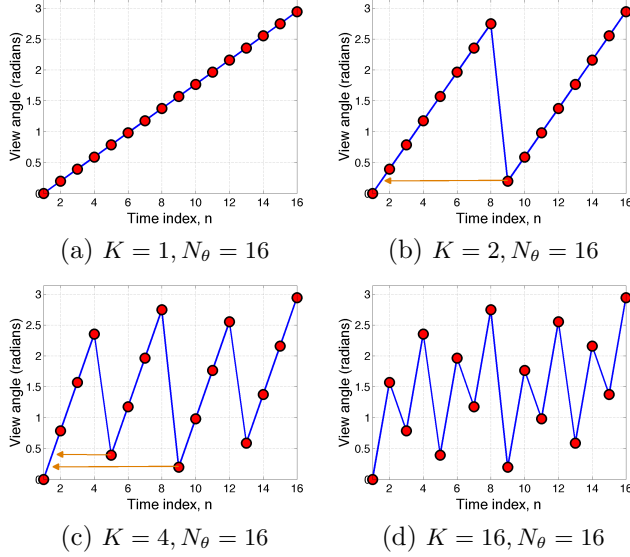


Figure 1: Illustration of interlaced view sampling pattern for different K values. (a-d) show $\theta_n \bmod(\pi)$ vs. time index, n , for $K = 1, 2, 4$, and 16 . The arrows show the relative difference between the angular values across sub-frames.

ble quality to what would be possible with traditional

Using TIMBIR, the growth of Al dendrites in an Al-Cu liquid alloy was examined. We examined the evolution of the complex morphology of the secondary dendrite arm structure of a dendrite growing into a supersaturated liquid. The sample evolution during solidification at a few times is shown in Fig.2.

A single dendrite forms on the side of the sample with primary arms growing out across the sample and four arms growing near the sample walls. By symmetry, all of these arms are growing in the $\langle 100 \rangle$ direction, as are the secondary and tertiary arms. The evolution of the volume fraction of the solid phase (f_s), shown in Fig.2d, indicates that the sample is significantly undercooled when nucleation occurred, leading to a rapid rise in the solid fraction to just over 20% solid by volume. At this point, the supersaturation is exhausted and further solidification is driven by the cooling rate of the furnace. The datasets that are analyzed in this work are in the solute supersaturation-driven growth regime, which closely matches with theoretical and simulation work on dendrite evolution behavior.

In order to satisfy the spatial Nyquist sampling requirement for each 3D time sample of a 4D reconstruction, it is typically necessary to collect approximately $N_\theta = N_p$ progressive views, where N_p is the number of sampled pixels perpendicular to the axis of rotation. In the traditional approach, these N_θ progressive views are taken in sequence while the sample is rotated continuously over π radians. The object is then reconstructed at a temporal rate of $F_s = F_c/N_\theta$ where F_c is the data acquisition rate.

In contrast to this traditional approach, TIMBIR uses an interlaced view sampling method where each frame of data consists of N_θ distinct views that are acquired over K interlaced sub-frames (see Fig. 1). Each sub-frame of data then consists of N_θ/K equally spaced views, but together the full frame of data contains all N_θ distinct views of the object. The key innovation is that by collecting the data in this interlaced format and using MBIR reconstruction, it is possible to reconstruct at a temporal frequency of $F_s = KF_c/N_\theta$, which is K times higher than would normally be possible. In typical experimental scenarios, we have used $K = 16$ to achieve $16\times$ the temporal resolution with compara-

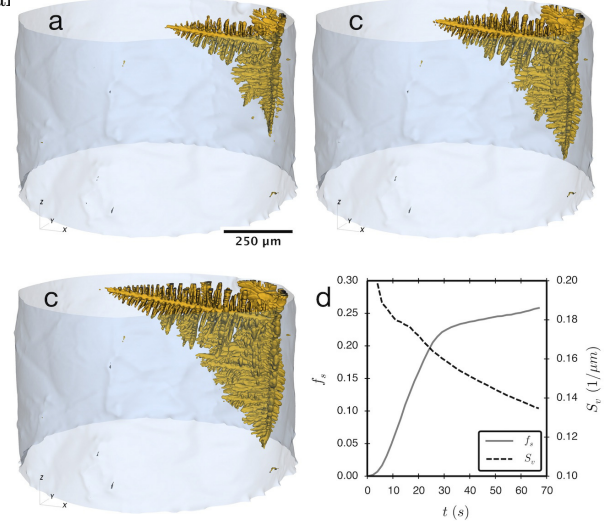


Figure 2: Morphology of the full sample at (a) 5.8, (b) 7.6 and (c) 9.4 seconds after the start of solidification. The dendrite that is growing out into the sample is the one that is isolated and analyzed. Solid fraction (f_s) and interfacial area per unit volume (S_v) are shown in (d).

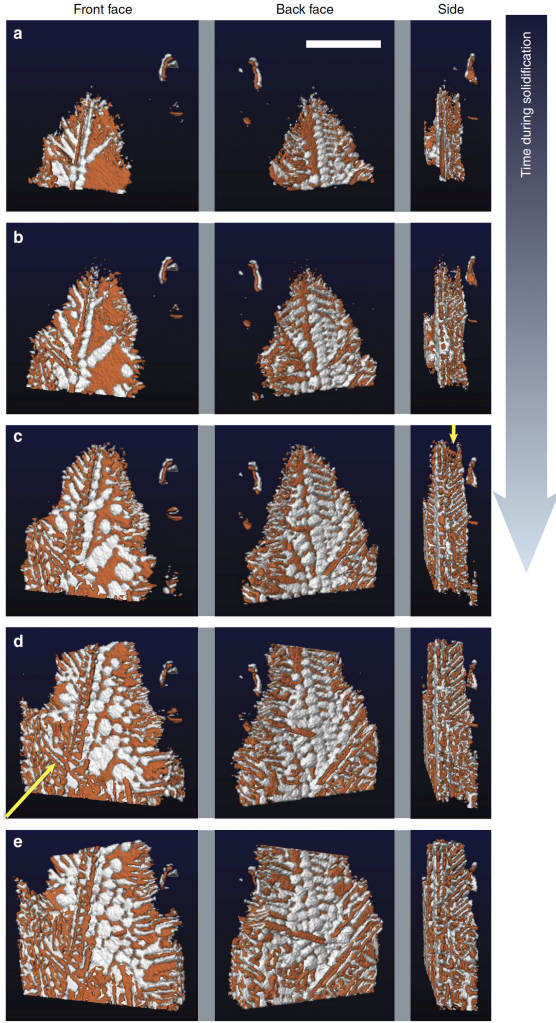


Figure 3: Morphology of the eutectic colony during the growth process. Reconstructions given at (a) 100, (b) 140, (c) 180, (d) 220 and (e) 260 s after the start of solidification. Shown are three views per time-step, corresponding to the front (0°), back (180°) and side (90°) of the eutectic colony. The arrow in the side view of c points to the tips of the Ge plates that lead the solidification event; the arrow in the front view of d points to another eutectic colony that impinges upon the one of interest. When viewed from either the front or the back, the interfacial morphology is markedly different from that seen in transparent organic materials: bulbous-like domains of Al envelope the surfaces of Ge as solidification proceeds. Scale bar, 100 μm .

existing models for irregular eutectic growth are fully adequate for describing the rich variety of anisotropic patterns that arise during crystallization. We find that defects play a critical role in the growth of the eutectic, in which pockets of metal engulf the exposed facets and control the overall growth rate of a eutectic colony. These experimental results identify a new mechanism for the growth of an irregular eutectic, and provide the key insights needed to model the crystallization of these technologically complex materials.

The surface area per unit volume of solid phase (S_v) is also shown in Fig.2. This quantity is inversely proportional to the average feature size in the system; thus, the decreasing S_v is indicative of the average feature size increasing. The decrease, however, is only about 30% of the value after 5 seconds of solidification; in comparison, the volume fraction of solid has increased by nearly a factor of 2.5. So, while the solid-liquid interfacial area increases dramatically during solidification, the volume fraction increases as well leading to a small change in S_v . The morphology of the dendrite has been quantified in a manner appropriate for comparison to future simulations [26]. We find a lack of self-similarity of the structure, power laws for the volume fraction transformed, surface area and surface area per volume that are different from those seen in transparent organic materials, and a markedly different side branch structure from that seen in transparent organic materials. The presence of tip splitting of the secondary arms implies that solute segregation should be present at levels that exceed those given by estimates of the secondary arm spacing.

We have investigated irregular eutectic alloy growth via 4D XRT, see Fig.3 which shows the morphology of the eutectic colony at five representative time-steps during the growth process [29]. As each time-step, we view the colony from its front (0°), back (180°) and side (90°). While plates of Ge are evident at the earlier stages of growth, bulbous-like domains of Al envelope its surfaces at longer times. One might suppose that these bulbs of Al impede the growth of the colony in the lateral (horizontal) direction, since the Ge crystals cannot grow through the solid metal layer. However, even before the emergence of the outermost bulbs of Al, the colony extends very slowly in the lateral direction. In fact, throughout the growth process, interfacial velocities are roughly 10 times lower in the lateral direction compared with the transverse (vertical). Any non-zero growth rate in the lateral direction at long times may be due to the growth and coarsening of the outermost Al bulbs, since the Ge plates do not become appreciably thicker. Through these experiments we have resolved a longstanding controversy in the field, and demonstrate that none of the

Stochastic Models for Microstructure Segmentation

Precise boundary information helps to characterize many important materials properties that depend on interfaces, boundary curvature, or topology. However, imaging noise and low contrast sometimes cause nearby boundaries to merge, hence creating a challenging task in the segmentation of microstructural elements such as dendrites.

Our work presented in [24] uses geometric representations for narrow gaps, or channels, in a stochastic modeling framework to detect channels in an image, and uses this information to correct merging defects in segmentation. Our approach uses a Marked Point Process (MPP) framework to detect the position, width, length and rotation of each channel. Compared to the conventional Markov random field model, which is defined on a fixed lattice, the advantage of using an MPP is that it can model randomly-located objects and their spatial relations.

Marked Point Process

A 2D marked point process $\mathbf{X} = \{X_1, \dots, X_n\}$ defined on space a $S = M \times K$ is a point process, where the positions of the points are defined on $K = [0, I_{width}] \times [0, I_{height}]$, a bounded subset of $\mathbb{R}^2$, and each point has associated with it a collection of random variables, known as marks, defined on a space M . Each marked point consists of a random spatial location and corresponding random variables that describe the shape of an object, such as an ellipse, a rectangle, or a line segment located at that spatial location. For example, if an image is composed of randomly located circular particles, we can model this image with the radius and locations and radii of all of the circles in the image. In this case, the i th marked point can be expressed as $X_i = (S_i, N_i)$, where S_i is the location and the mark N_i represents the radius of the circle object.

The probability density function of an MPP is given by the Gibbs distribution as:

$$p(\mathbf{x}) = \frac{1}{z_{mpp}} \exp(-U(\mathbf{x})) \quad (1)$$

where $U(\mathbf{x})$ is the Gibbs energy of a configuration $\mathbf{x}$ and z_{mpp} is a normalizing constant. To describe a density $p(\mathbf{x})$ conditioned on image data $\mathbf{y}$, Bayesian modeling is used, so

$$p(\mathbf{x}|\mathbf{y}) \propto p_{likelihood}(\mathbf{y}|\mathbf{x})p_{prior}(\mathbf{x}) \quad (2)$$

Then a Gibbs energy can be described as:

$$U(\mathbf{x}|\mathbf{y}) \propto U_D(\mathbf{y}|\mathbf{x}) + U_P(\mathbf{x}) \quad (3)$$

where $U_D(\mathbf{y}|\mathbf{x})$ is a data energy and $U_P(\mathbf{x})$ is a prior energy. The term $U_D(\mathbf{y}|\mathbf{x})$ can be further expanded as a sum of individual potentials over n objects in a configuration.

$$U_D(\mathbf{y}|\mathbf{x}) = \sum_{i=1}^n V_d(\mathbf{y}|x_i) \quad (4)$$

$V_d(\mathbf{y}|x_i)$ is a data potential for an individual object. If $V_d(\mathbf{y}|x_i) \leq 0$, the object x_i is a favorable candidate, since it increases the density $p(\mathbf{x}|\mathbf{y})$. For the prior energy $U_P(\mathbf{x})$, we use only single-object and two-object cliques. Then the prior energy can be expressed as:

$$U_P(\mathbf{x}) = \sum_{i=1}^n V_p(x_i) + \lambda_{int} \sum_{x_i \sim x_j} V_{int}(x_i, x_j) \quad (5)$$

$V_p(x_i)$ is a prior potential for an individual object, and $V_{int}(x_i, x_j)$ is an interaction potential, where a neighborhood relation $\sim$ is defined between a pair of objects and λ_{int} is a parameter to control the amount of contribution of the interaction potential to the prior energy.

The first microscope sample image is a René88 DT compound. The size of this image is 194×149 pixels. Figure 4 shows the results of applying the curved channel method. For this experiment, we set $\lambda_o = 34$, $\omega_f = 20$, $\omega_s = 1$, $\omega_d = 2$, $\omega_c = 8$, $\omega_{eo} = 1$.

We apply the channel information to improve segmentation results. To do this, we control the interaction parameter value in the EM/MPM segmentation algorithm according to the channel configuration. The main idea is that the energy cost for connecting regions found to be separated by a channel is increased during the segmentation procedure, to avoid merging dendrites separated by a channel. Figure 4 shows a comparison of the original EM/MPM and the EM/MPM after detecting channels and adapting the segmentation accordingly. These figures show that the modified EM/MPM corrects many bridged channel defects.

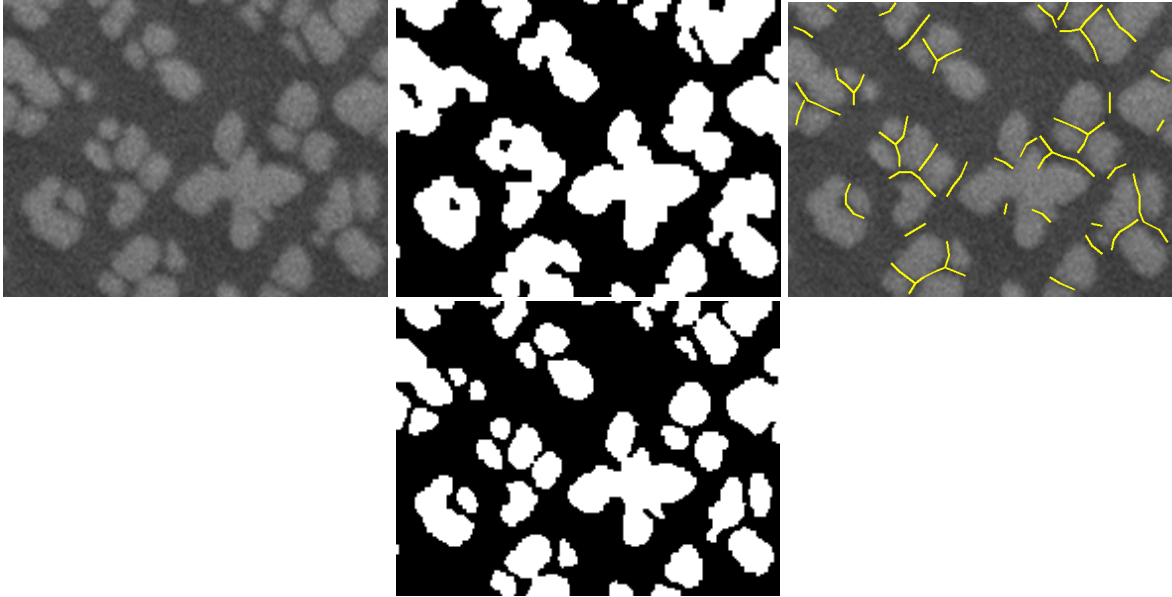


Figure 4: Results for sample image 1, René88 DT Compound: (a) Test image, (c) Original EM/MPM, (g) Curved channels (running time 40sec), (h) $\tilde{\beta}(1, a, b)$ of curved channel, (i) EM/MPM after detecting curved channels, Interaction parameters of all the EM/MPM results are set as 2.9 , which was determined experimentally to give a reasonable trade-off between noise and channel bridging.

The image presented in Figure 5 represents a slice of Pb-Sn volume obtained during a coarsening process. Its size is 398×528 pixels. Figure 5 shows that our method successfully models the gaps between neighboring dendritic arms and shows the improved segmentation after extracting information from the detected channels.

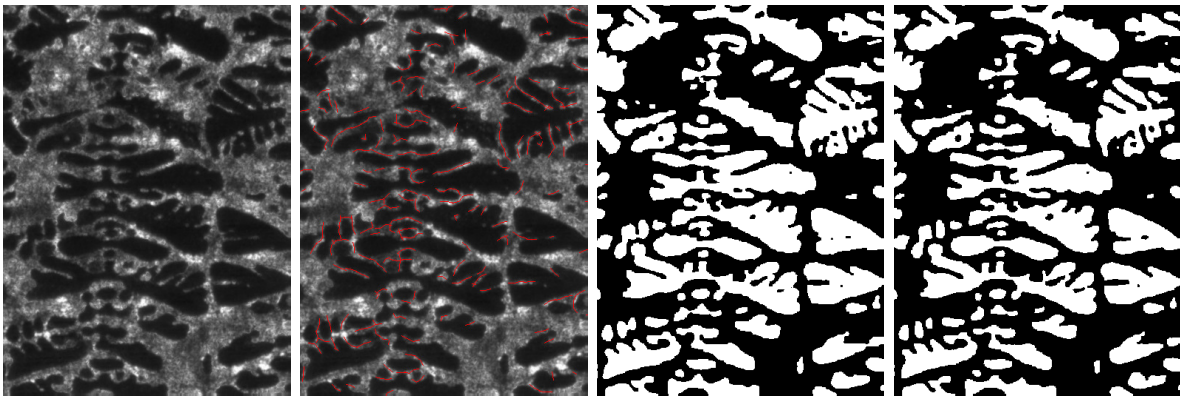


Figure 5: Results for sample image 3, Pb-Sn: (a) Test image, (b) Curved channels (c) Original EM/MPM, (d) EM/MPM after detecting curved channels. Interaction parameters of all the EM/MPM results are set as 2.8.

3 Two-Point Statistics of Microstructures

S.R. Kalidindi and P.W. Voorhees

Abstract: We extend the existing framework of two-point spatial correlations to allow for the quantification, analyses, interpretation and visualization of microstructure coarsening measured by time-resolved X-ray computed tomography. Specifically, extensions were made to facilitate (i) the incorporation of non-conventional local attributes such as solid-liquid interface, interface curvature, and interface velocity in the description of the local state, and (ii) the efficient computation of bulk spatial correlations when the local attributes are sparsely defined only at special locations in the three-dimensional volume (e.g., solid-liquid interfaces). We have explored multiple variants of spatial correlations, including Pearson correlation coefficients and two-point joint probabilities, and examined their relative merits in providing useful new insights into the coarsening process. Algorithmic enhancements needed to carry out these computations on the large datasets produced in the experiments are also described. The results demonstrate the remarkable ability of these new protocols in automated (unbiased) capture of the fourfold symmetry of the dendritic microstructure, and in providing quantitative and reliable estimates of the characteristic lengths associated with the dendritic microstructure (including the secondary and tertiary dendrite arm spacings, secondary dendrite arm diameter, and the solute diffusion length). These estimated quantities agree well with the direct measurements from the microstructure. The results also indicate that interfaces with high negative and near-zero mean curvature have long range spatial auto-correlations, whereas all values of the interfacial normal velocity are only auto-correlated in the short range in space.

Microstructures, which arise from inhomogeneities in composition or structure, are ubiquitous in materials ranging from condensed matters, to hydrodynamic systems, and to biological soft matters. Microstructures of materials can often have deterministic impacts on the properties of the materials on the macro-scale. Dendrites are one of the most frequently seen microstructures during solidification of metals and alloys. The morphologies of these dendritic microstructures can strongly affect the physical, chemical and mechanical properties of the subsequent products of many metallurgical technologies, such as casting. This makes the study of the evolution of dendritic microstructures a crucial part in predicting and controlling the properties of metallic materials.

However, like many other microstructures present in nature, dendrites are complex structures. They have irregular shapes, and are often highly interconnected. Moreover, as most real problems demand three-dimensional or even four-dimensional (including time) considerations, it makes it practically difficult to visualize the data, and even more so to analyze it. As a result, two-dimensional sections of the data are usually made in order to obtain salient attributes of the three-dimensional dataset, such as the secondary and tertiary arm spacings of the dendrite. As the system size grows larger, the dataset may become unwieldy to handle as a whole, and hence data processing and interpretation must be done in a piece by piece manner. This adds significantly to the challenges in arriving at meaningful global (bulk) descriptors of the microstructure that are also of high value in correlating to its performance characteristics.

A rigorous statistical framework that can capture efficiently and systematically the directionally resolved higher-order microstructural features (i.e., spatial correlations) is necessary for aggregating and curating the core materials knowledge in the form of processing-structure-property (PSP) linkages. Towards this end, we develop and demonstrate for the first time, a new and efficient set of protocols for the computationally efficient quantification of the spatial correlations between microstructural features defined on the interfaces [41]. These new protocols are built on the existing framework of two-point statistics that have been used previously to quantify the spatial correlations between local attributes defined in the bulk of the microstructure (as opposed to the attributes defined only on the interfaces in this work). Additionally, the new protocols are demonstrated using a scalable algorithm suitable for large and high-dimensional datasets.

In this work, we demonstrate the application of the concepts presented in the earlier sections on an experimental dataset of a dendritic microstructure in an Al-Cu alloy undergoing coarsening. The sample raw material is a directionally solidified Al-Cu alloy with 19 wt% of Cu. During the experiment, the sample was heated to 558°C (5°C above the eutectic temperature), forming a liquid-solid mixture with a constant amount of the two phases (volume fraction of liquid 0.4704). The sample was then held at that temperature, in which state the interfaces between liquid and solid can rapidly coarsen to decrease the total interfacial

area and thus total energy of the system. The isothermal condition was held for 79 min while the sample was being scanned by X-ray computed tomography (XCT).

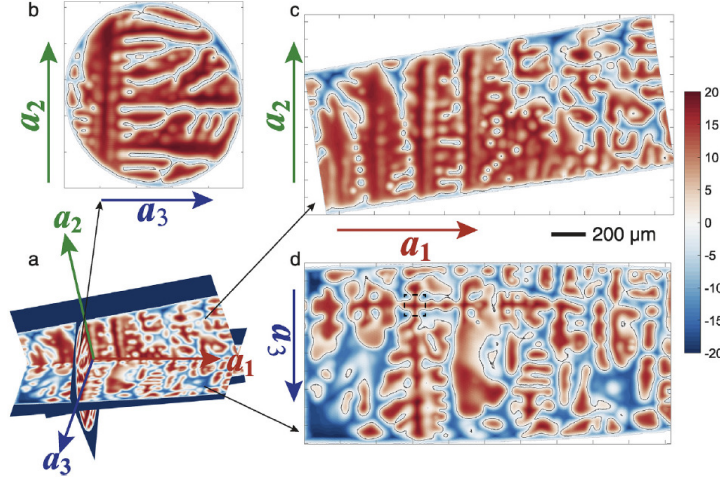


Figure 6: 2D slices of the signed distance function (SDF). The value of the SDF indicates the distance in pixels to the nearest interface, with positive values being the solid phase and negative being liquid. (a) 3D view of the 2D SDF slices. The three slicing planes are orthogonal to each other and chosen to be aligned with the primary dendrite arm (a_1), secondary dendrite arm (a_2), and tertiary dendrite arm (a_3) directions. The x , y , z are the original data axes. (bed) Enlarged 2D views of the SDF slices in (a). The black lines indicate the location of the interface (i.e. $SDF = 0$).

In order to visualize the microstructure, Fig.6 shows the signed distance function (SDF) that is used to implicitly define the interface locations. An SDF is an array of data in which the value at each voxel gives the distance to the nearest interface, with opposite signs on the two sides of the interface. In this study, we use positive values to represent voxels inside the solid phase, and negative values the liquid phase. Therefore, the interface is defined as where the SDF crosses zero. Two-dimensional slices of the SDF are shown in Fig.6 with zero value contour lines marked by black lines. Since the sample was directionally solidified along the axial direction of the cylinder, the primary dendrite arms are all aligned approximately in the axial direction of the cylindrical sample. Perpendicular to the primary arm are the secondary arms, and perpendicular to the secondary arms are the tertiary arms. This is shown in Fig.6, where the primary, secondary and tertiary arm directions are labeled as a_1 , a_2 and a_3 , respectively on the signed distance function plots. Note that the dendrite arm axes (a_1 , a_2 , a_3) are slightly different from the data axes (x , y , z). In Fig.6b and c, quaternary dendrite arms can be seen along directions roughly perpendicular to tertiary arms. However, due to the small spacing between the tertiary arms, the quaternary arms are short and tilted.

We first treat correlations with the mean curvature H and interfacial velocity V data as continuous local states and compute the two-point Pearson correlations. We present the autocorrelations of H and V and the cross correlation between H and V . All three correlation maps are shown in Fig.7 in forms of orthogonal two-dimensional slices, up to vectors whose components are smaller than 200 pixels $358\mu m$. The 2D slices shown in this figure are selected perpendicular to secondary a_2 and tertiary a_3 arm directions; compare with the sections of the microstructure volume shown in Fig.6d and c, respectively. In all of the three correlation maps ($H - H$, $V - V$, $H - V$), we see four-fold symmetric patterns, reflecting the four-fold symmetry of the dendrite arms. In the $H - H$ auto-correlation (Fig.7a,d), we see long range correlations along the secondary and tertiary dendrite arm axes (a_2 and a_3). More specifically, the two-point Pearson correlation function $H - H$ for values up to 0.2 up to a distance of $250\mu m$ along a_3 . This correlation distance is even larger for the a_2 direction, where the correlations is greater than 0.2 for distances as far as $350\mu m$. These long range correlations of the interfacial curvatures along the tertiary and secondary dendrite arms indicate that the secondary and tertiary dendrite arms are aligned parallel to each other (along the a_2 and a_3 axes, respectively), and that their interfacial shapes are similar along their respective axial directions. This can also be verified from the slices of the SDF in Fig.6. The continuous correlations come from the stems of

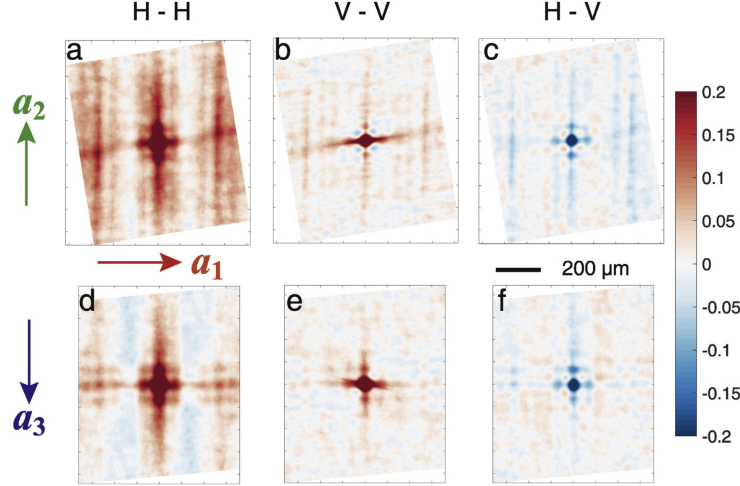


Figure 7: Two-dimensional slices of the two-point Pearson auto-correlation of interfacial mean curvature H (a,d) and velocity V (b,e), and two-point Pearson cross-correlation of H and V (c,f). The slices are selected perpendicular to the tertiary dendrite arms, and perpendicular to the secondary dendrite arms. These slices correspond to the 2D slices of the SDF in Fig. 6c and d, respectively.

the secondary and tertiary dendrite arms, which have extended structures along directions matching the strong correlation stripes. Along the a_1 axis, the Pearson autocorrelation coefficient of H shows periodic patterns with a periodicity of $270 \mu m$ (Fig. 7a,d). This indicates periodically repeated microstructures along the primary dendrite arm direction in the sample, which naturally suggests that this periodicity comes from the secondary branches on the primary arm. Therefore, this distance of $270 \mu m$ should correspond to the secondary dendrite

We have developed a two-point statistics method for analyzing microstructure datasets. This method includes two sets of spatial correlation tools: two-point Pearson correlation function, which measures how much two arbitrary spatial functions correlate in space on a scale of 1 to 1, and two-point probability function, which gives the joint or conditional probability distribution of two discretized local states in space. These two-point correlations allow us to retrieve important microstructure attributes in large and high-dimensional datasets in a physically interpretable form. It also provides an objective, reliable, and systematic way of measuring the characteristic length scales in the microstructure. Using this approach we have been able to extract the average spacing between secondary dendrite arms in complicated dendritic mixtures, the average width of these arms as well as the spacing between tertiary arms.

4 Data-driven approaches for structure-property linkages in high contrast composites

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Abstract: Data-driven methods are emerging as an important toolset in the studies of multiscale, multi-physics, materials phenomena. More specifically, data mining and machine learning methods offer an efficient toolset for extracting and curating the important correlations controlling these multiscale materials phenomena in high-value reduced-order forms called process-structure-property (PSP) linkages. Broadly speaking, PSP linkages can be cast in both directions of scale-bridging: (i) homogenization (going from smaller scales to larger scales) and (ii) localization (going from larger scales to smaller scales). In this project, data-driven approaches are implemented to model PSP linkages (i.e. homogenization and localization linkages) of high contrast composites. In particular, localization is studied by two approaches based on traditional machine learning methods, while deep learning is used to model homogenization.

4.1 Motivation & Major Goals

There has been a growing popularity in the use of data mining and machine learning methods in studies of various phenomena in materials science. In particular, parametric models are learned from massive amounts of collected data, either from laboratory experiments or from computational simulations, in order to represent, describe, and approximate process-structure-property (PSP) relationships for materials systems. Models built in such manner are often used as surrogate models for the more expensive and/or computationally intensive physics based models (e.g., thermochemical and microstructural evolution models). In contrast to the modeling style in physical models, where we explicitly specify equations, physical constraints, variable spaces to the extreme, data models often free the designers from such specifics. The logical and mathematical formula they use tend to form automatically or semi-automatically, with only the supply of data examples, and specifications of model structures, loss functions, and optimizers, to extract unknown correlations between inputs and outputs.

An important attribute of most materials systems employed in advanced technologies is that they exhibit heterogeneous structures at a multitude of hierarchical length scales. Consequently, most materials phenomena of interest are inherently multiscale, and the communication of the salient information between the hierarchical structure scales is the central challenge in any modeling and simulation effort. Historically, the multiscale materials modeling efforts have addressed either homogenization (communication of information from the lower length scale to the higher length scale) or localization (communication of information from the higher length scale to the lower length scale). Although both homogenization and localization have been studied extensively in literature using physically based approaches, recent work has identified the tremendous benefits of fusing these approaches with data-driven approaches. However, most of the prior effort has only addressed a limited number of the multiscale features. For example, many of the previous efforts are not readily extendable to high contrast composites (i.e., large differences in the properties of the microscale constituents in the composite).

One of the goals of this project was to use data-driven approaches to model PSP linkages (i.e. homogenization and localization) of high contrast composites. More specifically, localization was studied by two approaches based on traditional machine learning methods, while deep learning was used to model homogenization. All of the three works are described in the following sections.

4.2 Significant Results

4.2.1 Machine learning approaches for elastic localization linkages in high-contrast composite materials

This work [23] explores and presents multiple viable approaches for computationally efficient predictions of the microscale elastic strain fields in a three-dimensional (3-D) voxel-based microstructure volume element (MVE). Advanced concepts in machine learning and data mining, including feature extraction, feature

ranking and selection, and regression modeling, are explored as data experiments. Improvements are demonstrated in a gradually escalated fashion achieved by (1) feature descriptors introduced to represent voxel neighborhood characteristics, (2) a reduced set of descriptors with top importance, and (3) an ensemble-based regression technique.

The dataset used in this work is a high-contrast two-phase composite system. The contrast refers to the ratio of the elastic stiffness parameters of the constituent phases of the composite system, and this dataset has a contrast of 10. A total of 2500 microscale volume element (MVE) with varying volume fractions are included in this study. They are evenly distributed in 100 variations of volume fraction values, from 1.0% to 99.4%. Therefore, 25 MVEs are present in each variation, within which, 15 are used as calibration (for feature extraction, model training), and the remaining 10 are used for testing.

The performance of the models is evaluated by the mean absolute strain error (MASE) e in a MVE, defined as

$$e = \frac{1}{S} \sum_{s=1}^S \left| \frac{p_s - \hat{p}_s}{p_{imposed}} \right| \times 100\% \quad (6)$$

where $p_{imposed}$ denotes the average strain imposed on the MVE, and p_s and $\hat{p}_s$ denote the values of the strain in the voxel s from the FE model and the surrogate model developed in this work, respectively. This metric quantifies the average error for a single MVE microstructure.

Figure 8 schematically illustrates the main data-driven protocol for establishing a predictive model. It generally comprises of two key processes: (i) feature extraction, and (ii) construction of the regression model.

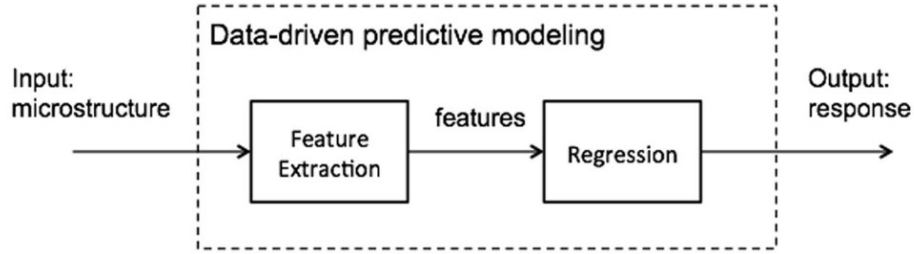


Figure 8: Flowchart of data-driven single-agent predictive modeling.

A set of features are designed to describe local information of targeted voxel. Figure 9 shows the definition of the set of features used in this work. Then, feature extraction is applied to select the most important features related to output, and different regression methods are used to train predictive models. The results show that the predictive model using Random Forest method trained on 57 selected features gives the best performance, which can achieve 13.02% average MASE on the testing set.

Symbol	Meaning	Count	Scope
$m_{s,l,t}$	Microstructure value of voxels at a neighbor level l , with index t , of a focal voxel at s $l = 1, \dots, 12$	179	Binary, {0,1}
pr_l^h	Fraction of voxels with microstructure phase h at neighbor level l	24	Real, [0,1]
Pr_l^h	Fraction of voxels with microstructure phase h up to neighbor level l	24	Real, [0,1]
$\hat{p}_{norm}^h$	The normalized impact of all 12 levels of neighbors of phase h $\hat{p}_{norm}^h = \sum_{l=1}^{12} T_l \cdot pr_l^h / \sqrt{l} + T_0 \cdot pr_0^h / 0.5$	2	Real
S_3	3-plane symmetry index	1	Real
S_9	9-plane symmetry index	1	Real

Figure 9: Definition of the set of features.

Figure 10 shows a comparison of FE and machine learning model predictions of the strain fields in an example MVE with the volume fraction of 50%. We can observe that the results of FE model and predictive model are almost indistinguishable.

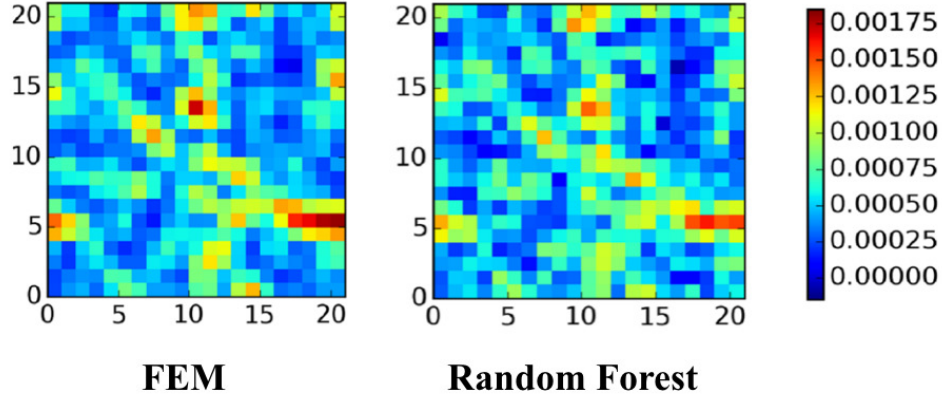


Figure 10: A comparison of FE and statistical model predictions of the strain fields in an example MVE with the volume fraction of 50 %.

4.2.2 Context aware machine learning approaches for modeling elastic localization in three-dimensional composite microstructures

In this work [42], we design a new data modeling approach that is explicitly hierarchical to be able to take advantage of the multiscale characteristics of a heterogeneous material structure. The design is to be manifested through the idea of context detection, which is a concept used in reinforcement learning and robotics to deal with non-stationary environments. Context detection is defined here as finding the right high-level, low-dimensional, knowledge representation in order to create coherent learning environments. Once different contexts are identified from data, one can build separate models out of each context group. This approach has many similarities with the divide and conquer scheme, which breaks a large, difficult problem into a set of small, simpler problems.

This work examines the advantage of building context aware learning systems by solving an elastic localization problem in high contrast composites. More specifically, we aim to provide a computationally efficient surrogate model, with parameters learned from data, to predict the microscale elastic strain field in any given voxelized three-dimensional (3-D) volume element of a high contrast composite subjected to a prescribed macroscale elastic strain (applied as a periodic boundary condition). We address the multiscale challenge by identifying and representing the higher level data distribution through context detection. In our designed two-stage system, the first stage attempts to find the contexts in data, while the second stage builds context specific learning models. We compare the results from this new data model with benchmarks from previous work in section 4.2.1 using same dataset and demonstrate that the two-layer data modeling scheme provides a viable approach for capturing the elastic localization linkages in high contrast composites.

The outline of such a learning structure is shown in Figure 11. The major advancement of this model is designed two layers of feature extraction: the macro-layer, and the micro-layer. The macro-layer features are generated to probe MVE-level similarity in structures. Once a representation of similarity is defined, the original set is divided into multiple subsets with low inter-similarity and high intra-similarity. MVEs within the same context group possess higher resemblance with each other, in terms of solely their structure representation, than those from different groups. In this work, we examines three macro features, which are volume fraction, data clustering and two-point statistics. Micro-layer features, on the other hand, specialize in learning the voxel level characteristics. We adopt selected features from previous section that gave the best performance as micro features. Random forest method is applied to train the predictive model.

The results show that using two-point statistics as macro features gives the best performance, which can achieve 8.04% average MASE on testing set. The prediction output of a center slice of a randomly selected MVE with 50% volume fraction is illustrated in Figure 12. The difference between a ground truth slice (annotated “FEM”) and a predicted slice (annotated “Multi-agent model”) is almost indistinguishable.

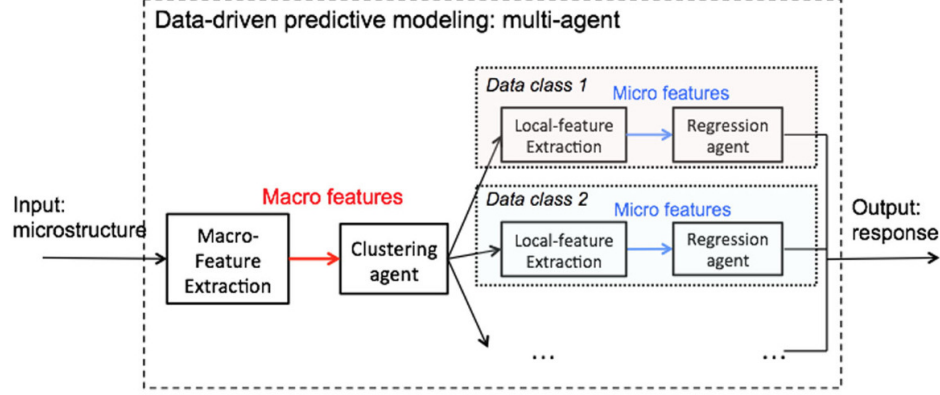


Figure 11: Flowchart of data-driven multi-agent predictive modeling.

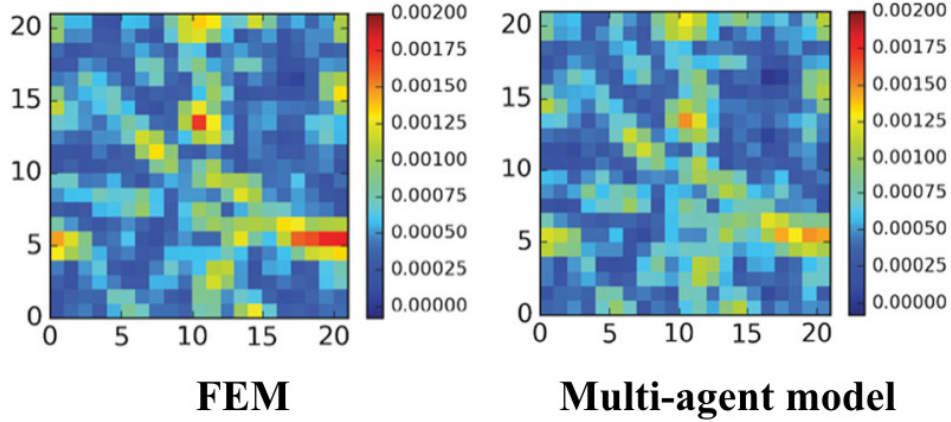


Figure 12: A comparison of FE and multi-agent model predictions of the strain fields in an example MVE with the volume fraction of 50 %.

4.2.3 Deep learning approaches for mining structure-property linkages in high contrast composites from simulation datasets

Traditional machine learning methods usually depend on intensive feature engineering, and have enjoyed some success in establishing the desired PSP linkages. In contrast, deep learning approaches provide a feature-engineering-free framework with high learning capability. In this work (under review), a deep learning approach is designed and implemented to model an elastic homogenization structure-property linkage in a high contrast composite material system. More specifically, a standalone 3-D convolutional neural network (CNN) is built for the first time to establish structure-property linkages for high contrast elastic 3-D composites using a completely feature-engineering free approach. An extensive analysis of CNNs with different numbers of convolution and pooling layers is performed.

In order to explore and evaluate the performance of CNN models in predicting the effective elastic properties of high contrast composites, we generated 8550 3-D MVEs from micromechanical finite element models with contrast of 50. In the effort to generate a rich morphological diversity in the generated set of MVEs, 3-D Gaussian filters with different covariances were employed. Some examples of MVEs with different microstructural details are shown in Figure 13. The MVE in part (a) is generated by a 3-D Gaussian filter with three identical diagonal entries in the covariance matrix. On the other hand, the microscale constituents of MVEs in part (b),(c) and (d) have clear directionality in x, y and z directions. To train the model and evaluation models performance, the ensemble of 8550 MVEs are split into 2 sets. 5700 MVEs are used for training the CNN, and rest 2850 MVEs are solely used as test set.

The model performance is evaluated by calculating mean absolute stiffness error (MASE) for both training and testing sets. MASE for a selected set (i.e. training or testing set) reflects the average error between the

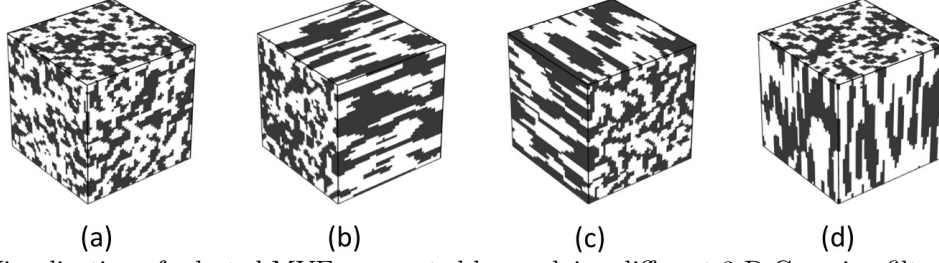


Figure 13: Visualization of selected MVEs generated by applying different 3-D Gaussian filters to a random number field.

predicted values and ground truth values calculated from FE models. This error metric is defined as

$$MASE = \frac{1}{N} \sum_{i=1}^N \left| \frac{S_i - \hat{S}_i}{S_{average}} \right| \times 100\% \quad (7)$$

where N denotes the total number of MVEs in the selected dataset, S_i and $\hat{S}_i$ represent ground truth effective stiffness and predicted effective stiffness for the i^{th} MVE, respectively. $S_{average}$ denotes the average effective stiffness of all the MVEs in the dataset. In other words, MASE can be calculated by dividing mean absolute error between ground truth values and predicted values by the average of effective stiffness in the dataset.

Different architectures of varying depths and different number of filters in each convolution layer of 3-D CNNs are explored to examine which architecture produce the best model for the present application. The depths of 3-D CNNs are gradually extended from 10 layers to 16 layers where multiple stacked convolution layers and pooling layers are added gradually. In other trials, the number of filters in each convolution layer was gradually increased for a fixed depth. The results show that a 14 layers 3-D CNN achieves the best performance (good accuracy without over-fitting). In the description of the architecture, if “convX” denotes a convolution layer with $X \times 3 \times 3 \times 3$ filters, “pool” a pooling layer and “fcX” a fully connected layer with X neurons. The best CNN can be described as *input – conv16 – pool – conv32 – pool – conv64 – pool – conv128 – pool – conv256 – pool – fc2048 – fc1024 – output*. Then, we carry out further exploration and optimization of the hyperparameters space for this architecture. The results shown that when batch size is 32 MVEs and no dropout is applied, the deep learning model gives the best performance.

We compare the results obtained from deep learning approach with the results obtained from the correlation based method, which is shown in Table 4. The proposed 3-D CNN improves the model performance of benchmark method by as much as 54% ($1 - 3.10/6.79$) in terms of testing MASE. The parity plots of both methods for both training and testing sets are shown in Figure 14. The top and bottom row depict the results of both methods for training and testing sets, respectively. The left and right columns correspond to CNN, correlation-based methods, respectively. From Figure 14, it is seen that the parity plots of 3-D CNN for both training and testing sets exhibit high accuracy and low variance. In particular, it is noted that the deep learning approach performs better in very low and very high effective stiffness values, where the benchmark approach produces significantly higher errors in its predictions.

Table 4: Performance comparison of predictive models

Method	Training MASE	Testing MASE	Training time	Testing time
3-D CNN	2.10%	3.10%	25 min	6.75 s
Correlation function based method	6.81%	6.79%	70 s	35 s

In addition, two software packages have been developed based on this work, which can be found in <http://cucis.ece.northwestern.edu/projects/MURI/workpred.html>. One is deuNet, which is a theano based deep learning software package. It is a general package that makes exploring different neural network architectures, training schemes, hyperparameters easy. The other one is MES Predictor, and it can be directly used to predict macroscale (effective) stiffness of high-contrast two-phase three-dimensional composite materials.

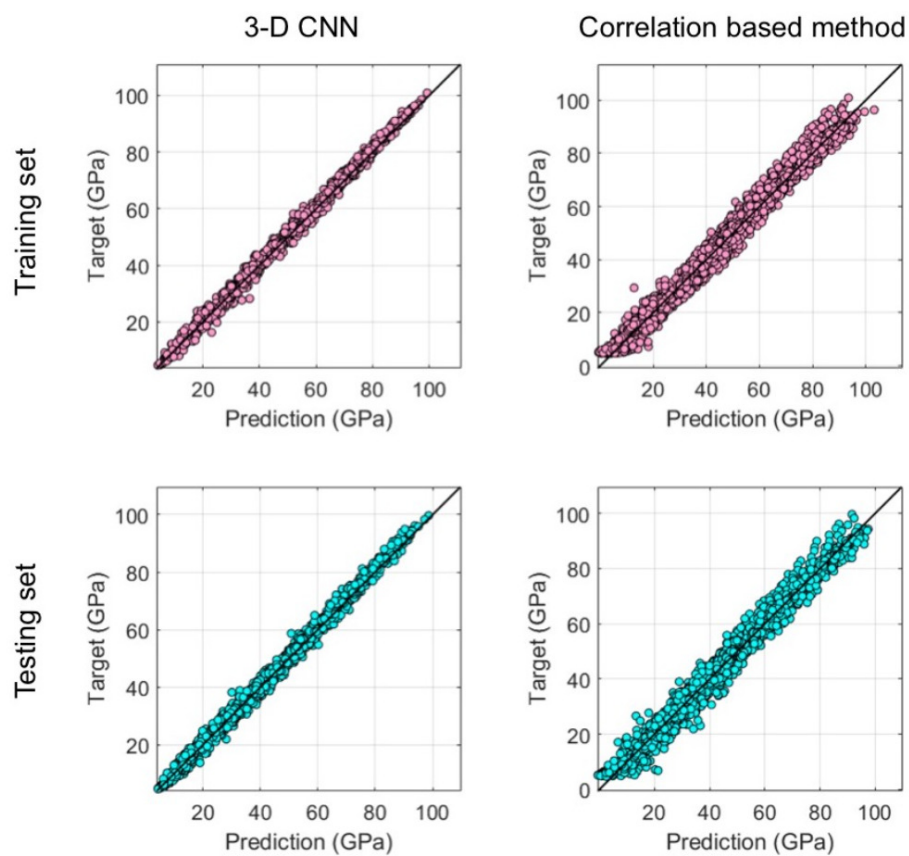


Figure 14: Parity plots of CNN and correlation based method. The top and bottom rows depict the results for training and testing sets, respectively. The columns represent the CNN and correlation based method, respectively.

5 A CNN architecture for the indexing of electron diffraction patterns

*Dipendra Jha, Ruoqian Liu, Saransh Singh, Reda Al-Bahrani, Wei-keng Liao,
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Abstract: Electron Backscatter Diffraction (EBSD) is one of an array of tools widely used by domain scientists to determine the crystal orientation of individual grains in polycrystalline materials. Current methods for indexing of diffraction patterns include the dictionary based indexing and the Hough transform. While the former is computationally very expensive, the latter method is highly sensitive to noise present in experimental patterns. In this project, we have designed and implemented several machine learning based prediction models for indexing EBSD patterns. We demonstrated how a deep learning approach can be leveraged to build a predictive tool that can learn the crystal orientations of polycrystalline materials from simulated EBSD patterns, and perform indexing of diffraction patterns from real experiments with better accuracy than dictionary based indexing.

5.1 Motivation and Major Goals

Since its development in the early 1990s, automated Electron Backscatter Diffraction (EBSD) has become the primary tool to determine the crystal orientation of crystalline materials across a wide variety of material classes [5]. The technique provides quantitative information about the grain size, grain boundary character, grain orientation, texture and phase identity of the sample by measuring the angular distribution of backscattered electrons using a combination of scintillator screen and a charge coupled device (CCD) camera. The schematic of the EBSD setup is shown in 15. The sample sits at a tilt of σ (typically 70°) with the camera tilted at angle θ_c (typically $0^\circ - 10^\circ$). A parallel beam of electron travels from the pole piece and interacts with the sample at point O . The backscattered yield is measured by the scintillator. The physics-based model predicts the backscattered yield based on the principles of quantum mechanics. Assuming the microscope is parametrized by $\mathcal{M}$, the geometry of the setup is denoted by $\mathcal{G}$ and the crystal under investigation is parametrized by $\mathcal{C}$, the forward model is given by

$$\mathcal{F} \equiv \mathcal{F}(\mathcal{M}, \mathcal{G}, \mathcal{C}). \quad (8)$$

Further details of the model can be found in [18]. An example experimental EBSD pattern from Iron with its corresponding physics-based simulation is shown in Fig. 15(b) respectively.

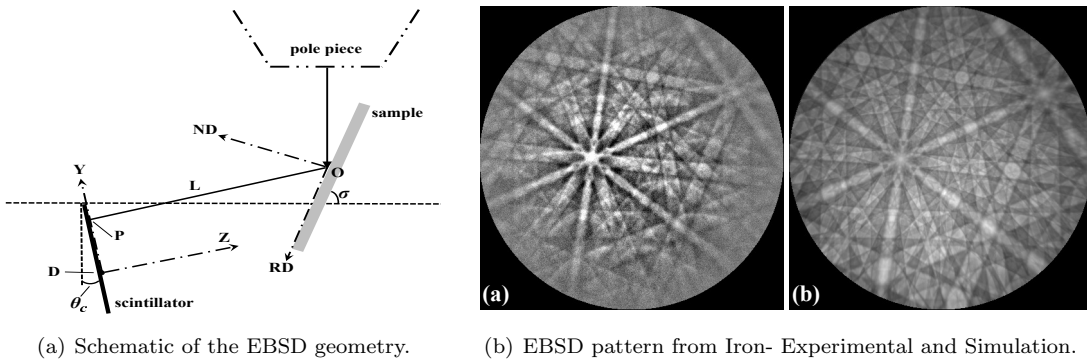


Figure 15: EBSD geometry and patterns.

There are two major techniques to indexing EBSD patterns, each with its advantages and drawbacks. These include the commercially available Hough-transform based approach and the newly developed Dictionary Indexing method. The commercially available solution to the indexing problem is performed using a feature detection algorithm [3]. A Hough transform of the diffraction is performed to identify linear features from the diffraction pattern. The angles between the extracted linear features are compared to a pre-computed look up table to determine the crystal orientation. This method has been very successful in indexing EBSD patterns and has led to significant advances in materials characterization. However,

the performance of this method quickly deteriorates in the presence of noise. Domain scientists also use a newly developed dictionary-based indexing approach for the prediction of crystal orientation from the EBSD patterns. Dictionary based indexing [28] is a nearest neighbor search approach in which the output angles correspond to the orientation angles of the closest EBSD pattern present in dictionary. This method has been shown to be very robust to noise in the diffraction pattern and outperforms the line feature based Hough transform method [11] for a wide variety of crystal classes. However, this approach is computationally very expensive, which limits the technique to be an off-line method, and a real time solution to the indexing problem is not possible using this approach.

In this project, our goal was to design and implement a machine learning based predictive model that can be trained using a simulation diffraction dataset, but can also predict the crystal orientations for experimental data, such that they have a minimum “disorientation” with respect to their ground truth. The project was carried out in two phases. In the first phase, we focused on designing convolutional neural network to learn and optimize for each of the individual crystal orientation angles [31]. In the second phase, we designed a convolutional neural network model for multi-output learn all the three orientation angles using one network and optimize for the mean disorientation [46].

5.2 Deep Learning Approach

5.2.1 Optimizing for Individual Angles

In the first phase [31], we constructed a deep convolutional neural network (CNN) that took the EBSD images as input, and learn to predict the three real-valued Euler angles as output. It used multiple convolutional layers that are used to take into consideration the spatial dependencies among image pixels, and fully connected layers for multi-layer regression. The dataset was composed of 333,227 simulated EBSD patterns of polycrystalline Nickel. The input data was a set of grey scale images with input pixel values between 0 (black) and 255 (white). Each image is associated with three target Euler angle values for determining the crystal orientation. We separately modeled for each Euler angle output, resulting in three different tasks. A random set of 300,000 samples was used for training, and 30,000 was used for testing each of these tasks.

A special loss function was designed to account for the periodicity of angular data (the fact that 0° is close to 359°) when measuring the difference between predicted outputs and the ground truth. Suppose a training set of m samples is given as $\{X_i, y_j^i\}_{i=1}^m$, where $X_i \in R^n$ denotes the i -th training sample, and y_j^i denotes the j -th output, $j = 1, 2, 3$, of the same training sample. For $j = 1, 3$ the output y^j is an orientation angle between 0° to 360° . For $j = 2$, y^j is bounded by a smaller range between 0° to 60° . To quantitatively measure the difference between the predicted angle $\hat{y}_i$ and ground truth y_i , while taking care of the periodicity of angular expressions, the loss L_i is computed from the predicted angle $\hat{y}_i$ and the ground truth y_i as:

$$L_i(y_i, \hat{y}_i) = \arccos(\cos(|y_i - \hat{y}_i|)) \quad (9)$$

where the j index in y is omitted because we used the same form across all three tasks. Such a loss function converted any angular difference between $[360^\circ, 360^\circ]$ to $[0^\circ, 180^\circ]$, or $[0, \pi]$ in radians. In CNN training, this customized loss function is used instead of the cross-entropy in classification. We designed and experimented with several CNN architectures, we present the best architecture- that is composed of 4 convolutional layers and 3 fully connected layers (including the last output layer), in Figure 16.

The best CNN network trained for the first angle had a normalized error rate of 0.007 on test data, after 300 epochs, that is a Mean Absolute Error (MAE) of 2.5° when predicting an angle between 0° and 360° . For the second and third Euler angles, the MAE in predictions were 1.8° and 4.8° respectively. The results were compared with the state-of-the-art benchmark of the dictionary based indexing presented in [28]. We replicated the method using the same split of training and test as used in deep net training, and obtained MAEs of 5.7° , 5.7° , and 7.7° , respectively for each angle. On an average our results were 54% better than the benchmark method. The evaluation in for this phase was carried out using simulated test samples which are very clear and pristine with no noise. The CNN network developed here treated the three Euler angles as independent. Rather than minimizing the difference between the three actual and predicted Euler angles, domain scientists are interested in minimizing the disorientation between the predicted and ground truth crystal orientations.

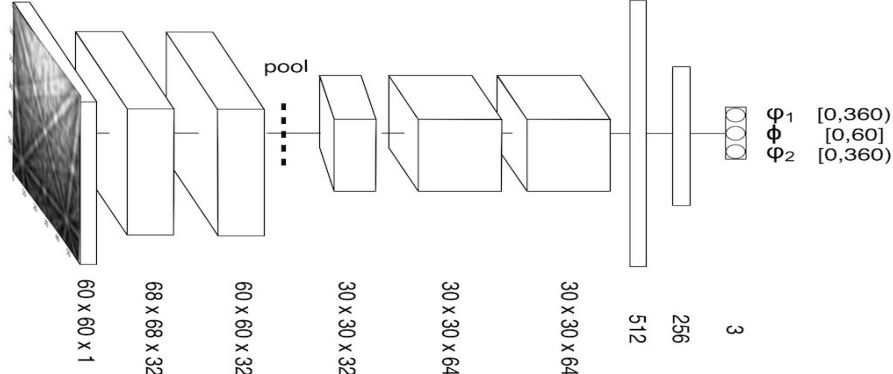


Figure 16: CNN architecture for optimizing for individual angles.

5.2.2 Optimizing for Disorientation

In the second phase of this project [46], our goal was to develop a machine learning based predictive model to predict the crystal orientations of the experimental diffraction patterns such that they have minimum disorientation with the ground truth. The ideal data driven approach for building a predictive model would be to train a machine learning model on the EBSD patterns from experiments. Nevertheless, experiments are very expensive; we have 1000 “experimental” diffraction patterns. Hence, we leveraged *only* the simulation EBSD patterns for training such that the model can predict the crystal orientations for the experimental EBSD patterns with minimum disorientation with respect to the true orientations. The training and test datasets were composed of EBSD patterns of polycrystalline Nickel. The training dataset contained two EBSD pattern dictionaries from simulation, one generated with a cubochoric sampling of $N = 100$ samples along the cubic semi axis, the other with $N = 50$. The first dataset has 333,227 patterns, the second one has 41,625 patterns. Combining them, 374,852 patterns were obtained to form the training set. The performance of the models were evaluated by indexing 1000 EBSD samples from real experiments with known orientations. The disorientation between the predicted and known Euler angles provides the efficacy of the approach.

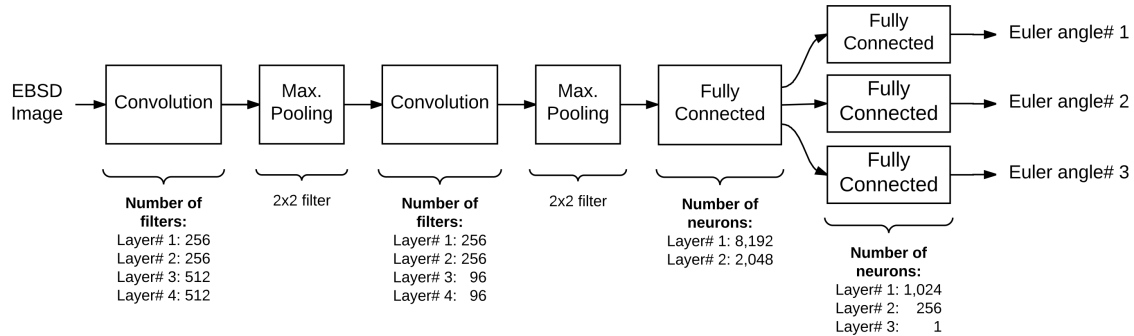


Figure 17: CNN architecture for learning multiple outputs.

It required optimizing for the domain specific function of mean disorientation between the predicted and the true crystal orientations. It was challenging due to two main reasons. First, the disorientation is the distance metric of a non-euclidean manifold. In the absence of symmetries in orientation, this metric is easy to compute using analytical expressions. However, the presence of crystal symmetries introduces degeneracies in the space and introduces discontinuities in the gradient of the disorientation metric with respect to the input orientations. It rendered the disorientation function inappropriate to be used for optimization using stochastic gradient descent for any deep learning model. Second, the original disorientation algorithm was computationally intensive; it takes one pair of predicted and true Euler angles and computes their

symmetrically equivalent orientations to find the disorientation (Equation 10):

$$\mathcal{D}(\mathbf{g}_1, \mathbf{g}_2) = \min_{i,j \in \mathcal{N}} \left\{ \arccos \left(\left(\text{tr} \left[(\mathcal{O}_c^i \mathbf{g}_1)^{-1} (\mathcal{O}_c^j \mathbf{g}_2) \right] - 1 \right) / 2 \right), \arccos \left(\left(\text{tr} \left[(\mathcal{O}_c^i \mathbf{g}_2)^{-1} (\mathcal{O}_c^j \mathbf{g}_1) \right] - 1 \right) / 2 \right) \right\}. \quad (10)$$

Due to the presence of crystal symmetry given by the set $\mathcal{O}_c$ with cardinality $\#\mathcal{O}_c = 24$ in our case, there exist 24 symmetrically equivalent orientations for both– the predicted and true orientations. This gave $24 \times 24 = 576$ symmetrically equivalent orientation pairs for each disorientation computation.

The disorientation computation contains $\arccos(x)$ function which is undefined for values outside its domain of $[-1, 1]$. We approximated it by putting an upper bound of 1 to the magnitude of all the values passed to the $\arccos(x)$ function. We implemented a differentiable approximation of the mean disorientation by building a computational tensor graph using TensorFlow [39]. We leveraged its auto-differentiation support for computing the gradients of the mean disorientation with respect to the Euler angles. The mean disorientation was optimized by training a deep learning model using the stochastic gradient descent algorithm [2]. However, it was very costly both in terms of processing time and memory transfer; it took around 24 hours to train our model for one epoch using a TitanX GPU with 12GB memory. This made it impractical to train a deep learning model using the sequential implementation in feasible time. We optimized it to process one mini-batch so that it could leverage the parallelization available in GPUs.

Optimizing for mean disorientation required learning the crystal orientation angles using a single model such that they can be used to optimize the mean disorientation. Figure 17 demonstrates a novel CNN model architectures– a branching model with individual and independent model components for each output. The first four model components are composed of multiple convolution layers and max pooling. Convolution layers captures the locally correlated features present in the input EBSD patterns; they learn the high level abstract features from the inputs. As the three outputs require similar learning capability, the branched model is composed of three classifier branches containing equal numbers of layers and parameters. As the outputs are correlated with each other, they all shared the same convolution outputs and the first fully connected component. The convolutional layers are the computationally expensive components; they extract high level features from the inputs that are required for learning all outputs. Sharing the convolutional layers kept the computational cost comparable to using conventional approach of using three different models for learning three different outputs. All models were implemented using python and TensorFlow [39]. They were trained using Titan X GPUs with 12GB memory. An extensive search was carried out to tune the hyperparameters such as learning rate, optimization algorithm, momentum and learning rate decay. The models were evaluated using the mean disorientation and mean symmetrically equivalent orientation absolute error (MSEAE). The MSEAE was computed by considering the periodicity of orientation angles as follows:

$$mseae_1(\vec{y}_s, \hat{\vec{y}}_s) = \frac{1}{n} \sum_{i=1}^n \sum_{j=1}^3 |y_{sj} - \hat{y}_{sj}|, \quad (11)$$

$$mseae_2(\vec{y}_s, \hat{\vec{y}}_s) = mseae_1(\vec{y}_s, \hat{\vec{y}}_s) \mod (2\pi), \quad (12)$$

$$MSEAE(\vec{y}_s, \hat{\vec{y}}_s) = \begin{cases} mseae_2(\vec{y}_s, \hat{\vec{y}}_s), & \text{if } mseae_2(\vec{y}_s, \hat{\vec{y}}_s) \leq \pi \\ = 2\pi - mseae_2(\vec{y}_s, \hat{\vec{y}}_s), & \text{else} \end{cases} \quad (13)$$

where $\vec{y}_s$ and $\hat{\vec{y}}_s$ are Euler angles of the symmetrically equivalent true and predicted orientations with minimum disorientation.

First, we experimented with several conventional loss functions to optimize the orientation angles. The model with MAE as loss function achieved the best accuracy in predicting the Euler angles in the experimental set; it outperformed the dictionary based indexing by an MSEAE of 40% as seen in Table 5. The three Euler angles exhibited different errors; the error for the second Euler angle being the lowest. Nevertheless, the mean disorientation for the test set did not reach such low value as the training set; it stopped improving after reaching 0.596° . The mean disorientation for the test set while using MSE as the loss function was 1.285° . Next, we optimized using the domain specific loss function of mean disorientation. Since our goal was to optimize for disorientation, we expected it to result in a better mean disorientation on both sets. However, the mean disorientation alone as the loss function did not work well; it achieved a mean disorientation of 1.224° on the test set.

Table 5: Mean Disorientation (MD) and mean symmetrically equivalent orientation absolute error (MSEAE) using different models and loss functions.

Model	Loss Function	Simulation Data	Experimental Data	
		MD	MD	MSEAE
Dictionary-based Indexing	-	-	0.652°	[0.6592°, 0.3534°, 0.6484°]
Deep Learning	MAE	0.064°	0.596°	[0.4039°, 0.1776°, 0.4426°]
Deep Learning	MSE	0.292°	1.285°	-
Deep Learning	Mean Disorientation	0.272°	1.224°	-
Deep Learning	MAE+Mean Disorientation	0.132°	0.548°	[0.7155°, 0.2194°, 0.7066°]
Deep Learning	MSE+Mean Disorientation	0.171°	0.658°	-

Since optimizing for mean disorientation did not perform well, we designed and experimented with several hybrid loss functions combined the mean disorientation with the conventional loss functions. The best loss function was the sum of the MAE and the mean disorientation. The model achieved a mean disorientation of 0.548° on the experimental set- 16% better than using dictionary based indexing.

6 Vector Field Electron Tomography

C. Bouman and M. De Graef

Abstract: Tomography has been used traditionally to reconstruct 3D scalar fields. In our research, we have applied Bayesian concepts to the reconstruction of 3D vector fields in the area of magnetic materials. Our approach allows for the accurate 3D reconstruction of both the magnetic vector potential and the magnetization of nano-scale magnetic samples, based on phase reconstructions from Lorentz transmission electron microscopy

6.1 Introduction

In conventional tomographic reconstructions, the object being reconstructed is of a scalar nature; mass density is perhaps the most frequent quantity being reconstructed, but in principle any scalar field could be handled. When the field has vector character, for instance the magnetization field of a magnetic material, or the polarization of a ferroelectric material, then a tomographic reconstruction requires additional experimental data, since one has to determine three numbers for each voxel instead of just one. Vector field tomography has been used in the past to reconstruct velocity fields in fluids based on transmission of acoustic waves []. In classical electromagnetism, the fundamental fields are the electric field $\mathbf{E}$ and the magnetic induction $\mathbf{B}$. When a magnetic object is studied by means for transmission electron microscopy, however, the interactions between the incident beam and the sample require a quantum mechanical formulation, which, in turn, requires the use of the more fundamental fields, namely the electrostatic potential V and the magnetic vector potential $\mathbf{A}$. In our application of vector field electron tomography (VFET), we aim to reconstruct, in 3D, all three components of either the magnetic vector potential $\mathbf{A}(\mathbf{r})$ or the magnetization $\mathbf{M}(\mathbf{r})$ as a function of position $\mathbf{r}$.

Consider a magnetic object of arbitrary shape, described by a shape function $D(\mathbf{r})$ which equals 1 inside and 0 outside the object. The function is described with respect to an origin that is located somewhere inside the object, possibly in the center-of-mass. The object has both a charge density, $\rho(\mathbf{r})$ (number of charges per unit volume), and a magnetization $\mathbf{M}(\mathbf{r})$ (total dipole moment per unit volume); the charge density is assumed to be a constant throughout the object, whereas for the magnetization only the magnitude, M_0 , of the vector field is constant. The charge density is measured in C/nm³, whereas the magnetization is expressed in A/nm. The charge density is related to an electrostatic potential through the following convolution integral:

$$V(\mathbf{r}) = \frac{1}{4\pi\epsilon_0} \iiint d\mathbf{r}' \frac{\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|}, \quad (14)$$

whereas the magnetic vector potential can be derived from the magnetization as:

$$\mathbf{A}(\mathbf{r}) = \frac{\mu_0}{4\pi} \iiint d\mathbf{r}' \mathbf{M}(\mathbf{r}') \times \frac{\mathbf{r} - \mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|^3}; \quad (15)$$

$\epsilon_0 = 8.854 \times 10^{-21}$ F/nm is the permittivity of free space, and $\mu_0 = 4\pi \times 10^{-7}$ N/A² is the permeability of vacuum. The electrostatic potential is measured in V, whereas the vector potential is expressed in units of Vs/nm. The goal of VFET is to determine both ρ and $\mathbf{M}$ as a function of position for a given sample, based on projected through-focus image series as a function of sample tilt angle.

When an electron wave travels through a region of space with a non-vanishing magnetic vector potential, $\mathbf{A}(\mathbf{r})$, then the phase of that wave is modified as follows:

$$\varphi_m(\mathbf{r}_\perp) = -\frac{e}{\hbar} \int_{-\infty}^{+\infty} \mathbf{A}(\mathbf{r}_\perp + \ell\boldsymbol{\omega}) \cdot \boldsymbol{\omega} d\ell, \quad (16)$$

where $\boldsymbol{\omega}$ is a unit vector along the electron beam propagation direction, and ℓ parameterizes the electron trajectory; $\mathbf{r}_\perp$ is a vector in the plane normal to $\boldsymbol{\omega}$. The prefactor can also be written as π/ϕ_0 , with $\phi_0 = h/2e$ the flux quantum ($\phi_0 = 2070$ T nm²). In general, there will also be an electrostatic contribution to the phase shift:

$$\varphi_e(\mathbf{r}_\perp) = \frac{\pi}{\lambda E} \int_{-\infty}^{+\infty} V(\mathbf{r}_\perp + \ell\boldsymbol{\omega}) d\ell, \quad (17)$$

where $V(\mathbf{r})$ is the electrostatic lattice potential (measured in Volt), λ is the (relativistic) electron wavelength, and E is the relativistic accelerating potential of the microscope. Note that both of these integrals can be regarded as tomographic projection integrals, so that acquisition of the two phase shifts as a function of object tilt should, in principle, permit for a 3D reconstruction of both potentials V and $\mathbf{A}$. Such reconstructions have been possible for a while, but they always use the “traditional” reconstruction approaches, i.e., filtered back-projection (FBP), simultaneous iterative reconstruction technique (SIRT), discrete algebraic reconstruction technique (DART), and so on. None of these approaches takes into account any prior information about the object being reconstructed. In the MURI program, we worked in a close collaboration between Carnegie Mellon University and Purdue University to apply Bayesian principles to the 3D reconstruction problem in the form of Model-Based Iterative Reconstructions (MBIR). Some of the underlying theory and a few selected results are described in the following sections [43, 45].

6.2 Theoretical Model

We introduce the MBIR approach by first formulating its framework for a generic tomography problem. Let $x \in \mathbb{R}^N$ be the discrete vector of an unknown image and $y \in \mathbb{R}^M$ be the discrete vector of projection measurements of x at a variety of angles. Then the MBIR approach performs a reconstruction by maximizing the joint probability distribution resulting from the likelihood function, $\mathcal{P}(y|x)$, and the prior distribution, $\mathcal{P}(x)$. This methodology is more commonly known as the maximum-a-posteriori (MAP) estimation technique [4]. Mathematically, the MAP estimate is expressed as:

$$\hat{x}_{\text{MAP}} = \underset{x}{\operatorname{argmin}} \{ -\log \mathcal{P}(y|x) - \log \mathcal{P}(x) \}. \quad (18)$$

We use the Poisson probability mass function to model the likelihood term. We use a second order Taylor series expansion of the logarithm of the Poisson distribution [4] to approximate the first term of eq. 18 as:

$$\log \mathcal{P}(y|x) \approx -\frac{1}{2}(y - Hx)^T W(y - Hx) + f(y), \quad (19)$$

where H is the forward projection matrix, W is a diagonal noise weighting matrix and $f(y)$ is a term independent of the optimization variable x . Additionally, H is an orthogonal matrix such that $H^T = H^{-1}$; hence, H^T denotes the back projection operator.

For the problem of reconstruction of magnetization vector fields, the forward model has the form

$$y = FHx + w,$$

where F is the sparse tomographic projection matrix, H is a non-sparse convolution matrix that performs linear space-invariant convolution, and w is the noise vector with covariance W^{-1} . In [45], we derive a general form for the structure of H of the form

$$z = Hx, \quad (20)$$

where $z = [z^{(u)t}, z^{(v)t}, z^{(w)t}]^t$, $x = [x^{(u)t}, x^{(v)t}, x^{(w)t}]^t$, and

$$H = \begin{bmatrix} 0 & H^{(w)} & -H^{(v)} \\ -H^{(w)} & 0 & H^{(u)} \\ H^{(v)} & -H^{(u)} & 0 \end{bmatrix}. \quad (21)$$

where $H^{(w)}$, $H^{(v)}$, and $H^{(u)}$ are three Green’s function operators that are efficiently implemented with FFTs.

The prior term in eq. 18 is modeled using a Markov Random Field (MRF) in:

$$\log \mathcal{P}(x) = - \sum_{\{i,j\} \in \mathcal{C}} b_{ij} \rho(x_i - x_j), \quad (22)$$

where $\mathcal{C}$ denotes the set of neighboring pixels, b_{ij} is a non-causal symmetric weighing filter that is normalized such that $\sum b_{ij} = 1$, and $\rho(\cdot)$ is the potential function. The choice of the MRF as the prior model is due

to its proven usefulness in image processing as well as in tomographic reconstructions [8]. In our work, we resort to a class of MRFs called the q -Generalized Gaussian Markov Random Field (q -GGMRF) to model the prior. Accordingly, the potential function in eq. 22 is given by:

$$\rho(\Delta) = \frac{|\Delta|^p}{p\sigma_x^p} \left(\frac{\left| \frac{\Delta}{T\sigma_x} \right|^{q-p}}{1 + \left| \frac{\Delta}{T\sigma_x} \right|^{q-p}} \right), \quad (23)$$

where $\Delta = x_i - x_j$, and p , q , σ_x and T are the q -GGMRF parameters. Typically, $1 \leq p \leq q \leq 2$ is used to ensure strict convexity of the potential function and, subsequently, of the MAP optimization [15]; for our work we use $q = 2$. When p is set to 2, the potential function is quadratic and the prior model facilitates a reconstruction with smooth edges. On the other hand, when p is close to 1, the prior model performs sharp edge preserving reconstructions. Similarly, σ_x is the variance of the prior distribution and its value is set to achieve a balance between noise and resolution. Finally, the constant T determines the approximate threshold of transition between low and high contrast regions.

Substituting the log-likelihood expression of eq. 19 and the prior model of eq. 22 into eq. 18, and considering W as the identity matrix, one obtains the MAP reconstruction to be the solution of the following optimization problem:

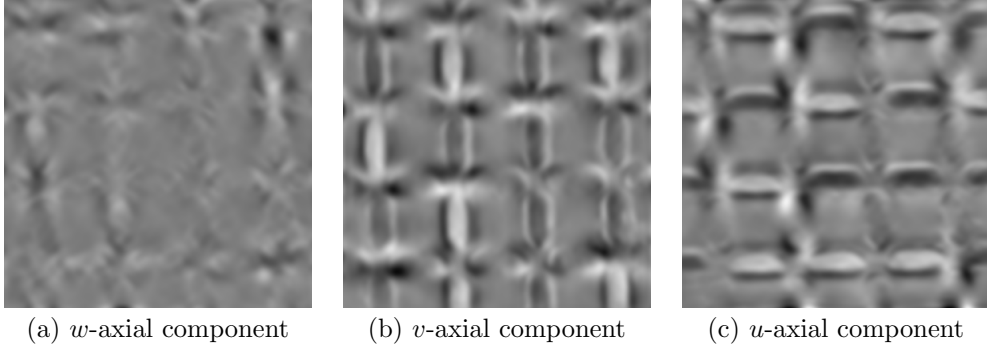
$$\hat{x}_{\text{MAP}} = \underset{x}{\operatorname{argmin}} \left\{ \frac{1}{2} \|y - Hx\|^2 + \sum_{\{i,j\} \in \mathcal{C}} b_{ij} \rho(x_i - x_j) \right\}, \quad (24)$$

where the cost (objective) function being minimized is given by

$$c(x) = \frac{1}{2} \|y - Hx\|^2 + \sum_{\{i,j\} \in \mathcal{C}} b_{ij} \rho(x_i - x_j). \quad (25)$$

The resulting optimization problem is difficult to solve directly, so we solve it by breaking into couple optimizations using the augmented Lagrangian formulation, and then using the alternate direction method of multipliers (ADMM) optimization approach.

Reconstruction of Magnetization using MBIR ($u - v$ axes)



Reconstruction of Magnetization using MBIR ($v - w$ axes)

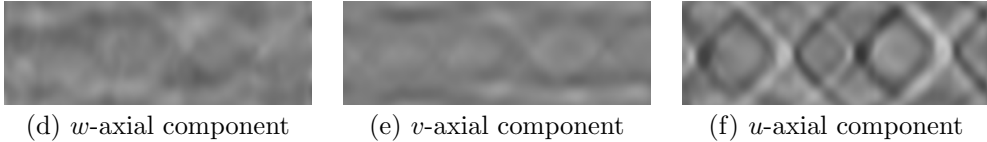


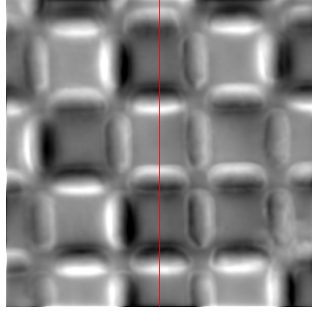
Figure 18: Reconstructions of magnetization and magnetic vector potential of a Ni-Fe sample using the conventional method and MBIR. (a-c) and (d-f) show the reconstruction of magnetization using MBIR along the $u - v$ plane and $v - w$ plane respectively. Previously, there was no algorithm in the literature for reconstructing 3D magnetization.

7 Results

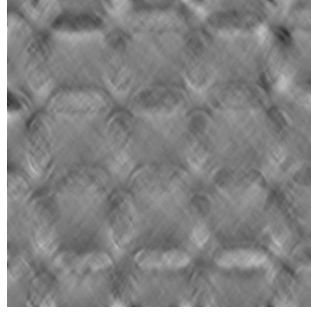
A dedicated Lorentz TEM equipped with a spherical aberration corrector was used to image a NiFe sample patterned into interacting islands using vector field electron tomography [10, 14]. At each tilt angle, the electron phase is recovered from measurements using the transport-of-intensity phase retrieval algorithm presented in [20]. The data consists of electron phase images at tilt angles ranging from -50° to 50° at steps of 1° for tilt across both the u -axis and v -axis.

Fig. 18 (a-f) shows the MBIR reconstruction of the 3D magnetization. There is no other conventional reconstruction shown since we could not find any other implementation of an algorithm that reconstructs 3D magnetization in the literature. We can see that the vertically aligned magnetic domains have magnetization oriented along the v -axis and the horizontally aligned magnetic domains have magnetization oriented along the u -axis.

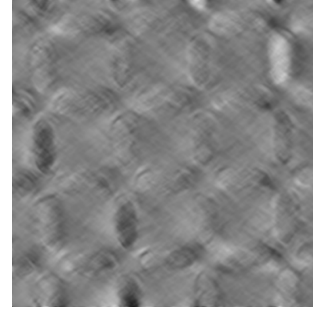
Reconstruction of Magnetic Vector Potential using Conventional Method ($u - v$ axes)



(g) w -axial component



(h) v -axial component



(i) u -axial component

Reconstruction of Magnetic Vector Potential using Conventional Method ($v - w$ axes)



(j) w -axial component

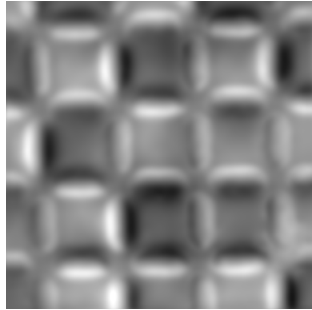


(k) v -axial component

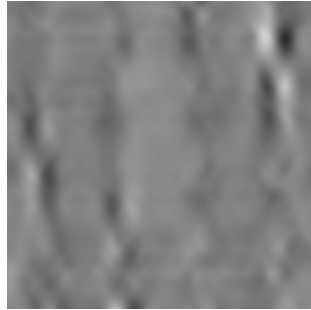


(l) u -axial component

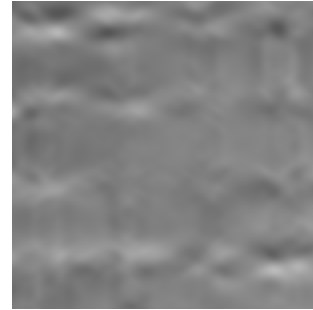
Reconstruction of Magnetic Vector Potential using MBIR ($u - v$ axes)



(m) w -axial component



(n) v -axial component

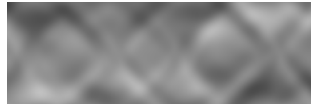


(o) u -axial component

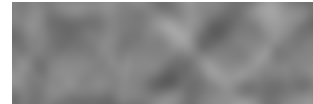
Reconstruction of Magnetic Vector Potential using MBIR ($v - w$ axes)



(p) w -axial component



(q) v -axial component



(r) u -axial component

Figure 19: (g-i) and (j-l) show the reconstruction of magnetic vector potential using the conventional method along the $u - v$ plane and $v - w$ plane respectively. (m-o) and (p-r) show the reconstruction of magnetic vector potential using MBIR along the $u - v$ plane and $v - w$ plane respectively. The 1st, 2nd, and 3rd columns show the vector field components oriented along the w -axis, v -axis, and u -axis respectively.

Fig. 19 (g-r) shows the reconstruction of the 3D magnetic vector potential using both MBIR and the conventional method presented in [10]. The conventional method results in prominent artifacts in the magnetic vector potential reconstruction of the v -axial component shown in Fig. 18 (h,k) and the u -axial component shown in Fig. 18 (i,l). Ideally, the u and v components of the magnetic vector potential must be zero. This is because the sample is engineered such that the w -axial component of magnetization is zero while the u -axis and v -axis components of magnetization do not vary along the w -axis. Hence, it follows that the u -axis and v -axis components of magnetic vector potential are zero.

8 Data Sampling and Linear Schemes for Microstructure Design with Vibrational Tuning Constraints

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Wei-keng Liao, Alok Choudhary, Veera Sundararaghavan and Ankit Agrawal*

Abstract: Microstructure sensitive design has a critical impact on the performance of engineering materials. While theoretical models for computing properties given the microstructure are known for a given alloy, inversion of these relationships to obtain microstructures that lead to desired properties is challenging, primarily due to the high dimensionality of microstructure space, multi-objective design requirement and non-uniqueness of solutions. Furthermore, the safety and performance requirements of critical components, as well as the cost of material and machining of components, make dovetailing of the microstructure based on the design objective imperative. This project addresses optimization of several microstructure design problems, and two broad sets of approaches are delineated: numerical and data-driven methods. Proposed methodologies provide schemes to overcome local optima, accelerate the optimization process as well as generate numerous optimal and near-optimal solutions that can accelerate materials development.

8.1 Motivation & Major Goals

Exploring and harnessing the association between processing, structure, properties, and performance is a critical aspect of new materials exploration. Variation in microstructure leads to a wide range of materials properties and that in turn impacts the materials performance. The performance of materials can be significantly improved by dovetailing the microstructure. Furthermore, in actual engineering problems, microstructure optimization problems are often constrained by design constraints. One of the primary goals of materials design is enhancing properties of a material in specific directions while sacrificing the properties in other directions where they are not as crucial to the design problem.

The microstructure modeling of this study is based on a one-point probability descriptor, orientation distribution function (ODF). The ODF measures the volume fractions of different orientations in a polycrystalline material. We use a finite element technique to discretize the ODF values in the Rodrigues orientation space. The finite element discretization is advantageous for design optimization since it can represent sharper textures, even single crystal designs. The goal is to find the best ODF distribution to optimize the macro-engineering material properties. Traditional techniques for materials design involves in numerical optimization techniques such as gradient based and genetic algorithms.

However, numerical methods suffer from few drawbacks. First, the efficiency of mathematical search deteriorates quickly as the candidate space grows, with a microstructure represented by hundreds of dimensions, traditional searches for microstructure design can be slow. Furthermore, these methods often suffer from getting stuck in a local optimum for non-convex constrained optimization objectives. Dimension reduction techniques are known to be influential in analyzing variable relationships in large datasets containing high variable dimensions. For optimization problems, data-driven methods can be considered to reduce the search space by limiting the number of design variables in search and finding patterns to form a superior searching strategy. Data-driven analysis from reasonable collection of variable-objective data instances could provide valuable insights towards the variable relations as well as discover directions that could lead to quick convergence.

8.2 Significant Results

This section discusses some of the notable results from the methodologies propounded as part of the project, with the first and second subsections discussing the numerical and data-driven approaches respectively.

8.2.1 Linear Schemes for Microstructure Optimization

The microstructure is modeled using the ODF values that are modeled using the finite element discretization. The problem of interest (illustrated in Fig. 20) includes a cantilever beam made of Galfenol, which has a

body-centered cubic (BCC) structure. The material is modeled using 145 ODF values, and 76 of them are independent. The design objective is determined as maximization of the yield stress while the first bending and torsional natural frequencies are constrained for vibration tuning. The main goal of the problem is to find the best microstructure design that maximizes the yield stress of the beam and satisfies the given vibration constraints.

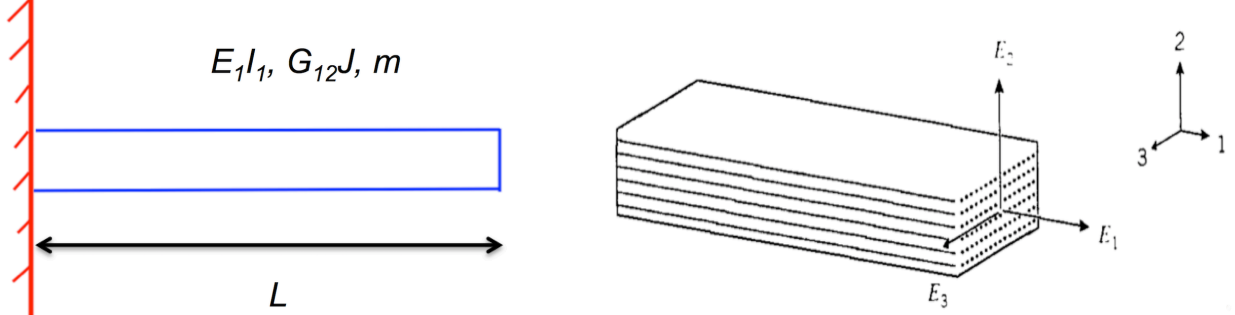


Figure 20: Geometric Representation of Galfenol Beam Vibration Problem

According to the coordinate system introduced in Fig. 20, the analytical equations of the first torsional and bending natural frequencies for an orthotropic material can be shown respectively as below:

$$\omega_{1t} = \frac{\pi}{2L} \sqrt{\frac{G_{12}J}{\rho I_p}} \quad (26)$$

$$\omega_{1b} = (\alpha L)^2 \sqrt{\frac{E_1 I_1}{m L^4}} \quad \text{and} \quad \alpha L = 1.87510, \quad (27)$$

where $G_{12} = 1/S_{66}$, $E_1 = 1/S_{11}$ and S being the compliance elements ($S = C^{-1}$). In these formulations, J is torsion constant, ρ is density, I_p is polar inertia moment, m is unit mass, L is length of the beam and I_1 is moment of inertia along axis-1. The computation of the yield stress using the upper bound approach is given in Eq. 28 respectively.

$$\langle \sigma_y \rangle = \int \sigma A dV \quad (28)$$

where A denotes the ODF and dV is the elemental volume. The mathematical formulation of the optimization problem is given below:

$$\max \quad \sigma_y \quad (29)$$

$$\text{subject to} \quad \int A dV = 1 \quad (30)$$

$$\text{subject to} \quad 19 \text{ Hz} \leq \omega_{1t} \leq 21 \text{ Hz} \quad (31)$$

$$\text{subject to} \quad 120 \text{ Hz} \leq \omega_{1b} \leq 122.5 \text{ Hz} \quad (32)$$

$$A \geq 0 \quad (33)$$

The optimization problem includes the unit volume constraint by definition (volume fractions should sum unity over the orientation space) as well as the constraints for the first natural frequencies to tune the beam vibration. To solve the problem, the length of the beam is taken as $L = 0.45$ m and the beam is considered to have a rectangular cross-section with dimensions $a = 20$ mm and $b = 3$ mm. The steps taken to optimize the microstructure are summarized below:

- The solution space is firstly reduced to a new space, called property closure. The property closure is the space of important material properties, and it includes all possible microstructure designs. Therefore the same optimization problem can be solved in a reduced space with less number of variables compared to the original solution space. The procedure for calculating the property closure is given next.

Property closures represent complete range of properties obtainable from the space of ODFs. These are approximated by the space between upper and lower bounds of the given property. Upper bound closure of stiffness values represent the range of properties obtainable by the upper bound homogenization relation. The hull maps the full range of upper bound values of a combination of stiffness values. The extreme textures were found to correspond to single crystals. A simple technique for constructing property closures (for the homogenization relations considered here) is by establishing the smallest convex region enveloping single crystal property points. Linear programming although more rigorous, is more intuitive for construction of property closures, since closures are obtained as a result of property maximization or minimization. Connecting faces on the closure may contain polycrystals that are explicitly identified by the LP approach. This approach is also well-suited for other problems, such as identification of textures with desired property combinations where several properties are optimized simultaneously. Let v_1, v_2 be the set of properties for which the closure is required. The closure for property v_1 is first found by obtaining the extremal values (v_{1max}, v_{1min}) . Then, property v_1 is discretized into m values $v_1^i, i = 1, \dots, m$ between v_{1max} and v_{1min} . The property closure of the combined set of properties (v_1, v_2) is found by executing a similar extremum LP problem at each point v_1^i with the additional constraint that $p_1^T A = v_1^i$. In general, the closure for a combined set of n properties $(v_1, v_2, \dots, v_n)$ is a n -dimensional volume found by executing an LP problem extremizing v_n at a set of discrete points $(v_1^i, v_2^j, \dots, v_{n-1}^l)$ in the closure area of $(v_1, v_2, \dots, v_{n-1})$. The corresponding LP problem for minimizing v_n is written below:

$$\begin{aligned}
\min_A v_n &= p_n^T A \text{ satisfying the constraints} \\
q^T A &= 1 \\
A &> 0 \\
p_1^T A &= v_1^i \\
p_2^T A &= v_2^j \\
&\dots \\
p_{n-1}^T A &= v_{n-1}^l
\end{aligned} \tag{34}$$

To maximize v_n another similar problem is executed where the objective is changed as $\min_A v_n = -p_n^T A$. The closure represents the range of properties obtainable when using the homogenization methodology. The infinite solutions can be represented as shown below:

$$X_i = X_1 + \lambda V_i, \text{ where } i = 1, 2, 3, 4, \dots, n \tag{35}$$

$$V_i = \text{Null}(C(:, i)) , \tag{36}$$

where Eq. 35 defines the infinite solutions, X_i , using one solution, X_1 , and null space vectors, V_i . n is the number of null space vectors. Even though the number of null space vectors is finite, the number of solutions are infinite since λ can be any number that satisfies the ODF positiveness constraint ($A \geq 0$). Since the optimization problem is solved in macro properties' space (property closure), and the macro properties' space is generated by the ODF values through averaging equations, any point inside this solution domain corresponds to a known set of ODF values. Therefore there is always at least one optimal ODF solution inside this domain. The solution strategy aims to find this optimum solution not only when it is unique but also when it is multiple.

- One solution of the problem should be computed to start the algorithm. The solution technique depends on either the problem is linear or not. For a linear problem, "one solution" can be computed solving a Linear Programming (LP) problem. However, sampling can be performed to find one solution of a nonlinear problem. The values of the microstructure dependent input parameters will be the same in all solutions if multiple solutions exist.
- As providing the same microstructure dependent property values, independent solution directions are computed using the Null Space approach of the linear solver. For a single solution problem, there is no existing solution direction since the single solution defines a point in the solution space.

- In case of having multiple solutions, these solutions are computed using "one solution" of the problem and the independent solution directions (Eq. 35).

The optimization problem of Galfenol beam vibration tuning has linear design objective and constraints. Therefore, the one solution to the problem could be found by solving an LP problem directly. The multiple solutions of this problem correspond to the designs having the same values for microstructure dependent input parameters (E_1 and G_{12}). The problem has 73 solution directions (76 optimization variables, 3 linear equations - 2 of them are for computation of E_1 and G_{12} , and 1 of them is for unit volume constraint) and these solutions are polycrystal designs. The parameters of the multiple optimum solution are given and compared to the best single crystal solution in Table 6. Some of the optimum microstructure designs are shown in Fig. 21. Since the linear solver was able to compute independent solution directions for Galfenol beam optimization problem, each design in Fig. 21 is different than the others and has different ODF values. However, they are still providing an identical maximum yield stress value and satisfying the design constraints.

Parameters	Optimum Polycrystal	Best Single Crystal
σ_y	384.1256 MPa	352.7132 MPa
E_1	210.2585 GPa	215.2910 GPa
G_{12}	78.4528 GPa	78.0022 GPa

Table 6: Optimization Results for Vibration Tuning of the Galfenol Beam

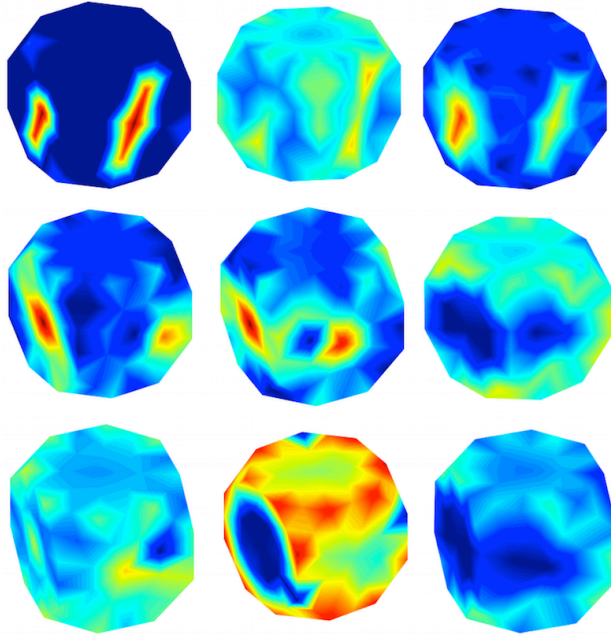


Figure 21: Some of the Optimum ODF Solutions

8.2.2 Data-Driven Microstructure Optimization

Galfenol Microstructure Optimization This work [42] attempts to identify the complete space of microstructures for yielding the desired combination of properties demanded by a given application. The problem involves microstructure design of magnetoelastic Fe-Ga alloy (Galfenol) for enhanced elastic, plastic and magnetostrictive properties. While theoretical models for computing properties given the microstructure are known for this alloy, inversion of these relationships to obtain microstructures that lead to desired

properties is challenging. The challenges are primarily due to the high dimensionality of microstructure space, complexity of the constraints and non-uniqueness of solutions. These challenges render traditional search-based optimization methods insufficient concerning both searching efficiency and result optimality. In this work, a route to address these challenges using a machine learning methodology is proposed. A systematic framework consisting of random data generation, feature selection, and classification algorithms are developed. Experiments with five design problems that involve identification of microstructures that satisfy both linear and nonlinear property constraints show that our framework outperforms traditional optimization methods with the average running time reduced by as much as 80% and with optimality that would not be achieved otherwise.

Table 22 illustrates the flow diagram of this methodology, and it consists of four steps (a) Random Data Construction (b) Search Path Refinement (c) Search Path Reduction (d) Enhanced Optimization. The dataset is generated using several randomization methods that exhaustively sample the search space. The motivation of introducing feature ranking into optimization is to obtain a specific search path in the form of a sorted order of variables before the start of a search, to improve the searching efficiency. For this problem, feature ranking is determined by voting across the ranks across four feature ranking algorithms. To reduce the search region of each variable, we build a rule-based classification tree to learn the most promising region of values for each variable. The data instances with desired function values are represented by the class ‘1’ and the contradictory class is labeled as ‘-1’. After a tree is constructed, we look for the leaf nodes with “-1”. We traverse from the root to each of the “-1” leaf nodes, and rules describing the are generated along the path. The searching effort is thus reduced to a more concentrated area on these variables. Compared to the original region of $[0, 1]$, the search region has been reduced significantly. A gradient-based line search can be conducted on an ordered list of variables, finding, one variable at a time, the value of it that optimizes the function from a reduced value space. Optimization becomes a much promising endeavor when the search space is reduced, and a pre-planned searching path is deployed.

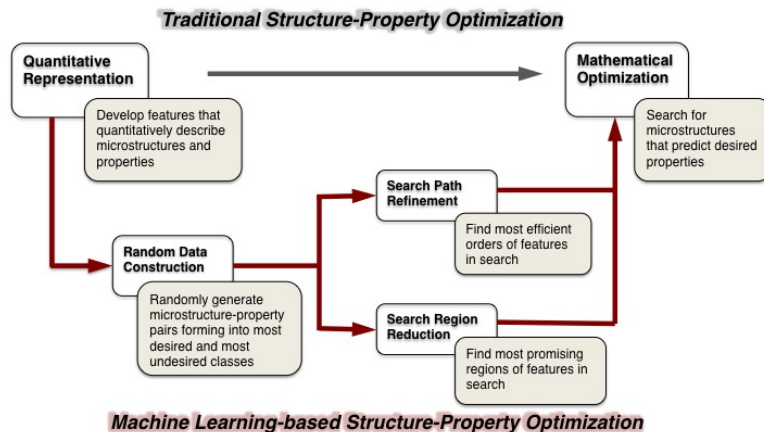


Figure 22: Framework of material structure optimization. The flow on top is the traditional search-based mathematical optimization method. The bottom is the machine learning based method we propose. Three additional steps are inserted to learn a refined and reduced search space.

The three baseline methods to compare with are: 1) an exhaustive search (eSearch) containing 1 million random searches, 2) a generalized pattern search, which can be considered as a smarter-than-exhaustive guided search (gSearch), 3) a traditional optimization algorithm, specifically linear programming (LP) is used for linear problems and genetic algorithms (GA) for nonlinear problems. Table 7 shows the comparison of results obtained by the aforementioned methods. As we can deduce from this comparison, for linear properties as yield stress (Y) and magnetostrictive strain (m), LP always provides a valid solution. It is the nonlinear problems such as Young’s modulus (E) and composite functions F_1 and F_2 that pose a more significant challenge for traditional optimization methods. Exhaustive search gives unstable results. Guided search tend to get stuck at local optima, which can sometimes be even worse than the naive exhaustive search (although the running time is much less). Surprisingly, GA has worked poorly on nonlinear problems (we also tried GA on linear problems, and the answer is never as good as LP). In fact, for nonlinear problems,

the guided search is often a better choice than GA.

Table 7: Comparison summary of optimal values for different design problems across methods

Property	Objective	eSearch	gSearch	LP/GA	ML
E (GPa)	Min	127.56	89.5667	136.25	85.9878
Y (MPa)	Max	301	306.54	353.11	353.11
m	Max	9.0034e-5	1.3797e-4	1.5498e-4	1.5498e-4
F_1	Max	1.0911e-7	1.2609e-7	1.0122e-7	2.9347e-7
F_2	Max	9.81	10	9.9987	10

A machine learning (ML)-based guided search aims to reduce the search space, that is, the set of candidate solutions, by using heuristics specific to the problem class, and was able to achieve a fine balance between accuracy and efficiency, and more importantly, it can find the complete set of solutions that none of the other methods can. Furthermore, a software has been developed based on this work, which can be found in <http://cucis.ece.northwestern.edu/projects/MURI/workopt.html>. It is a general optimization package that performs search space reduction given an objective function and mathematical constraints.

Vibrational Tuning of Galfenol This work [44] addresses optimization of microstructure design to maximize the yield stress of a Galfenol beam under vibration tuning constraints defined for the first torsional and bending natural frequencies. The presence of vibrational constraints to the design objective renders the previous methodology of guided pattern search infeasible. Pattern search approaches the optimal point by creating a mesh from a sequence of satisfying points, and is not able to converge for this problem as a mesh satisfying the constraints is not attainable. A two-step data-driven solution scheme 23 is proposed to find optimal microstructure satisfying performance requirements, and design and manufacturing constraints. The first phase of the approach involves developing and executing sampling algorithms to generate possible ODF solutions meeting the process limitations. The sampling algorithms i.e. *partition* and *allocation* scheme complement one another and ensure sampling the entire feature space. *Partition* warrants that different permutations of non-zero ODF dimensions are explored for a given set of ODF dimension. *Allocation* guarantees that all the ODF dimensions are explored sufficiently. There are two sets of objectives (Equations 1 and 2) with different vibrational constraints that were explored in this work. The flow-diagram for the proposed methodology is illustrated in Figure 23.

First set of constraints:

$$\text{subject to } 19.5 \text{ Hz} \leq \omega_{1t} \leq 21.5 \text{ Hz} \quad (37a)$$

$$\text{subject to } 120 \text{ Hz} \leq \omega_{1b} \leq 122.5 \text{ Hz} \quad (37b)$$

Second set of constraints:

$$\text{subject to } 21.5 \text{ Hz} \leq \omega_{1t} \leq 23.5 \text{ Hz} \quad (38a)$$

$$\text{subject to } 100 \text{ Hz} \leq \omega_{1b} \leq 114 \text{ Hz} \quad (38b)$$

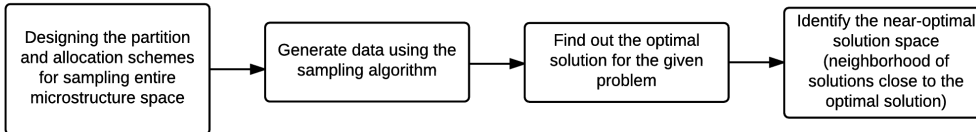


Figure 23: Flow diagram of the proposed methodology. Upper and lower bound approaches for both sets of constraints are repeated for both problems.

We evaluate the proposed data-driven approach in yielding optimal and near-optimal solutions and find that it outperforms or matches previous state-of-the-art methods and produces numerous near-optimal solutions which is one of the most significant contributions of this study. Table 8 presents the total number of near-optimal solutions, or in other words, solutions that are proximal to the optimal solutions. The near-optimal solutions of this problem correspond to different designs having same or similar values for yield

stress. The algorithms were executed to produce around 5 million valid (which obey all the constraints) solutions. For all the four problems (upper and lower bound approaches for two sets of constraints), 3-9 near-optimal solutions with a neighborhood of 10^{-4} (from the optimal solution) are discovered. Further, between 89-402 solutions in a neighborhood of 10^{-3} and between 147-1579 solutions in a neighborhood of $5 \cdot 10^{-3}$, across all the categories are identified. For the upper and lower bound approaches, our solutions are compared against the genetic algorithm based scheme and LP-based methods respectively.

The proposed data sampling approach based on the sampling algorithms surpassed the yield stresses obtained from genetic algorithm based solver for the upper bound approach (as shown in Table 9). In particular, we get an improvement of more than 25% for upper bound approach on the first set of objectives against the previous state-of-the-art approach. Additionally, the results for the lower bound are comparable to the optimal values achieved by the LP method (Table 10). It is important to note that only the LP solution (used for the lower bound approach by Acar et al. [37]) yields the theoretical maximum value in contrast to the genetic algorithm solver scheme used by them for the upper bound approach [36]. As described before, obtaining multiple optimal solutions are critical as traditional low-cost manufacturing processes can only generate a limited set of microstructures. While a single solution may not be economically feasible to manufacture, hundreds or thousands of near-optimal solutions can accelerate the speed of materials development. Therefore, it provides flexibility to produce solutions which are cost-effective selectively, and improve the overall efficiency of manufacturing immensely.

Table 8: Number of solutions within 0.01%, 0.1% and 0.5% of the optimal solutions. For each set of constraints (5 million valid data points were generated).

Constraint	Bound	within 0.01%	within 0.1%	within 0.5%
1	Upper	3	89	147
1	Lower	9	92	222
2	Upper	7	402	2015
2	Lower	3	116	1579

Table 9: Comparison of the maximum yield stress achieved for the 2 sets of constraints with the proposed approach and the previous state-of-the art genetic algorithm solver (GA) [36] approach for microstructure design with process constraints(upper bound Equations 37a, 38a). The yield stress σ_y , bending ω_{1b} and torsional ω_{1t} frequencies of the optimal solutions generated by both methods. The units for yield stress σ_y is MPa and the frequencies is Hz.

Constraint	Bound	σ_y (current)	σ_y (GA)	ω_{1b} (current)	ω_{1b} (GA)	ω_{1t} (current)	ω_{1t} (GA)
1	Upper	385.237	384.126	120.006	120.210	21.341	21.498
2	Upper	388.089	308.446	102.589	113.918	23.482	23.485

Table 10: Comparison of the maximum yield stress achieved for the 2 sets of constraints with the proposed approach and the previous state-of-the-art LP [37] approach for the microstructure design with process constraints (lower bound Equations 37b, 38b). The yield stresses σ_y , bending ω_{1b} and torsional ω_{1t} frequencies of the optimal solutions generated by both methods. The units for the yield stress σ_y is MPa and the frequencies is Hz.

Constraint	Bound	σ_y (current)	σ_y (LP)	ω_{1b} (current)	ω_{1b} (LP)	ω_{1t} (current)	ω_{1t} (LP)
1	Lower	385.113	385.650	121.272	120.020	21.344	21.500
2	Lower	387.134	387.259	106.519	100.000	23.477	23.499

Optimization techniques including the methods used by Acar et al. such as a genetic algorithm [36] or linear programming based scheme [37] lead to a unique microstructural solution or sometimes a few. One of the limitations of their approaches for vetting equivalent solutions is that it only searches for identical optimal value. However, for practical design applications, a near-optimal solution is adequate as long as the constraints are strictly obeyed, and the near-optimal solutions are proximal to the optimal solution. A future direction for reducing the dimensionality of microstructure space is highlighted that can accelerate

the process of achieving solutions satisfying all the constraints by isolating ODF dimensions that are mostly non-zero across a majority of near-optimal ODF solutions.

ML-Guided Optimization of α -Titanium In this work, we explore the microstructure optimization of multiple design problems for a Titanium panel. Two different mesh sizes to represent ODFs are investigated in this proposed work - 50 and 388. Three separate properties: coefficient of expansion α , stiffness coefficient C_{11} and yield stress σ are maximized, and four different design problems explored. The dimensionality of the search space increased considerably for a mesh size of 388 compared to the previous vibration tuning problem where the mesh size for Galfenol was 76. An ML-assisted sampling scheme was implemented where we use random forests to narrow down the search space by iteratively selecting the ODF dimension groups that deliver the maximal values. Figure 24 delineates the flow diagram of the optimization methodology.

Table 11 depict the number of near-optimal solutions obtained for one of the design problems. For the upper and lower bound approaches, our solutions are compared against the genetic algorithm based scheme and LP-based methods respectively. The proposed data sampling approach based on the sampling algorithms surpassed the yield stresses obtained from genetic algorithm based solver for the upper bound approach. Additionally, the results described in Table 11 for the lower bound are comparable to the optimal values achieved by the LP method. The proposed approach generates numerous near-optimal solutions (about 10^2 - 10^6 , i.e., 3 – 4 orders of magnitude higher than prior methods).

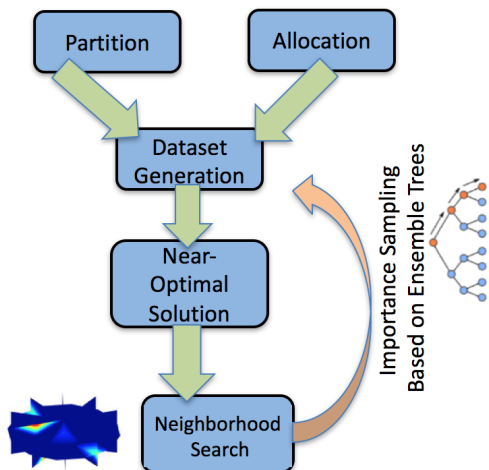


Figure 24: Flow diagram of our methodology. The green arrows depict the data generation process, and the orange arrow signifies the feedback-aware sampling.

Table 11: Number of solutions within 0.01%, 0.02%, 0.05% and 1% of the optimal solutions.

Bound	Mesh Size	ML-Guided Sampling			
		within 0.01%	within 0.02%	within 0.05%	within 1%
Upper	388	140	280	759	1.255×10^3
Lower	388	0	6.223×10^3	1.078×10^5	1.084×10^5

The proposed work provides a future direction for feedback aware sampling that can iteratively incentivize distinct ODF dimensions using feature selection using machine learning. This would yield ODF vectors with higher objective value, which can be investigated to accelerate the process of attaining optimal or near-optimal solutions.

9 Algorithms for the design of functional materials

Kaushik Bhattacharya, Marc De Graef and Richard D. James

Abstract As part of this project, the PIs have developed a suite of algorithms for the discovery of functional materials. These are adapted for the systematic discovery of functional materials based on highly reversible first-order phase transformations between one crystalline phase and another, which also can be accompanied by an abruptly changing polarization, magnetization or transport property. The algorithms developed under the project enable the seamless passage from high accuracy powder or micro-Laue X-ray measurements of crystal structure and lattice parameters to the discovery of a new highly reversible alloy or oxide, by a procedure that is described in this final report.

9.1 Introduction

Multiferroics are materials that combine at least two of the three “ferroic” properties ferroelectricity, ferromagnetism, and ferroelasticity. Despite an intense search over the past decade, there has been limited success in finding *single phase* (“intrinsic”) multiferroic compounds that exhibit strong ferroelectricity and ferromagnetism above room temperature, and are switchable. In the Mosaic MURI project we have instead followed a design route made possible by theoretical and algorithmic advances in the project: *multiferroic response driven by reversible phase transformation* (Figure 25). In this case elasticity, electricity, magnetism and temperature are coupled, and can be simultaneously controlled along with transport and optical properties. Such a broad combination creates a host of possibilities for novel sensors, actuators, microelectronic and optical devices, information storage media, energy storage media, magneto-electro-caloric and energy conversion devices.

The pervasive bottleneck with phase transformations, especially strong first order phase transformations, is reversibility. Fracture often occurs after a few cycles. Even the most reversible such alloy, binary NiTi, has significant migration of transformation temperature, typically $\sim 20^\circ\text{C}$ after a few hundred cycles, and exhibits complete failure after a few thousand cycles under stress-induced transformation with stresses as in Figure 26 or 27.

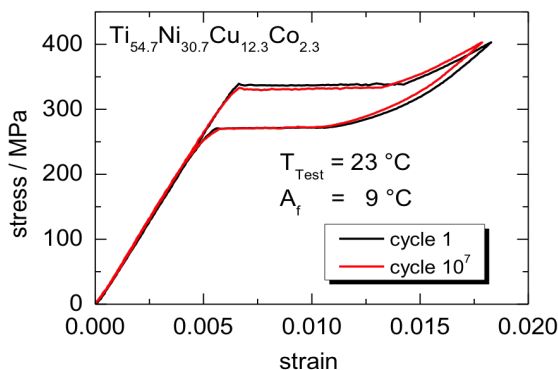


Figure 26: Demonstration [27] of reversibility of stress-induced full phase transformation, under demanding conditions of 400 MPa peak *tensile* stress (each cycle), in a TiNiCuCo alloy. Cycle 1 is plotted on the same diagram as cycle 10 million. This alloy satisfies to high accuracy the cofactor conditions [25, 17]

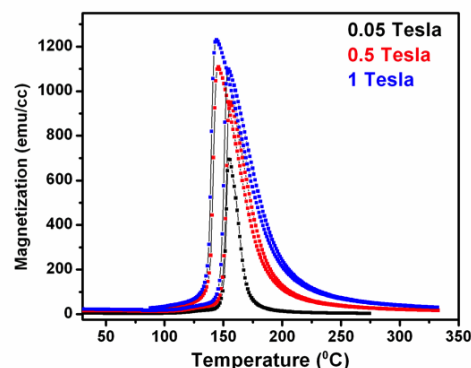


Figure 25: Multiferroism in the alloy $\text{Ni}_{45}\text{Co}_5\text{Mn}_{40}\text{Sn}_{10}$ [33]. Note the strong magnetism of the high temperature phase of this ferroelastic: pure iron has a magnetization of 1400 emu/cm^3 at 150°C . The high temperature phase also has low coercivity and high permeability.

As a direct result of work on the Mosaic MURI project, this limitation has been breached. Two alloys have emerged ([27, 25, 30] and Figures 26, 27) that demonstrate a new route to the reversibility of phase transformations, resolving the key limitation of “multiferroism by phase transformation”. These two alloys closely satisfies the *cofactor conditions* [2] derived previously by James and collaborators and studied extensively under the MURI project. The cofactor conditions are nongeneric conditions of compatibility between phases (“supercompatibility”) within the Geometrically Nonlinear Theory of Martensite, a theory developed by James and collaborators. In Figure 27 note the extreme behavior: *the peak stress is of the order of the yield stress of a Ni superalloy, the strain is an order-of-magnitude larger than the yield strain of such an alloy, and yet it exhibits 100,000 cycle reversibility with full recovery.*

A key aspect of this breakthrough [27, 16, 25] is that theory preceded synthesis. Theory identified special conditions on lattice parameters at which singular behavior is expected. Compositional changes were then designed to achieve the particular compositions at which these special conditions are satisfied. The whole procedure was enabled by the algorithms developed under the project and summarized here.

9.2 Supercompatibility

To summarize the state-of-the-art at this time, there are two levels of conditions of compatibility that have yielded record low hysteresis in strong first order phase transformations and record high levels of reversibility measured by cyclic repeatability of properties. These two conditions are:

$\lambda_2 = 1$ [6, 7, 12, 13]. Here, λ_2 is the middle eigenvalue of the transformation stretch tensor [35]. Satisfying $\lambda_2 = 1$ to high accuracy (1.0000 ± 0.0002) in a transforming material leads to exceptionally low hysteresis (Figure 29), with the width of the thermal hysteresis loop, $(1/2)(A_s + A_f - M_s - M_f) < 2^\circ \text{ C}$. Accurate satisfaction of $\lambda_2 = 1$ has been demonstrated in several metal alloy systems, including NiTiX alloys and various Heusler alloys (Figure 29). This is done by preparing alloys in either bulk or film form, accurate measurement of lattice parameters by X-ray methods, calculation of λ_2 using the Cauchy-Born rule, and then systematically changing composition and repeating these steps to achieve $\lambda_2 = 1$. The indications are that the reversibility of the transformation as measured by the migration of transformation temperature during cyclic transformation is also significantly improved by tuning the composition to make $\lambda_2 \approx 1$.

However, from the recent demonstrations, Figures 26,27, the cofactor conditions (see below) appear to have a stronger effect on reversibility. The physical meaning of $\lambda_2 = 1$ is that it allows for a perfect, untwinned interface between the phases, and therefore eliminates the stressed transition layer that ordinarily exists there. Low hysteresis of strong first order phase transformations enable the realization of multiferroic energy conversion devices and new concepts for sensors.

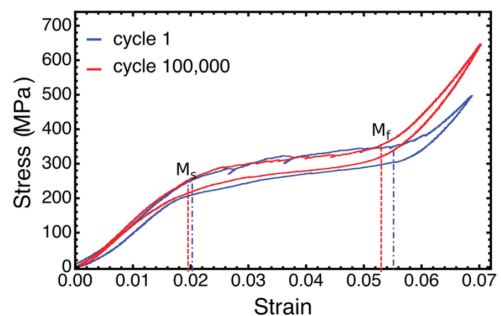


Figure 27: Demonstration [30] of 100,000 cycle reversibility of full phase transformation, under $> 500 \text{ MPa}$ peak compressive stress, $> 6\%$ strain on 2 micron pillars of a ZnAuCu alloy satisfying accurately the cofactor conditions. Note also the low stress hysteresis.

Cofactor conditions ([17, 16, 25, 6]). The *cofactor conditions* include the condition $\lambda_2 = 1$, as well as a second condition on lattice parameters, and an inequality (which is typically satisfied). The physical meaning

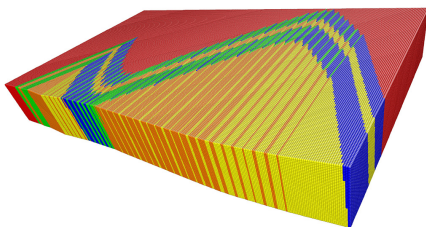


Figure 28: A predicted complex microstructure possible under the cofactor conditions. Austenite is red. The martensite consists of four variants. In each region the crystal structure is at its stress-free, minimum free energy state and exhibits large deformation, yet the regions fit together perfectly. Similar microstructures are seen in $\text{Zn}_{45}\text{Au}_{30}\text{Cu}_{25}$ [16].

of the cofactor conditions is that they are necessary and sufficient conditions that the crystallographic theory of martensite is satisfied *for any volume fraction of the twins* and also for many twin systems. The crystallographic theory of martensite is a broadly successful theory that describes in detail the low energy twinned and stressed interface between two crystalline phases. The cofactor conditions imply the existence of a great many additional microstructures involving both phases which have either low energy transition layers (in the sense of the crystallographic theory) or zero elastic energy [17]. An example is shown in Figure 28 which, despite the large distortions, contains zero elastic energy and exhibits perfect fitting. These zero-elastic-energy microstructures include nucleation mechanisms in which a nucleus of the martensite phase grows continuously from zero volume, and other zero-energy microstructures where the austenite grows continuously from zero volume

within a twin band of martensite [17]. In all these cases there is flexibility: some volume fractions can be freely changed keeping the microstructure at zero stress.

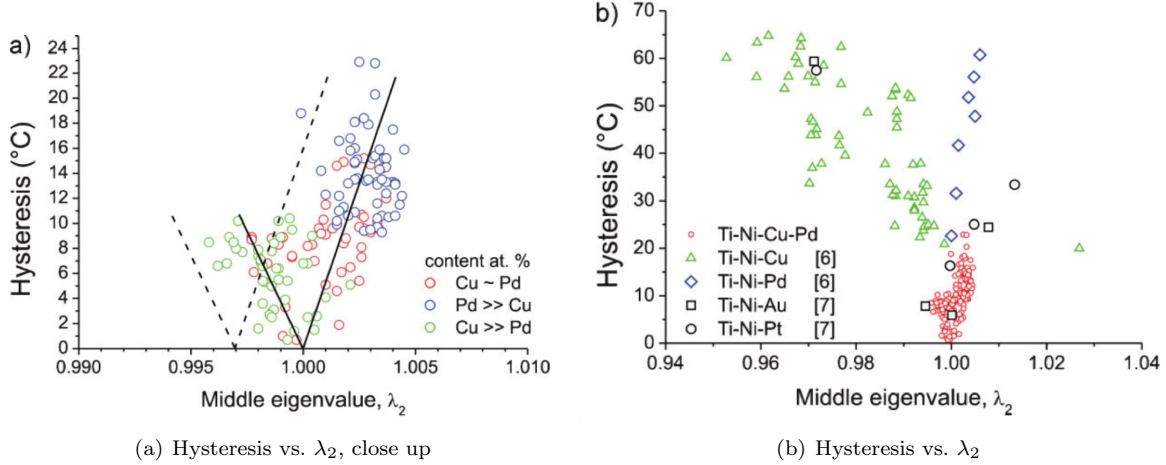


Figure 29: Measured thermal hysteresis vs. λ_2 in alloys from a combinatorial library (red and triangles) and from bulk alloys of TiNi(Cu, Pd, Pt, Au) [7, 12, 13]. Each marker corresponds to a different alloy. Figure (a) is a close-up of (b) near $\lambda_2 = 1$. Note that the size of the hysteresis can be reduced to near zero by tuning composition to make $\lambda_2 = 1$.

While many groups worldwide are currently pursuing these conditions in various alloy systems, currently, there are two alloys known that accurately satisfy the cofactor conditions: $\text{Zn}_{45}\text{Au}_{30}\text{Cu}_{25}$ ([16] and Figure 27) and the alloy shown in Figure 26, $\text{Ti}_{54.7}\text{Ni}_{30.7}\text{Cu}_{12.3}\text{Co}_{2.3}$ [27]. The first, $\text{Zn}_{45}\text{Au}_{30}\text{Cu}_{25}$ [16], was developed in the laboratory of James by systematic changes of composition. It exhibits the lowest hysteresis $(1/2)(A_s + A_f - M_s - M_f) \approx 0.2^\circ\text{C}$ measured in a bulk alloy with a strongly first order phase transformation ($> 5\%$ transformation strain)¹. It also exhibits many unusual features: nonreproducibility of microstructure from cycle to cycle, independence of microstructural changes across a grain boundary during cyclic transformation, an unprecedented shape memory effect at small scales (Figure 27), and complex microstructure but with few stressed transition layers [34]. It also exhibits exceptional reversibility under thermal or stress cycling [16, 30].

A fascinating aspect of $\text{Zn}_{45}\text{Au}_{30}\text{Cu}_{25}$ is its behavior at small scales [30]. In typical cases of pillar compression experiments with ≈ 1 micron pillars, such as those in NiTi [9], transformation is accompanied by the formation of a single large shear band having sharp re-entrant corners, and failure occurs after a few cycles. In contrast, in the cofactor alloy $\text{Zn}_{45}\text{Au}_{30}\text{Cu}_{25}$ the 2 micron pillar of the experiment of Figure 27 showed complex fine microstructures and a smooth boundary during transformation *even at micron scales* [30]. Evidently, interfaces cost so little energy, and there are so many possibilities, that bulk-like microstructures are observed even at scales at which interfacial and bulk transition layers are thought to be prohibitive.

9.3 Main algorithms

A flow chart of the alloy development procedure for supercompatible alloys is shown in Figure 30. This is schematic with regard to method of synthesis and characterization: sputtered (or MBE grown) films [27] or cold crucible methods sometimes replace arc melting, and micro-Laue methods replace powder X-ray methods [34]. Algorithms developed under the project are now being used for all the arrows in this figure, but the most critical (and most difficult) steps are X-ray methods $\rightarrow$ Lattice parameters $(\alpha, \beta, \gamma, \delta) \rightarrow$ Transformation stretch matrix $\rightarrow$ Variants $\mathbf{U}_1, \dots, \mathbf{U}_n$ and twin systems. The key problem is that X-ray methods give – at best and with considerable difficulty in these low symmetry

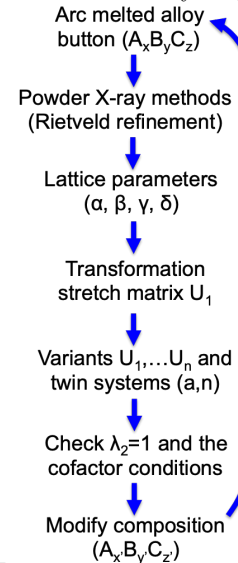


Figure 30: Flow chart of alloy discovery procedure. Each arrow is enabled by algorithms developed under the project.

¹This alloy has about 6% transformation strain.

systems – the primitive unit cell (fundamental periodicity) and space group².

In practice the primitive unit cell of the low symmetry martensite has much larger volume than the primitive unit cell of austenite. In this case a *sublattice unit cell* of austenite (having about the same volume as the martensite primitive unit cell) transforms to the primitive unit cell of martensite. But the ratio of volumes of the sublattice unit cell of austenite to the primitive unit cell of austenite can be large; this ratio is approximately 18 in the case of the cubic to monoclinic transformation in $\text{Zn}_{45}\text{Au}_{30}\text{Cu}_{25}$. The atoms within the (potentially large) sublattice unit cell of austenite shuffle to new positions in the martensite lattice.

Thus, there are two problems. First, there are a huge number of sublattices of austenite that have unit cells with about the same volume as that of martensite. Second, given one of them, there are infinitely many unit cells for that sublattice. Which one of them transforms to a given primitive unit cell of martensite? This is a classic problem in phase transformations that was already faced by Bain, who theorized that one should seek “a mode of atomic shift [that] requires minimum motion” [1]. His conjectured solution, the well-known Bain strain, is accepted today in a number of cases, but also is not the correct solution for many transformations, particularly in complex lattices.

As part of the Mosaic MURI project, we have developed an algorithm ([35], see also structrans.org) that rigorously finds the “a mode of atomic shift [that] requires minimum motion”, interpreted in a precise and physically sound way. “Minimum motion” is defined by a norm. Thus, given only results that come from the X-ray methods, the algorithm searches out all sublattices of austenite, and finds those with minimum atomic motion. Despite the fact that there are an infinite number of sublattices, the algorithm finds the exact minimum, because we first establish a rigorous, quantitative upper bound which reduces the search to a large but fine number of sublattice unit cells.

This algorithm is enhanced by calculations of λ_2 and the three cofactor conditions, calculation of all twin systems, calculation of all solutions of the crystallographic theory of martensite, and accurate display of these results (see structrans.org), also including unusual microstructures possible under the cofactor conditions.

Technology areas that may be initiated or impacted by this research are listed in Table 1.

Table 1: Potential DoD Technology Areas Enabled by Transformational Multiferroic Materials with Multi-Million Cycle Reversibility (expanded from Science **348**, p. 969)

Technology	Relevant Multiferroic Properties	Potential Applications
Actuation	Shape memory effect	Vehicle actuators Aerospace (UAV) actuators Robotic actuators Remote field-induced, medical devices Combustion chamber actuation (high T)
Sensing	Magnetoelectric properties Switching of dielectric tensor Switching of susceptibility	Mechanical, electrical, magnetic sensors Optical sensors, imaging, smart windows Power electronics
Information storage	Magnetoelectric properties	Magnetic bit switching by an electric field
Switches	Multiferroic properties	Electromagnetic and optical switches Active metamaterials
Direct energy conversion	Magnetization Polarization	Solar-thermal heat to electricity Waste heat conversion in digital devices Powering spacecraft
Energy storage power electronics	Antiferroelectric to ferroelectric	Compact energy storage capacitors
Solid-state cooling	Magnetization Polarization	Magnetocaloric refrigerators Electrocaloric refrigerators

²In practice the space group is guessed and fitting is done. The quality of the fit then suggests the most likely space group.

10 Markov random fields for microstructure synthesis: examples and validation

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S. Kalidindi, M. DeGraef, V. Sundararaghavan

Abstract: In this MURI task, we have pioneered an new methodology based on Markov Random Fields (MRFs) for quantification and reconstruction of material microstructures. In the MRF approach, microstructures are quantified using the conditional probability density for the state of a pixel given the known states of its neighboring pixels. If only the nearest neighbors are chosen, this amounts to modelling a microstructure as an Ising graph. While in Ising models, a lattice is constructed with pixels (with binary states) interacting with its nearest neighbors, in MRFs, pixels take up integer or vector states and interact with multiple neighbors over a window. The sampling of conditional probability of a pixel given the states of its known neighbors is based on a Markov chain. In the sampling method, a set of neighbor pixels are used as a template to determine the probability distribution function (PDF) of the unknown pixel[32]. To improve the computational efficiency, an alternate methodology based on optimization was also validated. The approach minimizes a neighborhood cost function that ensures that the local neighborhood of the Ising lattice taken along the x-,y- or z- directions through the 3D microstructure is similar to some neighborhood in the 2D lattice imaged along that plane. This reconstruction problem leads to anisotropic microstructures that have similar high order statistics[19], which is in contrast to other such works in literature that use assumptions of microstructural isotropy. The sampling approach and the optimization approach can also be applied in tandem for modeling time varying microstructures[38]. In this report, we primarily illustrate examples from this method as applied to spatial and temporal microstructure reconstruction.

10.1 Introduction

Some of early attempts at microstructure modeling were based on Ising models. In the Ising model, a $N \times N$ lattice (L) is constructed with values X_i assigned for each particle i on the lattice, $i \in [1, \dots, N^2]$. In an Ising model, X_i is a binary variable equal to either $+1$ or -1 (eg. magnetic moment). In general, the values X_i may contain any one of G color levels in the range $\{0, 1, \dots, G-1\}$ (following the integer range extension of the Ising model). A *coloring* of L denoted by $\mathbf{X}$ maps each particle in the lattice L to a particular value in the set $\{0, 1, \dots, G-1\}$. Ising models fall under the umbrella of *undirected graph models* in probability theory. In order to rewrite the Ising model as a graph, we assign neighbors to particles and link pairs of neighbors using a bond as shown in Fig. 31(a). The rule to assign neighbors is based on a *pairwise Markov property*. A particle j is said to be a neighbor of particle i only if the conditional probability of the value X_i given all other particles (except (i, j) , i.e., $p(X_i | X_1, X_2, \dots, X_{i-1}, X_{i+1}, \dots, X_{j-1}, X_{j+1}, \dots, X_{N^2})$) depends on the value X_j .

In the classical Ising model, each particle is bonded to the next nearest neighbor as shown in Fig. 31(a). For modelling microstructures, a higher order Ising model (Fig. 31(b)) is used. The particles of the lattice correspond to pixels of the 2D microstructure image. The neighborhood of a pixel is modeled using a square window around that pixel and bonding the center pixel to every other pixel within the window. Using this graph structure, a *Markov random field* can be defined as the joint probability density $P(\mathbf{X})$ on the set of all possible colorings $\mathbf{X}$, subject to a *local Markov property*. The *local Markov property* states that the probability of value X_i , given its neighbors, is conditionally independent of the values at all other particles. In other words, $P(X_i | \text{all particles except } i) = p(X_i | \text{neighbors of particle } i)$. The theory to sample from the conditional probability density $p(X_i | \text{neighbors of voxel } i)$ can be found in References [19, 32, 38]. In this chapter, we describe various example applications of this approach including generation of synthetic 2D and 3D microstructures and modeling temporal evolution of 2D microstructures using experimental data.

10.1.1 Example 1: Reconstruction of a Polycrystal

A polycrystalline structure was chosen for reconstruction. The free parameter in the reconstruction is the sampling window size which is taken to be $w = 13 \times 13$ pixels.

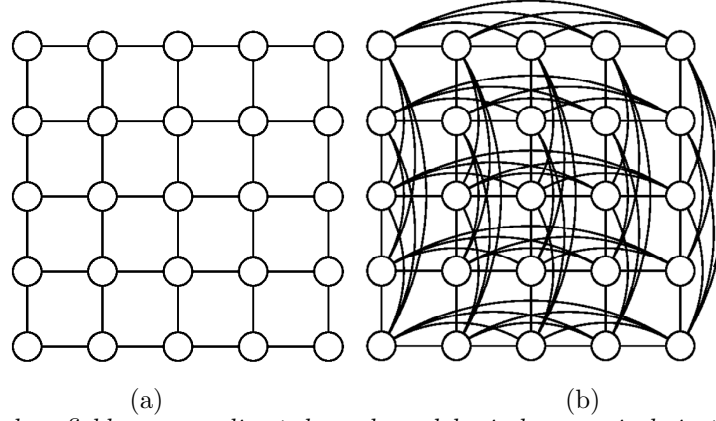


Figure 31: *Markov random field as an undirected graph model, circles are pixels in the image and bonds are used to connect neighbors: (a) Ising model with nearest neighbor interactions (b) Microstructure modeled by including higher order interactions in the Ising model.*

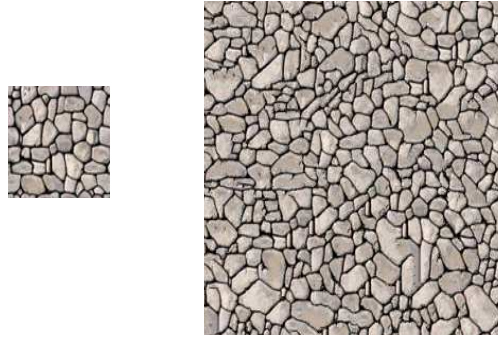


Figure 32: *Initial microstructure (left) and the synthesized microstructure (right) from our Markov random field code. Note that local features such as grain boundaries are effectively captured.*

We compared descriptors that describe the shape distribution of individual grains in the experimental and synthesized grains in Example 1, rather than the global (average) feature represented in the histograms of Fig. 33. Moment Invariants (MIs) are non-linear combinations of moments of an object shape that are invariant with respect to a class of coordinate transformations. For 1D distribution of data, the second order moment is analogous to standard deviation while the 4th order moment invariant is similar to kurtosis. For 2D shapes, DeGraef and coworkers have identified 2 MI's (ω_1, ω_2) of second order. Graphical representation for these two second order moment invariants in a x-y plot is known as Second Order Moment Invariant map (SOMIM). The SOMIM for the image in Fig. 32 and its reconstruction using MRFs is shown in Fig. 33. To compare the SOMIMs, we need to introduce an appropriate similarity metric. The modified Bhattacharya coefficient $H(p, q)$, also known as Hellinger distance, provides a metric to distinguish between two different density maps p and q . The regular Bhattacharya coefficient $\beta(p, q)$ is a measure of the similarity between two normalized distributions and can be written in discrete form as

$$\beta(p, q) = \sum_{i=1}^N \sqrt{p(i)q(i)}, \left(\text{with } \sum_{i=1}^N p(i) = \sum_{i=1}^N q(i) = 1 \right) \quad (39)$$

where the summation runs over the N bins of the SOMIM. The larger the value of β , more similar the two distributions are. The Hellinger distance $H(p, q)$ is defined as

$$H(p, q) = \sqrt{1 - \beta(p, q)} \quad (40)$$

Value of H for SOMIM is 0.47 for the polycrystalline case in Example 1. This value indicates good reproduction of polycrystalline shapes using the MRF synthesis approach.

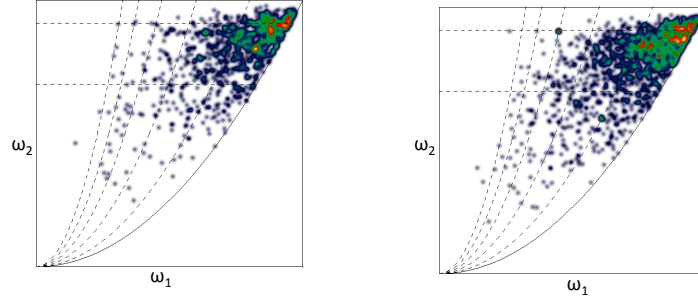


Figure 33: *SOMIM density map original(left) and synthetic(right) microstructure*

10.1.2 Example 2: 3D reconstruction of a two phase composite

The properties of the reconstructed image was studied based on an experimental dataset. The author provides a high resolution planar microstructure image (Fig. 34(a)) of a silver-tungsten composite with porous tungsten matrix and molten silver (volume fraction of silver phase $p = 20\%$). A 657×657 pixel region of the microstructure corresponding to $204 \mu\text{m}$ square area was converted to a black and white image for distinguishing the two phases. This was done by selecting a threshold color below which phases were set to white (the silver phase) and the rest of the image was set to black (the tungsten phase). The final black and white image is shown in Fig. 34(a)(inset). A $64 \mu\text{m}$ square cell within this image was chosen to reconstruct the 3D image.

An instance of the reconstructed microstructure is shown in Fig. 34(b,c) with the distribution of each phase shown separately. The two-point correlation measure, $S_{(2)}^i(r)$, of this image can be obtained by randomly placing line segments of length r within the microstructure and counting the fraction of times the end points fall in phase i . The auto-correlation function for the silver phase $\gamma(r) = \frac{S_{(2)}^1(r) - p^2}{p - p^2}$ of the reconstructed 3D microstructure and the experimental image are compared in Fig. 35(a). The decay in the two point correlation function is identical for the reconstructed image up until $3\mu\text{m}$, showing excellent reproduction of the short-range correlation. Although the longer range correlations match qualitatively, there is a drift seen as the distance between pixels increases. Both the excellent match in short range correlation and the small drift in the long range correlation can be explained based on the reconstruction algorithm, which models a stronger interaction of a center pixel to pixels in its immediate local neighborhood than pixels farther away. In effect, the algorithm gives a stronger weighting towards matching the short range correlations in the microstructure. Short range correlations carry the greatest weightage in determining mechanical properties such as elastic modulus, although long range correlations have been found to be important for phenomena such as surface roughening during plastic deformation. To test if the elastic properties are well captured in the reconstructed 3D microstructure, we compared against the experimental data of the elastic modulus as a function of temperature. The computed properties of the reconstructed 3D microstructure closely follow the experimentally measured Young's modulus as shown in Fig. 35(b) with an average error from experimental data of about 5 %.

10.1.3 EXAMPLE 3: Spatio-temporal sampling (2D + time) of grain growth

The temporal Markov Random Field (MRF) algorithm from [38] is used to synthesize the evolution of microstructure over a larger region given a small input movie. The movie is obtained from a phase field simulation of grain growth. The image size of the original gray-scale movie is 71×71 . The synthesized movies are double the size (142×142 pixels) but over the same time steps as the original phase field simulation. The snapshots from the original phase field simulation and the synthesized movies for initial, an intermediate and the final time are compared in Fig. 36. These synthesized microstructures correspond to window sizes of 5, 7 and 9 used in the MRF model. The window size is the adjustable parameter in the method for different microstructures. At window sizes much smaller than the correlation lengths, false matches lead to high noise in the reconstructions. The window size of 5 does not produce a good quality reconstruction as seen in the clusters of small grains that persist at longer times. At very high window sizes, not enough matching windows can be identified and in addition, more computations are needed that slow down the simulation. As seen in

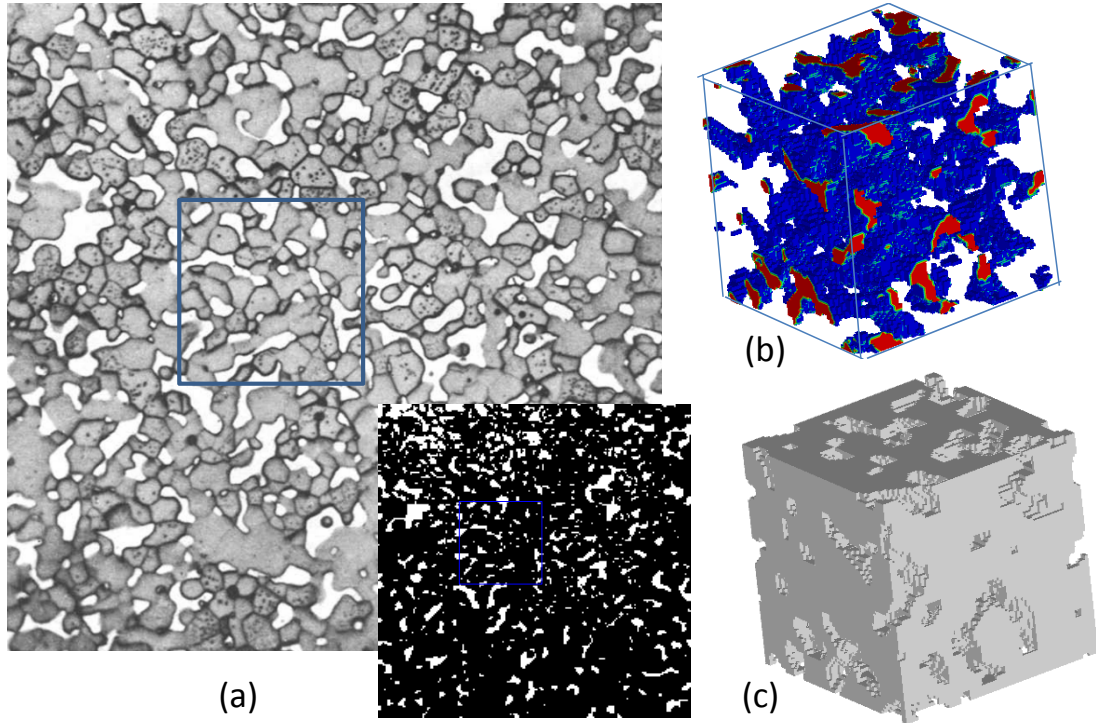


Figure 34: (a) Experimental Tungsten-silver composite image ($204 \times 236 \mu m$) from Umekawa et al (JMPS, 1965). The black and white image corresponds to a thresholded image with white representing the silver phase and black representing Tungsten. A $64 \mu m$ square cell shown in inset was used to reconstruct the 3D image. (b) A $64 \mu m$ length cell of reconstructed 3D microstructure of the experimental image showing silver distribution (c) The tungsten phase of the reconstructed microstructure.

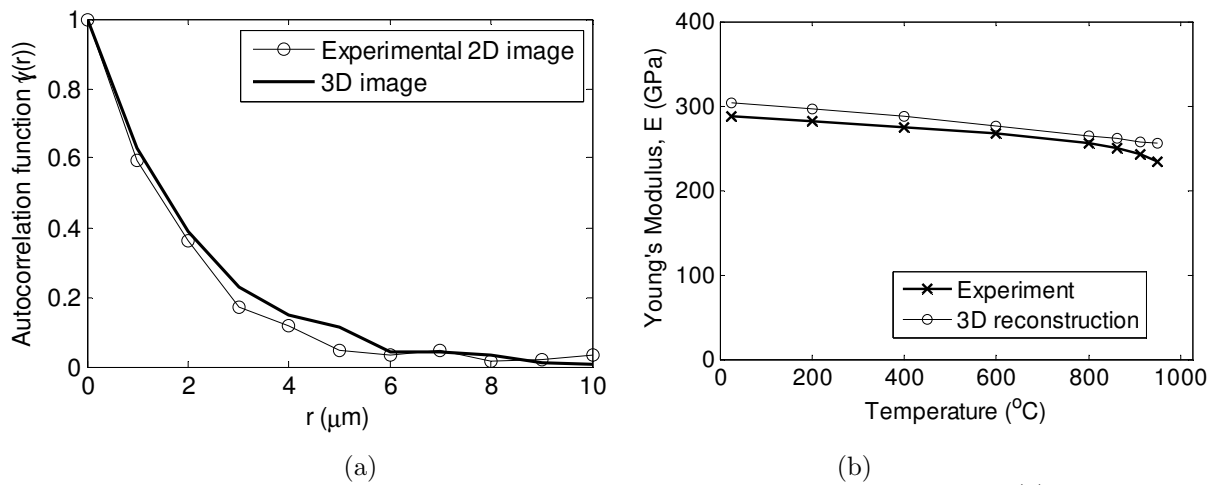


Figure 35: Comparison of properties of 3D reconstruction of Silver-Tungsten composite (a) The autocorrelation function for the silver phase (b) Experimental Young's modulus is shown along with the FEM results for the reconstructed 3D microstructure.

Fig. 36, the window sizes of 7 and 9 visually look similar to the phase field simulation. To quantitatively compare the grain size and shapes of the input and synthesized images, two global feature vectors were extracted from the input microstructure and compared to the synthesized microstructure (window size 9). The feature vectors are described below:

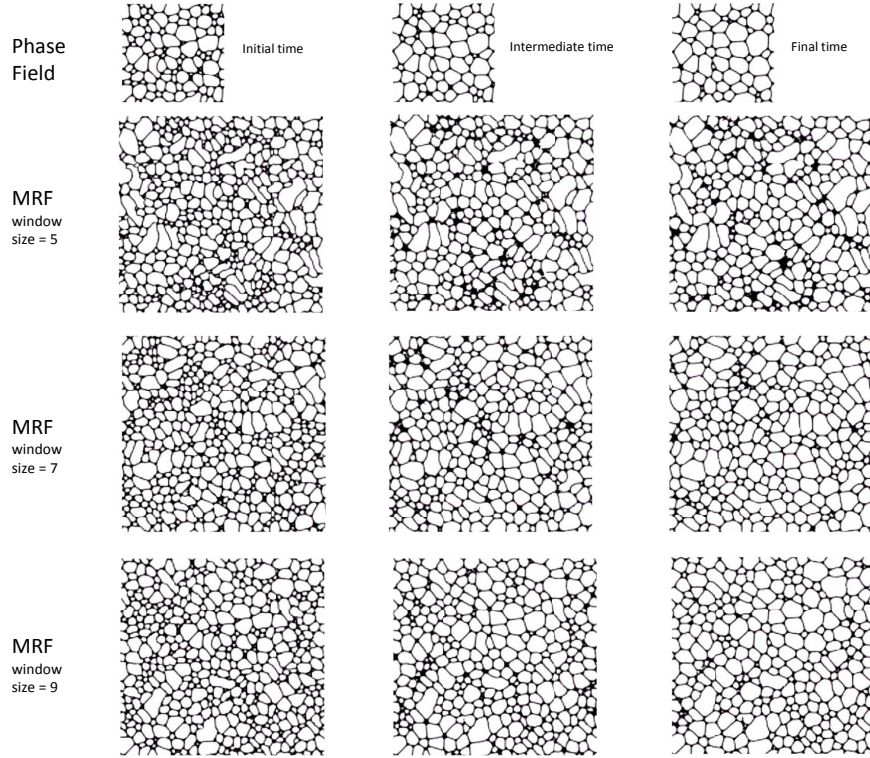


Figure 36: *Snapshots of the gray-scale movie are compared to the synthesized movie snapshots at initial, intermediate and final times. The synthesized microstructures are obtained using three different window sizes.*

1. Grain size and grain boundary perimeter statistics: The grain size and grain boundary (GB) perimeter of each grain is tabulated. A histogram containing the area or the perimeter of grains in the x-axis and the fraction of grains with the corresponding area (or GB perimeter) is plotted in the y-axis.
2. Grain shape statistics using the Rose of intersections: To obtain the rose of intersections, a network of parallel equidistant lines is placed over the microstructure image at several angles and the number of grain boundary intersections with each test line is measured. The histogram of intersections with the angle of orientation of the lines is called the rose of intersection.

The plot in Fig. 37(a,b) shows the fraction of grains as a function of grain area and grain perimeter (measured in pixels) for initial and final times. The rose of intersections graphs are illustrated using lines with 11 different angles at initial and final times. The shape histogram depicts the decrease in overall grain size with time (shrinking of the contour) while the overall grain shape as indicated by the contour shape remains mostly equiaxial. Fig. 37 shows that the synthesized microstructures are able to capture the grain size and shape statistics of the original microstructure evolution predicted by the phase field method. MRF sampling approach proposed here is significantly faster than performing a phase field simulation for larger spatial regions. We do not solve any differential equations and as such, are not constrained by the small time increments needed to maintain numerical stability.

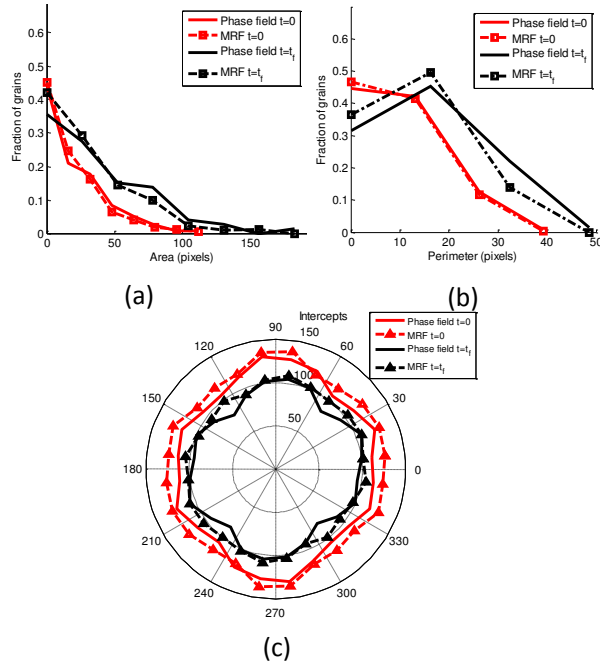


Figure 37: Comparison of statistical features of phase field and MRF algorithms (a,b) grain area and grain perimeter statistics at initial and final time (c) Rose of interactions at initial and final time

10.1.4 EXAMPLE 4 Microstructure embedding in CAD models

The 3D reconstruction methodology shown in this chapter can be extended to any geometry. In principle, one could use sampling and optimization methodologies to embed microstructures over an engineering (computer aided design (CAD)) model. To demonstrate this, we have embedded a microstructure (of stainless steel) into a CAD geometry in Fig. 38. The microstructure was obtained using serial sectioning and diffraction techniques. The microstructure is in the form of voxels colored by grain numbers. The 3D microstructure is then sampled onto the CAD geometry using an extension of the sampling method described in the previous sections. Such methodologies will be useful for developing 3D computational models of microstructures at a component level using limited experimentally sampled volumes.

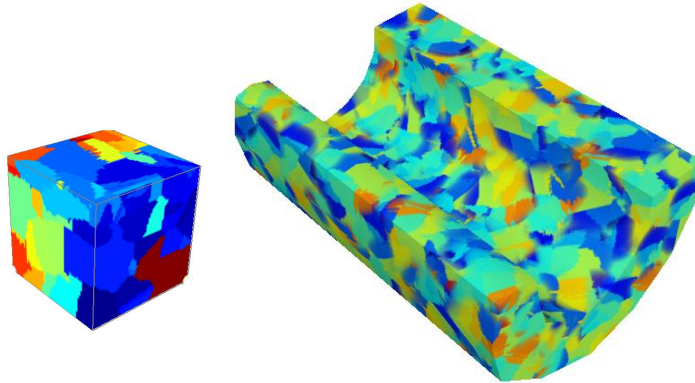


Figure 38: (left) AL6XN microstructure from serial sectioning, AL6XN RVE (Alexis Lewis, NRL) (right) Synthesized geometry

10.2 Conclusions

It is human intuition that different windows taken from a polycrystalline microstructure generally ‘look alike’. This can be quantified through an underlying stationary probability distribution that generates all possible microstructural windows. While quantifying this high dimensional joint probability distribution of all pixel colors is computationally intractable, we looked at sampling and optimization methods to model this distribution. For many microstructures, the probability of a pixel color depends only on the pixel state of its close neighbors. Thus, one could represent microstructures in the form of undirected probabilistic graphs called Markov Random Fields (MRFs) with pixels interacting with neighbors over a sampling window. Previous methods for reconstructing polycrystals using algorithms based on a common set of underlying features such as marginal histograms and point probability functions have often failed to capture the local information such as sharp grain boundaries. We find that the MRF approach is an attractive solution in this regard. We show that not only are the global features such as grain size/shape distribution captured but also the mechanical (elastic) properties of the synthesized microstructures as computed using finite element method were also found to closely reproduce the experimental values. We also presented an extension of this MRF algorithm for synthesizing microstructure evolution. A promising method for reconstructing diverse microstructures from two-dimensional microstructures imaged on orthogonal planes was also presented. The method is particularly promising for anisotropic cases where the x -, y - and z - slices look different. With these developments, we believe that Markov random fields presents an exciting avenue for practical reconstruction of spatio-temporal evolution of microstructures.

11 Software packages created in this program

MURI co-PIs

11.1 Mined-MatKit

A collection of materials informatics tools wrapped in easy to use GUIs (Fig. 39) for license-free deployment. Currently consists of the following:

- Microstructure Generation module. Creation of a vast variety of randomized microstructures under physical and statistical constraints;
- Fitting module. Used for establishing functions/linkages between material descriptors and process or property parameters;
- Segmentation module. Deals with the transition from raw imaging data to microstructure representations;
- Dimensionality Reduction module. Deals with the identification of dominant descriptors of microstructure from a large set of statistics;
- Spatial Statistics module. Deals with the calculation of spatial statistics under variety of scenarios and definitions.

Mined-MatKit Download Link: <https://github.com/MINED-MATKIT>

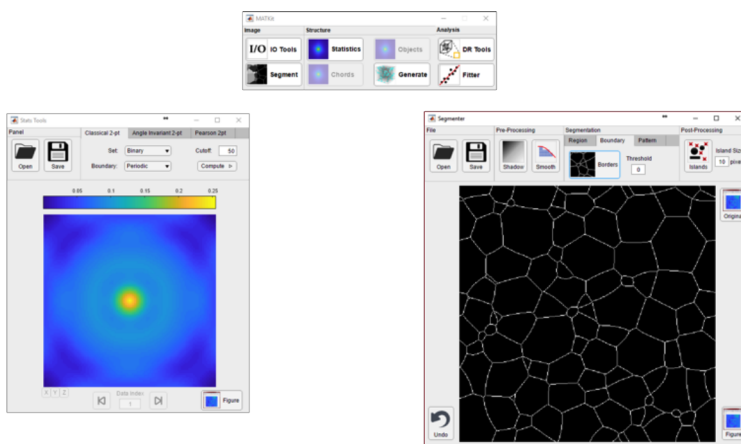


Figure 39: Selected screen shots of the MatKit interface.

11.2 Project Pages

Project-Pages is a Jekyll based scientific blogging tool, made easily accessible to the non-coder researcher through simple heavily opinionated templates, while preserving the freedom of the code savvy user to customize all functionality as they please. Project-Pages (Fig. 40) uses a specialized re-skin of Prose.io to provide a CMS-like overlay.

Organization Level Functionality:

- Adaptive Tags
- Text and Tag Based Search
- Ability to Manage Multiple Projects
- Members Page

- Mention Tracking via Altmetric
- Google Analytics
- Comments with Disqus
- Responsive Design

Post Level Functionality:

- Code Highlighting
- MathJax for Equations
- Presentation Layout using Reveal.JS with Easy to Use Jekyll Helpers
- MATLAB Layout with Direct to Post Export Function in MATLAB
- Jupyter Notebook Layout Directly from Notebook Files
- Mermaid.JS for Automated Workflow Visualization
- Projector Layout using DataProjector.JS to Visualize 3D Point Cloud Data from JSON Files
- ThreeJS Layout to Visualize 3D Surface Data

Project Pages Download Link: <https://github.com/projectpages>

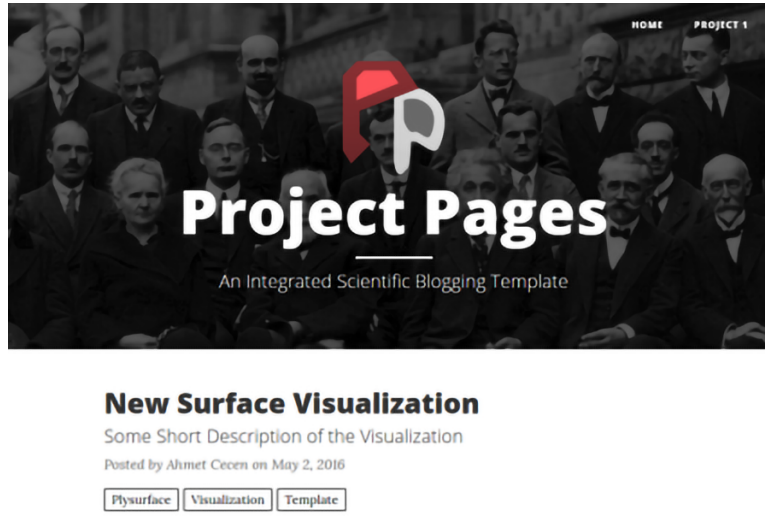


Figure 40: Screen shot of the Project Pages interface.

11.3 MicroFract

The open source MicroFract 2D and 3D codes formulate an image-based methodology for identifying the path of least resistance in the microstructure [40]. The user specifies the line direction of the macroscopic crack and a cohesive energy map corresponding to the weak and strong interfaces in the microstructure. The code outputs the crack trajectory as shown in the Figure below.

Microstructural crack trajectory is overall driven by the macroscopic crack direction, while microstructure resolves the crack along surfaces of low resistances: in effect, the crack follows a zig-zag trajectory following weak microstructural features. The excess energy to be supplied at the microstructural scale to form a crack of area A is taken as

$$E[A] = \int_A (\Gamma - \Phi) dA \quad (41)$$

where Γ is the cohesive energy along the crack path and Φ is the supplied macroscopic energy that acts against cohesion. We define the crack path to be the area A that minimizes this energy. In effect, we are searching for a path of least resistance that has both high stresses and low interfacial strength, and is thus amenable for fracture.

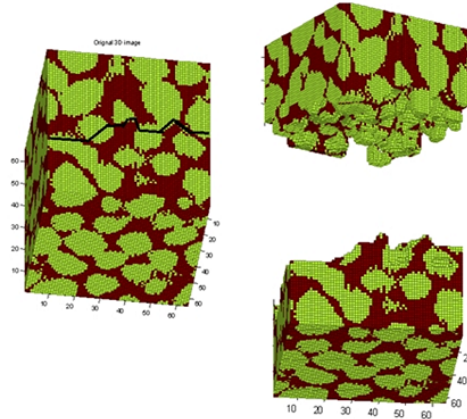


Figure 41: A 3D example of MicroFract code. The user inputs a microstructure cohesive energy map and a approximate crack line. The code outputs the 3D crack trajectory. The green particle is more brittle than the red matrix and the predicted crack maximizes its area along the brittle particles.

The problem of finding the crack area A that minimizes the above equation is an exercise in functional minimization. Computationally, the solution typically involves solution of a partial differential equation that tracks fracture surface evolution using gradient descent schemes, phase field parameters or level sets. The drawback of these approaches are that the solution may lead to a local minima and there is no guarantee of global minimum energy path. The primary purpose of this software is to introduce the use of graph cuts method to solve this problem. The above equation can be exactly solved (to get global minimum) by the max-flow min-cut theorem. The results are shown via 2D and 3D examples from literature and the code and examples are made available to users.

MicroFract Download Link: <http://umich.edu/~veeras/projects/MicroFract.html>

11.4 Pruned Search

Pruned Search is an enhanced searching algorithm for general optimization. It combines the concept of importance ranking and dimension reduction and generalizes them into meta-heuristics, to help better search for optimal objectives. The primary idea is to have the search force focus on a more promising path and reduce the irrelevant effort. The validity of this heuristic is rooted in two assumptions.

Firstly, we assume the desired function value depends only on a reduced, albeit unknown, set of variables. Secondly, we assume that the impact of each variable on the search value is different. Hence, there exists an optimal order regarding searching priority. The next task is obtaining an optimal search path, and at the same time, reducing the viable search region of each variable so that it is faster. For that, a representative set of data needs to be collected. We call that process - data distillation (Figure 42). Next, we perform complexity reduction on two branches, in parallel: one creates an ordered list of variables based on their impact towards the function, and the other reduces of the feasible region for each variable. The former is achieved through feature selection methods where a ranking is guaranteed. The latter is realized through examining a rule-based classifier and looking for the critical thresholds. After the above steps, meta-heuristics about searching are obtained. Optimization becomes a more promising endeavor after the search space is pruned. We then employ a simple line search algorithm that takes a prefixed searching order and replaces the original constraints with those obtained from the pruning process.

The Pruned Search tool requires a set of control variables x as inputs, an objective function in the form of $y=f(x)$, where y is the target of optimization and a set of constraints as bounds and functions of x . The example demo provides a case study for optimizing few materials properties. To use this software, following packages are required: MATLAB 7.6/2008a (or higher) with Pattern Recognition Toolbox, Python 2.7,

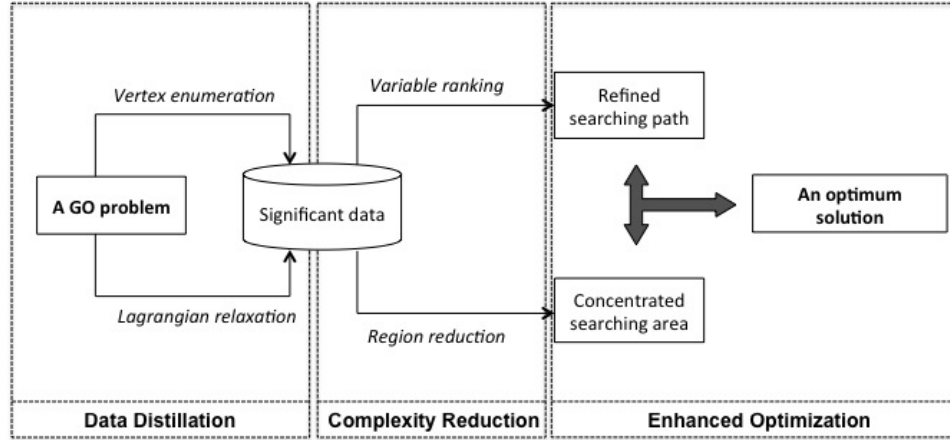


Figure 42: The method consists of three key processes. Data Distillation collects a significant and representative data set from an objective function. Complexity Reduction ranks the importance and prunes the search space for each variable. Finally, the Enhanced Optimization searches within the reduced space and seeks for the optimal solution.

Sklearn 0.14 (or higher), Scipy 0.7 (or higher), and Numpy 1.4 (or higher).

Pruned Search Download Link: <http://cucis.ece.northwestern.edu/projects/MURI/workopt.html>

11.5 MES Predictor

The open source MES Predictor is developed to establish the process-structure-property (PSP) linkages. More specifically, it's used to establishing high value structure-property homogenization linkages for high-contrast elastic 3-D composite microstructures. The model is based on 3-D convolutional neural network (CNN), and the source code is written in Python 2.7. In order to use this software, some packages are required, which are Python 2.7, Numpy, Sklearn, Keras, Tensorflow, Pickle and HDF5.

To use this software, what the algorithm requires as input are a numpy array. The shape of this numpy array is $(x, 51, 51, 51, 1)$ where x is the number of microscale volume elements (MVEs) and the dimension of microstructure should be three-dimensional (i.e. $51 \times 51 \times 51$). The CNN will establish the PSP linkages in the materials system and predict its macroscale (effective) stiffness.

The MES Predictor is currently command-line program, and it can only be used to make predictions. To make more contributions to the community, we are developing another software and an online application. The new software will also provide the source code that users can use to train their own customized CNN. The online application is based on MES Predictor, and users can just upload their data on the website to get predictions instead of downloading and installing all the packages and using command-line to make predictions.

MES Predictor Download Link: <http://cucis.ece.northwestern.edu/projects/MURI/workpred.html>

11.6 Plug&Play

We distribute Plug & Play as an open source Matlab software package. The software package supports a basic implementation of P&P for interpolation and in-painting applications, and is designed to be easily ported to applications such as tomography.

Plug&Play Download Link: <https://engineering.purdue.edu/~bouman/Plug-and-Play/>

11.7 TIMBIR

The time-interlace model-based iterative reconstruction (TIMBIR) algorithm is available as open-source C for MPI implementation on large scale computers. The software is designed to compute 4D reconstructions

of objects from view samples at arbitrary times/angle intervals. It is actively used by researchers at the Advanced Photon Source (APS).

Plug&Play Download Link: <https://engineering.purdue.edu/~bouman/software/tomography/>

11.8 Structural Phase Transformation Web Tools

The Structural Phase Transformation Web Tools (structrans.org) is a user-friendly web-based research and educational platform for structural phase transformation. The most straightforward usage of STRUCTRANS is to calculate the transformation stretch tensor and correspondence between two crystal structures, as described in this report. The basic algorithm is described in [35]. The underlying calculation is the following: given the lattice parameters and space groups of the two phases, related by a group-subgroup relation, find the minimum strain (measured in a reasonable norm [35]) that maps a sublattice of the austenite phase to the primitive lattice of the martensite phase. From this minimization the transformation stretch tensor is found.

Additional capabilities include the ability to generate the associated twin systems, compatible interfaces, solutions of the crystallographic theory of martensite, and to check conditions of supercompatibility ($\lambda_2 = 1$ and the cofactor conditions). Plans are in place to display the solutions by the visualization method described in [17].

Structural Phase Transformation Web Tools Download Link: www.structrans.org

11.9 EMsoft: a package for electron scattering simulations

The open source EMsoft package was developed to provide simulation algorithms for electron scattering, in particular diffraction experiments in the scanning electron microscope (SEM). The source code is mostly written in Fortran-2003, with bits in OpenCL (to drive GPU cards) and some interfacing code in C/C++. With the help of Mike Jackson and his coworkers at BlueQuartz Software (Dayton, OH), EMsoft has been set up with both a public (at GitHub) and private source code repository as well as a nightly build system to make sure that the code compiles on Mac OS X, Windows 10, and Linux platforms. The GitHub repository (URL at bottom of page) provides access to all the source code under a BSD 2 license, as well as all resource and template files; a number of program manuals are made available in PDF format from the repository, and a wiki page has detailed information about program input and several examples for EBSD, ECP, and TKD pattern simulations.

The majority of the EMsoft programs are currently command-line programs, and require a bit of a learning curve to properly use them. To make the interface more user-friendly, we have started the task of creating a graphical user interface (GUI) called EMsoftWorkbench. This GUI is under development by BlueQuartz, and can be accessed from their nightly build beta site (URL below). Currently, only the EBSD-related programs are available in the GUI; others will be added in the future, depending on the availability of funding.

The EMsoft package makes available all the dictionary indexing programs for EBSD, ECP, TKD, and PED modalities.

Download Link: <https://github.com/EMsoft-org>

EMsoftWorkbench Link: <http://www.bluequartz.net/binaries/EMsoft/experimental>

References

- [46] Dipendra Jha, Saransh Singh, Reda Al-Bahrani, Wei-keng Liao, Alok N. Choudhary, Marc De Graef, and Ankit Agrawal. “Extracting Grain Orientations from EBSD Patterns of Polycrystalline Materials using Convolutional Neural Networks”. *Microscopy and Microanalysis* 24 (2018), pp. 497–502.
- [45] K.A. Mohan, P. Kc, C. Phatak, M. De Graef, and C.A. Bouman. “Model-Based Iterative Reconstruction of Magnetization using Vector Field Electron Tomography”. *IEEE Trans. Comp. Imag.* 4.3 (2018), pp. 432–446.
- [44] A Paul, P Acar, R Liu, W Liao, A Choudhary, V Sundararaghavan, and A Agrawal. “Data Sampling Schemes for Microstructure Design with Vibrational Tuning Constraints”. *AIAA Journal (in press)* (2018).
- [43] P. Kc, K.A. Mohan, C. Phatak, C.A. Bouman, and M. De Graef. “3D reconstruction of the magnetic vector potential using model based iterative reconstruction”. *Ultramicroscopy* 182 (2017), pp. 131–144.
- [42] Ruoqian Liu, Yuksel C Yabansu, Zijiang Yang, Alok N Choudhary, Surya R Kalidindi, and Ankit Agrawal. “Context Aware Machine Learning Approaches for Modeling Elastic Localization in Three-Dimensional Composite Microstructures”. *Integrating Materials and Manufacturing Innovation* (2017), pp. 1–12.
- [41] Yue Sun, Ahmet Cecen, John W Gibbs, Surya R Kalidindi, and Peter W Voorhees. “Analytics on large microstructure datasets using two-point spatial correlations: Coarsening of dendritic structures”. *Acta Materialia* 132 (2017), pp. 374–388.
- [40] Veera Sundararaghavan and Siddhartha Srivastava. “MicroFract: An image based code for microstructural crack path prediction”. *SoftwareX* 6 (2017), pp. 94–97. ISSN: 2352-7110. DOI: <http://dx.doi.org/10.1016/j.softx.2017.04.002>. URL: <http://www.sciencedirect.com/science/article/pii/S2352711017300092>.
- [39] Martín Abadi, Ashish Agarwal, Paul Barham, Eugene Brevdo, Zhifeng Chen, Craig Citro, Greg S Corrado, Andy Davis, Jeffrey Dean, Matthieu Devin, et al. “Tensorflow: Large-scale machine learning on heterogeneous distributed systems”. *arXiv preprint arXiv:1603.04467* (2016).
- [38] Pinar Acar and Veera Sundararaghavan. “A Markov random field approach for modeling spatio-temporal evolution of microstructures”. *Modelling and Simulation in Materials Science and Engineering* 24.7 (2016), p. 075005. DOI: <http://dx.doi.org/10.1088/0965-0393/24/7/075005>. URL: <http://stacks.iop.org/0965-0393/24/i=7/a=075005>.
- [37] Pinar Acar and Veera Sundararaghavan. “Linear Solution Scheme for Microstructure Design with Process Constraints”. *AIAA Journal* 54.12 (2016), pp. 4022–4031. DOI: <http://doi.org/10.2514/1.J055247>.
- [36] Pinar Acar and Veera Sundararaghavan. “Utilization of a Linear Solver for Multiscale Design and Optimization of Microstructures”. *AIAA Journal* 54.5 (2016), pp. 1751–1759. DOI: <http://doi.org/10.2514/1.J054822>.
- [35] Xian Chen, Yintao Song, Nobumichi Tamura, and Richard D James. “Determination of the stretch tensor for structural transformations”. *Journal of the Mechanics and Physics of Solids* 93 (2016), pp. 34–43.
- [34] Xian Chen, Nobumichi Tamura, Alastair MacDowell, and Richard D James. “In-situ characterization of highly reversible phase transformation by synchrotron X-ray Laue microdiffraction”. *Applied Physics Letters* 108.21 (2016), p. 211902.
- [33] P L Kuhns, M J R Hoch, S Yuan, A P Reyes, V Srivastava, R D James, and C Leighton. “Magnetic-field-induced changes in superparamagnetic cluster dynamics in the martensitic phase of $\text{Ni}_{43}\text{Co}_7\text{Mn}_{40}\text{Sn}_{10}$ ”. *Applied Physics Letters* 108.25 (2016), p. 252403.
- [32] A Kumar, L Nguyen, M DeGraef, and V Sundararaghavan. “A Markov random field approach for microstructure synthesis”. *Modelling and Simulation in Materials Science and Engineering* 24.3 (2016), p. 035015. DOI: <http://doi.org/10.1088/0965-0393/24/3/035015>. URL: <http://stacks.iop.org/0965-0393/24/i=3/a=035015>.

- [31] Ruoqian Liu, Ankit Agrawal, Wei-keng Liao, Marc De Graef, and Alok Choudhary. “Materials Discovery: Understanding Polycrystals from Large-Scale Electron Patterns”. *Proceedings of IEEE BigData Workshop on Advances in Software and Hardware for Big Data to Knowledge Discovery (ASH)*. 2016, pp. 2261–2269.
- [30] Xiaoyue Ni, Julia R Greer, Kaushik Bhattacharya, Richard D James, and Xian Chen. “Exceptional resilience of small-scale $\text{Au}_{30}\text{Cu}_{25}\text{Zn}_{45}$ under cyclic stress-induced phase transformation”. *Nano letters* 16.12 (2016), pp. 7621–7625.
- [29] Ashwin J Shahani, Xianghui Xiao, and Peter W Voorhees. “The mechanism of eutectic growth in highly anisotropic materials”. *Nature communications* 7 (2016).
- [28] Yu H Chen, Se Un Park, Dennis Wei, Greg Newstadt, Michael A Jackson, Jeff P Simmons, Marc De Graef, and Alfred O Hero. “A dictionary approach to electron backscatter diffraction indexing”. *Microscopy and Microanalysis* 21.3 (2015), pp. 739–752.
- [27] Christoph Chluba, Wenwei Ge, Rodrigo Lima de Miranda, Julian Strobel, Lorenz Kienle, Eckhard Quandt, and Manfred Wuttig. “Ultralow-fatigue shape memory alloy films”. *Science* 348.6238 (2015), pp. 1004–1007.
- [26] JW Gibbs, K Aditya Mohan, EB Gulsoy, AJ Shahani, X Xiao, CA Bouman, M De Graef, and PW Voorhees. “The three-dimensional morphology of growing dendrites”. *Scientific reports* 5.11824 (2015).
- [25] Richard D James. “Taming the temperamental metal transformation”. *Science* 348.6238 (2015), pp. 968–969.
- [24] Dae-Woo Kim and Mary L. Comer. “Channel detection in microscope images of materials using marked point process modeling”. *IEEE International Conference on Image Processing*. 2015, pp. 3054–3058.
- [23] Ruoqian Liu, Yuksel C. Yabansu, Ankit Agrawal, Surya R. Kalidindi, and Alok N. Choudhary. “Machine learning approaches for elastic localization linkages in high-contrast composite materials”. *Integrating Materials and Manufacturing Innovation* 4.13 (2015), pp. 1–17.
- [22] K. A. Mohan, S. V. Venkatakrishnan, J. W. Gibbs, E. B. Gulsoy, X. Xiao, M. De Graef, P. W. Voorhees, and C. A. Bouman. “4D model-based iterative reconstruction from interlaced views”. *2015 IEEE International Conference on Acoustics, Speech and Signal Processing (ICASSP)*. 2015, pp. 783–787.
- [21] K Aditya Mohan, SV Venkatakrishnan, John W Gibbs, Emine Begum Gulsoy, Xianghui Xiao, Marc De Graef, Peter W Voorhees, and Charles A Bouman. “TIMBIR: A method for time-space reconstruction from interlaced views”. *IEEE Transactions on Computational Imaging* 1.2 (2015), pp. 96–111.
- [20] E. Humphrey, C. Phatak, A.K. Petford-Long, and M. De Graef. “Separation of electrostatic and magnetic phase shifts using a modified transport-of-intensity equation”. *Ultramicroscopy* 139 (2014), pp. 5–12. ISSN: 0304-3991. DOI: <http://dx.doi.org/10.1016/j.ultramic.2014.01.005>.
- [19] Veera Sundararaghavan. “Reconstruction of three-dimensional anisotropic microstructures from two-dimensional micrographs imaged on orthogonal planes”. *Integrating Materials and Manufacturing Innovation* 3.1 (2014), p. 19. ISSN: 2193-9772. DOI: 10.1186/s40192-014-0019-3. URL: <https://doi.org/10.1186/s40192-014-0019-3>.
- [18] Patrick G. Callahan and Marc De Graef. “Dynamical Electron Backscatter Diffraction Patterns. Part I: Pattern Simulations”. *Microscopy and Microanalysis* 19.5 (2013), pp. 1255–1265.
- [17] Xian Chen, Vijay Srivastava, Vivekanand Dabade, and Richard D James. “Study of the cofactor conditions: conditions of supercompatibility between phases”. *J. Mech. Phys. Solids* 61.12 (2013), p. 2566.
- [16] Yintao Song, Xian Chen, Vivekanand Dabade, T W Shield, and Richard D James. “Enhanced reversibility and unusual microstructure of a phase-transforming material”. *Nature* 502 (2013), p. 85.
- [15] Singanallur V Venkatakrishnan, Lawrence F Drummy, Michael A Jackson, Marc De Graef, Jeff Simmons, and Charles A Bouman. “A Model Based Iterative Reconstruction Algorithm For High Angle Annular Dark Field - Scanning Transmission Electron Microscope (HAADF-STEM) Tomography”. *IEEE Trans. on Img. Proc.* 22 (2013), pp. 4532–4543.

- [14] C. Phatak, A. Petford-Long, O. Heinonen, M. Tanase, and M. De Graef. “Nanoscale structure of the magnetic induction at monopole defects in artificial spin-ice lattices”. *Physical Review B* 83.17 (2011), p. 174431. ISSN: 1098-0121. DOI: 10.1103/PhysRevB.83.174431.
- [13] Robert Zarnetta, Ryota Takahashi, Marcus L Young, Alan Savan, Yasubumi Furuya, Sigurd Thienhaus, Burkhard Maaß, Mustafa Rahim, Jan Frenzel, Hayo Brunken, et al. “Identification of quaternary shape memory alloys with near-zero thermal hysteresis and unprecedented functional stability”. *Advanced Functional Materials* 20.12 (2010), pp. 1917–1923.
- [12] Zhiyong Zhang, Richard D James, and Stefan Müller. “Energy barriers and hysteresis in martensitic phase transformations”. *Acta Materialia* 57.15 (2009), pp. 4332–4352.
- [11] Claire Maurice and Roland Fortunier. “A 3D Hough transform for indexing EBSD and Kossel patterns”. *Journal of microscopy* 230.3 (2008), pp. 520–529.
- [10] C. Phatak, M. Beleggia, and M. De Graef. “Vector field electron tomography of magnetic materials: Theoretical development”. *Ultramicroscopy* 108.6 (2008), pp. 503–513. ISSN: 0304-3991. DOI: <http://dx.doi.org/10.1016/j.ultramic.2007.08.002>.
- [9] C P Frick, S Orso, and E Arzt. “Loss of pseudoelasticity in nickel–titanium sub-micron compression pillars”. *Acta Materialia* 55.11 (2007), pp. 3845–3855.
- [8] Jean-Baptiste Thibault, Ken Sauer, Charles Bouman, and Jiang Hsieh. “A Three-Dimensional Statistical Approach to Improved Image Quality for Multi-Slice Helical CT”. *Medical Physics* 34.11 (2007), pp. 4526–4544.
- [7] J Cui, Y S Chu, O Famodu, Y Furuya, J Hattrick-Simpers, R D James, A Ludwig, S Thienhaus, M Wuttig, Z Zhang, and I Takeuchi. “Combinatorial search of thermoelastic shape memory alloys with extremely small hysteresis width”. *Nature Materials* (5 2006), pp. 286–290. DOI: 10.1038/nmat1593.
- [6] R D James and Z Zhang. “A way to search for multiferroic materials with unlikely combinations of physical properties”. *Magnetism and Structure in Functional Materials, Springer Series in Materials Science*. Ed. by A. Planes, L Manósa, and A. Saxena. Vol. 9. Berlin: Springer-Verlag, 2005, pp. 159–175.
- [5] Brent L. Adams, Stuart I. Wright, and Karsten Kunze. “Orientation imaging: The emergence of a new microscopy”. *Metallurgical Transactions A* 24.4 (1993), pp. 819–831. ISSN: 1543-1940. DOI: 10.1007/BF02656503. URL: <http://dx.doi.org/10.1007/BF02656503>.
- [4] C. A. Bouman and K. Sauer. “A Generalized Gaussian Image Model for Edge-Preserving MAP Estimation”. *IEEE Trans. on Image Processing* 2.3 (1993), pp. 296–310.
- [3] N.C. Krieger Lassen. “Automatic crystal orientation determination from EBSPs”. *Micron and Microscopica Acta* 6 (1992), pp. 191–192.
- [2] Léon Bottou. “Stochastic gradient learning in neural networks”. *Proceedings of Neuro-Nimes* 91.8 (1991).
- [1] Edgar C Bain. “The Nature of Martensite”. *Trans. AIME* 70.1 (1924), p. 25.

Publications acknowledging MURI support

- [146] Ankit Agrawal and Alok Choudhary. “An online tool for predicting fatigue strength of steel alloys based on ensemble data mining”. *International Journal of Fatigue* 113 (2018), pp. 389–400.
- [145] Camilo Aguilar and Mary Comer. “A Marked Point Process Model Incorporating Active Contours Boundary Energy”. *Proceedings of Computational Imaging XVI*. 2018.
- [144] Gregory T. Buzzard, Stanley H. Chan, Suhas Sreehari, and Charles A. Bouman. “Plug-and-Play Unplugged: Optimization Free Reconstruction using Consensus Equilibrium”. *SIAM Journal on Mathematical Imaging* 11.3 (2018), pp. 2001–2020.
- [143] Ahmet Cecen, Hanjun Dai, Yuksel C Yabansu, Surya R Kalidindi, and Le Song. “Material structure–property linkage using three-dimensional convolutional neural networks”. *Acta Materialia* 146 (2018), pp. 76–84.
- [142] M. L. Comer and P. Voorhees. “Narrow gap detection for dendritic structures in materials images”. *Proceedings of Computational Imaging XVII*. 2018.
- [141] G. Dilshan P. Godaliyadda, Dong Hye Ye, Michael D. Uchic, Michael A. Groeber, Gregory T. Buzzard, and Charles A. Bouman. “A Framework for Dynamic Image Sampling Based on Supervised Learning”. *IEEE Trans. on Computational Imaging* 4.1 (2018), pp. 1–16.
- [140] Chun-Jen Hsueh and Kaushik Bhattacharya. “Optimizing microstructure for toughness: The model problem of peeling”. *Struct Multidisc Optim* 58 (2018), pp. 1067–1080.
- [139] Richard D James. “Materials from mathematics”. to appear in Bulletin of the American Mathematical Society. 2018.
- [138] Richard D James. “Symmetry, invariance and the structure of matter”. to appear in Proc. International Congress of Mathematicians. 2018.
- [137] Dipendra Jha, Saransh Singh, Reda Al-Bahrani, Wei-keng Liao, Alok N. Choudhary, Marc De Graef, and Ankit Agrawal. “Extracting Grain Orientations from EBSD Patterns of Polycrystalline Materials using Convolutional Neural Networks”. *Microscopy and Microanalysis* 24 (2018), pp. 497–502.
- [136] Tianyu Li, Mary Comer, and Josiane Zerubia. “A Connected-Tube MPP Model for Object Detection with Application to Materials and Remotely-Sensed Images”. *IEEE International Conference on Image Processing*. 2018.
- [135] K.A. Mohan, P. Kc, C. Phatak, M. De Graef, and C.A. Bouman. “Model-Based Iterative Reconstruction of Magnetization using Vector Field Electron Tomography”. *IEEE Trans. Comp. Imag.* 4.3 (2018), pp. 432–446.
- [134] A Paul, P Acar, R Liu, W Liao, A Choudhary, V Sundararaghavan, and A Agrawal. “Data Sampling Schemes for Microstructure Design with Vibrational Tuning Constraints”. *AIAA Journal (in press)* (2018).
- [133] Arindam Paul, Pinar Acar, Ruoqian Liu, Wei-keng Liao, Alok Choudhary, Veera Sundararaghavan, and Ankit Agrawal. “Data Sampling Schemes for Microstructure Design with Vibrational Tuning Constraints”. *American Institute of Aeronautics and Astronautics (AIAA) Journal* 56.3 (2018), pp. 1239–1250.
- [132] F. Ram and M. De Graef. “Phase Differentiation by Electron Backscatter Diffraction using the Dictionary Indexing Approach”. *Acta Materialia* 144 (2018), pp. 352–364.
- [131] Jin Yang and Kaushik Bhattacharya. “Augmented Lagrangian Digital Image Correlation”. *Experimental Mechanics*. To appear (2018).
- [130] Jin Yang and Kaushik Bhattacharya. “Combining image compression with digital image correlation”. *Experimental Mechanics* To appear (2018).
- [129] Jin Yang and Kaushik Bhattacharya. “Fast Adaptive Global Digital Image Correlation”. *Proceedings of the Society of Experimental Mechanics*. 2018.
- [128] Jin Yang and Kaushik Bhattacharya. “Multigrid adaptive mesh digital image correlation”. *Int. J. Num. Meth. Engng.* Submitted (2018).

- [127] Zijiang Yang, Yuksel C Yabansu, Reda Al-Bahrani, Alok N Choudhary, Surya R Kalidindi, and Ankit Agrawal. "Deep learning approaches for mining structure-property linkages in high contrast composites from simulation datasets". *Computational Materials Science* 151 (2018), pp. 278–287.
- [126] Zijiang Yang, Yuksel C. Yabansu, Reda Al-Bahrani, Wei-keng Liao, Alok N. Choudhary, Surya R. Kalidindi, and Ankit Agrawal. "Deep learning approaches for mining structure-property linkages in high contrast composites from simulation datasets". *Computational Materials Science* 151 (2018), pp. 278–287.
- [125] Shijie Zhang, Zhengtian Song, G. M. Dilshan P. Godaliyadda, Dong Hye Ye, Azhad U. Chowdhury, Atanu Sengupta, Gregory T. Buzzard, Charles A. Bouman, and Garth J. Simpson. "Dynamic Sparse Sampling for Confocal Raman Microscopy". *Journal Analytical Chemistry* 90 (2018), pp. 4461–4469.
- [124] Patrick Altschuh, Yuksel C Yabansu, Johannes Hötzer, Michael Selzer, Britta Nestler, and Surya R Kalidindi. "Data Science Approaches for Microstructure Quantification and Feature Identification in Porous Membranes". *Journal of Membrane Science* 540 (2017), pp. 88–97.
- [123] P.G. Callahan, M.P. Echlin, T.M. Pollock, and M. De Graef. "Reconstruction of Laser-Induced Surface Topography from Electron Backscatter Diffraction Patterns". *Microscopy and Microanalysis* 23 (2017), pp. 730–740.
- [122] P.G. Callahan, M.P. Echlin, T.M. Pollock, S. Singh, and M. De Graef. "3D Texture Visualization Approaches: Theoretical Analysis and Examples". *Journal of Applied Crystallography* 50 (2017), pp. 430–440.
- [121] P.G. Callahan, M.P. Echlin, T.M. Pollock, S. Singh, F. Ram, and M. De Graef. "3D Texture Visualization Approaches: Applications to Nickel and Titanium Alloys". *Journal of Applied Crystallography* 50 (2017), pp. 1267–1279.
- [120] Al'ona Furmanchuk, James Saal, Jeff Doak, Gregory Olson, Alok Choudhary, and Ankit Agrawal. "Prediction of Seebeck Coefficient for Compounds without Restriction to Fixed Stoichiometry: A Machine Learning Approach". *Journal of Computational Chemistry* (2017), pp. 1–11.
- [119] Kasthurirangan Gopalakrishnan, Siddhartha K. Khaitan, Alok Choudhary, and Ankit Agrawal. "Deep Convolutional Neural Networks with transfer learning for computer vision-based data-driven pavement distress detection". *Construction and Building Materials* 157 (2017), pp. 322–330.
- [118] Hanlin Gu, Lars Bumke, Christoph Chluba, Eckhard Quandt, and Richard D James. "Phase engineering and supercompatibility of shape memory alloys". *Materials Today* (2017).
- [117] Richard D James, Alessia Nota, and Juan JL Velázquez. "Self-similar profiles for homoeenergetic solutions of the Boltzmann equation: particle velocity distribution and entropy". *arXiv preprint arXiv:1710.03653* (2017).
- [116] P. Kc, K.A. Mohan, C. Phatak, C.A. Bouman, and M. De Graef. "3D reconstruction of the magnetic vector potential using model based iterative reconstruction". *Ultramicroscopy* 182 (2017), pp. 131–144.
- [115] Abhishek Kumar and Veera Sundararaghavan. "Simulation of magnetostrictive properties of Galfenol under thermomechanical deformation". *Finite Elements in Analysis and Design* 127 (2017), pp. 1–5. ISSN: 0168-874X. DOI: <http://dx.doi.org/10.1016/j.finel.2016.11.009>. URL: <http://www.sciencedirect.com/science/article/pii/S0168874X16305820>.
- [114] Ruoqian Liu, Yuksel C Yabansu, Zijiang Yang, Alok N Choudhary, Surya R Kalidindi, and Ankit Agrawal. "Context Aware Machine Learning Approaches for Modeling Elastic Localization in Three-Dimensional Composite Microstructures". *Integrating Materials and Manufacturing Innovation* (2017), pp. 1–12.
- [113] A. Paul, P. Acar, W. Liao, A. Choudhary, V. Sundararaghavan, and A. Agrawal. "Microstructure Optimization of Constrained Design Objectives using Feedback-Aware Data-Generation" (2017). in-preparation.
- [112] Evdokia Popova, Theron M Rodgers, Xinyi Gong, Ahmet Cecen, Jonathan D Madison, and Surya R Kalidindi. "Process-Structure Linkages Using a Data Science Approach: Application to Simulated Additive Manufacturing Data". *Integrating Materials and Manufacturing Innovation* 6.1 (2017), pp. 54–68.

- [111] F. Ram, S. Singh, S.I. Wright, and M. De Graef. “Error Analysis of Crystal Orientations Obtained by the Dictionary Approach to EBSD Indexing”. *Ultramicroscopy* 181 (2017), pp. 17–26.
- [110] Nicole M. Scarborough, G. M. Dilshan P. Godaliyadda, Dong Hye Ye, David J. Kissick, Zhang, Justin A. Newman, Michael J. Sheedlo, Azhad Chowdhury, Robert F. Fischetti, Chittaranjan Das, Gregory T. Buzzard, Charles A. Bouman, and Garth J. Simpson. “Dynamic X-ray Diffraction Sampling for Protein Crystal Positioning”. *Journal of Synchrotron Radiation* 24 (2017), pp. 188–195.
- [109] S. Singh, A. Leff, M. Taheri, and M. De Graef. “Applications of Forward Modeling to Refinement of Grain Orientations”. *Microscopy and MicroAnalysis* 23:S1 (2017), pp. 594–595.
- [108] S. Singh, F. Ram, and M. De Graef. “EMsoft: Open Source Software for Electron Diffraction/Image Simulations”. *Microscopy and MicroAnalysis* 23:S1 (2017), pp. 212–213.
- [107] Suhas Sreehari, S. V. Venkatakrishnan, Katherine L. Bouman, Jeffrey P. Simmons, Lawrence F. Drummy, and Charles A. Bouman. “Muti-Resolution Data Fusion for Super-Resolution Electron Microscopy”. *NITIRE at CVPR*. 2017, p. 213.
- [106] Yue Sun, Ahmet Cecen, John W Gibbs, Surya R Kalidindi, and Peter W Voorhees. “Analytics on large microstructure datasets using two-point spatial correlations: Coarsening of dendritic structures”. *Acta Materialia* 132 (2017), pp. 374–388.
- [105] Veera Sundararaghavan. *Graph theory based software for microstructure fracture: MicroFract2d and MicroFract3d*. <http://umich.edu/~veeras/projects/MicroFract.html>. Accessed: 2017-07-30. 2017.
- [104] Veera Sundararaghavan and Siddhartha Srivastava. “MicroFract: An image based code for microstructural crack path prediction”. *SoftwareX* 6 (2017), pp. 94–97. ISSN: 2352-7110. DOI: <http://dx.doi.org/10.1016/j.softx.2017.04.002>. URL: <http://www.sciencedirect.com/science/article/pii/S2352711017300092>.
- [103] J. Wittkamper, B. Xu, B. Kombaiah, F. Ram, M. De Graef, Kitchinm J.R., G.S. Roher, and P.A. Salvador. “Competitive growth of αPbO_2 and rutile polymorphs of SnO_2 on different orientations of Columbite CoNb_2O_6 substrates”. *Crystal Growth & Design* 17 (2017), pp. 3929–3939.
- [102] Yuksel C Yabansu, Philipp Steinmetz, Johannes Hötzer, Surya R Kalidindi, and Britta Nestler. “Extraction of reduced-order process-structure linkages from phase-field simulations”. *Acta Materialia* 124 (2017), pp. 182–194.
- [101] Jin Yang and Kaushik Bhattacharya. “Efficient Digital Image Correlation To Obtain Deformation Fields”. *Proceedings of the Society of Experimental Mechanics*. 2017.
- [100] Pinar Acar and Veera Sundararaghavan. “A Markov random field approach for modeling spatio-temporal evolution of microstructures”. *Modelling and Simulation in Materials Science and Engineering* 24.7 (2016), p. 075005. DOI: <http://dx.doi.org/10.1088/0965-0393/24/7/075005>. URL: <http://stacks.iop.org/0965-0393/24/i=7/a=075005>.
- [99] Pinar Acar and Veera Sundararaghavan. “Linear Solution Scheme for Microstructure Design with Process Constraints”. *AIAA Journal* 54.12 (2016), pp. 4022–4031. DOI: <http://doi.org/10.2514/1.J055247>.
- [98] Pinar Acar and Veera Sundararaghavan. “Utilization of a Linear Solver for Multiscale Design and Optimization of Microstructures”. *AIAA Journal* 54.5 (2016), pp. 1751–1759. DOI: <http://doi.org/10.2514/1.J054822>.
- [97] Ankit Agrawal and Alok Choudhary. “A Fatigue Strength Predictor for Steels Using Ensemble Data Mining”. *Proceedings of 25th ACM International Conference on Information and Knowledge Management (CIKM) (Demo)*. 2016, pp. 2497–2500.
- [96] Ankit Agrawal and Alok Choudhary. “Perspective: Materials informatics and big data: Realization of the “fourth paradigm” of science in materials science”. *APL Materials* 4.053208 (2016), pp. 1–10.
- [95] Ankit Agrawal, Bryce Meredig, Christopher Wolverton, and Alok Choudhary. “A Formation Energy Predictor for Crystalline Materials Using Ensemble Data Mining”. *Proceedings of IEEE International Conference on Data Mining (ICDM) (Demo)*. 2016, pp. 1276–1279.

- [94] Ahmet Cecen, Tony Fast, and Surya R Kalidindi. “Versatile algorithms for the computation of 2-point spatial correlations in quantifying material structure”. *Integrating Materials and Manufacturing Innovation* 5.1 (2016), p. 1.
- [93] M. Chapman, P.G. Callahan, and M. De Graef. “Determination of sample surface topography using electron back-scatter diffraction patterns”. *Scripta Mater.* 120 (2016), pp. 23–26.
- [92] Xian Chen, Yintao Song, Nobumichi Tamura, and Richard D James. “Determination of the stretch tensor for structural transformations”. *Journal of the Mechanics and Physics of Solids* 93 (2016), pp. 34–43.
- [91] Xian Chen, Nobumichi Tamura, Alastair MacDowell, and Richard D James. “In-situ characterization of highly reversible phase transformation by synchrotron X-ray Laue microdiffraction”. *Applied Physics Letters* 108.21 (2016), p. 211902.
- [90] Abhik Choudhury, Yuksel C Yabansu, Surya R Kalidindi, and Anne Dennstedt. “Quantification and classification of microstructures in ternary eutectic alloys using 2-point spatial correlations and principal component analyses”. *Acta Materialia* 110 (2016), pp. 131–141.
- [89] Tony Fast, Olga Wodo, Baskar Ganapathysubramanian, and Surya R Kalidindi. “Microstructure taxonomy based on spatial correlations: Application to microstructure coarsening”. *Acta Materialia* 108 (2016), pp. 176–185.
- [88] Gero Friesecke, Richard D James, and Dominik Jüstel. “Twisted X-rays: incoming waveforms yielding discrete diffraction patterns for helical structures”. *SIAM Journal on Applied Mathematics* 76.3 (2016), pp. 1191–1218.
- [87] Al’ona Furmanchuk, Ankit Agrawal, and Alok Choudhary. “Predictive analytics for crystalline materials: Bulk modulus”. *RSC Advances* 6.97 (2016), pp. 95246–95251.
- [86] Yaniv Ganor, Traian Dumitrică, Fan Feng, and Richard D James. “Zig-zag twins and helical phase transformations”. *Phil. Trans. R. Soc. A* 374.2066 (2016), p. 20150208.
- [85] G. M. Dilshan Godaliyadda, Dong Hye Ye, Michael D. Uchic, Michael A. Groeber, Gregory T. Buzzard, and Charles A. Bouman. “A Supervised Learning Approach for Dynamic Sampling”. *Proceedings of Computational Imaging XIV at the IS&T Electronic Imaging Symposium*. Vol. 8. 2016, pp. 1–8.
- [84] B.E. Jackson, J.J. Christensen, S. Singh, M. De Graef, D. Fullwood, E. Homer, and R. Wagoner. “Performance of dynamically simulated reference patterns for cross-correlation electron backscatter diffraction”. *Microsc. MicroAnal.* 22 (2016), pp. 789–802.
- [83] Yanfeng Jiang, Vivekanand Dabade, Lawrence F Allard, Edgar Lara-Curzio, Richard James, and Jian-Ping Wang. “Synthesis of α -Fe₁₆N₂ compound anisotropic magnet by the strained-wire method”. *Physical Review Applied* 6.2 (2016), p. 024013.
- [82] Dominik Jüstel, Gero Friesecke, and Richard D James. “Bragg–von Laue diffraction generalized to twisted X-rays”. *Acta Crystallographica Section A: Foundations and Advances* 72.2 (2016), pp. 190–196.
- [81] Surya R Kalidindi, David B Brough, Shengyen Li, Ahmet Cecen, Aleksandr L Blekh, Faical Yannick P Congo, and Carelyn Campbell. “Role of materials data science and informatics in accelerated materials innovation”. *MRS Bulletin* 41.8 (2016), pp. 596–602.
- [80] P L Kuhns, M J R Hoch, S Yuan, A P Reyes, V Srivastava, R D James, and C Leighton. “Magnetic-field-induced changes in superparamagnetic cluster dynamics in the martensitic phase of Ni₄₃Co₇Mn₄₀Sn₁₀”. *Applied Physics Letters* 108.25 (2016), p. 252403.
- [79] A Kumar, L Nguyen, M DeGraef, and V Sundararaghavan. “A Markov random field approach for microstructure synthesis”. *Modelling and Simulation in Materials Science and Engineering* 24.3 (2016), p. 035015. DOI: <http://doi.org/10.1088/0965-0393/24/3/035015>. URL: <http://stacks.iop.org/0965-0393/24/i=3/a=035015>.
- [78] Ruoqian Liu, Ankit Agrawal, Wei-keng Liao, Marc De Graef, and Alok Choudhary. “Materials Discovery: Understanding Polycrystals from Large-Scale Electron Patterns”. *Proceedings of IEEE BigData Workshop on Advances in Software and Hardware for Big Data to Knowledge Discovery (ASH)*. 2016, pp. 2261–2269.

- [77] Ruoqian Liu, Logan Ward, Chris Wolverton, Ankit Agrawal, Wei-Keng Liao, and Alok Choudhary. “Deep Learning for Chemical Compound Stability Prediction”. *Proceedings of ACM SIGKDD Workshop on Large-scale Deep Learning for Data Mining (DL-KDD)*. 2016, pp. 1–7.
- [76] K. Aditya Mohan, Xianghui Xiao, and Charles A. Bouman. “Direct Model-Based Tomographic Reconstruction of the Complex Refractive Index”. *Proceedings of the IEEE International Conference on Image Processing*. Best Student Paper Award (1st place). 2016, pp. 3176–9.
- [75] K.A. Mohan, K.C. Prabhat, C. Phatak, M. De Graef, and C.A. Bouman. “Iterative reconstruction of the magnetization and charge density using vector field electron tomography”. *Microsc. MicroAnal.* 22 (Suppl 3) (2016), pp. 1686–1687.
- [74] Xiaoyue Ni, Julia R Greer, Kaushik Bhattacharya, Richard D James, and Xian Chen. “Exceptional resilience of small-scale $\text{Au}_{30}\text{Cu}_{25}\text{Zn}_{45}$ under cyclic stress-induced phase transformation”. *Nano letters* 16.12 (2016), pp. 7621–7625.
- [73] Daniel P Satko, Joshua B Shaffer, Jaimie S Tiley, S Lee Semiatin, Adam L Pilchak, Surya R Kalidindi, Yoji Kosaka, Michael G Glavicic, and Ayman A Salem. “Effect of microstructure on oxygen rich layer evolution and its impact on fatigue life during high-temperature application of α/β titanium”. *Acta Materialia* 107 (2016), pp. 377–389.
- [72] Ashwin J Shahani, E Begum Gulsoy, Stefan O Poulsen, Xianghui Xiao, and Peter W Voorhees. “Twin-mediated crystal growth: an enigma resolved”. *Scientific reports* 6 (2016).
- [71] Ashwin J Shahani, Xianghui Xiao, Kwan Skinner, Matthew Peters, and Peter W Voorhees. “Ostwald ripening of faceted Si particles in an Al-Si-Cu melt”. *Materials Science and Engineering: A* 673 (2016), pp. 307–320.
- [70] Ashwin J Shahani, Xianghui Xiao, and Peter W Voorhees. “The mechanism of eutectic growth in highly anisotropic materials”. *Nature communications* 7 (2016).
- [69] S. Singh and M. De Graef. “Orientation Sampling for Dictionary-Based Diffraction Pattern Indexing Methods”. *Modeling and Simulations in Materials Science and Engineering* 24 (2016), p. 085013.
- [68] Suhas Sreehari, S. Venkat Venkatakrishnan, Brendt Wohlberg, Gregory T. Buzzard, Lawrence F. Drummy, Jeffrey P. Simmons, and Charles A. Bouman. “Plug-and-Play Priors for Bright Field Electron Tomography and Sparse Interpolation”. *IEEE Trans. on Computational Imaging* 2.4 (2016). IEEE Signal Processing Society 2018 Young Author Best Paper Award, pp. 408–423.
- [67] Philipp Steinmetz, Yuksel C Yabansu, Johannes Hötzer, Marcus Jainta, Britta Nestler, and Surya R Kalidindi. “Analytics for microstructure datasets produced by phase-field simulations”. *Acta Materialia* 103 (2016), pp. 192–203.
- [66] S.V. Venkatakrishnan, L.F. Drummy, M. Jackson, C.A. Bouman, J.P. Simmons, and M. De Graef. “A phantom-based forward modeling approach in support of model-based iterative reconstructions for HAADF-STEM tomography”. *Ultramicroscopy* 160 (2016), pp. 7–17.
- [65] Deepti Verma, Ben Hourahine, Thomas Frauenheim, Richard D James, and Traian Dumitrică. “Directional-dependent thickness and bending rigidity of phosphorene”. *Physical Review B* 94.12 (2016), p. 121404.
- [64] A. Wang and M. De Graef. “Modeling dynamical electron scattering with Bethe potentials and the scattering matrix”. *Ultramicroscopy* 160 (2016), pp. 35–43.
- [63] Logan Ward, Ankit Agrawal, Alok Choudhary, and Chris Wolverton. “A General-Purpose Machine Learning Framework for Predicting Properties of Inorganic Materials”. *npj Computational Materials* 2.16028 (2016).
- [62] S Yuan, PL Kuhns, AP Reyes, JS Brooks, MJR Hoch, V Srivastava, RD James, and C Leighton. “Phase separation and superparamagnetism in the martensitic phase of $\text{Ni}_{50-x}\text{Co}_x\text{Mn}_{40}\text{Sn}_{10}$ ”. *Physical Review B* 93.9 (2016), p. 094425.
- [61] H. Zhao and M. Comer. “A Hybrid Markov Random Field/Marked Point Process Model for Analysis of Materials Images”. *IEEE Transactions on Computational Imaging* 2.4 (2016), pp. 395–407. ISSN: 2333-9403. DOI: 10.1109/TCI.2016.2579601.

- [60] John M Ball and R D James. “Incompatible sets of gradients and metastability”. *Archive for Rational Mechanics and Analysis* 218.3 (2015), pp. 1363–1416.
- [59] Amartya S Banerjee, Ryan S Elliott, and Richard D James. “A spectral scheme for Kohn–Sham density functional theory of clusters”. *Journal of Computational Physics* 287 (2015), pp. 226–253.
- [58] M. Chapman, S. Singh, and M. De Graef. “Can EBSD be used for determination of grain boundary inclination?” *Microsc. MicroAnal.* 21 (Supp. 3) (2015), pp. 2039–2040.
- [57] Y.H. Chen, D. Wei, G. Newstadt, M. De Graef, J.P. Simmons, and A.O. Hero. “Parameter Estimation in Spherical Symmetry Groups”. *IEEE Sign. Proc. Lett.* 22 (2015), pp. 1152–1155.
- [56] Y.H. Chen, D. Wei, G. Newstadt, M. De Graef, J.P. Simmons, and A.O. Hero. “Statistical Estimation and Clustering of Group-invariant Orientation Parameters”. *Information Fusion (18th International Conference on)*. 2015, pp. 719–726.
- [55] JW Gibbs, K Aditya Mohan, EB Gulsoy, AJ Shahani, X Xiao, CA Bouman, M De Graef, and PW Voorhees. “The three-dimensional morphology of growing dendrites”. *Scientific reports* 5.11824 (2015).
- [54] Akash Gupta, Ahmet Cecen, Sharad Goyal, Amarendra K Singh, and Surya R Kalidindi. “Structure–property linkages using a data science approach: application to a non-metallic inclusion/steel composite system”. *Acta Materialia* 91 (2015), pp. 239–254.
- [53] Richard D James. “Materials science: Magnetic alloys break the rules”. *Nature* 521.7552 (2015), pp. 298–299.
- [52] Richard D James. “Taming the temperamental metal transformation”. *Science* 348.6238 (2015), pp. 968–969.
- [51] S. Kalidindi and M. De Graef. “Materials Data Science: Current Status and Future Outlook”. *Annual Review of Materials Research* 45 (2015), pp. 171–193.
- [50] Surya R Kalidindi. *Hierarchical materials informatics: novel analytics for materials data*. Elsevier, 2015.
- [49] Surya R Kalidindi and Marc De Graef. “Materials data science: current status and future outlook”. *Annual Review of Materials Research* 45 (2015), pp. 171–193.
- [48] D. W. Kim and M. L. Comer. “Channel detection in microscope images of materials using marked point process modeling”. *IEEE International Conference on Image Processing*. 2015, pp. 3054–3058.
- [47] Ruoqian Liu, Ankit Agrawal, Zhengzhang Chen, Wei-keng Liao, and Alok Choudhary. “Pruned Search: A Machine Learning Based Meta-Heuristic Approach for Constrained Continuous Optimization”. *Proceedings of 8th IEEE International Conference on Contemporary Computing (IC3)*. 2015, pp. 13–18.
- [46] Ruoqian Liu, Abhishek Kumar, Zhengzhang Chen, Ankit Agrawal, Veera Sundararaghavan, and Alok Choudhary. “A Predictive Machine Learning Approach for Microstructure Optimization and Materials Design”. *Nature Scientific Reports* 5.11551 (2015).
- [45] Ruoqian Liu, Yuksel C Yabansu, Ankit Agrawal, Surya R Kalidindi, and Alok N Choudhary. “Machine learning approaches for elastic localization linkages in high-contrast composite materials”. *Integrating Materials and Manufacturing Innovation* 4.1 (2015), p. 13.
- [44] Ruoqian Liu, Yuksel C. Yabansu, Ankit Agrawal, Surya R. Kalidindi, and Alok N. Choudhary. “Machine learning approaches for elastic localization linkages in high-contrast composite materials”. *Integrating Materials and Manufacturing Innovation* 4.13 (2015), pp. 1–17.
- [43] K. A. Mohan, S. V. Venkatakrishnan, J. W. Gibbs, E. B. Gulsoy, X. Xiao, M. De Graef, P. W. Voorhees, and C. A. Bouman. “4D model-based iterative reconstruction from interlaced views”. *2015 IEEE International Conference on Acoustics, Speech and Signal Processing (ICASSP)*. 2015, pp. 783–787.
- [42] K Aditya Mohan, SV Venkatakrishnan, John W Gibbs, E Begum Gulsoy, Xianghui Xiao, Marc De Graef, Peter W Voorhees, and Charles A Bouman. “4D model-based iterative reconstruction from interlaced views”. *Acoustics, Speech and Signal Processing (ICASSP), 2015 IEEE International Conference on*. IEEE. 2015, pp. 783–787.

- [41] K Aditya Mohan, SV Venkatakrishnan, John W Gibbs, Emine Begum Gulsoy, Xianghui Xiao, Marc De Graef, Peter W Voorhees, and Charles A Bouman. “TIMBIR: A method for time-space reconstruction from interlaced views”. *IEEE Transactions on Computational Imaging* 1.2 (2015), pp. 96–111.
- [40] Maryana I Nave, Jason P Allen, Yu-chen Karen Chen-Wiegart, Jun Wang, Surya R Kalidindi, and Konstantin G Kornev. “In situ X-ray nanotomography of metal surfaces during electropolishing”. *Scientific reports* 5 (2015), p. 15257.
- [39] D.J. Rowenhorst, A.D. Rollett, G.S. Roher, M.A. Groeber, M.A. Jackson, P.J. Konijnenberg, and M. De Graef. “Tutorial: consistent representations of and conversions between 3D rotations”. *Modeling and Simulations in Materials Science and Engineering* 23 (2015), p. 083501.
- [38] AJ Shahani, EB Gulsoy, VJ Roussochatzakis, JW Gibbs, JL Fife, and PW Voorhees. “The dynamics of coarsening in highly anisotropic systems: Si particles in Al-Si liquids”. *Acta Materialia* 97 (2015), pp. 325–337.
- [37] S. Singh and M. De Graef. “Automated dictionary-based indexing of electron channeling patterns”. *Microsc. MicroAnal.* 21 (Supp. 3) (2015), pp. 2041–2042.
- [36] S. Singh and M. De Graef. “GPU-based defect image simulations using the scattering matrix formalism”. *Microsc. MicroAnal.* 21 (Suppl. 3) (2015), pp. 2213–2214.
- [35] S. Sreehari, S. V. Venkatakrishnan, L. F. Drummy, J. P. Simmons, and C. A. Bouman. “Generation of spatial constraints for electron tomographic reconstruction”. *NSRC workshop for Big, Deep, and Smart Data Analytics in Materials Imaging*. 2015.
- [34] S. Sreehari, S. V. Venkatakrishnan, L. F. Drummy, J. P. Simmons, and C. A. Bouman. “Rotationally invariant non-local means for image denoising and tomography”. *Proceedings of the IEEE International Conference on Image Processing*. 2015, pp. 542–546.
- [33] S. Sreehari, S. V. Venkatakrishnan, J. P. Simmons, L. F. Drummy, and C. A. Bouman. “Non-local Prior Modeling for Tomographic Reconstruction of Bright Field Transmission Electron Microscopy Images”. *Microscopy and Microanalysis (M μ M ’15)*. 2015.
- [32] Suhas Sreeharia, S. V. Venkatakrishnan, Lawrence F. Drummy, Jeffrey P. Simmon, and Charles A. Bouman. “Advanced Prior Modeling for 3D Bright Field Electron Tomography”. *Computational Imaging XIII*. Vol. 9401. 2015, 940108 (12 pp.)
- [31] S.V. Venkatakrishnan, L.F. Drummy, M. Jackson, M. De Graef, J.P. Simmons, and C.A. Bouman. “Model-based iterative reconstruction for bright field electron tomography”. *IEEE Trans. Comp. Imag.* 1 (2015), p. 1.
- [30] A. Wang, A.C. Leff, M.L. Taheri, and M. De Graef. “A statistical dictionary approach to automated orientation determination from precession electron diffraction patterns”. *Microsc. MicroAnal.* 21 (Supp. 3) (2015), pp. 1247–1248.
- [29] S.I. Wright, M.M. Nowell, S.P. Lindeman, P.P. Camus, M. De Graef, and M.A. Jackson. “Introduction and comparison of new EBSD post-processing methodologies”. *Ultramicroscopy* 159 (2015), pp. 81–94.
- [28] M. Yan, M. De Graef, Y.N. Picard, and P.A. Salvador. “Electron channeling contrast imaging (ECCI) of anti-phase boundaries in coherently strained La_{0.7}Sr_{0.3}MnO₃ thin films on (110)-oriented SrTiO₃”. *Appl. Phys. Lett.* 107 (2015), p. 041601.
- [27] S Yuan, P L Kuhns, AP Reyes, J S Brooks, M J R Hoch, V Srivastava, R D James, S El-Khatib, and C Leighton. “Magnetically nanostructured state in a Ni-Mn-Sn shape-memory alloy”. *Physical Review B* 91.21 (2015), p. 214421.
- [26] Ankit Agrawal, Parijat D Deshpande, Ahmet Cecen, Gautham P Basavarsu, Alok N Choudhary, and Surya R Kalidindi. “Exploration of data science techniques to predict fatigue strength of steel from composition and processing parameters”. *Integrating materials and manufacturing innovation* 3.1 (2014), p. 8.
- [25] Ankit Agrawal, Parijat D Deshpande, Ahmet Cecen, Gautham P Basavarsu, Alok N Choudhary, and Surya R Kalidindi. “Exploration of data science techniques to predict fatigue strength of steel from composition and processing parameters”. *Integrating Materials and Manufacturing Innovation* 3.8 (2014), pp. 1–19.

- [24] M. De Graef. “Towards a uniform model for lattice defect image simulations”. *Microscopy and MicroAnalysis* 20 (suppl 3) (2014), pp. 1022–1023.
- [23] Lawrence Drummy, Singanallur Venkatakrishnan, Marc De Graef, Jeffrey Simmons, and Charles A. Bouman. “Model-Based Iterative Reconstruction for Multimodal Electron Tomography”. *Data Analytics for Materials Science and Manufacturing symposium, 2014 TMS Annual Meeting & Exhibition*. 2014.
- [22] G. M. Dilshan Godaliyadda, Gregory T. Buzzard, and Charles A. Bouman. “A Model-Based Framework for Fast Dynamic Image Sampling”. *Proceedings of the IEEE International Conference on Acoustics Speech and Signal Processing*. 2014, pp. 1822–1826.
- [21] Emine Begum Gulsoy, Ashwin J Shahani, John W Gibbs, Julie L Fife, and Peter W Voorhees. “Four-dimensional morphological evolution of an aluminum silicon alloy using propagation-based phase contrast X-ray tomographic microscopy”. *Materials Transactions* 55.1 (2014), pp. 161–164.
- [20] Yanfeng Jiang, Vivekanand Dabade, Michael P Brady, Orlando Rios, Richard D James, and Jian-Ping Wang. “9 T high magnetic field annealing effects on FeN bulk sample”. *Journal of Applied Physics* 115.17 (2014), 17A758.
- [19] Ruoqian Liu, Ankit Agrawal, Wei-keng Liao, and Alok Choudhary. “Search Space Preprocessing in Solving Complex Optimization Problems”. *Proceedings of IEEE BigData Workshop on Complexity for Big Data (C4BD)*. 2014, pp. 1–5.
- [18] Bryce Meredig, Ankit Agrawal, S Kirklin, J E Saal, J W Doak, A Thompson, Kunpeng Zhang, Alok Choudhary, and Christopher Wolverton. “Combinatorial screening for new materials in unconstrained composition space with machine learning”. *Physical Review B* 89.094104 (2014), pp. 1–7.
- [17] K. Aditya Mohan, S. V. Venkatakrishnan, Lawrence F. Drummy, Jeffrey Simmons, Dilworth Y. Parkinson, and Charles A. Bouman. “Model-Based Iterative Reconstruction for Synchrotron X-ray Tomography”. *Proceedings of the IEEE International Conference on Acoustics Speech and Signal Processing*. 2014, pp. 6909–6913.
- [16] Y.N. Picard, M. Liu, J. Lammatao, R. Kamaladasa, and M. De Graef. “Theory of dynamical electron channeling contrast images of near-surface crystal defects”. *Ultramicroscopy* 146 (2014), pp. 71–78.
- [15] D. Rořca, A. Morawiec, and M. De Graef. “A new method of constructing a grid in the space of 3D rotations and its applications to texture analysis”. *Modeling and Simulations in Materials Science and Engineering* 22 (2014), p. 075013.
- [14] Ashwin J Shahani, E Begum Gulsoy, John W Gibbs, Julie L Fife, and Peter W Voorhees. “Integrated approach to the data processing of four-dimensional datasets from phase-contrast x-ray tomography”. *Optics express* 22.20 (2014), pp. 24606–24621.
- [13] H Shi, R Delville, V Srivastava, R D James, and D Schryvers. “Microstructural dependence on middle eigenvalue in Ti–Ni–Au”. *Journal of Alloys and Compounds* 582 (2014), pp. 703–707.
- [12] Gal Shmuel, Adam Thor Thorgeirsson, and K Bhattacharya. “Wavelet analysis of microscale strains”. *Acta Mat.* 76 (2014), pp. 118–126.
- [11] J. P. Simmons, C. Przybyla, S. Bricker, Dae Woo Kim, and M. L. Comer. “Physics of MRF regularization for segmentation of materials microstructure images”. *IEEE International Conference on Image Processing*. 2014, pp. 4882–4886.
- [10] Veera Sundararaghavan. “Reconstruction of three-dimensional anisotropic microstructures from two-dimensional micrographs imaged on orthogonal planes”. *Integrating Materials and Manufacturing Innovation* 3.1 (2014), p. 19. ISSN: 2193-9772. DOI: 10.1186/s40192-014-0019-3. URL: <https://doi.org/10.1186/s40192-014-0019-3>.
- [9] S.V. Venkatakrishnan, M.-S. Hsiao, N. Garvin, M.A. Jackson, M. De Graef, J.P. Simmons, C.A. Bouman, and L.F. Drummy. “Model-based iterative reconstruction for low-dose electron tomography”. *Microscopy and MicroAnalysis* 20 (suppl 3) (2014), pp. 802–803.
- [8] F. Yang, F. Scheltens, D. McComb, D.B. Williams, and M. De Graef. “Absorption corrections for a four-quadrant SuperX EDS detector”. *Microscopy and MicroAnalysis* 20 (suppl 3) (2014), pp. 100–101.

- [7] H. Zhao, M. L. Comer, and M. De Graef. “A Unified Markov Random Field / Marked Point Process Image Model and Its Application to Computational Materials”. *IEEE International Conference on Image Processing*. 2014, pp. 1713–1716.
- [6] Xian Chen, Vijay Srivastava, Vivekanand Dabade, and Richard D James. “Study of the cofactor conditions: conditions of supercompatibility between phases”. *J. Mech. Phys. Solids* 61.12 (2013), p. 2566.
- [5] Oren Y Kanner, Doron Shilo, Jian Sheng, Richard D James, and Yaniv Ganor. “Ferromagnetic shape memory flapper for remotely actuated propulsion systems”. *Smart Materials and Structures* 22.8 (2013), p. 085030.
- [4] G Shmuel, A T Thorngiesson, and K Bhattacharya. “Applications of wavelets in the representation and prediction of transformation in shape-memory polycrystals”. *Proc. TMS Symposium on Data Analytics for Materials Science and Manufacturing*. Ed. by Simmons, C Bouman, F Fahroo, S R Kalidindi, J Knopp, and P W Voorhees. 2013, pp. 1–8.
- [3] Yintao Song, Kanwal Bhatti, Vijay Srinivasan, Chris Leighton, and Richard D James. “Thermodynamics of energy conversion via first order phase transformation in low hysteresis magnetic materials”. *Energy Environ. Sci.* 6.4 (2013), p. 1315.
- [2] Yintao Song, Xian Chen, Vivekanand Dabade, T W Shield, and Richard D James. “Enhanced reversibility and unusual microstructure of a phase-transforming material”. *Nature* 502 (2013), p. 85.
- [1] Singanallur V. Venkatakrishnan, Charles A. Bouman, and Brendt Wohlberg. “Plug-and-Play Priors for Model Based Reconstruction”. *IEEE Global Conference on Signal and Information Processing (Global-SIP)*. 2013, pp. 945–948.