APS Workshop 2: Advancing Materials Research: Synergies in Large Scale Simulations and APS-U Imaging and Microscopy Techniques

Wednesday, May 7, Morning

8:30 – 9:00 Ross Harder and Jon Tischler (X-ray Science, Argonne National Laboratory)
Introduction and Opening Remarks

9:00 – 9:30 Anter El-Azab (School of Materials Engineering, Purdue University)
Interfacing Continuum Dislocation Dynamics with DAXM Technique and Theory-inspired Future Experiments

9:30 – 10:00 Felix Hofmann (Department of Engineering Science, University of Oxford)
Irradiation-induced Structure and Property Evolution in Metals: Combining Large-scale Atomistic Simulations and X-ray Observations

10:00 – 10:30 Break

10:30 – 11:00 Subramanian Sankaranarayanan (Center for Nanoscale Materials, Argonne National Laboratory)
Probing Defect Dynamics through Integrated Molecular Simulations, Imaging, and Machine Learning

11:00 – 11:30 Youping Chen (University of Florida)
Concurrent Atomistic-continuum Simulation of Materials and Heterostructures from Synthesis to Microstructure and Properties

11:30 – 12:00 Anthony Rollett (Department of Materials Science and Engineering, Carnegie Mellon University)
Synchrotron X-ray Microscopy Combined with Simulation and Machine Learning

12:00 – 1:30 Lunch Break

Wednesday, May 7, Afternoon

1:30 – 2:00 Khaled SharafEldin (School of Materials Engineering, Purdue University)
Machine Learning of Dislocation Microstructure from DAXM Data and a Theoretical Perspective from Mesoscale Theory of Crystal Defects

2:00 – 2:30 Leora Dresselhaus-Marais (Geballe Laboratory for Advanced Materials, Stanford University)
Subsurface Imaging of Multi-timescale Defect Dynamics

2:30 – 3:00 Stephan O. Hruszkewycz (Materials Science, Argonne National Laboratory)
Guiding High-energy Coherent Diffraction and HEDM Measurements with Mesoscale Crystal Plasticity Modeling

3:00 – 3:30 Break

3:30 – 4:00 Mathew J. Cherukara (X-ray Science, Argonne National Laboratory)
AI-enhanced X-ray Imaging: Bridging Length Scales to Visualize Materials Dynamics

4:00 – 4:30 Paul Evans (Materials Science and Engineering, University of Wisconsin-Madison)
Solid-phase Crystallization of Complex Oxides: Complementarity of X-ray Nanobeam Diffraction and Molecular Simulation

4:30 – 5:00 Ian McNulty (Lund University)
Closing Discussion

5:00 Adjourn

Interfacing Continuum Dislocation Dynamics with DAXM Technique and Theory-inspired Future Experiments

Anter El-Azab[1]

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I will review the continuum theory of dislocation dynamics (CDD) and present some results relevant to the use of the high-resolution differential-aperture x-ray structural microscopy (DAXM) technique in probing the dislocation structure in deformed crystals. CDD is a theory of mesoscale deformation that has been successfully used to predict dislocation cell formation in FCC crystals in recent years. I will also discuss the combined use of CDD and DAXM (and related synchrotron-based methods) toward understanding the physics of self-organization of dislocations during plastic deformation of metals. In particular, I will discuss the potential use of high-resolution DAXM to measure the dislocation-dislocation correlations in crystals, which is critical for the development of generation II of the CDD theory.

Irradiation-induced Structure and Property Evolution in Metals: Combining Large-scale Atomistic Simulations and X-ray Observations

Felix Hofmann[1], Ivan Tolkachev[1], Suchandrima Das[2], David Yang[1,3], Samanyu Tirumala[1, 4], Daniel Long[1], Daniel Mason[4], Pui-Wai Ma[4], Sergei Dudarev[4], Max Boleininger[4], Ross Harder[5], Wonsuk Cha[5], Dina Sheyfer[5], Wenjun Liu[5], and Jon Tischler[5]

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Nuclear Fusion promises an almost limitless, low-carbon energy source. Materials inside future reactors will be exposed to high temperatures, large heat fluxes, and intense irradiation with neutrons and ions. This irradiation causes the formation of defects in the material that can degrade the structural performance and alter physical properties. For reactor design, the prediction of irradiation-induced material evolution remains a major challenge.

Using x-ray micro- and coherent diffraction, we have studied the evolution of irradiation damage in tungsten, the front runner material for fusion armor. With the aid of molecular dynamics simulations, we shed light on the underlying mechanisms that cause the dose-dependent lattice swelling and contraction observed in experiments [1, 2].

It has been proposed that nano-structuring may reduce irradiation-induced material evolution, as grain boundaries could act as sinks for irradiation damage. We explore this hypothesis using large-scale molecular dynamics simulations and x-ray diffraction measurements on iron [3]. Nano-crystalline material is made using severe plastic deformation and subsequently irradiated at room temperature. Our results show irradiation-induced annealing and grain growth. They further suggest that material under irradiation eventually converges to a critical stored energy density, irrespective of the initial starting state.

An interesting further dimension stems from the interaction of defects with hydrogen, and I will briefly present our very recent work on this [4].

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Probing Defect Dynamics through Integrated Molecular Simulations, Imaging, and Machine Learning

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Defects critically determine the performance of materials in a variety of different energy applications, yet their real-time behavior under operational conditions remains elusive. By integrating molecular simulations with high-resolution x-ray imaging and machine learning-driven data analytics, we aim to achieve a holistic, multiscale understanding of defect dynamics. Using several representative examples from 2D to bulk materials and modalities from microscopy to x-ray imaging, we demonstrate an improved understanding of defect structure/evolution and structural transition. Our combined approach can potentially enable direct visualization of atomic-to-mesoscale defect structures and interactions, provides quantitative insights into the mechanisms driving defect evolution, and offers predictive modeling capabilities, paving the way for the rational design of defect-tolerant materials.

Concurrent Atomistic-continuum Simulation of Materials and Heterostructures from Synthesis to Microstructure and Properties

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This presentation will introduce a concurrent atomistic-continuum (CAC) method that can be used to simulate the nonequilibrium processes of materials synthesis, defect formation, fracture, and phonon thermal transport. Building on Kirkwood's statistical mechanical theory of transport processes, the CAC formulation extends Kirkwood's theory for ensemble-averaged single-component single-phase hydrodynamical systems to a concurrent two-level structural description of crystalline materials [1, 2]. It links and unifies atomistic and continuum descriptions of conservation laws, stress, heat flux, and temperature. This naturally leads to a multiscale methodology that provides a unified treatment for microstructural, mechanical, and thermal transport processes, without the need of any empirical rules or parameters other than an interatomic potential. A massive parallel algorithm has been developed to implement the formulation in the LAMMPS codebase [3]. Numerical examples of concurrent multiscale simulation of the dynamic processes of heteroepitaxy, defect formation during direct bonding and heteroepitaxy [4], and collective behavior of interfaces, dislocations, and phonons [5], will be present, offering insights into the multiscale dynamic processes, as well as the mechanisms for defect formation, materials failure, and phonon scattering by defects and interfaces in epitaxial heterostructures.

This work is based on research supported by the US National Science Foundation under Award Numbers 2054607 and 2349160. The current version of the CAC method was developed in collaboration with Adrian Diaz, Yang Li, Boyang Gu, David McDowell, and Steve Plimpton.

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Synchrotron X-ray Microscopy Combined with Simulation and Machine Learning

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Additive manufacturing (AM), *aka* 3D printing, is a relatively new technology that has given rise to the "maker culture" and an intense interest in design. That has carried over into metals AM, which has jumped almost immediately into manufacturing of actual parts in a variety of alloys. In doing so it has liberated thinking about part design albeit within certain constraints and complex components have been deployed that were previously inaccessible. One example is described of the co-design of HX against printing constraints, alongside evolution in alloy choice. Another example is the development of a digital twin for processing-microstructure-fatigue in 3D printed metals. These practical applications expose the issue that the reliability of parts that must carry load depends on the internal micro-structure, acutely so in fatigue loading. This motivates detailed study of all aspects of materials microstructure ranging from defect structures to strain, all of which is ideally suited to the use of intense sources of high energy x-rays as only advanced light sources can deliver. Computed tomography (CT) has revealed the presence of porosity in all additively manufactured metals examined to date and confirmed that appropriate process control can limit it. CT has also provided data on surface condition which directly affects fatigue performance. High speed radiography reveals even more crucial details of how laser light generates vapor cavities that can deposit voids past a critical instability point. "Hot" cracking has been imaged as it happens during the solidification process, which offers the possibility finding printing recipes for alloys previously considered off-limits to 3D printing. High-speed, high-resolution diffraction microscopy in stainless steel, alloy 718, and Ti-6Al-4V reveals unexpected solidification and precipitation sequences. Diffraction microscopy reveals the highly strained nature of printed metals and how microstructure and internal strain state evolves during subsequent annealing and/or annealing. Permeating all these activities is direct simulation and machine learning as an invaluable aid to the researcher. Pores, for example, need to be understood in terms of their size, location, shape, and the stress + strain concentrations that occur under load. Direct simulation with elasto-viscoplastic micro-mechanics with crystal plasticity can be done with good efficiency but need to be reformulated as a reduced-order model for efficient use in workflows that can efficiently evaluate process variations in a digital twin.

Machine Learning of Dislocation Microstructure from DAXM Data and a Theoretical Perspective from Mesoscale Theory of Crystal Defects

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We present a study that leverages high-resolution differential-aperture x-ray structural microscopy (DAXM) to probe the local dislocation microstructure in deformed 304-steel at small strain, by measuring the lattice rotation and deviatoric elastic strain with a sub-micron resolution. For a single grain in a polycrystalline specimen, the measured lattice rotation field over the measured volume exhibited a multimodal distribution while the deviatoric elastic strain showed a single-mode distribution. An informed, unsupervised Cauchy mixture machine learning model was developed to resolve the multimodal distribution of the lattice rotation. By mapping the lattice rotation data associated with each Cauchy peak in the model back onto the measured volume, we identify contiguous regions of the crystal rotated near the average values corresponding to the peaks of the overall rotation distribution. These regions represent the grain subdivision in the microstructure. The dislocation density tensor was also computed, and its norm was laid over the rotation field to detect the subgrain boundaries. This step provided a validation of the Cauchy mixture model for the analysis of the lattice rotation distribution. The current study highlights the integration of advanced x-ray microscopy techniques with data-driven analysis methods to uncover detailed microstructure scales in deformed crystals.

Subsurface Imaging of Multi-timescale Defect Dynamics

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Connecting the multiscale behaviors of defects to properties is a longstanding challenge in materials science. TEMs have imaged dislocations and their dynamics for a long time, but uncertainties persist as to how those dynamics translate to subsurface dynamics, especially under different types of loading conditions. My group has developed the time-resolved versions of multiscale x-ray diffraction imaging techniques between synchrotrons and x-ray free electron lasers (XFELs). Using dark-field x-ray microscopy (DFXM) and x-ray topography, we can image strain fields hundreds of micrometers beneath a material's surface that encode defects and thermomechanical waves in crystalline materials – from ms to fs timescales. I will present my group's work developing these instruments, building the theory and computational simulations to interpret them, and selected findings that reveal opportunities in translating synchrotron and XFEL insights. Our work provides context of how multiscale subsurface defect dynamics describe material responses relevant to the mechanical and thermal properties of materials.

Guiding High-energy Coherent Diffraction and HEDM Measurements with Mesoscale Crystal Plasticity Modeling

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The simultaneous capabilities of the newly upgraded APS and the Leadership Computing Facility at Argonne offer exciting possibilities and can make a big impact in our approach to characterizing and modeling polycrystalline bulk structural materials ubiquitous in our everyday lives. This talk will introduce the synergistic aspects of crystal plasticity finite element (CPFE) simulations, high energy diffraction microscopy (HEDM), and high-energy Bragg coherent diffraction imaging (BCDI). We will cover opportunities to develop a workflow that integrates these modeling and measurement modalities, contexts in which the micron-scale resolution of HEDM and the nanoscale resolution of BCDI can reveal new aspects of materials, and materials science questions that can benefit from this approach.

AI-enhanced X-ray Imaging: Bridging Length Scales to Visualize Materials Dynamics

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Advanced photon sources with enhanced coherence, brightness, and spatial resolution are revolutionizing materials characterization by enabling unprecedented visualization of microstructural evolution across multiple length and time scales. However, the extraordinary increase in data volume and complexity poses significant challenges for traditional analysis methods. This talk will present emerging frameworks that synergistically combine large-scale simulations with artificial intelligence to transform how we process, analyze, and interpret advanced x-ray characterization data.

I will highlight three complementary approaches. First, I will discuss simulation-informed physics-aware deep learning frameworks for rapid 3D nanoscale coherent imaging, which dramatically accelerate phase retrieval processes for Bragg coherent diffraction imaging (BCDI). Second, I will present unsupervised learning methods for automated classification of relaxation dynamics in x-ray photon correlation spectroscopy (XPCS) that can enable direct correlation between microscopic dynamics and macroscopic properties. Third, agentic frameworks that can serve as intelligent intermediaries between experimental x-ray data and physics-based simulations.

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Solid-phase Crystallization of Complex Oxides: Complementarity of X-ray Nanobeam Diffraction and Molecular Simulation

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Complex oxides can be created in novel three-dimensional geometries and with otherwise challenging compositions by controlling the transformation between the amorphous and crystalline states in the solid state. This transformation is accompanied by phenomena that are unusual among crystal growth techniques. A particularly interesting series of questions arises because of combination of (i) the large (~10%) difference in density between the amorphous and crystalline states and (ii) long timescales for the plastic relaxation of applied stresses. We have created thin-film crystallization environments that probe the crystallization of $SrTiO_3$ under highly controlled geometries. X-ray nanobeam diffraction characterization revealed that crystallization over lateral distances of microns produces an unusual "rotating lattice" microstructure driven by preferential nucleation of specific dislocations [1]. The crystallized structures have piezoelectric and second-harmonic-generation signatures consistent with noncentrosymmetry [1]. The defect configuration is driven by the low-symmetry of stress at the amorphous crystalline interface. Complementary molecular simulations reveal other interface-driven effects, including a tilting of the interface and predictions of the stress and orientation-dependence of the growth rate. Beyond these initial experiments and nanoscale simulations, mesoscopic features of the crystallization continue to pose challenges that can be addressed by advances in structural characterization and computational predictions.

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