NREL

Autonomous Research for Real-World Science

ARROWS Workshop

May 19-21, 2025

Cecil H. & Ida Green Graduate and Professional Center
Colorado School of Mines
Golden, CO

Hosted by the National Renewable Energy Laboratory and Colorado School of Mines

Welcome

It is our distinct pleasure to welcome you to the **Autonomous Research for Real-World Science** (ARROWS) workshop, hosted by the National Renewable Energy Laboratory (NREL) and the Colorado School of Mines! We stand at a pivotal moment where the fields of materials science, chemistry, and autonomous experimentation are converging, creating unprecedented opportunities for discovery and innovation. AI-driven research methods and autonomous experimentation promise to fundamentally transform how we approach scientific challenges, accelerating the pace of development for practical, real-world solutions. ARROWS is dedicated to exploring this exciting intersection.

Over the next three days, we bring together leading experts to delve into the practical applications and future potential of autonomous systems in science. We will explore key themes including the grand challenges in materials and chemistry ripe for autonomous approaches, the development of domain-specific analytics and visualization tools, the intricacies of autonomous decision-making in experimental workflows, and the real-world technical implementations that bring these concepts to life. Our aim is not only to showcase cutting-edge research but also to foster new collaborations, identify critical challenges and opportunities, and collectively strategize pathways for groundbreaking research and future funding initiatives.

We are thrilled to have you join us for what promises to be a stimulating and productive workshop. We encourage active participation, open discussion, and the forging of new partnerships as we collectively shape the future of autonomous, real-world science.

Warm regards,

Steven R. Spurgeon
National Renewable Energy Laboratory
Colorado School of Mines

Hilary Egan
National Renewable Energy Laboratory

Andriy Zakutayev
National Renewable Energy Laboratory

ARROWS Workshop: Agenda

May 19-21, 2025 • Golden, CO

Arrive Sunday, May 18, 2025

Monday, May 19, 2025

7:30 – 8:30 am	REGISTRATION, Green Center, Colorado School of Mines
8:30 – 9:00 am	**Welcome & Meeting Overview** **Peter Green, Deputy Director,** *National Renewable Energy Laboratory* **Steven R. Spurgeon,** *National Renewable Energy Laboratory* **Hilary Egan,** *National Renewable Energy Laboratory* **Andriy Zakutayev,** *National Renewable Energy Laboratory*
9:00 – 10:30 am	***SESSION 1: Platforms for Autonomous Discovery*** *Chair: Andriy Zakutayev, National Renewable Energy Laboratory*
9:00 am	**Autonomous Materials Exploration for Thin-film and Solid Materials** **Taro Hitosugi,** *University of Tokyo*
9:30 am	**Data-Rich Autonomous Labs for Accelerated Materials Discovery** **Milad Abolhasani (Remote),** *North Carolina State University*
10:00 am	**AI, Microscopes, and the Quest for Better Materials** **Sergei V. Kalinin,** *University of Tennessee–Knoxville*
10:30 – 11:00 am	Discussion Break
11:00 – 12:30 pm	***SESSION 2: AI & Models for Autonomous Science*** *Chair: Robert White, National Renewable Energy Laboratory*
11:00 am	**Accelerating Design of Organic Materials with Autonomous AI agents** **Olexandr Isayev,** *Carnegie Mellon University*
11:30 am	**Automated Continuous-Flow Synthesis, Foundation Models, and Multi-Agent Systems for Accelerated Discovery of Polymers** **Nathaniel Park,** *IBM Research*
12:00 pm	**Advanced Materials Meet Causal Learning** **Ayana Ghosh,** *Oak Ridge National Laboratory*
12:30 – 1:30 pm	Working Lunch
1:30 – 3:00 pm	***SESSION 3: Synthesis Workflows & Integration*** *Chair: Davi Febba, National Renewable Energy Laboratory*
1:30 pm	**Enabling Autonomous Synthesis and Compositional Optimization in Epitaxial Film Growth via Machine Learning and Data Analytics** **Ryan Comes,** *University of Delaware*

Monday, May 19, 2025 Continued

2:00 pm	**AI for Accelerated Materials Discovery at Intel Labs** **Santiago Miret (Remote),** *Intel Company*
2:30 pm	**Real-Time Autonomous Combinatorial Experimentation – Nimble and Agile Approaches to Self-Driving Laboratories** **Ichiro Takeuchi,** *University of Maryland*
3:00 – 3:30 pm	Discussion Break
3:30 – 5:00 pm	**Breakouts: Identifying Gaps and Opportunities**
5:00 – 6:00 pm	Free Time / Travel to Mixer
6:00 – 8:00 pm	**Mixer, Buffalo Rose (Golden, CO)**

Tuesday, May 20, 2025

8:00 – 9:00 am	REGISTRATION
9:00 – 10:30 am	***SESSION 4: Autonomous Synthesis & Characterization*** *Chair: Robert Epps, National Renewable Energy Laboratory*
9:00 am	**Nanomaterials Discovery Through AI-Enhanced Characterization: Faster, Cheaper & Better?** **Vinayak Dravid,** *Northwestern University*
9:30 am	**Challenges in Self-Driving Laboratories: Curiosity-Based Explorations, and the Need for Human-in-the-Loop Workflows for Maximizing Robustness and Prior Knowledge** **Rama Vasudevan,** *Oak Ridge National Laboratory*
10:00 am	**Advancing Ionomer-Based Water Electrolysis: Integrated Characterization, Foundation Models for Segmentation, and Autonomous Optimization** **Dani Ushizima,** *Lawrence Berkeley National Laboratory*
10:00 – 10:30 am	Discussion Break
10:30 – 12:00 pm	***SESSION 5: In Situ Discovery and Visualization*** *Chair: Patrick Emami, National Renewable Energy Laboratory*
10:30 am	**Integrating Automation Tactics and Machine Learning to Direct Nanoscale and Atomic Scale Transformation Experiments** **Raymond Unocic,** *North Carolina State University*
11:00 am	**Towards Machine-Learning Enabled Automated Analysis of Heterogeneous Nanomaterials** **Mary Scott,** *Lawrence Berkeley National Laboratory*
11:30 am	**Practically Speaking: The Engines that Automate** **Matthew Olszta,** *Pacific Northwest National Laboratory*

Tuesday, May 20, 2025 Continued

12:00 – 12:30 pm	Discussion Break
12:30 – 1:30 pm	Working Lunch
1:30 – 4:30 pm	***NREL Facility Tours*** *Hosts: Steven Spurgeon, Andriy Zakutayev, Hilary Egan, Brooks Tellekamp*
1:30 – 2:00 pm	*Shuttle to NREL* Verified Tour Attendees
2:00 – 4:00 pm	**NREL Facility Tours** **(Combi Synthesis, Autonomous Microscopy, Supercomputing)**
4:00 pm	*Return to Mines*
4:30 – 5:00 pm	Free Time / Discussion Break
5:00 – 7:00 pm	**Poster Session & Refreshments**

Wednesday, May 21, 2025

*Day 3 will only consist of discussions and writing of a perspective article for those interested in participating.

8:30 – 9:00 am	Morning Coffee & Networking
9:00 – 11:00 am	**Breakout Writing Session Part 1:** **Collaborative Perspective Article**
11:00 – 11:45 am	Discussion Break / Group Photo
11:45 – 1:00 pm	**Breakout Writing Session Part 2:** **Collaborative Perspective Article**
1:00 – 2:00 pm	Working Lunch
2:00 – 2:15 pm	**Concluding Remarks** **Steven R. Spurgeon,** *National Renewable Energy Laboratory* **Hilary Egan,** *National Renewable Energy Laboratory* **Andriy Zakutayev,** *National Renewable Energy Laboratory*

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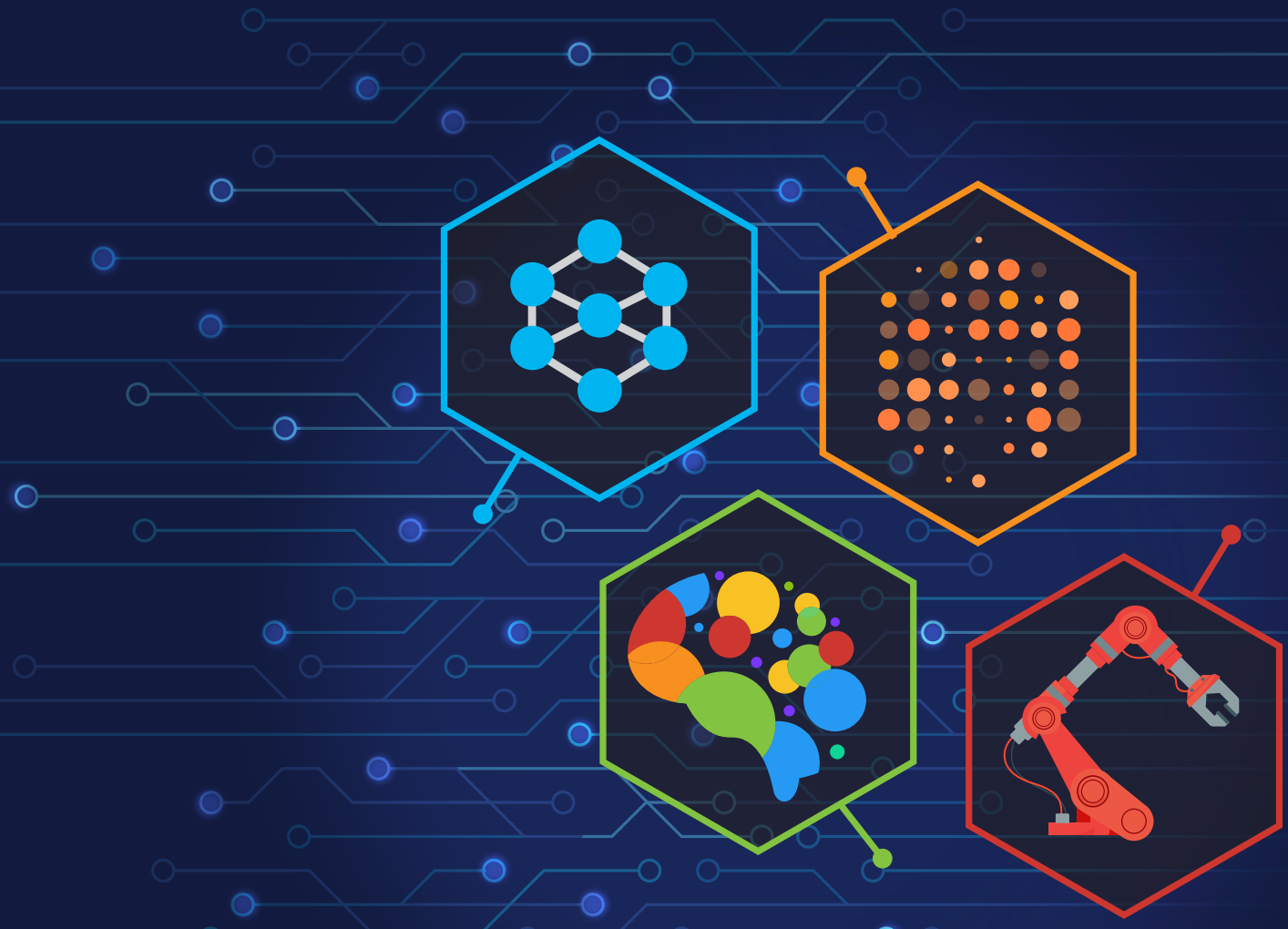
Abstracts



Data-Rich Autonomous Labs for Accelerated Materials Discovery

Milad Abolhasani[1]

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Quantum dots (QDs), semiconductor nanocrystals with size- and composition-dependent electronic and optical properties, offer significant potential for applications in renewable energy, quantum information processing, and optoelectronic devices. However, traditional synthesis and characterization methods for QDs are typically slow, resource-intensive, and challenging to scale, limiting rapid discovery and industrial implementation. To address these limitations, our group has developed *Data-Rich Autonomous Labs* that integrate modular fluidic reactors, robotics, automated experimentation, in-situ multi-modal characterization, and artificial intelligence (AI)-driven decision-making to substantially accelerate quantum dot discovery and precision manufacturing.
Autonomous labs have emerged as powerful platforms capable of rapidly exploring extensive synthesis-property parameter spaces of advanced functional materials. Recent advancements in autonomous experimentation have been primarily driven by *process intensification* principles, designed to streamline workflows through enhanced speed, safety, and resource efficiency. Specifically, the integration of microscale fluidic reactors and real-time characterization techniques enables precise control of reaction conditions, rapid equilibration, enhanced heat and mass transfer, and significant reduction in chemical consumption.

Despite these advancements, current autonomous lab platforms largely rely on steady-state experimental approaches, neglecting valuable insights available from transient-state conditions. Leveraging transient-state data can offer deeper mechanistic understanding of synthesis pathways and substantially enrich datasets available for AI-driven decision-making. This work introduces *dynamic flow experimentation* as an innovative approach for data-intensive exploration of QD synthesis parameters. By systematically varying reaction parameters during continuous operation and employing real-time *in-situ* characterization, these dynamic experiments generate comprehensive time-series data. This approach allows direct correlation of instantaneous flow conditions to steady-state equivalent residence times, significantly intensifying data generation rates.

In this talk, I will present a data-rich autonomous lab utilizing a dynamic experimentation method that operates at least 100 times faster than conventional steady-state synthesis techniques, simultaneously reducing chemical consumption by more than threefold. Using cadmium selenide (CdSe) QDs as a representative case study, I will demonstrate how this novel approach enables rapid and comprehensive mapping of synthesis parameters to material properties. Furthermore, the extensive dataset generated supports the development of robust digital twins, facilitating precise autonomous closed-loop experimentation to optimize multiple emission wavelengths for advanced photonic and energy applications.

Enabling Autonomous Synthesis and Compositional Optimization in Epitaxial Film Growth via Machine Learning and Data Analytics

Ryan Comes[1,2], Patrick Gemperline[2], Sumner Harris[3], and Rama Vasudevan[3]

[1.] University of Delaware, Department of Materials Science and Engineering, Newark, DE, USA
[2.] Auburn University, Department of Physics, Auburn, AL, USA
[3.] Oak Ridge National Laboratory, Center for Nanophase Materials Sciences, Oak Ridge, TN, USA

Machine learning (ML) has revolutionized the field of materials science, particularly in the synthesis of epitaxial films. By leveraging ML algorithms, researchers can optimize synthesis parameters, predict material properties, and enhance real-time monitoring techniques. *In situ* characterization during film growth provides an avenue to enable our goal of ML-aided film growth. The integration of ML in epitaxial film synthesis involves several key steps, starting with high-quality, real-time data collection from in situ monitoring techniques such as reflection high energy electron diffraction (RHEED). Our work focuses on addressing the challenges in analyzing RHEED data during molecular beam epitaxy (MBE) and pulsed laser deposition (PLD) synthesis.

By implementing advanced data analytics techniques such as principal component analysis (PCA) and k-means clustering [1], we have enhanced the precision of real-time monitoring with greater sensitivity to subtle features in the images. With a new residual sum-of-squares (RSS) alignment algorithm, we have enabled the quantitative comparison of RHEED videos between samples, facilitating the identification of optimal growth conditions and improving the reproducibility of epitaxial film synthesis.

Building on this foundation, have expanded into the application of deep learning to predict the stoichiometry of $Sr_{2x}Ti_{2(1-x)}O_3$ thin films using RHEED images acquired during PLD [2]. A gated convolutional neural network trained for regression of the Sr atomic fraction achieved accurate predictions with a small dataset of 31 samples. Explainable AI techniques revealed a previously unknown correlation between diffraction streak features and cation stoichiometry in $Sr_{2x}Ti_{2(1-x)}O_3$ thin films. This study highlights how ML can transform a ubiquitous in-situ diagnostic tool, typically limited to qualitative assessments, into a quantitative surrogate measurement of continuously valued thin film properties. Such methods are critically needed to enable real-time control, autonomous workflows, and accelerate traditional synthesis approaches.

Looking forward, the advancements presented in these studies pave the way for the future of autonomous material synthesis [3] and real-time monitoring. By leveraging ML, researchers can achieve efficient and reproducible epitaxial film synthesis, ultimately contributing to the goals of developing high-quality materials with tailored properties. The synergy between advanced data analytics and deep learning techniques offers a promising direction for the continued evolution of ML-assisted epitaxial film synthesis, enabling more precise control and accelerated discovery in materials science.

[1] P.T. Gemperline, et al. Journal of Vacuum Science and Technology A, **43** (2025), 032701.
[2] S.B. Harris*, P.T. Gemperline*, et al. Nano Letters, Article ASAP (2025),
[3] T. Kaspar, et al. Journal of Vacuum Science and Technology A, **43** (2025), 032702.
[4] This work was supported by the Center for Nanophase Materials Sciences (CNMS), which is a U.S. Department of Energy, Office of Science User Facility at Oak Ridge National Laboratory. P.T.G. and

R.B.C. gratefully acknowledge funding for RHEED analytics from the National Science Foundation Division of Materials Research under award DMR-2045993. P.T.G. also acknowledges support from the Department of Energy's Office of Science Graduate Student Research Program (DE-SC0014664). The authors also acknowledge assistance from Microsoft Copilot in the generation of this abstract.

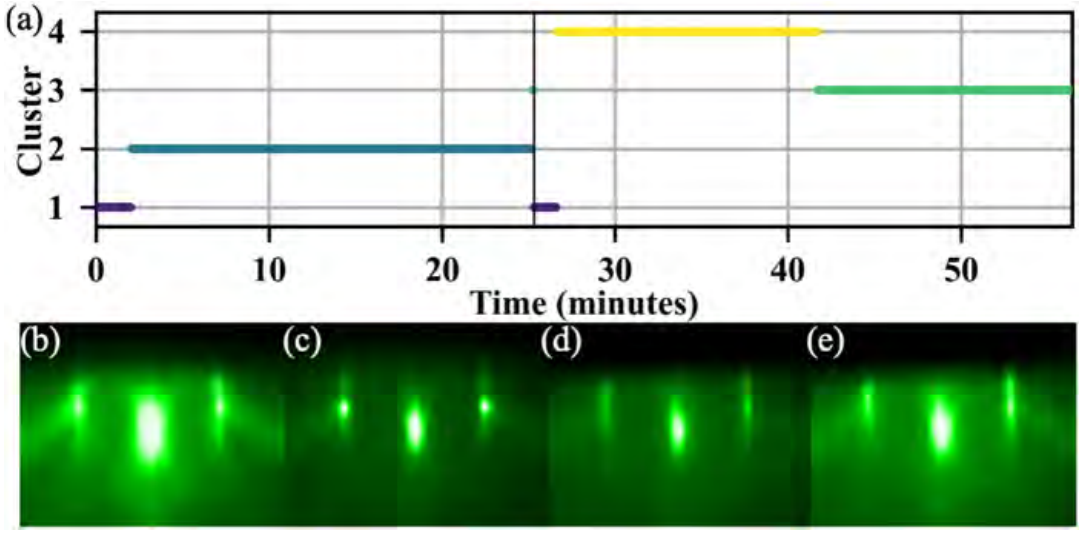

Figure 1. Results from the k-means clustering of a recording containing 2 LFO samples. The black vertical line at min represents the division between the samples. (a) A graph of the clusters over the course of the growths; (b)–(e) the centroid images corresponding to the clusters in (a) in an ascending numerical order left to right.

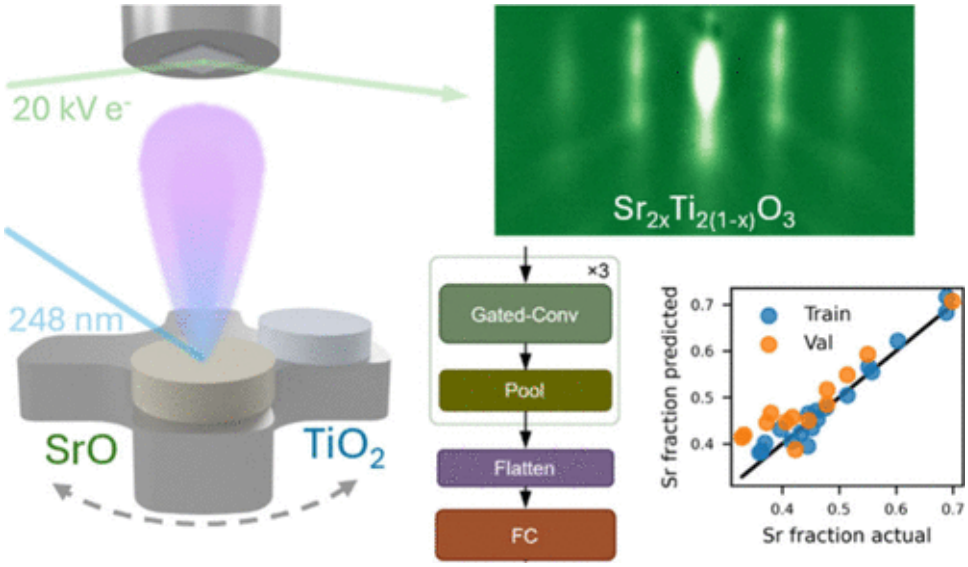

Figure 2. Overview of the stoichiometry estimation process for $Sr_{2x}Ti_{2(1-x)}O_3$ films based on RHEED images.

Nanomaterials Discovery Through AI-Enhanced Characterization:

(Faster, Cheaper & Better?)

Alfred Yan[1], Roberto dos Reis[1,2], & Vinayak P. Dravid[1,2,3]

[1]Department of Materials Science and Engineering, [2]The NU*ANCE* Center, [3]International Institute of Nanotechnology; Northwestern University, Evanston, Illinois 60208, **USA**

Materials characterization has long been recognized as the critical bottleneck in accelerating materials discovery and development, especially for diversity of crystalline materials. Traditional approaches to crystallographic analysis, compositional mapping, and electronic structure determination rely on sequential, time-consuming techniques that cannot keep pace with modern synthesis methods. This fundamental mismatch between synthesis and characterization throughput has severely limited our ability to navigate vast materials design spaces efficiently.

This challenge is particularly evident in "megalibraries", which represent massively parallel deposition of nanoparticles with varied compositions prepared via scanning-probe tip-based synthesis [1,2]. Here, the challenge is to "ration" or optimize the number of electrons that need to be utilized *spatially* to address characterization needs of millions of nanoparticles. Clearly, addressing this challenge requires innovative approaches that leverage automation and machine learning coupled to innate rapid analytical methods [3–5].

Among limited approaches, diffraction has the potential to be that "*one-stop-shop*" to elucidate the crystal structure of a nanoparticle down to its relevant details, such as point- and space-group symmetry. In this context, Transmission Kikuchi Diffraction (TKD), implemented within scanning electron microscopy (SEM), offers an ideal starting point by providing crystallographic information at significantly higher spatial resolution than conventional EBSD [6]. Our approach transforms TKD from a specialized technique into a potentially high-throughput analytical tool through automated data acquisition and machine learning-based pattern interpretation. This is enabled by neural networks trained on comprehensive dynamical simulations to identify subtle structural features that would remain hidden using conventional sequential techniques. A schematic depicting the data acquisition and training workflow is shown in Figure 1. This approach has the potential for rapid space group analysis even without prior knowledge of the sample composition.

Further, through multimodal data integration, we can simultaneously process TKD patterns with Energy Dispersive X-ray spectroscopy signals creating a unified characterization platform that reveals correlations between crystal structure and elemental distribution. This multimodal framework extends beyond SEM to transmission electron microscopy, where we've pioneered nD-STEM methodologies that capture diffraction, imaging, and spectroscopic dimensions concurrently, dramatically accelerating materials analysis while revealing previously inaccessible structure-property relationships. By integrating these automated characterization methods with predictive modeling, we establish closed-loop experimental workflows that enhance discovery efficiency across diverse application domains including energy, catalysis, and sensing.

The presentation will emphasize unprecedented opportunities with the convergence of machine learning and multimodal electron microscopy for accelerated materials exploration.

References/Citations:

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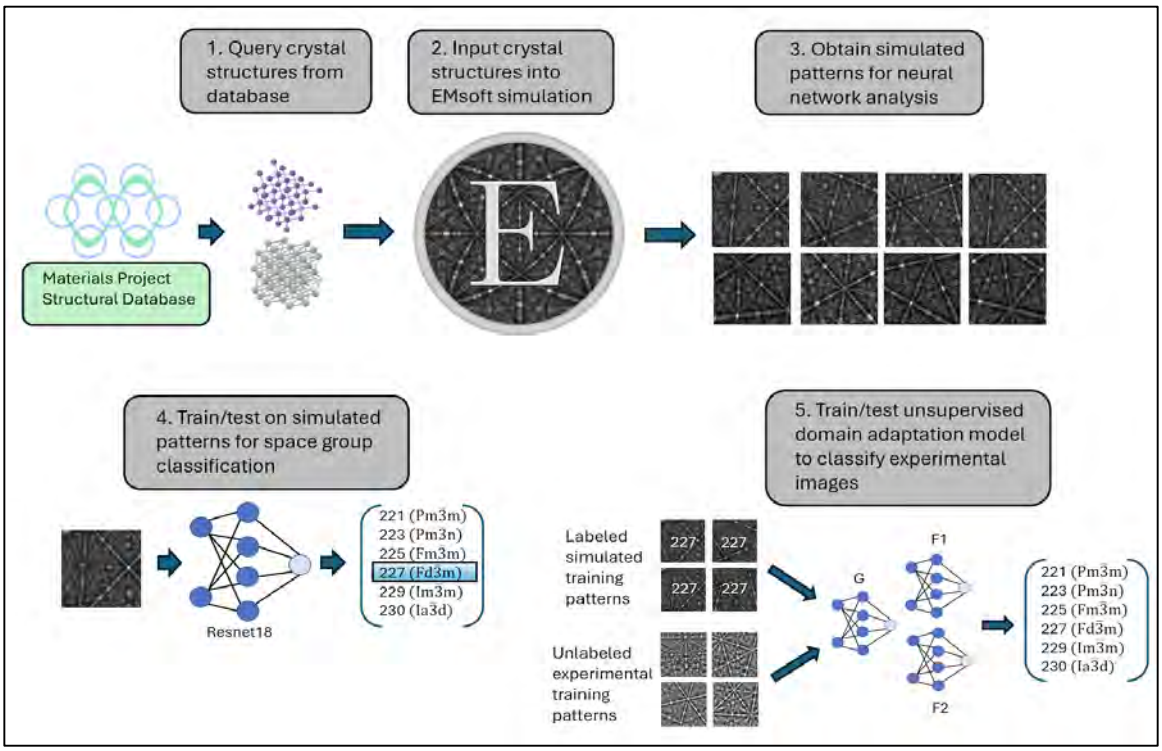

Figure 1: Schematic of workflow for developing neural network models to analyze the space group from a Kikuchi pattern.

Advanced Materials Meet Causal Learning

Ayana Ghosh[1]

[1.] Computational Sciences and Engineering Division, Oak Ridge National Laboratory, Oak Ridge, TN, USA

For decades, progress in materials science has come from a powerful partnership between theory and experiment. Theoretical models such as band theory help us understand how materials behave at a fundamental level, while experimental tools such as X-ray diffraction and electron microscopy allow us to observe and measure those behaviors in the real world. This collaboration has led to incredible advances, from semiconductors in computers to high-performance alloys in aerospace.

Today, a transformative frontier is taking shape — one that bridges materials science with modern data-driven technologies. The ongoing advancements in the landscape of artificial intelligence (AI) that have revolutionized fields of image recognition, natural language processing, and many others are now being used to accelerate the discovery, design of novel materials, process optimization. These methods help us analyze large datasets, identify hidden patterns, and make predictions about how a material will perform based on its structure or composition. The growing integration is helping to build a more efficient, predictive, and exploratory approach to materials research—one that could dramatically speed up innovation across energy, electronics, medicine, and beyond.

While these methods have made remarkable strides in detecting patterns in complex data, their reliance on statistical relationships often falls short in uncovering the fundamental physics or chemistry that govern material properties. These models excel at finding correlations but struggle to provide insight into the underlying mechanisms, limiting our ability to understand *why* and *how* material parameters influence specific properties. A significant challenge is the lack of explainability and interpretability in most ML models, including large language models (LLMs), which are designed to predict rather than finding scientific reasoning behind the predictions. This limitation underscores [1] the need for bringing in methods that not only identify patterns but also explain the underlying processes that govern material properties.

In our studies, we have been investigating, implementing and developing causal ML approaches for advanced functional materials that allow us to identify underlying cause-effect relations that govern physical properties of interest, test whether a hypothesized model is correct and quantify how changes in control variables affect outcomes. While verifying causal relationships can be challenging—often requiring validation from theory and simulations—suitable proxies can be designed to represent various scenarios and help establish robust structure-property relationships.

We have focused on perovskite oxides, known for their unique electronic, magnetic, and structural properties, essential for advanced electronic and energy devices. Our goal is to resolve key challenges that have so far eluded traditional simulations, experiments, and standard ML/DL methods, making these materials viable for practical applications. The origin of cation ordering [2] in these materials in the form of double perovskites (DPOs) has remained as a mystery for years since several factors such as cation radii and/or oxidation states, charge ordering, cooperative first order Jahn−Teller distortions of B-site cations (FOJT), A-site vacancies coupled with SOJT distortion, and tilt of BO_6/$B'O_6$ octahedra,

contribute to it. Bringing in the causal intuitions made it possible to not only pin down the necessary condition for tunable cation ordering but also establish quantifiable (previously unknown) stricture-property relationships between geometry, modes, and ordering. Comprehensive insights gained from this study shows that the trilinear coupling between structural modes representing tilt, rotation, and A-site antiferroelectric displacement in the Landau free-energy expansion, leads to formation of A-site clear layered ordering in hybrid improper ferroelectric oxides.

A follow-up study based on density functional theory (DFT) calculations [3,4] combined with finite-temperature *ab initio* molecular dynamics (AIMD) simulations, reveals that polarization switching in DPOs occurs through a two-step process, driven by out-of-phase rotation. Our results [5] indicate that switching via out-of-phase rotation leads to lower switching barriers than in-phase rotation and tilt for superlattices, complying with that found for layered DPOs. However, the challenge of pinpointing a mechanism responsible behind tuning the switching barrier is not straightforward to understand. Here, we have developed a causal reasoning workflow to uncover such physics.

In conclusion, although our causal workflows [6-8] have primarily been applied to perovskite oxides, the methodologies are intended to be broadly applicable to the materials design and optimization process. By providing a deeper understanding of atomistic mechanisms and material functionalities, these can be extended to other materials systems, enabling more efficient decision-making, prediction of properties, and the design of advanced materials across diverse applications in electronics, energy, and beyond.

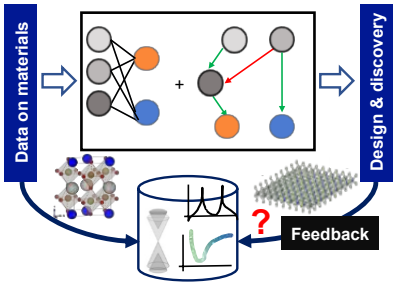

Figure 1. Schematic illustrating how causal models facilitate decision-making in materials design and discovery, aimed towards uncovering uncover fundamental physics and integration of theory into autonomous experimental workflows.

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[1] A Ghosh et al., Comput. Mater. Sci. **233** (2024), 112740.
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[9] The author acknowledges the funding from the Laboratory Directed Research and Development Program of Oak Ridge National Laboratory, managed by UT-Battelle, LLC, for the U.S. Department of Energy. ORNL is managed by UT-Battelle, LLC, for DOE under contract no. DE-AC05-00OR22725.

Autonomous Materials Exploration for Thin-film and Solid materials

Taro Hitosugi[1,2]

[1.] Department of Chemistry, The University of Tokyo, Tokyo, Japan
[2] School of Materials and Chemical Technology, Institute of Science Tokyo, Tokyo, Japan

Integrating machine learning and robotics into established research methodologies can accelerate materials science research significantly. Many studies have already demonstrated the potential of autonomous (self-driving) experiments in materials science [1, 2]. The rapid advancement of digital technologies is changing the way we conduct research.

Here, we discuss the status and prospects of data- and robot-driven materials research using autonomous experiments. We have developed an autonomous experimental system for thin-film materials. We constructed a system that automates sample handling, thin-film deposition, optimization of growth conditions, and data management. By using Bayesian optimization in conjunction with robots, our approach facilitates high-throughput experiments and generates comprehensive datasets that cover many aspects of materials (X-ray diffraction, Raman spectroscopy, scanning electron microscopy, optical transmittance measurement, electronic conductivity measurement). We tuned the hyperparameter for Bayesian optimization using the domain knowledge of chemistry; the number of trials to reach the global optimum is reduced [3, 4].

The system demonstrated the synthesis and optimization of the electrical resistance in Nb-doped TiO_2 thin films [5]. Moreover, this autonomous approach has enabled the discovery of new ionic conductors [6]. We discuss the potential impact of this technology in accelerating materials science research, particularly in solid materials.

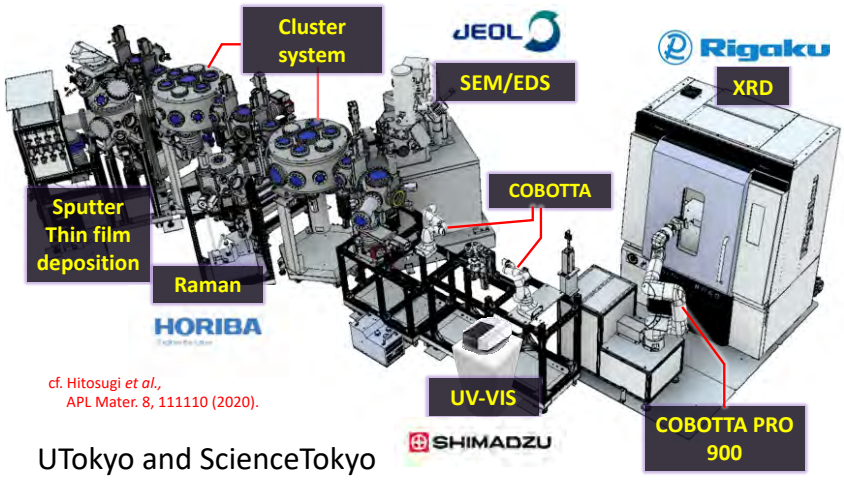

Figure 1. Autonomous experimental system for thin-film material exploration [7].

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[7] Hitosugi *et al.*, Digital Discovery, *submitted*.

Accelerating design of organic materials with autonomous AI agents

Olexandr Isayev[1],[2]

[1.]Department of Chemistry, Mellon College of Science, Carnegie Mellon University, Pittsburgh, PA 15213, USA
[2.] Department of Materials Science and Engineering, Carnegie Mellon University, Pittsburgh, PA 15213, USA

Automation of experiments in self-driving laboratories promises to revolutionize scientific research by enabling remote experimentation and improving reproducibility. However, maintaining quality control without constant human oversight remains a critical challenge. In this talk, we will provide an overview of the latest developments in machine learning and AI methods and application to the problem of molecular and material discovery at Isayev's Lab at CMU. We identify several areas where methods have the potential to accelerate chemistry research and disrupt traditional approaches.

HPLC remains one of the most critical methods for analytical and preparative separations in pharmaceutical, (bio-)chemical, and materials research. While modern HPLC instruments offer partial automation, day-to-day reliability still relies on expert monitoring of anomalies. This issue is even more pressing in academic automated laboratory settings, where instruments may be remotely shared among multiple researchers and institutions, leading to high throughput, which makes rare events more significant, especially for closed-loop experiments. Under these conditions, it is neither feasible nor cost-effective for highly trained operators to continuously supervise every single experimental run. Our study presents a generalized machine learning (ML) framework for anomaly detection that is protocol-agnostic and instrument-neutral.[1]

Recently we also proposed a novel ML-guided materials discovery platform that combines synergistic innovations in automated flow synthesis and automated machine learning (AutoML) method development. A software-controlled, continuous polymer synthesis platform enables rapid iterative experimental–computational cycles that result in the synthesis of hundreds of unique copolymer compositions within a multi-variable compositional space.[2] We are currently enhancing this platform with Reinforcement learning (RL) agents. Under the RL paradigm, an agent(s) is trained to select actions that maximize the cumulative sum of rewards, which, in the context of chemical discovery, is often consistent with a target property, structural feature, or function. RL agents can learn to suggest synthesis protocols, potential reactants, and experimental conditions by training via value-based or policy-based iterative schemes.

Here, a human-in-the-loop reinforcement learning (RLHF) approach is used to discover exceptional polyurethane elastomers that overcome pervasive stress–strain property tradeoffs.[3] Starting with a diverse training set of formulations, a coupled multi-component reward system was identified that guides RL agents toward materials with both high strength and extensibility. Through several rounds of iterative optimization combining RL predictions with human chemical intuition, we identified elastomers with more than double the average toughness compared to the initial training set.

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AI, Microscopes, and the Quest for Better Materials

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Materials shape the world around us, and the discovery of new materials defines the future of multiple technological frontiers. Breakthroughs in photovoltaics and energy storage are reshaping the global energy landscape, while extreme environments in hypersonics, rockets, and nuclear applications demand materials with unprecedented resilience. We are now at a pivotal moment where unprecedented investments in machine learning (ML) and artificial intelligence (AI) are poised to transform scientific discovery. Yet, among all areas of the physical sciences, materials science remains one of the most challenging for AI. The past two decades have demonstrated that simply scaling computation or synthesis by orders of magnitude is not enough to accelerate progress. The key lies in closing the loop between theory, hypothesis generation, experiment planning, synthesis, and characterization—while continuously refining theoretical models based on experimental feedback.

In this presentation, I will illustrate how ML-driven electron and scanning probe microscopies can be leveraged to uncover structure-property relationships in complex materials, extract fundamental physical laws governing ferroelectric polarization dynamics and property evolution across combinatorial libraries, and even manipulate matter on the nanometer and atomic level. Central to this approach is the concept of probabilistic reward functions, which enable autonomous research workflows while integrating human-in-the-loop decision-making. I will demonstrate how reward-based automated characterization can be used to close the materials discovery loop, orchestrate diverse characterization tools across shared chemical spaces, and co-navigate costly experiments and epistemic uncertainty-aware theoretical models. The special case here is operationalized materials and physics discovery in combinatorial libraries, where ML-enabled scanning probe microscope autonomously performs topography and spectroscopy tuning and combinatorial space exploration. I further discuss strategies to extend these strategies towards electron microscopy bypassing the sample preparation bottleneck. Looking ahead, this work lays the foundation for the automated lab of the future, where human intuition and AI-driven autonomy work in synergy to drive materials discovery at an unprecedented scale.

Sergei Kalinin is a Weston Fulton chair professor at the University of Tennessee, Knoxville. In 2022 – 2023, he has been a principal scientist at Amazon special projects (moon shot factory). Before then, he has spent 20 years at Oak Ridge National Laboratory where he was corporate fellow and group leader at the Center for Nanophase Materials Sciences. He received his MS degree from Moscow State University in 1998 and Ph.D. from the University of Pennsylvania (with Dawn Bonnell) in 2002. His research focuses on the applications of machine learning and artificial intelligence methods in materials synthesis, discovery, and optimization, automated experiment and autonomous imaging and characterization workflows in scanning transmission electron microscopy and scanning probes for applications including physics discovery, atomic fabrication, as well as mesoscopic studies of electrochemical, ferroelectric, and transport phenomena via scanning probe microscopy. When at ORNL, he led several major programs integrating the ML and physical sciences and instrumentation, including the Institute for Functional Imaging of Materials (IFIM 2014-2019), the first program in DOE

integrating ML and physical sciences, and the microscopy effort in INTERSECT program that realized first ML-controlled scanning probe and electron microscopes. At UTK MSE, he participated in building one of the first efforts in the country on ML-driven materials exploration. At UTK, his team has now realized fully AI-controlled SPM and STEM systems and co-orchestration workflows between multiple characterization tools for scientific discovery. He has also taught multiple courses on the ML for materials science and microscopy including Bayesian optimization methods. Sergei has co-authored >650 publications, with a total citation of ~58,000 and an h-index of ~118. He is a fellow of NAI, Academia Europaea, AAAS, RSC, AAIA, MRS, APS, IoP, IEEE, Foresight Institute, and AVS; a recipient of the Adler Lectureship (APS 2025), Duncumb Award (MSA 2024), Medard Welch Award (AVS 2023), Orton Lectureship (ACerS 2023), Feynmann Prize of Foresight Institute (2022), Blavatnik Award for Physical Sciences (2018), RMS medal for Scanning Probe Microscopy (2015), Presidential Early Career Award for Scientists and Engineers (PECASE) (2009); Burton medal of Microscopy Society of America (2010); 5 R&D100 Awards (2008, 2010, 2016, 2018, and 2023); and a number of other distinctions. As part of his professional services, he organized many professional conferences and workshops at MRS, APS and AVS; for 15 years organized workshop series on PFM, and served/s on multiple Editorial Boards including NPJ Comp. Mat., J. Appl. Phys, and Appl. Phys Lett.

AI for Accelerated Materials Discovery at Intel Labs

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Machine Learning (ML) methods that can process large amounts of heterogenous data have tremendous potential to accelerate the end-to-end discovery, synthesis, and characterization of novel materials to address global-scale challenges like clean energy, sustainable semiconductor manufacturing and drug discovery. In this talk, I will present an overview of Intel Labs' research efforts and community engagement efforts on ML for materials discovery along with technical deep dives focusing on two ambitious goals:

1. Machine Learning Interatomic Potentials (MLIPs): Accelerating scientific simulation by >100x using geometric deep learning and software tools to enable large-scale deployment of machine learning potentials for real-world simulations. Through the Open MatSci ML Toolkit [1], Intel Labs makes the training and deployment of MLIPs accessible by connecting relevant data sources with modern ML models and scalable deep learning training capabilities. In addition to the Open MatSci ML Toolkit, we have enabled the acceleration of equivariant deep learning methods through EquiTriton [2], an open-source implementation of Triton-based spherical harmonic kernels. The acceleration of equivariant models by EquiTriton has enabled us to train models at higher levels of expressivity of spherical harmonics kernels and study their relative importances when modeling materials properties.

2. Materials Science Language Models: Leveraging Large Language Models (LLMs) as scientific assistants to automate scientific tasks for materials discovery. While modern LLMs have made great progress in solving language-based tasks for a variety of fields, they still exhibit lack of understanding of the materials science domain. We have proposed some new methods to alleviate this gap, including new benchmarks (MatSciNLP [3]), multi-round instruction fine-tuning for the first billion-scale LLM for materials science (HoneyBee [4]), as well as a tool-augmented LLM that markedly improves the capabilities of diverse language models to perform materials science language tasks (HoneyComb [5]). Concurrently, we continue to showcase gaps and limitations of language models, such as property prediction dependent on geometry (MatText [6]), that require further research to enable important capabilities.

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Practically Speaking: The Engines that Automate

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Automation has always been with us since the dawn of invention. Groups or individuals, typically guided by necessity, adapted to their surroundings by creating new methods to assist their daily lives. Initially, these creations were often simple and many times crude, but they sparked new thoughts and pathways that inspired others. Automation is just one facet of invention, but it is often closer to mature development of inventive thought. It occurs when inventions are streamlined to a point where human intervention is often not necessary. For this reason, automation is typically considered for repetitive tasks that do not require creativity or innovation. In reality, the implementation of automation is more often a cost savings function. Can a process be designed that has reliability, speed, and accuracy that could effectively replace human interaction? In the sciences, which inherently require creativity, there is often considerable pushback; "A computer could never do my job!"

Automation has come for scanning transmission electron microscopy, and we must embrace it without a) alienating the field, b) losing sight of fundamental science principles, and c) developing black box technologies which can be applied without concern for data accuracy. In truth, through innovation we have been marching towards automation in the development of new tools and advanced microscopes. For instance, linescans that were initially collected step by step were replaced by automated rastering/scanning of the beam, and automated diffraction pattern collection was displaced by 4D STEM. In a certain light, these could be seen as advances that were just small steps towards full automation. These periodic advancements still require human interaction, and thus they were not truly considered automaton. There was still a "human in the loop", but at what point is that no longer a necessity? For newer, more advanced microscopes, this is becoming a reality, but what about the thousands of older microscopes that are still maintained and operated by humans for everyday analysis?

Our research goals are twofold: First, we aim to develop automated tools that still require human interaction, and second, we are working with platforms that are widespread and not confined to a single microscope manufacturer. There will always be an inherent distrust of automation from prior generations, and therefore creating interactive modalities that keep scientists engaged is important for development of next generation, fully automated tools. Additionally, development of these tools in more open source and accessible platforms provides a sandbox to encourage widespread development. While our work spans both machine learning integration and stage control, our discussion will focus on the later, as we feel it is crucial to have a platform for anyone to build upon. Understanding stage motion and broader microscope control is tantamount to being able to automate any experiment.

We will discuss stage movements for automated montaging[1-4] as well as performing automated oblique (e.g., combinations of alpha and beta tilts) tilt series[3] all through Gatan Microscopy Suite (GMS). The power of this approach is that it allows for full control of the sample in a standard double tilt holder (Figure 1). Combining multimodal observations (ADF/BF and EDS) across physical interfaces and determination of crystallographic phases on the order of nanometers makes this an extremely useful facet of the materials science toolbox. To optimize the use of multimodal approaches it is necessary to utilize machine learning techniques such as few shot analysis[5] coupled with integrated control of microscope platforms (e.g., GMS)[1]. If we cannot collect big data through stage automation, then AI tools are not practical.

Automation of scanning transmission electron microscopy (STEM) is necessary in assisting the development of next generation materials characterization. This will be important for a variety of materials including those under extreme environments. Materials that experience high temperatures and irradiation can undergo drastic transformations over the lifetime of the intended components. We will illustrate how studying void analysis of materials in corrosive environments (Figure 2) and under irradiation can benefit immensely from computed tilting and spectroscopy. The ability to quickly and accurately predict how materials properties can change in response to these stimuli can be extremely beneficial to safety and productivity. Most often, it is necessary to inspect these changes at the nanometer and atomic length scales to fully appreciate slight but meaningful modifications, but because techniques such as STEM are time consuming and expensive, it is imperative to automate not only data collection but analysis as well.

The future of automation in the electron microscope needs to be built upon an engine that can not only be utilized on current microscopes, but as well can be applied to future stage designs built towards enhanced stability, repeatability, and fidelity. We believe that by creating a sandbox for all microscopists to play in now will pay dividends well into the future.

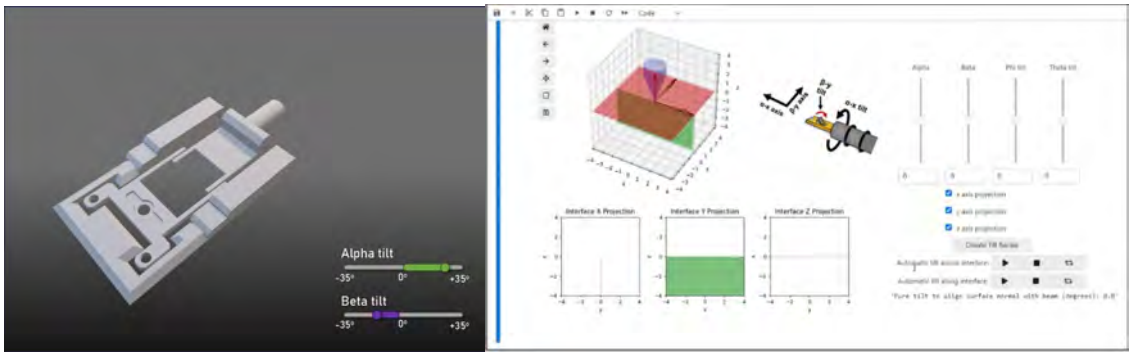
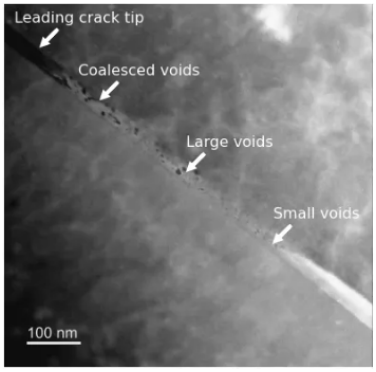


Figure 1. Cradle schematic on a JEOL ARM double tilt holder and corresponding tip tilt motion tracker.



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Automated Continuous-Flow Synthesis, Foundation Models, and Multi-Agent Systems for Accelerated Discovery of Polymers

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The integration of artificial intelligence (AI) and automated experimentation into research workflows for chemistry and materials has been heralded to reduce the tedious, labor-intensive nature of experimental research. Within the domain of polymers, there is promise that advances in these areas can significantly reduce the traditionally extended timelines needed to discover, develop, and commercialize new materials—particularly given the massive design space arising from the available structural and architectural diversity. However, there exist significant gaps between the increasing capabilities of automated synthesis platforms and AI systems and their practical implementation within experimental research workflows for polymer chemistry. To address these gaps and strive to realize an intelligent platform for polymer discovery, we have organized our research into three separate thrusts: 1) automated continuous-flow synthesis of polymers, 2) development of new foundation models for polymeric materials, and 3) integration of foundation models within multi-agent systems to facilitate human-AI interaction.

Continuous-flow synthesis is an excellent platform for facilitating automated experimentation within polymer chemistry. Controlling the polymer composition and architecture through a combination of reactor design and adjusting experimental conditions in real-time, offer capabilities to prepare highly tailored materials that would be difficult to prepare using batch reactors. Continuous-flow processes, however, can be complicated by extended residence times as a result of the higher dilution typically needed. This is problematic for automated experimentation as it can result in smaller throughput and increased potential for forming clogs. These issues can be mitigated by using highly active polymerization catalysts, which can reduce the residence times to milliseconds while maintaining a high degree of control over the molecular weight distribution and end group fidelity (Fig. 1A) [1,2]. Moreover, the combination of performant catalysts with sophisticated automation software facilitates the implementation and control of complex reactor designs—allowing multiple reactors to be run in parallel to scan polymer composition space and extract kinetic data on different catalyst systems.

The use of automated continuous-flow platforms necessitates integration of predictive models for polymer properties within the workflow, allowing a reduction in the overall experiment load and focusing on promising regions of composition space. Most models, however, are limited to a narrow scope of polymer types and only a few are generalizable to multiple prediction tasks. To accommodate the development of an improved foundation for polymer property prediction, we developed new methods of polymer representation using a domain-specific language, Chemical Markdown Language (CMDL)—allowing for accurate representation of the diversity of polymer architectures (Fig. 1B) [3]. Evaluation of this approach for catalyst and polymer design showed immense promise for the use of this representation system with transformer models. Further development of the representation system allowed for greater compatibility with existing polymer datasets, enabling the development of a new foundation model of polymeric materials [4]. This new 289 M parameter model demonstrated state-of-

the-art or near state-of-the-art performance on a variety of property prediction benchmark tasks.

Facilitating human–AI interactions is the key component for integration of AI systems within experimental workflows. Currently, the use of agentic systems powered by large language models (LLMs) provide immense capabilities in translating tasks defined by users using natural language into actionable plans for materials design and experiment execution. However, in highly technical domains, such as chemistry and materials, LLMs frequently suffer from a lack of depth of knowledge as well as restricted ability to accurately analyze chemical structures. To correct for this, we have developed methodology to facilitate structure and characterization data focused retrieval-augmented generation (RAG), allowing the LLM to receive relevant information for complex research tasks. Embeddings from chemistry and polymer foundation models were leveraged to create structure-focused vector databases, allowing facile structural similarity queries [5]. For characterization data, structure and image embeddings were leveraged to create a hybrid vector database collection capable of handling retrieval of either similar polymer structures or characterization data. These capabilities were integrated and benchmarked within an agentic workflow, demonstrating both their utility and the ability to leverage open-source LLMs to achieve comparable or better performance relative to closed-source models [6].

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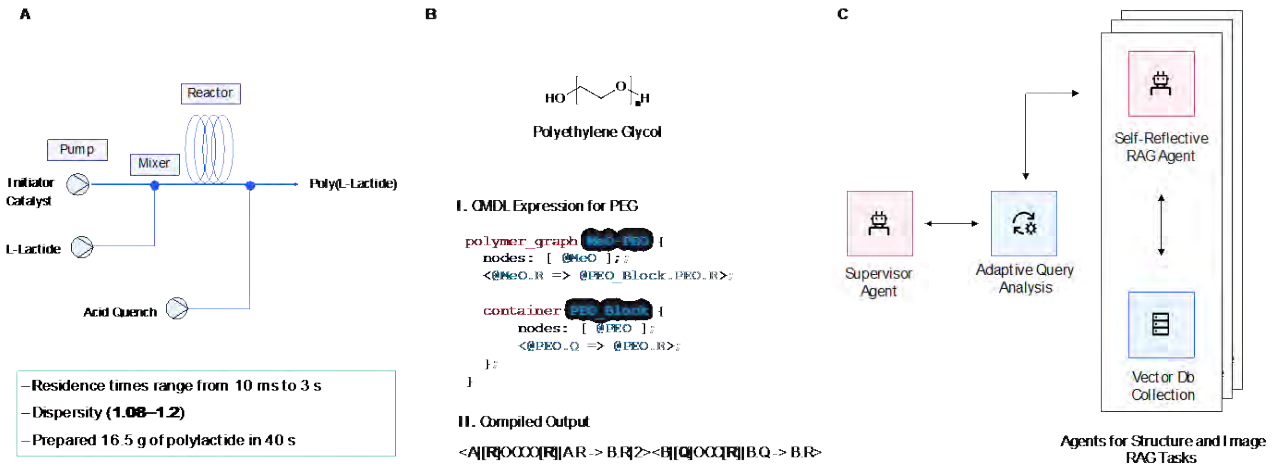

Figure 1. A) Schematic of a continuous-flow reactor used to synthesize L-lactide homopolymers. B) Example of a polymer graph expression written in CMDL (**I**) and the compiled output as serialized graph used within fine-tuning tasks for transformer models (**II**). C) Overview schema on agentic workflow utilizing semantic structural and image capabilities for RAG tasks.

Towards Machine-Learning Enabled Automated Analysis of Heterogeneous Nanomaterials

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Automated and autonomous characterization experiments are playing increasingly important roles in workflows for the discovery, design, and synthesis of materials. Recent work has illustrated successful robotic synthesis of nanomaterial populations [1, 2]. While the synthesized material is typically optimized for bulk performance and properties, individual particles are expected to exhibit heterogeneity. Given the importance that local, atomic-scale features play on nanomaterial properties, methods such as transmission electron microscopy (TEM) are needed to understand the relationship between structure and function in individual particles. Development of high throughput and automated TEM data taking and analysis will be critical to incorporating this technique into autonomous synthesis and characterization workflows.

Deep learning approaches to interpret scientific data are the subject of intense recent study. Specifically, recent work illustrated the successful use of convolutional neural networks (CNNs) as part of high throughput image analysis pipelines [3, 4]. In these examples, CNNs are used reduce input high resolution TEM imagine data to distributions of size and shape. This approach has the potential to relate the size and shape statistics of a nanoparticle population to bulk properties and synthetic protocols. Previous work has illustrated that CNNs require special consideration to avoid bias when implemented for TEM data analysis, and may not generalize well under differing experimental conditions [5, 6]. For example, data curation, CNN architecture, and performance metric choice can all play a role in the accuracy of CNN predictions on EM data. Customizing CNNs for scientific data can therefore be challenging, especially when dataset sizes are small.

Here, we present a systematic study of application of CNNs to EM data. We consider how architecture influences image classification and segmentation, and illustrate the role that data curation can play on the tradeoff between flexibility and accuracy of CNNs. We then show application of these methods to a large-scale synthesis study of Cobalt oxide nanoparticles[7]. By varying experimental parameters during synthesis, we produce cubic-shaped cobalt oxide nanoparticles with varying sizes, degrees of corner truncation, and face convexity. We illustrate successful strategies for application of CNNs to the hundreds of thousands of nanoparticles imaged in this study, and discuss error metrics and interpretation of the CNN results. The resulting statistical distributions of the nanoparticle enable us to understand the role of synthetic parameters on nanoparticle structure and shape, which ultimately will influence the catalytic behavior of these particles. These results provide intuition as to how neural network workflow design choices affect TEM image analysis, and provide guidance for researchers using CNNs for analysis of scientific images. Ultimately, this work is an example of large-scale EM analysis of hundreds of thousands or nanoparticles synthesized under a variety of conditions enabled by machine learning, and is an important step towards incorporating high throughput EM analysis into automated and autonomous nanomaterial synthesis workflows.

In particular, the degree of sample heterogeneity plays an important role: to obtain adequate statistical sampling of images of nanomaterials, the number of observations needed varies depending on the variety of features present. This is captured by the concept of 'representativeness', a property that describes whether a subset of data that captures the properties of the entire dataset. Even for seemingly self-similar populations of nanomaterials, large sample sizes are needed to observe subtle trends in size and shape statistics [7].

This work illustrates implementation of machine learning approaches for automated analysis of large nanomaterial datasets, with an emphasis on statistical sampling, proper curation of training data, and strategies to avoid bias in analysis. Ultimately, this work highlights important considerations for automated data analysis when sample or experimental conditions may be changing or unknown [8].

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Real-time Autonomous Combinatorial Experimentation – Nimble and Agile Approaches to Self-driving Laboratories

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Our philosophy for autonomous materials science is to build on existing capabilities of experimental instruments and bootstrap self-navigating systems in nimble and agile ways. Many materials synthesis and characterization tools around us are already sophisticated and automated. Automated is not autonomous, but automation is an important prerequisite for making autonomous systems. For example, by externally controlling beamline end-station instruments, we have previously demonstrated autonomous synchrotron diffraction [1] and neutron diffraction [2] (Fig. 1). In both cases, we were successful in reducing the overall number of experimental iterations and/or measurements by significant margins. We have also developed an inexpensive educational tool, which performs simple autonomous chemistry experiments [3].

The combinatorial library platform is conducive to active learning-driven autonomous experimentation. The array format with which materials samples of different compositions are laid out on combinatorial libraries facilitates sequential exploration of samples in a straightforward manner. Ability to synthesize and characterize arrays of materials sequentially one after another and in closed-loops can greatly enhance the efficacy of autonomous experimentation. Sometimes the exercise of self-driving combinatorial experimentation can be focused just on the characterization aspect: fabrication of some libraries such as thin-film composition spreads can be carried out quickly and without difficulties, but it is the quantitative evaluation of physical properties of interest which can be time and effort consuming for each individual sample. In such instances, it is effective to let the Gaussian processes dictate the sequence of measurements on the library. For example, in a previous experiment, we were able to find the composition of a phase change memory material with the largest bandgap contrast between amorphous and crystalline phases on a pre-fabricated ternary thin-film composition spread after measuring only a fraction of the entire composition range covered on the spread [1].

In another example, we have demonstrated real-time self-driving continuous cyclical interaction of experiments and computational predictions for materials exploration. In particular, we have performed rapid mapping of a temperature-composition phase diagram, a fundamental task for the search and discovery of new materials. Thermal processing and experimental determination of compositional phase boundaries in thin films are autonomously interspersed with real-time updating of the phase diagram prediction through the minimization of Gibbs free energies. The workflow was able to accurately determine the eutectic phase diagram of the Sn-Bi binary thin-film system on the fly from a self-guided campaign covering just a small fraction of the entire composition - temperature phase space, translating to a 6-fold reduction in the number of necessary experiments. This study demonstrated for the first time the possibility of real-time, autonomous, and iterative interactions of experiments and theory carried out without any human intervention [4].

We have also recently demonstrated autonomous control of unit cell-level growth of functional thin films implemented in combinatorial pulsed laser deposition. Dynamic analysis of reflection high-energy electron diffraction images is used to autonomously navigate multi-dimensional deposition parameter

space in order to rapidly identify the optimum set of growth parameters for fabricating the targeted materials phase. I will also discuss other autonomous experimentation projects we are carrying out including metal additive manufacturing. This work is performed in collaboration with M. Lippmaa, H. Liang, A. G. Kusne, A. McDannald, and J.-C. Zhao. This work is funded by NIST, ONR, SRC, and Department of Energy, Office of Science, Office of Basic Energy Sciences Energy Frontier Research Centers program under Award Number DE-SC0021118.

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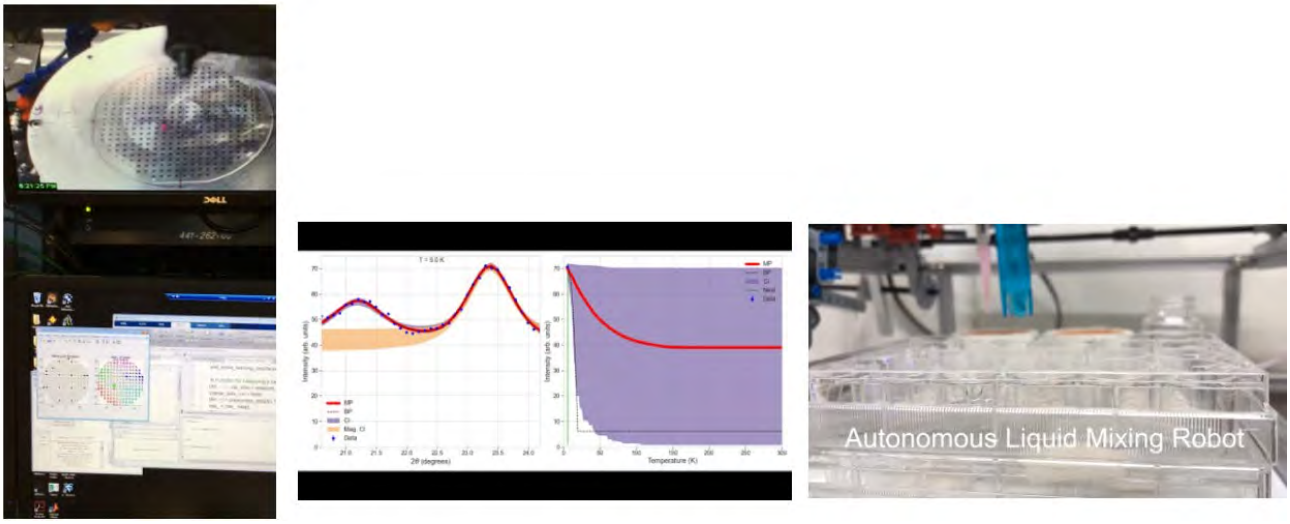

Figure 1. (left) CAMEO: Closed-loop autonomous materials exploration and optimization [1]; (middle) Autonomous neutron diffraction explorer (ANDiE) [2]; (right) LEGOLAS: LEGO based Low-cost Autonomous System for Education [3]

Integrating Automation Tactics and Machine Learning to Direct Nanoscale and Atomic Scale Transformation Experiments

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Recent advances in autonomous experimentation are rapidly transforming the capabilities of direct-write nanofabrication, enabling precise manipulation of matter from the atomic to nanoscale with an unprecedented degree of control. At the forefront of this transformation is the integration of automation and machine learning (ML) with scanning transmission electron microscopy (STEM)-based direct-write approaches, which together are redefining the way materials can be engineered and modified at the smallest length scales. In this work, we demonstrate a comprehensive methodology that couples real-time feedback-controlled STEM imaging with ML-based image analysis to guide atomic-scale transformations with high spatial and temporal resolution. Building upon prior developments in atomic defect engineering in two-dimensional (2D) materials,[1–4] and leveraging high-throughput, deep learning-enhanced STEM workflows, we deploy a closed-loop experimental platform.[5] This system employs deep convolutional neural networks (DCNNs) to classify evolving atomic configurations during dynamic electron-beam-induced transformations, enabling on-the-fly adjustment of beam irradiation parameters.[4,6] Such adaptive control allows for precision operations, including layer-by-layer material removal, the formation of sub-nanometer pores, and the stabilization of transient metastable phases within $Ti_3C_2T_x$ MXenes.[7]

In addition to beam-driven modifications in vacuum, we extend the concept of direct-write nanofabrication to hydrated environments using liquid-phase electron beam lithography.[8] This approach allows for chemical and structural manipulation from liquid precursors, expanding the scope of material systems and phenomena accessible to in situ experimentation. Our in situ observations under varying thermal and electron beam irradiation conditions reveal key mechanisms governing atomic mobility and beam-matter interactions, aligning with previously reported behaviors in thermally activated edge reconstructions in 2D transition metal dichalcogenides (TMDs). By combining real-time analysis, automated feedback control, and multimodal environmental capabilities, this work establishes a robust platform for autonomous, atomically precise fabrication. The resulting system serves as a powerful toolset for materials discovery, defect engineering, and dynamic structural modulation—offering new opportunities for scientific exploration in both fundamental and applied research contexts.

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Advancing Ionomer-Based Water Electrolysis: Integrated Characterization, Foundation Models for Segmentation, and Autonomous Optimization

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Low-temperature electrolysis using single-ion-conducting polymers (ionomers) offers strong prospects [1] for dynamic, modular operation and enhanced efficiency through differential pressure strategies. Among hydrogen production technologies, proton exchange membrane water electrolyzers (PEMWEs) stand out due to their enhanced operation, high conversion efficiency, high purity hydrogen (>99.99%), and rapid dynamic response. However, key challenges persist, hindering broader commercialization, notably the reliance on catalyst materials, thick durable separators/membranes, and limited fundamental understanding of complex interfacial phenomena at the ionomer/catalyst interface, particularly related to the oxygen evolution reaction (OER). Fundamental research into iridium oxide [2] is essential to reduce catalyst loading while maintaining performance. Iridium oxide catalysts represent 26–47% of the total system cost, primarily due to iridium scarcity. Commercial iridium oxide catalysts typically exist as amorphous IrOx or crystalline IrO2, each exhibiting trade-offs between catalytic activity and stability.

Our current work at CIWE (Center for Ionomer-based Water Electrolysis) [3] directly addresses these challenges by combining detailed experimental characterization with advanced computational modeling and artificial intelligence (AI) techniques. We have explored novel physical and electrochemical analyses of new iridium oxide catalysts (amorphous IrOx and crystalline IrO2 from Ishifuku Metal), employing scanning electron microscopy using focused ion beam (FIB-SEM), transmission electron microscopy (TEM), Raman spectroscopy, X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), and more. These comprehensive characterizations reveal distinctive structural properties; notably, crystalline IrO2 displays needle-like morphologies with significantly greater specific surface area (~110 $m^2\ g^{-1}$) compared to amorphous IrOx (~50 $m^2\ g^{-1}$) [2]. Electrochemical performance evaluations across varied catalyst loadings demonstrate important differences: amorphous IrOx exhibits distinct redox peaks indicative of surface activation processes occurring after extensive cycling, whereas crystalline IrO2 primarily shows double-layer capacitance behavior at lower voltages. According to Zenyuk's laboratory latest results [2], both catalysts demonstrate competitive polarization performances, with voltages of 1.92 V (amorphous) and 1.94 V (crystalline) at 5 A cm^{-2} for a loading of 0.85 mg cm^{-2}, accompanied by low Tafel slopes (43.8 mV dec^{-1} for amorphous IrOx, 53.7 mV dec^{-1} for crystalline IrO2).

Key scientific questions currently pursued by our team involve the application of computer vision techniques and machine learning (ML) tools. Specifically, we investigate whether AI/ML algorithms can improve materials characterization through accurate segmentation methods and morphological

quantification, addressing challenges such as measuring porosity, which is complicated by interference from adjacent layers, precise determination of layer thickness, assessing homogeneity, and detecting defects such as long cracks and bends. Additionally, qualitative analysis using advanced 3D visualization tools is being evaluated to determine if such immersive visualization methods can accelerate algorithm development and provide deeper insights into material structure and behaviors beyond traditional performance metrics like 2D plots of small sections, Dice coefficients and F1-scores.

We have investigated the use of the Segment Anything Model (SAM), a powerful foundation model for computer vision, to perform zero-shot segmentation in FIB-SEM and cryo-TEM imaging. Zero-shot methods like SAM are valuable due to their ability to generalize without requiring extensive labeled datasets. To enhance the performance and applicability of SAM in segmenting complex EM datasets, Gaussian Process (GP) optimization has been considered for efficient hyperparameter tuning. GP allows a systematic scan through hyperparameter spaces, quantifies uncertainty, and optimizes model performance, ensuring robust segmentation outcomes suitable for high-resolution structural analyses, and control for realization of the digital twin. Additionally, Gaussian mixture models (GMM) are being explored alongside SAM for comparative evaluation, and it will provide insights into alternative statistical approaches for segmentation and morphological characterization.

Our integrated approach also leverages domain-specific analytics and immersive visualization technologies, such as ASCRIBE Virtual Reality platform [4] powered by Unreal Engine and Meta Quest headsets, to intuitively monitor and interpret complex multimodal datasets in real-time. AI-driven autonomous decision-making frameworks using Gaussian Processes and zero-shot computer vision based on foundation models are likely to enhance experimental agility and adaptive optimization.

Despite these advancements, significant hurdles remain, including lack of data annotation for accuracy validation, complex data integration, noise suppression, parallelization of intensive tasks, such as parameter optimization, etc. Continued research [5] will focus on refining analytical and computational methodologies, reducing catalyst usage, and broadening the deployment of autonomous systems. Addressing these needs will further fundamental insights into ionomer-based interfaces and accelerate the transition toward more efficient, durable, and economically viable electrolyzer technologies.

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Challenges in self-driving laboratories: curiosity-based explorations, and the need for human-in-the-loop workflows for maximizing robustness and prior knowledge

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Self-driving laboratories promise to tremendously improve the rate of progress of the discovery and optimization of molecules and materials [1]. This is both from the increase in automation of the process, enabling many more experiments to be performed, as well as the integration of machine learning methods, such as Bayesian optimization algorithms (BO) that can rapidly find promising regions of the parameter space. This combination has led to an explosion of interest within autonomous and self-driving laboratories, and a wealth of literature reviews are now available on the topic [2,3].
At the Center for Nanophase Materials Sciences (CNMS) at Oak Ridge National Laboratory, we have been creating autonomous laboratories that automate scanning and electron probe microscopes as well as synthesis tools such as pulsed laser deposition (PLD) systems. In this talk, we will explore several challenges related to algorithms for autonomous synthesis and characterization and explain why a human-in-the-loop workflow, with human and AI collaboration, is required to maximize efficiency and robustness.

Specifically, we will begin with an exploration of automated synthesis and the need for human oversight, which can be provided through gated active learning approaches. In our autonomous pulsed laser deposition platform, it is possible to grow 2D materials via PLD and measure their optical properties via laser reflectance and gauge the quality of the films via in-situ Raman spectroscopy. This was used as the signal to optimize the PLD conditions (energy of the two lasers, the temperature of the substrate, and the pressure of the background gas in the chamber). Using traditional Bayesian optimization, it was shown that it was possible to find an optimal synthesis recipe in under 100 synthesis experiments [4]. However, this was beset by many 'failed' experiments where the growth regime was such that the film quality was poor.

Upon inspection, one of the challenges encountered was that the defined scalarizer function used for the optimization relied on the quality of the fitting of certain Raman peaks, and if the spectrum quality was poor, this would result in poor quality data fed into the BO algorithm, leading to sub-par performance. This speaks to the trouble with scalarizer functions in general which must be apriori defined: when there are unknowns in a true experiment, the quality of the output may deviate, and a method to tune the experimental trajectory on-the-fly is required. Thus, a new algorithm, termed "Dual-GP", consisting of a standard BO loop, as well as a secondary loop that includes a quality inspection that can either be achieved via a human operator, or some other method (e.g., an ML agent, some other traditional analytics based method, etc.) was proposed [5]. In the full optimization, the secondary quality assessment is used to confine the regions of the parameter space to the parts that are most promising, and to neglect the parts where the data is deemed of poor quality. This was shown to significantly improve the overall efficiency of the BO and reduce sample wastage, thereby increasing robustness in autonomous synthesis workflows.