AI/ML/Autonomous Experimentation for Thin Films Processing
Room 317 - Session AIML2-ThA

AI/ML-enhanced RHEED in Thin Film Growth

Moderators: Mr. Dorien Carpenter, University of Alabama at Birmingham, **Prof. Martin Seifrid**, North Carolina State University

3:30pm **AIML2-ThA-6 Turning RHEED into a Tool for Stoichiometry Analysis and Real-time Control of Epitaxial Film Growth Using Machine Learning**, ***Ryan Comes,*** University of Delaware **INVITED**

Progress in materials synthesis increasingly depends on combining real-time characterization with data-driven analysis and feedback. Reflection High–Energy Electron Diffraction (RHEED) offers a unique capability for monitoring thin film growth during deposition, yet its broader utility is constrained by limited dynamic range, signal saturation, and challenges in extracting quantitative information from diffraction images. To address this, we have developed a robust and extensible methodology to improve RHEED image quality, enable quantitative analysis, and support feedback-aware synthesis workflows. Weshowed that deep convolutional neural networks trained on RHEED patterns could accurately predict the cation stoichiometry of $SrTiO_3$ thin films during growth, with explainable AI tools revealing previously unrecognized correlations between diffraction streak intensity ratios and reciprocal-space spacing.[1] These results establish RHEED as a viable surrogate measurement for film stoichiometry.

To identify and track structural evolution during growth, we have also applied unsupervised machine learning approaches, including principal component analysis and k–means clustering, to time-resolved RHEED image sequences. Image drift correction and precise alignment of the specular reflection enable reliable frame-to-frame comparisons for $LaFeO_3$ and $SrIrO_3$ films synthesized by molecular beam epitaxy. These preprocessing and analysis steps establish a foundation for automated recognition of growth modes and cross-material comparisons.[2]

Our ongoing work focuses on the development of software for real time analysis of videos to provide ML-driven feedback to the human operator of the MBE system using reinforcement learning. This framework incorporates a pathway toward real-time synthesis feedback by integrating enhanced RHEED video streams with growth metadata such as fluxes, shutter states, chamber pressure, and substrate information. By combining unsupervised learning, deep neural networks, and real-time diagnostics, this work lays the groundwork for closed-loop thin-film synthesis capable of quantitative interpretation, adaptive control, and accelerated materials discovery.

[1] S.B. Harris, et al. *Nano Letters*, **25**, 5867–5874. (2025)

[2] P.T. Gemperline, et al. *JVSTA,* **43**, 032701 (2025)

4:00pm **AIML2-ThA-8 AI-Driven Recognition of RHEED Pattern Features for MBE Monitoring**, ***Vadim Gorshanov,*** DCA Instruments Oy, University of Turku, Finland; *Markku Rajala,* DCA Instruments Oy, Finland; *Milica Todorović,* University of Turku, Finland

Reflection High-Energy Electron Diffraction (RHEED) is a standard in situ characterization technique used to monitor surface crystal structure during thin film deposition methods such as Molecular Beam Epitaxy (MBE). RHEED patterns contain rich information about the surface ordering, but their interpretation is often manual or restricted to simple intensity tracking, which fails to fully capture the evolution of the surface. This work presents an approach to automating RHEED pattern analysis using semantic segmentation (pixel-wise labeling) to detect and classify diffraction features, including spots, streaks, and Kikuchi lines.

To enable such analysis, we compiled and annotated a ground-truth dataset of diverse RHEED patterns. This dataset was utilized to conduct a comparative study of modern deep learning architectures, benchmarking Convolutional Neural Networks (CNNs) and Transformer-based architectures paired with various encoder backbones. The goal of this comparison is to identify and optimize the architecture that maximizes accuracy to ensure reliable pattern monitoring. Results demonstrate that the optimized models robustly isolate diffraction features from complex backgrounds. Furthermore, the method exhibits strong generalization capabilities, successfully recognizing RHEED patterns of different material systems and imaging conditions not present in the training data. In addition, the image processing achieves frame rates suitable for real-time monitoring. The procedure enables the extraction of quantitative diffraction features from raw RHEED images and videos, paving the way for automated control in industrial RHEED workflows, as well as facilitating post-growth analysis.

4:15pm **AIML2-ThA-9 Toward LLM Driven Agentic Automation of RHEED Diagnostics in Thin Film Deposition**, ***Asraful Haque,*** *Rama Vasudevan, Christopher Rouleau, Sumner Harris,* ORNL

Real-time interpretation of reflection high-energy electron diffraction (RHEED) patterns during thin film growth remains a bottleneck for fully autonomous deposition systems. While individual machine learning tools have demonstrated promise for isolated RHEED analysis tasks, integrating them into a coherent, decision-capable framework has been challenging. Here we present an agentic automation architecture that orchestrates multiple AI/ML modules for RHEED diagnostics using large language model (LLM)-based agents as the reasoning backbone. The system integrates: (i) a YOLO (You Only Look Once) based object detection and classification pipeline for real time identification of RHEED pattern features including spot geometry, streak morphology; (ii) a structural similarity (SSIM) based algorithm for automated sample alignment and beam registration in RHEED; and (iii) an FPGA-DAC (Field Programmable Gate Array and Digital to Analog Converter) controlled electron beam deflection subsystem enabling automated rocking curve acquisition. The LLM agent layer interprets outputs from each module, coordinates decision logic across the diagnostic pipeline, and interfaces with the deposition control system to enable closed-loop feedback.

We demonstrate proof of concept autonomous operation where the agent sequences alignment, real-time pattern classification, and real-time rocking curve measurements without human intervention. This modular, agent-orchestrated approach represents a scalable path toward self-driving thin film growth platforms where natural language reasoning and computer vision together manage the complexity of in-situ diagnostics. This research was conducted at the Center for Nanophase Materials Sciences (CNMS), which is a DOE Office of Science User Facility at Oak Ridge National Laboratory.

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