NAMBE
Room Cummings Ballroom - Session NAMBE2-MoA
Advances in In Situ Characterization

Moderator: Zachary LaDuca, University of Wisconsin - Madison

3:30pm **NAMBE2-MoA-9 Principal Component Analysis of Rheed as an Indicator of Process Change During Molecular Beam Epitaxial Growth***, **Kurt Eyink**, Y. Zhang, K. Mahalingam, R. Bedford,* Air Force Research Laboratory, Materials and Manufacturing Directorate, USA

Reflection high energy electron diffraction (RHEED) is a ubiquitous sensor used in the epitaxial growth of materials. It is routinely used to determine the surface phase present during growth but has also been used to extracted many additional features such as: QD facet structures, surface roughness and strain relaxation to name a few. In addition to surface features, RHEED also possesses bulk components in the Kikuchi lines and therefore potentially linked to bulk material quality.In this presentation, we discuss Principle Component Analysis (PCA) applied to RHEED movies acquired during the growth of molecular beam epitaxy of GaAs(001). In particular we use of PCA on a series of images from a section of the growth and show the components determined in this section allow growth to be followed in subsequent images of the growth.In addition, we show these same components can be used to follow growth on a different wafer.We discuss changes which occur in RHEED due to the unique orientations every wafer has relative to the chamber/electron beam as well as the inability to exactly reproduce conditions such as focus, angle of incidence, and wafer orientation. Correction for these modifications need to be applied prior to projection onto existing PCA components however after application we demonstrate the ability to track growth under similar conditions to the first analysis. We also show that growths which deviates from this region cause an increase in residual using the projected images and show that this is a simple method to indicate when new processes are occurring.

3:45pm **NAMBE2-MoA-10 Automated Machine Learning of in-Situ RHEED Data Provides Real-Time Guidance for Materials Growth Optimization***, **Christopher Price**, J. Munro,* Atomic Data Sciences*; G. Zhou, Y. Li, C. Hinkle,* University of Notre Dame

Achieving fine control of process feedback during bottom-up synthesis is critical to achieve repeatable synthesis of materials using commercially scalable methods. Composition is controlled by adjusting synthesis parameters, but the relationships between these adjustments and the resulting sample characteristics are only established after sample growth with ex-situ characterization such as XPS, and vary between individual hardware installations. Using automated feature extraction for in-situ reflective high-energy electron (RHEED) patterns, we show that quantitatively predictive relationships can be established between in-situ and ex-situ characterization (XPS measurements) based on small sets of labeled training points within a specific material system. Non-obvious but straightforward correlations are shown to be predictive in quantitatively estimating composition on par with expert XPS analysis. The compositional estimates from RHEED data collected during growth can be delivered fast enough to make real-time adjustments to materials processing and influence the synthetic output. In this work, we outline the workflow and show that this strategy generalizes across two different materials systems without retraining of the core feature extraction models. A reduction in ex-situ experiments and quasi-real-time feedback to offer more precise synthetic control is demonstrated. These relationships provide guidance to develop hardware-specific recipe adjustments to unlock fine-tuned control of materials synthesis.

4:00pm **NAMBE2-MoA-11 On-the-Fly Analysis of RHEED Images During Deposition Using Artificial Intelligence***, **Tiffany Kaspar**,* Pacific Northwest National Lab*; J. Pope, S. Akers, H. Sprueill, A. Ter-Petrosyan, D. Hopkins,* Pacific Northwest National Laboratory

Modern synthesis methods enable the fabrication of an ever-expanding array of novel, non-equilibrium, and/or metastable materials and composites that may possess unique and desirable functionality. Thin film deposition by molecular beam epitaxy (MBE) can produce atomically precise (or nearly so) materials with a wide range of functional electronic, magnetic, ferroelectric/multiferroic, optical, and/or ion-conducting properties.We are working to employ artificial intelligence (AI)-accelerated analysis of in situ and ex situ data streams for on-the-fly feedback control of the MBE deposition process that will enable targeted synthesis of novel materials with desired structure, chemical stability, and functional properties.Here we present a machine-learning-enabled framework for analysis of reflection high energy electron diffraction (RHEED) pattern images in real time (one image per second).Our approach utilizes pre-trained neural networks for image preprocessing, statistical analysis to identify changepoints in the images over time, and network graph analysis methods to precisely identify and classify changes.We demonstrate this framework using RHEED images collected from the deposition of epitaxial oxide thin films such as anatase TiO_2 on $SrTiO_3(001)$.Advantages and disadvantages of our approach will be discussed, as well as its potential use as the basis for on-the-fly feedback control of deposition parameters.

4:15pm **NAMBE2-MoA-12 The Development of Order and Interfaces During Oxide MBE Growth: Real Time X-Ray Diffraction Measurements***, **Hawoong Hong**, D. Fong, A. Bhattacharya,* Argonne National Laboratory

While the atomic structure of interfaces in complex oxide heterostructures created by epitaxial growth has been investigated extensively, few studies have been conducted on *how* interfaces form at the initial stage of film growth. The dynamic aspects of the growth behavior can strongly influence the final interfacial atomic structure, which may lead to the emergence of interface-specific properties. Here, the structural development of La_2CuO_4 thin films grown by molecular beam epitaxy on a $LaSrAlO_4$ substrate is investigated by X-ray diffraction measurements with rapid scans over a volume of reciprocal space in addition to fixed-point measurements. The results show that the atomic structure of the interface becomes fully established after just a single unit cell growth; afterwards, changes occur only within the topmost half- to one unit cell of the growing film. However, diffraction intensity oscillations from simultaneous reflection high energy electron and X-ray measurements stabilize only after the growth of two unit cells, indicating that the growth front morphology continues to evolve until the start of the third unit cell. Our multimodal investigation provides new insights into the atomic processes of layered oxide interface formation, such information can be relevant to the engineering and optimization of functional layer structures.

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