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

AI/ML Tools in Molecular Beam Epitaxy & Flash Session

Moderators: Dr. Michael Nolan, Tyndall National Institute, University College Cork, **Ms. Campbell Sweet**, University of Missouri

11:00am **AIML2-ThM-13 AI/ML Techniques for Molecular Beam Epitaxy***, Stephanie Law,* Pennsylvania State University **INVITED**

MBE synthesis offers precise control over film thickness, composition, and crystallinity by tuning a range of growth parameters. While this tunability enables tailored synthesis, it also introduces significant complexity, resulting in multidimensional growth spaces. Identifying optimal growth parameters within these complex spaces typically involves trial-and-error experimentation, in which growers use a combination of intuition and grid search methods to guide the choice of growth parameters. An alternative way to navigate complex growth spaces is through the use of Bayesian Optimization (BO) or AI-driven hypothesis generation, which can explore parameter space more efficiently and find global optimal growth parameters. By leveraging probabilistic models such as Gaussian Process Regression (GPR), BO can statistically select the most informative experiments, balancing exploration of new growth parameter spaces with exploitation of known good parameters. AI-guided hypothesis generation can be used to read the extant literature and generate experimental campaigns. In this work, we apply BO to efficiently explore the growth space for In_2Se_3 thin films on Al_2O_3 substrates using MBE. By integrating prior experimental data and employing GPR, we employed a data-driven synthesis campaign that significantly reduced the number of required experiments. Leveraging BO resulted in an increase in the fraction of γ-In_2Se_3 from ~50% to >90% in fewer than 10 samples. To further understand the influence of individual growth parameters on model predictions, we used SHapley Additive exPlanations (SHAP), w hich indicated that the indium flux is the most important parameter in predicting the film polymorph fraction, pointing toward defects as a method of stabilizing the polymorph. We also employed AI-guided experimental design to explore growth parameter space for In(Ga)Sb quantum dots. This approach led us to a clear understanding of how QD morphology and density depends on growth parameters, and how QD photoluminescence is correlated with both. These findings highlight the potential of data-driven optimization in complex materials synthesis and further solidify the growing body of evidence supporting the use of ML for efficient thin film experimental design.

11:30am **AIML2-ThM-15 Human-in-the-loop Agentic Design of Experiments for Molecular Beam Epitaxy Synthesis of In(Ga)Sb Self-Assembled Quantum Dots in an InAs Matrix***, Molly McDonough,* Pennsylvania State University*; Priyanka Petluru, Aaron Muhowski,* Sandia National Laboratories*; Wesley Reinhart, Stephanie Law,* Pennsylvania State University

In(Ga)Sb self-assembled quantum dots (QDs) are a class of III-V nanostructures that have been studied primarily due to their broken-gap band alignment and emission wavelengths in the mid-wave (MWIR) and long-wave infrared (LWIR). The MWIR and LWIR regimes are crucial to many different technological areas including biosensing, trace gas analysis, environmental monitoring, and defense. However, options for compact, high efficiency emitters in this wavelength range are nascent. InSb and In(Ga)Sb self-assembled quantum dots (SAQDs) in an InAs matrix are a unique option for this wavelength regime due to their tunable emission wavelength by tailoring the lateral size and height of the SAQDs. Previous work on InSb SAQDs has yielded strong photoluminescence (PL) emission in the 3µm - 5µm range. For In(Ga)Sb SAQDs on InAs, emission out to 8µm has been successfully demonstrated.

Our work builds on latent knowledge by modeling the relationship between growth parameters, SAQD morphology and properties, and PL emission wavelength to create designer MWIR and LWIR emitters. We implement a human-in-the-loop agentic design of experiments process via tool-using large-language models to suggest growth conditions for maximal information gain per trial. The parameters explored here include how V/III beam equivalent pressure ratios, growth rate, deposition thickness, Ostwald ripening time, composition, and the use of bismuth surfactant influence our nanostructure morphology and emission characteristics. By this method, we achieve PL emission from In(Ga)Sb nanostructures in the 5µm to 7µm range. We observe the formation of In(Ga)Sb quantum dashes, which contrasts with existing studies on In(Ga)Sb quantum dots. The results presented here provide in-depth mapping of the synthesis-structure-property relationships in this system and more broadly demonstrate an effective and practical deployment of agentic hypothesis generation for synthesis exploration and device development.

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