We Have to Go Small to Go Big: Autonomous Multimodal Microscopy for Materials and Physics Discovery

Sergei V. Kalinin

University of Tennessee, Knoxville
E-mail address: sergei2@utk.edu

Materials discovery remains central to technological progress - from energy and quantum devices to sustainable manufacturing. Over the past two decades, theoretical workflows for high-throughput screening and predictive modeling have matured significantly, ushering in the era of "big data" and now ML in computational materials science. However, the true test of these predictions lies in experimental realization. In the last five years, experimental synthesis has undergone rapid transformation through lab robotics and scalable platforms such as combinatorial libraries. Yet, the critical bottleneck remains characterization. This is particularly the case for self-driving laboratories, where scaling demands rapid, quantitative insight into structure and function at sub-micron length scales and sub-second timescales.

In this presentation, I will describe our work in closing this loop by developing autonomous workflows for comprehensive mapping of composition–structure–property relationships. These workflows are based on the combination of bottom-up optimization workflows and top-down LLM-based reward design, heuristic identification, literature mining, and coding assistance. As a first example, I will demonstrate how scanning probe microscopy (SPM) can be quantified and fully automated for the exploration of combinatorial libraries of ferroelectric, photovoltaic, and electrochemical materials. These workflows are inherently non-myopic, involving multi-step sequences of spectroscopy optimization, adaptive experiment design, and structure–function discovery. All these capabilities are implemented through real-time feedback and decision-making loops on ML-enabled SPM, and are benchmarked against human operator baseline. We then extend these concepts to scanning transmission electron microscopy (STEM), which enables atomic-scale insights into structure, chemistry, and local functional properties. To address the sample preparation bottleneck inherent to STEM, we introduce the concept of random libraries: large and diverse sets of local environments enabling the exploration of complex, high-dimensional materials design spaces using statistical methods. To summarize, 20 years ago, the rise of big data revolutionized theory in materials science. Today, we are entering the era of "big materials", where we start the same scale-up for synthesis and characterization. Autonomous and intelligent characterization is the final step in realizing the vision of closed-loop, ML-driven materials discovery.