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From Event Detection to Physical Hypothesis Learning via Automated and Autonomous Microscopy

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Electron and scanning probe microscopies are by now the mainstay of areas ranging from fundamental condensed matter physics and materials science to chemistry and biology. In many cases, these imaging methods provide quantitative or semiquantitative information on materials structure and functionalities and allow for the exploration of broad range of responses to global (temperature, chemistry) and local (beam exposure, probe bias and pressure) stimuli. However, the connection between microscopy imaging, spectral data, and specific mechanisms of physical and chemical phenomena often requires extensive studies including multiple human-driven stages.

The recent emergence of the automated and autonomous microscopes enabled by custom- and manufacturer provided programming (e.g. Python) interfaces offers an opportunity to change this paradigm. However, the development of automated microscopy workflows requires machine learning methods that can guide and navigate exploration processes in the image, spectroscopy, or microscope parameter spaces. Classical Bayesian optimization-based strategies that now form the mainstay of the field are non-parametric, and typically do not reflect any prior physical knowledge, relying instead solely on the internal correlations within the data. This consideration strongly limits their applicability and often requires large volumes of training data, rendering practical applications intractable.

In this presentation, I will discuss strategies for automated experiment in microscopies starting with simple event detection with predefined action tables to active learning methods capable of physics-driven discovery of structure-property relationships, and learning the underpinning physical mechanisms given the list of possible hypotheses. While we present examples for Scanning Probe Microscopy, such workflows are universally applicable to other imaging techniques including (Scanning) ransmission Electron Microscopy (STEM) and mass-spectrometric chemical imaging.

As a first example of active learning of structure-property relations, we demonstrate Deep Kernel Learning of the relationship between polarization switching and non-linearities and domain structures in ferroelectric materials. In this approach, we use the prior information on domain structure within the image to guide the selection of locations for hysteresis loop or non-linearities measurements. This automated workflow discovers that on-field and off-field hysteresis loops are dominated by different underlying properties based on the largely different exploration paths and sampling points. Furthermore, an extension of DKL towards multimodal imaging is demonstrated, allowing elucidation of information channels that allows for best predictability of target physical functionality.

As a second development, we note that in many cases domain expertise allows for formulation of one or several possible hypotheses for materials behavior, coming with specific functional forms, constraints, and possible priors on parameter values. Hence, the ideal algorithm for exploring materials physics will

co-navigate experimental and theoretical spaces, exploring or optimizing the experimentally determined material's functionality while refining the hypotheses (here, functional form) of materials behavior within this parameter space. Here, we introduce an active learning approach based on co-navigation of the hypothesis and experimental space by combining the fully Bayesian structured Gaussian Processes with reinforcement learning policy refinement. This approach closely resembles the classical human-driven physical discovery, when several alternative hypotheses realized via models with adjustable parameters are tested during the experiment.

As a first example of hypothesis learning, we demonstrated this method for exploring concentration-induced phase transitions in the combinatorial libraries of Sm-doped $BiFeO_3$ using Piezoresponse Force Microscopy. Here, the compositions along the phase diagram are explored in automated fashion aiming to discover the phase transition mechanism. Looking forward, we believe that the proposed approach makes a strong case for the synergistic combination between scanning probe and other imaging methods with combinatorial material science. While traditionally combinatorial studies have been limited by the ability to quantify materials structure and functionality across sample libraries, recent advances in focused X-Ray methods and scanning probe microscopy (SPM) offer a solution. At the same time, the proposed hypothesis-driven Gaussian Processes framework developed here further allows incorporation and selection between physical models. Note that while the approach is implemented here for the 1D case, it is straightforward to extend it to higher-dimensional parameter spaces and more complex physical problems.

As a second example of hypothesis learning, we demonstrate this approach towards exploring ferroelectric domain switching. In these experiments, the SPM tip is used to create ferroelectric domains, the size of which is controlled by the magnitude and length of the bias pulse. We create a list of possible models (hypotheses) of material behavior. The target of the automated experiment is to most rapidly minimize the uncertainty in the hypothesis space by iteratively choosing experimental parameters. In this manner, the automated experiment adopts human-like decision-making and implements it as a part of the discovery workflow.

Overall, the combination of the physics-driven machine learning and externally-controlled SPM tools opens a fundamentally new opportunity for materials discovery [3].

References:

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