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Machine Learning-Driven Automated Scanning Probe Microscopy for Ferroelectrics

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Machine learning (ML) and artificial intelligence (AI) have been applied to determine the physical mechanisms involved in phenomena encoded within microscopy data [1], enabling ML/AI to rapidly become an indispensable part of physics research. However, the real-time connection between ML and microscopy—which enables automated and autonomous experiments for microscopy imaging and spectroscopy measurements—still lags. Until now, the search for interesting functionalities in microscopy experiments has been guided by auxiliary information from microscopy to identify potential objects of interest based on human intuition; the exploration and verification of physical mechanisms depend on human-based decision making, i.e., operators determine the parameters for subsequent experiments according to the previous experiment. Here, we developed a ML-driven automated experiment (AE) scanning probe microscopy (SPM) workflow (Figure 1) to learn the functionality and mechanism in materials in an automatic manner. We demonstrate the application of these ML-AE workflows by investigating ferroelectric materials, including studies of domain wall dynamics, domain switching mechanism, and the conductivity of topological defects.

First, we deployed a Deep Kernel Learning (DKL) framework in Piezoresponse Force Microscopy (PFM) to explore the domain structure and physical properties of ferroelectric materials during the experiment, which allows problem-specific tuning of workflow and operation in real-time [2]. The structure of the DKL kernel provides insight into the physics of the process. This approach is used to explore the relationship between polarization switching or nonlinearity and domain structure in ferroelectric materials (Figure 2). Second, we developed a hypothesis-learning-driven SPM workflow for the exploration of domain switching in classical ferroelectric materials, which allows autonomous identification of mechanisms of bias-induced domain switching [3]. In this workflow, several possible hypotheses describing the system's behavior are available to complement the automated experiment, the ML algorithm aims to establish the best model of the system's behavior within the smallest number of steps following a certain optimization policy. Third, a workflow is developed to study predefined objects in SPM measurements [4]. This workflow first maps a large field of view and identifies the objects of interest, it subsequently performs zoomed-in measurements on the objects automatically to investigate their properties.

We implemented these approaches in SPM for ferroelectric materials investigation, however, these approaches can be applied for a broad range of physical and chemical microscopy experiments. The workflows can be adapted to apply to a broad range of imaging and spectroscopy methods, e.g., electron microscopy, optical microscopy, and chemical imaging [5].

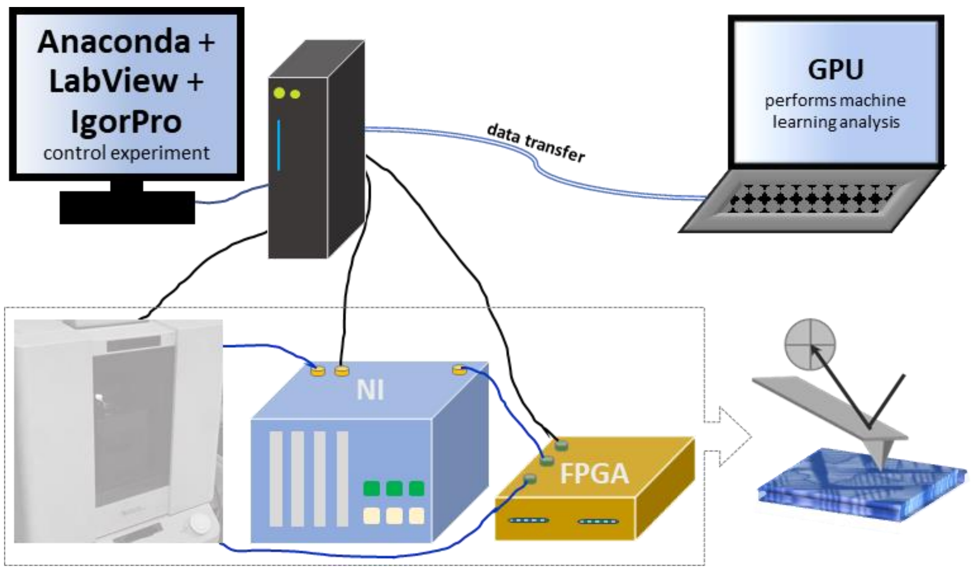

Figure 1. A schematic of the ML-driven automated SPM platform components. The platform includes an Oxford Instrument Asylum Cypher, an in-house LabView-based National Instruments hardware (LabView-NI), a Field Programmable Gate Arrays (FPGA), one computer for measurement control, and another computer with GPU (supercomputer) for machine learning analysis. In ML-SPM measurement, the FPGA controls the tip position by sending position as electrical signals to the Asylum Cypher and simultaneously sends a trigger to LabView-NI for measurement; LabView-NI generates excitation waveform and acquires data; The data transfer between analysis computer and measurement computer is enabled through a LAN cable.

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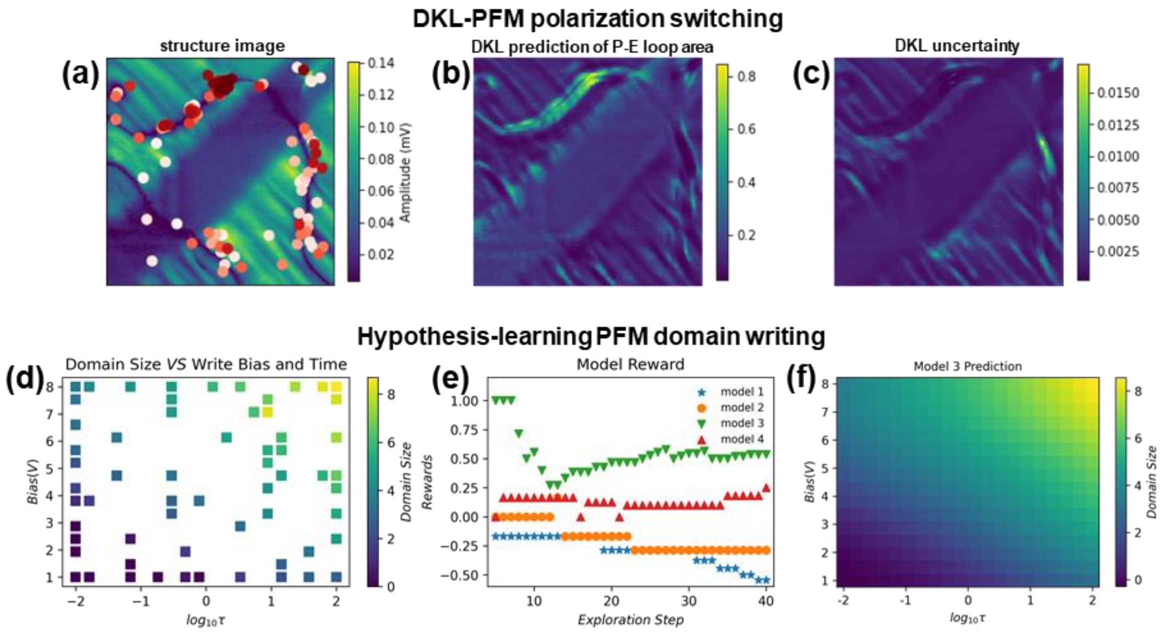

Figure 2. (a-c) DKL-PFM results of a $PbTiO_3$ thin film, the DKL exploration used (a) a PFM amplitude image as structure image, which indicates both a/c and c/c domains in the film. During experiment, DKL actively explores the relationship between domain structure and local hysteresis loop. (b-c) show the DKL predictions of hysteresis loop area based on 200 hysteresis loops and the corresponding DKL uncertainty, respectively. (d-f) Hypotheses-learning results of a $BiFeO_3$ thin film, which demonstrate the domain switching mechanism in relation to applied voltage and time. (d), experiment data of written domain size as a function of writing parameters including writing voltage and time. (e), model rewards during the hypotheses learning, indicating that model 3 gained a higher reward than other models, and its reward gradually increased at the latter part of the experiment. (f), prediction of domain size as a function of writing parameters by four different model 3.

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