Experimental discovery of structure–property relationships in ferroelectric materials via active learning

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Emergent functionalities of structural and topological defects in ferroelectric materials underpin an extremely broad spectrum of applications ranging from domain wall electronics to high dielectric and electromechanical responses. Many of these functionalities have been discovered and quantified via local scanning probe microscopy methods. However, the search has until now been based on either trial and error, or using auxiliary information such as the topography or domain wall structure to identify potential objects of interest on the basis of the intuition of operator or pre-existing hypotheses, with subsequent manual exploration. Here we report the development and implementation of a machine learning framework that actively discovers relationships between local domain structure and polarization-switching characteristics in ferroelectric materials encoded in the hysteresis loop. The hysteresis loops and their scalar descriptors such as nucleation bias, coercive bias and the hysteresis loop area (or more complex functionals of hysteresis loop shape) and corresponding uncertainties are used to guide the discovery of these relationships via automated piezoresponse force microscopy and spectroscopy experiments. As such, this approach combines the power of machine learning methods to learn the correlative relationships between high-dimensional data, as well as human-based physics insights encoded into the acquisition function. For ferroelectric materials, this automated workflow demonstrates that the discovery path and sampling points of on- and off-field hysteresis loops are largely different, indicating that on- and off-field hysteresis loops are dominated by different mechanisms. The proposed approach is universal and can be applied to a broad range of modern imaging and spectroscopy methods ranging from other scanning probe microscopy modalities to electron microscopy and chemical imaging.

The rapid evolution of scanning probe and electron microscopy techniques over the past three decades has revolutionized areas of science ranging from materials and condensed matter physics to chemistry and biochemistry. As such, the various microscopy imaging modes have now become a mainstay across virtually all scientific fields. Similarly, the combination of imaging and spectroscopic modes in these techniques has provided a wealth of information on structure–property relations in these dissimilar systems. Examples include scanning tunnelling microscopy and spectroscopy; dark and bright field imaging in electron microscopy and electron energy-loss spectroscopies; topographic imaging and force–distance curve measurements in atomic force microscopy; and electromechanical hysteresis loop measurements in piezoresponse force microscopy. These structure–property relationships in turn yield a wealth of information on the underpinning physical, chemical and biological mechanisms.

The locations for spectroscopic measurements are very often selected manually on the basis of the perceived (by human operator) interest in specific locations, as identified via features in a structural image. This point-and-click selection can be based on-field-specific intuition, curiosity and in special cases on a specific hypothesis. Alternatively, measurements can be performed in spectroscopic grid modes where the spectral data is collected over a uniform sampling grid. These in turn necessitated development of the linear and non-linear dimensionality-reduction methods for analysis of such multidimensional data, ushering exploratory machine learning methods into imaging areas; however, these imaging modalities are characterized by substantial disparities in acquisition times for the spectroscopic and structural measurements. Correspondingly, the spatial density of the information is limited. Although post-acquisition pan-sharpening methods based on compressed sensing, Gaussian process and so on have been developed, these approaches do not change the fundamental limitation of the spectroscopic imaging methods. Similarly, correlative learning of structure–property relationships implemented via im2spec approach requires the availability of the full dataset, and implicitly assumes that the material properties did not change as the result of measurements.

The rapid progress in the computer vision methods enabled by the advent of the deep learning a decade ago, as well as a wave of interest towards autonomous driving systems, has stimulated strong interest in autonomous microscopy, with several notable opinion pieces over the past three years. However, realization of this vision necessitates solution of three intertwined problems, including direct control of the microscope operation via external electronics, development of machine learning algorithms enabling the automated experiment and—perhaps less obviously—identifying the specific problems that the automated experiment seeks to resolve. Until now, this last problem has been largely overshadowed by the first two.

The direct control of microscopes has been available for decades, typically developed in the context of atomic and particle
For imaging, the adaptive non-rectangular scanning approach was demonstrated by Ovchinnikov and co-workers in 2009. More recently, Huang et al. and Stores et al. have demonstrated the combination of machine learning algorithm with atomic force microscopy (AFM) enabling autonomous operation of AFM without the need of human intervention in imaging modes, with the automated experiment playing the role of (pre-trained) feature identifier.

However, the second key component of the automated experiment is the strongly coupled problem of machine learning algorithms with specific physics. The automated experiment targeting the mechanisms of the ferroelectric domain wall pinning on structural defects will generally pursue a different strategy to the experiment exploring the interactions between ferroelectric and ferroelastic domain walls. We have recently reported the detailed analysis of the machine learning perspectives in automated/autonomous experiments in microscopy. In particular, we noted that the automated experiment itself is defined only in the context of past knowledge and seeks to discover new information or minimize uncertainties in the known system behaviour. This coupled machine learning and physics problem in the context of active learning makes the automated experiment a highly domain-specific problem.

Until now the automated experiments have been implemented using either human-based features engineering or simple deep convolutional neural network-based image recognition. For example, in piezoresponse force microscopy (PFM), the automated experiment was introduced based on a line-by-line feedback system employed during classical rectangular scanning by Kelley et al.; termed FerroBot. FerroBot has shown the feasibility of the automated experiment in PFM by using simple operator-defined features of interest. Finally, a Gaussian process/Bayesian optimization (GP/BO) framework has been recently developed to control probe trajectory via leveraging the explored information and scanning sequence.

At the same time, the classical GP/BO routines that underpin most automated experiments over the past years are based purely on the data available through the specific experiment and are further limited to low-dimensional signals. As such, they are ill-suited for the active learning of the structure–property relationships.

Here we implement the deep kernel learning (DKL)-based experimental workflow for active discovery of structure–property relationships in ferroelectric materials. This approach combines the power of machine learning to establish the correlative relationships between multidimensional datasets, and human-based physical reasoning to establish targets for exploration based on observations and their uncertainties. Here the relationship between the local domain structure and hysteresis loop is explored and future measurement locations are selected based on learnt relationships between local domain structure and polarization-switching behaviour.

To illustrate the principle of the DKL applications in experiment, we first implement DKL using a pre-acquired high-density band excitation piezoresponse spectroscopy (BPS) imaging dataset, which provides the known ground-truth image. Here, as a model system we have chosen a PbTiO₃ (PTO) thin film grown on (001) KTaO₃ substrates with a SrRuO₃ conducting buffer layer by the metalorganic chemical vapour deposition method, as reported by H. Morioka and colleagues. Figure 1a shows the topography image of the PTO sample, illustrating clear ferroelastic domain wall pattern. The clearly visible corrugations on the sample surface are associated with the ferroelastic domain walls between the domains with different polarization orientations. Here the lattice mismatch across the single domain walls gives rise to strain and hence deformations. The superposition of deformations form multiple domain walls give rise to ripple-like structure.

Figure 1b,c shows the corresponding band excitation PFM amplitude and phase images, which indicate the existence of both in-plane a domains and out-of-plane c domains. In the former, the polarization vector is parallel to the surface and thus the associated electromechanical response amplitude is close to zero. At the same time, the response amplitude in the latter is high. Finally, the phase image indicates whether the polarization vector is parallel or anti-parallel to the surface normal.

We analyse the high-density polarization loop measurements to explore polarization dynamics in this system. In these measurements, the direct current component of the tip bias follows the triangular waveform, inducing domain nucleation and growth below the tip. The resultant changes of the PFM signal are recorded as the local hysteresis loop. The shape of the hysteresis loop thus reflects the mechanism of local polarization switching as affected by the ferroelastic walls, structural defects and so on. The resultant three-dimensional data array can be analysed to extract descriptors of the hysteresis loop such as area under the loop, coercive and nucleation biases and so on, which can be plotted as two-dimensional maps. A high grid density (100×100) polarization image is shown in Fig. 1d, where a similar domain structure is visible. Figure 1e,f displays ferroelectric polarization hysteresis loops from the points marked on Fig. 1d.

The combination of the PFM domain structure image and hysteresis loop mapping allows one to reconstruct structure–property relationships, defined here as a correlative link between the local ferroelastic domain structure and hysteresis loop shape. We have previously demonstrated the use of machine learning methods (specifically, an encoder–decoder type of neural networks) to build such relationships for ferroelectric and plasmonic structures. These correlative relationships allow answering questions such as: what responses are possible in a given system? What structures are necessary to maximize certain aspects of the response? And so on. However, as with any correlative method, these answers are valid only for the in-distribution data (meaning for the same material under the same microscope settings), and do not allow answering counterfactual and intervention questions. Some of these can be answered via transition from correlative to generative physical models, but these raise further questions of theory–experiment matching.

Furthermore, these analyses are limited to the case in which the full dataset is available a priori, that is, they allow for analysis of the data after the experiment. However, it is not guaranteed that the pre-acquired datasets sampled the most interesting locations, and the number of possible measurements are limited by acquisition time and probe stability. By contrast, in an active machine learning setting, only the full topographic and PFM images are available, and information contained in these images is used to select the locations for spectroscopic measurements. Further locations are identified on the basis of the examination of spectroscopic data. For example, the operator can learn that: the in-plane a–domain regions do not have a measurable hysteresis loop; all a–c–domain structures have similar switching behaviours; and irregular domain edges or junctions may possess interesting dynamics. The subsequent selection of the target locations can then be based on these observations and curiosity (exploration) or perceived usefulness (exploitation). Importantly, this approach is the basis of DKL.

The DKL is based on GP regression and can be represented as a combination of the GP with deep neural networks. The GP generally refers to an indexed collection of random variables, any finite subcollection of which has a joint multivariate Gaussian distribution. A GP is completely determined by its mean and covariance functions, with the latter determining the functional form and strength of interaction between the points in the input space. A common application of the GP is in a regression setting in which it can be used for reconstructing data from sparse observations with quantified uncertainty. Note that the uncertainty is important when gaining quantitative insights into physical behaviours, and locations with high uncertainty can indicate the presence of new physical mechanisms. Specifically, given the dataset \( D = \{ x_i, y_i \}_{i=1,...,N} \), where \( x \) and \( y \) represent inputs/features...
and outputs/targets, respectively, the GP probabilistic regression model with a standard squared exponential kernel of the form $k(x, x') = \sigma^2 \exp\left(-\frac{1}{2l^2}(x - x')^2\right)$ is defined as

$$y \sim \text{MultivariateNormal}\left(0, K\left(x, x', \sigma, l\right)\right)$$  \hspace{1cm} (1a)

$$\sigma \sim \text{logNormal}(0, s_{\text{const}})$$  \hspace{1cm} (1b)

$$l \sim \text{logNormal}(0, s_{\text{const}})$$  \hspace{1cm} (1c)

where $K$ denotes a function that computes a kernel matrix such that $K_{ij} = k(x_i, x_j)$ for the sampled kernel hyperparameters and $s_{\text{const}}$ is a positive number. The GP model can be trained either using a Markov chain Monte Carlo algorithm on the model to get posterior samples for the GP parameters or via a variational inference. It is commonly assumed that there is an observation noise such that $y_{\text{noisy}} = y + \epsilon$, where $\epsilon$ is a normally distributed noise with zero mean and variance $\sigma^2_{\text{noise}}$. Practically, this noise gets absorbed into a computation of the covariance function.

Once the GP model parameters have been learned, they can be used to make predictions on new inputs. This is accomplished by sampling from the multivariate normal posterior over the model outputs at the provided new inputs $x^*$:

$$f_* \sim \text{MultivariateNormal}\left(\mu^\text{post}, \Sigma^\text{post}\right)$$  \hspace{1cm} (2a)

where $\theta = [\sigma, l]$ and

$$\mu^\text{post} = K(x_*, x|\theta)K(x, x|\theta)^{-1}y$$  \hspace{1cm} (2b)

$$\Sigma^\text{post} = K(x_*, x_*,|\theta) - K(x_*, x|\theta)K(x, x|\theta)^{-1}K(x, x_*)$$  \hspace{1cm} (2c)

The GP predictive mean ($f_*$) and variance ($\nabla f_*$) can be used to select the next measurement point(s) via a so-called acquisition function $\text{acq}(f_*, \nabla f_*|\theta)$, so that $x_{\text{next}} = \text{argmax}(\text{acq})$. The acquisition function reflects the measure of interest to specific region on the basis of the expected function value and uncertainty, balancing exploration and exploitation. Implementation-wise, one generally starts with a few sparse observations and trains a GP model. A prediction on the new inputs—which usually represent all the unmeasured points in a selected parameter space—is then made and used to derive an acquisition function for sampling the next query point. This approach is referred to as BO or active learning. For structural imaging in microscopy, the parameter space corresponds to a two-dimensional grid over a chosen scan area; for spectroscopic measurement, a third dimension corresponding to the energy axis can be added.

A substantial limitation of standard GP-based active learning is that it does not scale well with dimensionality of the parameter space. As a result, for many common hyperspectral measurements in 3D–5D space, the GP training and inference may take so long (even with modern graphics processing units) that it is faster to perform the measurement by simply sampling all of the points (that is, the standard way of doing measurements). Another limitation is that the standard GP does not, strictly speaking, learn representations of data that preclude use of prior knowledge from different experimental modalities to assist in the experiment (something that a (good) experimentalist does all of the time). In the context of the ferroelectric domain studies detailed here, the simple GP/BO does not use the information on the pre-existing domain structure to build the relationship between these and switching behaviours.

To address these issues, here we have adapted a deep kernel learning approach where a neural network is used to convert high-dimensional input data into a set of low-dimensional descriptors on which a standard (base) GP kernel operates (Fig. 2a). We formally define our deep kernel as

$$k_{\text{DKL}}(x, x'|\theta) = k_{\text{base}}(g(x)|w), g(x'|w)|\theta)$$  \hspace{1cm} (3)
where $\mathbf{w}$ are the weights of the neural network $g$. The deep kernel hence operates in the latent (embedding) space learned by a neural network from the (potentially high-dimensional) data and can be referred to as the data-informed kernel. The parameters of the neural network and GP base kernel are learned simultaneously by maximizing the model evidence via a stochastic variational inference\(^5\). The trained DKL GP model is then used for obtaining predictive mean and variances following the equation (2), that is, using the standard GP kernel $k(z, z'|\theta)$ operates (where $z$ are the embedded/latent features). The BO loop. At the training step, the DKL-based GP regression model (for brevity, referred to as DKL model) is trained using a small number of observations where inputs are topographic image patches and outputs are the corresponding spectra or scalar values of a specific property derived from the spectra. At the prediction step, a trained DKL model is used to predict spectra at every coordinate in the topographic image for which there is no measured spectra. Importantly, this method outputs both the expected mean value, $\hat{f}_m$, of the property of interest and the associated uncertainty, $\mathcal{V}[f_m]$, which are used to derive an acquisition function $\text{acq}(\hat{f}_m, \mathcal{V}[f_m])$ for selecting the next measurement point (see b). Note that if the output is a vector-valued function (such as spectra) it needs to be scalarized before passing to the acquisition function such that the exploration process is controlled by a single descriptor that is a function of the predicted functionality and its uncertainty. In this manner, the human operator defines what physical functionality is targeted during the experiment. Note that exploration can be based both on physical and information-theoretical criteria, that is, targeting variability of observed behaviours (curiosity learning).

To illustrate the DKL approach, we first implement it on the pre-acquired dataset. Here we use random sampling with 15% of measured points for training the DKL model that subsequently makes predictions on the full dataset. Figure 3 shows the results of DKL analyses on the pre-acquired BEPS data (as shown in Fig. 1d,e). Both analyses on hysteresis loop area and width indicate good reconstructions of loop area and width maps (Fig. 3c,k) compared with the ground-truth maps (Fig. 3b,j). The embedded variables highlight the tiny (Fig. 3g,o) and large domains (Fig. 3h,p), respectively. Figure 3q–s shows several examples demonstrating the reconstruction of hysteresis loops versus ground-truth loops.

**Fig. 2 | Schematic illustration of active learning with DKL.** Here the algorithm has access to the full topographic data and uses BO to learn the relationship between the domain structure within the patch and hysteresis loop. a, The inner structure of the DKL model. A feedforward neural network $g$ parametrized by $\mathbf{w}$ projects the potentially high-dimensional input data $X$ into the low-dimensional latent space, $g(X|\mathbf{w})$, in which a standard GP kernel $k(z, z'|\theta)$ operates (where $z$ are the embedded/latent features). b, The BO loop. c, At the training step, the DKL-based GP regression model (for brevity, referred to as DKL model) is trained using a small number of observations where inputs are topographic image patches and outputs are the corresponding spectra or scalar values of a specific property derived from the spectra. d, At the prediction step, a trained DKL model is used to predict spectra at every coordinate in the topographic image for which there is no measured spectra. Importantly, this method outputs both the expected mean value, $\hat{f}_m$, of the property of interest and the associated uncertainty, $\mathcal{V}[f_m]$, which are used to derive an acquisition function $\text{acq}(\hat{f}_m, \mathcal{V}[f_m])$ for selecting the next measurement point (see b). Note that if the output is a vector-valued function (such as spectra) it needs to be scalarized before passing to the acquisition function such that the exploration process is controlled by a single descriptor that is a function of the predicted functionality and its uncertainty. In this manner, the human operator defines what physical functionality is targeted during the experiment. Note that exploration can be based both on physical and information-theoretical criteria, that is, targeting variability of observed behaviours (curiosity learning).
We further explore the effect of the number of sampling points for random sampling. Here we show the DKL reconstruction of the coercive field, hysteresis loop area and width on the basis of 1%, 3%, 5% and 10% of random sample data (Fig. 4a–c). Note that in this approach DKL aims to reconstruct the relationship between the local domain structure and the hysteresis loop properties on the

**Fig. 3 | DKL on the pre-acquired dataset for 15% of measured locations.** a–h. DKL analysis of loop area. a, 15% randomly sampled loop area data for training session. b, Ground-truth loop area map. c, DKL-predicted loop area. d, DKL uncertainty map. e, f, Absolute error map of DKL prediction (e) and the histogram distribution of errors (f). g, h. The embedded latent maps of the trained DKL; the embedding dimensions were set to 1 (g) and 2 (h). i–p. Same as a–h, but for DKL analysis of loop width. q–s. Examples of DKL prediction on hysteresis loops from three different locations with different domain structures, showing DKL-predicted loops and ground-truth loops.
basis of a fully known collection of image patches, and patch–spectrumpairs available for only a (very small) fraction of the data. Withthis relationship established, it aims to reconstruct the spectra properties and their uncertainties for the full dataset. Figure 4d–f shows the quality of the reconstruction as the mean squared error (MSE) between the reconstruction and the ground truth as a function of the number of sampled data points. Note that in Fig. 4a, the coercive field is the average of the positive and negative coercive fields; in Fig. 4b,c, the loop area and width are from off-field hysteresis loops. Further results on the separated positive and negative coercive fields, and the on-field loop area, are shown in Supplementary Fig. 1. Further details on how the random sampling of data affects the DKL reconstruction is shown in Supplementary Videos 1–5.

We further illustrate the transition from reconstruction based on predefined (for example, random or low-density grid) sampling to science-driven discovery and experimental implementation of this approach. Here the critical new element is the definition of the scalar descriptor that reflects the physical behaviours we are interested in and use to guide the exploration process. For the hysteresis loop shown in Fig. 5a, these can be parameters such as loop area or width, coercive field and nucleation bias, or more complex descriptors such as quality of functional fit by predefined function, fit parameters and so on. Alternatively, the exploration can be based on information-theoretical criteria such as growth of entropy of the dataset (that is, curiosity learning). The key element here is that the acquisition function summarizing the degree of interest to a specific behaviour allows human-level decision making searching for specific physical signatures, and allows to incorporate associated uncertainties.

Figure 5b,c shows the DKL-BO navigation sequence with the acquisition function based on hysteresis loop area and loop width. Figure 5d,e shows the DKL reconstruction of loop area and width with 0.5%, 1%, 5% and 10% of the points where the DKL-BO suggested to perform measurements. The reconstruction quality can be evaluated by comparison with the random sampling points in Fig. 4. The reconstruction process can be reproduced through the provided code in the Methods.

Finally, we deploy the DKL discovery workflow on the operational microscope. Here we combined the DKL discovery workflow in Jupyter notebook with an in-house LabView-based script for National Instruments hardware (LabView-NI) to control the tip position for BEPS waveform generation and data acquisition. First we performed a band excitation PFM (BEPFM) measurement to acquire the domain structure image, which will be used to...
generated domain structure image patches for DKL. At the beginning of BEPS hysteresis loop measurement, the sampling point is initialized by the LabView-NI at a random location to obtain hysteresis loop. The Jupyter notebook then analyses the hysteresis loop and the corresponding pre-acquired domain structure image patch to train the DKL model. The DKL was trained for 200 steps, that is, until 200 pixels worth of data were captured.

The process is then repeated. A schematic of this specific workflow used is shown in Fig. 6a. Figure 6b,c shows BEPFM amplitude and phase images with grid sizes of 256×256, which will be used to generate image patches for DKL. It can be seen that the PTO film contains both in-plane $a$ domains and out-of-plane $c$ domains, as demonstrated previously. The grid size of the generated image patches for DKL-BO is 20×20, whereas the BEPS measurement was performed at a grid size of 237×237. The DKL-BO process was continued for 200 steps, that is, until 200 pixels worth of data were captured.

We used the image patches generated from the BEPFM results in Fig. 6b,c to guide the discovery workflow using on- and off-field hysteresis loop areas. The discovered points are labelled. The detailed discovery processes of acquisition function images with labelled exploration points are shown in Supplementary Videos 6 and 7. Interestingly, the DKL-BO-sampled points for on-field hysteresis loop area are concentrated around the $c^*/c^*$ ferroelectric domain walls (Fig. 6b), whereas the points for off-field area are concentrated around $a/c$ ferroelastic domain walls (Fig. 6c), demonstrating the potential of this approach to discover different behaviours based on predefined exploration targets. This is indicative of the different properties included in the on- and off-field hysteresis loops.

With the obtained 200 hysteresis loops and the BEPFM domain structure images, we are able to make predictions on the hysteresis loop area maps. Figure 6d,e shows the DKL prediction of on- and off-field hysteresis loop area maps, respectively, along with the DKL uncertainty. The domain structures are visible in the predicted loop area maps, indicating the hysteresis loop is associated with the domain structure.

These observations can be readily rationalized (but not predicted) on the basis of the known physics of ferroelectric domain walls. Here the larger polarization mobility in the vicinity of the 180 walls results in more substantial hysteresis loop opening in the on-field measurements. At the same time, the off-field measurements detect only the slowly relaxing (on the measurement time scale) components, which is indicative of the stronger pinning at the ferroelastic walls. These behaviours generally agree with some of the prior observations of similar systems\textsuperscript{[46,47].}

To summarize, we have developed the DKL approach that allows the physical discovery in automated experiment. Compared with the classical BO-based strategies that use a single (or small number) of scalar descriptors to guide the navigation process and do not incorporate the prior knowledge, this approach uses the data contained in structural images to identify the locations of the spectroscopic measurements, and identifies new locations and builds the structure–property relationships simultaneously. This discovery process is guided by the acquisition function that
is constructed from predicted behaviour and its uncertainty, and reflects the target of the experiment. This target can be optimization of specific property, similarity to a given model, or novelty discovery. In this manner, we combine the power of correlative machine learning methods to establish relationships between multidimensional dataset and derive corresponding uncertainties, and human physics-based decision making encoded in the choice of the acquisition function.

Here we implemented this approach for PFM measurement to investigate the relationship between polarization hysteresis and ferroelectric/ferroelastic domain structures. The obtained results show different exploration path and sampled points when the DKL is guided by on- and off-field hysteresis loops, indicating that the structure–hysteresis relationship varies under different circumstances (on- or off-field). We also note that in principle the deep neural network part of the DKL can be pre-trained on previous experimental data from the same or similar systems, somewhat equivalent to the transfer learning approach; however, this will necessitate the stringent analysis of the out-of-distribution drift effects (for example, due to different microscope settings).

Similarly, this workflow can be readily extended to other scanning probe microscopy (SPM) modalities, including current–voltage curve or relaxation measurements. We expect that most substantial benefit will be achieved for measurements with the readily identifiable connection to materials physics (for example, signature of Majorana fermions in STM), large acquisition times and especially destructive measurements such as nanoindentation and irreversible electrochemical measurements. Beyond SPM, similar approaches can be used to techniques such electron microscopy, chemical and mass-spectroscopic imaging, and nanoindentation and micromechanical testing. Finally, the DKL approach can be implemented over more complex parameter spaces, for example for material discovery in combinatorial spread libraries or molecular systems.

Methods

Data analysis. The detailed methodologies of DKL analysis on pre-acquired data are established in Jupyter notebooks and are available from https://git.io/JRspC. A standard MLP with three hidden layers containing 1,000, 500 and 50 neurons was used as a feature extractor in the DKL model.

PTO sample. The PTO film was grown by chemical vapour deposition on a SrRuO3 bottom electrode on a KTaO3 substrate.

BEPM and BEPS measurements. The PFM was performed using an Oxford Instrument Asylum Research Cypher microscope with Budget Sensor Multi75E-G Cr/Pt-coated AFM probes (~3 N m–1). Band excitation data are acquired with a National Instruments DAQ card and chassis operated with a LabView framework.

DKL-PFM implementation. The DKL deployment notebook for BEPS measurement is available from https://git.io/JRspC, which can be adapted for other modalities.

Data availability

The data that support the findings of this study are available at https://git.io/JRspC (https://zenodo.org/badge/latestdoi/393505955).

Code availability

The code of this study is available at https://git.io/JRspC (https://zenodo.org/badge/latestdoi/393505955).

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Author contributions

S.V.K conceived the project and M.A.Z. realized the DKL-BO workflow. Y.L. performed detailed analyses with basic workflow from M.A.Z. Y.L. deployed the DKL to PFM measurement and obtained results. R.K.V. and K.P.K. helped with the
deployment. H.F. provided the PTO sample. All authors contributed to discussions and the final manuscript.

Competing interests
The authors declare no competing interests.

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