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FerroNet: Machine Learning Flow for Analysis of Ferroelectric and Ferroelastic Materials

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Ferroelectric and multiferroic materials are among the most fascinating materials classes, due to both broad spectrum of applications and dazzling array of physical phenomena they exhibit. For over 70 years, ferroelectric structure and the nature of the order parameter were explored using the macroscopic scattering methods, providing the information on the average structure and symmetries, as well as correlate disorder. In the last decade, multiple studies of polarization behavior in ferroelectrics were reported using the direct imaging of atomic coordinates via the (Scanning) Transmission Electron Microscopy ((S)TEM), where atomic coordinates were used to map polarization field. However, implied in the analyses to date were the existence of macroscopically-defined polarization field as a (single) order parameter in the system and the relationship between the mesoscopic polarization and local atomic coordinates was postulated based on macroscopic models. In more complex analyses, the bulk form of the Ginzburg Landau free energy was additionally adopted as determined from macroscopic thermodynamic and scattering studies. However, in many materials systems such as morphotropic and relaxor ferroelectrics, the nature of the order parameter itself and hence corresponding free energy expansions are actively debated. Correspondingly, of interest is the question whether these descriptors can be obtained from the experimental data, as opposed to being postulated.

Here we explore the nature of the building blocks in the morphotropic ferroelectric systems using the statistical analysis of the atomically-resolved STEM data. Using the deep-learning analysis, we identify the localization of atomic columns from noisy experimental data in the form of the probability density field. We further explore the use of several linear and non-linear statistical unmixing techniques including Gaussian mixture models and independent component analysis to build the library of structural distortions and the associated domain structures.

As a first step of the analysis, we adopted the deep learning neural network analysis to convert noisy experimental data into atomic coordinates of different atomic species. We used a U-net-like fully convolutional neural network (FCNN) supplemented by the dilated convolutions in the bottleneck layer, which allows to perform the simultaneous mixed-scale de-noising of the atomic image and separation of atomic columns with different intensities into different “channels” (classes). This approach allows rapid identification of atomic positions based on local contrast. The output of the FCNN is a probability density field, which shows a probability of each pixel belonging to a given type of atomic column.

To get fundamental insight into nature of the elementary building block in the material, we generated the local neighborhoods of size d from the output of FCNN for each site in the lattice, as shown in Fig. 1 (a). These sub-images are centered at the center of mass of each individual column, and hence are robust with respect to intrinsic factors such as large scale strains and distortions, and extrinsic factors such as microscope drift. On this stage, the image is transformed from 2D object to the set of sub-images c_n, where $n = i,j$ defines the lattice site at which sub-image is centered.

Based on the initial analysis of information content in the system (set of sub-images c_n) via principal component analysis (Fig. 1), we further implemented the set of linear (non-negative matrix factorization

(NMF), independent component analysis (ICA), Gaussian Mixture model (GMM)) and non-linear (manifold learning) methods for the analysis of the data, all of which allow for a certain form of physical constraints. Of these, NMF separates the mixture in non-negative components, corresponding to positive intensities for the image. ICA operates to "decrease the Gaussianity" and maximize the variability between components. GMM seeks to represent the data as probability to belong to a specific component of the model mixture. The best results for the *linear* clustering/decomposition analysis on our data were obtained using the NMF, which clearly showed presence of different domain structures. We emphasize that our approach works in the presence of high levels of noise and large image distortions where the "standard" methods may fail.

The analysis is implemented in the form of Google Colaboratory notebook [1] and is fully available to the STEM community [2].

References:

[1] https://github.com/pycroscopy/AICrystallographer.
[2] The authors acknowledge funding from the US Department of Energy, Office of Science, Basic Energy Sciences, Division of Materials Science and Engineering (CN, SVK). The work was performed at the Center for Nanophase Materials Sciences, which is a US DOE Office of Science User Facility.

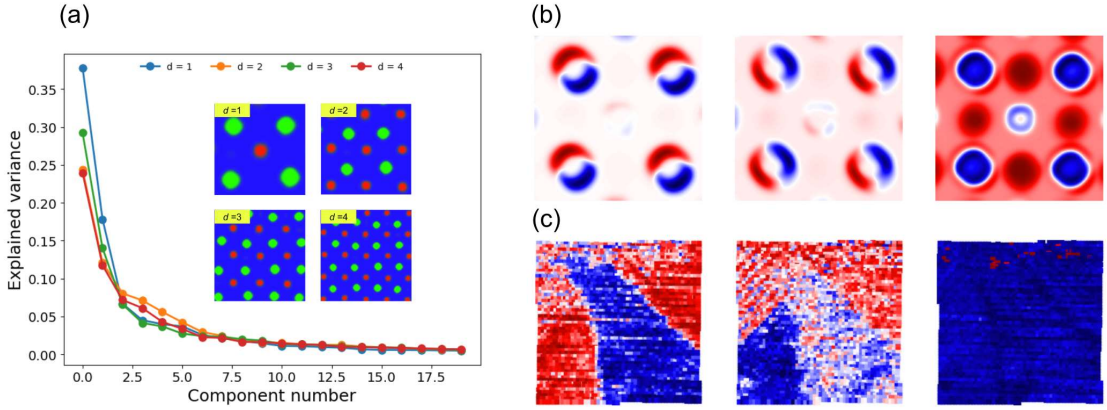

Figure 1. Analysis of ferroelectric distortions in $La_xBi_{1-x}FeO_3$. (a) Principal component analysis (PCA) scree plot corresponding to different size of local descriptors (set of sub-images) extracted from the neural network output (see the insets; "red" and "green" are the two sublattice classes, "blue" is a background class). (b, c) PCA decomposition of the local image descriptors extracted from the neural network output into three components with the associated eigenmodes (b) and loading maps for each eigenmode (c) , which are consistent with the projected [101] axis A-site displacements characteristic of $BiFeO_3$.

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