Learning from Imperfections: Predicting Structure and Thermodynamics from Atomic Imaging of Fluctuations

Lukas Vlcek[2], Maxim A. Ziatdinov[3], Artem Maksov[3], Alexander Tselev[1], Arthur P. Baddorf[3], Sergei V. Kalinin[3], Rama K. Vasudevan[3]

An equilibrium phase diagram of a chemical system or an alloy graphically shows regions, usually in respect to temperature and chemical composition, where single phases or structures are stable, and regions where two or more phases must coexist. A phase diagram is an indispensable tool for understanding and predicting system behavior under different conditions. Phase diagrams are generally based on experimental data or thermodynamic calculations with a single sample being used to derive information on a single point in a diagram. Therefore, construction of phase diagrams and identification of special points on phase diagrams demand significant experimental efforts, and frequently not all compositions can be investigated in equal detail. Construction of phase diagrams based on fewer experimental data would significantly facilitate the search for and discovery of new materials.

This work demonstrates an approach where an atomically-resolved image of a single sample is used for determination of structural and chemical variations of a compound, which then analyzed with methods of statistical testing, statistical physics, as well as machine learning algorithms to obtain a predictive model of the material for a range of chemical compositions. The approach makes a direct use of the imperfections, impurities, and stochastic details of material structure present in atomically resolved microscopic images. The developed framework (Figure 1) was applied to infer effective atomic interactions driving Ca and La segregation in a La5/8Ca3/8MnO3 thin film. The results demonstrate that atomic-scale studies of a single composition can provide information on a finite area of the chemical space, and this information can be used to reconstruct material properties in a finite composition and temperature range. The optimized model is further analyzed by a variational autoencoder to detect anomalous behavior in the composition phase.

1 — Department of Physics & CICECO, University of Aveiro
2 — Materials Sciences and Technology Division, Oak Ridge National Laboratory, USA
3 — Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, USA

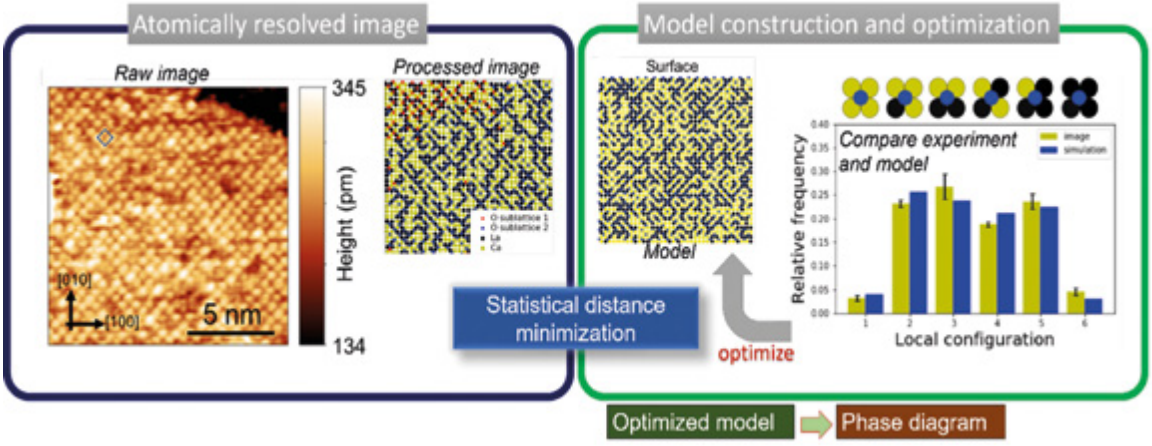

FIGURE 1
a) TEM micrograph showing the The proposed workflow involves processing the data from atomic-scale imaging with account of data from macroscopic measurements and first-principles theory. Atomic resolution images (such as the scanning tunneling microscopy image shown on the left) provides information on location and composition of the atoms. The image is processed to facilitate the comparison with the generative model. The generative model is optimized to minimize the statistical distance between the model and the experimental data. The result after optimization is a model that can be used to predict the system behavior as a function of, e.g., chemical potential or temperature. These data can be fed directly into artificial intelligence algorithms to determine any anomalies in the phase diagram and to highlight points for further investigation.