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Big, deep, and smart data from atomically resolved images: exploring the origins of materials functionality

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The rapid progress in high resolution (scanning) transmission electron microscopy, scanning probe microscopies, and associated spectroscopic techniques now provides a wealth of information on materials structure and functionality. The examples include atomically resolved structural images containing data on atomic positions with picometer precision, tunneling and EELS spectra containing information on local chemical properties and electronic structure, and many others. However, this progress brings forth the challenge of converting the large volumes of local structural and spectroscopic data into materials-specific information to further develop knowledge and insight into physics of specific materials systems.

Here, we discuss the pathways for systematically exploring structural and spectroscopic imaging data to understand materials functionality. By now, the foundations of "big data" techniques for image analysis in (S)TEM and SPM fields are well established, including the compressed sensing approaches for image and spectral reconstruction, pansharpening for image-spectral fusion, and multivariate techniques for spectral unmixing. However, in many cases these techniques are developed from a purely statistical perspective and are not directly connected to fundamental physics and chemistry of materials.

In this presentation, I will demonstrate several approaches to develop a physics-based statistical techniques for analysis of (S)TEM and STM imaging and spectroscopic data. The introduction of hard-physics based constraints in linear unmixing problems allows separation of local responses in EELS and CITS into a physically meaningful information channel. A similar approach combined with a sliding Fourier transform (Figure 1) can be used for effective image segmentation, phase and defect identification, and order parameter mapping in atomically resolved images. The synergy of the two approaches can be used for direct structure-property relationship mining from correlation analysis of the registered spectral and structural data. We believe that systematic implementation of these approaches combined with the implementation of direct data streaming from imaging tools and development of key-word and content searchable data bases will enable systematic structure property correlation studies, opening this information for comparison and reuse. I will further discuss the use of atomically resolved data to reconstruct the thermodynamic properties of materials using statistical matching between images. This approach will be illustrated for an example of lateral segregation in the transition metal oxide solid solutions (see Figure 2).

Finally, I will delineate several pathways for using this information for optimization and design of functional materials by constraining the number of theoretically explored atomic configurations, providing feedback to model selection and parameter refinement in theory, and developing systematic property-synthetic pathways correlation.

References:

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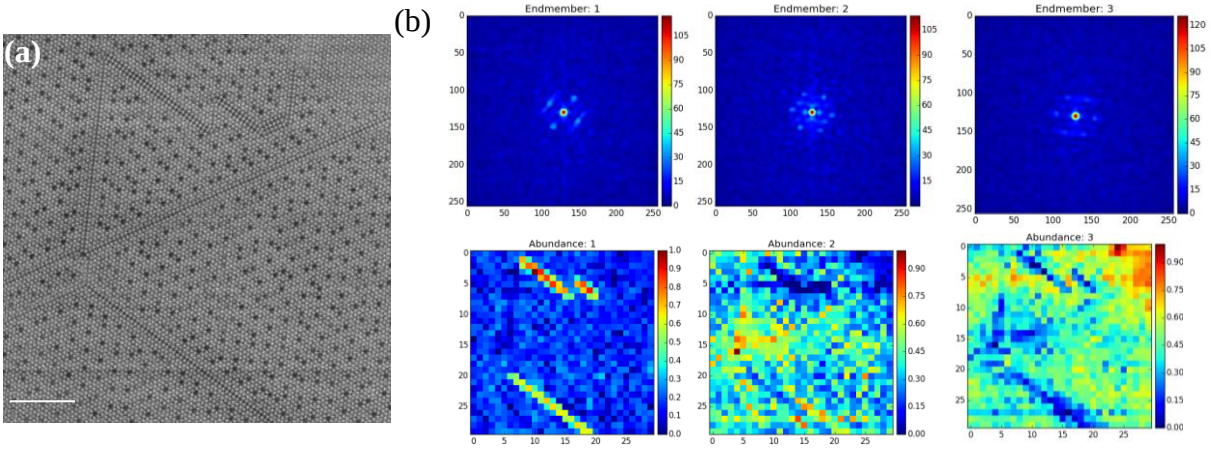

Figure 1. (a) STEM-HAADF image of the M2 phase of a Mo-V-Sb-Ta oxide catalyst. (b) N-FINDR Unmixing after sliding FFT was applied to the image in (a), and clearly identifies the dislocations, majority phase and defects of interest. Scale bar in (a) is 5nm.

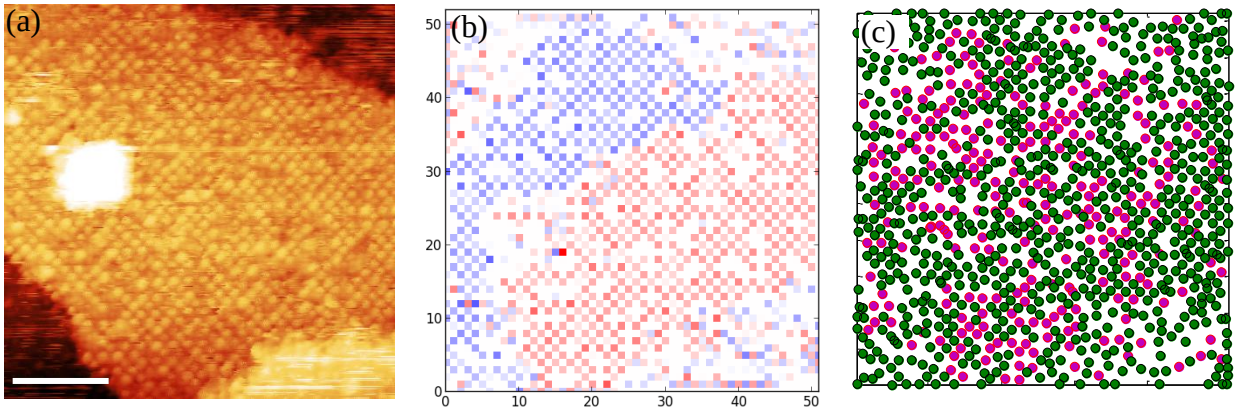

Figure 2. (a) STM topography of $La_{5/8}Ca_{3/8}MnO_3$ thin film. Scale bar, 5nm. (b) Identification of two oxygen sub-lattices in (a) by 2D pair distribution functions between centers of mass. (c) Segmentation by use of Gaussian fits and thresholding on intensity, which would correspond to sub-surface dopants and/or defects. This can be analyzed subsequently to determine level of (in-plane) dopant segregation.

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