Combining first principles computation and advanced characterization to understand disorder and interfaces in batteries

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The use of first principles density functional theory has been extremely fruitful in the study of crystalline materials for batteries, culminating in thousands of publications, thousands of structures and phase diagrams available online through the Materials Project,[1] and discovery of completely new cathode materials.[2] In the operation of real batteries however, disorder and amorphous phases are prevalent in the bulk and especially at interfaces. In these cases, advances in computational approaches as well as the combination of computation and advanced characterization techniques are needed to unravel the complex atomic arrangements.

In this talk, I will discuss how first principles computation is used to understand order-disorder transitions in the lithiation of crystalline silicon as a Li-ion battery anode[3] and in the charge reaction of Li_5FeO_4 in Li-O_2 cells.[4] In addition, I will discuss complexity at interfaces in surface-functionalized[5] and nanostructured[6] electrode materials. Strategies to create and validate atomistic models of disordered structures will be discussed and compared. In all of these examples, the importance of advanced characterization techniques such as x-ray absorption, scattering, and reflectivity, and how they are used in conjunction with first principles computation, will be highlighted.

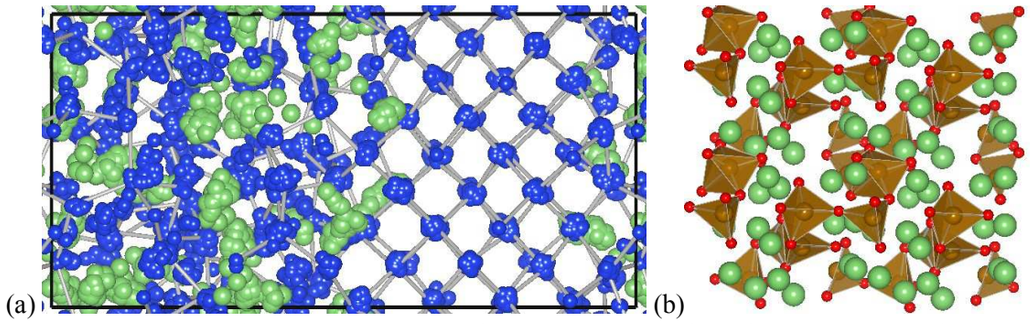

Figure 1: Amorphous and disordered structures in the bulk and at interfaces: (a) crystalline-amorphous interface between lithiated and unlithiated Si, (b) disordered structure from charging Li_5FeO_4.

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[1] materialsproject.org
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[5] In collaboration with P. Rajput, K. Puntambekar, M. Hersam, M. Bedzyk, T. Fister, P. Fenter, H. Yildirim, and J. Greeley.
[6] In collaboration with T. Fister and P. Fenter.