Abstract Submitted
for the MAR13 Meeting of
The American Physical Society

Exploring the electronic structure and dynamics of lithium compounds through first-principles interpretation of X-ray absorption spectra[1] DAVID PRENDERGAST, TOD PASCAL, XIN LI, JINGHUA GUO, Lawrence Berkeley National Laboratory, YI LUO, KTH Royal Institute of Technology — In situ X-ray spectroscopy will reveal fundamental details of elecltrochemistry in working cells, provided that the data is interpretable. To this end, we are developing first-principles methods to simulate core-level absorption spectra of molecules, condensed phases, and interfaces with explicit inclusion of dynamics. We validate this approach by application to various lithium compounds that may be present in the solid electrolyte interphase (SEI) and to lithiation of graphite in the anode. Our calculations reveal that instantaneous broken symmetry about the x-ray excited atom may be evident in the resulting spectroscopy and highlights both dynamical and static disorder in these materials. Furthermore, we observe complex anisotropic interactions upon charge transfer between lithium and graphite that contradict a simplistic view of intercalation in terms of complete electron transfer and the rigid band approximation.

[1]This work was supported by the U.S. Department of Energy under Contract No. DE-AC02-05CH11231

David Prendergast
Lawrence Berkeley National Laboratory

Date submitted: 09 Nov 2012 Electronic form version 1.4