Abstract Submitted
for the MAR14 Meeting of
The American Physical Society

First-principles interpretation of core-level spectroscopy of photoelectrochemical materials and processes[1] SRI CHAITANYA DAS PEMMARAJU, DAVID PRENDERGAST, Lawrence Berkeley National Laboratory — We present two case studies of first-principles theoretical methods applied in conjunction with experimental core-level spectroscopy measurements to investigate the electronic structure and dynamical processes in molecular and interfacial systems relevant to photoelectrochemical (PEC) technologies. In the first [1], we study the core-level and valence spectroscopies of two zinc(II)-porphyrin based Donor-pi-Acceptor (D-p-A) [2] dyes using the occupancy-constrained excited electron and core-hole (XCH) [3] approach and time-dependent density functional theory (TDDFT) simulations. In the second, we use constrained DFT and TDDFT to interpret measured transient core-level shifts in time-resolved femtosecond x-ray photoelectron spectroscopy, investigating the dynamics of the electron injection process from a N3 dye molecule chemisorbed onto a ZnO substrate. These studies illustrate the utility of first-principles methods in guiding the design of better PEC materials. References: [1] Zegkinoglou, I et al, J. Phys. Chem. C, J. Phys. Chem. C, 2013, 117, 13357 [2] Yella, A. et al, Science 2011, 334, 629. [3] Prendergast, D and Galli, G; Phys. Rev. Lett. 2006, 96, 215502.

[1]This work was performed at the Molecular Foundry, LBNL, supported by the Office of Science, Office of Basic Energy Sciences, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

Sri Chaitanya Das Pemmaraju
Lawrence Berkeley National Laboratory

Date submitted: 15 Nov 2013 Electronic form version 1.4