Abstract Submitted
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Band-edge engineering of Silicon by Surface Functionalization: a Combined Ab-initio and Photoemission Study[1] YAN LI, Computational Science Center, Brookhaven National Laboratory, LESLIE OLEARY, NATHAN LEWIS , Department of Chemistry, California Institute of Technology, GIULIA GALLI, Department of Chemistry and Department of Physics, University of California, Davis — The electrode material choice is limited in solar to fuel formation devices because of the requirement of band-edge matching to the fixed fuel formation potential. This limitation can be relieved via band-edge engineering. The changes of band-edge positions of Si electrodes induced by the adsorption of H-, Cl-, Br- and short-chain alkyl groups were investigated by combining density functional (DFT), many-body perturbation theory (MBPT), and ultraviolet photoelectron spectroscopy. The band edge shifts are related to the formation of surface dipole moments, and determine the barrier height of electrons and holes in doped silicon surfaces. We find that the trends of the sign and magnitude of the computed surface dipoles as a function of the adsorbate may be explained by simple electronegative rules. We show that quasi-particle energies obtained within MBPT are in good agreement with experiment, while DFT values may exhibit substantial errors. However computed band edge differences are in good agreement with spectroscopic and electrical measurements even at the DFT level of theory. [1] Y. Li and G. Galli, Phys. Rev. B 82, 045321 (2010). [2] Y. Li, L. O'Leary, N. Lewis and G. Galli, to be submitted.

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Yan Li
Computational Science Center, Brookhaven National Laboratory

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