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Energy Level Alignment at Aqueous GaN and ZnO Interfaces[1]
MARK S. HYBERTSEN, CFN, Brookhaven National Laboratory, NEERAV KHARCHE, JAMES T. MUCKERMAN, Chem. Dept., Brookhaven National Laboratory — Electronic energy level alignment at semiconductor-electrolyte interfaces is fundamental to electrochemical activity. Motivated in particular by the search for new materials that can be more efficient for photocatalysis, we develop a first principles method to calculate this alignment at aqueous interfaces and demonstrate it for the specific case of non-polar GaN and ZnO interfaces with water. In the first step, density functional theory (DFT) based molecular dynamics is used to sample the physical interface structure and to evaluate the electrostatic potential step at the interface. In the second step, the GW approach is used to evaluate the reference electronic energy level separately in the bulk semiconductor (valence band edge energy) and in bulk water (the $1b_1$ energy level), relative to the internal electrostatic energy reference. Use of the GW approach naturally corrects for errors inherent in the use of Kohn-Sham energy eigenvalues to approximate the electronic excitation energies in each material. With this predicted interface alignment, specific redox levels in water, with potentials known relative to the $1b_1$ level, can then be compared to the semiconductor band edge positions. Our results will be discussed in the context of experiments in which photoexcited GaN and ZnO drive the hydrogen evolution reaction.

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