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Electronic properties of semiconductor-water interfaces: Predictions from *ab-initio* molecular dynamics and many-body perturbation theory¹

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Photoelectrochemical cells offer a promising avenue for hydrogen production from water and sunlight. The efficiency of these devices depends on the electronic structure of the interface between the photoelectrode and liquid water, including the alignment between the semiconductor band edges and the water redox potential. In this talk, we will present the results of first principles calculations of semiconductor-water interfaces that are obtained with a combination of density functional theory (DFT)-based molecular dynamics simulations and many-body perturbation theory (MBPT). First, we will discuss the development of an MBPT approach that is aimed at improving the efficiency and accuracy of existing methodologies while still being applicable to complex heterogeneous interfaces consisting of hundreds of atoms [1,2]. We will then present studies of the electronic structure of liquid water and aqueous solutions using MBPT, which represent an essential step in establishing a quantitative framework for computing the energy alignment at semiconductor-water interfaces [3-5]. Finally, using a combination of DFT-based molecular dynamics simulations and MBPT, we will describe the relationship between interfacial structure, electronic properties of semiconductors and their reactivity in aqueous solutions through a number of examples, including functionalized Si surfaces [6] and GaP/InP surfaces in contact with liquid water.

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