Photoelectrochemical Water Splitting in Nanoscale Semiconductors

Fuel generation by photoelectrochemical (PEC) water splitting is a promising renewable energy technology and also a complex and fascinating mesoscale science problem. A critical advance in this field has been the advent of surface engineering approaches, which allows the processes of light absorption, charge transport, and interfacial energetics, reactivity, and stability can be independently tuned. Still, to develop fully optimized systems we need to understand the full time-dependent evolution of charge carriers. For example, impulsive light absorption within the absorption depth of the material is followed by an initial transport process that occurs on picoseconds timescales before minority carriers to reach the reactive interface. Using in situ time-resolved optical measurements, we are able to show that this ultrafast transport efficiency can vary greatly with nanoscale structure and material quality, and be an overall limiting factor in the incident-photon-to-current efficiency (IPCE). Here we demonstrate how changes in the interfacial chemical and electronic properties, including the addition of amorphous surface layers and other modifications to the interfacial depletion layer modifies the overall *ps*-to-*μs* charge dynamics.