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Elucidating how photoexcited semiconductor nanocrystals drive multi-electron redox catalysis

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Abstract

Colloidal semiconductor nanocrystals are remarkably versatile materials that exhibit a high degree of tunability in electronic structure, optical spectra, and surface properties. My research group is focused on the photophysics and photochemistry of nanoscale semiconductors with a particular emphasis on light-driven processes involved in multi-electron redox reactions. This seminar will focus on two stories. First, I will discuss coupling of semiconductor nanocrystals with redox enzymes to photochemically drive reactions such as reduction of H^+ to H_2 and N_2 fixation to make NH_3 , and carbon-carbon bond formation. Using time-resolved spectroscopy over a broad range of timescales (100 fs – 10 μ s), in conjunction with kinetic modeling, we examine charge transfer between photoexcited nanocrystals and enzymes to identify structural and chemical parameters that govern the overall photochemical reactivity. The second part of the seminar will focus on the dynamics of photoexcited holes in nanocrystals and the implications of those dynamics on oxidation photochemistry. In particular, I will discuss the spatial dynamics of trapped holes on nanocrystal surfaces and the consequences of their behavior on charge transfer.