Driving multielectron redox catalysis with photoexcited nanocrystals

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The synthetic tunability of electronic structure and surface chemistry of semiconductor nanocrystals make them attractive light absorbers for light-driven chemistry. A variety of architectures have been constructed where nanocrystals are coupled with multielectron redox catalysts to drive reactions like H2 generation, CO2 reduction, N2 reduction, and water oxidation with light. In these systems, light absorption in nanocrystals is followed by charge transfer to catalysts, which then use them for redox transformations, and/or to sacrificial carrier scavengers. Interfacial charge transfer between the nanocrystal and the catalyst and its competitiveness with other relaxation pathways in nanocrystals are of paramount importance to the overall photochemical reactivity. In this talk, I will focus on our efforts to elucidate both the kinetics of charge transfer and the kinetics and mechanisms of the competing photophysical pathways in nanocrystal-based systems for light-driven multielectron chemistry. This work entails transient absorption spectroscopy measurements, extensive kinetic modeling to extract rate constants of relevant processes in these heterogeneous systems, and, for some systems, contributions from theory.