Trapped hole diffusion in semiconductor nanocrystals

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Semiconductor nanocrystals exhibit remarkable tunability in electronic structure and chemical properties that have over the years led to developments in fundamental science as well as a number of applications. One of the persistent and ubiquitous features of excited state dynamics in multiple nanocrystalline materials is the fast and efficient trapping of photoexcited holes to the particle surface. The transformation of a photoexcited hole from a delocalized carrier to one that is localized to a surface site impacts processes such as electron-hole recombination and charge transfer. We recently reported that the trapped photoexcited holes are mobile on surfaces of CdS and CdSe nanocrystals, undergoing random-walk diffusion by hoping between chalcogen sites on the particle surface. This presentation will focus on our work in discovering and understanding the process of trapped hole diffusion. Specifically, experimental evidence for trapped hole diffusion in non-uniform nanorods of CdS and CdSe from transient absorption spectroscopy will be described. Temperature-dependent transient absorption measurements are consistent with a small polaron hopping description of trapped hole hopping close to room temperature, but not at low temperatures. Measurements of impacts of solvent and surface-capping ligand on trapped hole motion will also be discussed, as well as the evidence for diffusion-limited hole transfer.

References

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