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Nonradiative-decay mechanisms in CdSe nanoparticles: MUPPETS (multiple population-period transient spectroscopy) in excitonic systems[1] MARK BERG, KALYANASIS SAHU, SEAN KERN, Dept of Chemistry and Biochemistry, University of South Carolina — Nonradiative decay in semiconductor nanoparticles on the picosecond to nanosecond time scale is complex and poorly understood. Here, two-dimensional (2D) incoherent spectroscopy (MUPPETS) is applied to these processes in CdSe nanoparticles. For the first time, MUPPETS is extended to multilevel, excitonic systems to yield an analog of 2D coherent correlation spectroscopies. In core-only CdSe particles, the transfer of an excited electron from the core to the surface follows a highly dispersed, power-law decay in 1D measurements. 2D-MUPPETS measurements show that the rate dispersion is not due to relaxation nor due to multi-step kinetics, but results solely from particle-to-particle heterogeneity in the barrier to the surface. A model in which surface defects are distributed within the dipolar electric field of the particle accounts for the power-law decay. A second study of CdSe:ZnS core-shell particles uses correlation MUPPETS to distinguish biexcitons from photoproducts with a fast relaxing single exciton. Even when both species have similar lifetimes, they are distinguishable by having opposite signs and different symmetries in the two time intervals of a 2D experiment. Potential correlations between biexciton and exciton rates are sought, but are not found.

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Mark Berg
Dept of Chemistry and Biochemistry, University of South Carolina

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