"Extracting Charge Carriers from Colloidal Quantum Dots: Challenges and New Perspectives"



INORGANIC SEMINAR

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Abstract

Charge (Hole) Extraction

Surface Trapping

Nanometer-sized inorganic semiconductor crystals, better known as "quantum dots", offer a high potential for many opto-electronic applications, but their use has so far been limited mostly to light-emitting schemes, where advances in surface passivation have served to limit non-radiative recombination processes. Whereas the solid-state formalism is very well suited to describe the photophysics of quantum dots idealized as perfect particle-in-a-spherical-box models, there are challenges associated with understanding the behavior of quantum dots as electron donors and/or acceptors, a regime quite clearly outside the realm of the idealized infinite potential barrier at the surface of the quantum dot. In applications involving interfacial charge transfer between the semiconductor confined band states and outside redox-active species (photovoltaics, photocatalysis/photoredox, etc...), colloidal quantum dots that are effectively insulated from their surroundings cannot be used, and the complexity associated with surface states needs to be explicitly addressed. One of several important challenges limiting the use of QDs is the efficient extraction of holes from photoexcited QDs, a process that is often severely limited by competition with trapping to surface defect sites that are ubiquitous in QDs. We show that stable nitroxide free radicals can efficiently extract holes from II-VI chalcogenide QDs. By changing the structure and functionalization of these radicals, different aspects of electronic charge transfer can be monitored, including a wide tunability of the interfacial charge-transfer rate constants, from sub-picosecond to tens of nanoseconds, which is related to variations in electronic coupling and driving force. We also show the role that inhomogeneously-distributed surface traps play in interfacial photophysics of colloidal quantum dots, and how reasonable physical models can be obtained to describe the photophysical behavior of QDs with electronically-active surfaces.

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