MRSEC SEMINAR SERIES

"How bright is your quantum dot? Inducing multi-exciton emission with nanostructured gold."

Colloidally synthesized quantum dots (QDs) hold tremendous potential for low-cost processing and fabrication of optoelectronic devices. In addition to their size-tunable electronic structure and high photo-stability, their ability to support multiple optical excitations is being explored as a strategy for developing more efficient solar cells, brighter LEDs and even as sources of quantum entangled photons. Very often, we find that multi-excitons in QDs are poor light emitters due to a fast non-radiative decay pathway that competes with radiative recombination and makes multi-excitonic features difficult to observe using fluorescence. However, recent work in our group has demonstrated that gold nanostructures, which are able to collect and focus light over very small length scales, can effectively switch on multi-exciton emission when positioned in proximity to QDs. This effect has previously been observed for single excitations in QDs and molecules, but the ability to manipulate emission and absorption cross sections of multiply excited states using gold is leading us to some important insights about the nature and dynamics of multi-excited states in QDs.

In the first part of this seminar I will discuss recent single particle fluorescence data that show dramatic enhancement of QD multi-exciton fluorescence lifetimes in the presence of gold. We have demonstrated both bi- and tri-exciton emission from QDs that, in the absence of gold, exhibit neither. Some of the factors that control the magnitude of this plasmonic coupling will be illustrated and the implications for optoelectronic devices will be discussed. In the second part of the talk I will demonstrate a newly developed multi-pulse time-resolved fluorescence technique with which we are able to separate the recombination dynamics of multiply excited QDs from the dynamics of singly excited QDs in an ensemble solution. This allows us to selectively determine the effect of plasmonic structures on the emission and absorption rates of multi-excitons and makes it much easier to study the physical and chemical factors that control their recombination dynamics.

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