Efficient biexciton emission in single CdSe nanocrystals

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Abstract

Quantum-confined nanoparticles have been increasingly investigated over the past decade due to the superior efficiency and tunability of their emission wavelength from the ultraviolet to the near infra red. Among those nanoparticles, colloidal CdSe nanocrystals (NC) are particularly attractive for many applications such as nanoscale electronics, laser technology, quantum cryptography, and biological fluorescent labeling.

A detailed understanding of the NCs band-edge exciton fine structure is crucial for these applications. While intensive experimental and theoretical work has been performed to describe the size dependence of the exciton fine structure in nearly spherical NCs, the shape dependence has received much less attention despite recent advances in NC growth methods which lead to a greater control over shape distribution. Pioneering theoretical and experimental investigations [1, 2] have indicated that the shape dependence of NCs can be as important as the size dependence in terms of tuning their electronic and optical properties. The elucidation of these shape effects remains an experimental challenge which can be addressed by the optical study of individual NCs, where ensemble averaging over shape and size distributions is suppressed.

Shape and size effects also govern the optical response of NCs in the multiexcitonic regime, where potential applications such as optical gain are envisaged [3]. Despite the important role that biexcitons play in the optics of NCs, it has been practically impossible to observe the biexciton recombination line in the PL of CdSe NCs under continuous wave excitation, because of efficient nonradiative Auger recombination [4].

This presentation will be focused on our recent magneto-optical and time-resolved spectroscopic investigations of single commercial qdot655 streptavidin conjugates NCs (comprising a core of CdSe capped by a ZnS layer) as a function of temperature. The remarkable photostability of these NCs at low temperature led us to unveil the spectral and temporal signatures of the emission from the lowest exciton-fine-structure states [5,6], trion emission [7] and biexciton emission [8].

Because of the NCs shape distribution, we find various band-edge exciton fine structures that are consistent with theoretical predictions for elongated NCs. Furthermore, contrarily to what was anticipated for "standard" CdSe-based core shell NCs, we show evidence for spectral and temporal signatures of highly efficient radiative biexcitonic recombinations in this type of NCs. Special attention will also be paid to the attractive trion (charged exciton) emission properties for potential applications in quantum information processing.

References

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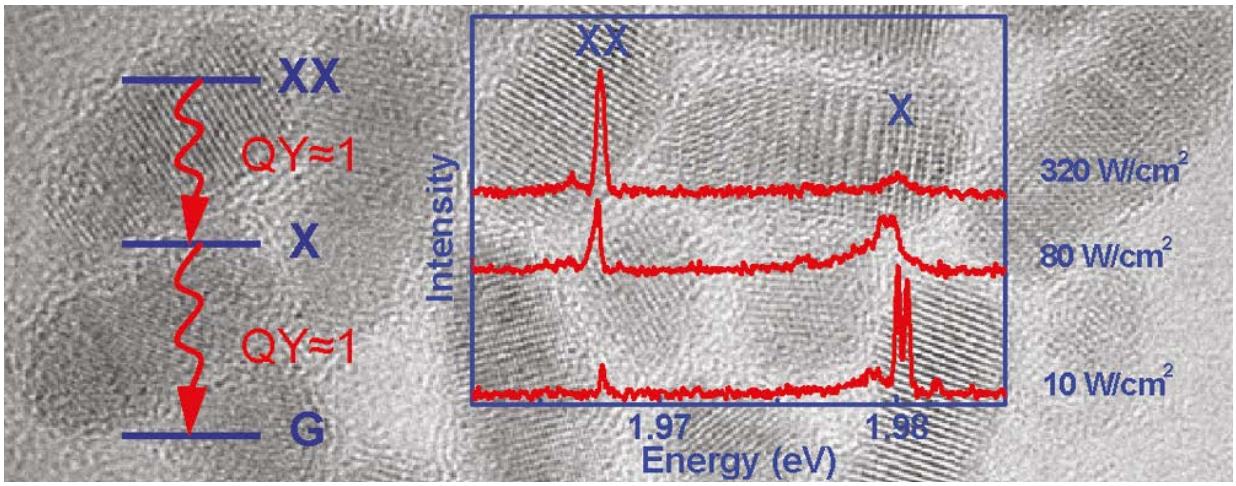
Figure



Emission spectrum of a single CdSe/ZnS nanocrystal at 2 K, showing evidence for strong radiative biexciton recombination.