Optical Probing of Non-Radiative Relaxation Processes of Confined Multiexcitons in Semiconductor Quantum Dots

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Abstract

We use polarization-sensitive, temporally resolved, intensity cross-correlation measurements to identify and study the triexciton-biexciton-dark exciton radiative cascades in self-assembled semiconductor quantum dots (QDs). These radiative cascades are indirect cascades involving a non-radiative phonon-assisted relaxation process between the two photon emissions. Our measurements reveal that the radiative cascade efficiency strongly depends on the polarization of the first emitted photon. We explain and model this dependence in terms of two interactions: the electron-heavy-hole exchange interaction and resonant electron- LO phonon Frölich interaction [1]. Our results shed new light on spin preserving and non-preserving relaxation processes involved in the thermalization of excited multiexciton states.

The ground-state triexcition [2] is formed by two spin-paired electrons and heavy holes in their ground spin singlet energy level and an additional unpaired electron hole pair in their respective first excited energy level. If the spins of the unpaired carriers are parallel, then there are two options for s-shell e-h pair recombination. Both leave an excited biexciton state in the QD. This biexciton then relaxes non-radiatively, either by spin flip or by spin flip-flop processes, before a second recombination occurs. After this, a single spin-parallel electron-heavy hole pair (dark exciton) remains in the QD. Our detailed model calculations [1] provide a novel way to accurately calculate the phonon assisted relaxation rates and thereby to quantitatively model the polarization-sensitive intensity cross-correlation measurements of these indirect radiative cascades.

[1] Y. Benny, et al. Phys. Rev. B **89**, 035316 (2014).
[2] E. R. Schmidgall, et al. Phys. Rev. B **90**, 241411 (2014).