Mechanism and dynamics of biexciton formation from a long-lived dark exciton in a CdTe quantum dot

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Abstract

Numerous optical studies of various quantum dot (QD) systems established a toolbox of techniques effective in characterization of single-dot photoluminescence (PL) spectra. In particular, measurement of the PL spectrum as a function of the excitation intensity is often employed to distinguish between different excitonic complexes [1]. The results are usually discussed in terms of a power-law behavior of the PL intensity. The simplistic stochastic model states for example that the formation of the biexciton requires a coincidence of two exciton formation events [2], and thus the biexciton PL intensity should increase quadratically with the excitation power. In real experiments, however, the biexciton PL intensity does not follow this prediction and exhibits less steep dependence [3].

In this work we analyze the dependence of a single CdTe QD PL on pulsed excitation intensity and demonstrate that in the wide range of intensities the key mechanism of the biexciton formation involves a dark exciton [4]. This long-lived state can store a photoexcited electron-hole pair until another pair is injected and form the biexciton. A primary consequence of this mechanism is nonquadratic excitation power dependence of the biexciton PL intensity.

The discussed mechanism opens interesting possibilities due to the fact that two electron-hole pairs forming a biexciton are excited by different laser pulses, which can be controlled independently. In particular, we exploit it to study an effect of carrier spin polarization on the dynamics of biexciton formation. Our results evidence that the dynamics of this formation is mainly governed by the effect of the electron spin blockade.

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