Pis'ma v ZhETF, vol. 123, iss. 6, pp. 425 – 426

Delayed photoluminescence blinking of single semiconductor quantum dots: novel experiments and modeling[1)]

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Submitted 6 February 2026
Resubmitted 6 February 2026
Accepted 7 February 2026

The photoluminescence (PL) kinetics of semiconductor nanocrystals (quantum dots, QDs) includes a surprisingly wide range of times. The presence of shallow metastable trap states leads to the appearance of a long tail on the PL decay curve, the so-called delayed PL, at times ranging from hundreds of nanoseconds to microseconds. Surprisingly, intensity and lifetime of the delayed PL from a single QD notably fluctuate in time. In this work, delayed PL of single colloidal core/shell CdSeS/ZnS QDs was in depth studied by time-gated PL traces analysis. Observed delayed PL blinking allowed us to study the shallow trap states. We show that single QD PL in active trap state is characterized by mono or bi-exponential decays with times altering from 60 ns to 600 ns, that is much exceed exciton lifetime in bright state ~ 30 ns. Simulation allowed us to conclude that the absence of an exciton component in the kinetics of delayed PL can be explained by the fast (~ 100 ps) charge carrier Auger trapping process on a shallow trap state. We associate this state with a localized defect at the inner core/shell QD interface. Also, we demonstrate that the variation in the lifetimes and delayed PL decay shapes can be described entirely by a change in the thermal de-trapping rate.

DOI: 10.7868/S3034576626030177

Quantum dots (QDs) starting from their discovery [1] are among the most promising nanoscale structures for the development and application of advanced technologies [2]. Their outstanding optical properties make them ideal for creating next-generation optoelectronic devices, such as lasers [3, 4], efficient white light sources [5, 6], high-efficiency solar cells [7, 8], luminescent markers for biovisualization [9–11], and single-photon sources [12–14].

Study of semiconductor nanocrystals (QDs) at the single-particle level allows for investigation of photophysical and spectral phenomena that are not detectable in ensembles, where the data are strongly averaged. These effects include spectral diffusion [15–17] as well as PL blinking [18], which are processes of stochastic variation of PL wavelength and PL intensity. In addition to blinking and spectral diffusion, QDs also show significant lifetime enhancement effects – delayed PL [19]. Studies of single CdSe/CdS QDs have shown that delayed PL is not uniform in time, and experiences significant changes in intensity – known as "blinking" of delayed PL [20]. However, the question of the origin of the delayed component of PL and its blinking remains unanswered to date.

In this paper, we provide a detailed study of the delayed PL blinking in single semiconductor colloidal CdSeS/ZnS QDs, focusing on the variation of delayed PL kinetics in time. We show that single QD PL in active trap state is characterized by mono or bi-exponential decays with times altering from 60 to 600 ns, that is much exceed exciton lifetime in bright state ~ 30 ns. The performed Monte Carlo simulation of charge recombination dynamics in QDs based on modified shallow trap models allows us to describe the observed PL decay features and verify the previously proposed mechanism of charge trapping by shallow traps. Simulation allowed us to conclude that the absence of an exciton component in the kinetics of delayed PL can be explained by the fast (~ 100 ps) charge carrier Auger trapping process on a shallow trap state. We associate this state with a localized defect at the inner core/shell QD interface. The release time of charge carriers from the trap can be much shorter than the calculated lifetime. This suggests that the observed lifetimes do not

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necessarily require long-term charge retention in the trap: there could be multiple cycles of trapping and de-trapping processes before radiative recombination event. Simulation also showed that the distribution of the delayed PL lifetimes can be reproduced by changing the de-trapping rate k_{dt}. This variation in the de-trapping rate may be due to the electric fields caused by the appearance and fluctuations of charges on the QD surface.

Funding. Authors acknowledge support for the research project from the Ministry of Education and Science of the Russian Federation "Photonic Technologies in Science and Education: Materials, Methods, and Tools for Ultra-High-Sensitivity Optical Sensing and Microscopy". I. Eremchev and A. Naumov acknowledge support for the research project from the Ministry of Education and Science of the Russian Federation (Grant # FFUU-2025-0004).

Conflict of interest. The authors of this work declare that they have no conflicts of interest.

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