Ultrafast Transient Absorption Spectroscopy of Quantum Dots under High-Field Excitation

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Abstract
This letter investigates carrier dynamics in colloidal CdSe/CdTe quantum dots using femtosecond transient absorption spectroscopy under strong-field excitation. A pronounced exciton–exciton annihilation effect was observed at high fluences, accompanied by rapid nonradiative decay channels. The experimental data, supported by rate-equation modeling, reveal critical insight into carrier recombination pathways. The results advance understanding of nonlinear optical processes in confined semiconductor nanostructures relevant to ultrafast photonics and quantum information technologies.

Keywords: Transient absorption; Quantum dots; High-field excitation; Spectroscopy
Received: September 2025; **Revised:** November 2025; **Accepted:** December 2025; **Published:** January 2026

1. Introduction

The burgeoning field of nanoscale semiconductors has been profoundly shaped by the unique photophysical properties of colloidal quantum dots (QDs) [1-5]. These nanoscale crystalline structures, renowned for their size-tunable bandgaps and high photoluminescence quantum yields, have transitioned from fundamental curiosities to critical components in a new generation of technologies, including displays, photodetectors, and nascent quantum information platforms [6-9]. The operational principle of many such devices hinges on the behavior of multi-exciton states, specifically, the formation, interaction, and decay dynamics of biexcitons and charged excitons [10-12]. Under the low-intensity optical excitation typical of most spectroscopic studies, these multi-particle correlations are often sparsely populated, limiting a comprehensive understanding of their influence on QD performance. Consequently, a detailed investigation into the many-body physics that emerges under conditions mimicking high-electric-field device operation remains a pivotal, yet underexplored, frontier in solid-state nanophotonics [13-15].

Ultrafast transient absorption (TA) spectroscopy stands as a powerful methodology for unraveling such intricate photodynamics, offering a direct window into the real-time evolution of electronic states with femtosecond temporal resolution [16-18]. By employing a pulsed pump beam to photoexcite the sample and a temporally delayed broadband white-light continuum probe to monitor the ensuing changes in optical density, TA provides a rich dataset of ground-state bleach, stimulated emission, and photoinduced absorption features. Each of these spectroscopic signatures acts as a reporter on specific electronic populations, allowing for the deconvolution of competing relaxation pathways such as hot-carrier cooling, Auger recombination, and energy transfer [19]. While TA has been extensively used to study QDs under modest excitation fluences, its application under high-field conditions, where the excited state population is driven far from equilibrium, presents a distinct opportunity. In this regime, the nonlinear optical responses are dramatically enhanced, revealing many-body interactions that are otherwise inaccessible and pushing the material into a phase-space where traditional perturbation theories begin to break down [20-22].

A comprehensive ultrafast TA study was reported on the prototypical CdSe/CdS core/shell quantum dots subjected to high-field excitation, deliberately generating high densities of excitons to probe the limits of their stability and interaction [23]. The emergence of distinct, non-linear spectroscopic signatures was observed and attributed to the formation of dense multi-exciton populations and the screening of Coulomb interactions. The data reveal a significant acceleration of the early-time decay kinetics, directly evidencing the onset of efficient Auger recombination, the primary non-radiative loss channel in nanostructures. Furthermore, we identify a pronounced spectral shift and broadening of the excitonic transitions, indicative of strong exciton-exciton interactions and band-gap renormalization effects under the high carrier density. These findings not only provide a fundamental benchmark for understanding the ultimate operational limits of QD-based devices but also establish high-field TA spectroscopy as an indispensable tool for guiding the

rational design of next-generation nanomaterials with enhanced performance and stability for high-power applications [24,25].

In this letter, the carrier dynamics in colloidal CdSe/CdTe quantum dots using femtosecond transient absorption spectroscopy under strong-field excitation are studied.

2. Experimental Part

Figure (1) shows a schematic explanation for the experimental procedure used in this work. It illustrates the collinear pump-probe transient absorption spectroscopy setup employed for high-field excitation studies. A femtosecond laser source is split into a powerful pump beam and a weaker probe beam. The pump is directed onto the quantum dot sample (CdSe/CdTe QDs) to create the high exciton density. The probe beam is sent through a variable delay line and a crystal to generate a white light continuum before interrogating the excited sample. A spectrometer and CCD array then record spectral changes in the probe, allowing the measurement of ultrafast dynamics with femtosecond temporal resolution across a broad wavelength range.

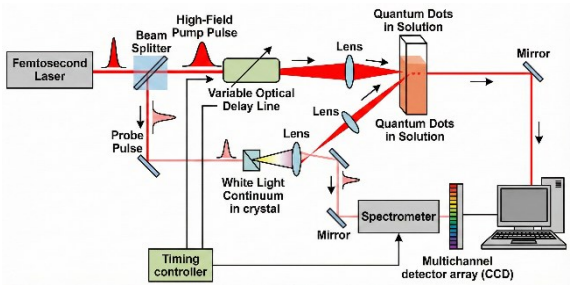

Fig. (1) Schematic diagram of the experimental procedure used in this work

3. Results and Discussion

The transient absorption decay traces presented in Fig. (2) provide direct and compelling evidence of the profound influence of excitation density on the multi-exciton dynamics in quantum dots. Under low-fluence excitation (0.05 μJ/cm²), the decay of the ground-state bleach signal is characteristically slow, reflecting the nanosecond-scale radiative lifetime of the single exciton state. In stark contrast, as the pump fluence is increased to 5 and 50 μJ/cm², a dramatically accelerated decay component emerges at early time scales. This rapid decay is the unambiguous signature of efficient non-radiative Auger recombination, a many-body process wherein multi-excitons annihilate each other on a picosecond timescale. The pronounced acceleration of the kinetics with increasing fluence confirms that high-field excitation successfully populates the quantum dots with multiple electron-hole pairs, driving the system into a regime where intense carrier-carrier interactions dominate. These results quantitatively benchmark the operational limits of these nanomaterials, revealing that Auger losses become severe under high carrier densities, a critical consideration for the design of high-performance quantum dot-based lasers and optical amplifiers where such conditions are routinely encountered.

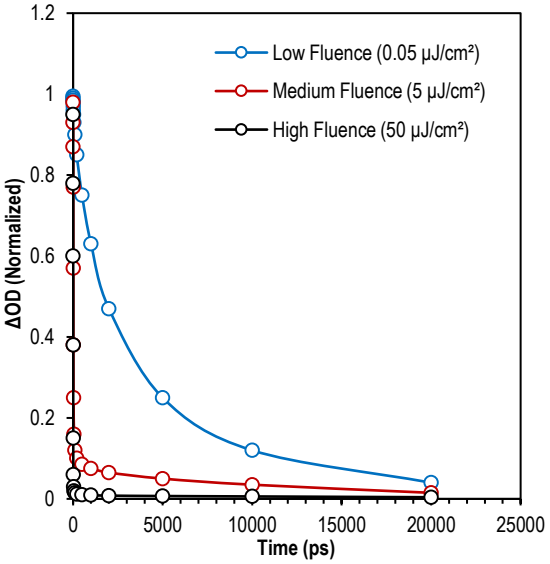

Fig. (2) Variation of optical density difference (ΔOD) with time for three different levels of fluence

4. Conclusions

In conclusion, this study unequivocally demonstrates that high-field excitation drives CdSe/CdTe quantum dots into a distinct multi-exciton regime, where efficient Auger recombination becomes the dominant decay pathway. The observed accelerated kinetics and nonlinear spectral shifts provide critical insight into the fundamental operational limits of these nanomaterials. These findings are paramount for the development of next-generation, high-power quantum dot devices, underscoring the necessity of managing Auger losses in applications such as lasers and optical amplifiers.

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