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Ultrafast Spectroscopy of CdS/CdSe Quantum Dots

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Abstract—Results from the nonresonance spectroscopy of CdS/CdSe quantum dots (composites of CdSe–CdS nanoparticles (core–shell)) are presented. The nonlinear optical properties of CdS/CdSe QDs in PMMA are studied with fs pulses at 1053 nm using the transient lens technique. QDs generate rapidly oscillating signals with amplitude rise times of around 200 fs and decay times of around 500 fs, while pure PMMA polymer only generates an oscillating signal whose envelope coincides with its autocorrelation function.

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INTRODUCTION

Due to their unique chemical and physical properties, semiconductor nanoparticles now find wide practical application [1]. In these materials, the quantum size effect affects the widths of band gaps, making them desirable in manufacturing optoelectronic emitters (LEDs), single-electron transistors (SETs), displays on quantum dots (QD LEDs), lasers, and so on [2, 3]. Among the known semiconductor nanoparticles, nanocrystallites of semiconductors CdSe and CdS are of particular interest, due to their unique optical properties. The evolution of photoinduced electrons and holes depends strongly on the size of the nanocrystallite and the properties of its surface. As a rule, recorded nonlinear responses have the picosecond relaxation times associated with processes of exciton–phonon interaction [4]. Along with this, there are Auger recombination processes [5] that produce faster subpicosecond decays of the observed optical responses.

Numerous ways of synthesizing semiconductor nanoparticles and controlling their size distribution were described in [6]. One problem during synthesis is the aggregation into clusters tens and hundreds of nanometers in size. This strongly suppresses the quantum-size effect in nanoparticles and makes their use in optoelectronics and other areas of nanotechnology impossible. The core–shell method was used to solve this problem. This allowed us to obtain quantum dots 5 nm in size, consisting of CdSe nuclei coated with a CdS shell. Such nanoparticles are not subject to aggregation and retain all of their optical properties, due to the quantum-size effect.

In this work, we used femtosecond nonresonance spectroscopy to observe the nonlinear optical response of synthesized quantum dots with ultrashort relaxation times of less than 500 fs.

EXPERIMENTAL

CdSe nanoparticles were synthesized via the colloidal method in a water–glycerin solution [7], after which a coating based on cadmium acetate and thioacetamide was applied to the CdSe nuclei. According to data from luminescent spectroscopy, the average size of composites of CdSe–CdS nanoparticles (core–shell) was 5 nm. The quantum dots obtained in this manner were mixed in pyridine with poly(methyl methacrylate) (PMMA) in a ratio of 5% by weight using an ultrasonic mixer. The resulting suspension was then poured onto a rotating quartz substrate (a technique known as spin coating) and dried at 60°C. The resulting sample was a film 0.3 mm thick. A PMMA film 0.3 nm thick on a quartz substrate and with no quantum dots was used for comparison.

The familiar pump–probe optical scheme was used in our experiments to record optical responses with femtosecond resolution. This scheme is based on two laser pulses. The first intense laser pulse excites the studied medium, and the second weak laser pulse probes the state of the medium after a fixed time. A ytterbium solid-state femtosecond TeMa laser (central wavelength, 1053 nm; duration, 170 fs; pulse repetition rate, 70 MHz; Yb: KGW; pumping with white LEDs using SESAM as a bleaching absorber) was used as our radiation source. Laser pulses with energies of 20 nJ (pumping) and 0.1 nJ (probing) were focused into a sample section with a diameter of ~ 50 μm. The delay time between pulses was controlled by an optical delay line in increments of 0.2 fs. A diaphragm is placed in the probing beam in front of the photodetector. This diaphragm blocks ~50% of the radiation with no pumping.

The experiment is based on the spectroscopic technique of an induced lens during nonresonance excitation (Fig. 1). A laser beam with a Gaussian intensity