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Nonresonant Transient Refractive Index Spectroscopy in Semiconductor Quantum Dots

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Abstract. We report transient refractive index change in semiconductor nanoparticles dispersed in polymethylmethacrylate matrix via pump-probe experiment. At lower pump intensities the detected signal consists of the pulse autocorrelation-shaped part and another part delayed by 300 fs. The latter's relative intensity depends on the pump level. However in CdS monocrystal the detected signal was found to lack this second feature completely.

Keywords: quantum dots, semiconductor nanoparticles, transient lens, nonlinear refractive index

The femtosecond laser radiation (central wavelength of 1050 nm, pulse duration of 100 fs and repetition rate of 70MHz) was split into pump and probe beams. Both pump and delayed probe beam were focused at the sample. Then the probe beam was collimated and directed to the photodiode through small aperture so only a fraction of it passed. The excitation wavelength was non-resonant.

The samples we used were CdSe-CdS nanoparticles in polymethylmethacrylate matrix composites and CdS monocrystal. CdSe quantum dots were synthesized through a colloid method in a water-glycerin mixture. The CdS shell layer based on cadmium acetate and thioacetamide was coated on the CdSe cores. The average nanoparticles diameter estimated as 5 nm (via luminescence specroscopy)

In CdS monocrystal the signal we detected mimicked the laser pulse autocorrelation function (obtained through noncollinear second harmonic generation) with full width at half maximum (FWHM) of 200 fs. When pump energy varies in the range of 1.4×10^{-11} J — 1.4×10^{-8} J the amplitude of the signal changes but not its shape. In CdSe-CdS apart from autocorrelation function there is a strong side signal with FWHM of 200 fs shifted towards positive delays by 300 fs. Intensity relations of those two components depend on the pump intensity: at 1.4×10^{-11} J they are well separated and the amplitude of the second part is 3 times smaller than the first one. As the pump increases, the second part grows stronger and eventually both components merge into one with FWHM of 500 fs.

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