Photoluminescence and nonlinear transmission peculiarities of the CdTe/CdSe nano-tetrapods

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Colloidal branched nano-heterostructures have attracted a great deal of attention owing to their elaborated morphology and electronic properties that differ from the spherical particles of the same materials. Among all the anisotropic nanoparticles the type-II tetrapod-shaped crystals are particularly interesting because of long-range-photoinduced charge separation and hence their potential photovoltaic applications. In this work photoluminescence (PL) properties and nonlinear transmission of tetrapod-shaped CdTe/CdSe nanocrystals were investigated at the one-photon stationary excitation regime. CdTe/CdSe nanocrystals consist of the CdTe tetrapod-shaped core with the CdSe tips grown selectively on the CdTe arms ends [1]. The average length of the CdTe tetrapod arms was 12±1 nm and the length of CdSe tips was 11±1 nm. Average thickness of both the CdTe arms and CdSe tips was about 3 nm. Pumping was realized by the Nd^{3+}:YAP laser second or third harmonic pulses (λ = 540 nm, τ=10 ns or λ = 360 nm, τ=9 ns, respectively). The neutral filters were utilized to vary the pump intensity from 1 kW/cm^2 to 14 MW/cm^2. As a probe signal the broadband PL of the Coumarin-120, Coumarin-7, Kiton Red and Oxazine-720 Perchlorate dyes were used.

Three well-resolved maxima were found experimentally in the PL spectra of the CdTe/CdSe nanocrystals at the one-photon stationary excitation of excitons. The presence of well-pronounced emission band in the 650-850 nm region is associated with the radiative recombination from the “charge-transfer” states (CT-states) involving spatially separated electrons and holes in the CdTe and CdSe domains of the nanocrystals. Two maxima in the PL spectra centered at the wavelengths 595 nm and 677 nm correspond to the lowest $1Sh_{3/2}$-$1S_e$ exciton’s states in the CdSe and CdTe domains, respectively. The saturation of the CT-PL occurs much faster than the saturation of the excitonic PL due to the significant difference in the radiative lifetimes of the CT- and excitonic states. PL saturation can be associated with the state-filling effect and Auger recombination processes of excitons for high pump intensities. Also a giant blue shift (≈129 meV) of the CT PL spectra with the growth of pump intensity was observed [2]. The shift occurs due to the increase of the exciton radius as a result of the exciton phase space filling. Differential transmission (DT) spectra were measured to investigate the nonlinear properties of the nano-tetrapods. DT spectra demonstrated the presence of superposition of the exciton’s transitions bleaching originating from the different structural domains of the heterostructured CdTe/CdSe nanocrystals, which confirm the observed separated excitonic emission bands in the PL spectra. It was found that saturation of the nonlinear absorption occurs at lower pump intensities than the saturation of CT PL and excitonic PL.

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[1] R. Vasiliev, D. Dirin, M. Sokolikova, J. Mater. Res. **26**, 1621 (2011).
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