Nonlinear absorption and photoluminescence of direct and charge-transfer excitons in CdTe/CdSe nanotetrapods

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The photoluminescence (PL) peculiarities and the excited state absorption of the tetrapod-shaped CdTe/CdSe nanocrystals (Fig.1a) are studied in the single-photon excitation regime [1]. Tetrapod-shaped type II heterostructured nanocrystals composed by different materials are among the most intriguing low dimensional semiconductor nanostructures with anisotropic shape [2]. From a fundamental point of view, heterostructured nanotetrapods allow more efficient long-range induced separation of photoexcited electrons and holes.

We demonstrate the pump intensity dependent giant blue shift (≈129 meV) of the indirect transition (charge-transfer – CT) emission band (Fig.1b) caused by the exciton's space filling effect and the modification of energy level structure by the induced internal electric field in both domains. The PL lines of the CdTe and CdSe domains, originating from direct electron-hole transitions were observed in addition to the tunable PL across the indirect gap (Fig.1b). Differential transmission spectra reveal the superposition of the exciton's transitions bleaching originating from different structural domains of the CdTe/CdSe nanocrystals, which confirms the observed separated excitonic emission bands in the PL spectra. The observed features of the PL and differential transmission spectra allow to determine and to analyze possible exciton's relaxation channels in the CdTe/CdSe nanocrystals (Fig.1c). The first-principles calculations reveal that the ground states of both electrons and holes are localized in the CdSe domain, but the charge carrier spatial separation is still present, as holes are localized at the interface [3]. This picture is proven by the time-resolved PL measurements from the sub-picoseconds to tens of nanoseconds range, which demonstrate the presence of a picosecond time scale corresponding to the indirect excitons being much smaller than the previously observed value.

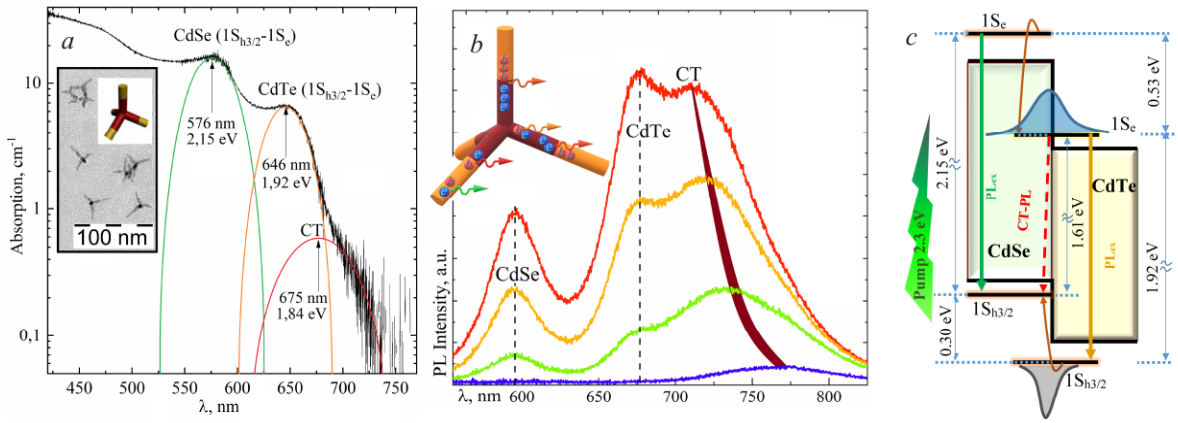

Fig. 1 a) The linear absorption spectrum of the tetrapod-shaped CdTe/CdSe nanocrystals. Gaussian bands corresponding to the CdSe and CdTe excitonic absorption bands (green and yellow lines) and charge-transfer (CT) band (red line). The inset shows transmission electron microscopy (TEM) image of the nanotetrapods and its sketch: red color demonstrates the core and the arms; yellow color shows the tips. b) PL spectra of the CdTe/CdSe nanotetrapod's colloidal solution for different pump intensities. c) Energy scheme of the CdTe/CdSe heterostructure. The scheme shows the band gap and the ground state energy levels for holes and electrons and schematically demonstrates the spatial localization of the electron and hole wave functions. Vertical (direct excitons) and inclined (indirect excitons) lines with arrows show main transitions, which contribute to the PL spectrum.

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