Title: Advanced spectroscopic characterization of quantum dot ensembles

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Abstract:

Semiconductor quantum dots (QDs) are small crystallites whose sizes (of the order of nm) cause spatial confinement of carriers in all 3 dimensions. As result, QDs often reveal very different physical properties in comparison with their bulk counterparts. From the optical point of view, the broadening of bandgap with QD-size shrinking is particularly interesting. It is a purely quantum mechanical effect that results from quantum confinement (QC), i.e. dimensional limitations of excitons. A strong spatial confinement leads to a relaxation of momentum (Heisenberg uncertainty principle), consequently, larger overlap of the wave-functions of carriers results in significant increase of probability of radiative recombination. Therefore ensembles of QDs are promising candidates for new generations of photonic and photovoltaic devices.

This PhD thesis is primary focused on detailed spectroscopic characterization of ensembles of direct (PbS) and indirect (Si) semiconductor QDs in both colloidal (toluene) and matrix-embedded (oxide or oxinitrides multilayers) forms. The oleic-acid capped PbS QDs were purchased commercially. Si QDs with dodecyl-functionalization were synthesized by non-thermal plasma method and the solid samples with Si QDs were fabricated by annealing multilayers of altering Si-rich and stoichiometric oxides deposited by the PECVD method (both types of Si QDs were fabricated and provided by our foreign collaborators).

The main aim of this work was to gain an insight on some fundamental problems in ensembles of QDs with an ambition to improve their optical characteristics, namely absorption and emission efficiencies. The optical properties of samples were characterized by time-integrated and time-resolved (TR) photoluminescence (PL) spectroscopies. The analysis of PL spectra and the position of their peak changing with size and material of QDs proved the QC-related origin of PL. Using the set-up with an integrating sphere we experimentally studied absolute quantum yield (QY) in dependence on various parameters (QD concentration etc.). To assess the absorption strength of Si QDs, namely absorption cross-section, we theoretically and experimentally developed PL modulation technique that is based on TR onset and decay transients. We paid special attention to careful investigation and setting of the optimum experimental parameters for TR PL experiments. By variation of the temperature and inter-QD distance in multilayer samples with Si QDs we proved the dependence of both absorption and emission properties on those parameters and determined the optimum oxide barrier thickness to maximize PL brightness. In addition, we developed a novel procedure to derive size-selected decays from spectrally resolved ones and retrieved 100% efficient size fraction of Si QDs in colloidal suspension of nearly fully-radiative QDs. These results were used for estimation of internal quantum efficiency (IQE) in solid samples. Finally, comparing results of IQE with PL QY we demonstrated and estimated the fraction of dark QDs in ensembles of QDs. Further investigation should be focused on understanding the physical properties of dark QDs to minimize their number and maximize the emission efficiency of ensembles of QDs.

Keywords: quantum dots, lifetime, quantum yield, internal quantum efficiency, absorption cross-section