Colloidal quantum dot field-effect transistors

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Summary

Many, if not all, of the modern technological developments became possible due to scientific progress in the field of semiconductors, which arises in the early XIX century. This field, that was considered to be 'suicidal' because of low reproducibility and technical challenges at the beginning of XX century, brought us the cleanest material, ever produced by humans, – crystalline silicon. Integrated circuits, made on silicon are now used for most of the electronic devices, are at the heart of computers and of photosensors.

Novel electronic materials, that are able to substitute traditional crystalline semiconductors for lower costs and easier fabrication, have been in the focus of many research projects over the last years. Quantum dots occupy a significate niche in this research, due to their simple synthesis, solution processing, and high performance. In particular, strong absorption of visible and near-infrared light makes them potential successor of crystalline and amorphous silicon photodetectors. Another advantage of quantum dots is their tuneable bandgap that depends on the physical size of the nanoparticles. Recently, many challenges have been overcome, such as photoluminescence blinking of the nanoparticles and poor conductivity of the film, what open a way for utilizing the quantum dots in displays, photodetectors, solar cells, field-effect transistors, and other optoelectronic devices.

However, despite great success in optoelectronics, quantum dot solids require additional work to overcome the remaining challenges, inherent to the nature of the material. As synthesized, colloidal quantum dots generally are capped with long aliphatic molecules (ligands), to ensure the passivation of the surface and stable dispersion of the nanoparticles in organic solvents. When deposited in a film, these ligands represent dielectric barriers, suppressing the charge transport through the film. To obtain charge transport between quantum dots, the original ligands are exchanged into smaller entities, either in solution or in pre-deposited film. However, the passivation of the surface by new ligands often is imperfect, thus causing the formation of surface defects. Due to the large surface to volume ratio, these defects result in a number of energy states within the band gap of the quantum dot film.

Additionally, the charge transport is readily influenced by device fabrication conditions, the stoichiometry of the surface and the nature of the ligands.

The aim of this thesis is to give fundamental insights into the charge transport process in quantum dots film and demonstrate high-performance optoelectronic devices, such as double gate field effect transistors, logic circuits, light emitting transistors and photodetectors.

In Chapter 2 the first double-gate quantum dot field effect transistor is introduced. The fabricated devices in a double-gate configuration are characterized by smaller hysteresis and higher electron and hole mobilities respect to single gate ones. The simultaneous use of the two gates allows superior control of the threshold voltage shift and of the polarity of the PbS CQDs FET by effective channel tuning. Further, we demonstrated that CQDs are fully compatible with multiple gate technology, expanding the possibilities of the circuit design in future CQDs-based integrated circuits.

The course for the integration was further followed in Chapter 3, where is demonstrated the first hybrid inverter, consisting of transistors using PbS CQDs as n-type material and single wall carbon nanotubes (SWCNTs) as p-type. The pair of FETs, gated with P(VDF-TrFE-CFE)/PMMA high-k polymer bilayer, demonstrates complementary characteristics, allowing using them in CMOS-like electronic devices. The inverter features sub-1V operation with the highest reported static gain (76 V/V) and noise margins (80%, calculated following maximum equal criteria principle), as for fully solution-processed devices.

In Chapter 4 we explore the first quantum dot light emitting field-effect transistor (QDLEFET) with solid state gating. We show that it is possible to combine near-infrared light generation and the current switching properties in a single transistor. The QDLEFET shows state-of-the-art switching performance, with an electron mobility of 0.06 cm2 V^{-1} s^{-1} and an on-off ratio of 10^4. At room temperature, the external quantum efficiency (EQE) of the light generation is $1.3 \cdot 10^{-5}$. To get further insights into the origin of relatively low EQE and the charge transport details, we performed low-temperature experiments, combining electroluminescence (EL) and conductivity studies. We found, that the EL EQE increases drastically with decreasing temperature and reaches 1% at temperatures lower than 100K. The origin of this increase is the decrease of the exciton dissociation rate, governed by phonon-assisted hopping of charge carriers between transport energy levels in CQDs film. We further strengthened our discussion by performing the conductivity measurements, which showed Mott-type variable range hoping to be the charge transport mechanism in the PbS CQDs films. Additionally, we were able to identify the presence of hole trapping within the bandgap, the presence of this trapping states is influencing largely the hole transport in ambipolar PbS QDs FETs.

After demonstrating the concept devices in chapters 2,3 and 4, in chapter 5 we show the full compatibility of PbS CQDs with upscale industrial techniques, such as optical lithography and wet etching. We demonstrate for the first time, that lithographically etched PbS CQDs film can be used as a material for high-performance photosensitive transistors. Further, we develop a method of lithographical fabrication of the top contact electrodes, avoiding the formation of injection barriers. The cut-off frequency of PbS QDs FETs was measured and reported for the first time, with the value of 400kHz. The transistors, additionally, demonstrate responsivity to near-infrared light. These findings show that PbS CQDs can be used as a material for optoelectronic devices, operating in the medium frequency range.

In summary, this thesis demonstrates the set of various devices, made of PbS CQDs, along with the fundamental description of optical and electrical processes occurring in PbS CQDs films. Our findings, along with the ones of others, suggests that CQDs have great prospects on the way to commercialization into viable technologies, especially in fields were traditional semiconductors show strong limitations, as the emission and detection of light in the near-infrared spectral region.