Colloidal core-shell quantum-dots in photovoltaic cells

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Colloidal quantum dots (CQDs) show a potential utilization as the building blocks of photovoltaic cells (PVCs). They exhibit size dependent band-gap energy, relatively large cross section of absorption (10^{-14}-10^{-15}cm^2), and photo stability over a length of time. In particular the IV-VI (PbSe, PbS), the InAs (P) or the CdTe CQDs show an optical absorption within the near infrared (600-3500 nm) with a tail into the visible regime, thus, can harvest a large spectral range of the sun-light. These CQDs also show a relatively long excited state lifetime (<msec of the single exciton), sufficient duration for the charge extraction. The group IV-VI CQDs, in particular also present low effective masses of the electrons and holes (~0.1 m_0), offering an option for a high mobility of the carriers. These CQDs act as the photo-active antennae in the PVCs, that must be coupled to charge transporters (e.g., TiO_2, electrolytes, polymers) or directly to the collecting electrodes (e.g., ITO, FTO, SnO_2). The lecture at the Quantsol 2009 meeting will include a description of a few preliminary attempts in impregnating CQDs into a sensitized PVCs (replacing the dyes in a dye-sensitized solar cell), or packed in a p-i-n configuration between conducting electrodes. Both alternatives required some chemical modification of the CQDs surfaces before the PCVs assembly. It should be noted that the colloidal procedure supply CQDs with organic surfactants, that are long and hydrophobic molecules, creating a potential barrier for a charge transport. Thus, a ligand exchange procedure were carried out, inducing a capsulation with a short-length molecules and occasionally also with conjugated or/and functional groups. Those chemical properties enable a better adhesion to the transporters and improved transport properties. Further on, the exterior ligands have a certain influence on the alignment of the CQDs Fermi energy with respect to that of the transporters.

One of the major concerns in the use of CQDs is related to the formation of excitons or multiple-excitons by a sun-light radiation (referring only to those multiple excitons that are formed by a sequential filling of the band states). It is very well known that multiple excitons in CQDs of type-I primarily decay via nonradiative Auger relaxation at a time scale of 10-100 ps, due to the enhancement of Coulomb interactions inside dot with a typical diameter of 3-4 and surfactant molecules of low dielectric constant, and thus, will not be of any benefit in PVCs. This work shows a suppression of an Auger relaxation and an extension of multiple excitons' lifetime in quasi-type-II CQDs. For example, recently, we unambiguously showed the emission of neutral biexciton (BX), triexciton (TX) and quandraexciton (QX) in a single CQD under continuous-wave excitation in CdTe/CdSe core-shell CQDs, using micro-photoluminescence (μ-PL) spectroscopy. The results revealed a remarkable spectral stability, approaching a blinking-free behavior. The excitons' binding energies were evaluated by a second order perturbation theory, considering Coulomb many-body

interactions in a quasi-type II CQDs, with a partial separation of the carriers' radial density distribution. The multiple exciton dependence of the excitation power was compared to a kinetic model, considering probability of states' population, a model that showed a close agreement with the experimental observations, and enabled to supply an estimate of the multiple excitons' lifetimes (from a few tenths to a few hundred seconds, proposing suppression of an Auger relaxation which permits extraction of the carriers in PVCs).

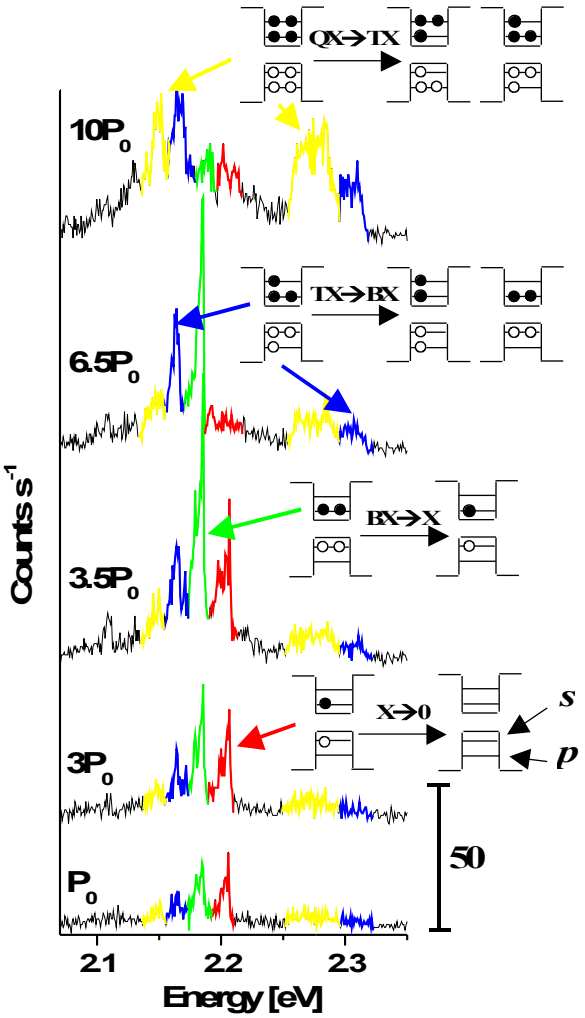

Fig. 1: micro-PL spectra of CdTe/CdSe CQDs, measured at various laser powers. The possible multiexciton recombination processes are given in the diagrams.

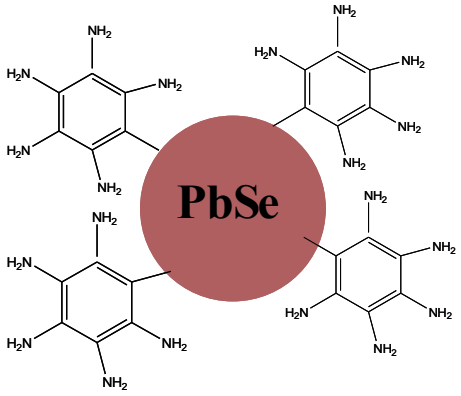

Figure 2: PbSe CQD capped with conjugated organic ligands,

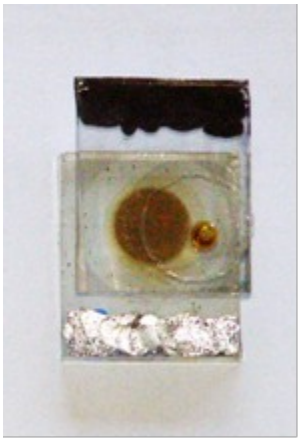

Fig.3: A Picture of a CQDs-sensitized photovoltaic cell