The significant of alloy colloidal quantum dots and quantum rods: electronic tunability, unique thermal and chemical stability, non-blinking behavior, carriers' multiplication

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Colloidal quantum dots (CQDs) attract worldwide scientific and technological attention, due to the ability to engineer their optical properties by the variation of their size. However, several important applications, such as biological tagging and photovoltaic cells, impose a limit on their size, yet demand tunability and thermal stability of the optical band edge. This work introduces a new class of heterostructures, comprised of PbSe and CdTe cores or rods, coated by $PbSe_xS_{1-x}$ or $CdTe_xSe_{1-x}$ shells, with different internal division, with a radial gradient composition, which offer a control of the band edge properties by varying the CQDs' composition. The continuous-wave and transient photoluminescence measurements over a wide temperature range (1.4 - 300 K) revealed a distinct behavior of the heterostructures with respect to that of pure cores or rods: (i) increase of the emission quantum yield; (ii) red-shift of the absorption edge, however, a decrease of the emission Stokes shift; (iii) alleviation of a dark exciton recombination, *viz*, a reduction of an exchange interaction; (iv) tuning of the radiative lifetime with shell width and composition; (v) reduction of the band edge temperature coefficient, dE/dT, *viz*, induction of thermal stability. The k•p envelope function calculation, considering abrupt or smooth alloying continuation of the potential at the core (rod)-shell interface, revealed a delocalization of the hole wavefunction over the entire volume of the CQDs, as a partial explanation for the marked tunability, nonetheless, preserving a desired size (see scheme below).

Single dot measurements of the alloyed heterostructures showed a unique spectral stability, exhibiting blinking-free behavior. Furthermore, the work resolved well-resolved neutral single-exciton, biexciton, triexciton, and quadraexciton emission bands in the μ-PL spectrum of a single CdTe/CdSe core-

shell CQD, pumped by a continuous-wave excitation and exhibiting nearly blinking-free behavior. The multiple excitons were generated by a sequential filling of the s- and **p**-electronic shells with the increase of the excitation power. Identification of the multiexciton emission bands was based on the evolution of the μ-PL spectrum with a change in the pumping power, showing dominancy of single and biexciton bands at the lowest pumping power, which fade away upon the increase of the pumping intensity, and these bands were replaced by a higher-order excitons at the highest pumping attained. Multiple excitons are of significant interest in gain devices as well as in photovoltaic applications. Despite the band-edge variations promoted by the internal division and composition of heterostructures, multiple exciton generation process (contrary to a sequential filling discussed above), was found to be less efficient than expected.

Scheme: Charge distribution between a core and a shell