Excitons' fine structure in the micro-photoluminescence spectrum of a single doped/undoped core/shell colloidal quantum dot

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The scientific and technological interest in colloidal quantum dots (CQDs) over two decades was served around their special electronic properties, exhibiting atomic-like density of states manipulated by variation of size and shape - those making them attractive for various opto-electronics and biological applications. Nowadays a new profit is recognized, implementing the CQDs as efficient component in quantum computing and spintronics device, anticipating generation of single or multiple excitons with defined spin orientation in undoped or magnetically doped CQDs. Hence, this emerging interest requires a detailed investigation of excitons' fine structures, including exploration of spectral split and polarization properties of excitons' recombination emission.

This work aimed to resolve the fine structure of neutral single and multiple excitons (bi-exciton, triple-exciton, quantra-exciton) in the micro-photoluminescence (μ-PL) spectrum of an isolated un doped CdTe/CdTexSe1-x core/shell CQD, with an extremely thin shell (~ monolayer). The CdTe/CdTexSe1-x CQDs exhibit a quasi type-II electronic alignment, showed in the past a reduced Auger rate, a unique spectral stability with a blinking free behavior, viz., they are the most suitable CQD for the current study. Mn-doped CdTe/CdTexSe1-x CQDs were prepared in a control manner, positioning the dopant either precisely either at the core or at the shell regime. The multiple excitons in undoped/doped CQDs were generated by a sequential filling of the states with an increase of excitation power, when recorded at 4.2 K. The fine structure was resolved by following the linear or circular polarization components detected at various angles with respect to a laboratory frame. Representative spectra of an undoped CQD with diameter of 4 nm, recorded at various excitation powers, are shown in Figure 1. The exciton notations follow the diagram given in Panel B of the figure. Panel C approves the spectral stability, while Panel D guarantees detection of a single CQD, using photon autocorrelation measurement. The observations revealed the following information: (a) determination of the confinement geometry and its influence on a single and multiple excitons' fine structure; (b) polarization nature (circular, linear or elliptical) of the multiple exciton recombination processes; (c) the direct and indirect (exchange) Coulomb interactions in CdTe based CQDs; (c) additional fine structure split in doped CQDs by the Mn unpaired spins, (d) theoretical evaluation of the excitons' binding energy, exchange interaction energy and Mn-exciton interaction.

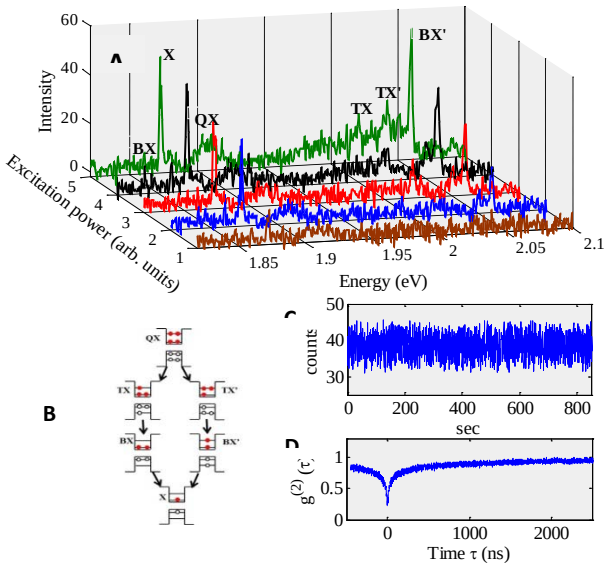

Figure 1: (A) μ-PL spectra of a single CdTe/CdTe$_x$Se$_{1-x}$ CQD recorded at various laser excitation powers, varying from 0.1μW – 10 μW; (B) Schematic drawing of the electronic configuretion of a single exciton X, bi-excitons BX and BX', triple excitons TX and TX'and quatra-exciton. The most probable electron-hole recombination routes are designateded by the black arrows. The appearance of the various excitons in the μ-PL spectra are labeled in Panel (A). (C) Plot of the fluorescence intensity trace vesus time, suggesting a non blinking behavior; (D) Plot of the *cw*