SINGLE- AND MULTI-EXCITONS IN SEMICONDUCTOR NANOCRYSTAL QUANTUM DOTS

Prof. Efrat Lifshitz, Schulich Faculty of Chemistry, Russell Berrie Nanotechnology Institute, and Solid State Institute, Technion, Haifa, 32000, Israel

Multiexciton generation and recombination in colloidal semiconductor nanocrystals quantum dots (NQDs) have been the subject of a special scientific and technological interest in recent years due to their importance in gain devices, photovoltaic cells and single photon light sources. However, multi-excitons in core NQDs are "squeezed" into a volume comparable to a single bulk exciton, and experience low dielectric screening by the insulating surrounding ligands. Under those conditions, strong exciton-exciton (X-X) attractive Coulomb interaction takes place, leading to the so-called Auger relaxation. This process stimulates quenching of the multi-exciton radiation within a few ps, inhibiting their detection in time-integrated cw-PL experiments, and generates luminescence intermittency (blinking) in a single NQD detection. Alternatively, NQDs hetero-structures, characterized by the inclusion of additional semiconductor epitaxial layer between the core and the surfactants (named core-shell NQDs), may be pre-engineered to facilitate a unique condition that would control the Auger process and extend the multi-excitons' lifetime to the ns scale. Here we show, for the first time, well-resolved tetraexciton, triexciton, biexciton and a single exciton bands in the micro-photoluminescence spectrum of a blinking-free (90%) single CdTe/CdSe core-shell semiconductor nanocrystal quantum dot. The multiexcitons were generated by a continuous-wave laser with $\hbar\omega<2E_g$, sequentially filling the *s*-shell and *p*-shell. The *s*-shell (*p*-shell) recombination emission was red (blue)-shifted with respect to that of a single-exciton, and showed additional splitting in the presence of an external magnetic field. Representative example of the shell filling with the increase of the laser power is giving in the Figure below. The blinking free condition was achieved by a structural and dielectric matching and a partial carriers' delocalization at the core-shell interface. This investigation supplied information that was previously obscured in ensemble measurements, with tremendous benefit for future use in NQDs-based technologies.

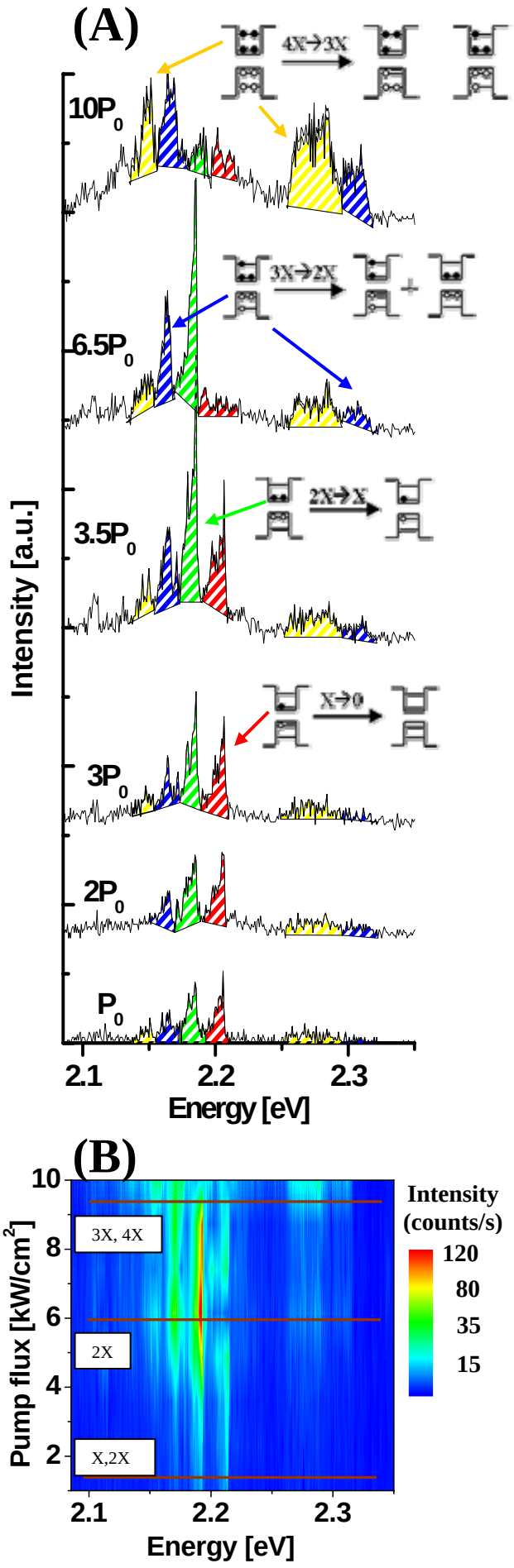

Figure: A sequence of μ-PL spectra of a single CdTe/CdSe core-shell NQD, excited with a variable laser power (when P_0 = 10 μW). The spectra consist of 4-6 bands, shaded in different colours, and their corresponding recombination transitions are drawn schematically in the adjacent diagrams (notations are explained in the text). **Inset:** A contour plot of the μ-PL intensity (see side ruler) versus the emission energy and the laser power, *P*.