Exciton Escape in CdSe Core-Shell Quantum Dots: Implications for the Development of Nanocrystal Solar Cells

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The demonstration of inexpensive growth of CdSe quantum dots (QD's) and their subsequent size and shape control [1] opened new opportunities for the development of low cost and high efficiency QD solar cells. It was recently proposed that the absorption of a single photon in CdSe QD's might generate one or more electron-hole (e-h) pairs through direct carrier multiplication processes. It was suggested that such processes might raise the upper limit of energy conversion up to 66% [2,3]. The device functionality depends on the escape of the generated e-h pairs through quantum tunneling across the shell layer in order to produce currents in the external circuit. However, the time scale of the out-tunneling process of the e-h pairs must be faster than recombination processes. By disregarding the complicated atomic structure at the QD interface, there are two obstacles affecting the out-tunneling of e-h pairs: the shell barrier and coulomb interaction.

In this work, we investigate the tunneling dynamics of e-h pairs in CdSe core-shell QD's aiming the determination of the best conditions (shell material and thickness, and QD sizes) for optimal energy conversion. Our theoretical model is based on the solution of the time-dependent two-particle hamiltonian described within the effective mass approximation. Our results indicate that the tunneling lifetime of a 10 nm wide QD with 0.9 nm shell thickness is approximately 0.01 ns and 1.0 ns for CdS and ZnS as shell material, respectively. This difference of two orders of magnitude is explained by the confinement barrier heights. Variations of the shell thickness in the range 0.3-1.5 nm modifies the tunneling lifetimes by one (three) order(s) of magnitude for CdS (ZnSe) shell layers regardless the QD size and external electric fields. By comparing these data with the recently measured recombination lifetimes in CdSe QD's of few nanoseconds, we conclude that the efficiency of QD solar cells depends on very restrict growth control in order to obtain the appropriate QD sizes and shell thicknesses.

[1] X. Peng *et al.*, Nature **404**, 59 (2000).
[2] M. Califano, A. Zunger and A. Franceschetti, Nano Lett. **4**, 525 (2004).
[3] M. Califano, A. Zunger and A. Franceschetti, Appl. Phys. Lett. **84**, 2409 (2004).