Increased MEG Efficacy via Large Bi-exciton Binding Energy in CdSe/CdTe Type II Core/Shell QDs

S. Tomić[1]*, E. J. Tyrrell[1], J. Miloszewski[1]
[1]University of Salford, Joule Physics Laboratory, Manchester M5 4WT, UK,
s.tomic@salford.ac.uk
* Corresponding Author

Keywords: *MEG, bi-exciton binding, colloidal quantum dots, correlations, type II.*

Abstract

Theoretical predictions indicate that multi-exciton generation (MEG) has the potential to enhance the efficiency of a single gap cell from 33% to 42% [1]. Full realization of this potential requires that the energy threshold for MEG be minimized. An attractive interaction between excitons reduces the threshold by the biexciton binding energy, Bxx, but this has been found to be small (-10 meV) for type I QDs [2]. Previous calculations of Bxx in type II CdSe/CdTe QDs have found a large repulsion between excitons [3]. Here, we show that, by taking into account CI, combinations of core diameter and shell thickness can be found for a CdSe/CdTe core/shell QD that result in large values of Bxx<0.

Our theoretical methodology is based on an 14-band **k.p** Hamiltonian, with correct atomistic symmetry, C2v, of the zinc-blend material [4]. Excitonic states were found using the full CI method, that incudes explicitly the effects of Coulomb interaction, exact exchange and correlations between many-electron configurations. Particular attention was paid to accurate modeling of the dielectric constant variation through the structure as well as surface polarization effects on core/shell and shell/solvent interfaces, obtained from ab initio time-dependent density functional theory [5].

We conclude that: (i) it is not possible to predict biexciton binding using the Hartree approximation alone; it can only be predicted with a full CI Hamiltonian [6]; (ii) CI predicts Bxx = -70 meV for structures with 0.5 nm thick shell that agrees with experiment [7]; (iii) by ignoring the dielectric confinement, it is not possible to predict biexciton binding for structures with shell thickness > 0.75 nm; (iv) by changing the solvent dielectric constant from 1 to 2 the variation in the Bxx binding energy is as big as 100 meV; (v) a proper calculation of Bxx requires the inclusion of correlations and surface polarization effects but the effect of self-polarization is negligible. The strong biexciton binding found is explained by the stronger reduction in the Columbic repulsion between holes than reduction in the attraction between electrons and holes upon the addition of the CdTe shell layer. The Aufbau principle and Hund rule reveal this to be a consequence of 4 fold degeneracy of the h-ground state imposed by symmetry of the structure.

[1] V.I. Klimov, Appl Phys Lett **89**, 123118 (2006)
[2] R. D. Schaller, J. M. Pietryga, & V. I. Klimov, Nano letters, 7, 3469 (2007)
[3] A. Piryatinski at al, Nano Lett **7**, 108 (2007)
[4] S Tomić and N Vukmirović J. Appl Phys **110**, 053710 (2011)
[5] L. Bernasconi, S. Tomić et al, Phys Rev B **83**, 195325 (2011)
[6] E.J. Tyrrell and S. Tomić, J. Phys. Chem. C (2015) (accepted)
[7] D. Binks, S. Tomić, et al, (in preparation)