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Effect of Shell and Shell Thickness on Photoluminescence (PL) of a CdSe/ZnS Core – Shell Quantum Dot

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Abstract. The Photoluminescence (PL) intensity is theoretically calculated for a CdSe/ZnS Core – Shell Quantum Dot incorporating WKB approximation. Coating CdSe bare quantum dots with a ZnS layer indicates an enhancement in the PL intensity remarkably with a red-shift in spectra. The PL intensity increases with initial shell growth and then it starts to decrease if outer shell growths continue. Smaller Core – Shell Quantum Dots shows greater PL intensity with more red-shift in PL spectra.

1. Introduction

Modern technological developments have made it possible to prepare three dimensionally confined quantum dot (QD) structure. Self assembled QDs, colloidal QDs are examples of it. In each case the surface to volume ratio of the number of atoms in QDs is very large as compared to their bulk counterpart. The dangling bonds of the surface atoms in QDs can be passivated by either using suitable organic capping or by deposition of inorganic material of appropriate band gap energies and lattice matching on it. Deposition of an additional layer of inorganic material on QDs is called as Shell and such Quantum Dots are popularly known as Core – Shell QDs (CSQDs). CSQDs find variety of applications ranging from biological tagging to single photon emitters, entangled Q-bit registers etc. Some basic linear optical properties of QDs e.g. appearance of discrete absorption spectra reveal the evidence of excitonic state in a QD. Due to large confinement effect the separation of optically induced electron-hole pairs in QDs becomes appreciably small and gives rise to stronger Coulombic interaction as compared to bulk. In small QDs the confinement dominates over the Coulombic interaction energy resulting into the blue shifting of electron-hole states. Embedding the core of a QD in a medium of different dielectric constant can have profound effect on its electronic and optical properties. In CSQDs the electron and hole wave functions may penetrate in the shell region. The extent of penetration depends on the properties of the core and shell material. The electrons (holes) experience a barrier potential equivalent to the conduction (valence) band offset of a core and shell. A proper choice of core and shell material gives an additional parameter to control the electronic and optical properties of CSQDs. In general, deposition of shell causes red-shift in PL peak and improves the PL quantum efficiency due to the proper passivation of surface dangling bonds and nonradiative recombination sites with strong confinement of electrons and holes inside the core [1]. The basic requirements to determine the PL intensity are the excitation mechanism which can generate population in various excited states of the system and radiative as well as nonradiative recombination.
 processes. In this paper, we have theoretically investigated the linear optical properties like PL intensity of two different CSQDs by taking into account the temperature dependence of energy levels dephasing mechanisms and exciton-phonon interaction.

2. Theoretical Formulation:
We consider a type – I spherical core shell quantum dot with the band gap energy of the core material being smaller than that of the shell material. The geometry and the dimensions of the dot is illustrated in Fig.(1). Here, a is the radius of the core and b is the radius of the CSQD as a whole such that the annular shell thickness is \( d = (b-a) \). The two-dimensional electron and hole confinement potentials are given by [2, 3]

\[
V_{e,h}(r) = \frac{V_{c}}{a^2} \left( r^2 - a^2 \right)
\]

(1)

\( r \) being the quasi-particle position satisfying the condition \( a < r < b \). The subscripts \( e, h, c \) and \( v \) denote the electron, hole, conduction band and valence band, respectively. \( V_c \) and \( V_v \) are the conduction and valence band offsets between the core and the shell. The single particle wave functions under such situation can be described using WKB approx. as [2, 3, 4]
In order to describe the optical transitions in a three level system, we use the density matrix approach. It is appropriate to describe the optical properties of the quantum dot by taking into account the optical transitions among the ground, excitonic and biexcitonic states. The direct ground to biexciton state transitions is considered to be optically forbidden. The transition dipole moment operators corresponding to the transitions between the exciton to ground states ($\mu_{\text{ex}}$) and biexciton to exciton states ($\mu_{\text{exb}}$) are defined as [2, 5]

$$
\phi_i = \begin{cases} 
\frac{A_i}{\sqrt{\kappa_i}} \exp \left[ - \int_{-a}^{x} k_i dx \right] & ; \quad -b < x < -a \\
\frac{A_i}{\sqrt{\kappa_i}} \sin \left[ \int_{-a}^{x} k_i dx + \frac{\pi}{4} \right] & ; \quad -a < x < a \\
\frac{A_i}{\sqrt{\kappa_i}} \exp \left[ - \int_{x}^{b} k_i dx \right] & ; \quad a < x < b 
\end{cases}
$$

(2)

$$
\mu_{\text{ex}}(a, b) = \frac{e p_{\text{ex}}}{\hbar \omega_{\text{ex}}} \left| \phi_i^*(a, b) \phi_i(a, b) \right| \quad (3)
$$

and

$$
\mu_{\text{exb}}(a, b) = -\frac{1}{2} \frac{e p_{\text{exb}}}{\hbar \omega_{\text{exb}}} \left| \phi_i^*(a, b) \phi_i(a, b) \right| \quad (4)
$$

Here, ($\omega_{\text{ex}}$) and ($\omega_{\text{exb}}$) are the transition frequencies corresponding to exciton and biexciton levels. They are temperature sensitive and taken as described in [2, 6]. $p_{\text{ex}}$ is the interband transition momentum matrix element [7]. Using the density matrix formulation described in [3, 7] we have calculated theoretically the PL intensity for a CdSe/ZnS CSQD.

3. Conclusion

Photoluminescence (PL) spectra of CdSe/ZnS Core – shell quantum dot with varying ZnS shell thickness are presented in Fig. 2. The PL intensity arises due to the radiative decay from the exciton state and is proportional to the vacancy in the ground state and the occupation in excitonic state [2]. Following the similar theoretical formulation as described in [2, 3, 6] we calculated the PL intensity for two different CdSe/ZnS CSQDs having core radius 2 nm and 3 nm. The shell thickness (d) varies from 0.2a to 0.6a. Here in these two cases we have changed the core radius (a) keeping the core radius to shell thickness ratio (i.e. a: d) same. We find that for smaller core radius as the external shell growth starts, the PL intensity increases and also shows a red-shifted nature. For a shell thickness of 0.6a the intensity starts to decrease. For a bigger core size (a = 3 nm) the PL intensity increases for 0.2a thick ZnS shell layer and starts to decrease if we put another layer of same thickness (d = 0.4a). The red-shift in the PL spectra is also smaller than the first case as one can see from Fig. 3. The reason of decreasing PL intensity due to a thick external shell is described below.
In small CSQDs, there is greater possibility that the carrier particles can tunnel and spread over the shell region also, which is the main reason behind the red-shift in PL spectra. Again in case of a CdSe/ZnS CSQD, primarily due to the shell layer, the carrier particles experience a strong confinement potential which causes a huge electron-hole Coulomb interaction. First-principle study
also exhibit large electron-hole Coulomb interaction in different CSQDs due to strong confinement [8]. The localization of carrier wave functions in core region is a major factor responsible to increase the PL intensity. The reduction in the PL quantum efficiency is observed as shell thickness is increased further. The reason behind the decrease in PL intensity is due to the lattice mismatch and other radiation less relaxation processes which become important when a thick ZnS shell is grown around the CdSe core. To conclude, we can sum up that, the electron and hole wave functions get modified inside the core due to the presence of shell in conventional semiconductor core-shell QD. WKB approximation method is more appropriate to select the exact wave function for the electrons and holes. The knowledge of the effect of ZnS shell formation on CdSe QD will help to fabricate various optoelectronic devices.

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References