Physica B 466-467 (2015) 101–106

Contents lists available at ScienceDirect

Physica B

journal homepage: www.elsevier.com/locate/physb

Optical properties of type-I PbSe/CdSe core/shell quantum dot

S.N. Saravanamoorthy [a], A. John Peter [b,*], Chang Woo Lee [c,1]

[a] Department of Physics, Devanga Arts College, Aruppukottai 626101, Virudhunagar, India
[b] PG and Research Department of Physics, Government Arts College, Melur 625106, Madurai, India
[c] Department of Chemical Engineering, College of Engineering, Kyung Hee University, 1732 Deogyeong-daero, Gihung, Yongin, Gyeonggi 446-701, South Korea

ARTICLE INFO

Article history:
Received 11 January 2015
Received in revised form
1 April 2015
Available online 8 April 2015

Keywords:
Exciton
Binding energy
Oscillator strength
Recombination rate
Absorption coefficient
Core/Shell quantum dot

ABSTRACT

Electronic properties and optical properties of exciton in a PbSe/CdSe core/shell quantum dot are investigated taking into account the spatial confinement effect. The present model is based on the PbSe/CdSe quantum dot for type-I confinement regime. The dielectric mismatch effect and the self polarization potential are taken into consideration in the PbSe/CdSe quantum dot nanostructure. Polarization charges are incorporated at the interface of the core/shell materials. Numerical calculations on the electronic and optical properties are found with the ratio of radius of inner and outer shell materials for various shell radii. The exciton binding energy and the interband optical transition energies are computed using variational formulism within the single band effective mass approximation. The oscillator strength and the recombination life time are determined with the ratio of radius of core to shell materials taking into account the dielectric mismatch between the materials. The nonlinear absorption coefficients and the changes of refractive index are computed for the ground and first excited state using compact density matrix method. The obtained results are found to be in good agreement with those reported by other investigators.

1. Introduction

Lead chalcogenide IV-VI semiconducting materials are given attention due to their potential applications in opto-electronic devices such as tunable mid-infrared diode lasers. The optical gain and threshold properties are considered to be important parameters for the design to achieve high efficiency lasers [1] and the wave guiding devices [2]. These materials show large nonlinearity and the fast response time (ps) which can be used in optical signal switches [3]. PbS/PbSe quantum dots are promising materials for photovoltaic solar cells for probing infrared spectrum [4–6], they exhibit suitable spectral emission which is used for optical communication wavelength [7] and biological markers [8,9]. They are the excellent candidates for the potential applications in the near-infrared region wavelengths which can be tailored from 1 μm to beyond 2 μm. These materials show strong nonlinear optical properties displaying higher refractive index (nearly six orders) than the conventional Si and GaAs materials. Hence, they can be used for all optical signal processing systems [10,11]. The other important material parameters are lattice constants, mobilities, carrier concentrations, composition with the inner and outer materials in a heterostructure, band offsets, temperature dependent band gap, energy dependent effective masses, larger dielectric constants and energy-dependent refractive indices.

The bulk effective exciton Bohr radius of PbSe (409 Å) is larger than the typical nanocrystal size and this property leads to a strong quantum confinement. This effect strongly influences the electronic and optical properties. The above parameters are required to design any nano-device for the optimum qualities with the maximum quantum efficiency. The electronic structure of any semiconducting material in a heterostructure is determined by the effective mass, dielectric constant and the barrier height [12]. A quantum dot in which all the three dimensions are confined in the nanometer ranges in which the quantum confinement effect dominates. The charge carriers confined in the quantum dot exhibit exotic behavior. They show size dependent electronic and optical properties. Narrow quantum dots show higher photon energies more than the wider dots with the same content. Core/shell is an example for a quantum dot. Core is made up of a semiconducting material (PbSe) which is coated with the lattice matched shell material (CdSe) in order to improve its optical properties. These core/shell materials show novel optical properties better than the bare quantum dots [13–15]. PbSe related colloidal core/shell heterostructures have been investigated for optoelectronic applications [16]. Nonlinear optical properties associated with the intersubband transitions in some core/shell quantum dots have been investigated with the influence of the

* Corresponding author. Fax: +91 4522415467.
E-mail address: a.john.peter@gmail.com (A. John Peter).
[1] Co-corresponding author.

http://dx.doi.org/10.1016/j.physb.2015.04.005
0921-4526/

shell thickness, impurity, and dielectric environment [17,18].

In the present work, numerical calculations on exciton binding energies and some nonlinear optical properties of exciton in a PbSe/CdSe core/shell quantum dot are discussed taking into account the geometrical confinement effect. The computations are done with the various shell radii and the different ratio of radii of inner core to the shell materials. The effect of dielectric mismatch is included between the core and shell material. The exciton binding energy and the interband optical transition energies are found using variational approach with the single band effective mass approximation. The oscillator strength and the radiative transition life time are obtained taking into consideration of effects of geometrical confinement and the dielectric mismatch. The nonlinear absorption coefficients and the changes of refractive index are studied for the ground and first excited state using compact density matrix method. The paper is organized as follows. In Section 2, the theoretical model of obtained eigen functions and eigen energies of lowest binding energies and the related nonlinear optical properties are explained. The results and discussion are presented in Section 3. The summary of the obtained results are briefed in the last Section.

1.1. Theoretical framework

The present system consists of a core (PbSe) with the inner radius, R_C and the dielectric constant (ε_c) embedded on an outer shell (CdSe) material having radius, R_S and the dielectric constant (ε_s). The two particle Hamiltonian, within the single band effective mass approximation, is given by

$$H = \sum_{i=e,h}\left[-\frac{\hbar^2}{2m_i^*}\nabla^2 + V_i(r_i)\right] - U(r_e, r_h) + W(r) \tag{1}$$

where $i=e$, h refer the carrier choice, $V_i(r_i)$ is the confinement potential of each section. The first term represents the kinetic energy operator. $U(r_e, r_h)$ is the potential energy term with the inclusion of polarization charges induced at the boundary of the core/shell quantum dot. $W(r)$ is the self energy term. m_i^* is the effective mass of the charge carrier (electron and hole) which is given by

$$m_{e(h)}^* = \begin{cases} m_{e(h)(core)}^* & r < R_c \\ m_{e(h)(shell)}^* & r > R_s \end{cases} \tag{2}$$

where R_c and R_s are the radius of PbSe inner core and the outer CdSe shell material respectively. $m_{e(h)(core)}^*$ and $m_{e(h)(shell)}^*$ are the effective masses of core and shell material respectively. The Coulomb potential energy with the inclusion of polarization of charges is given by

$$U(r_e, r_h) = -\frac{e^2}{\varepsilon\left|\vec{r_e} - \vec{r_h}\right|} - \frac{e^2(\varepsilon_c - \varepsilon_s)}{\varepsilon_c}\sum_{l=0}^{\infty}\frac{r_e^l r_h^l}{R^{2l}}\frac{l+1}{l\varepsilon_c + (l+1)\varepsilon_s}P_l(\cos\theta) \tag{3}$$

where $P_l(\cos\theta)$ is the Legendre polynomial and $P_l(\cos\theta)$ is the angle between $\vec{r_e}$ and $\vec{r_h}$. The dielectric constant of the inner core material is greater than the shell material. It follows as

$$\varepsilon(r_e, r_h) = \begin{cases} \varepsilon_c & r < R_c \\ \dfrac{\varepsilon_s(r^3 - R_s^3) + \varepsilon_s(R_s^3 - R_c^3) + \varepsilon_c R_c^3}{R_s^3} & r > R_s \end{cases} \tag{4}$$

where ε_c and ε_s are the dielectric constants of core and the shell materials respectively. The effects of the polarization charges induced on the quantum dot arise due to the dielectric mismatch

Table 1
Material parameters* used in the calculations.

Parameter	PbSe	CdSe
E_g (eV)	1.137	1.572
m_e^* (m_0)	0.07	0.13
m_h^* (m_0)	0.06	0.45
ε	25	9.5
C_{11}(GPa)	123	74.1
C_{12}(GPa)	19.3	45.2
C_{44}(GPa)	15.9	13.4
C_{13} (GPa)	12.31	39
C_{33} (GPa)	96.37	84.3
e_{31} (C/m^2)	21.2	0.347
e_{31} (C/m^2)	17.3	–0.16
P_{SP} (C/m^2)	0.29	0.006
a (nm)	0.6126 0	4299

* Parameters taken from Ref. 32.

[18]. The values of dielectric constants are appended in Table1.

The self interaction potential, $W(r)$, comes out from the interaction between the exciton and its image charge. This arises due to the dielectric mismatch between the inner core and the shell material. This polarization energy depends on the relative distance between the exciton and the center of the system. Thus, the self polarization potential is given by [19,20]

$$W(r) = \frac{e^2(\varepsilon_c - \varepsilon_s)}{2\pi\varepsilon_c R_c}\sum_{k=0}^{\infty}\frac{k+1}{k\varepsilon_c + (k+1)\varepsilon_s}\frac{r^{2k}}{R_c^{2k}} \tag{5}$$

The barrier potentials without the strain effect, $V_{e(h)}(z)$, related to the band offsets in the PbSe/CdSe structure are given by

$$V_{e(h)}(z) = \begin{cases} V_{e(h)}^0 & r < R_c \\ V_{e(h)}^0 & r > R_s \end{cases} \tag{6}$$

where $V_{e(h)}$ is the barrier height of conduction (valence) band. 70% contribution to the conduction band and 30% to the valence band between PbSe/CdSe materials are assumed in the calculations.

The calculations of the strain effects are included through the confinement potential barriers. In fact, the lattice mismatch between the inner core and outer shell materials is incorporated. And hence, the total confinement potential in the conduction band with the inclusion of strain effects, between the core and shell material, is expressed as [21]

$$V_e = V_{e(h)}^0 + a_c(\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}) \tag{7}$$

where a_C is the conduction band hydrostatic deformation potential and $V_{e(h)}^0$ is the barrier potentials of conduction and valence bands without the strain inclusion which have been denoted in Eq.(6). The confinement potential in the valence band follows as

$$V_h = V_h^0 - a_v(\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}) + b(\varepsilon_{zz} - \varepsilon_{xx}) \tag{8}$$

where a_v is the valence band hydrostatic potential and b is the shear deformation potential. The heavy hole has been involved in the numerical computations since many experimental results use heavy hole excitons. The components in strain tensor equations are expressed as

$$\varepsilon_{xx} = \varepsilon_{yy} = \frac{a_c - a_s}{a_c} \tag{9}$$

where a_c is the lattice constant of inner core material and a_s is the lattice constant of shell material. We assume that the present work is oriented along growth direction as