

Mechanisms of Electroluminescence in Silicon Nanostructures

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Abstract The mechanism of electroluminescence (EL) in silicon nanostructure (NC) is investigated. The influence of quantum confinement effect (QCE) on the radiative recombination rate and the radiative recombination time is calculated. We found that the radiative recombination rate depends on the diameter d of the spherical crystallites and the radiative recombination rate decreases with decreasing size of the Si-QDs. As a result, the EL emission intensity enhanced due to QCE.

Keywords Electroluminescence, Quantum confinement effect, Radiative recombination, Quantum dots

1. Introduction

Semiconductor quantum dots (QDs) are the leading materials regarding high-density electronic devices. QDs are nanocrystals with nanometric size (ranging in size from 2 to 10 nm in diameter) provide a nearly zero-dimensional system, where carriers are confined in all spatial directions [1]. They have attracted much attention due to their applications in light emitting diodes (LED), biological imaging etc [2, 3]. Silicon nanostructure (Si-ns) is one of the candidates for photonic material. The reduced size to nanoscale results in new quantum phenomena that yield some modification in material properties because of quantum confinement effects of electrons and holes in the material [4].

Electroluminescence (EL) is a phenomenon of light emitting in certain materials such as semiconductors due to an electric field or passage of an electric current through the semiconductor. It occurs due to radiative recombination of excited electrons with holes and as a result photon are released. EL is an optical property which depends on the nature of the material. In silicon quantum dots the EL depends on the size and the EL intensity is higher for smaller size [5]. The optical properties of silicon quantum dots specially the EL is affected by different factors such as the preparation methods and conditions (laser ablation, deposition temperature, annealing, and oxidation process etc) [6]. In addition to these factors, the QCE is the one which affects the optical phenomena in nano range systems that can changes the EL spectra [4].

One of the important parameter that determines the optical

and transport properties of the of QDs is the energy gap between the highest occupied and lowest unoccupied molecular orbitals (HOMO-LUMO) levels. In EL phenomena carriers-excitation is due to the applied electric field. When an electron in the HOMO gets enough energy, it can jump to the LUMO and the emission happens when the charge carriers (electrons and holes) recombine. The electron-hole recombination time (life time of exciton) is strongly dependent on the particle size [7].

Recently, there has been great interest in developing quantum dot (QD) light emitting devices (LED) since the work of Colvin et al [8] for the application of displays with quality images and low power consumption. The fabrication of QD-LED based display by Kim et al proves the feasibility of this type of display [9]. Due to their high photoluminescence quantum yields, QD-LED also used for solid state lighting applications. The fabrication of different colored QD-LED by Anikeeva et al demonstrated the possibility of EL in the visible wavelength range [10].

In this paper, we present a theoretical model for the calculation the radiative recombination rate and life time for Si-QDs EL phenomena. The second section discusses the model of optical transition and formulation of the problem. The results of our calculation are presented and discussed in section three and section four gives the conclusion.

2. Model of the System

In order to formulate and describe the EL phenomena from the QD structures, we consider the QDs as ensembles of nanometric size spherical particles, having a well-defined size distribution. We assume both excitation and emission process for electrons takes place inside the QDs. The QDs system will have a quasi-direct band structure owing to the quantum confinement effect (QCE) and the valance and conduction bands are represented as the HOMO and LUMO

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Published online at <http://journal.sapub.org/ajcmp>

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respectively (fig. 1).

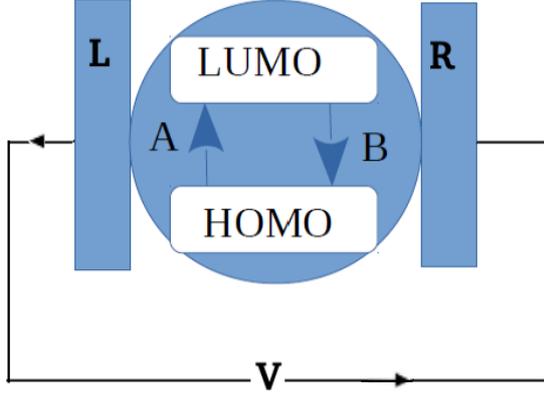


Figure 1. Direct transition between the HOMO and LUMO states of the QD. high energy electrons are generated and excited to the LUMO state and relaxed (path A). The relaxed carriers recombine radiatively to ground states and giving PL (path B). L and R represents the left and right contact leads to the QDs

Electronic transition between the HOMO - LUMO is induced due to the application of external electric field. Since electric field is an electromagnetic field (EMF), it is assumed to be of the form

$$E(t) = \Re[E_o(e^{-i\omega t} + e^{i\omega t})] \quad (1)$$

We begin by considering a Hamiltonian which describe the interaction of the electric field with the electronic state. To describe the electric field, we introduce a vector potential in the electric dipole approximation of the form $A(t) = A_o \hat{\theta}(e^{-i\omega t} + e^{i\omega t})$ is the polarization unit vector. Then Hamiltonian reads as:

$$H = \frac{1}{2m} \left(p - \frac{e}{c} A \right)^2 + \phi(r), \quad (2)$$

where e is the charge of an electron and c is the speed of light. Because of gauge in variance, the choice of these potentials is not unique. For simplicity, we choose the Coulomb gauge in which $\nabla \cdot A$ and $\phi = 0$.

Since the operator p as an operator does not commute with A and for the purpose of calculating linear optical properties we neglect the term $[e^2 A^2 / 2mc^2]$ from the expansion of $[(p - e/c)^2]$. Considering A as a classical vector, the Hamiltonian can be written as

$$H = H_o + H_I \quad (3)$$

where $H_o = p^2 / 2m$ is the unperturbed Hamiltonian and the interaction Hamiltonian H_I is given by

$$H_I = - \left(\frac{e}{mc} \right) A \cdot p. \quad (4)$$

We first assume that the vector potential A is weak enough that we can apply time-dependent perturbation theory (in the form of the Fermi Golden Rule) to calculate the transition probability per unit volume for an electron in the HOMO state $|h\rangle$ with energy E_h and wavevector k_h to the LUMO state $|l\rangle$ with corresponding energy E_l and wavevector k_l . We also assume that the system at $t = 0$ is in its ground state and the transition between HOMO-LUMO state of N identical atoms is described by the Bloch wave functions of

the form

$$|h\rangle = U_{kh}(r) e^{ik_h \cdot r}, \quad (5)$$

$$|l\rangle = U_{kl}(r) e^{ik_l \cdot r}. \quad (6)$$

where the indices h and l indicate the HOMO and LUMO states respectively.

The matrix element (H_{hl}) of the perturbation H_I is given by

$$H_{hl} = \langle l | H_I | h \rangle = \left(\frac{-ie\hbar}{mc} \right) \langle l | A \cdot \nabla | h \rangle. \quad (7)$$

Using the Bloch wave functions in equation (5) and equation (6), equation (7) can be written in integral form as

$$H_{hl} = \left(\frac{-ie\hbar}{mc\Omega} \right) \left[\int e^{i(k_h - k_l)} U_{kl}^*(r) U_{kh}(r) A \cdot (ik_h) \right] dr - \left(\frac{ie\hbar}{mc\Omega} \right) \left[\int e^{i(k_h - k_l)} U_{kl}^*(r) A \cdot \nabla U_{kh}(r) \right] dr \quad (8)$$

The integral of the first term in equation (8) which contains the term $U_{kl}^*(r) U_{kh}(r)$, vanishes because $U_{kl}(r)$ and $U_{kh}(r)$ are orthogonal and for direct transition $k_h = k_l$ then equation (10) can be written as

$$H_{hl} = \left(\frac{e}{mc} \right) A \cdot \left[\left(\frac{i\hbar}{\Omega} \right) \int U_{kl}^*(r) \nabla U_{kh}(r) dr \right] \quad (9)$$

The term in the square bracket in equation (9) is called the momentum operator (the electric dipole transition element), p_{hl}

$$p_{hl} = \left(\frac{i\hbar}{\Omega} \right) \int U_{kl}^*(r) \nabla U_{kh}(r) dr \quad (10)$$

In terms of the momentum operator, the interaction Hamiltonian matrix element H_{hl} then becomes

$$H_{hl} = - \left(\frac{e}{mc} \right) A \cdot p_{hl} \quad (11)$$

The emission and absorption of energy by charge carriers (electrons/holes) is essentially a scattering phenomenon between initial state $|h\rangle$ and final state $|l\rangle$. The electric field is the time dependent perturbation which induces this event. The radiative transition rate (R) for energy emission and absorption per unit time obtained by substituting equation (11) into the Fermi Golden Rule:

$$R = \frac{2\pi}{\hbar} \sum_{\omega} |H_{hl}|^2 \delta(E_h(\omega) - E_l(\omega) \pm \hbar\omega) \quad (12)$$

The momentum matrix element in equation (10) is not strongly dependent on k so we shall replace it by the constant $|p_{hl}|^2$. Equation (12) can then be simplified to read

$$R = \frac{2\pi}{\hbar} \left(\frac{e}{mc} \right)^2 |A|^2 \sum_{\omega} |p_{hl}|^2 \delta(\hbar\omega_{hl} \pm \hbar\omega) \quad (13)$$

where $\hbar\omega_{hl} = E_h(\omega) - E_l(\omega)$.

The oscillator strength f_{hl} , which is the measure of the radiative probability of a quantum mechanical transition between two atomic levels (HOMO and LUMO), is related to the momentum matrix element as

$$f_{hl}(\omega) = \frac{2}{m\hbar\omega_{hl}} |p_{hl}|^2 \quad (14)$$

Using $E_o = \omega/c A_o$ and substituting equation (14) in to equation (15), the radiative recombination rate becomes

$$R = \frac{\pi e^2 E_o^2}{m\omega_{hl}} f_{hl}(\omega_{hl}) \quad (15)$$

The size dependence of the oscillator strength (f_{hl}) with the crystallite diameter d is usually assumed to be of the form [11]

$$f_{hl}(\omega_{hl}) = \frac{\gamma}{d^\beta}, \quad (16)$$

where γ is arbitrary proportionality constant and the power exponent β depends on the material property as well as range of the crystallite size being used, and the values of β had been taken to be 5 by Sanders et al [12] and 6 by Khurgin et al [13].

Assuming each atom in the quantum dots contains at least one excited carriers due to the applied external electric field, the number of excited carriers (N_c) in the QDs is proportional to its volume. i.e $N_c \sim \Omega$. For spherical QDs of diameter d , the volume is $\Omega = (\pi/6)d^3$. Combining these assumption and facts we arrive at $N_c \sim (\pi/6)d^3$. For every transition in the QD, the oscillator strength is proportional to $1/d^\beta$ (equation 16), so the total oscillator strength over the volume is proportional to N_c/d^β . Therefore, for equation (16) we have

$$f_{hl}(\omega_{hl}) \sim \frac{\pi\gamma}{6} d^{3-\beta}, \quad (17)$$

Using equation (19) into equation (17), the radiative recombination rate R is given by

$$R = \eta \left(\frac{e^2 E_0^2}{m\omega_{hl}} \right) d^{3-\beta} \quad (18)$$

where η is a proportionality constant parameter. The radiative recombination time (τ) is the inverse of the radiative recombination rate (R), $\tau = 1/R$.

3. Results and Discussion

In this section, we present the main results that we obtained based on model calculation in section 2. Various parameters are considered here. The dot size is taken in the range 1nm-10nm. The parameter β is taken in the range 5-6. It is known that the band gap Si-QD is size dependent. It is reported recently that the band gap of Si QD ensemble, follows a fit with equation, $E_g = 1.13 + 13.9/d^2$, where d is the dots diameter [14]. Even if the band gap depends on size for our numerical calculation we use $E_g = 2.75\text{eV}$ [15]. The applied electric field E_0 is taken to be 10^3 V/cm .

As can be seen from figure 2, the radiative recombination rate (R) of silicon quantum dots increases as the size of Si-QDs decreases and we also observe that R increases with values of β . To obtain an insight of the effect of size on the radiative recombination rates and hence on the EL spectral profile of the system, both excitation and emission processes for electron hole pairs occur in side the Si-QDs are considered. The EL emission in visible range is observed when optical transitions between HOMO-LUMO gaps are induced by the electric field.

If the radiative recombination rate is relatively high, a pair comprised an excited electron and hole can recombine via radiative emission process. On the other hand, for low radiative recombination rate case the non-radiative

recombination events degrade the performance of light emission. The QCE changes the energy level spectrum to a discrete level structure so that the radiative recombination rate is enhanced. Our result is in good agreement with experimental and theoretical findings [7].

The size dependent radiative life time that we obtain has power relation with the diameter of spherical quantum dots. From figure 3 one can observe that the radiative lifetime of Si-QDs decreases rapidly with the decrease of the Si-QDs diameter. As a result, the EL emission spectra shifted to the visible range. In addition, the variation of radiative life time with the values of β indicates the EL depends on the properties of the Si-QDs.

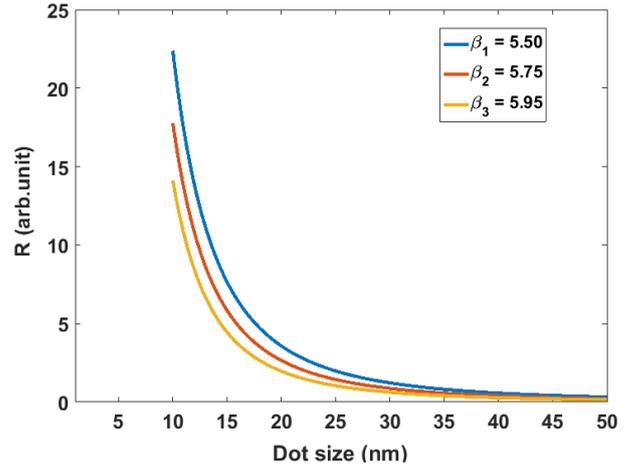


Figure 2. Variation of radiative recombination rate with size of the quantum dot

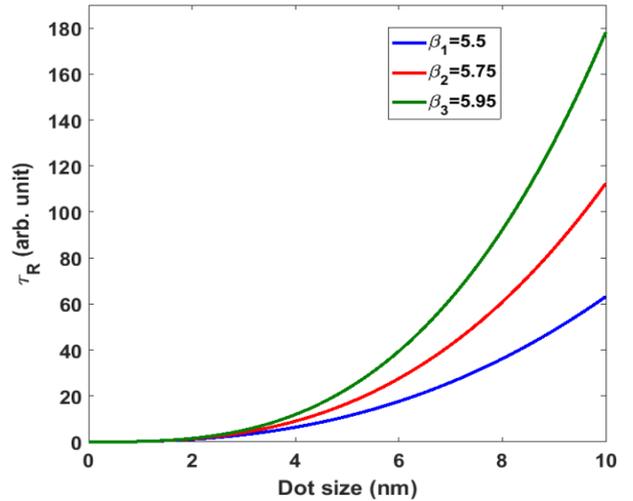


Figure 3. Size dependent radiative life time

4. Conclusions

In conclusion, the effect of the QCE on the EL of Si-QDs were investigated theoretically. Enhanced radiative recombination rate is observed in small-sized Si-QDs. It is also seen that the recombination life time decreases as the Si-QDs size decreases owing to the QCE.

ACKNOWLEDGEMENTS

The author would like to thank Wollo university college of Natural Science.

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