Effect of Quantum Confinement on The Wavelength of CdSe, ZnS And GaAs Quantum Dots (Qds)

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Abstract: The effect of confinement on quantum dots (QDs) of CdSe, ZnS and GaAs on the wavelength has been studied using the Brus equation at various confinement radii. It is observed that the QDs of CdSe and GaAs possess the trait for possible extension of their wavelength to match the IBSC systems for communication in the optical C band.

Index Terms— quantum dots, exciton, Brus, confinement, heterostructures, thinfilms, photoluminescent

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1 INTRODUCTION

With the advent of semiconductor nanostructures and the discovery of their greater physical properties, extensive research has been conducted to make use of these reduced dimensionality structure properties for further applications. The study of low-dimensional semiconductor heterostructures quantum dots (QDs) is one of the main subjects in condensed matter Physics owing to their application to optoelectronic devices like light emitting diodes and lasers [1], [2], solar cells [3], [4]. In particular QDs, haven achieved great confinement that cannot be reached with bulk semiconductors or other nanostructures [5]. Recent report by Ustinov et al, pointed out the improvements made on the quality of lasers produced from the injection of self-organized InAs/GaAs QDs [6]. In his report low-threshold frequencies have been demonstrated using InAs embedded in GaAs quantum well (QW). It has been found that further improvement of the laser characteristics can be achieved by increasing the surface density of QDs, the energy separation between the QD energy levels, the matrix states and improving the uniformity of the QD ensemble. On the contrary, using the quantum dots in the active region of optoelectronic devices allows us to extend the optical emission range toward long wavelengths due to increased localization energy of carriers in quantum dots as compared to QW. Recently the QD of InAS have been shown to reach a wave length between 1.33-1.5 µm [7], [8], [9], for high speed communication systems due to the prediction of a zero band offset for one of the carrier types at the QD/barrier heterojunction. The intermediate band solar cell (IBSC) was achieved using several systems on several substrates for example, Miska et al; was able to realize this using InAs/InP(113)B [7]; while Enzmann et al, was able to optimize the growth rate using several methods on InAs/AlGaInAs lattice matched to InP(001) [8]. Pancholi et al, achieved a similar growth by using InAs/GaAsSb [9]. All of these are achievable with self-organised quantum dots with low densities (≤ 10µm^{-2}). This low density QD could be used for single photon generation in the optical C-band in order to minimize losses in commonly used glass fibre for telecommunications transmission. In this research, the effect of changing the radius (size) of quantum dot on the wavelength of light emitted will be computed theoretically using the Brus equation. Our result will be compared with other low density heterostructures as observed experimentally.

2 THEORETICAL FRAMEWORK

The particle-in-a-box model gives an insight into the behavior of the electrons inside the QDs and also a quantum mechanical description of the size dependent frequency of light emission from QD [10]. The simplest form of the particle-in-a-box model considers a one-dimensional system.

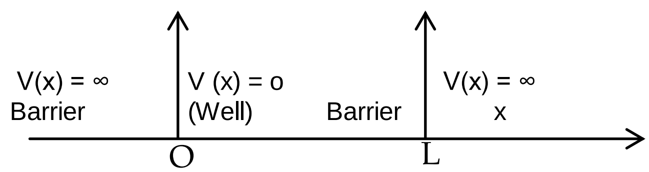

Fig. 2.0: Infinite one-dimensional (potential) square well.

The potential energy in this model is given as.

$$V(x) = \begin{cases} 0, 0 < X < L \\ \infty, otherwise \end{cases} \quad (2.0)$$

Where:

L = length of box

X = position of particle within the box.

Picture a particle such as an electron bouncing around inside a one-dimensional box of length L (Fig. 2.0). However, the “correct” picture in modern physics (quantum mechanics) is that of an electron wave reflecting back and forth from the walls of the box.

The wave function ψ (x,t) can be found by solving Schrödinger equation for the system:

$$\frac{i\hbar\partial\psi(x,t)}{\partial t} = \frac{-\hbar\partial^2\psi(x,t)}{2m\partial x^2} + V(x)\psi(x,t) \quad (2.1)$$

Where: ħ = Reduced (normalized) Planck’s constant, m= Mass of particle, i= Imaginary unit and t= Time

Inside the box, no forces act upon the particle, which means that the part of the wave function inside the box oscillate through space and time with the same form as a free particle.

$$\psi(x,t) = Ae^{i(kx-\omega t)}$$

$$e^{i(kx-\omega t)} = ASin(kx) + BCos(kx) \quad (2.2)$$

Where: A and B = arbitrary complex number, K = wave number and ω = angular frequency. The wave is a probability wave and the amplitude or size of the wave function at a given position is related to the probability of finding a particle there by:

$$P(x,t) = |\Psi(x,t)|^2 \quad (2.3)$$

The waves must have a node at the edges of the box (since electrons cannot escape the box and its probability must varnish at the edge), so only certain wavelengths fits inside. Also, the amplitude of the wave function may not "jump" abruptly from one point to the next [10]. These two conditions are only satisfied by wave functions with the form.

$$\psi_n(x,t) = \begin{cases} \mathrm{ASin(knx)e^{-i\omega t}}, \mathrm{o} < x < L \\ 0, \text{otherwise} \end{cases} \quad (2.4)$$

Where: n = positives whole number, k = wave number
The wave number is restricted to certain, specific values given by:

$$K_n = \frac{n\pi}{L} \quad (25)$$

Where: L= size of the box and n= 1, 2, 3, 4……
K is related to the wavelength by:

$$K_n = \frac{2\pi}{\lambda_n} \quad (2.6)$$

Equating equations (2.5) and (2.6) gives

$$\lambda_n = \frac{2L}{n} \quad (2.7)$$

Equation (2.7) above shows how the wavelength λ of the electron wave function depends on the size of a one-dimensional "box" of length L. Momentum P of the particle can be calculated from its wavelength as follows.

$$p_n = \frac{h}{\lambda_n} = \frac{nh}{2L} \quad (2.8)$$

Finally, the kinetic energy E of each allowed state n for the electron can be computed as:

$$E = \frac{p^2}{2m} = \frac{n^2h^2}{(2L)^2(2m)} \quad (2.9)$$

$$E_n = \frac{n^2h^2}{8mL^2} \quad (2.10)$$

The energy E_n of the electron states varies inversely as the square of the box L^2. Therefore as the box gets smaller, the energy for each state increases. The one-dimensional model of a particle in a box can easily extended to a three dimensional box, which is more relevant to describing the behavior of QDs (confinement in three dimension). In three dimensions, the energy of the particles (electron) in equation (2.2) becomes.

$$E = E_x + E_y + E_z = \frac{h^2}{8m}\left(\frac{n_x^2}{L_x^2} + \frac{n_y^2}{L_y^2} + \frac{n_z^2}{L_z^2}\right) \quad (2.11)$$

Where: x, y, z, = three orthogonal directions, n =energy levels and can take on integer values greater than or equal to one, m=mass of particle (electron) and L_x, L_y, L_z= dimensions of the box in the x, y, and z directions. One important feature of the particle-in-a-box model is "confinement energy" the lowest possible energy for the particle is not zero; (rather, it is: $E = \frac{h^2}{8mL^2}$).

This energy increase with decreasing size of the box. The confinement energy is observed in quantum dots through an increase in the energy of the band gap. The band gap for bulk CdSe at 300k is Eg =1.74ev. The energy of the band gap is greater for CdSe quantum dots. The confinement energy for the quantum dots sample is equal to the band gap energy minus 1.74ev [11]. Another feature of the particle-in-a-box model is that the energy spectrum is discrete rather than continuous. Only certain energies are allowed for the electron. This also happens in quantum dots; the density of states gets peaked at certain energies. This can be observed in the absorption spectrum of quantum dots [11]. In the computation of the emission energy states, the overall Brus equation was used [5].

$$\Delta E(R) = E_g(R) + \frac{h^2}{8R^2}\left(\frac{1}{m_e^*} + \frac{1}{m_h^*}\right) \quad (2.12)$$

Where

ΔE = the emission energy, E_g= band gap energy, R= radius, h= Planck constant m^*_e = effective mass of excited electron and m^*_h = effective mass of excited hole For the assumptions made in arriving at equation (2.12), the reader is referred to a review article on this subject by Chukwuocha et al, [5] and a paper that appears in the journal [12].

3 Results

For the simulation in the confinement regime, we present the values of the parameters used and the results obtained with the dimensionless and physical quantities in this research. With the ground state confinement energy, emission energy and the assumed radius for this present work taken from reference number [5]. This we have used to compute the wavelength as a function of radius for three different semiconductor quantum dots that we have studied. In arriving at the results, several parameters were used, for Cadmium Selenide, Zinc Sulphid and Gallium arsenide as in table 3.1 below.

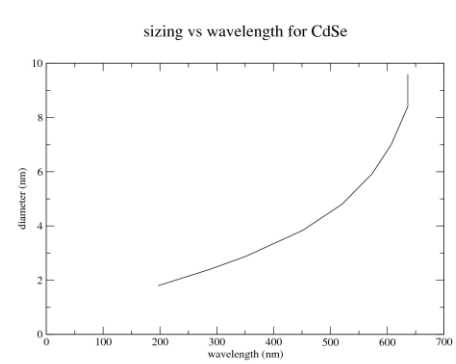

Fig. 3.1 showing the wavelength for CdSe at various confinement radii

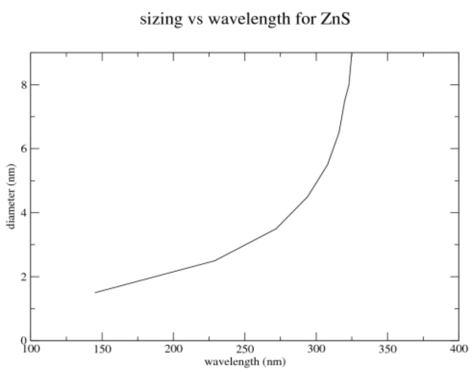

Fig. 3.2 showing the wavelength for ZnS at various confinement radii

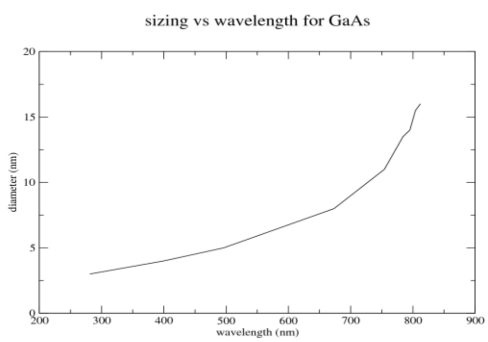

Fig. 3.3 showing the wavelength for GaAs at various confinement radii

4 Discussion of Results

The plots (Figures (3.1), (3.2), and (3.3)) for the three different semiconductors show an exponential dependence of wavelength of light emitted on size of quantum dot. One can conclude that the larger the dot, the redder (lower energy) its fluorescence spectrum would be. Conversely, smaller dots emit bluer (higher energy) light. The coloration is directly related to the energy levels of the quantum dot. Quantitatively speaking, the band gap energy that determines the energy (and hence colour) of the fluorescent light is inversely proportional to the size of the quantum dot. Larger quantum dots have more energy levels which are also more closely spaced. i.e., the energy levels form a near continuum (weak confinement regime). A closely look at the plots shows also that CdSe and GaAs has the potential to reach the desired wavelength in the IBSC systems. We have computed stand alone dots with a view of ascertaining their plausible usage for the IBSC system and the dots of CdSe and GaAs shows a remarkable usage if they were embedded in QW substrate or quantum wire as the case may be. Van Driel showed that the lifetime of fluorescence is determined by the size of the quantum dot [13]. Larger dots have more closely spaced energy levels in which the electron-hole pair can be trapped. Therefore, electron-hole pairs in larger dots live longer causing larger dots to show a longer lifetime. From the experimental observations of Harbold and monica, with transmission electron microscopy, the following results were obtained for average radius of cadmium selenide QDs. This result corresponds to the different colours (wavelength) that make the visible spectrum [11].

TABLE 3.1
MATERIAL PARAMETER USED FOR COMPUTATION

QDs	m^*_e	m^*_h	E_{bulk} at 300k	a_B
CdSe	$0.13m_o$	$0.45m_o$	1.7 eV	6 nm
ZnS	$0.34m_o$	$0.23m_0$	3.68 eV	5nm
GaAs	$0.063m_o$	$0.51m_o$	1.424 eV	10nm

Table 3.1 showing material parameter used for the computation of the confinement energies at various radii which is less than the bohr radius a_B [5].

5 Conclusion

A closely look at the computed wavelength from figures 3.1 to 3.3 show that CdSe and GaAs can be extended to reach the desired wavelength for the IBSC systems. This can be achieved by embedding the QD in a quantum well substrate or the wire substrate. Also from our simple model a slight deviation values corresponding to lower threshold of the wavelength was obtained. This slight deviation between the experimentally observed data from Harbold and Monica and the simple model proposed is attributed to the following:

1. Spherical shape assumption-in reality, quantum dots of shapes such as cones, pyramids, domes, disks, ellipsoids etc. exist.
2. The weak coulombic interactions that were ignored though should be considered at very small radius (size).
3. Ground state was considered in our model, while peak emission energy was considered by Harbold and Monica.
4. The radius given by the transmission electron microscopy (TEM) was an average value.

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TABLE 4.1
SIMULATION RESULTS FOR WAVELENGTH

Radius (nm)	colour	Experimental observed wavelength (nm) [11]	Computed wavelength (nm)
2.15	Green	495-570	488
2.60	Yellow	570-590	542
3.18	Orange	590-620	589
3.44	Red	620-750	605

Table 4.1: showing our computed result for CdSe and the one reported by experimental observations [11].