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LINEAR AND NONLINEAR OPTICAL PROPERTIES OF GaAs QUANTUM DOT

BY

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This is my original work has not been presented for a degree in any other university and that all sources or materials used for the thesis have been duly acknowledged.

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This thesis has been submitted for examination with my approval as University advisor.

Dr. _____

To my Beloved Family.

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Abstract

In this thesis we have studied theoretically the linear and nonlinear optical properties of one electron GaAs quantum dot using the Schrdinger equation for rigid confinement. The studies on the linear and nonlinear optical properties involving lowest transition energy in quantum dot have the potential for promising device applications. In semiconductor quantum dots, the nonlinear optical properties are known to be greatly enhanced as compared to bulk semiconductors. The work presented in this thesis presented the linear and nonlinear optical properties such as the real and imaginary part of dielectric constant. The index of refraction and the absorption coefficient are studied. The theoretical investigation of the optical properties in spherical one electron quantum dot will lead to a better understanding of the properties of quantum dots. Thus it is found that the absolute strength of resonance is indeed large compared to typical absorption behavior of the bulk materials. It is observed that decrease the size of the quantum dot lead to a blue shift in peak positions of the absorption coefficient, refractive index and dielectric constant. A decrease in the total absorption coefficient and refractive index change is observed with increasing optical intensity. The present study would be a useful contribution to experimental work on linear and nonlinear optical properties of GaAs quantum dot. .

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Chapter 1

Introduction

1.1 Statement of the Problem

In this thesis the lowest transition energy for GaAs quantum dot will be calculated by solving the Schrodinger equation for rigid confinement. The shift in lowest transition energy for a QD will be compared with its bulk counterpart. Employing this transition energy, its linear and nonlinear optical properties such as the real and imaginary part of dielectric constant the index of refraction and the absorption coefficient will be studied. Size dependent optical property will be investigated.

1.2 Objectives

The main objectives of this work is to use the quantum mechanical formulation to study the linear and nonlinear optical properties associated with intraband transitions in the single electron charged GaAs quantum dot. It has two parts.

1. **Linear optical properties of GaAs quantum dot.**

    - To describe the energy versus radius of one electron GaAs quantum dot
    - To study the real and imaginary part of dielectric constant.
    - To study the optical property of GaAs quantum dot with its bulk counterpart.

2. **non linear optical properties of GaAs quantum dot**

    - To describe the nonlinear absorption coefficient and refractive index of GaAs quantum dot.

1.3 Significance of the study

Studying the linear and nonlinear optical absorption coefficients and refractive index in GaAs quantum dot will lead to a better understanding of its optical properties. Thus theoretical studies may have profound consequence' about practical application of the electro optical devices such as optical swithces and also has an extensive application in the optical communication. Moreover understanding the basic property of GaAs quantum dot will help for developing new generation technological devices.

Chapter 2

Theoretical background of the study

2.1 GaAs quantum dot

GaAs is one of the most technologically important and most investigated compound lll-V compound semiconductor materials and it is formed by combining As (group-V) and Ga (group-lll) elements from the periodic table. It was first produced by Goldschmidit in 1920's [1]. However, its properties remained unknown up to 1952. The crystal structure of GaAs is zincblende type, in which a face-centered cubic lattice (fcc) of As with Ga atoms positioned on the body diagonals. Ga(As) atoms is displaced from the As(Ga) atom by a distance $(\frac{a}{4} + \frac{a}{4} + \frac{a}{4})$ along the body diagonal, where a is the lattice constant of GaAs given by [2] $a = 0.565325 + 3.8810^{(} - 6)(T - 300k)nm$, Where T is the temperature in Kelvin.

Both the conduction band minima and the valence band maxima of GaAs lie at the same value vector at k=0. Therefore the transition of an electron from the valence band to the conduction band does not need photon assistance. According to the energy band diagram definition the nature of the GaAs bandgap is direct. The direct band gap ensures excellent optical properties of GaAs as well as superior electron transport in the conduction band. This property makes GaAs superior over si, which has an indirect band gap for devices used in optoelectronic applications. Moreover, GaAs possesses high carrier mobility than

si, and it is preferably used in high frequency devices. Electrical properties of GaAs material makes it suitable to be used as a semi-insulating substrate for integrated circuits do to its high resistivity compared to si material. In addition, due to the fact that energy gap of GaAs is higher than that of si. ($E_g^{GaAs} = 1.424ev > E_g^{Si}$=1.1ev at 300k), the GaAs devices are more reliable to operate at high temperatures than is devices [3].

Semiconductor quantum dots(QDs) are nanoscale materials in which the motion of an electron is confined in all three directions. This leads to quantum-size effects its electronic properties might not always be dominated by quantum size effects. A typical quantum dot contains between a few hundred and a few thousands atoms, and its electronic properties can be explained only invoking quantum confinement.

In GaAs quantum dot the transition leads to the formation of three dimensional systems. The obtained QD have typical dimension of approximately 15-30nm in diameter and 4-5nm in height size the smaller the potential well the greater the energy between the states in the QD. The material composition determines the band edges of the semiconductor and within the depth of the confinement potential [4].

There are a number of areas, where it has been predicted that the implementation of QDs should result in improved device performances. One example result is the quantum dot laser . That ideally should be intensive to temperature variations if separation distance between QD levels is larger than the thermal energy. QD lasers with promising performance have also been fabricated [5].

The QD infrared photodector is supposed to have good dector characteristics, since the discreteness of the energy states should result in a decreased dark current. Secondly, for intraband transitions in a QD infrared photodector, light can be observed in any angle of incidence. While a corresponding QW based detector only can absorb light polarized along the QW confinement direction [6].

A further step in developing QD based detectors is to be embedded them in a QW structure, so called dots in a well photo detector. The detectors intraband transitions are the between QD and QW states, which allows higher flexibility in device tuning [7] .

Other interesting QD-based devices are emitters of entangled photon-pairs and single photons emitters, which are central components in the developments of quantum cryptography and quantum information applications. Single photon emission from QDs was first shown using structures, where the QD was embedded in a micro cavity in resonance with the QD transition [8]. Subsequently, single photon emission has been achieved by resonant excitation into QD excited states allowing spin-state preparation [9].

Quantum dot consists of a nanoscale crystal from a special class of semiconductor materials, which are crystals, composed of chemical materials. The size of quantum dot ranges from several to tens of nanometers ($10^{-9}m$) in diameter, which is about 10-100 atoms. A quantum dot can contain from a single electron to several thousand electrons. Since the size of the quantum dot is designable a quantum dot behaves similarity to atoms and is often referred to as artificial atoms or giant atoms. The dots have nonlinear properties that are unique to them by increasing the intensity of light, heat or electricity being pumped into them. They can have an ultrafast(less than IPs), change in both the absorption coefficients and refractive index this gives then great switches behavior [10].

A quantum dot nanostructure that confines the motion of the charge carriers in all three spatial directions leading to discrete quantized energy level due to quantum confinement effects. The first experimental evidence and theoretical descriptions of 3D quantum confinement was published in the early 1980 with semiconductors nanocrystals[11].

The confinement in this case is formed by the presence of the interface between different semiconductor materials. The semiconductor quantum dot is buried in another semiconductor matrix while the band gap of the matrix material is larger than that of the dot material consequently the electron energy level and the heavy hole energy level and quantized and lifted relative to the band edge of the bulk material [10-11]. Here the heavy whole energy level is considered because it is the lowest level in the valence band in most common semiconductors used for the realization of quantum dots.

A quantum dots has electronic properties between those of bulk materials and discrete molecules. The energy quantization of both electrons and holes depends on the size,

shape and composition of quantum dots, as well as the intrinsic properties of quantum dot with matrix materials. In particular, the energy band gap of quantum dots is size dependent.

2.2 Quantum confinement and density of states

Nanostructures can be used to tune the band gap through quantum confinement. Quantum size effect (confinement) refers to the fact that carriers (electrons and holes) are confined by potential barriers in small region of space where the dimensions of the confinement are less than the deBroglie wavelength of these charge carriers. The length scale below which strong quantization effects occur ranges from 5-25nm for typical semiconductors [12].
In general, it is the physical confinement of charge carriers that lead to the creation of discrete energy levels. In the simplest case, these discrete energy levels can be calculated by solving the Schrdinger equation with infinite barrier at one-dimensional confinement. Certain other shape of higher dimensional confinement can also be treated theoretically. In regions of zero potential, the Schrdinger equation is:

$$\frac{\hbar^2}{2m^*}\nabla^2\psi = \mathbf{E}\psi \tag{2.1}$$

Where ψ are solutions of the wave functions of electron,$\hbar$ the planck constant, m^* is effective mass of electron and the energy level given by:

$$\frac{\hbar^2}{2m^*}k^2 = \mathbf{E} \tag{2.2}$$

The magnitude, k, of the wave vector is related to the wavelength λ, by$k = 2\pi/\lambda$. Reduced dimensionality leads to the change in density of energy states (Dos), which refers to the number of quantum states per unit energy [13].
Integrating the Dos of the quantum over a range of energy will produce a number of

states in that energy range. So Dos affects the Properties of quantum dots [14]. In bulk materials charge carriers are continuously distributed to higher energy states, while size quantization leads to an increase of the Dos near the Sub-band edge in QWs. With further reducing dimensionalities for NWs injected charge carriers concentrate in an increasingly narrow energy range near the sub-band edge. In QDs carriers (electrons and holes) are confined in three-dimensions, allowing zero dimensions (0D) in their degrees of freedom, and creating atom- like levels with discrete (Dirac delta) densities of states. Furthermore, with the same dimensionality, both energy levels and related Dos are affected by the shape of nanostructures. For example, the energy levels of ideal cubic shape QD can be simply calculated from the famous quantum mechanical model of the particle in a box problem [15].

Structure	Quantum Confinement	Number of free Density
Bulk	0	3
Quantum Well/Super Lattice	1	2
Quantum Wire	2	1
Quantum Dot/Nanocrystal	3	0

Table 2.1: Classification of quantum confined structure

Density of states

The density of states refers to the number of quantum states per unit energy. In other words the density of states indicates how densely packed quantum states in a particular system, which is the number of states per unit volume L^D in the energy $\epsilon + \epsilon + d\epsilon$ divided by d where L is the linear size of the system and D can be dimension. The basic idea here is that while restricting the motion of electron in one, and three dimensions gives rise to the discrete energy levels, in situations such as quantum well and quantum wire, there are additional degrees of freedom along unconfined directions. As a result one can get the concepts of density of states. So the calculation of density of states provide a more detail description of what the electrons structures as well as the energies of 3D,2D,1D and 0D materials looks like [16].

Density of states in three Dimensions (Bulk Materials)

In three dimensions, there is no confinement in the direction of motion of electrons. The only constrains is the periodic boundary conditions. Consider the volume in k-space $V_k = \frac{4}{3}\pi k^3$ and then each allowed value of k-space occupies a volume of

$$\left(\frac{2\pi}{L_x}\right)\left(\frac{2\pi}{L_y}\right)\left(\frac{2\pi}{L_z}\right) = \frac{8\pi^3}{V} = k_x k_y k_z \tag{2.3}$$

The number of modes (N) in the sphere is then:

$$N = \frac{V_k}{k_x k_y K_z} = \frac{\frac{4}{3}\pi k^3}{8\Pi^3} L_x L_y L_z \tag{2.4}$$

Assuming that the particle is an electron and we consider the spin of degeneracy (up and down), then we multiply N by 2

$$N' = 2\left(\frac{\frac{4}{3}\pi k^3}{8\Pi^3} L_x L_y L_z\right) = \frac{\pi k^3}{3\pi^3} L_x L_y L_z \tag{2.5}$$

This is the total number of states in sphere from the definition of the density, which is the number of states per unit volume, we get

$$\rho = \frac{N'}{L_x L_y L_z}\frac{k^3}{3\pi^2} \tag{2.6}$$

Since the density of states $\rho'(\epsilon)$ is defined as the number of allowed states within energy $d\epsilon$, thus

$$\rho'(\epsilon) = \frac{d\rho}{d\epsilon} = \frac{d\left(\frac{k^3}{3\pi^2}\right)}{d\epsilon} = \frac{1}{3\pi^2}\frac{d(k^3)}{d\epsilon} \tag{2.7}$$

where $k = \sqrt{\frac{2m\epsilon}{\hbar^2}}$ alternatively $k^3 = \left(\frac{2m\epsilon}{\hbar^2}\right)^{3/2}$

The expression becomes,

$$\rho'(\epsilon) = \frac{1}{3\pi^2}\left(\frac{2m}{\hbar^2}\right)^{3/2}\frac{d\epsilon^{3/2}}{d\epsilon} = \frac{1}{3\pi^2}\left(\frac{2m}{\hbar^2}\right)^{3/2}\frac{2}{3}\epsilon^{1/2} = \frac{1}{3\pi^2}\left(\frac{2m}{\hbar^2}\right)^{3/2}\frac{2}{3}\sqrt{\epsilon} \tag{2.8}$$

The above equation tells that the density of states in 3D is square root dependent on energy. It is known that the unconfined wave functions within 3D box are plane waves in all three dimensions. Therefore, the wave functions could be described as

$$\psi(x,\ y,\ z) = Ae^{ik_x X}e^{ik_y Y}e^{ik_z Z} \tag{2.9}$$

Substituting in the Schrödinger equations gives

$$\left(\frac{-\hbar^2}{2m}\right)\left(\frac{d^2}{dx^2}+\frac{d^2}{dy^2}+\frac{d^2}{dz^2}\right)\psi = E\psi \tag{2.10}$$

which gives

$$\frac{\hbar^2}{2m}(k_x^2+k_y^2+k_z^2) = E \tag{2.11}$$

After rearranging, energies of the system are

$$E = \frac{\hbar^2 k^2}{2m} \tag{2.12}$$

where $k = \sqrt{k_x^2+k_y^2+k_z^2}$ and $k_{x,\ y,\ z} = \frac{2m}{L}{}_{x,\ y,\ z}$

$$\rho'_{3D} = \frac{1}{2\pi^2}\left(\frac{2m}{\hbar^2}\right)\sqrt{\varepsilon-\varepsilon_g} \tag{2.13}$$

The 2D DOs: Quantum wells confined in 1D

In quantum wells, the electrons motion is confined in to two dimensions and unconfined in the other dimensions; let's assume its motion in the Z-direction. Thus, the total energy of the system is the sum of the energy along the quantized direction and the energy along the other two directions, therefore the Schrödinger equations for the carrier of effective mass m^* of a semi conductor quantum well is given by

$$\left[\frac{\hbar^2}{2m^*}\nabla^2 + V(x,\ y)\right]\psi(x,\ y,\ z) = E\psi(x,\ y,\ z) \tag{2.14}$$

The electron wave function is

$$\psi(x,\ y,\ z) = \psi_n(z)e^{i(k_x+k_y)} \tag{2.15}$$

the total energy of the carrier is given by [17]

$$E_n = \frac{\hbar^2}{2m^*}(k_x^2 + k_y^2) + \frac{\hbar^2 n^2 \pi^2}{2m_e L_z^2} \tag{2.16}$$

where $k^2 = k_x^2 + k_y^2 = \frac{n\pi}{L},\ k_x = \frac{2\pi}{L_x}$ and $k_y = \frac{2\pi}{L_y}$

Suppose now two areas in the k-space is given as $A_k = k^2$, the area of a given mode is then $k_x k_y$ with the total number of modes (N)

$$N = \frac{\pi k^2}{4\pi^2} L_x L_y = \frac{k^2 L_x L_y}{4\pi} \tag{2.17}$$

And again multiplying N by 2 account for the electron spin to get the total number of states

$$N' = 2\left(\frac{\pi k^2}{4\pi^2} L_x L_y\right) = \frac{k^2 L_x L_y}{4\pi} \tag{2.18}$$

We know that the total number of states per unit area gives, the densityρ, i.e.$\frac{N'}{L_x L_y}$, recalling that $k = \sqrt{\frac{2m\varepsilon}{\hbar}}$ substituting the equation gives $\rho = \frac{m\varepsilon}{\hbar^2\pi}$ which is the number of states per unit area. From the definition of the energy density, the density of states per unit area per unit energy becomes

$$\rho' = \frac{d\rho}{d\varepsilon} = \frac{m}{\hbar^2\pi} \tag{2.19}$$

One can observe that the energy density of the sub-band given by the above equation (2.17) for each successive k_2, there will be an additional $\frac{m}{\hbar\pi}$ term and hence another sub-band. Therefore, the density of states can be written as

$$\rho'_{2D} = \frac{m}{\hbar\pi} \sum \theta(\varepsilon - \varepsilon_n) \tag{2.20}$$

If a given semiconductor material with small energy gap is sandwiched between energy barriers from a semiconductor material with a large energy gap and layers thickness (are just a few atomic layer), the carriers are confined to the layered with lower energy. This situation gives the so called quantum wells [16-17].

Based on the above facts, QWs can be formed by preparing materials such as GaAs (gallium arsenide) sandwiched between two layers of materials with a wider band gap, like AlAs (Aluminum Arsenide) and etc. From the knowledge of quantum mechanics, it can be clear that the spectral properties of quantum wells are derived from confinement of charge carriers (electrons and holes). This shows that quantum confinement effect plays essential role to determine the spectral properties of quantum wells [18].

Another consequence on the quantum confinement is the modification of the density of states (Dos). In quantum wells the general shape of density of states modified as a step functions because of quantum confinement [16-17].

In addition the possibility of manipulating the sizes associated with the quantum well, changing the semiconductor material composition can also be a tool to fine tune the property of these quantum structures [17-18].

One of the peculiar features of quantum wells is the restriction on the movement of the electron into two directions and the other direction being confined. This in turns will result in allowed energy bands, whose energy positions are dependent on the height and width of the barrier. Therefore, restrictions on the movement of electrons affect electrons energy.

The most useful techniques for studies of the properties of nanostructures are based on the optical transitions between the discrete electronic levels / band gaps and thereby also an increase in gap energy due to confinement effects [16-18].

The 1D DOS: Quantum Wires Confined in 2D

In the situation where, the electrons motion is confined in two dimensions and only have one degree of freedom (say the x-direction), one can get quantum wire, Schrödinger equation is

$$\frac{-\hbar^2}{2m^*}\nabla^2\psi(x,\ y,\ z) = E\psi(x,\ y,\ z) \tag{2.21}$$

The wave function is given by

$$\psi(x,\ y,\ z) = \psi_{nl}(y,\ z)e^{i(k_x)} \tag{2.22}$$

the total energy of the system is can be written as

$$E_{nl}(k_x) = E_{nl} + \frac{\hbar^2}{2m_e}k_x^2 = \frac{\hbar^2\pi^2}{2m_e}\left(\frac{n^2}{l_z^2} + \frac{n^2}{l_y^2}\right) + \frac{\hbar^2k_x^2}{2m_e} \tag{2.23}$$

Furthermore along the confined directions$k_z = \frac{2\pi}{L_z}$, and $k_y = \frac{2\pi}{L_y}$ from the concept of quantum mechanics, the length in the k-space is given as L=2k. The number of modes along this length is$N = \frac{2k}{2\pi/L_x}$which is the number of states along a line is. Now if we consider an electron, ensure to take into account the spin degeneracy.

Now if we consider an electron, ensure to take into account the spin degeneracy

$$N = 2N = \frac{2kL_x}{\pi} \tag{2.24}$$

The number of states per unit length calls if density is given by

$$\rho = \frac{N'}{L_x} = \frac{2k}{\pi} = \frac{2}{\pi}\sqrt{\frac{2m\varepsilon}{\hbar^2}} \tag{2.25}$$

Therefore, based on similar definition given above, the energy density becomes

$$\rho' = \frac{d\rho}{d\varepsilon} = \frac{2}{\pi}\sqrt{\frac{2m\varepsilon}{\hbar^2}}\frac{d\sqrt{\varepsilon}}{d\varepsilon} \tag{2.26}$$

Thus, the complete expression taking in to account all m, and n are combinations for the energy density for quantum wires is written as

$$\rho'_{1D} = \frac{1}{\pi}\sqrt{\frac{2m}{\hbar^2}}\frac{1}{\sqrt{\varepsilon - \varepsilon_{n,m}}}\theta(\varepsilon - \varepsilon_{n,m}) \tag{2.27}$$

Where again θ is the heavy-side function. It is clear to observe that the density of states is inversely proportional to the square root of energy.

In quantum wires it is clear to observe that the motion of electrons is restricted only in one direction while leaving the other two directions confined. Thus, quantum wires can be defined as extremely narrow wires in which the free motion of carriers is only possible in one direction, along the wire, but confined in the other two directions. It can also defined as structures that have a thickness or diameter constrained to tens of nanometers or less and an unconstrained length [19].

These structures are usually referred to us one-dimensional (1D) materials and have many interesting properties that make them different from their corresponding bulk materials in the sense that QWRs are confined laterally and thus occupies discrete energy levels/ bands which are by far different from their original bulk materials [20].

The transport properties in quantum wires are affected by wire diameter, material compositions, quality of the crystal and etc . Optical properties raises in QWR's often show intense features at specific energies near singularities in the joint density of states formed under strong quantum confinement. Optical methods provide an easy and sensitive tool for measuring the electronic structures of nano wires, since optical measurements are sensitive to quantum effects [19-20].

The 0D-DOS: Particles confined in 3D

The 0D-Dos is spherical case because the motion of the particle confined in all directions. The density of states is basically a series of Delta-functions . The schrödinger equation with in the quantum box become

$$\frac{-\hbar^2}{2m}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}\right)\psi(x,\ y,\ z) = E\psi(x,\ y,\ z) \tag{2.28}$$

$$\psi_{nx,\ ny,\ nz}(x,\ y,\ z) = \sqrt{\frac{8}{L_x L_y L_z}} \sin\left(\frac{n_x \pi x}{L_x}\right) \sin\left(\frac{n_y \pi y}{L_y}\right) \sin\left(\frac{n_z \pi z}{L_z}\right) \tag{2.29}$$

the Eigen value for a specific Eigen function is given by

$$E_{nx,\ ny,\ nz} = \frac{\hbar^2 \pi^2}{2m^*} \left(\frac{n_x^2}{L_x^2} + \frac{n_y^2}{L_y^2} + \frac{n_z^2}{L_z^2}\right) \tag{2.30}$$

Where the fact that $E_{nx,\ ny,\ nz}$ is the total particle energy because of 3D confinement. A remarkable feature of a quantum box is when two or more of the dimensions are the same ($L_x = L_y = L_z$) more than one wave function corresponds to the same total energy [17]. The density of states in 0D (quantum dot) is written as

$$\rho'_{0D} = \theta(\varepsilon - \varepsilon_{l,n,m}) \tag{2.31}$$

When considering the density of states for a quantum dot structure, no free motion is possible because there is no k-space to be filled with electrons and all available states exist only at discrete energies.

Degrees of freedom	Dispersion *(KE)*	Density of state	Effective DS
3(bulk)	$E = \frac{\hbar^2}{2m^*}(k_x^2 + k_y^2 + k_z^2)$	$\rho_{DOS}^{3D} = \frac{1}{2\pi^2}\left(\frac{2m^*}{\hbar^2}\right)^{3/2}\sqrt{E - E_c}$	$N_c^{3D} = \frac{1}{\sqrt{2}}\left(\frac{m^* KT}{\pi\hbar^2}\right)^{3/2}$
2(slab)	$E = \frac{\hbar^2}{2m^*}(k_x^2 + k_y^2)$	$\rho_{DOS}^{2D} = \frac{m^*}{\pi\hbar^2}\sigma\sqrt{E - E_c}$	$N_C^{2D} = \frac{m^*}{\pi\hbar^2}KT$
1(wire)	$E = \frac{\hbar^2}{2m^*}(k_x^2)$	$\rho_{DOS}^{1D} = \frac{m^*}{\hbar\pi}\sqrt{\frac{m^*}{2(E-E_c)}}$	$N_C^{1D} = \sqrt{\frac{m^* KT}{2\pi\hbar^2}}$
0(box)	—	$\rho_{DOS}^{0D} = 2\sigma(E - E_c)$	$N^{0D} = 2$

Table 2.2: Density of states for semiconductors with 3,2,1 and 0 degree of freedom for propagation of electrons.

2.3 One electron quantum dot with rigid confinement

The phenomenon of the quantum confinement (i.e. the size quantization) can be observed in systems where the motion of electrons or other particles (holes, excitons, etc) is re-

stricted at least in one dimension by some potential energy profile such a system is usually referred to as a "low dimensional" system [20-21].

The energy spectra and the wave functions localization depend on the type of restrictions in one, two or three dimensions, as well as on the size of the nanostructure. In particular the quantum phenomena start to noticeable when the lateral extension of the potential will becomes comparable to the particle wave length. In order to better understand this concept, let us consider electrons. As elementary particles they exhibit the wave-particle duality of matter following the "deBrogile" relation. When immersed in a rigid the electrons are treated as a particle having an effective mass m^* accounting for the periodicity of the crystal potential. Its linear momentum P can be written in terms of its wave-like nature$p = \hbar k$, where $\hbar$ is the Dirac's constant (the plank's constant divided by 2π and k represents its wave number, associated to the deBorgile wave length $\lambda = 2\pi/k$. Electrons in a bulk solid are treated as particles not feeling the borders by imposing the periodic boundary conditions, so that the wave functions and the energies are not affected by the real spatial extension of the solid [20-21].

When the solid dimensions approach the electron wavelength, the permitted wave functions and energies undergo a series of restrictions in terms of continuity and absolute values. The system starts to be considered as quantized for a single electron spherical quantum dot [21].

2.3.1 The Particle in a Box

Modeling our electronic material as a box allows us to ignore atoms and assume that the materials are perfectly homogeneous [22]. We will consider boxes in different dimensions: three dimensions (typically bulk materials), 2-d (quantum wells), 1-d (quantum wires), or 0-dimensions (quantum dots).The label quantum here refers to the confinement of electrons [22].

When we say that an electron is confined in a low dimensional material we mean that critical dimensions of the material are on the order of the wavelength of an electron. We

know that when particles are confined, their energy levels become discrete. In quantum dots, electrons are confined in all three dimensions. In the confined directions we assume that the electron described by a plane wave [23].

2.3.2 Energy Eigenvalues of an electron in rectangular quantum box

Analyzing quantum particles in a box and bulk materials requires that solving the Schrodinger equation in 1-D, 2-D and 3-D. The Schrodinger equation in 1-D is given by:

$$E\psi(x) = \left(\frac{-\hbar^2}{2m} + \frac{d^2}{dx^2} + V(x)\right) + \psi(x) \tag{2.32}$$

The equation can be extended to higher dimensions (3D) as the kinetic energy operator in 3-D is given above as:

$$\hat{T} = \frac{\hat{P}_x^2}{2m} + \frac{\hat{P}_y^2}{2m} + \frac{\hat{P}_z^2}{2m} = \frac{-\hbar^2}{2m}\left(\frac{d^2}{dx^2} + \frac{d^2}{dy^2} + \frac{d^2}{dz^2}\right) \tag{2.33}$$

Where $\hat{P}_x, \hat{P}_y, \hat{P}_z$ are the components of the momentum on the X, Y, and Z axes respectively.

$$V(x,y,z) = \{^{0,\ 0<x<L,\ 0<y<L,\ 0<z<L}_{\infty,\ \text{otherwise}} \tag{2.34}$$

The above potential can be separated into x, y and z dependent terms

$$V(x,y,z) = V(x) + V(y) + V(z) \tag{2.35}$$

For a potential of this form the Schrodinger equation can take the following forms.

$$\frac{-\hbar^2}{2m}\left(\frac{d^2}{dx^2} + \frac{d^2}{dy^2} + \frac{d^2}{dz^2}\right)[V(x) + V(y) + V(z)]\psi(x,y,z) = E\psi(x,y,z) \tag{2.36}$$

where

$$E = \left[\frac{-\hbar^2}{2m}\right]\left(\frac{d^2}{dx^2} + \frac{d^2}{dy^2} + \frac{d^2}{dz^2}\right)[V(x) + V(y) + V(z)] = E_x + E_y + E_z \tag{2.37}$$

The wave function can also be written as:

$$\psi(x, y, z) = \psi(x) + \psi(y) + \psi(z) \tag{2.38}$$

The wave function $\psi(x, y, z)$ must vanish at the wall of the cubic box. The solution for one dimensional potential is given by:

$$\chi(x) = \sqrt{\frac{2}{L}\sin\left(\frac{n_x\pi}{L}x\right)n_x} \tag{2.39}$$

and the corresponding energy Eigenvalues becomes

$$E_{nx} = \frac{\hbar^2 n_x^2}{2m^* L^2} \tag{2.40}$$

From the expression above we can write the normalized three dimensional Eigenfunctions and their corresponding energies as:

$$\psi_{nx,ny,nz}(x, y, z) = \sqrt{\frac{8}{L^3}}\sin\left(\frac{n_x\pi}{L}x\right)\sin\left(\frac{n_y\pi}{L}y\right)\sin\left(\frac{n_z\pi}{L}z\right) \tag{2.41}$$

$$E_{nx,ny,nz} = E_x + E_y + E_z = \frac{\hbar^2\pi^2}{2m * L^2}\left(n_x^2 + n_y^2 + n_z^2\right) \tag{2.42}$$

Where n_x, n_y, n_z=1,2,3 The ground state energy becomes

$$E_{lll} = \frac{3\pi^2\hbar^2}{2m * L^2} = 3E_1 \tag{2.43}$$

Where $E_l = \frac{\pi^2\hbar^2}{2m^*}$, is the zero potential energy of a particle in one dimensional box. The first excited state has three possible sets of quantum numbers n_x, n_y, n_z (2,1,1),(1,2,1),(1,1,2)

corresponding to three different states $\psi_{211}(x,y,z),\ \psi_{121}(x,y,z)\text{and}\psi_{112}(x,y,z)$. The first quantum state is given by:

$$\psi_{211}(x,y,z) = \sqrt{\frac{8}{L^3}} \sin\left(\frac{2\pi}{L}x\right) \sin\left(\frac{\pi}{L}y\right) \sin\left(\frac{\pi}{L}z\right) \tag{2.44}$$

Chapter 3

MATERIALS AND METHODS

The study is devoted to the theoretical study of the linear and nonlinear optical properties of semiconductor GaAs quantum dot. The materials which we have employed were high capacity computer, Kile/Latex software source code and MATLAB is used for developing. Program code for plotting the thesis result and the roots are numerically determined by a program written in MATLAB. Mathematica software also used to solve long analytical and numerical expressions in our scientific work.

3.1 Methodology

The data we used in this simulation depends on materials properties of semiconductor quantum dot are based on experiment works those had been performed with other scholars.

3.1.1 Analytical method

In this thesis one of the important methods solving the problem analytically, the linear and nonlinear optical properties of GaAs quantum dot will be calculated.

3.1.2 Graphical and Numerical method

Graphically and numerical methods are the designed approaches to obtain quantitative solutions as well as qualitative description of the result.

Chapter 4

Energy Eigenvalues and optical properties of a spherical GaAs quantum dot

In this thesis the lowest transition energy for spherical GaAs quantum dot (QD) is calculated by solving the Schrödinger equation for rigid confinement. the shift in lowest transition energy for a QD is larger than the bulk system and this makes it useful for application from florescent labeling to light emitting diodes. Employing this transition energy its linear and saturable non linear optical properties such as the real and imaginary part of dielectric constant, the index of refraction and the absorption coefficient are studied.

4.1 Mathematical Formulation of the problem

The Schrödinger equation [18] for three dimensional spherical motions is,

$$\frac{-\hbar^2}{2m}\nabla^2(r,\theta,\varphi) + V(r,\theta,\varphi) = E\psi(r,\theta,\varphi) \tag{4.1}$$

Where

$$V(r)_{nl} = \{^{0 \quad \text{for} r<R}_{\infty \quad \text{for} \quad r\geq R} \tag{4.2}$$

The well known solution of this Schrödinger equation for rigid confinement is

$$\psi(r,\theta,\varphi) = Y_{lm}(\theta,\varphi)\frac{1}{R}\sqrt{\frac{2}{r}}\frac{J_{l+1/2}(\beta_{nl}r)}{J_{l+3/2}(\beta_{nl}R)} \tag{4.3}$$

Where, R is the radius of a QD and β_{nl} is the n^{th} zero of $\mathbf{J_{l+1/2}}(\beta_{\mathbf{nl}}\mathbf{R})$.

4.2 Quantum shift of the first transition for 2nm GaAs quantum dot

For 2nm radius of spherical GaAs quantum dot the resulting quantum shifts of the first transition are calculated employing the roots of Bessel function from the equation.

$$J_{l+1/2}(\beta_n R) = 0 \tag{4.4}$$

The function $J_{l+1/2}(\beta_n R)$ is the $(l+1/2)^{th}$order spherical Bessel function and the coefficient β_{nl} is the n^{th} zero of $J_{l+1/2}(\beta_n R)$. The roots are numerically determine by a program written in MATLAB. The results are illustrated in table 4.1

No of zero	l=0	l=1	l=2	l=3	l=4
1	3.14159	4.49341	5.76346	6.98793	8.18256
2	6.28319	7.72525	9.09501	10.4171	11.7049
3	9.42478	10.9041	12.3229	13.6980	15.0397

Table 4.1: Roots of $J_{l+1/2}(\beta_{nl}R) = 0$ for l=0,1,2,3 and 4

The conduction band effective electron mass for GaAs QD is 0.067 m_0(where $m_0$is the free

electron mass) [24]. The electron energy levels are given by

$$E_{nl} = E_{00}\left[\frac{\beta_{nl}}{\beta_{00}}\right]^2 = \frac{5.6144\times^{-14}}{R^2}\left[\frac{\beta_{nl}}{\beta_{00}}\right]^2 cm^2 - ev \tag{4.5}$$

The spacing between the lowest two energy levels is 1.457 eV. This is the quantum shift of the first transition for spherical GaAs quantum dot of 2nm. The variation of energy with respect to radius of GaAs QD is illustrated in Fig. 4.1

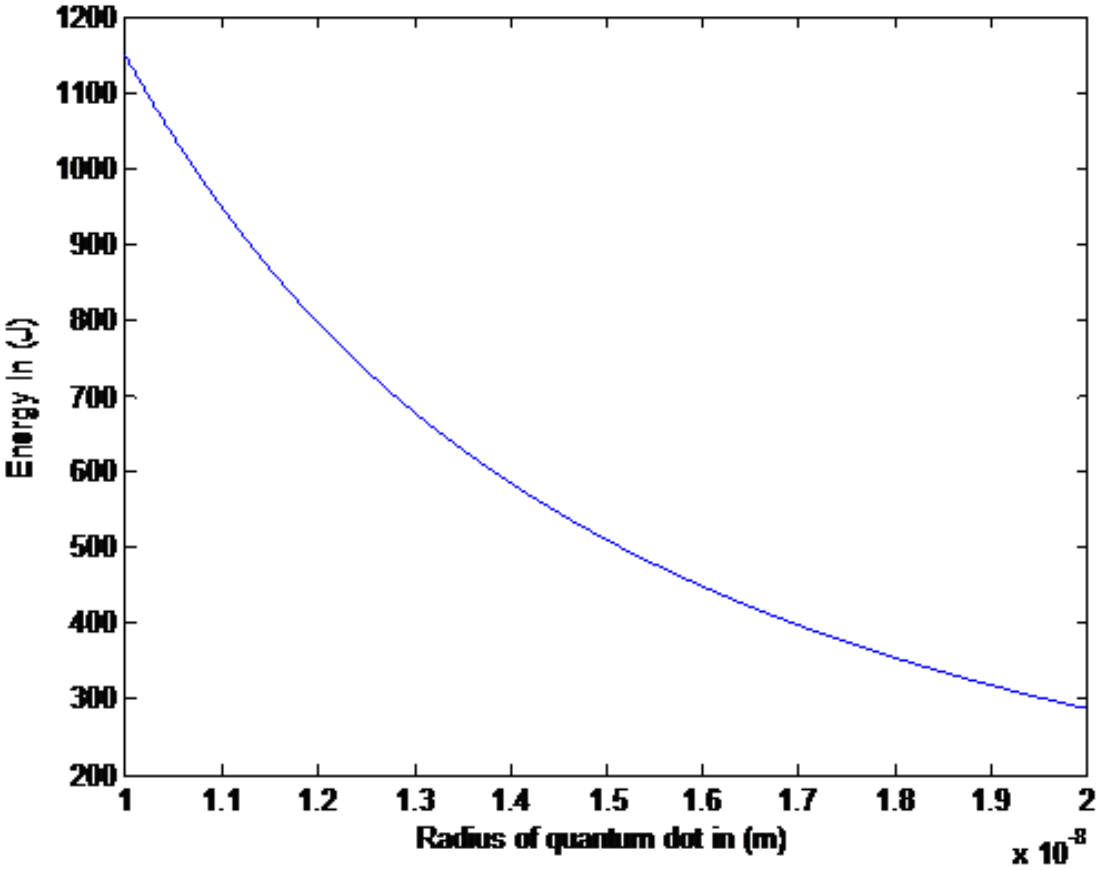

Figure 4.1: Energy versus radius of one electron GaAs quantum dot

It clearly verifies that reducing the size of QD increases the energy gap of the QD. The 1s electron state increases in energy with decreasing the QD radius R, while the 1s hole state decreases. The band gap therefore grows and can be tune over a wide range by changing

radius R. For the smallest radii, there is a shift in the lowest transition energy.

4.3 Linear optical properties of a spherical GaAs quantum dot

If the QD is subject to an oscillating electric field the perturbing Hamiltonian,$H(t) = H_p e^{i\omega t}$, induces a quantum transition from the initial state i of energy E_i to the final state of energy f of energy E_f. Using Fermi's golden rule, the transition probability for a QD [13, 18, and 22] is,

$$W(\omega) = \frac{2\pi}{\hbar}\frac{e^2 A_0^2}{m^{*2}c^2} \mid P_{c\nu} \mid^2 \frac{1}{\nu}\rho^{oD} \tag{4.6}$$

Where A_0 is the amplitude of the vector potential, P_{cv} is the optical matrix element, V is the volume of the QD. The moment matrix element is described as [22]

$$\mid P_{cv} \mid^2 = \frac{3m_e^2 E_x(E_x + \Delta x_0)}{2(3E_x + 2\Delta x_0)}\left(\frac{1}{m_e^*} - \frac{1}{m_e}\right) \tag{4.7}$$

Where, E_x is the band gap of the QD with one electron, Δx_0 the spin- orbit splitting of valence bands and m_e^* is the electron effective mass and m_e is the electron mass. For 2 nm radius of a spherical GaAs QD E_x= 2.887eV, Δx_0=0.34eV and m_e^* =0.067m_e . The optical absorption coefficient (α) is proportional to the number of optical transitions per unit volume and time. It is given by absorbed energy per unit time $\hbar\omega W(\omega)$. divided by energy flux,

$$\frac{A_0^2\omega}{\lambda} = \frac{A_0^2\omega^2 n(\omega)}{2\pi c} \quad \text{and} \quad \alpha(\omega) = \frac{\hbar\omega W(\omega)}{A_0^2 W^2 n(\omega)} 2\pi c$$

$$\alpha(\omega) = \frac{1}{V}\frac{4\pi^2 e^2}{m^{*2}\omega n(\omega) c} \mid p_{c\nu} \mid^2 \rho^{0D} \tag{4.8}$$

The optical absorption coefficient in terms susceptibility can also be written as

$$\alpha(\omega) = \frac{4\pi\omega}{cn(\omega)} Im\chi \tag{4.9}$$

Relating equation (4.15) and equation (4.16) we obtain,

$$Im\chi = \frac{f}{V}\sum_{n,\ l}(2l+1)\delta(E - E_{enl} - E_{hnl}) \tag{4.10}$$

where $f = \frac{2\pi e^2\hbar^2}{m^{*2}E^2} \mid p_{c\nu} \mid^2$

The linear optical properties such as real and imaginary part dielectric constant, and the absorption coefficient are determined for GaAs QD. In GaAs QD the allowed states are separated by large energies. So that transitions due to screening, exchange and band gap normalization are difficult and strongly in habited [23,24]. For the same reason. It is difficult to polarize the excition. In general it is possible to have two electrons or holes in any given energy level because the individual electron and hole states are spin degenerate. This means however that there is no exchange interaction between excited particles in a given energy level. As they have different spins. Hence there are identically no exchange effects if we only create electrons and holes in a given conduction level, respectively. This will be the cause for the absorption of the lowest quantum dot transition. In the vicinity of the resonance the dielectric constant ($\epsilon = \epsilon_1 + i\epsilon_2$) for quantum dot [26-29] is

$$\varepsilon = \varepsilon_\infty + \frac{\beta(\delta + i)}{1 + \delta^2} \tag{4.11}$$

where $\beta = \frac{4\pi f}{\hbar\Gamma_{xV}}$ and $\delta = \frac{(\hbar\Omega_x - \hbar\omega)}{\hbar\Gamma_x}$ With $\hbar\omega$ being the photon energy E, $\hbar\Omega$ being the lowest transition energy E_x, $\hbar\Gamma_x$ being the broadening E_b , and E_∞ the back ground dielectric constant. After some manipulation, the real and imaginary part of the dielectric constant for a quantum dot is given by

$$\epsilon_1 - \epsilon_\infty = \left(\frac{9\pi e^2\hbar^2 E_x(E_x + \Delta x_0)}{E_b R^3 E^2(3E_x + \Delta x_0)}\right)\left(\frac{m_e - m^*}{m_e m_e^*}\right)\left[\frac{\frac{(E_x - E)}{E_b}}{1 + \left(\frac{E_x - E}{E_b}\right)^2}\right] \tag{4.12}$$

$$\epsilon_2 - \epsilon_\infty = \left(\frac{9\pi e^2\hbar^2 E_x(E_x + \Delta x_0)}{E_b R^3 E^2(3E_x + \Delta x_0)}\right)\left(\frac{m_e - m^*}{m_e m_e^*}\right)\left[\frac{1}{1 + \left(\frac{E_x - E}{E_b}\right)^2}\right] \tag{4.13}$$

with the knowledge of ϵ_1 and ϵ_2, the index of refraction and the absorption coefficient are given by:-

$$n_r(\omega) = \left[\frac{1}{2}\epsilon_1(\omega) + \frac{1}{2}\{[\epsilon_1(\omega)]^2 + [\epsilon_2(\omega)]^2\}^{\frac{1}{2}}\right]^{\frac{1}{2}} \tag{4.14}$$

$$\alpha(\omega) = \frac{\omega\epsilon_2(\omega)}{c\left[\frac{1}{2}\epsilon_1(\omega) + \frac{1}{2}\{[\epsilon_1(\omega)]^2 + [\epsilon_2(\omega)]^2\}^{\frac{1}{2}}\right]^{\frac{1}{2}}} \tag{4.15}$$

The curves in Fig.4.2 are drawn according to (4.12) and (4.13) for a QD, with $E_x = 2.887ev, E_b = 0.5mev, \epsilon_\infty = 10, \Delta x_0 = 0.34ev$ and the radius of QD a= 2nm. They show the real and imaginary part of the dielectric constant.

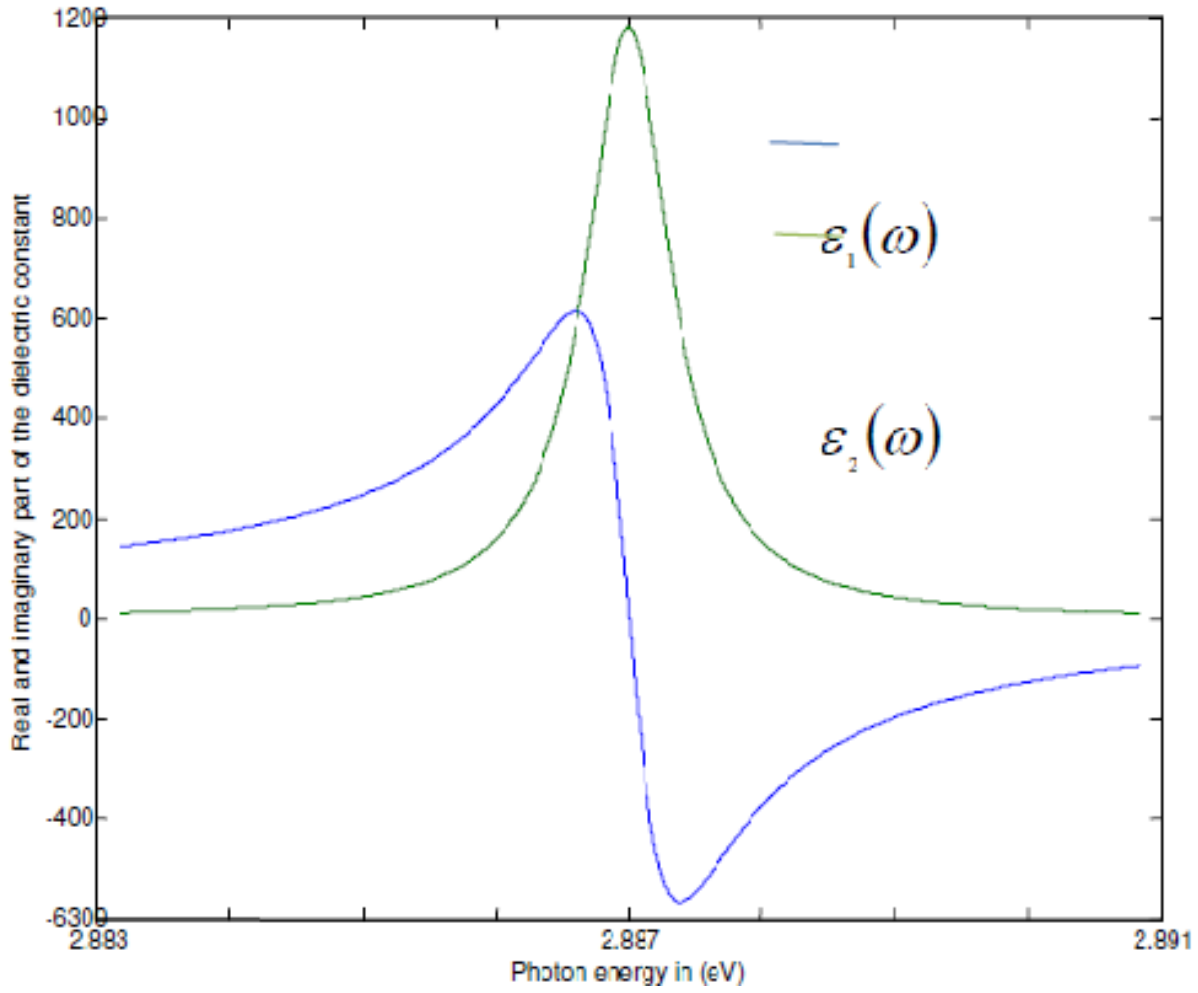

Figure 4.2: Real and imaginary part of linear dielectric constant for GaAs quantum dot

The absolute strength of the resonance is indeed large as compared to typical absorption and refraction behavior in the bulk material. Figures 4.3A and 4.3B demonstrate the linear refractive index and absorption coefficient respectively. These graphs are drawn according to (4.14) and (4.15). From figure 4.4 one can see that reducing a size of the dot increases the energy gap and results in the blue shift. This property makes the QD applicable from florescent labeling to light emitting diodes.

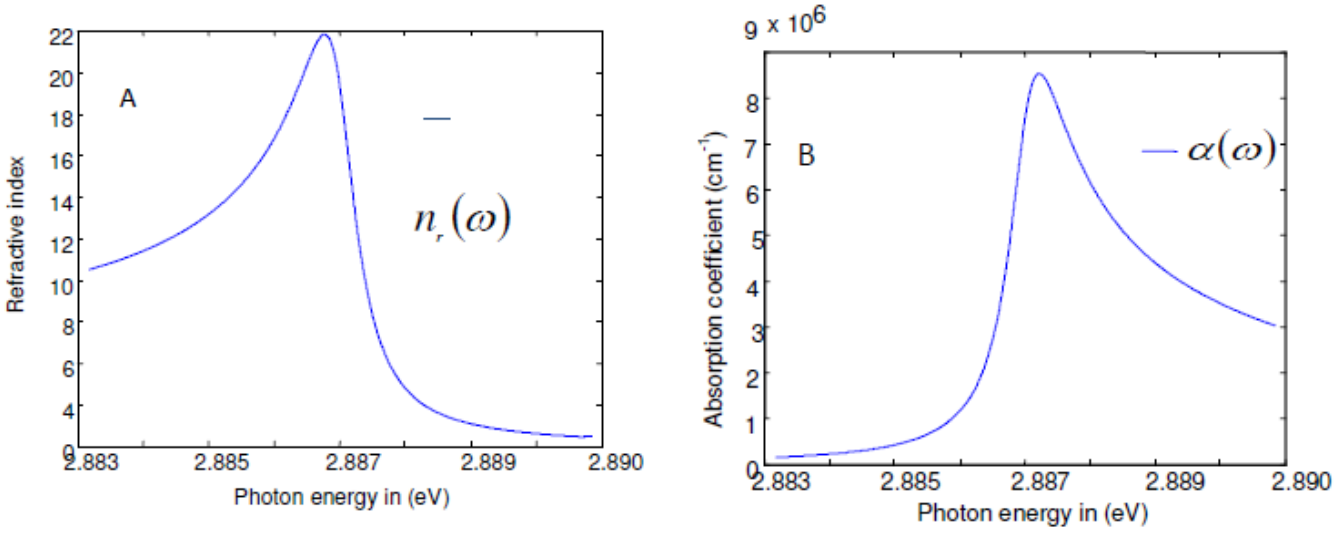

Figure 4.3: Linear refractive index (A) and absorption coefficient (B) versus photon energy for GaAs QD with one electron.

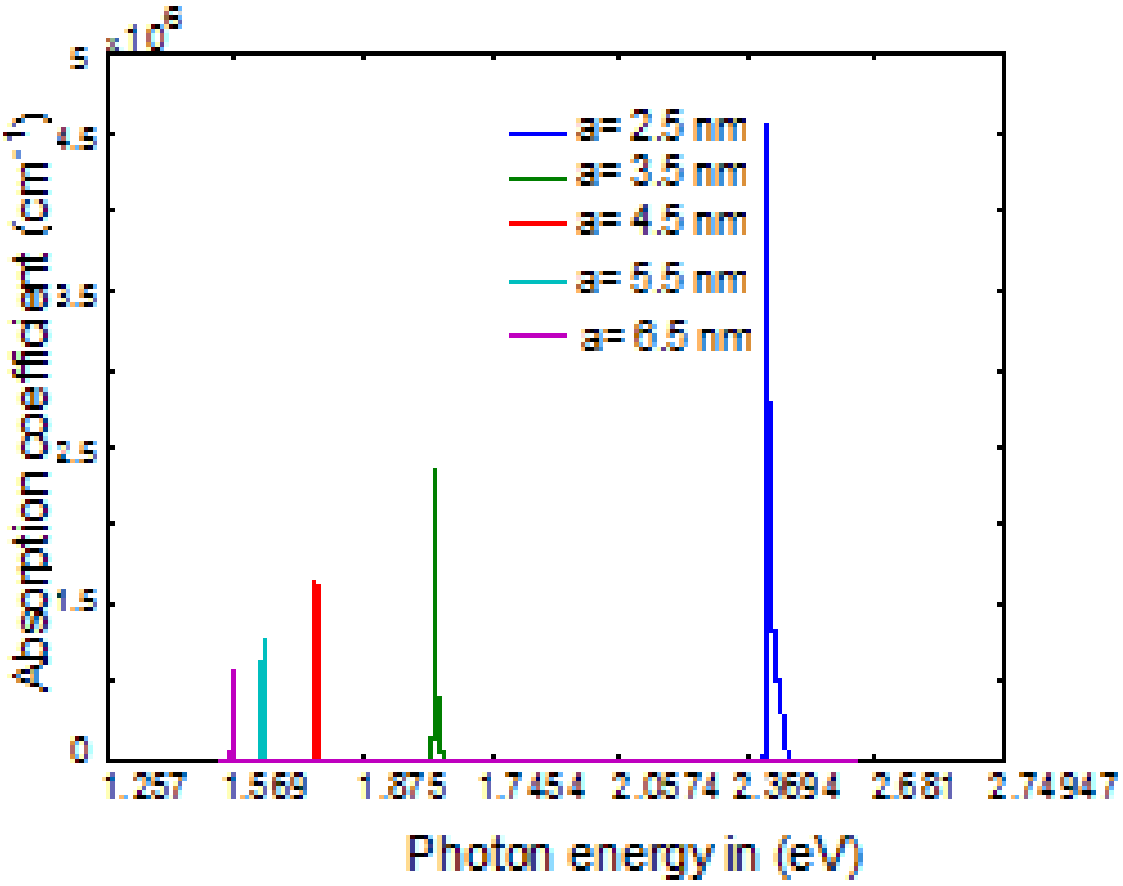

Figure 4.4: Linear absorption coefficient for one electron GaAs QD

4.4 Nonlinear optical properties of a spherical GaAs quantum dot

Since exchange and Coulomb effects are rigorously negligible, the absorption saturation of the lowest transition in the QD can be treated like a pair of two level systems, one for each spin. As the result the absorption saturates as

$$\epsilon = \epsilon_\infty + \frac{\beta(\delta + i)}{1 + \delta^2 + I/I_s} \tag{4.16}$$

where $I = E_0^2$ is the intensity with E_0 being the peak amplitude of the oscillating electric field inside the QD. $I_s = \frac{\hbar}{(\beta_\tau V)}$ is the saturation intensity, with being the recombination time. The absolute changes in absorption and consequently in the refractive index of a

QD can be much larger before the transition saturates in contrast to bulk system. The real and imaginary part of the dielectric constant for this case is

$$\epsilon_1 - \epsilon_\infty = \left(\frac{9\pi e^2\hbar^2 E_x(E_x+\Delta x_0)}{E_b R^3 E^2(3E_x+\Delta x_0)}\right)\left(\frac{m_e - m_e^*}{m_e m_e^*}\right)\left[\frac{\frac{(E_x-E)}{E_b}}{1+\left(\frac{E_x-E}{E_b}\right)^2 + I/I_s}\right] \tag{4.17}$$

$$\epsilon_2 = \left(\frac{9\pi e^2\hbar^2 E_x(E_x+\Delta x_0)}{E_b R^3 E^2(3E_x+\Delta x_0)}\right)\left(\frac{m_e - m^*}{m_e m_e^*}\right)\left[\frac{1}{1+\left(\frac{E_x-E}{E_b}\right)^2 + I/I_s}\right] \tag{4.18}$$

The intensity dependent refractive index and absorption coefficient according to (4.14), (4.15), and (4.17) and (4.18) are described in Fig 4.5. From the figures one can observe that the saturable nonlinear refractive and absorption coefficients decrease with increasing intensity. For QDs the nonlinear absorption completely saturates after enough refractive index changes or absorption changes takes place to make a given device switch in contrast to the bulk system. This property may be important to utilize a QD as a system for optical bistability and optical switching.

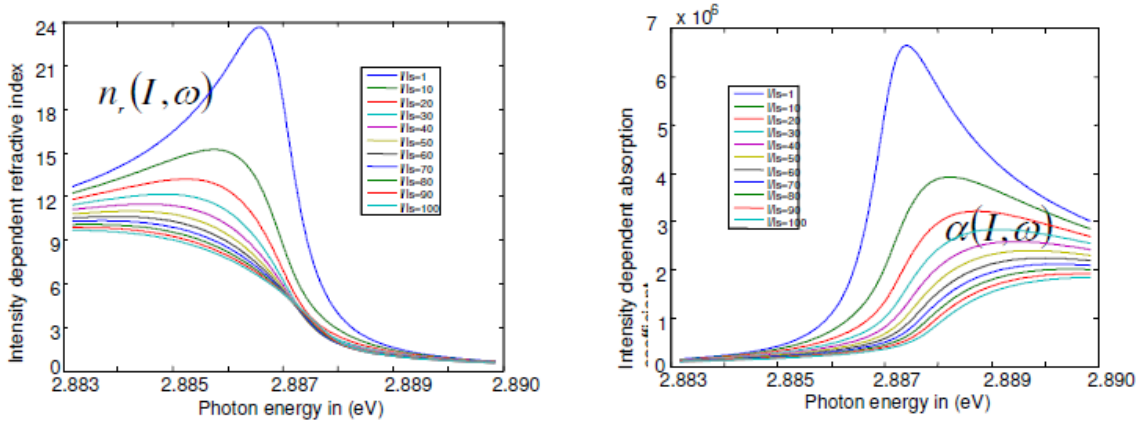

Figure 4.5: Intensity dependent refractive index and absorption coefficients for one electron GaAs QD.

The magnitude of the change in absorption per absorbed photon per unit volume is not significantly different in the QD from that of Bulk exactions for a given line width. Although the absorption in the QD is larger, the volume in which one photon must be absorbed

for saturation is correspondingly smaller, the real advantage of the QD is perhaps that the absolute changes in absorption and consequently in the refractive index can be much larger before the transition saturates. This is particularly important for nonlinear refractive effects, where in bulk material the readily saturable excitonic nonlinear absorption often completely saturates before enough index change or absorption change takes place to make a given device switch. Furthermore, this theory predicts that relatively complete saturation of absorption should be possible in the QD, because there are no completing mechanisms such as band gap renormalization that inhabit deep absorption saturation in bulk materials.

The present result are useful for further understanding the optical properties of one electron GaAs QD, and we hope that the theoretical study can make a significant contribution to experimental studies and practical applications.

Chapter 5

Conclusion

In this thesis we have studied the linear and nonlinear optical properties of one electron GaAs quantum dot. We have observed the real and imaginary part of dielectric constant the index of refraction and the absorption coefficient of one electron spherical GaAs quantum dot. The lowest transition energy of 2nm radius of spherical GaAs quantum dot is about 2.887ev and is much greater than to its bulk counterpart. Decrease in the absorption coefficient and refractive index change is observed with the increase of the optical intensity. Decreasing the radius of the quantum dot increases the energy gap and results in the blue shift. This shows that, in principle it is possible to use artificial materials with ever desirable optical properties.

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