Optical Properties of Three Dimensional Two Electron Zinc Oxide(ZnO) Quantum Dot in Parabolic Confinement

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DECLARATION

I here by declare that this thesis is my original work and has not been presented in any other university, and that all sources of material used for the thesis have been dully acknowledged.

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By

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Approved by the Examination Committee.

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Abstract

In this thesis, we discussed about the optical properties of three dimensional two electron zinc oxide (ZnO) quantum dot in parabolic confinement. Quantum dots are the minimum limit of low dimensional semiconductor systems in which the charge carriers (electrons and holes) are confined in all three directions of space. Quantum dots (QDs) refer to nanostructures made from semiconductor materials. Quantization of energy is the unique property of artificial atoms (quantum dots) and real atoms which makes quantum dots to be useful in studying the properties of systems at the atomic size scale. We discussed about the ground and excited state property of three dimensional(3D) two electron zinc oxide (ZnO) quantum dot in parabolic confinement. The ground and excited state energy eigenvalues of three dimensional(3D) two electron Zinc oxide(ZnO) quantum dot in parabolic confinement are obtained by using variational method. We use the variational method since the coupling constant is not small enough. Based on the obtained ground and excited state energy eigenvalues, the real and imaginary part of the third order susceptibilities are magnified for an increment of the confining frequency.

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Introduction

1.1 Back Ground of the Study

Since 1960, scientists are looking for very small sized (semiconductor nanostructures) that are applicable in many field of science and engineering. Among these scientists, a famous man Recard Feynman [1] declared that, "there is plenty of room at the bottom ", showing his vision towards small sized. In his work, he predicted that the miniaturization of large sized to nanometer length scales would result in the discovery of new technology which is called nanotechnology. The field of semiconductor nanotechnology began in the late 1960's and 1970's. The nanometer length scale materials are called low dimensional systems [2]. Quantum dots (QDs) generally refer to nanostructures made from semiconductor materials, in which carriers are confined in all spatial dimensions. Modern semiconductor processing techniques allowed the artificial creation of quantum confinement shape of few electrons. Because of the similarity between atoms and quantum dots they are called artificial atoms. However, there are considerable differences in physical characterizations between them. The confining potential of nucleus is singular but that

of quantum dots is nonsingular, which makes the parabolic confinement approximation possible and artificial atoms' typical dimensions range from nanometers to few microns, are much larger than real atom.

Low dimensional nanometer sized system describe those materials that exhibit different structure from the bulk semiconductor as a results of restricting the motion of a particle with the full three dimensions[3]. When the dimensions of material are reduced down to the nanoscale their properties often change from that of their bulk counterparts. If the size of the particle is very small comparable to bulk, then the available energy states become discrete and for charge carriers in semiconductor materials, the effective band gap between valence and conduction bands increases. If this confinement is only significant in one direction, then a quantum well is established.

In recent years considerable experimental and theoretical interest has been focused on the study of artificially structured atoms, such as quantum dots, where few electrons are confined in all three directions [4].

If the semiconductor is significantly confined in two dimensions, then the structure is quantum wire. Finally, if the semiconductor is strongly confined in all three directions; then quantum dot is created [5]. Low dimensional semiconductor materials for nanoscale have attracted wide interest due to their potential applications in many different fields. When the extent of large scaled is reduced in one or more the physical, chemical, electrical and optical properties can be dramatically altered. Quantum wells and quantum dots (QDs) with one electron are widely used in the nanophysics[6]. The modern technology gives a possibility to design quantum dots

with two and more electrons, where the coulomb interaction between them must be considered [7]. Due to the nanoscale localization of electrons, the coulomb interaction can exceed the average kinetic energy of electrons, which considerably complicates the analytic solutions of the Schrodinger equation.

The potential interest of studying nanostructure materials are due to essentially tenability of the physical properties by varying the particle size and shape. Quantum size effects also become prominent in semiconductor nanoparticles when the dimensions are reduced to sizes comparable to bulk semiconductor. If the dimensions are decreased sufficiently, the band gap of the semiconductor nanoparticles due to quantum confinement, altering the light absorption and emission properties of these nanoparticles that are now known as quantum dots [8].

Since 1991 carbon based on structured material were discovered like nanotubes, nanocrystal diamond, and group I-VI compound semiconductor. Group I-VI semiconductor nanoparticles have been extensively studied for their applications in displays, high density storage devices, photovoltaic and biological labels. One of the major effects is on optimizing the emission properties of the wide-band-gap semiconductor materials due to the increasing demand for high brightness light source operating in the ultraviolet (UV) region. Some examples of nanostructures based wide band gap semiconductor is, such as Zinc oxide(Zno), Gallium nitride(GaN) and Silicon Carbide(SiC). Zinc oxide(ZnO) is a I-VI compound semiconductor with a wide band gap of 3.37ev at room temperature.

One important consequences of its wide band gap is the transparency of ZnO to visible light, which promotes its applications as transparent conducting electrodes

in light emitting and photovoltaic devices. Zinc oxide exhibits high resistance to radiation, which makes it a suitable material for space applications [9].

1.2 Statement of the Problem

Most of the studies that are mentioned in the back ground of the study are limited to study the optical properties of one electron quantum dot in parabolic confinement. However, the two electron quantum dot has not been studied and this motivated us to investigate the optical properties of two electron quantum dot in parabolic confinement. The main purpose of this study is to calculate energy eigenvalues of three dimensional two electron quantum dot in parabolic confinement using variational techniques. In addition, we want to study the low dimensional properties of two electron quantum dot which exhibits different properties from its bulk counterpart due to quantum confinement effect.

1.2.1 Basic Research Questions

✓ How can we evaluate the energy eigenvalues of three dimensional two electron Zinc oxide quantum dot at ground state and first excited state?

✓ How can we determine the linear optical susceptibility for three dimensional two electron Zinc oxide quantum dot?

✓ What is the third order non-linear optical susceptibility of three dimensional two electron Zinc oxide quantum dot?

1.3 Objectives

1.3.1 General objective

The general objective of this study is:

✓ To study the quantum confinement effect of quantum dots and to determine the ground state and first excited state energy eigenvalues and to investigate optical properties of three dimensional two electron Zinc oxide quantum dot in parabolic confinement.

1.3.2 Specific objectives

- To evaluate the ground and excited state energy eigenvalues of three dimensional two electron Zinc oxide quantum dot.
- To determine the linear optical susceptibility of three dimensional two electron quantum dot.
- To calculate the third order non-linear optical susceptibility of three dimensional two electron Zinc oxide quantum dot.

1.4 Significance of the Study

This work helps us:

✓ To study peculiar optical properties of three dimensional two electron Zinc oxide(ZnO) quantum dot in parabolic confinement semiconductor in comparison to their bulk counterparts.

✓ To study the optical properties of three dimensional two electrons quantum dot in parabolic confinement which leads to know the effect of exchange and columbic energy on the quantum dot.

✓ To study the qualitative and quantitative theory of two electron quantum dot.

✓ The three dimensional two electron Zinc oxide(ZnO) quantum dot with tunable optical properties have got a remarkable technological relevance for various applications in various fields of science.

✓ In this work we shall able to understand the unique properties and various applications of Zinc oxide quantum dot(QD) and we get knowledge of how to calculate ground and excited state energy eigenvalues of three dimensional two electron Zinc oxide(ZnO) quantum dot in parabolic confinement.

1.5 Scope of the Study

The research is intended to calculate energy eigenvalues of three dimensional two electron Zinc oxide quantum dot and to determine linear and nonlinear optical susceptibilities by using density matrix formalism.

1.6 Limitations of the Study

Due to the time constraint, lack of internet and laboratory the scope of the study is limited to determine the optical properties of three dimensional two electron Zinc oxide quantum dot in parabolic confinement using density matrix formalism quantum mechanics technique.

2

Review of Literature

2.1 Introduction

In general, quantum dots(QDs) refer to nanostructures made from semiconductor materials, in which carries are confined in all spatial dimensions. Modern semiconductor processing techniques allowed the artificial creation of quantum confinement shape of few electrons. Because of the similarity between atoms and quantum dots they are called artificial atoms.

A three-dimensional harmonic oscillator with frequencies in rational ratios (RHO) and a Coulomb system are benchmarks for the hidden symmetries which account for the accidental degeneracy's of their quantum spectra. Recent advances in nanotechnology create a remarkable opportunity to trace their dynamical interplay in microscopic systems like quantum dots, which confine a few electrons to a space of a few hundred nanometers. Indeed, competition between a confining potential, approximated quite well by the harmonic oscillator (HO) and the repulsive electron-electron interaction produces a rich variety of phenomena, for example, in a two-electron quantum dot (QD) under a perpendicular magnetic field. Although most

of the symmetries are expected to be broken when the HO and the Coulomb potential are combined, in particular cases the Coulomb system and the RHO may have common symmetries. These symmetries were rediscovered in the analysis of laser-cooled ions in a Paul trap and of the hydrogen atom in the generalized van dear Waals potential. Here we demonstrate that the hidden symmetries could be observed in a two-electron QD under a tunable perpendicular magnetic field if the effective confinement potential is indeed the three dimensional (3D)HO[10].

2.2 Classification of Low Dimensional Materials

Low-dimensional structures are usually classified according to the number of reduced dimensions they have. More precisely, the dimensionality refers to the number of degrees of freedom in the particle momentum. Accordingly, depending on the dimensionality, the following classification is made:

Three-dimensional (3D) structure or bulk structure: No quantization of the particle motion occurs, i.e., the particle is free. There is no motion of free electrons and energy is quantized.

Two-dimensional (2D) structure or quantum well: Quantization of the particle motion occurs in one direction, while the particle is free to move in the other two directions. Where the motion of particles is restricted in one dimension, but they are free to move in the other two dimensions. Hence they are 2D structure.

One-dimensional (1D) structure or quantum wire: Quantization occurs in two directions, leading to free movement along only one direction. Where the motion of

particles is restricted in two dimensions, but they are free to move in one dimension. Hence they are 1D structure.

Zero-dimensional (0D) structure or quantum dot (sometimes called "quantum box"): Quantization occurs in all three directions [11]. Where motion of particles is restricted in all the three dimensions. They are zero-dimensional (0D). Semiconductor quantum dots which are confined in all the three directions show quantization of energy levels. Self assembled quantum dots, among all the types of quantum dots, self assembled quantum dots are attractive due to the potential applications in opto-electronic devices especially in optical lasers [12]. The development of semiconductor nanostructures, such as quantum boxes and dots, by the use of different techniques has attracted much theoretical and experimental attention [13].

2.3 Physics of Quantum Dots

The difference between quantum dot and its corresponding bulk semiconductor will be explained more by quantum mechanics. Classical mechanics fails to describe about the position and motion of localized particles in a quantum dot because of its deterministic view which cannot be applied to atoms. In quantum mechanics, the motion of charge carriers will be described by wave function. The quantum confinement will split the bulk conduction and valence bands in to a series of discrete energy levels in spherical quantum dots. Due to quantum dot confinement electrons will be excited from the valence band to the conduction band creating a hole in the valence band. The absence of negative charge in the valence band makes the holes effective charge to be positive, thus attracting electrons. This

pair's forms hydrogen atom that is loosely bound. In the study of quantum wires and quantum wells the 1D and 2D potential well models could be applied to predict energy levels due to the confinement. In the case of quantum dots exaction will be applied. In strong confinement, the size of quantum dots is comparable to the effective Bohr radius(a_o) of the exciton [14] which is given by:

$$a_o = \frac{\epsilon \hbar^2}{me^2} \tag{2.1}$$

where ϵ is dielectric constant,m is effective mass of exaction,$\hbar$ is the reduced planks constant and e is electronic charge.

Quantum mechanics describes that electrons in the semiconductor crystal has a wave like representation. The motion of electrons in quantum dot is confined to a region with dimensions comparable to the De Broglie wave length(λ_{DB}) of the particle. The wave length of this wave is inversely proportional to electrons momentum (19, 14, 18).

$$\lambda = \frac{h}{P} = \frac{2\pi\hbar}{P} \tag{2.2}$$

Now, we can estimate the critical size confinement using Heisenberg uncertainty principle,

$$\triangle X \triangle E \quad \cong \quad \hbar \tag{2.3}$$

where $\triangle X$ and $\triangle P$ are uncertainties of position and momentum of electron. According to Heisenberg uncertainty principle position(x) and momentum (p) of electron are correlated in that confinement to electron in space and effective mass of

electron where the uncertainty in energy is related to the momentum the crystal by:

$$E(k) = \frac{\hbar^2 k^2}{2m^*} \tag{2.4}$$

where, and we know that the crystal momentum is given by,$p = \hbar\, k$ which implies that $P = \sqrt{2m^* \triangle E}$. Size of quantum confinement of particle can be written as:

$$\triangle X = \frac{\hbar}{\sqrt{2m^* \triangle E}} \tag{2.5}$$

Quantum confinement excites electrons from the valence band to conduction band causing an increase band gap energy of quantum dots which enhances the electrical and optical application of quantum dots. For quantum effect to work properly in the actual devices the spacing of energy levels $\triangle E$ is related to the momentum p must be large in comparison to the thermal activation energy $K_B T$ where K_B is Boltzmann constant and T is the absolute temperature [19]. For room temperature operation this implies that

$$\triangle E >> K_B T \tag{2.6}$$

2.4 Optical Properties

A full understanding of the optical properties of semiconductors is thus deeply rooted in the foundation of modern state physics. The optical properties of a semiconductor can be defined as any property that involves the interaction between electromagnetic radiation or light and the semiconductor, including absorption, diffraction, polarization, reflection, refraction, and scattering effects. From the macroscopic viewpoint, the interaction of matter with electromagnetic radiation

is described by Maxwell's equations. The optical properties of matter are introduced into these equations as the constants characterizing the medium such as the dielectric constant, magnetic permeability, and electrical conductivity. From our optical viewpoint, we choose to describe the solid by the complex dielectric function $\varepsilon(\omega)$. The complex refractive index is the complex square root of the dielectric function. Its real and imaginary parts are the refractive index n and the extiniction coefficient K. These optical constants describe an electromagnetic wave in the medium of propagation; the refractive index n gives the phase shift of the wave, and the extinction coefficient k gives the attenuation of the wave. The field of optical spectroscopy is a very important area of science and technology since most of our knowledge about the structure of atoms, molecules, and solids is based upon spectroscopic investigations. Since the early 1950s, detailed knowledge about the various eigenstates present in semiconductors has emerged including energy bands, excitonic levels, impurity and defect levels, densities of states, energy level widths(lifetimes), symmetries, and changes in these conditions with temperature, pressure, magnetic field, electric field, etc. Most optical properties of semiconductors are integrally related to the particular nature of their electronic and vibrational structures. These electronic and vibrational dispersion relations are in turn related to the type of crystallographic structure, the particular atoms, and their bonding. The optical properties of semiconductors are often subdivided into those that are electronic and those that are vibrational (lattice related) in nature. The electronic properties concern processes involving the electronic states of the semiconductor, while the lattice properties involve vibrations of the lattice (absorption and creation

of phonons). Lattice properties are of considerable interest for heat dissipation, electronic transport, and lifetimes (broadenings) of electronic states, but it is the electronic properties which receive the most attention in semiconductors because of the technological importance of their practical applications. Modern-day semiconductor optoelectronic technololologies include lasers, light-emitting diodes, photodectetors, optical amplifiers, modulators, switches, etc, all of which exploit specific aspects of the electronic optical properties. The phenomena usually studied to obtain information on the optical properties of semiconductor are absorption, reflection or ellipsometry, photoconductivity, emission, light scattering, and modulation techniques[20].

In a direct band gap semiconductor, large numbers of electrons are excited from the valence band to conduction band. In semiconductor physics a direct band gap means that the minimum of the conduction band lies directly above the maximum of the valence band in the k space. In contrast, an indirect semiconductor refers to a semiconductor with a band gap in which the minimum energy in the conduction band is shifted by a k vector relative to the top of the valence band. Indirect transitions (nonvertical Transition) semiconductors such as Ge and Si have indirect gaps where the maximum valence band energy and minimum conduction band energy do not occur at the same k value. In this case, the electron cannot make a direct transition from the top of the valence band to the bottom of the conduction band because this would violate conservation of momentum. Such a transition can still take place but as at two-step process requiring the cooperation of another particle and which can then be described by second-order perturbation theory. Semicon-

ductor nanowires (NWs) are usually characterized by anisotropic optical properties like optical absorption, photoluminescence, photoconductivity, as well as generation of higher harmonics, and strongly depend on the polarization of exciting light [25]. Quantum dots, among all the low dimensional semiconductors, are quite interesting due to its quantum confinement and their exotic and unusual electronic and optical behavior [28]. A lot of work has been concentrated to study their linear and non-linear optical properties. The absorption coefficients and the changes of refractive index, among non-linear optical properties, have been discussed [29-31]. Further, there are many experimental reports which bring out the nonlinear optical properties of quantum dots and the enhancement of the biexciton binding energy [32], biexciton lasing [33] and a non- monotonic size dependence of the optical nonlinearity [34].

2.4.1 Nonlinear optics

Nonlinear optics is the study of phenomena that occur as a consequence of the modification of the optical properties of a material system by the presence of light. Typically, only laser light is sufficiently intense to modify the optical properties of a material system. The beginning of the field of nonlinear optics is often taken to be the discovery of second-harmonic generation by Franken et al. (1961), shortly after the demonstration of the first working laser by Maiman in 1960. Nonlinear optical phenomena are ”nonlinear” in the sense that they occur when the response of a material system to an applied optical field depends in a nonlinear manner on the strength of the optical field. For example, second-harmonic generation occurs as a

result of the part of the atomic response that scales quadratically with the strength of the applied optical field. Consequently, the intensity of the light generated at the second-harmonic frequency tends to increase as the square of the intensity of the applied laser light. In order to describe more precisely what we mean by an optical nonlinearity, let us consider how the dipole moment per unit volume or polarization P (t), of a material system depends on the strength E(t) of an applied optical field[11]. When light beam incident on optical medium, it may be: reflected, propagated and transmitted. Absorption occurs during the propagation if the frequency of the light is resonant with the transition frequencies of the atoms in the medium. When light of sufficient energy shines on to a material, it induces transitions of electrons from occupied (lower level) states below the Fermi energy to unoccupied (higher level) states above the Fermi energy. Photon is absorbed by exciting an electron from a lower level to higher level. Therefore optical absorption can lift electrons from completely full valence band to empty conduction band. Exciting electrons in a solid in addition to by photon absorption is exciting electron by other electron. This done by shining a beam of mono energetic electron at a sample. The conduction band and valence band are separated by an energy called band gap $E_g = E_c - E_v$, where Ec and Ev are energy of conduction band and valence band respectively. Absorption can occur at $\hbar\omega > E_g$. Absorption cannot occur at $\hbar\omega = E_g$. The lowest energy at which absorption can occur is given by difference in energy, $\hbar\omega_n = E_{en} - E_{hn}$, between the lowest state in the well on the conduction band and the lowest in the well in the valence band. Absorption can occur at higher energy. The strongest quantization occurs between corresponding states in

two bands, so that $n_e = n_h = n$. The strong absorption occurs at the frequency given by, $\hbar\omega_n = E_{n2} - E_{e1}$. The energy of the photons matches the difference in electric energy level. In solids this re-emission process is called luminescence. In other words luminescence is the general name given to the process of spontaneous emission of light by excited atoms in solid state materials. The photon emitted has a smaller energy than the absorbed photon. This reduction in the photon energy is called Stake's shift. Note Materials with direct band gaps are better than those with indirect band gaps because their absorption is higher [27]. In a direct band gap semiconductor, large numbers of electrons are excited from the valence band to conduction band. In semiconductor physics a direct band gap means that the minimum of the conduction band lies directly above the maximum of the valence band in the k space. In contrast, an indirect semiconductor refers to a semiconductor with a band gap in which the minimum energy in the conduction band is shifted by a k vector relative to the top of the valence band. Atoms jump to an excited stated state by absorption of photon, and then relax to an intermediate state, before re-emitting a photon by spontaneous emission as it falls back to the ground state. The optical properties of semiconductors at low light levels are often referred to as linear properties in contrast to the nonlinear optical properties described later [20]. There are many physical processes that control the amount of absorption or other optical properties of a semiconductor. In turn, these processes depend upon the Wavelength of radiation, the specific properties of the individual semiconductor being studied, and other external parameters such as pressure, temperature, etc. Just as the electrical properties of a semiconductor can be controlled by purposely

introducing impurity dopants (both p and n type) or affected by unwanted impurities or defects, so too are the optical properties affected by them. Thus, one can talk about intrinsic optical properties of semiconductors that depend upon their perfect crystalline nature and extrinsic properties that are introduced by impurities or defects. Properties such as the refractive index, absorption coefficient and reflectivity are independent of optical power [27]. Linear optics [23]:

Optical properties, such as the refractive index and the absorption coefficient are independent of light intensity.

The principle of superposition, a fundamental tenet of classical mechanics, holds. If input A produces response X and input B produces response Y then input (A+B) produces response $X + Y \rightarrow image$. The frequency of light cannot be altered by its passage through the medium. Light cannot interact with light; two beams of light in the same region of a linear optical medium can have no effect on each other. Thus light cannot control light. This approximation is only valid at low power levels [12]. With a high power laser it is possible to enter a different realm of behavior called nonlinear optics. Nonlinear optics is the study of phenomena that occur as a consequence of the modification of the optical properties of a material system by the presence of light [24]. In other words nonlinear optics is the study of the interaction of intense laser light with matter [14]. Typically, only laser light is sufficiently intense to modify the optical properties of a material system [24]. The beginning of the field of nonlinear optics is often taken to be the discovery of second-harmonic generation. We are concerned with the effects that light itself induces as it propagates through the medium. Nonlinear optical phenomena are ”nonlinear” in the

sense that they occur when the response of a material system to an applied optical field depends in a nonlinear manner on the strength of the optical field. For example, second harmonic generation occurs as a result of the part of the atomic response that scales quadratically with the strength of the applied optical field. Consequently, the intensity of the light generated at the Second-harmonic frequency tends to increase as the square of the intensity of the applied laser light. Nonlinear optics (When E is very high) [23]: Light can control light; photons do interact the optical properties of materials are described through the real and imaginary parts of the dielectric constant r. The dielectric constant is derived from the polarization p of the medium. In linear optics, we assume that P depends linearly on the electric field of the light wave, so that we can write:

$$\overrightarrow{p} = \epsilon_o \chi \overrightarrow{E}(t) \tag{2.7}$$

where χ is the electric susceptibility.

A full understanding of the optical properties of semiconductors is thus deeply rooted in the foundations of modern solid-state physics [14]. Low dimensional systems have revolutionized semiconductor physics. They rely on technology of hetrostructure, where the composition of a semiconductor can be changed on the scale of nanometers. For example sand which of GasAs between two layers of Alx-Gax-As acts like an elementary quantum well. The energy levels are widely separated if the well is narrow, and all electrons may be trapped in lowers levels. Motions parallel to the layers is not defected, however, so the electrons remain free in those directions. The result is a two dimensional gas and holes can be trapped in the same way. The

reduction of the size of a semiconductor system at nanometer scale gives rise to quantum size effects which can significantly change the energy spectrum of electrons and their behaviors. When one or more of the dimensions of a solid are reduced significantly its physical and chemical characteristics notably depart from those of solids. With reduction in size novel electrical, mechanical, magnetic and optical can be introduced. Low dimensional systems are usually classified according to the number of freedom in the particles-momentum.

In bulk (3D) structures the particles are free to move in all directions. Accordingly depending on dimensionality in two dimensional (2D) structures quantum well, the quantization of the particle motion occurs only in one direction, while the particles are free to move in the other two directions. In one dimensional semiconductor system (quantum) wire quantization's occurs in two directions leading to free movement along only one direction. However, in zero dimension (0D) semiconductor structure or quantum dot the quantization occurs in all three directions or energy is quantized. Quantum wells are thin layered semiconductor structures in which we can observe and control many quantum mechanical effects [15].

Because of our ability to tune the quantum dots in a precise fashion, they offer a wide variety of usage. Quantum dots have excellent electrical and electrical properties. They are therefore, attractive components for integration in to electronic devices. One advantage they have over traditional optoelectronic materials is that they exist in the solid state; solids tend to be more compact, easily cooled and allow for direct charge injection. In addition, quantum dots can interconvert light and electricity in a tunable manner dependent on crystal size. This allows for easy

wavelength selection. This is a significant improvement over Silicon-based materials require modification of their chemical composition to alter optical properties. Quantum dots can be used to absorb and emit light efficiency at any wavelength. This property enables them to form new kinds of lighting and improve the current laser technologies. Another problem with conventional layers in the need for cooling, and that the pulses are relatively slow. These parameters can be improved by the use of quantum dots. Quantum dots also can be used to produce efficient white light [16].

2.4.2 Linear and nonlinear optical susceptibilities

In order to describe more precisely what we mean by an optical nonlinearity, let us consider how the dipole moment per unit volume or polarization $\widetilde{P}(t)$, of a material system depends on the strength of an applied optical field. In the case of conventional (i.e., linear) optics, the induced polarization depends linearly on the electric field strength$\widetilde{E}(t)$ in a manner that can often be described by the relationship

$$\widetilde{P}(t) = \epsilon_o \chi^{(1)} \widetilde{E}(t) \tag{2.8}$$

where the constant of proportionality $\chi^{(1)}$ is known as the linear susceptibility and ϵ_o is the permittivity of free space. In nonlinear optics, the optical response can often be described by expressing the polarization $\widetilde{P}(t)$ as a power series in the field strength $\widetilde{E}(t)$ as

$$\widetilde{P}(t) = \epsilon_o[\chi^{(1)}\widetilde{E}(t) + \chi^{(2)}\widetilde{E}^2(t) + \chi^{(3)}\widetilde{E}^3(t) + ...] \tag{2.9}$$

The quantities $\chi^{(2)}$ and $\chi^{(3)}$ are known as the second- and third-order non- linear optical susceptibilities, respectively. For simplicity, we have taken the fields $\widetilde{P}(t)$ and $\widetilde{E}(t)$ to be scalar quantities. In general, the nonlinear susceptibilities depend on the frequencies of the applied fields, but under our present assumption of instantaneous response, we take them to be constants. We shall refer to $\widetilde{P}^{(2)}(t) = \epsilon_o \chi^{(2)} \widetilde{E}^2(t)$ as the second-order nonlinear polarization and $\widetilde{P}^{(3)}(t) = \epsilon_o \chi^{(3)} \widetilde{E}^{(3)}(t)$ to as the third-order nonlinear polarization. The linear optical susceptibility is the first order optical susceptibility only. From the second order to n^{th} number of orders are called non-linear optical susceptibilities [10].

2.5 Density Matrix Formalism

We calculate the nonlinear optical susceptibility through use of the density matrix formulation of quantum mechanics [10]. We use this formalism because it is cabable of treating effects, such as collisional broadening of the atomic resonances that cannot be treated by the simple theoretical formalism based on the atomic wave function. Let us begin by reviewing how the density matrix formalism follows from the basic law of quantum mechanics. If a quantum mechanical system (such as an atom) is known to be in particular quantum mechanical state that we designate S, we can describe all of the physical properties of the system interms of the wave function $\psi_S(r,t)$ appropriate to this state. This wave equation obeys the Schrodinger equation

$$i\hbar \frac{\partial \psi_S(r,t)}{\partial t} = \hat{H}\psi_S(r,t) \tag{2.10}$$

where $\hat{H}$ denotes the Hamiltonian operator of the system. We nassume that $\hat{H}_o$ can be represented as

$$\hat{H} = \hat{H}_o + V(t) \tag{2.11}$$

where $\hat{H}_o$ is the Hamiltonian for a free atom and $V(t)$ represents the interaction energy.

In quantum mechanics density matrix is defined as

$$\hat{\rho} = \mid \psi_S(r,t) >< \psi_S(r,t) \mid \tag{2.12}$$

where $\hat{\rho}$ is density matrix and it will be applied to calculate first order linear optical susceptibility and third order nonlinear optical susceptibility.

The third order non-linear optical interactions(i.e those described by a $\chi^{(3)}$ susceptibility) can occur for both centrosymmetric and noncentrosymmetric media. In order to describe more precisely what we mean by an optical non-linearity, let us consider how the dipole moment per unit volume, or polirization $\overrightarrow{P}(t)$ of a material system depends on the strength $\overrightarrow{E}(t)$ of an applied optical field.

The third-order correlation to the density matrix is given by[11]

$$\rho^{(3)}_{nm} = exp[-(\omega_{nm} + \gamma_{nm})t] \int_{\infty}^{t} \frac{-i}{\hbar} \left[\hat{v}, \hat{\rho}^2\right]_{nm} exp(i\omega_{nm} + \gamma_{nm})t'dt' \tag{2.13}$$

where the commutator can be represented explicity as

$$[\hat{v}, \hat{\rho}^2]_{nm} = -\sum_{v} (\mu_{nv}\rho^{(2)}_{vm} - \rho^{(2)}_{nv}\mu_{vm}).\hat{E}(t) \tag{2.14}$$

Expressions for $\rho^{(2)}_{vm}$ and $\rho^{(2)}_{nv}$ are very complicated, we use the abbreviated notation introduced there:

$$\rho^{(2)}_{vm} = \sum_{t} \sum_{pq} K_{vlm} exp(-i(\omega_p + \omega_q)) \tag{2.15}$$

where K_{vml} has been displayed explicitly. We also represented the electric field as

$$\widetilde{E}(t) = \sum_{r} E(\omega r) e^{-i\omega_r t} \tag{2.16}$$

The commutator thus becomes

$$[\hat{v}, \hat{\rho}^2]_{nm} = -\sum_{vl}\sum_{pqr}[\mu_{nv}.E(\omega_r)]K_{vml}e^{-i(\omega_p+\omega_q+\omega_r)t}$$

$$+\sum_{vl}\sum_{pqr}[\mu_{vm}.E(\omega_r)]K_{nv}e^{-(\omega_p+\omega_q+\omega_r)t} \tag{2.17}$$

The nonlinear polarization oscillating at frequency $\omega_p + \omega_q + \omega_r$ is given by

$$P_k(\omega_p+\omega_q+\omega_r) = \epsilon_0 \sum_{hij}\sum_{pqr}\chi^{(3)}_{kjih}(\omega_p+\omega_q+\omega_r,$$

$$\omega_r, \omega_q, \omega_p) X E_j(\omega_r) E_i(\omega_q) E_h(\omega_p) \tag{2.18}$$

where $\omega_p, \omega_q, \omega_r$ are permutation frequencies.

2.6 The Electronic States of Quantum Dots

The qualitative analysis of the possible states of quantum dots with two or more electrons using the general theory of systems of identical particles including the weak magnetic interactions due to the interactions of the electron spins with each other and with orbital motion. We consider theoretical frame work for the two dimensional quantum dots. We take into account for electron-electron interaction with the coulomb potential, the Pauli- exclusion principle and external fields. We can reduce the number of spatial dimensions from three to two, that is from three dimensions one dimension is fixed by using the manufacturing techniques, which

forces the electrons to occupy a planar region. Quantum dots with this confining potential are often humorous parabolic quantum dots, due to the parabolic nature of the potential.

3

Materials and Methodology

To carry out the study it is important to use the following method and materials. These are: study site and period, method of approach, materials used, data analysis and ethical considerations.

3.1 Study Site and Period

The study would have been conducted at Jimma University, which is the first institution in Ethiopia in designing community based education, in department of physics from september 2016 to september 2017.

3.2 Method of Research

To achieve the stated objectives and problem, analytical methods with computational analysis would be used.

3.3 Materials

To study the given problem, we tried to carry out a deep survey of literatures such as journals, papers and books that are related to the title of the problem.

3.4 Graphical(Computational)

We used MATLAB and MATHEMATHICA software for graphical and computational verification of the analytical results.

3.5 Ethical Issues

To be legal for collecting all the information and materials for the study, it is important to have a permission letter. Therefore, I have got a letter of a permission from ethical committe of the college.

4

Result and Discussion

In this thesis we investigate the optical properties of three dimensional ZnO quantum dot. We first solve analytically the schrodinger equation to obtain the energy levels and wave functions. Using the obtained energy eigenvalues and wave functions, we calculate the optical properties of a spherical quantum dot. The purpose of this study is to obtain the changes in the linear optical susceptibility and third order nonlinear optical susceptibility of the quantum dot(QD).

4.1 Introduction

Quantum dots (QDs) and quantum wells (QWs) with one electron and their optical properties are widely studied. However, there is possibility to put two,three,and more electrons in a quantum dot, where coulomb interactions are taken into account. For determining the energy eigenvalues of two electron ZnO quantum dot, we employed variational method. We calculated the ground and first excited state energies of two electron quantum dots (QDs).

4.2 Mathematical Formulation of the Problem

- The hamiltonian of two interacting electrons in three dimensional quantum dot is given by:

$$\hat{H} = -\frac{\hbar^2}{2m*}\frac{d^2}{dr_1^2} - \frac{\hbar^2}{2m*}\frac{d^2}{dr_2^2} + \frac{1}{2}m*\omega^2 r_1^2 + \frac{1}{2}m*\omega^2 r_2^2 + \frac{e^2}{\epsilon \mid \overrightarrow{r_1} - \overrightarrow{r_2} \mid} \quad (4.1)$$

Using $\overrightarrow{r_1} = \sqrt{\frac{\hbar}{m*\omega}}\rho_1$, $\overrightarrow{r_2} = \sqrt{\frac{\hbar}{m*\omega}}\rho_2$, $dr_1 = \sqrt{\frac{\hbar}{m*\omega}}d\rho_1$, $dr_2 = \sqrt{\frac{\hbar}{m*\omega}}d\rho_2$ by substituting these equations in to Eq.(4.1) the dimensionless hamiltonian is:

$$\hat{H} = -\frac{1}{2}\frac{d^2}{d\rho_1^2} - \frac{1}{2}\frac{d^2}{d\rho_2^2} + \frac{1}{2}\rho_1^2 + \frac{1}{2}\rho_2^2 + \frac{\lambda}{\mid \overrightarrow{\rho_1} - \overrightarrow{\rho_2} \mid} \quad (4.2)$$

$$\hat{H} = -\frac{1}{2}(\nabla_1^2 + \nabla_2^2) + \frac{1}{2}(\rho_1^2 + \rho_2^2) + \frac{\lambda}{\mid \overrightarrow{\rho_1} - \overrightarrow{\rho_2} \mid} \quad (4.3)$$

where ρ_1 and ρ_2 are dimensionless co-ordinates of the first and the second electrons, which are measured in units of $\sqrt{\frac{\hbar}{m*\omega}}$, with m* is the effective mass of the electron and ω is the frequency of a confining potential, $\lambda = \frac{1}{\epsilon}\sqrt{\frac{me^4}{\hbar^2}\frac{m*}{m}\frac{1}{\hbar\omega}}$ is the coupling constant of coulomb interaction of two electrons, and ϵ is the dielectric constant of a quantum dot. For $\omega = 10THz$, $\frac{m*}{m} = 0.21$, and $\epsilon = 8.5$ for ZnO, we get $\lambda = 1.1$. Due to this large coupling constant, variational technique is more appropriate.

- The trial ground state wave function for two electron quantum dot is chosen as

$$\psi_o(\overrightarrow{\rho_1}, \overrightarrow{\rho_2}) = Aexp{-}\frac{\beta}{2}(\rho_1^2 + \rho_2^2) \quad (4.4)$$

where A is normalization constant and β is variational parameter[35].

- The trial ground state wave equation is the combination of the two single spatial particle state, so we can write Eq.(4.4) separately as

$$\psi_o(\rho_1) = A_1 exp(-\frac{\beta}{2}\rho_1^2) \tag{4.5}$$

$$\psi_o(\rho_2) = A_2 exp(-\frac{\beta}{2}\rho_2^2) \tag{4.6}$$

where A_1 and A_2 are normalization constants of the first and the second electrons respectively with $A = A_1 A_2$.

The normalized wave function should satisfies:

$$< \psi_o(\rho_1) \mid \psi_o(\rho_1) > = 1 \tag{4.7}$$

$$< \psi_o(\rho_2) \mid \psi_o(\rho_2) > = 1 \tag{4.8}$$

The above Eqs.(4.7) and (4.8) can be expressed as in position space integral (using spherical coordinate)

$$\int \psi_o^*(\rho_1)\psi_o(\rho_1)\rho_1^2 d\phi_1 sin\theta_1 d\theta_1 d\rho_1 = 1 \tag{4.9}$$

$$\int \psi_o^*(\rho_2)\psi_o(\rho_2)\rho_2^2 d\phi_2 sin\theta_2 d\theta_2 d\rho_2 = 1 \tag{4.10}$$

$$4\pi A_1^2 \int_0^\infty e^{-\beta\rho_1^2}\rho_1^2 d\rho_1 = 1 \tag{4.11}$$

$$4\pi A_2^2 \int_0^\infty e^{-\beta\rho_2^2}\rho_2^2 d\rho_2 = 1 \tag{4.12}$$

Evaluating the above equations Eq. (4.11) and (4.12) and solving for normalization constant we obtain

$$A_1 = \left(\frac{\beta}{\pi}\right)^{\frac{3}{4}} \tag{4.13}$$

$$A_2 = \left(\frac{\beta}{\pi}\right)^{\frac{3}{4}} \tag{4.14}$$

$$A = A_1 A_2 \tag{4.15}$$

$$A = \left(\frac{\beta}{\pi}\right)^{\frac{3}{4}} \star \left(\frac{\beta}{\pi}\right)^{\frac{3}{4}} \tag{4.16}$$

$$A = \left(\frac{\beta}{\pi}\right)^{\frac{3}{2}} \tag{4.17}$$

From Eq.(4.4) the normalized trial ground state wave function becomes[36]

$$\Psi_o(\overrightarrow{\rho}_1, \overrightarrow{\rho}_2) = \left(\frac{\beta}{\pi}\right)^{\frac{3}{2}} exp[-\frac{\beta}{2}(\rho_1^2 + \rho_2^2)] \tag{4.18}$$

- The trial excited state wave function for two electron quantum dot is chosen as

$$\psi_1(\overrightarrow{\rho}_1, \overrightarrow{\rho}_2) = Be^{-\frac{\gamma}{2}(\rho_1^2+\rho_2^2)}\rho_1 cos\theta_1 \rho_2 cos\theta_2 \tag{4.19}$$

where B is normalization constant and γ is variational parameter to be optimized.

- The trial excited state wave equation is the combination of the two single spatial particle state, so Eq(4.19) separately as

$$\psi_1(\overrightarrow{\rho_1}) = B_1 e^{-\frac{\gamma}{2}\rho_1^2}\rho_1^2 cos\theta_1 \tag{4.20}$$

$$\psi_1(\overrightarrow{\rho_2}) = B_2 e^{-\frac{\gamma}{2}\rho_2^2}\rho_2^2 cos\theta_2 \tag{4.21}$$

where B_1 and B_2 are normalization constants of the first and the second electrons respectively with $B = B_1 B_2$

The normalized wave function should satisfies:

$$< \psi_1(\overrightarrow{\rho_1}) \mid \psi_1(\overrightarrow{\rho_1}) > = 1 \tag{4.22}$$

$$< \psi_1(\overrightarrow{\rho_2}) \mid \psi_1(\overrightarrow{\rho_2}) > = 1 \tag{4.23}$$

The above equations Eq. (4.22) and (4.23) and solving for normalization constants by the same step we have followed for ground state obtain

$$B = B_1 B_2 \tag{4.24}$$

$$B = \frac{2\gamma^{\frac{5}{2}}}{\pi^{\frac{3}{2}}} \tag{4.25}$$

From Eq.(4.19) the normalized trial excited state wave function becomes

$$\psi_1(\overrightarrow{\rho_1}, \overrightarrow{\rho_2}) = \frac{2\gamma^{\frac{5}{2}}}{\pi^{\frac{3}{2}}} e^{-\frac{\gamma}{2}(\rho_1^2+\rho_2^2)} \rho_1 cos\theta_1 \rho_2 cos\theta_2 \tag{4.26}$$

4.3 The Ground State Energy Level of Two Electron Quantum Dot (QD)

- The ground state energy is given by

$$E^{(0)} = < \psi_0(\rho_1) \mid -\frac{1}{2}\frac{d^2}{d\rho_1^2} \mid \psi_0(\rho_1) > + < \psi_0(\rho_2) \mid -\frac{1}{2}\frac{d^2}{\rho_2^2} \mid \psi_0(\rho_2) > +$$

$$< \psi_0(\rho_1) \mid \frac{1}{2}\rho_1^2 \mid \psi_0(\rho_1) > + < \psi_0(\rho_2) \mid \frac{1}{2}\rho_2^2 \mid \psi_0(\rho_2) > +$$

$$< \psi_0(\rho_1, \rho_2) \mid \frac{\lambda}{\mid \overrightarrow{\rho_1} - \overrightarrow{\rho_2} \mid} \mid \psi_0(\rho_1, \rho_2) > \tag{4.27}$$

where $-\frac{1}{2}\frac{d^2}{d\rho_1^2} = -\frac{1}{2}\nabla_1^2$ and $-\frac{1}{2}\frac{d^2}{d\rho_2^2} = -\frac{1}{2}\nabla_2^2$

$$E^{(0)} = K_1 + K_2 + V_1 + V_2 + V_{12} \tag{4.28}$$

with K_1 and K_2 are the kinetic energy of the first and the second electrons respectively, V_1 and V_2 are the potential energy of the first and the second electrons respectively and V_{12} is the coulomb interaction energy. When we solve Eq.(4.27) one

by one separately

$$K_1 = < \psi_0(\rho_1) \mid -\frac{1}{2}\frac{d^2}{d\rho_1^2} \mid \psi_0(\rho_1) > \tag{4.29}$$

$$K_1 = < \psi_0(\rho_1) \mid -\frac{1}{2}\nabla_1^2 \mid \psi_0(\rho_1) > \tag{4.30}$$

$$K_1 = -\frac{1}{2}\left(\frac{\beta}{\pi}\right)^{\frac{3}{2}} \int e^{-\frac{\beta}{2}\rho_1^2}\left[\frac{\partial^2}{\partial\rho_1^2} + \frac{2}{\rho_1}\frac{\partial}{\partial\rho_1} + \frac{1}{\rho_1^2}\frac{\partial^2}{\partial\theta_1^2} + \frac{1}{\rho_1^2}cot\theta_1\frac{\partial}{\partial\theta_1}\right]e^{-\frac{\beta}{2}\rho_1^2}d^3\rho_1$$

$$K_1 = -\frac{1}{2}\left(\frac{\beta}{\pi}\right)^{\frac{3}{2}} \int e^{-\frac{\beta}{2}\rho_1^2}\left[\frac{\partial^2}{\partial\rho_1^2} + \frac{2}{\rho_1}\frac{\partial}{\partial\rho_1}\right]e^{-\frac{\beta}{2}\rho_1^2}d^3\rho_1$$

$$K_1 = -\frac{1}{2}\left(\frac{\beta}{\pi}\right)^{\frac{3}{2}} \int_0^\infty \int_0^\pi \int_0^{2\pi} e^{-\frac{\beta}{2}\rho_1^2}\Big(- 3\beta e^{-\frac{\beta}{2}\rho_1^2} + \beta^2\rho_1^2 e^{-\frac{\beta}{2}\rho_1^2}\Big)\rho_1^2 d\rho_1 sin\theta_1 d\theta_1 d\phi_1$$

$$K_1 = 4\pi\Big(- \frac{1}{2}\Big)\Big(\frac{\beta}{\pi}\Big)^{\frac{3}{2}} \int_0^\infty e^{-\frac{\beta}{2}\rho_1^2}\Big(- 3\beta e^{-\frac{\beta}{2}\rho_1^2} + \beta^2\rho_1^2 e^{-\frac{\beta}{2}\rho_1^2}\Big)\rho_1^2 d\rho_1$$

$$K_1 = \Big(4\pi\Big)\Big(- \frac{1}{2}\Big)\Big(\frac{\beta}{\pi}\Big)^{\frac{3}{2}}\Big(- \frac{3\sqrt{\pi}}{8\sqrt{\beta}}\Big)$$

$$K_1 = \frac{3}{4}\beta \tag{4.31}$$

The same procedure for the second particle also gives us the same average kinetic energy. Similarly solving for K_2

$$K_2 = \frac{3}{4}\beta \tag{4.32}$$

The total kinetic energy is

$$K = K_1 + K_2 \tag{4.33}$$

$$K = \frac{3}{4}\beta + \frac{3}{4}\beta$$

$$K = \frac{3}{2}\beta \tag{4.34}$$

The energy due to the confining potential is

$$V = V_1 + V_2 \tag{4.35}$$

$$V_1 = \frac{1}{2}\left(\frac{\beta}{\pi}\right)^{\frac{3}{2}} \int_0^{\infty}\int_0^{\pi}\int_0^{2\pi} e^{-\beta\rho_1^2}\rho_1^2\rho_1^2 d\rho_1 sin\theta_1 d\theta_1 d\phi_1$$

$$V_1 = \frac{1}{2}\left(\frac{\beta}{\pi}\right)^{\frac{3}{2}} (4\pi)\int_0^{\infty} \rho_1^4 e^{-\beta\rho_1^2} d\rho_1$$

$$V_1 = 2\pi\left(\frac{\beta}{\pi}\right)^{\frac{3}{2}}\left(-\frac{\sqrt{\pi}}{4}\right)\left(-\frac{3}{2}\beta^{-\frac{5}{2}}\right)$$

$$V_1 = \frac{3}{4\beta} \tag{4.36}$$

The same procedure for the second confining potential also gives us the same value. By solving for V_2,

$$V_2 = \frac{3}{4\beta} \tag{4.37}$$

The total confining potential is obtained by substituting Eq.(4.36) and (4.37) in to Eq.(4.35)

$$V = \frac{3}{4\beta} + \frac{3}{4\beta}$$

$$V = \frac{3}{2\beta} \tag{4.38}$$

- The non interaction part of the ground state energy is given by the sum of total kinetic energy and potential energy:

$$E^{nonint} = K + V \tag{4.39}$$

$$E^{nonint} = \frac{3}{2}\beta + \frac{3}{2\beta}$$

$$E^{nonint} = \frac{3}{2}\left(\beta + \frac{1}{\beta}\right) \tag{4.40}$$

The coulomb part is

$$T = \lambda \int \frac{\mid \psi_0(\rho_1) \mid^2 \mid \psi_0(\rho_2) \mid^2}{\mid \rho_1 - \rho_2 \mid} d^3\rho_1 d^3\rho_2 \tag{4.41}$$

$$T = \lambda(4\pi)^2 \left(\frac{\beta}{\pi}\right)^3 \int_0^\infty \frac{e^{-\beta(\rho_1^2+\rho_2^2)} \rho_1^2 d\rho_1 \rho_2^2 d\rho_2}{\rho_{12}} \tag{4.42}$$

Using spherical harmonics[35]

$$\frac{1}{\rho_{12}} = \frac{1}{\rho_1} \sum_{l=0}^{\infty} \left(\frac{\rho_2}{\rho_1}\right)^l P_l(cos\theta), \rho_1 > \rho_2 \tag{4.43}$$

$$\frac{1}{\rho_{12}} = \frac{1}{\rho_2} \sum_{l=0}^{\infty} \left(\frac{\rho_1}{\rho_2}\right)^l P_l(cos\theta), \rho_1 < \rho_2 \tag{4.44}$$

where $P_l(cos\theta)$ is legendre polynomial, θ is the angle between $\overrightarrow{\rho_1}$ and $\overrightarrow{\rho_2}$. According to the addition theorem for spherical harmonics

$$P_l(cos\theta) = P_l(cos\theta_1)P_l(cos\theta_2) + 2\sum_{m=1}^{l} \frac{(l-m)!}{(l+m)!} P_l^m(cos\theta_2) cosm(\phi_1 - \phi_2)$$

where θ_1,ϕ_1 and θ_2,ϕ_2 are the polar angles of the of the vectors $\overrightarrow{\rho_1}$ and $\overrightarrow{\rho_2}$ respectively. Using equations (4.43) and (4.44), the integral becomes

$$T = \lambda(4\pi)^2 \left(\frac{\beta}{\pi}\right)^3 \int_0^\infty \left[\int_0^{\rho_1} \frac{1}{\rho_1} e^{-\beta(\rho_1^2+\rho_2^2)} \rho_2^2 d\rho_2 + \int_{\rho_1}^\infty \frac{1}{\rho_2} e^{-\beta(\rho_1^2+\rho_2^2)} \rho_2^2 d\rho_2 \right] \rho_1^2 d\rho_1$$

$$T = \lambda(4\pi)^2 \left(\frac{\beta}{\pi}\right)^3 \left(\frac{\sqrt{\pi}}{4\beta^{\frac{3}{2}}}\right) \left(\frac{1}{2\sqrt{2\beta}}\right)$$

$$T = \lambda\sqrt{\frac{2}{\pi}}\beta^{\frac{1}{2}} \tag{4.45}$$

As the result, the ground state energy is by or substituting Eqs.(4.40) and (4.45) in to Eq.(4.28) becomes

$$E^{(0)} = \frac{3}{2}\beta + \frac{3}{2\beta} + \lambda\sqrt{\frac{2}{\pi}}\beta^{\frac{1}{2}} \tag{4.46}$$

The minimum energy will be obtained for the condition,

$$\frac{dE^{(0)}}{d\beta} = 0$$

$$\frac{dE^{(0)}}{d\beta} = \frac{d}{d\beta}\left(\frac{3}{2}\beta + \frac{3}{2\beta} + \lambda\sqrt{\frac{2}{\pi}}\beta^{\frac{1}{2}}\right)$$

$$\frac{dE^{(0)}}{d\beta} = \beta^2 + \frac{\lambda}{3}\sqrt{\frac{2}{\pi}}\beta^{\frac{3}{2}} - 1 = 0$$

$$\beta^2 + \frac{\lambda}{3}\sqrt{\frac{2}{\pi}}\beta^{\frac{3}{2}} - 1 = 0 \tag{4.47}$$

The numerical solution of β, we have done before the value of $\lambda = 1$,

$$\beta^2 + \frac{1}{3}\sqrt{\frac{2}{3.14}}\beta^{\frac{3}{2}} - 1 = 0 \Rightarrow \beta = 0.88$$

The numerical solution of ground state energy is calculated as

$$E^{(0)}(\lambda) = \frac{3}{2}(0.88) + \frac{3}{2(0.88)}\lambda\sqrt{\frac{2}{3.14}}(0.88)^{\frac{1}{2}} \tag{4.48}$$

$$E^{(0)}(1) = \frac{3}{2}(0.88) + \frac{3}{2(0.88)}\sqrt{\frac{2}{3.14}}(0.88)^{\frac{1}{2}} \tag{4.49}$$

$$E_0(1) = 3.77\hbar\omega \tag{4.50}$$

λ	1	1.5	2	2.5	3
β	0.883	0.834	0.791	0.752	0.718
$E_{(0)}$	3.773	4.143	4.502	4.852	5.194

Table 4.1: The ground state energy E_0 and variational parameter β for different λ.

4.4 The Excited State Energy Level of Two Electron Quantum Dot

- The excited state energy is given by

$$E^{(1)} = < \psi_1(\rho_1) \mid -\frac{1}{2}\frac{d^2}{d\rho_1^2} \mid \psi_1(\rho_1) > + < \psi_1(\rho_2) \mid -\frac{1}{2}\frac{d^2}{\rho_2^2} \mid \psi_1(\rho_2) > +$$

$$< \psi_1(\rho_1) \mid \frac{1}{2}\rho_1^2 \mid \psi_1(\rho_1) > + < \psi_1(\rho_2) \mid \frac{1}{2}\rho_2^2 \mid \psi_1(\rho_2) > +$$

$$< \psi_1(\rho_1,\rho_2) \mid \frac{\lambda}{\mid \overrightarrow{\rho_1} - \overrightarrow{\rho_2} \mid} \mid \psi_1(\rho_1,\rho_2) > \tag{4.51}$$

where $-\frac{1}{2}\frac{d^2}{d\rho_1^2} = -\frac{1}{2}\nabla_1^2$ and $-\frac{1}{2}\frac{d^2}{d\rho_2^2} = -\frac{1}{2}\nabla_2^2$

$$E^{(1)} = K_1 + K_2 + V_1 + V_2 + V_{12} \tag{4.52}$$

The same procedure for the first excited state energy as the ground state energy or by solving Eq.(4.51) we obtain $K_1 = \frac{5}{4}\gamma$ and $K_2 = \frac{5}{4}\gamma$

The total kinetic energy for the first excited state energy is given by:

$$K = K_1 + K_2 \tag{4.53}$$

$$K = \frac{5}{4}\gamma + \frac{5}{4}\gamma$$

$$K = \frac{5}{2}\gamma \tag{4.54}$$

The energy due to the confining potential is given by $V_1 = \frac{5}{4\gamma}$ and $V_2 = \frac{5}{4\gamma}$

The total potential energy is

$$V = V_1 + V_2 \tag{4.55}$$

$$V = \frac{5}{4}\left(\frac{1}{\gamma} + \frac{1}{\gamma}\right)$$

$$V = \frac{5}{2\gamma} \tag{4.56}$$

The non interacting part of the first excited state energy is given by:

$$E^{nonint} = \frac{5\gamma}{2} + \frac{5}{2\gamma} \tag{4.57}$$

The coulomb part of first excited state energy is calculated by same procedure we follow for ground state energy

$$V_{12} = \frac{49\lambda\sqrt{\gamma}}{30\sqrt{2\pi}} \tag{4.58}$$

By substituting Eqs.(4.57) and (4.58) in to (4.52)

$$E^{(1)} = \frac{5\gamma}{2} + \frac{5}{2\gamma} + \frac{49\lambda\sqrt{\gamma}}{30\sqrt{2\pi}} \tag{4.59}$$

The minimum energy will be obtained for the condition, $\frac{dE^{(1)}(\lambda)}{d\gamma} = 0$

$$\frac{d}{d\gamma}\left(\frac{5\gamma}{2} + \frac{5}{2\gamma} + \frac{49\lambda\sqrt{\gamma}}{30\sqrt{2\pi}}\right) = 0 \tag{4.60}$$

$$\gamma^2 + \frac{4\lambda}{150\sqrt{2\pi}}\gamma^{\frac{3}{2}} - 1 = 0 \tag{4.61}$$

The numerical solution for $\lambda = 1$, first $\gamma^2 + \frac{4(1)}{150\sqrt{2(3.14)}}\gamma^{\frac{3}{2}} - 1 = 0$, by solving this equation the value of γ is given by $\gamma = 0.94$ and $E_1(\lambda) = (0.94)^2 + \frac{4(1)}{150\sqrt{2(3.14)}}(0.94)^{\frac{3}{2}} - 1$

$$E_1(1) = 5.64\hbar\omega \tag{4.62}$$

λ	1	1.5	2	2.5	3
γ	0.939	0.911	0.885	0.860	0.837
$E_1(\lambda)$	5.641	5.954	6.263	6.566	6.866

Table 4.2: The first excited state energy E_1 and variational parameter γ for different λ when two electrons are in the state n=0, l=1, m=0.

4.5 Optical Properties of Three Dimensional Two Electron Quantum Dot

To study the optical properties of three dimensional two electron quantum dot we used the density matrix approach. Density matrix is a very useful tool that enlarges the concept of state from a vector in which all the properties of a quantum system can be extracted. Density matrix formalism is used to calculate the changes of refractive index and absorption coefficient of the quantum dot corresponds to an optical transition between the ground and excited states. The quantum dot is excited by applying an electromagnetic field,

$$E(t) = Ee^{i\omega t} + Ee^{-i\omega t} \tag{4.63}$$

Let us consider that the incident electromagnetic wave is polarized along z-direction of the quantum dot. Then the, time dependent equation of the density matrix operator ρ is given by

$$\frac{\partial \rho}{\partial t} = \frac{1}{i\hbar}\left[\hat{H}_0 - qzE(t), \rho\right] - \Gamma(\rho - \rho^{(0)}) \tag{4.64}$$

With H_0 is the hamiltonian of the quantum dot in the absence of electromagnetic field $\overrightarrow{E}$,q is the charge , $\rho^{(0)}$ is the unperturbed density matrix operator and Γ is the phenomenological damping constant due to collision among electrons and electron phonon interactions. Equation (4.64) can be solved using a usual iterative method[10].

$$\rho(t) = \sum_n \rho^{(n)}(t) \tag{4.65}$$

With $\frac{\partial \rho_{ij}^{(n+1)}}{\partial t} = \frac{1}{i\hbar}\left[(\hat{H}_o, \rho^{(n+1)})_{ij} - i\hbar\Gamma_{ij}\rho_{ij^{(n+1)}}\right] - \frac{1}{i\hbar}\left(qz, \rho^{(n)}\right)_{ij} E(t)$

Considering the two level electronic system. The electronic polarization P(t), and susceptibility $\chi(t)$ are defined by the dipole operator μ and the density matrix ρ

$P(t) = \epsilon_0\chi(\omega)\overrightarrow{E}e^{-i\omega t} + \epsilon_0\chi(-\omega)\overrightarrow{E}^* e^{i\omega t}$

$$P(t) = \frac{1}{V}Tr(\rho\mu) \tag{4.66}$$

where V and ρ are the volume and the one electron density matrix of the system respectively, ϵ_0 is the permitivity of the free space and the symbol Tr denotes the summation over the diagonal elements of matrix. The mathematical relations of the linear $\chi^{(1)}$ and the nonlinear $\chi^{(3)}$ susceptibility are obtained. The linear term is,

$$\epsilon_0\chi^{(1)}(\omega) = \frac{\rho_v \mid \mu_{10} \mid^2}{E_{10} - \hbar\omega - i\hbar\Gamma_{01}} \tag{4.67}$$

The third order term is

$$\epsilon_0\chi^{(1)}(\omega) = -\frac{\rho_v \mid \mu_{10} \mid^2 \mid E \mid^2}{E_{10}-\hbar\omega-i\hbar\Gamma_{01}}\left[\frac{4 \mid \mu_{10} \mid^2}{(E_{10}-\hbar\omega)^2+(\hbar\Gamma_{01})^2}-\frac{(\rho_{11}-\rho_{00})^2}{(E_{10}-i\hbar\Gamma_{01})(E_{10}-\hbar\omega-i\hbar\Gamma_{01})}\right]$$

The real part of first order susceptibility is given by:

$$Re\chi^1(\omega) = \frac{1}{\epsilon_0} \mid \mu_{10} \mid^2 \rho_v\left[\frac{E_{10}-\hbar\omega}{(E_{10}-\hbar\omega)^2+(\hbar\Gamma_{01})^2}\right] \tag{4.68}$$

The imaginary part of first order susceptibility is given by:

$$I_m\chi^{(1)}(\omega) = \frac{1}{\epsilon_0}\frac{\mid \mu_{10} \mid^2 \rho_v\Gamma_{01}}{(E_{10}-\hbar\omega)^2+(\hbar\Gamma_{01})^2} \tag{4.69}$$

The real part of third order susceptibility is given by:

$$\begin{aligned} Re\chi^3(\omega) = {} & \frac{-\mu c}{2n_r\epsilon_0} \mid \mu_{10} \mid^2 \rho_v\left[\frac{I}{[(E_{10}-\hbar\omega)^2+(\hbar\Gamma_{01})^2]^2}\right] \\ & \left[4(E_{10}-\hbar\omega) \mid \mu_{10} \mid^2 -\frac{(\rho_{11}-\rho_{00})^2}{(E_{10})^2+(\hbar\Gamma_{01})^2}\right. \\ & \left(\left(E_{10}-\hbar\omega)[E_{10}(E_{10}-\hbar\omega)-(\hbar\Gamma_{01})^2\right]\right. \\ & \left.\left.-(\hbar\Gamma_{01})^2(2E_{10}-\hbar\omega)\right)\right] \end{aligned} \tag{4.70}$$

where ρ_v is the carrier density in this system, μ is the permeability of the system, $E_{ij} = E_i - E_j$ is the energy interval of two different electronic states, μ_{ij} is the matrix

elements that is defined by $\mu_{ij} = |< \psi_i \mid q_x \mid \psi_j >|$ (i,j=1,2), and c is the speed of light. The imaginary part of third order susceptibility is given by:

$$I_m\chi^{(3)}(\omega) = -\left(\frac{I}{2\epsilon_R n_r c}\right)\frac{\mid \mu_{10} \mid^2 \rho_v \hbar\Gamma_{01}}{[(E_{10}-\hbar\omega)^2+(\hbar\Gamma_{01})^2]^2}$$

$$\left[4 \mid \mu_{10} \mid^2 - \frac{\mid (\rho_{11}-\rho_{00}) \mid^2 [3E_{10}^2 - 4E_{10}\hbar\omega + \hbar^2(\omega^2-\Gamma_{01}^2)]}{E_{10}^2+(\hbar\Gamma_{01})^2}\right] \tag{4.71}$$

4.6 Graphical Interpretation of the Optical Susceptibility

The numerical values of the parameters used in graphical illustrations are the carrier density $\rho_v = 5X10^{22}m^3$, the intensity, $I = 6X10^8\frac{\omega}{m^2}$,the medium refractive index $n_r = 2.92$, the back ground dielectric constant $\epsilon_R = 8.5$, for ZnO, the confining frequencies ($\omega = 8.5X10^{14}s^{-1}$, $1.3X10^{15}s^{-1}$, $1.71X10^{15}s^{-1}$), the energy differences ($E_{10} = 1.67X10^{-19}J$, $2.57X10^{-19}J$, $3.36X10^{-19}J$), the dipole moments ($\mu = 1.3X10^{-28}c-m$, $1.42X10^{-28}c-m$, $1.51X10^{-28}c-m$) and the transition frequencies($\omega_{10} = 1.6X10^{15}s^{-1}$, $2.45X10^{15}s^{-1}$, $3.2X10^{15}s^{-1}$).

In Figure 4.1, the real part of the third order nonlinear susceptibility as a function of photon energy is illustrated based on above parameters. From the figure it can be observed that as the confining frequency of the quantum dot increases, the transition frequency increases and the magnitude of the third order nonlinear susceptibility increases.

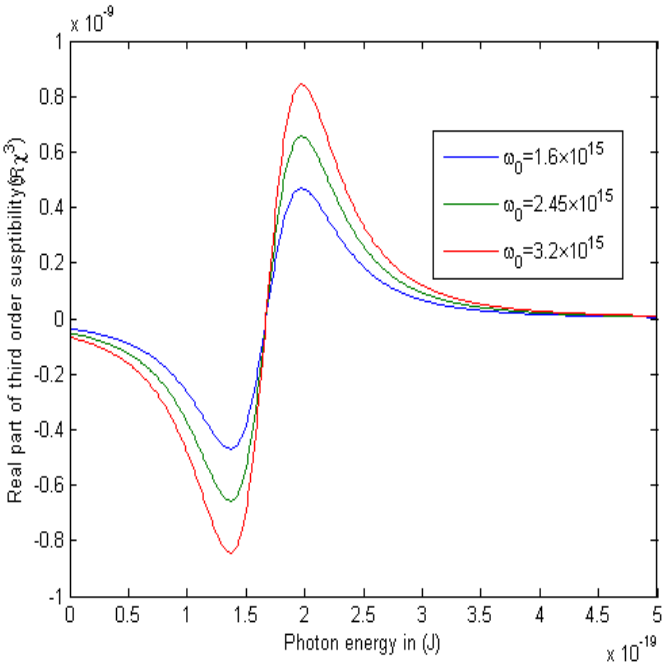

Figure 4.1: The plot of real part of third order optical susceptibility versus photon energy

In Figure 4.2, the imaginary part of the third order nonlinear susceptibility as a function of photon energy is described with respect to the above mentioned parameters. The result confirms that, as the confining frequency increases, the transition frequency increases and the magnitude of the imaginary part of the third order susceptibility is magnified. Where ω_0 is the confining frequency.

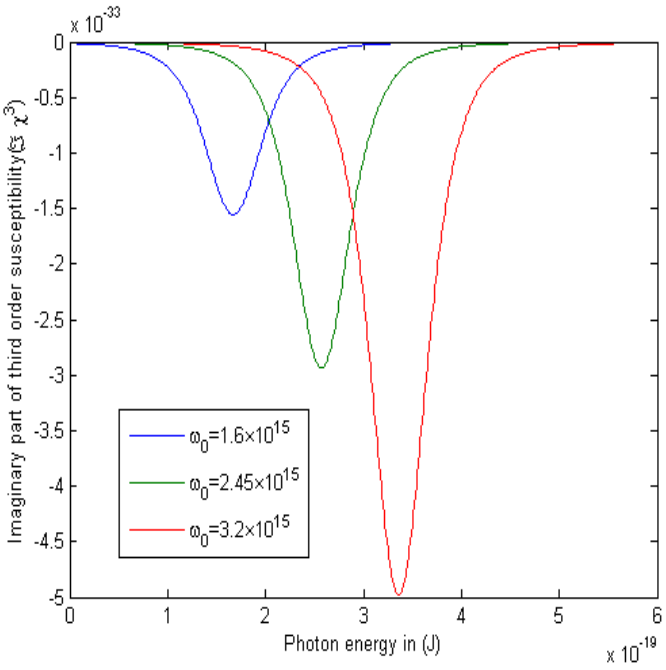

Figure 4.2: The plot of imaginary part of third order optical susceptibility versus photon energy

5

Conclusion

In this thesis we have developed the hamiltonian and trial wave functions for three dimensional two electron Zinc oxide(ZnO) quantum dot with parabolic confinement. Based on these hamiltonian and trial wave functions the ground and excited state energies are calculated using variational technique.

Finally, employing the calculated values of the ground and excited state energies for the coupling constant $\lambda = 1$, we determine the real and imaginary part of third order susceptibilities. The result shows that the magnitude of the real part of the third order susceptibility increases as the confining frequency increases. Moreover, the magnitude of the imaginary part of the third order susceptibility increases as a confining frequency which is related to the transition frequency increases.

Bibliography

[1] R.P.Feynman, *"There is plenty of room at the bottom,"*Am. Phys. Society vol.**23**, PP5-22(1960)

[2] F.M Peters and O.Hipolito, low dimensional semiconductor structures, Brazilian .J.Phys. vol.22, 3, PP183-193(1992)

[3] Michael .R.Galley, Quantum Phenomena in low dimensional systems .Fundamental of physics vol.II, PP.1-23 (1997).

[4] M.Dineykhan and R.G Nazmitdinov, two electron quantum dot in a magnetic field: Analytical results pp (1996).

[5] Charles Kittle, Intro.to solid states physics (8th), John Willey and Sons, Inc pp .518 -562 (2005).

[6] R.P Feynman. Caltech Engineering and Science 23, pp.5-22. (1960)

[7] John Basco Balagaru and B.G Jeya prakash, electrical and electronics - semiconductor nano devices, pp.3-29, (1983)

[8] John Davis, The physics of low dimensional semiconductor, Cambridge University press (1998)

[9] Klaus D .Sattler, nanoparticles and quantum dots. London New York, pp.518-562 (2005)

[10] Robert W.Boyd, Nonlinear optics, Rochester, NewYork, third ed. pp1- (2007)

[11] N.S.Simonovic, Hidden symmetries of two electron quantum dots in magnetic field (2004)

[12] X.F.Wang. phys.Lett.A 364, 66 (2007)

[13] SJari and R.Benaceur exaction in parabolic quantum dots in electric and magnetic fields (1994)

[14] Rolf Kole, ester Groeneveld, Daniel Van Maekel -Berg, Andries Meijrink and Celsode Mello Donega, ch2 : size effects on semiconductor nanoparticles , Springer - verlag , pp14-40 Berlin (2014)

[15] Miller. D.A.B, "Quantum well optoelectronic switching devices"Int .J.of high speed electronics Vol. 19-46 (1990)

[16] D.F. Sulis bury," Quantum dots that produce white light could be the light bulbs Successor" http: // www .physorg.com news 7421.htm

[17] *S.S.Rink D.A.B Miller, D.S Chemila*, phys .Rev. B35, 8114 (1986)

[18] Garnett W .Bryant, phys rev .B521 (1995)

[19] *L, W, Ping, Xia o, Jing-Lin, Y.J, WhenYu Yi-Fu and Wang -Zi-Wu,chin* .phys .Rev .B19, 047102 (2010).

[20] Mark Fox, optical properties of solids, oxford university press, third ed. Pp1 (2001)

[21] VladimirV. Mitin, Dmitry I. Sement Sov and NAizamiz. Vagidov, Quantum mechanics for nanostructures, Cambrige University press, NewYork,pp 105-239 (2010)

[22] D. Bera, L.Qian, Teng - kuan Tseng and P.H. Holloway, Quantum dots and their multimodalApplications: material 3, pp2277 -2290 (2010)

[23] Matyas Molnar "Nonlinear Optics Semiconductor Harmonic Generation" Unpublished thesis.

[24] Zis. ASW Wong, V Thanasi, R Jos's Ramakrishna and GHHO, Controlled synthesis and application of ZnO nanoparticles, nanorods nanospheresin dye sensitized solar cells, IOP, Vol.20 pp.045604-045610(2009)

[25] Richard Cisek, Virginijus Barda Harry E .Ruda, and Alexander Shik, "Nonlinear optical properties of nanowires ", Vol. 17, pp1-8, (2011)

[26] R.John Basco Balaguru professor, Quantum wells, Quantum wires, Quantum dots, quantum limit of conductance capacitance and Hall Effect, PP.3(1983)

[27] Tian tian, "optical properties of low dimensional semiconductor materials "unpublished thesis.

[28] Cui-Hong Liu and Bai-Ru Xu, Phys.Lett.A.372, 888 (2008)

[29] C.J. Zhang, K.X. Guo,Z.ELu,Physica E 36, 92 (2007)

[30] I. Karabulut, S. U nlu, H. Safak, Phys. Status Solidi B 242, 2902 (2005)

[31] L. Liu, J.J. Li, G.G. Xiong, Physica E 25, 466 (2005)

[32] Y. Masumoto, S. Okamoto, and S. Katayannagi, Phys. Rev. B50, 18658 (1994)

[33] Y. Masumoto, T. Kawamura, and K. Era, Appl. Phys. Lett. 62, 225 (1993)

[34] T. Kataoka, T. Tokizaki, and A. Nakamura, Phys. Rev. B48, 2815 (1993)

[35] Menberu Mengesha, Quantum mechanical and optical properties of assymmetric double wells and two electron quantum dots (2011).

[36] Mulugeta Duressa, The low lying states of two electron quantum dot with parabolic confinement (2011).